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Coupling between semiconductor quantum dots and two-dimensional surface plasmons

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When CdSe/CdS core/shell quantum dots (QDs) are dispersed onto a two-dimensional (2D) textured Ag grating, their luminescent behavior is dramatically modified. Excited QDs can resonantly excite surface plasmon polaritons (SPPs) propagating along the modulated metallic surface. The measurements of PL spectra and time-resolved PL spectra clearly indicate the interaction between QDs and 2D SPPs. The electromagnetic interaction between QDs and the 2D corrugated metal surface makes the luminescence be enhanced, polarized, and well directed.

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In recent years, semiconductor quantum dots (QDs) have attracted a lot of interest due to their optical properties.[1,2] Their spontaneous-emission behaviors depend not only on their own intrinsic properties (such as size, shape, composition, and surface morphology),[3] but also on the nature of their surrounding environment, more specifically on the photonic mode density at their location.[4,5] Previous experiments have found that the radiative lifetime of QDs near a smooth semiconductor (or metal) surface exhibits a damped oscillatory dependence on the separation between the QDs and the surface.[6] If the QDs are very close to the surface, the energy of the excited QDs will be nonradiatively transferred to the semiconductor/metal, and their luminescence will be strongly quenched. However, it was recently reported that the luminescence of QDs on a "rough" gold film is dramatically enhanced via electromagnetic interactions with the surface plasmon polaritons (SPPs).[4] The experiment with dye molecules on a one-dimensional (1D) metallic grating also demonstrated the modified optical radiation.[7] SPPs are nonradiative transverse magnetic (TM) modes that propagate along a metal surface. If the metal surface is periodically modulated on the scale of the light wavelength, there is an energy gap in the mode dispersion.[8] A two-dimensional (2D) modulation of the metal surface can generate a "full photonic band gap,"[9] which is similar in origin to the photonic band gap that occurs in photonic crystals.[10] If a light emitter is placed inside an optical medium with a photonic band gap, its luminescent behavior can be significantly affected due to the modification of the photonic mode density at its location.[11] Here, we report our experimental investigations on the electromagnetic interaction between single QDs and SPPs generated on a 2D textured metal surface. The radiative behaviors of CdSe/CdS core/shell QDs are dramatically modified due to their efficient coupling with the SPPs. These studies can impact not only on our fundamental understanding of the interactions between SPPs and nanoscale semiconductor light emitters, but also on potential applications in optoelectronic devices.[12,13]

The 2D textured metal structure was fabricated by evaporating a 200-nm-thick silver film onto a 2D grating, which was obtained by a holographic technique on the surface of a 330-nm-thick azobenzene containing polymer film that had been deposited onto a glass substrate.[14] The inscribed grating has a sinusoidal profile with the grating spacing varying between 300–400 nm in both dimensions. Figure 1 is the typical scanning electron micrograph (SEM) of the 2D textured Ag surface. The structured metallic surface consists of hexagonally arranged "dots," whose height is ~30 nm. The left inset shows the schematic of the surface, in which the direction ψ is defined. Three samples, with grating spacing of 300, 320, and 400 nm (which are labeled hereafter as samples *A*, *B*, and *C*, respectively), were chosen for the current study. The QDs used in the study were colloidal CdSe/CdS core/shell QDs with a ~4 nm CdSe core diameter and a three-monolayer CdS shell, whose photoluminescence (PL) peak is at ~610 nm, as shown in the right inset of Fig. 1. The QDs' initial quantum yield is ~0.5. Single QDs were dispersed onto the metallic substrate by spin coating from a very dilute solution of the QDs in tolunce. The average distance between adjacent QDs was estimated to be more than 10 μm, which prevents any direct interaction between the adjacent QDs.[15] The angularly dependent reflectivity of the metallic structure does not exhibit any observable change after depositing the QDs on its surface, indicating the low density of QDs on the sample surface. For comparison in optical measurements, a reference sample was also prepared by depositing QDs with the same concentration onto a smooth silica substrate.

The PL decay curves were recorded by employing a time-correlated photon-counting system with a pulse laser beam (wavelength of 400 nm, ~1 ps duration, and 16.4 MHz repetition rate) as the excitation source. PL spectra were recorded by a liquid-nitrogen-cooled charge coupled device (CCD) which was mounted behind a monochromator and the samples were excited by the 514.5 nm line of an Ar^+ laser. The PL measurement system used in the study is the standard far-field microscopy technique in a backscattering configuration.[16] All measurements were carried out at room temperature. Luminescence collected by the PL microscopic system is estimated to be from ~10 QDs. Due to the spectral diffusion and PL blinking behavior of colloidal QDs,[17] the obtained PL spectral shape would not be stable if the CCD exposure gating time is too short. For the reference sample (i.e., on the smooth silica), the PL spectra were not stable for a CCD integration time of less than 1.0 sec. On the other hand, in the case of QDs on the 2D metallic surface, the PL spectra became stable for a CCD integration time of about

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Generalized dark-state polaritons for photon memory in multilevel atomic media

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We generalize the concept of dark-state polariton in multilevel electromagnetically induced transparency systems. We show that the quantum states of light for the pulses can be mapped onto more than one collective atomic polarization states of the multilevel atomic system, which can act as a quantum state copier or divider. Such dark-state polaritons potentially have applications in quantum information processing.

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Optical memories using classical techniques of spin and photon echoes are quite powerful as far as the high capacity of data storage is concerned [1–6]. However, direct applications of these techniques for quantum memories are very much limited as the number of photons required is larger than number of atoms in the system. Quantum memories are essential in quantum information processing, which involves physically transporting or communicating quantum states between different nodes of a quantum network [7]. It has been well recognized [8] and now experimentally demonstrated [9] that photons can be used as ideal carriers of quantum information and ensembles of atoms can act as long lived storage and processing units. The basic requirement for a reliable quantum memory system is its capability of storing and releasing quantum states on demand at the level of individual qubits, which puts stringent conditions on coherent transfer of information among photons and atoms.

Among many methods for coherently controlling the photon-atom interactions, the techniques based on electromagnetically induced transparency (EIT) [10] have been shown to be very promising in quantum state manipulations [11] and enhanced nonlinearity applications [12]. EIT systems can be transparent for the probe light beam at certain frequencies due to quantum interference and provide a large variation in linear dispersion within the transparency window [13]. The sharp change in linear dispersion can lead to substantial group-velocity reduction of light [14] to preserve quantum states of the slowed down light pulses, thus allowing the atomic medium to act as a temporary storage or buffering device for the quantum states of light. Physically, the photon storage in an EIT system is due to the formation of so called dark-state polariton (DSP), which is a mixture of electromagnetic signal field and the collective polarization of the atomic system, controlled by the strong coupling beam. It is the mixing angle of the signal field and the collective atomic polarization, which determines the group velocity of the signal pulse propagating in the atomic medium. By adiabatic following of DSP it is possible to reduce the group velocity of light pulses and to convert the photon states into the metastable atomic-polarization state. The stored light field in the collective atomic polarization can be recovered by manipulating the mixing angle of the DSP components by controlling the coupling beam. The concept of DSP was first introduced in Raman adiabatic passage by Mazets and Matisov [15] and then formulated for the three-level EIT system by Fleischhauer and Lukin [16]. Recent experiments have demonstrated photon storage using the EIT system consisting of three-level atoms in Λ configuration due to dynamic group-velocity reduction via adiabatic following in DSP [9].

In this work we extend the concept of a simple DSP to a generalized dark-state polariton (GDSP) for a multilevel atomic system having more than one dark state. One such example is the inverted-Y configuration involving four atomic levels that can easily be realized in rubidium atoms [17]. As one will see in the following paragraphs that such a system can preserve quantum states of light in two different collective atomic polarization states of the same atomic medium and thus provide channelization or bifurcation of photon memory. The two different collective atomic polarization channels contributing to GDSP can be used to retrieve back the light pulses on demand at different times or at the same time. In a way this system acts as a copier or divider of the quantum memory.

A schematic diagram of the closed four-level atomic system in inverted-Y configuration is depicted in Fig. 1. Levels $|a\rangle$, $|b\rangle$, and $|c\rangle$ are in a three-level Λ-type configuration and levels $|b\rangle$, $|a\rangle$, and $|d\rangle$ form a three-level ladder-type configuration. The transition from $|b\rangle$ to $|a\rangle$ (with frequency ω_{ab}) interacts with a signal field of amplitude $\hat{\varepsilon}(z,t)$ [defined below in Eq. (4)] and frequency ν. A coupling field of frequency ν_{ac} (considered to be classical) drives the transition $|c\rangle$ to $|a\rangle$ (frequency ω_{ac}) with Rabi frequency $\Omega_c(z,t)$, while a pumping field of frequency ν_{ad}, which is also considered to be classical, drives the transition $|a\rangle$ to $|d\rangle$ (frequency ω_{ad}) with Rabi frequency $\Omega_d(z,t)$. If there is no coupling (pump-

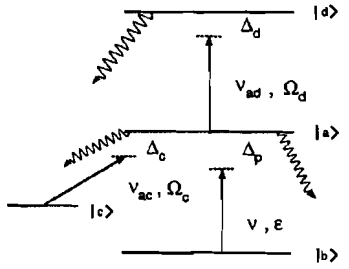

FIG. 1. Schematic diagram of a four-level atomic system in inverted-Y configuration. Here, ε, Ω_c, and Ω_d define signal, coupling, and pumping fields, respectively.

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Enhancement of the cavity ringdown effect based on electromagnetically induced transparency

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We show that the unique absorption and dispersion properties of the electromagnetically induced transparency can be used effectively to relax the conditions for observing the cavity ringdown effect (CRE), which can be useful in applications of CRE in ultrasensitive detection of chemical species. A more straightforward and simple method is used to model the interesting CRE. © 2004 Optical Society of America

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When an optical cavity is excited by a monochromatic wave and the cavity length is scanned faster than the cavity round-trip time, the cavity output field profile shows an amplitude oscillation in its normal decay curve, leading to a phenomenon known as the cavity ringdown effect (CRE).[1,2] Such oscillation is found to originate from the interference between the original input laser field and the intracavity circulating field, and the oscillation frequency can be controlled by the cavity scan speed.[1,2] Many interesting applications of CRE have been found, including measuring ultraslow velocities of cavity mirrors,[1] measuring ultrasensitive direct absorption,[3,4] studying the chemical kinetics and absorption bands of molecules,[5–7] and measuring the concentration of minor species in flames.[8,9]

Although the CRE is useful in practical applications, it suffers from serious limitations because of the stringent requirements for high cavity finesse, long cavity length, and fast cavity scan speed. For observation of the CRE, the cavity scan rate needs to be faster than the photon round-trip time inside the optical cavity, which typically demands that the cavity finesse be at least 10^5 for a cavity scan rate of 1 μm/s. In this Letter we propose to use an intracavity medium with electromagnetically induced transparency[10–12] to relax the strong conditions mentioned above and, therefore, enhance the CRE, which will promote potential applications that use this phenomenon. An EIT medium can have a sharp dispersion change near its EIT resonance,[13] which can slow down the speed of photons inside the optical cavity.[14–16] Typically an intracavity medium with sharp dispersion will be accompanied by significant absorption, which damps the ringdown oscillation.[1] However, an EIT medium is ideal in this regard, since it has a large dispersion change without any absorption at exact EIT resonance.

In previous works describing the CRE, the electric field inside the resonator at any instant was obtained by summing up all the wave components that underwent multiple reflections.[1,2] Here we present a more straightforward method of modeling the intracavity electric field. In what follows we use α_p ($|\alpha_p|^2$ is the average photon flow, expressed in number of photons per second) to denote the intracavity field. Consider a ring cavity consisting of four mirrors, as shown in Fig. 1. M_1 and M_3 are input and output mirrors with intensity reflectance R. M_2 and M_4 are perfect reflectors. M_2 is mounted on a piezoelectric transducer for cavity length scanning. The change of intracavity probe field α_p during a round-trip time duration τ_0 is due to the driving field $\alpha_p{}^{\text{in}}$, to the cavity decay γ_{cav}, and to the round-trip phase shift Φ_{cav}:

$$\tau_0 \frac{d\alpha_p}{dt} = \sqrt{R}\,\alpha_p{}^{\text{in}} - \gamma_{\text{cav}}\alpha_p + i\Phi_{\text{cav}}\alpha_p . \qquad (1)$$

For an empty cavity the total round-trip phase shift is proportional to the geometrical length of the cavity and can be expressed as $\Phi_{\text{cav}} = 2\pi(d_0 + v_{\text{cav}}t)/\lambda_p$, where d_0 is the initial cavity length, v_{cav} is the cavity scan speed, and λ_p is the wavelength of the input field. After substitution of Φ_{cav} into Eq. (1), it be solved analytically to give

$$\alpha_p(t) = \frac{\alpha_p{}^{\text{in}}\sqrt{\pi R}}{\sqrt{2i\tau_0 a}} \exp\left[\frac{(\gamma_{\text{cav}} - iat)^2}{2i\tau_0 a}\right] \times \left[\text{erf}\left(\frac{\gamma_{\text{cav}}}{\sqrt{2i\tau_0 a}}\right) - \text{erf}\left(\frac{\gamma_{\text{cav}} - iat}{\sqrt{2i\tau_0 a}}\right)\right], \qquad (2)$$

where d_0 is ignored (the cavity is in resonance with the field initially), $a = 2\pi v_{\text{cav}}/\lambda_p$, and erf is the error function. Under certain conditions (which will be

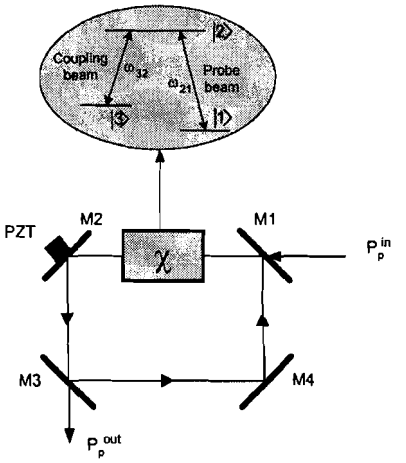

Fig. 1. Four-mirror optical ring cavity with an intracavity atomic medium. The three-level Λ-type system is shown in the oval at the top. PZT, piezoelectric transducer.

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On the bichromatic excitation of a two-level atom with squeezed light

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Abstract. Analytical results for the dynamical evolution of a single two-level atom coupled to an ordinary heat bath and bichromatically excited with two finite bandwidth squeezed fields are presented by solving the Heisenberg equations of motion. Photon statistics of the system in terms of the second-order intensity-intensity correlation function are also discussed. Transient fluorescent intensity as well as intensity correlation function exhibit oscillatory phenomenon even in the weak field limit. The latter also shows enhanced delayed bunching effect. All these effects are sensitive to the bandwidth of squeezed light.

PACS. 42.50.Ct Quantum description of interaction of light and matter; related experiments – 42.50.Dv Nonclassical states of the electromagnetic field, including entangled photon states; quantum state engineering and measurements

1 Introduction

Studies related to the dynamical evolution, the photon statistics, spectral and radiative properties of one and many two-level atoms embedded in a broadband squeezed bath have been the topics of keen interest in quantum optics. Gardiner [1] studied the interaction of a two-level atom with a broadband squeezed bath and predicted unequal polarization quadrature-decay rates. Carmichael, Lane and Walls [2] were able to discover a significant phenomenon of sub-natural linewidth in the fluorescence spectrum of a driven two-level atom, in the presence of squeezed light. Atomic absorption spectrum was discussed by Ritsch and Zoller [3] in the presence of colored squeezed vacuum. Since then many interesting results in atom-squeezed field interaction have been reported which include both two and three-level atoms interacting with broad bandwidth or narrow bandwidth squeezed baths [4, 5]. In a recent study the interaction of a two-level atom with the squeezed vacuum of bandwidth smaller than the natural atomic linewidth was considered and the hole burning and the three-peaked structure in spectra of fluorescence and transmitted field were predicted [6]. These results essentially show that squeezed fields having pairwise correlations and anisotropic noise distribution can give rise to interesting phenomena including novel features in spectral properties of atoms, formation of pure states and photon statistics. A more realistic model of finite bandwidth squeezed light interacting with a single two-level atom has been studied by Vyas and Singh [7] and Lyublinskaya and Vyas [8] where the source of the squeezed light employed was a degenerated parametric oscillator (DPO) operating below threshold and a homodyned DPO [9]. In another work, a two-level atom inside an optical parametric oscillator has been considered and hole and dips in the fluorescence and transmitted light has been observed [10]. The interest in other sources of squeezed light as well as its applications in a wide variety of areas has continued unabated [11–13].

The interaction of a single two-level atom with bichromatic driving field has also been studied extensively both theoretically and experimentally [14]. These studies were motivated by the observations that the bichromatic nature of the driving field can lead to a number of novel features which are different from the monochromatic case. For example, the fluorescence intensity exhibits resonances at subharmonics of the Rabi frequency and different spectral characteristics when compared with the usual Mollow triplet. Recently, some new calculations for resonance fluorescence and absorption spectra of a two-level atom driven by bichromatic field have been reported [15]. Also, reported are the effects of broadband squeezed reservoir on the second order intensity correlation function and squeezing in the resonance fluorescence for a bichromatically driven two-level atom [16]. Coherent population trapping and Sisyphus cooling under bichromatic illumination have also been studied [17]. In another recent work the electromagnetically induced transparency (which normally occurs in three-level atoms) has been demonstrated in a two-level atom excited by a bichromatic field (one strong and one weak field) and possibility of squeezed-light generation has also been discussed [18].

In this work, we study the interaction of a single two-level atom with a bichromatic electromagnetic field that is

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Polarization spectroscopy of InGaAs/GaAs quantum wires grown on (331)*B* GaAs templates with nanoscale fluctuations

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Using (331)*B* GaAs templates with nanoscale fluctuations, we have fabricated InGaAs/GaAs quantum wires (QWRs) with a density of $\sim 2.0\times 10^6$ cm^{-1} and the degree of polarization as high as ~28%. In the samples with weak lateral confinement, we observed thermal delocalization of carriers from the one-dimensional QWR states to the two-dimensional quantum-well states with increasing temperature, which is almost absent in QWR samples with strong lateral confinement.
 [DOI: 10.1063/1.1637721]

Low-dimensional quantum wells (QWs), quantum wires (QWRs), and quantum dots (QDs), are important quantum systems for both the study of fundamental physics and the potential applications in optoelectronic devices. Among these nanostructures, the one-dimensional (1D) QWRs are the smallest dimensional structures that can be used for efficient transport of electrons, and also as polarization-sensitive nanoscale photodetectors that may be useful in various fields.[1] Compared with the well-fabricated and -studied two-dimensional (2D) QWs and zero-dimensional (0D) QDs, the 1D QWRs have only become the focus of intensive investigations recently. This is partly due to the reason that fabrication of 1D QWRs has always been a challenge for crystal growth technology since a higher structural ordering is required, and also the underlying mechanisms for anisotropic growth along one direction are still not completely understood.[2]

In the past decade, many methods have been adopted to fabricate semiconductor QWRs, in which self-organized growth is one of the most promising approaches since it can effectively avoid the processing damage and contamination that might be introduced by some other fabrication processes, such as ultrafine lithography and chemical etching. Specifically, molecular beam epitaxy (MBE) growth of self-organized InGaAs/GaAs QWRs on high-index GaAs substrates has been shown to be an easy process to fabricate QWR structures with high lateral density, high uniformity, and high optical quality.[3–5] Basically, in the conventional process of growing narrow InGaAs/GaAs QWs,[4,5] the use of a high-index substrate would result in a flat, lower interface and a corrugated upper interface, which may act as lateral potentials for the carriers, hence, the formation of 1D QWRs. The realization of ordered QWR structures can be attributed to the surface reconstruction or atomic rearrangement observed on high-index surfaces with a high density of multiatomic steps or microfacets.[6]

In our previous work,[7] we observed that GaAs (331)*A* and (331)*B* surfaces are both faceted on a nanometer scale, containing (110) and (111) facets, and it was further proposed that the resulting highly anisotropic ridge-like surfaces could be used in the fabrication of high-quality and high-density QWR structures. Here, we report the MBE growth of InGaAs/GaAs QWRs on high-index (331)*B* GaAs templates with nanoscale fluctuations. Photoluminescence (PL) from these QWR structures at 8 K shows the degree of polarization to be as high as ~28%, which indicates good 1D carrier confinement. The density of these QWRs is estimated to be $\sim 2.0\times 10^6$ cm^{-1}, which meets the requirement of high optical gain for laser applications.[3,5] In the samples with weak lateral confinement, we observed thermal delocalization of carriers from the one-dimensional QWR states to the two-dimensional quantum-well states with increasing temperature, which is almost absent in the QWR samples with strong lateral confinement.

The samples used in our experiment were grown using a solid source MBE system (Riber 32) under an As beam equivalent pressure of 1×10^{-5} Torr. The surface evolution during MBE growth was characterized by *in situ* reflection high-energy electron diffraction (RHEED) and scanning tunneling microscopy (STM). A GaAs buffer layer, with a thickness of 500 nm, was first grown on the *n*-type GaAs (331)*B* substrate at a temperature of 610 °C. As shown in the bottom of Fig. 1, the resulting surface morphology, with straight ridge-like corrugation bounded by the (111) and (110) facets, was quenched to 540 °C as a template for the subsequent InGaAs overgrowth. The surface corrugation has an average lateral periodicity of ~5.0 nm with a vertical amplitude of ~1.0 nm. A detailed description of preparing such (331)*B* GaAs templates with nanoscale fluctuations can be found in Ref. 7. During the InGaAs overgrowth at 540 °C, atomically flat surfaces were developed for all the samples with different deposition thickness (>1.0 nm), as monitored from the RHEED patterns and STM images. Finally, a 20 nm GaAs cap layer was grown at 540 °C for each sample to maintain the smooth InGaAs surface configuration. The QWR structures are thus formed at thick parts in the InGaAs/GaAs QW with a corrugated bottom InGaAs-on-GaAs interface and a smooth top GaAs-on-InGaAs interface, which is schematically shown in Fig. 1. The structure of as-grown QWRs can be viewed as a totally "inverted" counterpart compared with

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Anisotropic photoconductivity of InGaAs quantum dot chains measured by terahertz pulse spectroscopy

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We report results of time-resolved terahertz (THz) pulse spectroscopy experiments on laterally ordered chains of self-assembled InGaAs quantum dots photoexcited with 400 nm, 100 fs laser pulses. A large anisotropy in the transient photoconductive response is observed depending on the polarization of the THz probe pulse with respect to the orientation of the dot chains. Fast (3.5–5 ps) and efficient carrier capture into the dots and one-dimensional wetting layers underneath the dot chains is observed below 90 K. At higher temperatures, thermionic emission into the two-dimensional wetting layers and barriers becomes significant and the anisotropy in the photoconductive signal is reduced. © *2004 American Institute of Physics.*
[DOI: 10.1063/1.1807959]

Carrier capture in self-assembled quantum dots (QDs) has been an extremely active area of research in recent years, in part due to the potential applications for tunable, efficient QD laser structures[1] and photodetectors.[2] Often, carriers are injected into the barrier or wetting layers (WLs) and must be captured by the QDs before they can radiatively recombine. These capture mechanisms are therefore intimately linked to the operating parameters of QD lasers and other QD photonic devices.

Recently, a high degree of linear ordering of QDs was achieved in an $In_{0.36}Ga_{0.64}As/GaAs$ superlattice by controlling strain in the Stranski–Krastanow growth process.[3,4] Researchers were able to fabricate aligned dot chains with an average length of 0.9 μm, as shown in Fig. 1(a), with dot densities of $\sim 1.8\times 10^{10}$ cm^{-2} per layer. This structure leads to very different potential profiles parallel (||) and perpendicular ($\perp$) to the dot chains. This letter demonstrates that polarized, subpicosecond, far-infrared light pulses can be used to probe anisotropic carrier transport resulting from this ordering.

The sample was grown on a semi-insulating (SI) GaAs [001] substrate with a 150 nm GaAs buffer layer deposited by molecular beam epitaxy at 580 °C. A 15-layer structure of $In_{0.36}Ga_{0.64}As/GaAs$ was then grown at 540 °C, each layer containing densely packed chains of QDs with an average diameter and height of 45 nm and 5 nm, respectively. The transmission electron microscopy (TEM) plan-view image of Fig. 1(a) shows the dot chains running in the $[1\bar{1}0]$ direction. TEM images also reveal the existence of a one-dimensional (1D) WL with an estimated height of 1.5–2 nm directly underneath each dot chain, all sitting on top of a ~0.7 nm thick two-dimensional (2D) WL. A schematic of this structure is shown in Fig. 1(b), and further details on growth and characterization of this sample can be found in Ref. 3.

We use time-resolved terahertz spectroscopy (TRTS) to probe the ultrafast carrier dynamics in this QD structure as a function of temperature, excitation density, and direction either parallel or perpendicular to the dot chains. TRTS has been used previously to investigate transient photoconductivity (PC) and carrier dynamics with subpicosecond time resolution in a variety of samples including bulk and thin film semiconductors,[5,6] insulators,[7] organic crystals,[8] and semiconductor nanostructures such as InP nanoparticle arrays, self-assembled InAs/GaAs quantum dots,[9–11] and GaAs/AlGaAs multiple quantum wells.[12] Specifically, the terahertz (THz) pulse used in this technique is sensitive to the product of free carrier density and carrier mobility. Once carriers become captured by traps or localized QD states, they are no longer mobile and therefore do not contribute to attenuation of the THz pulse transmitted through the sample. TRTS is therefore an ideal probe of carrier capture dynamics in QD systems, complementing traditional techniques such as photoluminescence rise time measurements.

While the details of the experimental setup have been described elsewhere,[6,13] the basic technique is as follows. The output from a ~0.7 mJ/pulse, 800 nm, 1 kHz, amplified

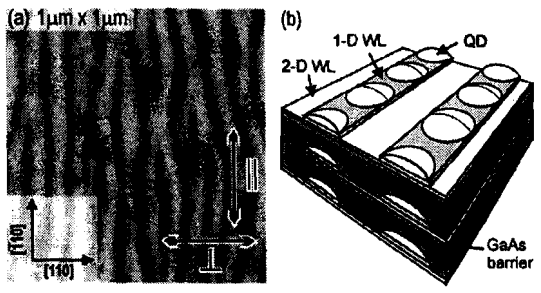

FIG. 1. (a) TEM image of QD sample surface morphology. THz polarization parallel (||) and perpendicular ($\perp$) to the dot chains is indicated. (b) Schematic diagram of the sample (not to scale) showing arrangement of QDs on 1D WLs.

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Controlling dynamic instability of three-level atoms inside an optical ring cavity

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Dynamic instability in the transmission field of an optical ring cavity containing three-level Λ-type rubidium atoms is studied in detail both experimentally and theoretically. The onset and periodicity of such dynamic oscillations in the cavity field can be controlled by the experimental parameters, such as intensity and frequency detuning of the coupling field and/or cavity field. Such nonlinear dynamic behavior is caused by competition between optical saturation of the cavity field and optical population pumping by the coupling field in the three-level atomic system.

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I. INTRODUCTION

Dynamic instability was observed and carefully studied in the upper branch of the optical bistability (OB) curve in a system with two-level sodium atoms inside an optical cavity [1]. This dynamic effect was attributed to the mechanism known as Ikeda instability [2], which appears in analyzing the stability of the steady states in OB curves, assuming a medium response time much faster than the cavity round trip time. In this limit and under certain parametric conditions, a sequence of period doubling bifurcation leading to a region of apparently aperiodic dynamic oscillation was observed. Multimode instabilities as well as oscillatory instabilities leading to optical turbulence in the OB from a two-level atomic system were also reported [3]. The off-resonant-mode instability in mixed absorptive-dispersive optical bistability was studied and it was shown that a portion of the lower transmission branch could also be unstable in addition to the upper-branch instability found in the system with pure absorptive bistability [4]. The observation of instability due to the onset of the cavity side mode was reported in a bistable optical system with a homogeneously broadened two-level medium [5]. Other kinds of self-oscillation and instability were also observed using different two-level atomic systems inside an optical resonator [6]. All the above mentioned theoretical modelings and experimental observations were carried out in two-level atomic systems. A different kind of dynamic instability was observed in the transmission field of an optical cavity consisting of a cold cloud of cesium atoms [7]. In this system the degenerate Zeeman sublevels of the $6S_{1/2}$ $F=4$ and $6P_{3/2}$ $F'=5$ states interact with two cavity fields pumped by one input circularly polarized laser beam. The instability was considered to be caused by competition between optical pumping to the state $6S_{1/2}$ $F=4$, $m_F=4$ from all other Zeeman sublevels and optical saturation of the transition from the state $6S_{1/2}$ $F=4$, $m_F=4$ to the state $6P_{3/2}$ $F'=5$, $m_{F'}=5$. The observed oscillatory behavior in the cavity output field was modeled as a quasi-two-level system interacting with one cavity field and only qualitative comparison was made between the observed phenomenon and a simplified theoretical model [7]. The complexities in that system are mainly caused by the degenerate Zeeman sublevels involved and the trapping and the repumping laser beams for creating the cold atomic cloud. The major limitation of that experiment was the use of only one cavity input (circularly polarized) beam to provide both linearly polarized optical pumping and probing (or saturation) beams, which prevented the two competing physical processes from being independently adjusted to systematically study the dynamic instability.

Recently, we have experimentally observed similar dynamic instability in a system consisting of three-level atoms inside an optical cavity, as shown in Fig. 1, and demonstrated dependence of such dynamic behaviors on the intensity of

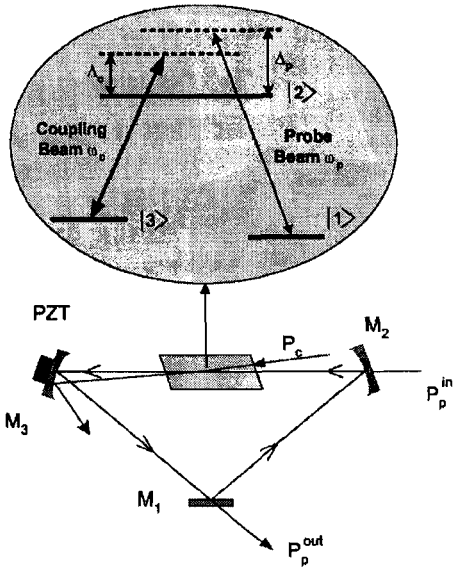

FIG. 1. Sketch of a three-level Λ-type atomic system and the optical ring cavity. M_1, M_2, and M_3 are mirrors; PZT is a piezoelectric transducer; $\Delta_c=\omega_c-\omega_{23}$, and $\Delta_p=\omega_p-\omega_{21}$ are the coupling and probe frequency detunings, respectively; P_c is the coupling laser beam; P_p^{in},P_p^{out} are the input and output probe laser beams, respectively.

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Suppression of radiative decay of CdTe quantum dots in a photonic crystal with a pseudogap

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Abstract. Observation-angle dependence of the spontaneous emission lifetime of CdTe quantum dots (QDs) embedded in a pseudogap photonic crystal (PC) film has been demonstrated. Comparison of two PC films with different photonic band-gaps (PBGs) differentiates the PBG effect from the electronic and/or chemical interactions between CdTe QDs and the host medium. This lifetime modification of QDs by a PC with pseudogap can be very useful in applications for optoelectronic devices such as QD lasers and QD switches.

Photonic crystals (PCs) are one-, two- or three-dimensional artificially engineered periodic structures [1–9], in which the refractive index varies on the length scale of optical wavelength. Analogous to the wave-like propagation of electrons in a crystalline structure, the propagation of light in a PC is diffracted for a certain range of wavelengths, leading to the formation of a photonic band-gap (PBG). The propagation of photons with energies within the PBG is not allowed throughout the PC. Much attention has been paid to the optical properties of the PCs in recent years owing to their potential applications in optoelectronic devices. PCs can efficiently modify the spontaneous emission (SpE) of light emitters embedded in them. For example, because of the PBG effect, SpE of emitters inside PCs can be amplified [3, 4], which is necessary for the realization of lasing without a threshold [10]. During the past few years, many kinds of light emitters, such as fluorescent dye molecules [4, 5], semiconductors [3, 6, 7], ions [8] and metals [9], have been filled into PCs to study the altered optical properties. However, few reports [11] have focused on the modified optical properties of semiconductor quantum dots (QDs) embedded inside the PCs. Semiconductor colloidal QDs are intermediate in size between single molecules and bulk solid-state materials and are of great interest for both fundamental research and technical applications owing to their relatively reproducible and controllable synthetic chemistry. It has been shown that the SpE of colloidal QDs in a PC can be strongly modified [12]. Furthermore, the controllable size (less than 10 nm), strong size-dependent optical emission and narrow photoluminescence (PL) width of colloidal QDs make them ideal candidates to prove the interactions between the SpE of light emitters and the PBG effect of PCs. It has long been proposed that the colloidal QDs can be used as active laser

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Modulation transfer in an electromagnetically induced transparency system

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Near the condition of electromagnetically induced transparency in a three-level system, the absorption experienced by the weak probe laser beam interacting with one transition is very sensitive to the frequency detuning of the strong-coupling laser beam coupled to another transition. When the frequency detuning of the coupling laser is modulated at different frequencies, the resulting change in probe absorption as a function of that modulation frequency can be described by a transfer function. The effects on the transfer function from various experimental parameters are calculated numerically. Experimental measurements are shown to be in good agreement with the predicted behavior.

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I. INTRODUCTION

Systems capable of exhibiting electromagnetically induced transparency (EIT) [1,2] have enjoyed recent popularity for slowing light [3,4], quantum information processing [5], photon storage [6,7], all-optical switching [8–10], and all-optical buffering. Since EIT is basically formed by a strong-coupling laser beam together with a probe laser beam (for three-level EIT systems), both linear (absorption and dispersion) [2,11] and nonlinear (Kerr nonlinearity) [12] optical properties of the probe beam can be effectively controlled by the coupling beam. In the steady state, the absorption reduction of the probe beam at EIT resonance can be considered to be a narrow-bandwidth frequency filter [13]. In the case of slowing down the group velocity of light pulses, very rapid dispersion change is required to achieve large group velocity reduction [3,4,11], which demands a very narrow EIT spike. However, such a narrow EIT window restricts the frequency bandwidth of the interacting pulses and, therefore, is only useful for slowing relatively long pulses (microsecond pulses in Refs. [3,4,14]). Similar conditions also apply to storing of optical pulses in such EIT media [6,7]. These competing trade-off conditions, i.e., needing sharp dispersion for slowing down group velocity of pulses versus requiring large bandwidth to accommodate short pulses, are ultimately important in using EIT media for all-optical buffering in practical optical communication systems.

Another interesting application of EIT media is in all-optical switching. Various methods have been proposed, and some have been experimentally demonstrated by using the enhanced nonlinearity in EIT media. All-optical switching between two steady states of the probe field with three-level atoms inside an optical cavity was experimentally demonstrated both inside [8] and outside [9] the optical bistability region. Such all-optical switching was achieved by making use of the greatly enhanced Kerr nonlinearity due to self-phase modulation near the EIT resonance [12]. Optical switching due to cross-phase modulation in four-level atomic systems was also proposed [15] and experimentally demonstrated in atomic vapors [10,14]. A study of optical switching using EIT in a vapor cell by modulating the Rabi frequency of the coupling beam found that the resulting amplitude of the switching signal decreases significantly as the modulation frequency increases [16]. Recently, an experiment using stimulated Raman adiabatic passage to make fast optical switching was reported [17], which showed that the atomic absorption changes quickly (depending on the effective Rabi frequencies of the coupling and switching beams) when the switching pulse is on. Studies of transient EIT effects have also indicated that the coupling Rabi frequency is the limiting factor for rapidly changing absorption as a three-level atomic system approaches its steady state [18,19].

For practical applications of using EIT effects in all-optical communication, such as all-optical buffering, optical storage, and all-optical switching, as well as enhancing nonlinearity with slowed light pulses, one needs to address the important questions of both speed and bandwidth of the systems. For some specific applications, a fine balance between these two competing requirements (narrow EIT window for increasing dispersion to slow down the pulse speed versus broad bandwidth for short pulses as well as to achieve fast modulation speed) must be accomplished by carefully choosing the parameters of the system. In this paper, we present a systematic study of the dynamic responses of the probe laser beam to the frequency modulation of the coupling beam in the three-level EIT system, which basically gives the transfer function of the EIT medium as a function of the modulation frequency. By performing such detailed studies, both theoretically and experimentally, we are able to identify the limiting factors for the responding bandwidth and switching speed of such EIT systems, which can have significant implications in the applications of EIT media in many aspects of optical communications. By identifying the major limiting factors for the fast switching and modulation transfer in a simple EIT system, we can design or engineer optimal EIT systems (or media) for desired applications.

In Sec. II, a general theoretical model is used to study the effects of various parameters on the modulation transfer from the coupling beam to the probe beam. Both cases with and without the Doppler effect are considered and discussed. Experimental measurements of the transfer function of the three-level EIT medium in rubidium vapor are presented in

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Atomic-coherence effect on the Jaynes–Cummings model with atomic motion

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The nonlinear transient effects, similar to self-induced transparency and adiabatic following, are studied for a moving two-level atom that is entering into an ideal microwave cavity in a coherent superposition of its states. The atom undergoes a one-photon transition in the cavity, sustaining a spatial field distribution for a single-mode coherent (or thermal or Fock state) field. For some particular choice of parameters of atomic coherence, removal of an appreciable amount of field energy from the cavity could be observed. © 2004 Optical Society of America

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1. INTRODUCTION

The Jaynes–Cummings model (JCM) describing the interaction of a two-level atom with a single quantized electromagnetic field mode in an ideal cavity has been generalized and extended in several interesting directions for many years.[1–7] A very important and significant generalization of JCM is to include the atomic motion in it. By doing so, the effects of field structure of the cavity mode sustained in the cavity can be addressed.[8–10] This model is a most elementary one for studying the interaction of a single two-level atom with an electromagnetic pulse. When mode structure is taken into account, some interesting nonlinear transient effects, which are similar to self-induced transparency and adiabatic following, can be observed in the atomic inversion.[11] Here we examine these transient effects, arising from the mode structure for a moving two-level atom that is injected in a coherent superposition of its states and undergoes a one-photon transition in a single-mode coherent (or thermal or Fock state) field. The motivation behind this study is from the recent cavity quantum electrodynamics (QED) experiments that use an atomic beam passing along the axis of a cylindrical cavity so that one can study the interactions of an atom with different cavity-field modes. In Ref. 8, these effects have been examined without the atomic coherence. As we will see in the following, some particular phases of the atomic coherence can make the outgoing atomic beam (from the cavity) be with a positive atomic inversion, leading to removal of field energy stored in the cavity. In a previous work, the interaction of a two-level atom (which is in a coherent superposition of its states) with an arbitrary field in an ideal cavity was considered in the framework of standard JCM (without any atomic motion included) under the "trapping-state" condition of the interaction time.[12,13] The cavity field evolved to a pure state that had been termed as a tangent/cotangent state of the electromagnetic field.[13,14] Also in the recent past, many interesting experiments were carried out related to the atomic coherence,[15,16] which motivated us to look into the effects of atomic coherence in a true cavity QED situation.

We will neglect the cavity damping in our discussion of the model, which is a reasonable assumption in the microwave regime, where cavities of very high quality factor ($Q \sim 4 \times 10^{10}$) have been achieved.[17] The typical interaction times for the Rydberg atoms (used in microwave cavity experiments having very large dipole moments) are of the order of 10^{-5} s, and this is three orders of magnitude shorter than the lifetime of photons in a typical high-Q microwave cavity, which is ~1/100 s. Thus the assumption of negligible cavity damping when the atom enters such cavity is reasonable. Also, the radiative lifetimes of the Rydberg atoms are large enough, specifically when the circular Rydberg states are employed in the study (about tens of a millisecond), that we can also neglect the radiative damping in our model. The stability of interaction times in the experiments related to the observation of trapping-state dynamics of the micromaser[17] is uncertain within 2–3% because of the effective method employed in the velocity-selection process of the atoms. The transverse velocity spread (with respect to the cavity axis) is very small so that most atoms are moving only along the cavity axis. In the experiment of Ref. 17, an uncertainty of interaction time of 2 μs over the average interaction time of 80 μs was reported, and this uncertainty is not mainly due to velocity spread. Note that the stability of interaction times and minimum transverse velocity spread are essential in observing trapping states of electromagnetic fields in the micromaser, and hence our predictions could be tested in similar kinds of experiments.

The paper is organized as follows. The model under consideration is discussed in Section 2. The analytical solutions of the model under exact resonance condition and in far-off resonance conditions are presented in Section 3. We give numerical solutions under different field

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On the dynamical evolution of a three-level atom with atomic motion in a lossless cavity

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Abstract

The dynamical evolution of a moving three-level atom in Λ-configuration interacting with two quantized modes (or one quantized mode and a classical field) in coherent states inside an ideal cavity is studied using density matrix equations. The spatial field mode structures of the cavity are taken into account in this study. We also discuss analytic results under certain parametric conditions pertaining to nonlinear transient effects similar to self-induced transparency, adiabatic following, etc. The possibility of realizing an optical switching in such system is also described.

PACS: 42.50.Ct; 32.80.Qk; 42.50.Pq

Keywords: Cavity QED

1. Introduction

The Jaynes–Cummings model (JCM) [1] describing interaction between a two-level atom and a single quantized mode of electromagnetic field in an ideal cavity has been a center of attraction in quantum optics during last few decades as many of its predictions could be experimentally realized. Discussions related to several interesting generalizations of this model are now available in the literature [2] and the model is still promising in many applications such as motion of ions in trap, and in the design of hardwares (such as logic switches) for realizing quantum computation, etc. A very significant and noteworthy generalization of JCM is to include the effect of atomic motion so that the spatial mode structure of the cavity field could be incorporated into this model [3]. In the standard JCM, the interaction between a constant electric field and a stationary two-level atom is considered. However, when the spatial structure of the cavity field mode is taken into account, the nonlinear transient effects, similar to self-induced transparency (SIT) and adiabatic following (AF), could be observed [3]. This kind of model provides

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Effects of side-coupling on the phase response of cascaded microring all-pass filters

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Abstract

All-pass filters are useful devices that allow phase correction or equalization of signal without introducing any amplitude distortion. We present a new design scheme in which a grating is introduced in the interaction region of the two adjacent microrings in a cascaded multiring structure. The analysis accounts for the counter-directional coupling between the two waveguides. Theoretical analysis shows that this side-coupling can effectively double the filter order thus either give better performance with the same filter order or decrease the filter dimensions with the same performance.

PACS: 42.79.Gn; 42.82.Et

Keywords: All-pass filters; Delay lines; Dispersion; Coupled mode theory

1. Introduction

Lossless all-pass filters (APFs) are linear systems which have a constant amplitude response over all frequencies and a phase response that varies with frequency. Thus, APFs are, generally speaking, used as phase equalizers or dispersion compensators in optical communication systems. Ideally, the frequency response of an APF can be written as $H(\omega) = |H(\omega)| \exp[j\varphi(\omega)]$, where $H(\omega) =$ constant and $\varphi(\omega)$ can be made arbitrarily close to any desired phase response. For optical communication systems, the group delay (first derivative of the phase response) and higher-order dispersions (second and higher derivatives) are of great importance to system performance, and an optical device that allows the correction of any order dispersion is very attractive for practical applications. Besides, by using more than one stage to form high-order filters, one can get even greater design flexibility and better system performance. Recently, optically implemented APFs have received considerable attention and have found applications in dispersion compensation [1–4],

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Simple method for frequency locking of an extended-cavity diode laser

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We have developed an extended-cavity tunable diode laser system that has a small linewidth and a large output power (more than 90% of the free-running power) whose operating frequency can be conveniently locked to a transition line of Rb atoms. Based on flat-mirror feedback and frequency self-locking and with weak feedback, we have achieved a continuous frequency detuning range greater than 900 MHz and a short-time linewidth stability of better than 0.4%. By using a two-step locking procedure we not only can lock the laser frequency but also can detune the frequency to any desired value. The locking is quite sturdy and rugged. © 2004 Optical Society of America

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1. Introduction

Semiconductor diode lasers are becoming increasingly more versatile tools in atomic physics and spectroscopy research owing to their reliability in giving high power and broad wavelength coverage while they steadily decrease in cost.[1] The light from diode lasers is very bright relative to the sizes of these lasers and of other laser sources. One can easily get several watts or hundreds of milliwatts of power from a laser diode operating under continuous wave conditions. In many cases a narrow linewidth and smooth wavelength tunability are desirable characteristics of an ideal spectroscopic source. Whereas diodes are compact and inexpensive devices, sometimes they do not readily exhibit these required attributes for high-resolution spectroscopy. The free-running diodes have some undesirable properties because of their short semiconductor cavities; e.g., their frequencies are highly sensitive to changes in temperature and injection current, and they have poor tunability. Thus it becomes necessary to improve the performance of diode lasers before they can be used in any atomic spectroscopy experiment to produce reliable and significant data. Frequency stabilization methods have been developed based on various technologies such as optical feedback,[2,3] external cavities,[4,5] injection locking,[6,7] and electronic feedback.[8,9] Hybrid systems that use more than one of the methods mentioned above are also possible.[10] Some other methods use spatial mode interference[11] and the Zeeman effect.[12,13] However, the most commonly used method to achieve substantial linewidth reduction and frequency stabilization is to operate the laser in a long external cavity that can provide frequency-selective optical feedback.[1] The number of photons in the cavity of a typical diode laser (100-mW output power; 0.1-mm cavity length operating at near infrared wavelengths) is of the order of 10^5, whereas for a typical gas laser this number is of the order of 10^7. Because of the low number of photons inside the cavity, diode lasers are more susceptible to the feedback mechanism than are normal lasers. A particularly simple design uses the feedback from a diffraction grating mounted in the Littrow configuration.[14] Difficulties in such design are related to grating alignment and the need to adjust the distance between the collimating lens and the laser within a few micrometers. Also, diodes must be antireflection coated on the output facet, a highly expensive procedure, to ensure stable operation in the presence of the strong feedback from the grating.[14]

In this paper we present a method for constructing an extended-cavity diode laser that uses a flat mirror rather than a diffraction grating to provide the optical feedback. The advantage of this method lies in its simplicity of design and in the narrow linewidth of

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Vacuum Rabi splitting for multilevel electromagnetically induced transparency system

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Abstract. We discuss the vacuum Rabi splitting (VRS) from multilevel atoms under electromagnetically induced transparency condition within the framework of linear absorption-dispersion theory. Sharp resonance features superimposed on usually occurring VRS doublet to three-peaked structure spectra are obtained here which can be engineered to have absorptive, dispersive, or dip like profiles according to the choice of system parameters such as radiative damping constants, atomic detunings and driving field strengths etc.

PACS. 42.50.Pq Cavity quantum electrodynamics; micromasers – 42.50.Ar Photon statistics and coherence theory

1 Introduction

Vacuum Rabi splitting (VRS) is considered as one of the important landmarks to achieve non-perturbative regime of cavity quantum-electrodynamics (QED) [1]. VRS has been studied from microwave regime to optical regime and from multi-atomic system to a single atom system. Experiments of cavity QED in non-perturbative or strong coupling regime for single atom were reported in the microwave domain using Rydberg atoms [2–7]. In one of the pioneering work of cavity QED, Haroche et al. [2] experimentally demonstrated self-induced Rabi oscillations in a collection of N Rydberg atoms inside a resonant millimeter-wave cavity. In this experiment the atoms exchanged energy back and forth at a rate $2\wp\varepsilon_0\sqrt{N}/\hbar$ (where $\wp$: dipole matrix element of atomic transition, ε_0: electric field per photon) in the microwave cavity. With the strong coupling regime it is understood that the atom-field coupling constant (g_0) is much larger than the radiative decay rate of atom (γ_a) as well as the rate of cavity damping of field (γ_{cav}). For single atom this implies $g_0 \gg \gamma_a, \gamma_{cav}$ and for N atoms it means that $g_0\sqrt{N} \gg \gamma_a, \gamma_{cav}$. With the developments in cavity QED experiments, the strong coupling regime was obtained in optical domain also. In the optical domain, observations of the VRS for $N \gg 1$ atoms were reported in the experiments by Raizen et al. [8] and Zhu et al. [9]. The number of atoms were brought down to the level of one atom on the average in cesium atomic beam experiment [10] for observing VRS in the optical domain. In another experiment, using rubidium Rydberg atoms in a superconducting niobium microwave cavity, VRS corresponding to $N = 3$ atoms had been observed [6]. The physical origin of VRS can well be explained using the dressed state picture [11]. The name 'vacuum Rabi splitting' suggests a phenomenon associated with quantum mechanics but it has been explained using the classical theory of linear absorption-dispersion [9]. In other words, the VRS is just a normal-mode splitting of coupled harmonic oscillator in which one of the oscillator is single mode field of the cavity and another oscillator is atomic dipole. Very recently, the related effect of Rabi oscillations has got a profound importance in the semiconductor systems of quantum wells, quantum dots, and micro-cavities etc. [12–14].

Discovery of the phenomenon of electromagnetically induced transparency (EIT) in the last decade has given a great deal of impact in the areas of quantum and nonlinear optics by providing control for the speed of light in terms of group velocity modification, lasing without inversion, potential to fabricate efficient electro-optic devices etc. [15]. The EIT is exhibited as a result of quantum interference and the induced atomic coherence in multilevel atomic system is responsible for the modification of linear absorption and dispersion of the atomic-medium [16,17]. The EIT systems also exhibit enhanced non-linear processes, e.g., harmonic generation [18], four-wave mixing [19], two-photon absorption [20], all-optical switching [21], optical multistability [22], slow light [23], and electromagnetically induced focusing [24].

Here we report the VRS spectrum within the frame work of classical model of linear absorption-dispersion for

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Hysteresis loop with controllable shape and direction in an optical ring cavity

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We have experimentally observed a "backward" (clockwise rotating) hysteresis cycle in the system of an optical ring cavity containing three-level (Λ-type configuration) rubidium atoms. The shape and direction of the observed hysteresis cycles can be easily controlled with experimental parameters. Such an interesting phenomenon is caused by the greatly modified absorption, dispersion, and nonlinear optical properties of the three-level atomic medium, due to the induced atomic coherence.

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The phenomenon of hysteresis is ubiquitous in magnetic, optical, electronic, mechanical, chemical, and biological sciences. A hysteresis cycle typically goes counterclockwise (noted as "forward") as experimentally demonstrated in many systems, such as ferroic materials [1], superconductors [2], spin glasses [3], semiconductors [4], polymers [5], porous media [6], granular systems [7], organic radicals [8], nanosystems [9], elastoplastic systems, and shape memory alloys [10]. The mechanisms for having such forward hysteresis cycles are different for different systems. For example, in ferromagnetic materials, the moving out of domain walls separating regions of antiparallel spins needs the increase of magnetic field, so the hysteresis (magnetization as a function of applied magnetic field) thus observed is a function of the work required to displace the domain walls, which ensures forward hysteresis cycles in such systems. In the system of two-level atoms contained in an optical resonator, the bistable curves in the input-output intensities are either due to the saturated absorption [absorptive optical bistability (OB)] or dispersion (dispersive OB) [4,11]. The appearance of the absorptive OB arises from the saturated absorption of the medium and the feedback effect due to the optical cavity, while the dependence of refractive index nonlinearly on the input intensity of the field is responsible for the dispersive OB. Simple arguments show that such OB curves should have forward hysteresis cycles, as observed previously [4,11].

Here, we report our experimental demonstration of a backward (clockwise) hysteresis cycle in a system of three-level (Λ-type) atoms, as shown in Fig. 1(a), inside an optical cavity. Using experimental parameters, such as coupling field frequency detuning, optical cavity detuning, and atomic number density, the shape and transition from a forward to backward hysteresis cycle can be well controlled. The controllability is provided by the unique abilities to manipulate the absorption, dispersion, and nonlinearity in such three-level atomic systems displaying electromagnetically induced transparency (EIT) owing to induced atomic coherence and quantum interference [12,13] among the atomic states, as well as the use of the optical cavity to provide the feedback mechanism [4,11]. Our study may act as a catalyst in search of such interesting behaviors in other systems exhibiting hysteresis (as listed in Refs. [1–10]), where appropriate physical mechanisms may be uncovered.

In optical science the hysteresis cycle is normally associated with optical bistable systems. Optical bistablities were studied in the late sixties [14]. The absorptive OB was predicted [15] in 1969 but any OB was experimentally observed in 1976 in sodium vapor [16]. Since then OBs are extensively studied in two-level atomic systems both theoretically and experimentally [4,11,17]. The underlying physical

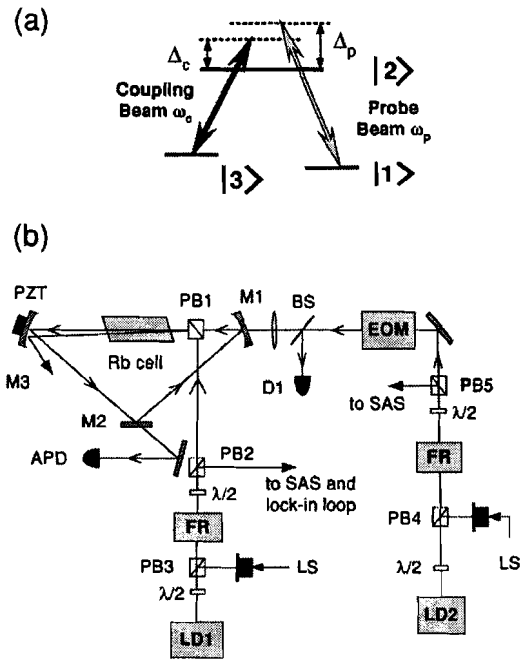

FIG. 1. (a) Diagram of the three-level atomic system. (b) The experimental arrangement used for studying optical bistability and optical multistability in ^{87}Rb atomic vapor: M1-M3 are mirrors of optical ring cavity; PZT is the piezoelectric transducer; LD1 and LD2 are coupling and probe lasers, respectively; PB1-PB5, polarizing cubic beam splitters; BS, beam splitter; EOM, electro-optic modulator; λ/2, half-wave plates; FR, Faraday rotator; D1, detector; APD, avalanche photodiode detector; LS, locking signal from reference Fabry–Perot cavity; and SAS is a saturation atomic spectroscopy set up.

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Effect of spontaneously generated coherence on the dynamics of multi-level atomic systems

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Abstract

The effect of spontaneously generated coherence on dynamical evolution of a multi-level atomic system consisting of three upper levels and a lower level is studied. For degenerate upper levels the population trapping in these levels increases in the steady state.

PACS: 42.50.Lc; 32.50.+d; 32.80.-t

Keywords: Quantum interference; Spontaneously generated coherence

The phenomena of atomic coherence and quantum interference in many quantum optical systems have been discussed extensively during the past decade. These phenomena are instrumental in giving rise to some interesting observations in quantum optics such as lasing without inversion [1], electromagnetically induced transparency [2], refractive index enhancement [3], absorption cancellation, population inversion without emission, modification of spontaneous emission process [4–6], and threshold reduction in optical bistability [7] etc., just to name a few.

When two excited levels decay spontaneously, they can strongly affect each other and give rise to interference, which modifies the spontaneous emission process. The effect of spontaneous-emission-induced interference on the population dynamics was studied in a Λ-type three-level atomic system by Javanainen [5]. On the other hand, the effect of such quantum interference on the V-type three-level atomic system was investigated by Zhu et al. [6], in the studies of population dynamics and spontaneous emission spectrum. They predicted that the population in the upper levels does not decay exponentially due to such interference. Also, this kind of interference can give rise to spectral narrowing and dark line in the spectrum. In this Letter we consider a four-level atomic system, as shown in Fig. 1, such that three upper levels are coupled by the same vacuum modes. We study the population dynamics in this system affected by the spontaneously generated coherence (SGC) among the decaying transitions. We also generalize our result to the cases with more than three upper levels under the condition that all the upper levels are degenerate.

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Controlling dynamic instability of three-level atoms inside an optical ring cavity

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Dynamic instability in the transmission field of an optical ring cavity containing three-level Λ-type rubidium atoms is studied in detail both experimentally and theoretically. The onset and periodicity of such dynamic oscillations in the cavity field can be controlled by the experimental parameters, such as intensity and frequency detuning of the coupling field and/or cavity field. Such nonlinear dynamic behavior is caused by competition between optical saturation of the cavity field and optical population pumping by the coupling field in the three-level atomic system.

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I. INTRODUCTION

Dynamic instability was observed and carefully studied in the upper branch of the optical bistability (OB) curve in a system with two-level sodium atoms inside an optical cavity [1]. This dynamic effect was attributed to the mechanism known as Ikeda instability [2], which appears in analyzing the stability of the steady states in OB curves, assuming a medium response time much faster than the cavity round trip time. In this limit and under certain parametric conditions, a sequence of period doubling bifurcation leading to a region of apparently aperiodic dynamic oscillation was observed. Multimode instabilities as well as oscillatory instabilities leading to optical turbulence in the OB from a two-level atomic system were also reported [3]. The off-resonant-mode instability in mixed absorptive-dispersive optical bistability was studied and it was shown that a portion of the lower transmission branch could also be unstable in addition to the upper-branch instability found in the system with pure absorptive bistability [4]. The observation of instability due to the onset of the cavity side mode was reported in a bistable optical system with a homogeneously broadened two-level medium [5]. Other kinds of self-oscillation and instability were also observed using different two-level atomic systems inside an optical resonator [6]. All the above mentioned theoretical modelings and experimental observations were carried out in two-level atomic systems. A different kind of dynamic instability was observed in the transmission field of an optical cavity consisting of a cold cloud of cesium atoms [7]. In this system the degenerate Zeeman sublevels of the $6S_{1/2}$ $F=4$ and $6P_{3/2}$ $F'=5$ states interact with two cavity fields pumped by one input circularly polarized laser beam. The instability was considered to be caused by competition between optical pumping to the state $6S_{1/2}$ $F=4$, $m_F=4$ from all other Zeeman sublevels and optical saturation of the transition from the state $6S_{1/2}$ $F=4$, $m_F=4$ to the state $6P_{3/2}$ $F'=5$, $m_{F'}=5$. The observed oscillatory behavior in the cavity output field was modeled as a quasi-two-level system interacting with one cavity field and only qualitative comparison was made between the observed phenomenon and a simplified theoretical model [7]. The complexities in that system are mainly caused by the degenerate Zeeman sublevels involved and the trapping and the repumping laser beams for creating the cold atomic cloud. The major limitation of that experiment was the use of only one cavity input (circularly polarized) beam to provide both linearly polarized optical pumping and probing (or saturation) beams, which prevented the two competing physical processes from being independently adjusted to systematically study the dynamic instability.

Recently, we have experimentally observed similar dynamic instability in a system consisting of three-level atoms inside an optical cavity, as shown in Fig. 1, and demonstrated dependence of such dynamic behaviors on the intensity of

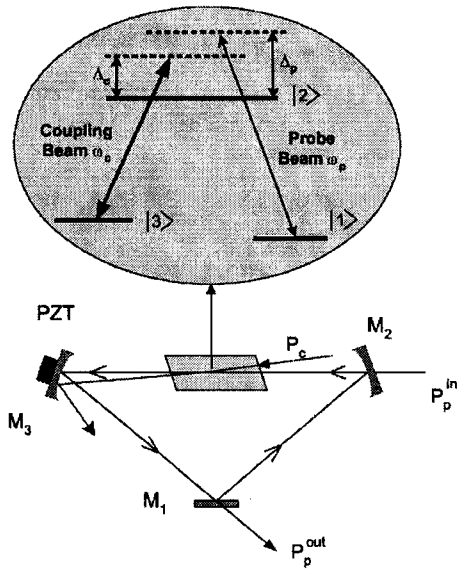

FIG. 1. Sketch of a three-level Λ-type atomic system and the optical ring cavity. M_1, M_2, and M_3 are mirrors; PZT is a piezoelectric transducer; $\Delta_c=\omega_c-\omega_{23}$, and $\Delta_p=\omega_p-\omega_{21}$ are the coupling and probe frequency detunings, respectively; P_c is the coupling laser beam; P_p^{in},P_p^{out} are the input and output probe laser beams, respectively.

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Optical limiting and enhanced optical nonlinearity in boron-doped carbon nanotubes

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Abstract

The optical limiting characteristics of pure and boron-doped multi-walled carbon nanotubes (MWNTs) are studied and compared at wavelengths of 532 and 1064 nm. Fluence-dependent transmission and Z-scan measurements using 8 ns pulses show that the boron-doped MWNT suspensions exhibit a stronger optical nonlinearity with fluence than pure carbon nanotube suspensions in both the visible and near-infrared wavelength ranges. The nonlinear response for both materials is dependent on reaggregation within the suspension under study.

Optical limiting has been an active field of research for the last decade due to growing needs in eye protection from intense laser exposure, as well as for protective applications in optical sensors [1]. While carbon nanotubes are known primarily for their extraordinary electronic and mechanical properties [2,3], researchers have observed strong and broad-band optical limiting in single-walled and multi-walled nanotube suspensions [4,5], as well as in solid nanocomposites [6,7]. There are some important review articles which have made a full coverage on the nonlinear optical properties of (doped) carbon nanotubes [8,9]. Such nonlinear optical transmission in carbon nanotubes has been shown to be strongly dependent on the host media, width and wavelength of light pulses. A general fluence limiting model gaining wide acceptance is as follows. The incoming pulse is quite long at several nanoseconds so the leading edge of the pulse beginning to heat the nanotube rapidly. At the lowest fluences thermal transfer to the solvent leads to the creation of 'mirco-bubbles.' However, as the fluence begins to rise, desorption of oxygen, amorphous carbon and other species attached to the nanotube surface begins to dominate thermal energy transfer. Finally, at the highest fluences, ablation of the nanotube itself begins to take place forming the so-called 'micro-plasma' [10,11]. Scattering becomes nonlinear since the higher light intensity creates more 'bubbles' (used in the general context), which cause the transmission to go down. We note that view this will have specific consequences, namely pulse width dependence of the limiting phenomena and pulse repetition rate dependences of the limiting phenomena, both of which are observed. While a simple version of this mechanism has been used to explain optical limiting in carbon black suspensions (the fluence regimes are not as well defined as in the nanotube case because the material ablates more easily), the effect is thought to be enhanced in carbon nanotubes because of their large aspect ratios that allow them to behave as effective antennae.

Nonlinear transmission characteristics have also been observed in nanotubes embedded in solids (nanocomposites) or soluble samples [6,7] and the effects are similar to or even stronger than suspension samples. Nonlinear absorption was proposed to explain the strong optical limiting effects in such system, though the effects of heat transfer to the matrix were not broadly addressed in that work [6].

An instructive approach to understanding the various fluence dependent mechanisms involved in optical

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Applied Physics B
Lasers and Optics

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Optical bistability in a three-level semiconductor quantum-well system

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ABSTRACT Optical bistable behavior in a unidirectional ring cavity (or a Fabry–Pérot cavity) containing a semiconductor quantum well, described as a three-level ladder-type system with similar transition energies, has been studied. The system interacts with a strong driving field which is in two-photon resonance with the intersubband transition and thus simultaneously drives all three levels into phase-locked quantum coherence. The threshold for switching to upper branch of the bistable curve is found to be reduced due to the presence of quantum interference. Such system can be used for making efficient and fast all-optical switching devices.

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1 Introduction

There is a great deal of interest in the phenomena of optical bistability (OB) and optical switching (OS) in view of their potential applications for optoelectronic devices [1–3]. This warrants exploring nonlinear interactions of lasers with different kinds of systems such as atomic vapors [1, 3] and semiconductor structures that include quantum wells (QWs) [4–9], so that one can develop practical optoelectronic devices which can have fast switching times and at the same time, be compact in size. Research efforts to study OB and OS with structured semiconductors started since long. These materials contain excitons and biexcitons having similarities with the model of a two-level atomic system. Such two-level atomic models can be used to describe excitonic nonlinear properties when excitons have sharp absorption peaks distinguishable from the continuum states [5, 8]. The interest to study OB and OS in QWs or related condensed-matter systems is due to the giant optical nonlinearity ($|\chi^{(3)}| > 10^{-2}$ esu) of excitonic origin [5], short relaxation times, low switching energies, and short switching times between OB branches [8].

The OB has been extensively studied in thick semiconductor Fabry–Pérot (FP) resonators [4, 5, 7–9] and its origin is attributed to large dispersion in the semiconductor materials. The refractive index of the semiconductor in the FP resonator varies with field intensity and shifts the energy of the optical mode sustained in the resonator, and hence provides a positive feedback. The photons are weakly coupled to the excitons and the proper frequency detuning of the field relative to the transition frequency of the medium is responsible for observing OB. Another mechanism for OB in semiconductor FP resonator is attributed to bleaching of the exciton Rabi splitting in the strongly coupled microcavity [10] which originates from cavity polaritons or mixed exciton–photon states. Intersubband OB induced by resonant tunneling in an asymmetric double QW has been studied theoretically by Stockman et al. [7] using density matrix formalism and hysteresis curves were obtained for both the charge transfer probability as well as the optical absorption as a function of light intensity. In another theoretical work of Batista et al. [7], strongly driven far infrared transitions in n-doped GaAs/AlGaAs QWs are studied and it has been shown that the intersubband transition can undergo Hopf bifurcation and period doubling bifurcation under appropriate parametric conditions. Attempts were made to establish analogies between certain important phenomena of atomic three-level systems and their semiconductor counterparts [11–13]. Some of these important phenomena of interest in semiconductor QWs are electromagnetically induced transparency (EIT) [11], enhanced nonlinear processes, lasing without inversion (LWI) [12], and optical Stark effect [13]. Note that observing coherent phenomena in these systems is restricted because of the large dephasing rates in semiconductors ($\sim 10\,\mathrm{ps}^{-1}$). Important advantages of using QWs are that their transition energies, dipole moments and symmetries can be engineered as desired and the dipole moments of intersubband transitions are large. Some Fano interference schemes have been used to demonstrate the viability of quantum interference and EIT in QW systems. EIT was recently demonstrated in a n-doped InGaAs QW (with an AlInAs barrier) which can be described as a three-level ladder-type system [11] as shown in Fig. 1a. A strong driving field, which is in two-photon resonance with the system and simultaneously drives all three states into coherence, results in a 'locking' of quantum phases. This system gives rise to an enhanced transparency feature in the absorption spectrum. The details of the experiment and simulation of EIT for such system in semiconductor QW are available in [11] and the aim of our current study is to look for OB in such system under the similar parametric conditions. Our motivation is to show, with a fairly

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Environmental Effects on Photoluminescence of Highly Luminescent CdSe and CdSe/ZnS Core/Shell Nanocrystals in Polymer Thin Films

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We report our systematic studies of the environmental effects on the photoluminescence (PL) from colloidal CdSe nanocrystals (NCs) embedded in polymer thin films. The highly luminescent bare-core CdSe NCs were significantly more robust than the core/shell CdSe/ZnS NCs against photoannealing under inert environments, while the core/shell NCs are more resistant to photooxidation. For the case of bare-core NCs, we show the importance of the initial photoactivation as the proper treatment for subsequent studies, a step that is basically irrelevant in the case of core/shell NCs. By measuring the PL wavelength shift, PL line width, and PL intensity, we investigated the dynamic changes of the emission properties of these NCs under different environments, including argon, oxygen, air, water vapor, and wet oxygen, and under different excitation conductions to reveal the photoinduced nature of the interactions between the nanocrystal surface and the environment. Mechanisms related to photoactivation, photooxidation, and PL enhancement are briefly discussed.

I. Introduction

Colloidal semiconductor nanocrystals (NCs) have attracted a lot of interest recently in the fields of physics, chemistry, biology, and engineering. The ability to control the optical properties of these NCs (through their size and shape) has made this field of material science very interesting for fundamental understanding and technological applications. Most importantly, such engineered NCs with controlled optical properties will impact applications that employ the emission properties of these NCs such as biological labeling,[1] optical and optoelectronic devices (such as optical switches, solar cells, LEDs, and lasers),[2−10] and gas sensing.[11] Incorporating these NC structures in such devices and applications requires the understanding of their PL behaviors under different environmental and photoexcitation conditions.

The surface of colloidal semiconductor NCs and the immediate surroundings (environment) are very important in determining their PL properties.[12−15] Previous investigations performed on NC systems dispersed in solutions have shown evidence for the important roles of reagent adsorption on the surface of the colloidal NCs and on their emission properties in the photoelectron-transfer processes.[16−18] Many applications require the NCs to be dispersed on thin films. Investigations of the effects of the adsorption of different gaseous analytes (on the surface of the colloidal NCs) on the emission properties of colloidal NCs in polymer thin films are also of great importance and have attracted some interest recently.[19−23]

These reports provide a good deal of scientific insight into the effect of different gaseous environments on the PL properties of different systems of CdSe NCs dispersed onto thin films. These investigations include studying the effect of water molecules and other gases on the PL from CdSe NC monolayers,[19] studying the influence of the surrounding atmosphere on the PL and blinking behavior and the effect of photooxidation and photobleaching of CdSe and CdSe/ZnS NCs embedded in polymer thin films,[20−22] and the study of the photooxidation of monolayers of CdSe NCs by time-resolved PL spectroscopy.[23] However, these studies did not provide decisive conclusions on some of the important issues. For example, it is not clear whether the highly luminescent CdSe bare-core NCs are good candidates for fabricating LEDs, lasers, and other optoelectronic devices, in comparison to the commonly used CdSe/CdS and CdSe/ZnS core/shell NCs.[8,12,13,24,25] Also, we recently reported that CdSe bare-core NCs embedded in polymer thin films showed significant photoactivation, brighter and sharper PL upon above-band gap photoradiation.[11] In this report, we will show that omitting photoactivation could be the actual reason behind the apparent discrepancies in the literature dealing with environmental effects of the PL of semiconductor NCs.[11,19−23]

In general, the dynamic properties of the PL signals have not been thoroughly and systematically investigated previously, and such detailed studies are necessary for research in colloidal NCs, at least for the purposes of comparison between different NC systems and for potential applications.

In this work, we report our systematic studies of the PL properties, such as wavelength shift, PL line width, and PL intensity, from colloidal CdSe NCs embedded in acoustic and electronic highly insulating polymer thin films. We use two kinds of NCs, i.e., bare-core CdSe NCs and CdSe/ZnS core/shell NCs, and study their PL properties under different atmospheric environments, such as vacuum, air, Ar, O_2, and H_2O, and under different photoirradiation conditions. We first characterize the photoactivation stage,[11] which is needed to precede the PL-based investigations for these colloidal bare NCs. We demonstrate the photostability of the NCs under inert

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Piezoelectric effect in elongated (In,Ga)As islands on GaAs(100)

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The piezoelectric (PZ) effect is demonstrated for the elongated three-dimensional (In,Ga)As islands grown on a GaAs (100) substrate. The photoluminescence (PL) spectrum is studied as a function of excitation intensity. With increasing excitation intensity, a blue shift and a linewidth reduction of the PL peak from the (In,Ga)As islands are observed. The observed phenomena are attributed to the screening of the internal strain-induced PZ field in the (In,Ga)As islands.

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Since Smith first predicted a large piezoelectric (PZ) effect in strained III-V or II-VI semiconductor heterostructures,[1] investigations have focused on the study of the PZ effect in semiconductors.[2–8] One reason for this interest is that the strain-induced PZ field can modify the band structure of semiconductors, and therefore, offers another design parameter for fabricating novel optical and electronic devices. For pseudomorphic heterolayers, the PZ polarization is determined by the symmetry of the lattice of the substrate and the orientation of the surface.[2] For example, for zincblende III-V semiconductor materials, the PZ polarization is along the growth direction for pseudomorphic layers grown on the (111)-oriented substrate,[1,2] while it vanishes for those grown on the (100)-oriented substrate.[3] For other non-(100) substrate orientations, the PZ polarization and PZ field usually can have both vertical (growth direction) and lateral (perpendicular to growth direction) components.[2] This is probably the reason why the overwhelming majority of previous investigations of the PZ effect in semiconductors were performed on structures grown on the (111)-oriented surface[4–6] and on high index substrates in general.[7,9,10] However, there have been some recent efforts in realizing the PZ effect on the (100)-oriented substrate for applications. For example, one approach to generate the PZ effect on the (100)-oriented substrate is to pattern the as-grown structure along the $[0\bar{1}1]$ direction[8] while another is to apply surface acoustic waves.[11] In this paper, we discuss a different approach based on the fabrication of three-dimensional (3D) structures, such as, quantum dots and quantum wires.

Quantum dot and wire structures have raised considerable interests because of their unique characteristics for optoelectronic and electronic device applications. Self-organized 3D islands based on the Stranski-Krastanov (SK) growth mode has proved to be a successful approach to fabricate quantum dot and wire structures. The shape of these SK islands can be controlled by changing the growth conditions[12] making it possible to directly engineer quantum dot and quantum wire structures. For example, long and uniform (In,Ga)As/GaAs quantum wires based on elongated islands have been fabricated using a superlattice (SL) growth scheme plus an annealing process.[13] Recently, the PZ effect has been observed in self-organized quantum dots grown on different high index surfaces.[9,10] However, these same studies also demonstrate that self-organized quantum dot structures grown on the GaAs (100) substrate do not show the PZ effect.[9,10] In this work, we demonstrate that the PZ effect is in fact observed in elongated 3D (In,Ga)As islands grown on a GaAs (100) substrate. The evidence for this observation is based on characterization of a sample with elongated islands by excitation intensity dependent photoluminescence (PL) spectroscopy. Specifically, a blue shift and a linewidth reduction of the PL peak from the (In,Ga)As islands are observed to take place with increasing optical excitation intensity. We attribute both the shift and the linewidth reduction to the screening of the internal PZ field in the elongated islands. As a comparison, a similar but more symmetrical shaped quantum dot structure was investigated in the same way and a band filling effect rather than the PZ effect was observed.

A typical sample was grown on a semi-insulating GaAs (100) substrate by molecular beam epitaxy (MBE). After the sample was introduced into the MBE growth chamber, the native oxide was desorped at 580°C followed by overheating the substrate to 600°C. After keeping the substrate at 600°C in As_4 atmosphere for 10 min, the sample was cooled down to 580°C and a 150-nm-thick GaAs buffer layer was deposited. The substrate was then cooled down to 540°C and a 15-period (In,Ga)As/GaAs superlattice was grown. Each time, immediately after the deposition of the (In,Ga)As layer, three monolayers of GaAs was grown without interruption to suppress In segregation and 20.3-nm GaAs was then added with growth interruption of 10 sec. Finally, a top (In,Ga)As layer was grown in order to characterize the surface morphology. The whole growth process was monitored by *in situ* reflection high-energy electron diffraction (RHEED). The RHEED pattern became spotty after the growth of (In,Ga)As layer indicating the appearance of 3D islands. The structural characterization was performed *ex situ* by x-ray diffraction (XRD) and atomic force microscopy (AFM).

Figure 1(a) shows the ω-2θ scan of a double crystal XRD around the GaAs (400) reflection measured with an open detector. Eight satellites appear within the measurement range indicating the good structural quality of the sample. The SL period of the sample was determined to be 23.7 nm from the spacing between the satellites. Figure 1(b) shows the simulation by the XRD dynamic theory assuming a coherent growth of the (In,Ga)As on GaAs. Good agreement between the experimental and the simulated curves is observed. The obtained thicknesses of (In,Ga)As and GaAs layers are 2.5 and 21.2 nm, respectively, and the In composition 0.28, which is in agreement with the values from RHEED

Modified spontaneous emission of CdTe quantum dots inside a photonic crystal

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The angular dependence of the spontaneous emission of CdTe quantum dots (QDs) inside a photonic crystal with a pseudogap is reported. The sensitive dependences of the radiative lifetime and the photoluminescence spectrum of CdTe QDs on the observation angle demonstrate the effect of the photonic bandgap on the spontaneous emission of the QDs. © 2003 Optical Society of America

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Photonic crystals (PCs) are periodic structures that strongly modify the dispersion relation of light. There has been great interest in controlling the atomic and molecular spontaneous emission (SpE) rate with three-dimensional (3D) PCs.[1-6] Previous experiments found significant modifications of the photoluminescence (PL) spectra of atoms and molecules in 3D PCs,[3] but the influence of the photonic bandgap (PBG) on the SpE lifetime is still in question. An early experiment with dye molecules in dilute colloidal crystals showed a variation of the SpE lifetime of 1.75 times.[4] However, it was argued that this remarkable variation was caused mainly by electronic and chemical interactions between dye molecules and the medium, instead of a pure PBG effect.[5] Another experiment with colloidal crystals doped with dye molecules found that the PBG effect on the SpE lifetime was very small (~2%).[6] A PC with a pseudogap exhibits an angularly dependent optical spectrum, so one can resolve this controversy by measuring the angle dependence of the SpE lifetime of light emitters in the PC.

Colloidal II–VI quantum dots (QDs) exhibit some unique optical and electrical properties. The SpE of a QD depends not only on the dot's properties (i.e., size, shape, and surface structure, etc.) but also on the nature of its surrounding environment, more specifically, on the photonic mode density at its location.[7] Compared with those of dyes, the PL peaks of QDs can be controlled by choice of the dot size to match the PBG, and the PL width of QDs is narrower, which makes QDs ideal as probe light emitters in PCs. The modification of the PL spectrum of CdS QDs in a PC was recently reported.[8] In this Letter we report the modification of the SpE lifetime of CdTe QDs in a PC film with a pseudogap. The PC thin film used in our experiment is a fcc crystal of air spheres in silica, which was grown along the [111] direction on a silica plate, as described in detail in Ref. 9. Although there are some defects and cracks, the macroporous film has long-range ordering in an area of 5 mm × 5 mm, which makes it possible to measure the SpE lifetime of light emitters in the film with angular dependence. The observed angular dependence of the SpE lifetime confirms the PBG effect on the SpE lifetime.

With the pseudo stop gap of our PC film around 530 nm, the diameter of the chosen colloidal CdTe dots is ~3 nm, so that their PL peak (~525 nm) is located within the gap. The size distribution of the dots is ~5%, and the PL quantum yield is ~30%. To incorporate the CdTe dots into the PC, we immersed the air-sphere crystal film in a toluene solution of dots for 1 h to promote the adsorption of CdTe dots at the SiO_2–air interfaces of the voids. The concentration of the solution was low ($\sim 10^{-7}$ mol/l) to make stacking of dots more unlikely. Then the sample was rinsed repeatedly in toluene to remove the dots adsorbed near the external film surface, and it was dried and sealed in a transparent container with ambient Ar for optical measurements. With a pulsed laser beam (wavelength of 400 nm, ~1-ps duration, 0.05 μJ per pulse, and 16.4-MHz repetition rate) as the excitation source, PL spectra were recorded by a liquid-nitrogen-cooled CCD mounted behind the monochromator. The radiative lifetime was measured by a time-correlated photon-counting system with a time resolution down to 400 ps. All measurements were carried out at room temperature.

Figure 1 shows the transmission spectrum of the PC film at normal incidence and the PL spectrum of CdTe QDs in solution. The PC film exhibits a clear stop band with the center position at 530 nm. The FWHM of the stop band is ~60 nm. The narrow FWHM (~10% of the center frequency of the bandgap) reveals that the macroporous silica film is of high crystalline quality.[10,11] Because of the finite thickness (~5 μm) of the PC film, the stop band is not complete. The low transmission of the passband is caused mainly by the absorption of silica, the reflections of the film and the substrate surfaces, the scattering of the macroporous surface, etc. After incorporation with CdTe QDs, the transmission spectrum of the PC film does not exhibit variation, indicating that the density of CdTe dots is very low in the voids of the PC film. The PL of the CdTe QDs peaks around 525 nm, with a FWHM

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Hidden resonant excitation of photoluminescence in bilayer arrays of InAs/GaAs quantum dots

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Photoluminescence (PL) of self-organized quantum dots (QDs) in bilayer InAs/GaAs structures is studied with a fixed seed layer and spacer, but variable second-layer coverage. Careful line shape analysis reveals modulation in the high-energy tail of the seed-layer PL spectrum. The oscillation-like behavior is reproducible with variations in both the temperature and optical excitation energy. These oscillations are attributed to carrier relaxation through inelastic phonon scattering from the wetting layer to the QD excited states. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1606109]

Carrier relaxation, including tunneling through potential barriers, represents one of the most fundamental processes described by quantum kinetics. Recently, tunneling between layers of semiconductor quantum dots (QDs) has attracted attention in view of the development of the growth capability to realize vertically aligned or stacked QD geometries.[1–4] It is also of interest because a good understanding of the different excitation and decay channels of excited states in multilayer QD structures will have important consequences for their potential application as emitter or detector arrays.

For stacked QD structures, the coupling strength between QD layers is controlled by systematic variation of the barrier thickness between layers and/or control over the dot size or composition from one layer to the next. For example, if the barrier is thick enough, the electronic levels in the layers of dots will have carrier recombination in each layer separately. However, if the barrier is reduced the carrier wave function in each dot in one layer can spread beyond the barrier into an adjacent dot in another layer, and as such coupling is established between the levels of QDs in bilayer structures. In this case carriers can be photogenerated in a given well and tunneling to an adjacent vertically aligned dot can occur if the tunneling time is less than the recombination time. In similar manner, by varying the QD size or composition, the energy separation between the two tunneling QD layers can be used to control the tunneling probability.

While due attention has been directed toward the coupling between closely stacked QD layers,[5–9] the role of the corresponding wetting layer (WL) in such vertically aligned structures has been relatively less explored. In order to distinguish different contributions to the relaxation of photoexcited carriers we consider a structure composed of two layers of weakly coupled InAs/GaAs QDs. This geometry allows us to discriminate between the coupling QDs in adjacent layers and the coupling between QDs in the seed layer and the WL associated with QDs of the adjacent or second layer.

The samples were grown using a solid-source molecular beam epitaxy chamber coupled to an ultrahigh vacuum scanning tunneling microscope (STM). The structures consist of two InAs layers. Each sample was grown on a GaAs (001) substrate, followed by a 0.5 μm GaAs buffer layer and 10 min annealing at 580 °C to provide a nearly defect-free atomically flat surface. The seed QD layer is then grown by depositing 1.8 monolayer (ML) of InAs at a growth rate of 0.1 ML/s, As_4 partial pressure of 8×10^{-6} Torr, and substrate temperature of 500 °C. This is followed by 50 ML GaAs deposited on top of the seed QD layer. The second QD layer was then added. The InAs deposition coverage in the second layer was varied from 1.8 to 2.7 ML for different samples used in our measurements. Each sample for optical study was finally capped with a 150 ML GaAs layer. The samples were structurally characterized by plane-view STM and cross-sectional transmission electron microscopy (XTEM). The photoluminescence (PL) was studied in temperature range of 10–300 K using the 514.5 nm line of an Ar^+ laser for GaAs excitation, a Ti–sapphire laser for WL excitation, as well as a HeNe laser for intermediate energy excitation range, thus spanning excitation densities from 0.1 to 20 W/cm^2.

Figure 1(a) shows a typical XTEM image of the sample with 1.8 and 2.4 ML depositions for the seed layer and the second layer, respectively. Statistical analyses of XTEM images show the resulting sample to be a weakly vertically correlated (~50% for 50 ML GaAs spacer thickness) double-layer QD structure, designed with a significantly different average QD size in the seed layer compared to that in the second layer. The QDs in the second layer are nearly twice the size of those in the seed layer due to additional deposition, as well as to the influence of the strain field from the seed layer.[3,10] The STM statistical analysis [Fig. 1(b)] indicates a QD size distribution of 4 ± 1.5 nm for the height, 20 ± 3 nm for the width, and a dot density of about 4.5×10^{10} cm^{-2} in the seed layer. The dot density in the second layer is variable over the range of $2.5–4\times10^{10}$ cm^{-2},[10] depending on the InAs coverage.

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High-efficiency blue-light generation by frequency doubling of picosecond pulses in a thick $KNbO_3$ crystal

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We report an experimental demonstration of highly efficient single-pass second-harmonic generation from 859 nm to 429.5 nm with picosecond pulses in a thick $KNbO_3$ crystal. Both the conversion-efficiency and quantum-noise properties of the generated blue pulses are measured at various pump intensities under a strong focusing condition. We find that the variation of the conversion efficiency of the picosecond second-harmonic generation is an oscillatory function of the input pump intensity (with a maximum efficiency of 56.5%) and is sensitive to the position of the input beam focus in the crystal. The quantum noise on the blue beam can be reduced below the shot-noise limit by 20% at low input power.

OCIS codes: 190.2620, 270.6570.

1. INTRODUCTION

Second-harmonic generation (SHG) is an attractive optical frequency conversion process to generate coherent light at shorter wavelengths.[1] Very high conversion efficiency has been reported under extremely high pump intensities,[2–4] i.e., an efficiency of 92% from 1064 nm to 532 nm in bulk nonlinear crystals with a typical pump intensity of a few GW/cm^2.[2] Recently, single-pass traveling-wave (TW) SHG with cw mode-locked pulses has attracted attention because of low average input power with high peak intensity, and conversion efficiency higher than 60% has been obtained both in bulk nonlinear crystals[5–8] (NLCs) and in quasi-phase-matched nonlinear waveguides.[9,10] Highly efficient TW SHG at a low average input power is an interesting nonlinear optical system that not only conveniently provides a coherent light source at short wavelengths, but also allows generation and observation of amplitude-squeezed light that has less-intensity noise than the standard shot-noise limit.[7,11] In fact, 6.7% (0.3-dB) amplitude squeezing in SHG was observed with a conversion efficiency of 15% in the TW SHG experiment with a type II phase-matched bulk KTP crystal.[11] In an experiment with a $LiNbO_3$ waveguide,[9] 16.8% (0.8-dB) squeezing in the transmitted fundamental field and 7.7% (0.35 dB) squeezing in the generated harmonic light were observed with a conversion efficiency approaching 60%. In the TW SHG with a thick potassium niobate ($KNbO_3$) crystal pumped by femtosecond pulses, a slope efficiency of 300% nJ^{-1} for harmonic conversion was achieved[5] and a 20% (1.0-dB) squeezing in the generated harmonic light was observed with a conversion efficiency of 60% at ~90 mW average input power.[7,12]

In this paper, we study highly efficient single-pass SHG in a thick, noncritically phase-matched $KNbO_3$ crystal pumped by picosecond pulses and measure the quantum-noise properties of the generated blue pulses. We show that the TW SHG process in $KNbO_3$ with picosecond pulses is quite different from that with femtosecond pulses. Much less peak intensity is necessary to obtain the high conversion efficiency for picosecond pulses under the strong focusing condition. We found that the variation of the conversion efficiency of the picosecond SHG is an oscillatory function of the input pump intensity (with a maximum efficiency of 56.5%) and is sensitive to the position of the input beam focus in the crystal. The quantum noise on the blue beam can be reduced below the shot-noise limit by 20% at low input power.

2. BACKGROUND

$KNbO_3$ is an attractive nonlinear crystal that has a large nonlinear coefficient and noncritical phase matching (no spatial walk-off) under appropriate conditions for low-power optical applications.[13–15] However, this crystal also has a large group-velocity mismatch (α = 1.2 ps/mm for the wavelength conversion from 860 nm to 430 nm) and a narrow phase-matching bandwidth of $\Delta\nu = 0.88/\alpha L$ (~0.2 nm for a crystal length of L = 10 mm),[6] which generally limit it from short-pulse, wideband applications. In the previous TW SHG experiments with femtosecond pulses,[5–8] the input spectral width (~10 nm for a Gaussian pulse with duration time t_p = 120 fs) is much broader than the phase-matching bandwidth. Thus the temporal walk-off length $l_T = t_p/\alpha$ (~100 μm), over which a fundamental pulse and a harmonic pulse miss temporal overlap from each other completely, is much

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Controlled steady-state switching in optical bistability

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Optical switching has been achieved between two steady states of optical bistability generated in a system with three-level atoms inside an optical cavity. The optical power switching is controlled by adding short positive (switching up) or negative (switching down) pulses to the input intensity. The coupling laser beam in the three-level atomic system is used to control the threshold value and the width of the hysteresis cycle, which can adjust and optimize the optical switching process. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1600833]

Optical bistability in atomic systems has been extensively studied both theoretically and experimentally since the early 1980s.[1] The quantitative comparisons between the detailed experimental results and theoretical calculations with various experimental parameters provided good understanding of the underlying physical mechanisms for optical bistability in two-level atomic systems.[2] Such bistable behavior between the input and output intensities of an optical resonator with two-level atoms is the result of nonlinearity of the intracavity atomic medium, such as saturation and Kerr nonlinear effects, and feedback of the optical intracavity field from the cavity mirrors.[1] Driving the interest in such bistable behavior was the possibility of using it for applications in optical switching, optical transistors, and optical storage elements, which are essential for optical computing and communications. Optical bistability has also been observed in semiconductor and other materials.[3,4] Optical switching in optically bistable multiple-quantum-well semiconductors and other systems has been demonstrated,[4,5] which made use of the bistable transmission behavior of thin film structures.

In recent years, optical bistability in systems with three-level atoms inside optical cavities has been theoretically studied[6] and experimentally demonstrated.[7,8] The major advantages of using three-level, instead of two-level, atoms as the nonlinear medium inside an optical cavity can be threefold. The first one is to make use of the atomic coherence induced in the three-level atomic systems, which can greatly modify the absorption, dispersion, and nonlinearity of the system,[9,10] thereby reducing the cavity input threshold power for optical bistability.[6] The second advantage is the controllability added by the coupling laser beam for such three-level atomic systems, as shown in the bubble of Fig. 1. By changing the intensity or the frequency detuning of the coupling beam, one can control the threshold value and the hysteresis cycle width of the bistable curve.[11] The last one is the simpler experimental setup with a three-level atomic system compared to a two-level atomic system. Due to severe Doppler effect, atomic beams or cold atomic samples were needed to observe optical bistability in two-level atomic systems inside an optical cavity.[2] However, by using the two-photon Doppler-free configuration in the three-level atomic system,[9] e.g., the two laser beams copropagate in the Λ-type three-level atomic medium, the first-order Doppler effect is eliminated. Therefore, one can easily observe optical bistability in an atomic vapor cell[7,11] without the need for a complicated vacuum system for atomic beams.

These advantages provide us with a unique opportunity to make optical switching between the two steady states of the bistable curve by adding positive and negative pulses to the cavity input intensity. In this letter, we describe such experimental demonstration carried out in a rubidium atomic vapor cell inside an optical ring cavity. Although optical switching was previously demonstrated in the three-level atomic system,[12] the actual switching action happened outside the bistable region, and the switching was controlled by the coupling laser beam, instead of by the cavity input beam, as will be shown here. The physical mechanisms for these two types of optical switching are very different.

The atomic medium used for the experiment is ^{87}Rb in the three-level Λ-type configuration, as shown in the bubble of Fig. 1. Level $|1\rangle$ $(F=1, 5^2S_{1/2})$ and level $|2\rangle$ $(F'$

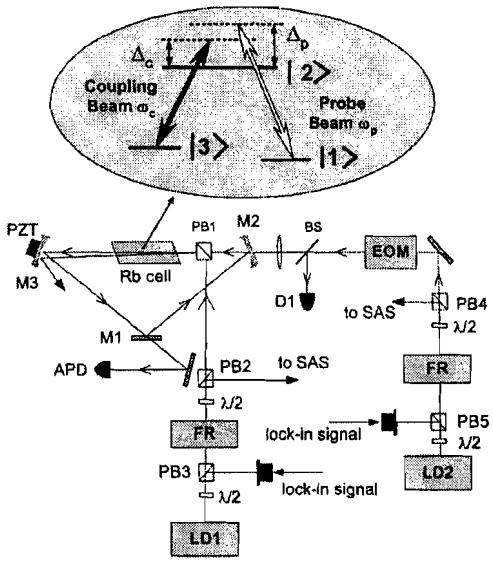

FIG. 1. The experimental setup, M1–M3 are mirrors of optical ring cavity; LD1 and LD2 are coupling and probe lasers, respectively; PB1–PB5 are polarizing cubic beam splitters; λ/2, half-wave plates; FR, Faraday rotators; D1, detector; APD, avalanche photodiode detector; SAS represents the saturation atomic absorption setup. Inside the bubble is the three-level Λ-type atomic system in the D_1 lines of ^{87}Rb.

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Effect of spontaneously generated coherence on optical bistability in three-level Λ-type atomic system

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Abstract

Optical bistability is studied for a three-level atomic system in Λ-configuration contained in an optical ring cavity and the effects of spontaneously generated coherence in the presence of two arbitrary coherent fields are investigated. Condition for observing optical multistable behavior in this system has also been specified.

PACS: 42.65.Pc; 42.65.Sf; 42.50.Gy

In recent years some new types of coherence produced due to decays of closely lying states have been discussed [1–5]. One of the main phenomena associated with these coherences is the modification of the line shapes of the spectral lines. In Λ-type three-level atomic system the spontaneously generated coherence (SGC) due to interaction with the vacuum bath of the radiation field has been discussed by Javanainen [4]. The important finding in the study of Ref. [4] is the disappearance of the dark state due to SGC. The SGC is very sensitive to the alignment of dipole moments of two transitions with respect to each other. The effects of SGC on electromagnetically induced transparency (EIT) [6] and coherent population trapping [7] phenomena were also examined [8]. Suppression of spontaneous emission [2] and subnatural linewidths [3] due to SGC were predicted in the three-level atomic systems.

Optical bistability (OB) was extensively studied initially in the two-level system comprising of alkali atomic beams inside an optical resonator where a single mode beam circulating [9,10]. The experimental demonstration of OB in this system gave a great thrust to many potential applications such as optical transistors, memory elements, and all optical switches. This led to further investigation of OB in three-level atomic system inside an optical cavity both theoretically [11,12] as well as experimentally [13]. Multistable (multiple hysteresis) behaviors has also been observed in Fabry–Perot cavities filled with atoms having several degenerate or nearly degenerate sublevels in the ground state and driven by linearly polarized light [14]. Most of these experiments used magnetic fields and high pressure buffer gases, and relied on Zeeman coherence as an efficient mechanism for the observed behavior. More specifically, optical trista-

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Photo-oxidation-enhanced coupling in densely packed CdSe quantum-dot films

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Photo-oxidation of densely packed monolayer of CdSe quantum dots (QDs) was studied by time-resolved photoluminescence (PL) spectroscopy. Electrons yielded in QDs by the strong laser-pulse irradiation can assist the oxidation of CdSe QDs. Such rapid photo-oxidation does not introduce more nonradiative defects, instead, it enhances the coupling between QDs through surface modification and the PL intensity can be significantly improved. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1590735]

In recent years, colloidal quantum dots (QDs) have generated much interest for both fundamental research and technical applications due to their relatively reproducible and controllable synthetic chemistry and strong size-dependent optical properties.[1,2] An important feature of these QDs is the relatively large surface-to-volume ratio.[3] The complex surfaces[4,5] of the colloidal semiconductor nanocrystals usually are chemically and optically unstable.[6] In ambient environment, water molecules adsorbed on CdSe QDs lead to native oxidation of their surfaces.[7] The slow oxidation process yields some nonradiative defects and decreases the fluorescent properties.[7] Another important process closely related to the surface of QDs is the aging due to photoirradiation.[7,8] To resolve the origin of the photo-oxidation process of colloidal QDs is a challenging task and will have big impacts on the stability of QD devices as well as on describing the surface structures of these colloidal nanostructures.

Colloidal QDs dispersed in solid films present more opportunities for potential device applications than those in solvents.[9] When QDs are densely packed in solid films, the coupling of electronic excitations between proximal QDs could play an important role in the electronic and optical properties.[9,10] In this letter, we report our experimental study of photo-oxidation of densely packed monolayer of CdSe QDs by time-resolved photoluminescence (PL) spectroscopy. We found that strong laser-pulse irradiation can induce rapid surface photo-oxidation, which does not increase more nonradiative defects, instead, it can enhance the coupling between QDs and significantly increase the PL intensity. These results indicate that the optical properties and the stability of colloidal QDs can be improved by proper photoirradiation, which are useful for optoelectronic device applications.

The method of synthesizing the high quantum-yield (QY) colloidal CdSe QDs used in this work was described in detail in Ref. 11. The CdSe QDs used here have an average diameter of 4.5 nm with a size distribution of about 5% and a QY of about 85%. CdSe QDs' monolayer films were deposited onto fused silica substrates by a similar method described in Refs. 9 and 10. The inset of Fig. 1 shows the transmission electron microscopy (TEM) image of a typical part of a sample film. CdSe QDs can self-assemble into a densely packed array due to their nondispersive size distribution. The samples were placed inside a vacuum chamber with optical windows for optical measurements. The irradiation laser pulses, obtained by frequency doubling a mode-locked Ti–Sapphire picosecond laser, were at $\lambda = 420$ nm with a 16.4 MHz repetition rate and 6.3 pJ per pulse. The laser intensity on the sample is ~ 0.5 kW/cm^2. PL and time-resolved PL were obtained by a liquid-nitrogen-cooled charge coupled device camera and a time-correlated photon-counting system, respectively. All measurements were carried out at room temperature.

The irradiation of the 420 nm laser pulses has no obvious effect on the QDs when the sample is placed in vacuum. However, if the sample film is placed in the ambient air environment, the photoirradiation induces a blueshift in PL peak position and a significant change in PL intensity. The PL spectra are strongly affected by the irradiation time of the samples in air. Figure 1 shows the evolution of PL spectrum of the 4.5 nm QD film for different irradiation time. Figure 2 shows the peak shift and PL intensity of the PL spectrum

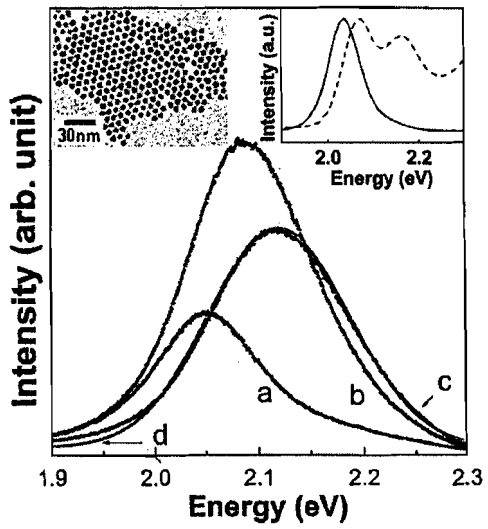

FIG. 1. Evolution of PL spectrum of 4.5 nm CdSe QDs (with 85% QY) under photoirradiation. Curves (a), (b), and (c) are PL spectra with photoirradiation times of 0, 2, and 60 s, respectively. The gray line (d) is the best Guassian fit of curve (c), which indicates that PL spectrum deviates from the symmetric shape. The right inset is for the PL (solid line) and absorption spectra (dashed line) of the initial QDs and the left inset is the typical TEM image of the sample.

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Electromagnetically induced transparency and its dispersion properties in a four-level inverted-Y atomic system

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Abstract

We consider a four-level atomic system in inverted-Y configuration and study the phenomenon of electromagnetically induced transparency (EIT) in this system under various parametric conditions in order to demonstrate controllability of the EIT and its dispersion properties.

PACS: 42.50.Hz; 32.80.Wr; 42.65.Ky

Keywords: Electromagnetically induced transparency; Quantum interference

1. Introduction

In recent past a great deal of attention has been paid to observe and understand the phenomenon of electromagnetically induced transparency (EIT) and related effects in multi-level atomic systems [1–3] interacting with two or more electromagnetic fields. The closely related phenomenon with EIT is coherent population trapping (CPT) [1]. In an usual CPT situation the two electromagnetic fields interacting with the atom are of almost equal strength and the quantum interference (QI) arises from these two fields. On the other hand, in usual EIT situation, one of the fields is much stronger than the other so that the QI is mostly governed by the stronger field. In the terminology of EIT [1–3], the weaker field is called *probe* field and the stronger field is called the *coupling* field. The EIT systems are potentially useful in applications such as electro-optical devices, sharp dispersion to control speed of light, nonlinear optics, and lasing without inversion. The induced atomic coherence in multi-level atomic systems cannot only modify the linear absorption and dispersion properties, but also enhance the nonlinear optical processes such as four-wave mixing [4,5], harmonic generation [6,7], and two-photon absorption [8] and all optical switching [9]. The QI can also modify the refractive index properties of a medium. It is due to the Kramers–Kronig relation, any change in absorption of medium will be accompanied by change in the dispersion of the medium and that leads to phenomena like electromagnetically induced focusing [10] and slow light [11]. By making use of atomic coherence, the nonlinear optical properties of an atomic medium can be modified and controlled. Recently, enhanced third-

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Effect of quantum interference on optical bistability in the three-level V-type atomic system

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The phenomenon of optical bistability is studied for the three-level atomic system in V-configuration confined in a unidirectional optical ring cavity, and the effects of quantum interference and coupling field are investigated. The possibility of obtaining optical multistability in the system by controlling quantum interference and coupling field strength is also discussed.

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The phenomenon of quantum interference is central to many new effects recently discovered in quantum optics. Harris *et al.* [1] considered the V-type three-level atom and demonstrated that the absorption rate can become zero due to the destructive interference and thus it is possible to have light field amplification without population inversion [2]. It was also shown that the quantum interference can lead to line narrowing, black dark line, and removal of spectral emission of driving field frequency in the emission spectrum [3]. It can also produce ultranarrow spectral lines in the fluorescence spectra of a three-level atom [4]. Fluorescence quenching has been experimentally observed in the sodium dimers [5]. Many other related phenomena with quantum interference, e.g., the electromagnetically induced transparency (EIT), refractive index enhancement, modification of spectral features of three-level systems featuring dark resonances, etc., were also studied in recent years [6].

Optical bistability (OB) has been extensively studied both experimentally and theoretically in the recent past [7]. Most of the experimental studies in OB have been devoted to two-level alkali atoms confined in an optical resonator [7,8]. The theoretical models of OB have considered the interaction of a collection of two-level atoms with a single-mode field [7,8]. The perpetual interests in OB and associated phenomena stem from the fact that these phenomena could have wide range of applications such as in optical transistors, memory elements, and all optical switches. Also bistable behaviors were studied theoretically [9,10] and observed experimentally [11] in three-level atomic systems inside optical cavities in recent years.

In the literature, the possible realization of optical multistability (OM) has also been mentioned, which involves interaction of a nonlinear medium with two different optical fields. In particular, Kitano [12] reported optical tristability in a three-level Λ-configuration interacting with two different modes of cavity under the limiting condition of large atomic detuning and no saturation. This work was generalized by Savage *et al.* [13] to include saturation in the dispersion limit, and they predicted that the asymmetric state becomes unstable and gives rise to self-oscillations and a different kind of optical turbulence. Later on Arecchi *et al.* [14] included the effect of ground-state coherence and reported not only the tristability but also higher-order bistability. Recently, a theoretical calculation for OB has been carried out for three-level atoms confined in an optical cavity, and the effect of coherence generated in the system by a control field is studied and possibility of generating multistability is also discussed [9].

Here, in this work we discuss the role of quantum interference in the phenomenon of OB in a three-level V-type atomic system confined in an optical resonator. The atomic system consists of two excited sublevels of same parity and a single ground level of different parity. The effect of quantum interference in spontaneous emission from the upper two levels is included in this investigation of OB. Such a model with quantum interference from spontaneous emission shows great enhancement of the population inversion in one of the optical transitions and can lead to substantial radiation amplification and existence of vacuum-induced quasitrapped states [15]. However, the previous theoretical works studying OB in three-level atomic systems did not include quantum interference in the decay channels in their models.

The model of the three-level atom considered here is depicted in Fig. 1. It is a closed V-type configuration with one single ground-state $|1\rangle$ and two closely lying upper states $|2\rangle$ and $|3\rangle$. The transition between $|1\rangle$ and $|2\rangle$ (with resonant frequency ω_{12}) is mediated by the probe laser field E_P (frequency ω_1), while the transition $|1\rangle$ to $|3\rangle$ (with resonant frequency ω_{13}) is driven by another laser field E_C (frequency ω_2) called coupling field in this work. The atomic dynamics of the system can be described by the Liouville equation for the density operator and the density matrix equations [15] with all decay terms included under rotating-wave approximation are

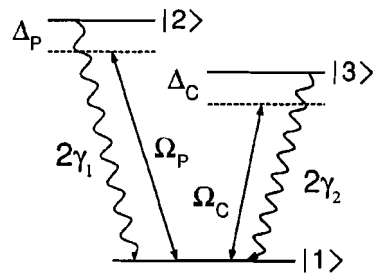

FIG. 1. Schematic diagram of a three-level V-type atom.

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Novel Linear and Nonlinear Optical Properties of Electromagnetically Induced Transparency Systems

Min Xiao

Invited Paper

Abstract—**We describe some interesting linear and nonlinear optical properties of three-level electromagnetically induced transparency (EIT) systems, such as absorption reduction, sharp dispersion change, and enhanced Kerr nonlinearity. These novel optical properties are very useful in enhancing efficient nonlinear optical processes, which can find applications in optoelectronic devices. We present some experiments done in our group in the past few years with three-level atomic systems, especially more recent experiments with EIT medium inside an optical cavity.**

Index Terms—**All-optical switching, atomic coherence, Kerr nonlinearity, nonlinear optics, slow light.**

I. Introduction

ELECTROMAGNETICALLY induced transparency (EIT) and its applications in multilevel atomic systems have been active research fields for the past decade [1], [2]. Since the first experimental realizations of EIT phenomena in strontium [3] and lead [4] hot atomic vapors with high-power pulsed lasers, significant advances were made in demonstrating the reduction of absorption and enhancement of dispersion near the EIT resonance. One such advance was to use low-power continuous-wave (CW) diode lasers as both the coupling and probe lasers. By making use of the two-photon Doppler-free configurations for the coupling and the probe beams in three-level atomic systems, significant absorption reduction was demonstrated in both three-level ladder-type [5] and Λ-type [6] inhomogeneously broadened atomic systems inside an atomic vapor cell at room temperature. Simple theoretical calculations were carried out for such ideal EIT systems and detailed quantitative comparisons were made with the experimentally measured results [5], [6]. The use of Doppler-free configuration allowed the direct observation of quantum interference effect in such EIT system with a very weak coupling intensity [7], which would have been buried by the Autler–Townes or ac-Stark effect under strong coupling intensity. Dispersion enhancement at the EIT resonance was then demonstrated in the three-level EIT system and a reduction in the group velocity of light by a factor of thirteen was inferred from the sharp dispersion slope [8], which had been pushed down to 90 m/s in atomic vapor with buffer gas by a better selection of atomic sublevels and use of diode lasers with locked frequency and narrower linewidths [9], [10]. Another advancement in demonstrating EIT was the use of cold atoms as the EIT medium [11]. With cold atoms, the Doppler effect was eliminated and the group velocity reduced to a mere 17 m/s [12]. With such cold atomic system, which has a longer coherence time, storage of photons via atomic coherence effect in collective atomic medium was experimentally demonstrated [13]. Such manipulations of linear absorption and dispersion properties in the coherent EIT media have profound implications in making ideal optical materials, especially in solid media [14].

With reduced absorption at atomic resonance, EIT media can also be used to enhance nonlinear optical processes near resonance, such as harmonic frequency generation [15], [16], nondegenerate four-wave mixing [17]–[19], and optical bistability and dynamic instability [20]. By making use of the greatly enhanced nonlinearity in multilevel atomic media, absorptive optical switching in a four-level N-type atomic system [21] and controllable all-optical switching in a three-level Λ-type atomic system inside an optical cavity [22] were experimentally demonstrated recently. Making more efficient nonlinear optical processes will have potential applications in optoelectronic and photonic devices.

In this paper, we describe a few experimental studies done in our group in three-level atomic systems in the past few years. Section II shows the linear absorption and dispersion properties in three-level inhomogeneously broadened atomic systems with weak CW diode lasers in the two-photon Doppler-free configurations. We also describe the linewidth narrowing of an optical cavity due to such highly dispersive intracavity medium. Section III is devoted to the nonlinear optical properties in such EIT systems including enhancement of nondegenerate four-wave mixing in three-level Λ-type and four-level double Λ-type systems, measurements of enhanced Kerr nonlinear index of refraction, and optical bistability and dynamic instability with EIT medium inside an optical cavity. In Section IV, we present an experimental demonstration of an efficient

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Optical Multistability in Three-Level Atoms inside an Optical Ring Cavity

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Optical multistability in an optical ring cavity filled with a collection of three-level Λ-type rubidium atoms has been experimentally demonstrated. The observed multistability is very sensitive to the induced atomic coherence in the system and can evolve from a normal bistable behavior with the change of the coupling field as well as the atomic number density. The underlying mechanism for the formation of such multistability is also discussed.

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Atomic optical bistability (AOB) in two-level atomic systems has been extensively studied both experimentally and theoretically in the past 20 years [1–5]. Such optical bistable behavior arises from the nonlinear interaction between a collection of two-level atoms and the field mode inside an optical cavity. Because of the Doppler effect, atomic beams were used for observing such AOB in these two-level atomic systems [2,3,5]. Many interesting phenomena in these AOB systems such as dynamic instabilities, squeezed states, cavity QED, and higher-order quantum correlation were all experimentally observed [6]. Also, bistable behaviors were predicted [7] and observed [8] in three-level atomic systems in recent years. Typically, those observed AOB phenomena were divided into two distinct categories, i.e., absorptive and dispersive bistability which have quite different characteristics [1–5].

Optical multistability (OM) was predicted and observed in systems involving interactions between nonlinear media and two different optical cavity field modes [9–12]. In particular, Kitano *et al.* [9] calculated optical tristability in a three-level Λ configuration under the limiting condition of large atomic detuning with no saturation, which was experimentally observed by Cecchi *et al.* [10]. This work was generalized by Savage *et al.* to include saturation in the dispersive limit [11]. Later, Arecchi *et al.* included the effect of ground-state coherence and reported not only tristability but also higher-order bistability [12]. The tristability generated here [12] involved a two-photon process via the cooperation of a bistability generation process. Several groups reported the observations of multistable/multiple hysteresis behaviors in a Fabry-Perot cavity filled with atoms having several degenerate or nearly degenerate sublevels in the ground state and driven by linearly polarized light [13–15]. In the transmitted light polarization switching occurs and three different states of polarization can dominate, giving rise to a transition from tristability to symmetry breaking bifurcation attributed to a change in cavity length or atomic density in the experiment of Giacobino [13]. The intricate multistable behavior, as observed by Nalik *et al.* [14], was a combined effect of Zeeman pumping and electronic excitation. Most of these experiments used magnetic fields and buffer gases, and relied on the Zeeman coherence as an efficient mechanism for the observed AOB/OM.

In this Letter, we report the experimental observation of a new kind of OM in a three-level Λ-type atomic system in a Rb vapor cell inside an optical ring cavity under different conditions. A simple physical model is given for understanding the appearance of such OM in this interesting system of electromagnetically induced transparency (EIT) [16,17] inside an optical cavity, in which AOB and dynamic instability were observed previously [8]. By varying the frequency detuning of the coupling laser beam or the atomic number density, transition from normal optical bistable behavior to the multistable one is achieved and controlled. Our ability of observing such OM stems from the enhanced Kerr nonlinearity due to atomic coherence in such a composite system [18] and the controllability of the coupling laser beam. Another advantage of our experimental setup is the use of an atomic vapor cell inside the optical cavity instead of atomic beams or a cold atomic sample, so we can increase the atomic density to a high value, which is essential for observing such OM. Because of the two-photon Doppler-free configuration used in our experiment, i.e., copropagation of the cavity field and the coupling field for the EIT transition, the first-order Doppler effect is eliminated [17]. With controllability of the intensity and frequency detunings of the coupling and cavity fields, as well as the atomic density, we can manipulate the absorption, dispersion, and nonlinear optical properties of the atomic medium inside the optical cavity, which provides us a unique opportunity for studying the OM in this system.

The three-level Λ-type atomic system and the experimental setup used in the experiment are similar as in Ref. [8]. The lower two levels of the atomic system designated as $|1\rangle$ and $|3\rangle$ correspond to the $F = 1$ and $F = 2$ states of $5S_{1/2}$ in ^{87}Rb, respectively, and the upper level $|2\rangle$ corresponds to the $F' = 2$ state of $5P_{1/2}$. The probe laser (frequency ω_p) is tuned near the transition frequency of $|1\rangle$ to $|2\rangle$, and the coupling laser (frequency

Photoluminescence upconversion in colloidal CdTe quantum dots

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Efficient photoluminescence (PL) up-conversion has been observed in colloidal CdTe quantum dots with an energy gain of as high as 360 meV. Compared with the normal PL, the peak energy of this up-converted PL (UCPL) shows a redshift of about 80 meV, and the corresponding radiative lifetime becomes nearly twice as long. This UCPL is attributed to the carrier recombination involving surface states mainly through a thermal excitation process.

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Photoluminescence (PL) up-conversion, the observation of luminescence with energies higher than those of the excitation photons, has been widely observed in the bulk[1] and heterostructures[2–5] of semiconductors, quantum wells,[6] porous silicon,[7] self-assembled quantum dots (QDs),[8] and colloidal QDs.[9,10] It was proposed that the existence of intermediate states with energies resonant with or lower than those of the excitation photons is a prerequisite for these up-converted photoluminescence (UCPL) processes. Carriers must be excited to these intermediate states by the first excitation photon and further to the higher energy states through various underlying mechanisms such as Auger excitation,[7] two-step two-photon absorption (TS TPA),[1–3,5,6] or the thermal effect.[9,10] Most of these intermediate states are "observable,"[2–7] i.e., in addition to the UCPL emission, one can also see the normal PL emission from these states with energies lower than those of the excitation photons. However, in some cases of self-assembled and colloidal QDs, these intermediate states can hardly be observed since carriers cannot efficiently populate them by the above-band-gap excitation of normal PL.[8] In a way, UCPL can provide a unique method for detecting the existence of these "defect" states in semiconductor quantum nanostructures and investigating their properties, which are usually different from those of the band-gap states.

Semiconductor colloidal QDs are characterized by a large surface-to-volume ratio due to their small particle sizes, as well as the surface complexity originating from the passivation ligands used to neutralize the surface dangling bonds. Surface states have long been invoked to be responsible for some complex phenomena that could not be explained by using the concept of band-gap states only. For example, the almost universal occurrence of a biexponential distribution in the radiative lifetime strongly implies the existence of surface states in colloidal QDs.[11,12] In this sense, time-resolved measurements can provide one, but only *indirect*, way to probe the surface states in colloidal QDs, and consequently the existence of them is still controversial.[13] However, if one can observe a UCPL signal from colloidal QDs, this problem will be easily resolved since the demand of intermediate states can only be fulfilled by the existence of surface states. Recently, UCPL signals were observed in colloidal InP,[9] CdSe,[9,10] and CdTe (Ref. 10) QDs, which have provided a strong proof of the existence of surface states in colloidal QDs and opened up a new opportunity for the systematic study of these surface states.

Here, we report on our optical studies of efficient UCPL in colloidal CdTe QDs with an energy gain of as high as 360 meV. Compared with the normal PL, the UCPL signal shows a redshift of about 80 meV in its peak energy, while the excitation spectroscopy of the UCPL (UCPLE) measurement at this peak energy showed a broad excitation band with energies below those of the band gap. Time-resolved measurements were utilized to study the time-resolved dynamics of this UCPL, whose radiative lifetime now becomes nearly twice as long as that of the normal PL. Finally this UCPL is attributed to the recombination of carriers from the band-gap states and surface states mainly through a thermal excitation process.

The CdTe QD samples were synthesized[14] in a non-coordinating solvent 1-octadecene (ODE) with controllable sizes. A mixture of CdO, tetradecylphosphonic, and technological-grade ODE was heated to 300 °C to get a clear solution. A solution of tellurium dissolved in tributylphosphine and ODE was quickly injected into this hot solution, and then the reaction mixture was allowed to cool to 240 °C for the growth of CdTe QDs. The unconverted cadmium and tellurium precursors and ODE were carefully separated out before the optical measurements. Typically, the size distribution was between 5% and 8% determined by using the transmission electron microscopy. Here we selected the sample with the diameter of 4.3 nm, which has a normal PL peak around 660 nm, for our optical measurements. The solution containing the QDs in chloroform ($CHCl_3$) solvent was sealed in an ultraviolet-visible (UV-Vis) cuvette initially under nitrogen gas environment to prevent photo-oxidation for the PL, absorption, UCPL, UCPLE, and time-resolved measurements at room temperature. The absorption spectrum, as shown in the inset of Fig. 1, gives the optical densities at different wavelengths, from which we estimate the QD density in this sample to be $\sim 2.7\times 10^{-6}$ M^{-1}. A Ti:sapphire pulsed laser (~1-ps duration, 0.05 μJ per pulse, and a 16.4-MHz repetition rate) with the photon energy varying from 1.442 eV (860 nm) to 1.699 eV (730 nm) was used as the excitation source for the UCPL measurement, while its second-harmonic light beam (at 365 nm) was used for the normal PL measurement. The UCPLE spectrum was ob-

InGaAs/GaAs three-dimensionally-ordered array of quantum dots

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We report on the first fabrication of (In,Ga)As/GaAs quantum dots with both vertical and lateral ordering forming a three-dimensional array. An investigation of the photoluminescence spectra from the ordered array of quantum dots, as a function of both temperature and optical excitation intensity, reveals both a lateral and vertical transfer of excitation. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1596712]

Recently, electronic and optoelectronic devices based on quantum wires and quantum dots (QDs) have been the subject of interest due to their potential applications as lasers, detectors, or photonic crystals.[1] As a result, there has been an extensive effort to manipulate and control the position, size, shape, and density of QDs.[2–5]

In this letter, we report on the fabrication of a vertically and laterally ordered (In,Ga)As/GaAs QD stack forming a three-dimensional QD array. The fabrication is achieved by utilizing multilayer vertical stacking grown at elevated temperature.[6–11] An investigation of the photoluminescence (PL) spectra from these ordered arrays of QDs, as a function of both temperature and optical excitation intensity, reveals both a lateral and a vertical transfer of excitation.

The samples used in the experiments were grown on semi-insulating GaAs [001] substrates, with a miscut angle smaller than 0.05°, using solid source molecular beam epitaxy. After the native oxide was desorbed, a 150-nm-thick GaAs buffer layer was grown at 580 °C. The substrate was then cooled down to 540 °C for the growth of the multilayer dot structure. After a 2-nm-thick $In_{0.36}Ga_{0.64}As$ layer was grown, three monolayers of GaAs were deposited without growth interruption to suppress In segregation. Then, after an 8 s interruption, 16 nm of GaAs was grown. The period of the superlattice was 15.

The samples were investigated using plane-view and cross-sectional transmission electron microscopy (XTEM) in order to examine vertical and lateral ordering was performed using a JEOL JEM2000FX microscope operated at 200 kV. PL studies were performed in a variable-temperature helium cryostat (4–300 K), under the excitation of the 514.5 nm line of a continuous-wave argon-ion laser. The PL signal was analyzed using a 0.5 m single-grating spectrometer, and detected using a photomultiplier tube.

Figures 1(a) and 1(b) show dark-field plan-view TEM and dark-field XTEM images of a typical multilayered QD sample, respectively. The XTEM image [Fig. 1(b)] shows an almost perfect vertical island correlation, and the plan-view TEM image [Fig. 1(a)] indicates that within each layer, the QDs were slightly elongated along $[1\bar{1}0]$ direction and densely packed in long lateral chains ($\sim 1-2$ μm in length). The effective 2D wetting layer (2DWL) thickness (about 0.7 nm) is the same for all layers. The average diameter of the QDs is about 45 nm while the average height is about 5 nm. The average distance between QDs within a chain is about 20 nm. The separation between a QD in one chain and the nearest QD in a neighboring lateral chain is about 70–80 nm. The QDs in each chain appear to sit on a common InGaAs base with an estimated height of about 1.5–2 nm. The vertical correlation of the QDs is due to the vertical strain field between the buried and the subsequent QDs, while the lateral ordering of the QDs is related to the strain-field-modulated surface along the $[1\bar{1}0]$ direction and the enhanced adatom migration length at the elevated growth temperature.

According to the XTEM, the tunnel barrier thickness between the vertically aligned QDs (defined as the average distance between the island tip in one layer and the island base in the second InGaAs layer) is about 10 nm and is approximately two times smaller than the barrier thickness between neighbor dots within a lateral chain. Based on calculations using the Wentzel–Kramers–Brillouin approximation and treating the dots as thin quantum wells,[12,13] the electron tunneling time is estimated to be $\tau_t \sim 0.8$ ns for square barriers of about 10 nm wide. In comparison, the typical radiative recombination time τ_r for InGaAs quantum dots is about 0.5–2 ns.[3,14] So, at low temperature, τ_t is comparable

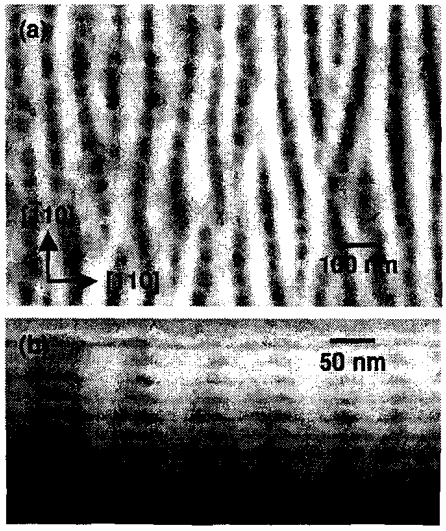

FIG. 1. (a) Dark-field plane-view TEM image of the sample obtained with $g=200$. (b) $[1\bar{1}0]$ XTEM image of the sample.

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Controlling optical bistability in a three-level atomic system

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We have experimentally studied the optical bistable behavior in an optical ring cavity filled with a collection of three-level Λ-type rubidium atoms, interacting with two collinearly propagating laser beams. The bistability so observed is very sensitive to the induced atomic coherence in this electromagnetically induced transparency system or consequently to the altered nonlinearity in the system and, thus, can easily be controlled by changing the intensity and the frequency detuning of the coupling field.

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In the recent past, a great deal of attention has been paid to observing and understanding the phenomenon of optical bistability (OB) in two-level atoms confined in an optical cavity, due to its potential applications in all-optical switches, memories, and optical transistors [1]. The saturation of atomic transition and intensity-dependent refractive index of the media are attributed to two different physical mechanisms for absorptive OB and dispersive OB, respectively [1–3]. The bistable states of the optical cavity output appear only for a definite range of input intensities [4] and the intensity of the upper branch of the bistable curve shows an instability behavior for some parametric conditions [5]. The cold atomic cloud of cesium atoms is yet another system which shows OB as well as instability in which the Zeeman sublevels in the $6S_{1/2}$, $F=4$ to $6S_{3/2}$, $F=5$ transition of cesium atoms interact with a circularly polarized laser beam in an optical cavity. However, there are limitations to this experiment because of the lack of control due to only one laser beam being employed for both optical pumping and saturation, and the optical cavity was not locked in observing OB and instability [6].

Recently, electromagnetically induced transparency (EIT) and related effects in multilevel atomic systems have attracted great attention [7–9]. The induced atomic coherence in multilevel atomic systems can not only modify the linear absorption and dispersion properties, but can also enhance the nonlinear optical processes such as four-wave mixing [10,11], harmonic generation [12,13], and two-photon absorption [14]. More recently, the enhanced third-order Kerr-nonlinear index of refraction (n_2, in the expression of $n=n_0+n_2|E|^2$) was measured near the EIT condition, as well as near the more general coherent population trapping (CPT) conditions in three-level Λ-type rubidium atoms inside an optical cavity [15]. It is interesting to note that at the exact EIT or CPT condition, i.e., when both the coupling and probe frequency detunings are zeros, n_2 is zero. However, if one of the frequency detunings is even slightly tuned to be nonzero, the value of n_2 is greatly enhanced as compared to the situation when there is no coupling beam present (as in a two-level situation). Detailed studies of the variations of n_2 with intensities and frequency detunings of both coupling and probe beams were reported earlier [15], which motivated us to further explore the bistable characters in this EIT system with the help of the enhanced nonlinearity due to atomic coherence.

The present work is built upon our preliminary observation (which mainly studied the instability behavior) [16] and is a further experimental investigation of the OB as functions of various parameters in the composite system consisting of three-level Λ-type Rb atoms and an optical ring cavity. The aim of this work is to experimentally demonstrate the controllability of OB behaviors in this system by varying the intensity and the frequency detuning of the coupling laser beam. These additional experimental controls provided by the coupling laser beam do not exist in the previous systems using two-level atoms for OB [1–5] or in the system with multi-Zeeman-levels but using only one laser beam [6]. Another advantage of our experimental setup is its simplicity with atoms in a vapor cell (no atomic beam or cooled atomic sample under vacuum is needed) in two-photon Doppler-free configuration to overcome the first-order Doppler effect [9]. The enhanced nonlinearity here due to atomic coherence can relate to the effect reported in an earlier experiment with sodium vapor filled in a Fabry-Perot cavity [17], where the two different frequencies for the atoms in a particular group of velocity distribution is provided by counterpropagating beams. Controlling OB can have practical applications in all-optical switches, memories, transistors, and logic circuits, which have no need for optical-electronic-optical conversion of signal information. Due to the enhanced Kerr nonlinearity in the three-level atomic system owing to atomic coherence, the switching thresholds of such devices can reduce down considerably, and hence controlling becomes easier and more efficient, which is also essential in order to process optical signals faster and reliably at very low intensity levels of light. Some interesting theoretical results for OB have been reported for three-level atoms confined in an optical cavity under various conditions [18].

The experiment was carried out in a three-level Λ-type system of ^{87}Rb atoms using the D_1 lines of $5^2S_{1/2}\rightarrow 5^2P_{1/2}$ transitions as depicted in the bubble of Fig. 1. The probe laser beam (i.e., the cavity field with frequency ω_P) is tuned to the atomic transition $|1\rangle$ ($F=1,5^2S_{1/2}$) to $|2\rangle$ ($F'=2$,

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Surface-Related Emission in Highly Luminescent CdSe Quantum Dots

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ABSTRACT

We report our experimental studies of surface-related emission in highly luminescent CdSe quantum dots (QDs) with controlled quantum yield and photooxidation by time-resolved photoluminescence measurements. This kind of surface-related emission, with a radiative lifetime of tens of nanoseconds, implies the involvement of surface states in the carrier recombination process of such highly luminescent CdSe QDs.

Colloidal semiconductor quantum dots (QDs) are a class of nanocrystals intermediate between single molecules and bulk solid-state materials and are of great interest for both fundamental research and technical applications.[1,2] One of the best-studied semiconductor quantum systems is the CdSe quantum nanostructure. Its 3D quantum confinement of both electrons and holes leads to strongly size- and shape-dependent optical and electronic properties,[3−5] which make them ideal candidates for tunable emitters in such applications as biological imaging[6] and single-photon sources.[7] One important concern in these applications is the radiative lifetime, which, for example, determines the average time interval between two consecutive emitting photons from the single-photon source and, consequently, the speed of opto-electronic devices. Compared with the typical ~1-ns radiative lifetime in bulk materials, the radiative lifetime of CdSe QDs usually ranges from several to tens of nanoseconds.[6−10] One interesting feature of the photoluminescence (PL) decay of colloidal QDs is the almost universal occurrence of a biexponential time distribution in the radiative lifetime. Typically, a shorter lifetime is on the time scale of several nanoseconds, and a longer one is tens of nanoseconds. The shorter lifetime can be attributed to the intrinsic recombination of initially populated core states,[8,9,11,12] but the possible origin of the longer lifetime is still in question.

One distinct feature of colloidal QDs is the large surface-to-volume ratio due to the decreasing particle size, and it has long been believed that the surfaces of colloidal QDs play important roles in carrier relaxation and recombination processes.[8,11,13] For example, a femtosecond broad-band transient absorption technique was used to show that electron relaxation was controlled by surface passivation, compared to surface-independent hole relaxation.[8] One direct way to understand the role of the surface in the PL decay dynamics fully is to change the surfaces of QDs systematically while monitoring the corresponding PL decay. Recently, we successfully realized the control of quantum yield (QY) from a few percent to as high as 86% in colloidal CdSe QDs via surface reconstruction.[14] This controllable high QY of QDs using the new synthesis procedure[14] provided us with a rare opportunity to study the correlation between the surface quality and the PL decay dynamics systematically.

In this letter, we investigate the biexponential distribution of the radiative lifetime in colloidal CdSe QDs with changing surface quality realized either by controlling the QY in synthesis or by introducing photooxidation with laser irradiation. The amplitude ratio of the longer (15−25 ns) to the shorter (2−5 ns) lifetime components in the PL decay-time distribution increases with increasing QY, as measured in vacuum. We also observed a decreasing amplitude ratio of the longer to shorter lifetime components when monitoring the PL decay in the process of photooxidation. Using previously established models of photooxidation,[15] it can be concluded that the recombination of electrons and holes on the surface gives rise to this surface-related emission with longer lifetime, which likewise implies the involvement of surface states.

The method of synthesizing CdSe QDs with controlled high QY was described in detail elsewhere.[14] It was found that under a largely biased initial Se/Cd ratio of precursors in solution the PL QY of CdSe QDs during their growth increases to a certain maximum value and then decreases, as shown in the inset of Figure 1. A series of samples with increasing diameters from 3.5 to 5.0 nm and different QYs (>30%) were taken from the solution during the growth process. For comparison, another QD sample with a QY of ~8% was prepared by the common method without using a

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Photoactivated CdSe Nanocrystals as Nanosensors for Gases

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ABSTRACT

The photoluminescence (PL) of high-quality CdSe nanocrystals (NCs) incorporated into polymer thin films was found to respond reversibly and rapidly to environmental changes upon photoradiation above their absorption onset. The PL of the NCs may be dramatically enhanced or diminished depending on the properties of the gases. Photostimulation was found to be necessary for the response, which likely makes the original dense-ligands monolayer on the surface of the NCs permeable to gases by the activation of the vibration modes of the NC lattice through photon–phonon coupling.

Bulk and thin-film semiconductors are plausible materials for gas-sensing technologies.[1] In general, sensing methods based on bulk crystals rely on the interactions between their surface atoms and the environment. This principle implies that nanocrystals (NCs)[2,3] and other nanostructures[4] should be ideal candidates for sensing purposes because of their extremely large surface-atom ratio. However, unlike those nanostructures grown by gas-phase techniques,[5] high-quality colloidal semiconductor NCs have barely been explored as gas-sensing materials. This is so because high-quality colloidal semiconductor NCs are generally coated by a dense monolayer of organic ligands that acts as a substantial barrier for the diffusion of gas molecules between the environment and the surface of the NCs.

We report here that photoexcitation of the electronic energy states of semiconductor NCs can make the ligands monolayer readily permeable to gas molecules. Under photoirradiation, the PL properties of the NCs responded to the environment in a reversible, rapid, and species-specific fashion. Such responses can be readily detected with several tens of NCs incorporated into the polymer thin films. This photostimulated response is likely due to the photon–phonon coupling of the optical absorption and emission processes occurring in the NCs.[6] The results demonstrated in this letter open the door to a new type of sensitive and simple nanosensors that are compatible for integration.

Colloidal semiconductor NCs are nanometer-sized fragments of the corresponding bulk crystals synthesized in solution.[2,3] When a photon is absorbed by a semiconductor NC, an exciton (an electron–hole pair) is generated. If the physical dimensions of the NC are smaller than the intrinsic size of the exciton generated inside the particle, then the photogenerated electron and hole will have a great chance to recombine and emit another photon with relatively low energy. For an efficient emission event, there should be no deep traps for the photogenerated electron and hole. The possible traps in those highly crystalline semiconductor NCs are generally from the surface atoms that are missing at least one chemical bond. For this reason, the surface atoms must be optimally constructed/reconstructed and passivated with some surface ligands. Consequently, any change in the surface environment of NCs may significantly affect their PL properties, which could potentially be exploited for the development of gas-sensing technologies.

Recently, a reliable and simple method was introduced to achieve optimal surface reconstruction for colloidal CdSe NCs.[7] Those NCs isolated at the "PL bright point"[7] in the growth course can possess as high as an 85% PL quantum yield with a narrow emission profile (Figure 1a). Furthermore, the emission properties of those NCs are unusually stable in solution. As with other types of colloidal NCs, the emission properties of these bright CdSe NCs are very sensitive to the nature of the surface ligands. In solution, the PL quantum yield of the CdSe NCs can be irreversibly altered from 85% with the original amine ligands to nearly 0% if the surface ligands are changed to thiols.

All measurements were performed with several tens to several hundreds of NCs (Figure 1b) with a narrow size distribution synthesized by the method reported previously[7] using a standard single molecular spectroscopy setup reported previously.[8] Thin-film samples were prepared by spin casting a dilute solution mixture of the NC solution (in chloroform, $CHCl_3$) with 1% poly(methyl methacrylate) (PMMA) onto

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Dependence of enhanced Kerr nonlinearity on coupling power in a three-level atomic system

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We study the enhanced Kerr-nonlinear coefficient in a three-level Λ-type atomic system for various coupling-beam powers. The Kerr-nonlinear coefficient behaves very differently in the strong and the weak coupling power regions and changes sign when the coupling or probe frequency detuning changes sign. Comparisons of Kerr-nonlinear coefficients as functions of probe frequency detuning, coupling power, and coupling frequency detuning are presented. © 2002 Optical Society of America

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Nonlinear optical processes can be greatly enhanced by atomic coherence in three-level atomic systems. In the past few years several experimental demonstrations of enhanced nonlinear optical processes in multilevel atomic systems were reported.[1–5] These experiments prompted recent theoretical studies of nonlinear optics at the single-photon level in three-level or four-level atoms.[6–8] More recently, the enhanced Kerr-nonlinear coefficient (n_2) of a three-level atomic system was directly measured by use of an optical ring cavity under conditions of electromagnetically induced transparency (EIT); i.e., the coupling intensity is much larger than the probe intensity.[9] These measurements provide a good understanding of the enhanced Kerr nonlinearity near EIT resonance and the possibility of optimizing nonlinear optical processes. However, enhancing the nonlinear optical processes is a property that is not unique to conditions of EIT alone. It also occurs near more-general coherent population trapping conditions for an arbitrary ratio of coupling power to probe power in three-level atomic systems.

In this Letter we present our experimental measurements of enhanced n_2 under general conditions of coherent population trapping when the coupling beam's power is varied from nearly equal to the probe power to much larger than the probe power. At the exact condition of coherent population trapping, i.e., when both the coupling frequency and the probe frequency detunings are zero, n_2 is zero. However, when one of the frequency detunings is tuned slightly off resonance, n_2 can be enhanced as much as 2 orders of magnitude compared with the value of n_2 when there is no coupling beam and can also change sign depending on the frequency detunings of the coupling and the probe beams. n_2 behaves quite differently in the strong- and the weak-coupling-beam limits.

Our experimental setup is the same as the one used for measuring n_2 under EIT conditions.[9] A rubidium vapor cell (5 cm in length and heated to 67 °C) with Brewster windows wrapped in a magnetic-shielding metal sheet is placed in an optical ring cavity (with a three-mirror configuration) with a length of 37 cm. The probe beam enters the ring cavity through one mirror (concave, with $R = 10$ cm and a transmissivity of 3%) and circulates inside the cavity. It exits the cavity through an output coupler with 1% transmission. The coupling beam is introduced through a polarization beam splitter cube inside the optical cavity with an orthogonal polarization to the probe beam. The coupling beam is misaligned from the probe beam at an angle of ~2°, so it does not circulate inside the ring cavity. The radii of the coupling and the probe beams at the center of the atomic cell are estimated to be 700 and 80 μm, respectively. The empty-cavity finesse is ~100 but degrades to ~50 after insertion of the atomic cell (with atoms far from resonance) and the polarization beam splitter cube.

We consider the three-level Λ-type atomic system in ^{87}Rb $D1$ lines. The coupling beam with frequency ω_c is coupled to near the $5S_{1/2}, F = 2$–$5P_{1/2}, F' = 2$ transition of frequency ω_{23}, and the probe beam with frequency ω_p is tuned near the transition from $5S_{1/2}, F = 1$ to $5P_{1/2}, F' = 2$ (frequency, ω_{12}). The coupling and probe frequency detunings are defined as $\Delta_p = \omega_p - \omega_{21}$ and $\Delta_c = \omega_c - \omega_{23}$, respectively, and their values are set by use of saturation absorption spectroscopy. The essential scheme that saves us from experiencing large Doppler broadening at such temperatures in an atomic cell is the use of a two-photon Doppler-free configuration[10,11] setup by propagation of the coupling and probe beams in the same direction. The technique of using an optical cavity to measure a Kerr-nonlinear coefficient has advantages over other (such as Z-scan) methods for atomic systems, because it can work with low laser intensity and long atomic cells. The well-defined cavity mode also prevents spatial changes in the probe beam owing to the nonlinearity in our experiment.

With ω_c and ω_p locked to certain values, we scan the length of the optical ring cavity across resonance by applying a ramp voltage to a piezoelectric transducer mounted upon the third cavity mirror (high reflector). When the coupling beam is blocked, the cavity transmission profile is basically symmetric, as shown in Fig. 1(a), with $\Delta_p = +40$ MHz and an intracavity peak power of 6 μW (corresponding to an intracavity peak Rabi frequency of $\Omega_p = 2\pi \times 11$ MHz). When the coupling beam is turned on with a power of 1.1 mW (corresponding to an average Rabi frequency of $\Omega_c = 2\pi \times 17$ MHz inside atomic cell) and $\Delta_c = 0$,

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Photoluminescence from single CdSe quantum rods

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Abstract

We report here the first observation of photoluminescence from single CdSe quantum confined nanorods (quantum rods). Luminescence imaging spectra of individual rods were obtained for two samples, with average aspect ratio of 2 and 4, respectively. Typical room-temperature luminescence spectral line widths of the rods in both samples are less than 60 meV, which are about the same as that of high-quality CdSe/CdS core/shell single quantum dots. Single quantum rods also exhibit luminescence intermittence as observed in single-dot systems.

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Keywords: Photoluminescence; Quantum nanocrystals; Single quantum rods; Imaging spectra; CdSe; Intermittence

Colloidal semiconductor nanocrystals have received much attention in recent years [1–4]. Their size-dependent optical properties with band edge tunable through visible wavelength range make them of particular interests for fundamental studies as well as potential applications. Single quantum-dot spectroscopy has been used to eliminate the effects of inhomogeneous broadening and ensemble averaging in a number of nano-sized semiconductor systems [5–7] and has revealed many new physical phenomena in single CdSe based nanocrystals, such as ultra-narrow line-shapes, luminescence intermittence, shifting of emission spectra, and a highly polarizable emitting state in the presence of strong local electric fields [8]. The strong polarized emissions have been observed in single CdSe [9] and CdS [10] nanocrystals.

It has been recently demonstrated that the shape of CdSe nanocrystals can be manipulated by controlling the growth kinetics [11]. The resulting particles can be from a nearly spherical morphology to a rod-like one. The rods can grow up to 200 nm long with an aspect ratio of up to 50 [12]. Studies of CdSe quantum dots suggested that electronic structures and optical properties of band edge have strong dependence not only on the size but also on the shape of nanocrystals [13]. The measurements on CdSe ensemble quantum rods have shown that Stokes shift is noticeably larger than that of the corresponding quantum dots, and being even larger with the increase of rod lengths [11]. Also the polarization of the emission of these rods exhibits significant difference in comparison with the CdSe dots [11]. Studies of these rod- and wire-like structures can provide a testing ground

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Controlling light by light with three-level atoms inside an optical cavity

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We present our experimental demonstration of controlling the cavity output intensity of one laser beam with the intensity of another laser beam in a composite system consisting of a collection of three-level Λ-type rubidium atoms and an optical ring cavity. When the intensity of the controlling beam is modulated with a square waveform, the cavity output power switches on and off (with a distinction ratio better than 20:1) between two steady-state values. This all-optical switching effect is the result of combined absorption and enhanced Kerr nonlinearity near resonance in such three-level atomic systems because of atomic coherence and can find applications in optical communication and optical computation. © 2002 Optical Society of America

OCIS codes: 190.1450, 190.3270, 230.1150, 030.1670, 160.4330.

Controlling light with light is essential in all-optical communication and optical computing. Optical bistability in systems containing saturable two-level atoms inside an optical cavity was considered to be an ideal mechanism for achieving controllable optical switching.[1] The optical switching of the cavity output field between the two steady states in the bistable region can be controlled by the input field intensity pattern. However, such optical switching action is not easy to achieve, although other effects such as optical bistability,[1,2] instability,[3] and quantum statistical properties[4,5] in such composite systems (involving two-level atoms and an optical cavity) were all studied many years ago. The important issue is what should be used to control the ON–OFF states of the cavity output power. In the traditional optical bistability systems the only controllable optical beam is the input field, which is part of the bistable curve in the input–output plot. In this case the cavity field intensity is controlled not by another laser beam but by its own input field.

The situation is different when the two-level atomic medium is replaced with a three-level electromagnetically induced transparency system,[6–8] in which the controlling (coupling) beam can be used to control the switching (probe) beam. In this Letter we present our experimental demonstration of a true all-optical switch in a system containing three-level atoms inside an optical ring cavity. Although optical bistability exists in such a system,[9,10] the switching does not occur between the two steady states of a single bistable curve; instead, it occurs between two distinct steady-state curves corresponding to setups with and without the controlling beam. The difference in the two steady-state curves is caused by the combined effects of absorption change and enhancement of Kerr nonlinearity that results from atomic coherence near resonance.[11] This is different from the absorptive photon switching demonstrated earlier in a four-level atomic system.[12]

The three-level Λ-type rubidium atomic system is the same as used in Ref. 10, where $F = 1$ (state $|1\rangle$) and $F = 2$ (state $|3\rangle$) states of $5S_{1/2}$ are the two lower states and $F' = 2$ (state $|2\rangle$) of $5P_{1/2}$ serves as the upper state. The controlling laser beam (with frequency ω_c) couples states $|3\rangle$ and $|2\rangle$, whereas the switching beam (with frequency ω_p) interacts with states $|1\rangle$ and $|2\rangle$. The controlling frequency detuning is defined as $\Delta_c = \omega_c - \omega_{23}$, and the switching frequency detuning from the atomic transition is defined as $\Delta_p = \omega_p - \omega_{12}$, where ω_{23} and ω_{12} are atomic transition frequencies from states $|2\rangle$ and $|3\rangle$ and from states $|1\rangle$ and $|2\rangle$, respectively. The experimental setup is shown in Fig. 1 with an atomic vapor cell containing such three-level atoms placed inside a three-mirror optical ring cavity. Both the controlling (LD1), and the switching (LD2) lasers are single-mode diode lasers that are current and temperature stabilized. The frequencies of these two diode lasers are further stabilized by use of optical feedback through servo-loop-controlled mirrors. Parts (~10%) of the switching and the controlling beams are split by polarizing beam splitters PB4 and PB2 to a saturation absorption spectroscopy (SAS) setup to monitor the frequency detunings of the two lasers. The optical ring cavity is composed of three mirrors. The flat mirror M2 and the concave mirror M1 (R = 10 cm) have ~1% and ~3% transmission, respectively. The third cavity mirror [concave with (R = 10 cm)] is

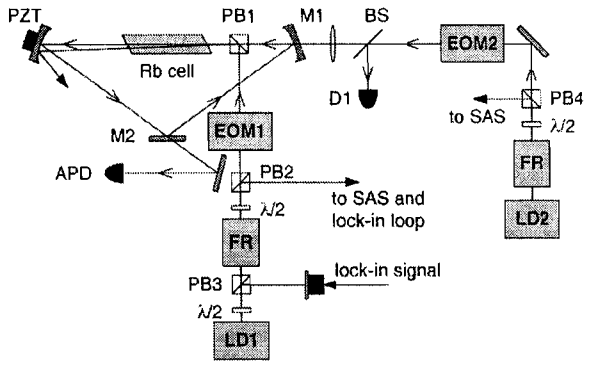

Fig. 1. Experimental setup: FRs, Faraday rotators; APD, avalanche photodiode; BS, beam splitter; D1, diode laser 1; PB1, PB4, polarizing beam splitters. Other abbreviations defined in text.

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Modification of spontaneous emission from CdSe/CdS quantum dots in the presence of a semiconductor interface

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The spontaneous-emission lifetime of CdSe/CdS core–shell quantum dots was studied as a function of the distance between the dots and a polished Si surface. The experimental results reveal a significant modification of the spontaneous-emission rate of the quantum dots by the Si surface. © 2002 Optical Society of America

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The spontaneous-emission characteristics of a particle depend not only on the properties of the particle itself but also on the nature of its surrounding environment, more specifically, on the photonic mode density at its location.[1–6] Spontaneous emission from rare-earth ions, dye molecules, atoms, and semiconductor structures can all be influenced in microcavities or near mirror surfaces.[4–7] For example, Drexhage found that the spontaneous-emission lifetime (τ_{se}) of Eu^{3+} ions near a silver mirror has a damped oscillatory dependence on the separation between the ions and the mirror.[4] Chance *et al.*[5] (CPS) proposed a classic dipole model to explain Drexhage's data and found good agreement. Experiments with InGaAlAs/GaAs quantum dots confined in planar microcavities demonstrated some control of the spontaneous-emission rates of these quantum dots.[7] Control of spontaneous emission from semiconductor structures may be useful in future applications of optoelectronic devices.

Colloidal II–VI semiconductor nanocrystals have been well studied because of their relatively reproducible and controllable synthetic chemistry and their strong size-dependent optical properties.[8–11] However, little is known about their photophysical properties near the surface of a semiconductor, especially near silicon (Si), despite their important applications to a number of diverse fields such as biological chips[10] and solar energy conversion.[11] In this Letter we examine the spontaneous-emission lifetime of CdSe/CdS core–shell nanocrystals as a function of their distances from a polished Si surface. CdSe/CdS dots were selected because of their good surface passivation. Our experimental data indicate a significant modification of the spontaneous-emission rate of the quantum dots by the Si surface.

The fabrication of colloidal CdSe/CdS dots was described in detail in Ref. 8. CdS shells with as many as three monolayers of thickness were grown upon CdSe cores with diameters of ~4.0 nm. Figure 1 shows the typical photoluminscence (PL) spectrum of dots in a toluene solution. Samples for our τ_{se} measurements were prepared by spin coating of a dilute solution of nanodots in toluene with 1% poly(methyl methacrylate) (PMMA) onto SiO_2 films thermally grown upon Si wafers. The average distance between quantum dots within the PMMA film was ~50 nm. The Si wafers were P type, ~30Ω cm^{-1} and (100) oriented. The thickness of the thermal SiO_2 films (d_{SiO_2}) was varied in a range of approximately 20–400 nm. The PMMA films containing CdSe/CdS dots were ~20 nm thick and, thus, the average separation (d) between the dots and the Si surface can be considered to be $d = d_{SiO_2} + 10$ nm. The inset of Fig. 1 shows a schematic diagram of the sample structure.

PL decay measurements were taken at room temperature by use of a time-correlated photon-counting system. The excitation pulses, obtained by frequency doubling of a mode-locked Ti:sapphire picosecond laser, were at $\lambda = 400$ nm, with a 16.4-MHz repetition rate and 6.3 pJ of energy per pulse. Figure 2 shows typical PL decays of CdSe/CdS dots upon a SiO_2/Si substrate [Fig. 2(b)] and directly upon a silica substrate [Fig. 2(a)]. These PL decay curves can be fitted well by a single exponential function in the first 30-ns time delay. It can be seen that there are some minor deviations from a single exponential for the long time delay, which are due to another time-decay component with a larger time constant. Experiments with time

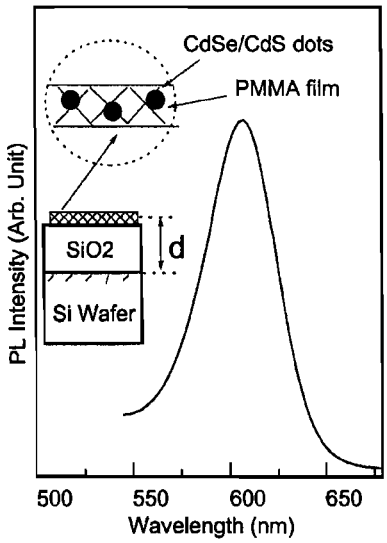

Fig. 1. Photoluminescence spectrum of the quantum dots in a toluene solution. Inset, schematic diagram of the sample structure.

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Photoluminescence study of carrier transfer among vertically aligned double-stacked InAs/GaAs quantum dot layers

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Photoluminescence (PL) properties of self-organized quantum dots (QDs) in a vertically aligned double-layer InAs/GaAs QD structure are studied as a function of temperature from 10 to 290 K. The QDs in a sample with a 1.8 ML InAs seed layer and a second 2.4 ML InAs layer are found to self-organize in pairs of unequal sized QDs with clearly discernible ground-states transition energy. The unusual temperature behavior of the PL for such asymmetrical QD pairs provides clear evidence for carrier transfer from smaller to larger QDs by means of a nonresonant multiphonon-assisted tunneling process in the case of interlayer transfer and through carrier thermal emission and recapture within one layer. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1510157]

The effect of temperature on both energy relaxation and carrier transfer mechanisms in semiconductor quantum dots (QDs) has been a subject of extensive investigations.[1–6] In general, it is reported that, as expected, the QD photoluminescence (PL) intensity decreases with increasing temperature due to carrier escape from the dot.[3] However, these investigations also report, rather unexpectingly, a redshift of the PL peak position and a decreasing PL linewidth with increasing temperature.[1,4–6] This unusual and interesting behavior has been explained by enhanced carrier relaxation between QDs due to several reasons including carrier thermionic emission,[3] carrier transport through the wetting layer (WL),[2] and tunneling mechanisms.[1] The effect of temperature can get even more interesting when the QD array exhibits a size distribution that shows more than one maximum, e.g., a bi- *or* multimodal QD size distribution either within one layer[7–10] or across multiple layers.[11,12] Despite such interesting possibilities and significant potential applications a complete picture of the energy and carrier transfer in such multimodal systems is still not available.[7,10]

In this letter we present a detailed study of the carrier transfer between two InAs QD families with different size distribution but separated from each other by a thin layer of GaAs. The particular QD system under investigation is a vertically aligned double-layer InAs/GaAs QD structure with different sized QDs in the first layer compared to the second layer.

Our samples were fabricated using a solid-source molecular beam epitaxy chamber coupled to an ultrahigh vacuum scanning tunneling microscope (STM). The growth structure consists of two InAs layers containing QDs, which was repeated eight times, in a GaAs matrix. All samples were grown on GaAs (100) substrates, followed by a 0.5 μm GaAs buffer layer and 10 min annealing at 580 °C to provide a nearly defect free atomically flat surface. The first QD layer was then added by depositing 1.8 ML of InAs with a growth rate of 0.1 ML/s, an As_4 partial pressure of 8×10^{-6} Torr, and a substrate temperature of 500 °C. This was followed with 16 nm of GaAs deposited on top of the first QD layer while the growth temperature was changed from 500 to 520 °C. The second QD layer was then added by depositing 2.4 ML of InAs. The resulting samples are vertically correlated double-layer QD structures with different QD sizes in each layer.[13,14] The substrate temperature was then reduced from 520 to 500 °C during a 40 nm GaAs growth, which was used to separate the pair of QD layers from seven additional pairs.[15] As seen by STM, the dot density in the bottom layer of the pair is about 4.5×10^{10} cm^{-2} while the density in the second layer is about 2.5×10^{10} cm^{-2}. Meanwhile, the top islands are nearly double the size of the bottom islands due to the additional deposition and higher growth temperature.

The PL was excited by the 514.5 nm line of a continuous wave Ar^+ laser. We applied excitation densities in the range 0.01–20 W/cm^2. The samples were mounted in a close-cycle cryostat, which allows measurements in the temperature range from 10 to 300 K. The PL signal was detected with a LN_2 cooled Ge photodiode using phase-sensitive detection.

Figure 1 shows the low-temperature PL spectrum from the 8× double-layer sample A. Also shown in the same figure are the PL spectra of two reference samples B and C containing multiple layers of only one of the two InAs QD double-layers, i.e., either sample B (2.4 ML) or sample C (1.8 ML). For samples B and C, the main PL peak can be fitted by a single Gaussian, indicating that the observed dot formation has only one dominant size. Sample B shows a single PL peak at an energy of 1.16 eV with the full width at half maximum (FWHM) of 50 meV while for sample C, the PL peak is at 1.27 eV with FWHM ~120 meV. These data are in agreement with expected values for the given growth conditions.[7]

The PL spectrum from the double-layer stacked InAs/GaAs QDs (sample A) shows a pronounced double-peak structure. This can be attributed to the total contribution in PL signal from QD ensembles of both layers. Indeed, a line

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Atomic coherence induced Kerr nonlinearity enhancement in Rb vapour

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Abstract. The Kerr nonlinear index of refraction for rubidium atoms is measured by using an optical ring cavity with and without electromagnetically induced transparency (EIT). Significant enhancement and inhibition of the Kerr nonlinear index is observed near resonance with EIT. The nonlinear index of refraction is measured as functions of probe and coupling frequency detunings, respectively, with and without the presence of EIT. A simple theoretical calculation including Doppler broadening is presented and is found to be in good qualitative agreement with the experimentally measured results.

1. Introduction

Multi-level atomic systems with electromagnetically induced transparency (EIT) [1–4] have been shown to enhance nonlinear optical processes such as frequency conversion [5–8] and four-wave mixing [9–11]. Enhanced third-order Kerr-type nonlinearity $\chi^{(3)}$ generated in multi-level atomic systems due to atomic coherence together with reduced absorption is responsible for such efficient nonlinear optical processes. Such enhanced Kerr nonlinearity also has various useful applications such as in cross-phase modulation for optical shutters [13] and self-phase modulation for generation of optical solitons [14]. In recent years, EIT and enhancement of linear and nonlinear susceptibilities with reduced absorption in multi-level atomic systems have attracted much interest [1–23]. Large nonlinear susceptibilities at low light powers are desirable for realization of photon blockades and single-photon nonlinear devices [13, 20–22]. To understand and optimize these nonlinear optical processes, one needs to have precise knowledge of the Kerr nonlinear coefficient n_2 as functions of other parameters such as pumping power and frequency detuning. Although many papers have been published on enhancing nonlinear optical processes, no direct experimental measurements of $\chi^{(3)}$ or n_2 have been reported, so far, in such multi-level atomic systems with atomic coherence conditions satisfied.

This paper presents experimental results in measuring the nonlinear Kerr index of refraction for three-level rubidium atoms inside an optical ring cavity with

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Coherent Microwave Generation in a Nonlinear Photonic Crystal

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Abstract—**We propose a new approach for generating coherent microwaves in a nonlinear photonic crystal through optical rectification. In a photonic crystal, the reciprocal vector and dispersion of the group velocity can be used to both compensate for the velocity mismatch between the generated microwave and the pump light. We show that coherent microwave radiation from kilohertz to terahertz can be generated through this approach by designing a suitable structure.**

Index Terms—**Coherent microwave, nonlinear photonic crystal, optical rectification, quasi-phase-matching.**

I. Introduction

OVER the last few decades, a great deal of attention has been given to materials with artificial periodic structures [1], i.e., superlattices. Among them are photonic crystals and quasi-phase-matched (QPM) materials. In a photonic crystal, the dielectric constant is varied periodically resulting in a dispersion relation that exhibits a band structure [2]. As a result, the propagation of light with a frequency in the band gap is suppressed. Some novel laser geometries [3], [4] and waveguide devices [5] have being constructed by making use of this frequency selectivity. Recent work on nonlinear photonic crystals has focused on the third-order nonlinearity [6]–[8]. While second-order nonlinear optical frequency conversion has not been well studied in such a structure [9], QPM materials—especially their most famous representative, periodically poled $LiNbO_3$ (PPLN)—have been carefully investigated [10], [11]. In PPLN or its analogs, the second-order nonlinear optical coefficient is periodically modulated, while its dielectric constant is uniform, leading to compensation of the phase mismatch between the input and the generated beams [11]. Due to the similarities in structure and some of the physical principles governing photonic crystals and QPM materials, a QPM material could be viewed as a special case of a nonlinear photonic crystal [12].

Among second-order nonlinear optical effects, optical rectification has been known for decades [13], but it has not been as widely studied as other nonlinear effects, such as second harmonic generation (SHG), difference-frequency generation (DFG), or optical parametric oscillation. However, recent reports show that optical rectification can be used to generate ultra-short electrical pulses with a high repetition rate [14], and terahertz radiation [15], [16].

In this paper, the optical rectification effect in a nonlinear photonic crystal is investigated. It is shown that coherent microwaves can be efficiently generated in a nonlinear photonic crystal through optical rectification. In addition, an intrinsic relationship between a nonlinear photonic crystal and a QPM material is established by considering optical rectification. The structural parameters of several coherent microwave sources are also calculated.

II. Theoretical Analysis

Although there is progress in generating coherent microwaves [17], [18], pursuing a highly efficient and simple coherent microwave source with good spatial and spectral characteristics is still a large challenge. Here, we consider optical rectification of a modulated light beam as a source of coherent microwaves. For a modulated light beam propagating through a nonlinear crystal, a modulated electric field is generated through optical rectification [14], so that every point where the pump light passes is a source of microwaves, where the frequency of the microwaves is determined by the repetition rate of the pump light. The generated microwaves will interfere with each other in any arbitrary direction, and the final microwave intensity along a specific direction depends on the relative phase difference between the microwaves. Obviously, if the light pulse's velocity is equal to the velocity of the microwave, all generated microwaves will interfere with each other constructively in the forward direction, and efficient coherent microwaves will be generated. Here, we should point out that it is the group velocity not the phase velocity of the pump light that should match the microwave's phase velocity, because it is the light pulses, and not the continuous wave (CW) light, that excites microwaves. Velocity matching is the prerequisite condition for coherent microwave generation through optical rectification. Unfortunately, for most materials, the optical refractive index is different from that of a microwave frequencies. The constructive interference condition thus cannot, in general, be fulfilled, making effective microwave generation difficult.

Two methods can be used to solve the velocity-matching problem: control of the group velocity of the pump light or control of the phase velocity of the microwaves. In this paper, we demonstrate that these two approaches can both be realized in a nonlinear photonic crystal.

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Effects of a highly dispersive atomic medium inside an optical ring cavity

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Abstract. Atomic media inside an optical cavity can significantly alter the spectral response of the cavity. Both theoretical and experimental examinations are made of the cavity transmission with a highly dispersive intracavity multi-level atomic medium. It is found, owing to the reduced absorption and steep dispersion change accompanying electromagnetically induced transparency in such a multi-level atomic medium, that the cavity linewidth can be made much narrower than the empty cavity linewidth. Cavity linewidth narrowing is measured as a function of both the coupling beam power and the atomic density. These experimental results are in good agreement with the theoretical predictions.

1. Introduction

The interactions between electromagnetic fields and atomic media can be greatly enhanced inside an optical cavity owing to the extended cavity lifetime of photons. For this reason, optical cavities are often used to enhance nonlinear optical effects as well as to achieve ultra-sensitive spectroscopic measurements. The transmission properties of an optical cavity containing an atomic medium are highly dependent on the atomic absorption and dispersion. In recent years, it has been demonstrated that one can modify the absorptive and dispersive properties of a weak probe field near an atomic transition frequency in multi-level atomic systems by applying a strong coupling field at another atomic transition [1–6]. This effect, known as electromagnetically induced transparency (EIT), is due to atomic coherence and quantum interference induced by probe and coupling fields together in three-level atomic systems, and is the underlying mechanism for such phenomena as the slowing down of the group velocity of light to a few metres per second [5–9], lasing without inversion [10], enhanced index of refraction [11], optically written waveguides in atomic vapour [12], and photon storage [13].

When multi-level atomic media are put into an optical cavity, the atomic flourescence spectrum and cavity transmission are shown to be modified owing to atomic coherence and quantum interference [14]. The greatly reduced absorption and sharp, linear dispersion change in such systems have led researchers to predict novel cavity transmission effects such as optical bistability [15], squeezing [16],

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Electronic structure transformation from a quantum-dot to a quantum-wire system: Photoluminescence decay and polarization of colloidal CdSe quantum rods

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Radiative lifetime, polarization, and the global Stokes shift of colloidal CdSe quantum rods with aspect ratios from 1.9 to 3.8 are measured at room temperature. The radiative lifetime and the global Stokes shift show nonmonotonous dependence on the aspect ratio of the nanorods, and strong linear polarization in emission sharply appears as the aspect ratio crosses a turning point. The features of radiative lifetime and polarization versus aspect ratio in these nanorods indicate a transformation of the electronic structure from a zero-dimensional quantum-dot system to a one-dimensional quantum-wire system. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1529086]

Nanometer-size semiconductor dots, rods, tubes, and wires exhibit electronic and optical properties that sensitively depend on both their sizes and shapes, and are both fundamentally and technologically interesting.[1–8] One of the best-studied semiconductor nanocrystallites is a colloidal CdSe quantum nanostructure.[8] Its relatively reproducible and controllable synthetic chemistry and strong size-dependent optical properties have made it the subject of many detailed spectroscopic studies and allowed detailed comparisons between experiments and theories. Recently, it has been demonstrated that the shape of the colloidal CdSe nanocrystals can be manipulated from a nearly spherical morphology to a rodlike one by controlling the growth kinetics.[5] The quantum rod is an intermediate form between the zero-dimensional (0D) quantum dot and the one-dimensional (1D) quantum wire, and can be used to explore how the electronic structures evolve from a 0D quantum system to a 1D quantum system.[4,6,9] For example, an empirical pseudopotential calculation predicts that when a CdSe dot with a diameter of 3.8 nm is elongated, a crossover of the two highest occupied electronic states occurs at an aspect ratio of 1.36.[6] Single-molecule spectroscopy measurements on CdSe rods confirmed a sharp transition from a nonpolarized to a linearly polarized emission at an aspect ratio of about 2, which is believed to be a consequence of such level crossover.[4,6]

Time-resolved spectroscopy is a powerful technique to probe energy relaxation and recombination dynamics in CdSe nanostructures.[7,8] In this letter, we report our experimental study of the shape dependence of the time-resolved photoluminescence (PL) spectroscopy of CdSe nanorods. The measured radiative lifetime first decreases and then increases as the aspect ratio of the nanorods increases. The nonmonotonous change of the radiative lifetime indicates the occurrence of a transformation of the electronic structure of the nanorods with the increase of the aspect ratio. Manipulating the radiative lifetime of colloidal nanostructures may have potential applications in biophotonic sensors.[10]

The colloidal CdSe quantum rods were synthesized by the injection of precursor molecules into a hot surfactant, which was described in detail elsewhere.[11,12] Shape control of the nanorods was achieved by the manipulation of the growth kinetics to make the growth rate along the long axis at least 2 orders of magnitude faster than that along the short axis. Figure 1 shows the transmission electron microscopy (TEM) images of two of the used nanorod samples. The size distribution, as determined by measuring the TEM images of more than 500 particles for each sample, is less than 5% in diameter and less than 8% in length for each sample. The nanorods for our experiment have the aspect ratios from 1.9 to 3.8, with a similar short-axis dimension of around 4.5 nm. Absorption spectra were taken from these nanorods in toluene solution. The samples for PL and radiative lifetime measurements were prepared by spin coating a clean fused silica coverslip with a dilute solution of nanorods in toluene fol-

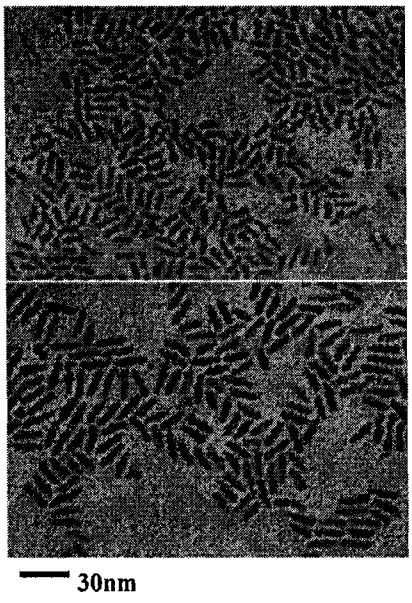

FIG. 1. TEM images of two CdSe rod samples with aspect ratios of 2.5 (up) and 3.3 (down), respectively.

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PHYSICAL REVIEW A, VOLUME 65, 051802(R)

Controlling the cavity field with enhanced Kerr nonlinearity in three-level atoms

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We experimentally demonstrate cavity field control by another laser beam with three-level Λ-type atoms inside an optical ring cavity. By adjusting the frequency detuning of the controlling beam (coupled to one of the atomic transitions), the intensity of the cavity field interacting with another atomic transition is switched on and off. Such all-optical switching between two steady states is caused by the enhanced Kerr nonlinearity due to atomic coherence in such system.

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The essential element in all-optical communication and optical computation is to achieve effective and fast all-optical switching. Multilevel electromagnetically induced-transparency (EIT) systems are ideal for such applications, since the linear and nonlinear optical properties of the probe beam can be dramatically changed by another (controlling) laser beam due to atomic coherence [1–11]. When such an EIT medium is placed inside an optical cavity, many other interesting effects, such as cavity linewidth narrowing [12], optical bistability [13,14], and dynamic instability [13], appear. The Kerr-nonlinear index of refraction of a three-level Λ-type atomic system can be greatly enhanced near resonance (by two orders of magnitude in some frequency detuning regions) and can change sign with small changes (tens of megahertz) in the frequency detunings of the controlling beam or cavity (probe) field [15]. Such enhancement in Kerr nonlinearity is caused by the atomic coherence induced in this EIT medium.

With the knowledge of these dramatic changes of the Kerr-nonlinear index of refraction near resonance, we demonstrate that the cavity-field intensity can be controlled by the frequency detuning of the controlling field at a relatively low optical power. With a small frequency change of the controlling beam (24 MHz), the cavity-field intensity switches with a switching ratio better than 30:1 and a switching time of few microseconds. Such all-optical switching is fundamentally interesting and can have applications in all-optical communication, optical logic gates, and optical information processing.

The experiment is performed in a composite system consisting of a collection of three-level Λ-type rubidium atoms, as shown in Fig. 1, in an atomic vapor cell and an optical ring cavity. $F=1$ (state $|1\rangle$) and $F=2$ (state $|3\rangle$) states of $5S_{1/2}$ are the two lower states and $F'=2$ state (state $|2\rangle$) of $5P_{1/2}$ serves as the upper state. The controlling (coupling) laser beam (with frequency ω_c) couples states $|3\rangle$ and $|2\rangle$ while the switching (probe) beam (with frequency ω_p) interacts with states $|1\rangle$ and $|2\rangle$. The frequency detuning of the controlling beam is defined as $\Delta_c=\omega_c-\omega_{23}$, where ω_{23} is the atomic frequency of the $|3\rangle$ to $|2\rangle$ transition, and the frequency detuning of the switching beam from its atomic transition is defined as $\Delta_p=\omega_p-\omega_{12}$. As we have previously demonstrated, the Kerr-nonlinear index of refraction n_2 changes dramatically both in magnitude and in sign near resonance [15]. These changes can be used to control the steady-state behaviors of the system. By changing the frequency detuning of the controlling beam, we can control the nonlinearity of the system and, therefore, make the output intensity of the cavity field to be at different steady states. Such action constitutes a controllable all-optical switching in this composite atom-cavity system.

A simplified sketch of the experimental setup is shown in Fig. 2 with an atomic vapor cell containing three-level atoms placed inside a three-mirror optical ring cavity. Both the controlling (LD1) and the switching (LD2) lasers are single-mode diode lasers that are current and temperature stabilized. The frequencies of these two diode lasers are further stabilized by using optical feedback through servo-loop-controlled mirrors. Parts (about 10%) of the switching and controlling beams are split by polarizing beam splitters PB2 and PB4 to a saturation absorption spectroscopy setup for monitoring the frequency detunings of the two lasers from their respective resonances with atomic transitions. The flat mirror $M2$ and the concave mirror $M1$ ($R=10$ cm) have about 1% and 3% intensity transmissions, respectively. The third cavity mirror (concave with $R=10$ cm) is mounted on a piezoelectric transducer (PZT) with reflectivity larger than 99.5%. The finesse of the empty cavity is about 100 with a free spectral range of 822 MHz (cavity length is 37 cm). A third laser (reference beam) is used to lock the frequency of the optical cavity to a Fabry-Perot cavity, which can be tuned easily by a PZT mounted on one of the mirrors. The ru-

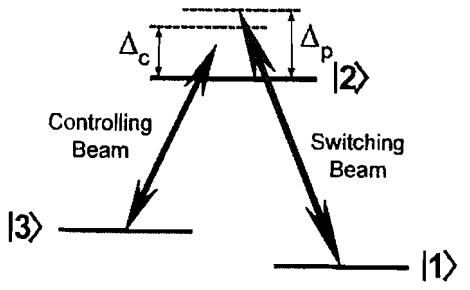

FIG. 1. Sketch of a three-level Λ-type atomic system.

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Lattice contraction in free-standing CdSe nanocrystals

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Colloidal CdSe nanocrystals (NCs) with high photoluminescent quantum yield are fabricated by a method adopting a large initial Se:Cd ratio of the precursors in the solution. X-ray diffraction and Raman measurements confirm that there exists lattice contraction in the free-standing wurtzite CdSe NCs. The lattice contraction indicates the occurrence of surface optimization/reconstruction during the growth, which results in the high quantum yield of the obtained CdSe NCs. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1507613]

Colloidal II–VI semiconductor nanocrystals (NCs) are of increasing interest because of their applications as optoelectronic, photochemical, and nonlinear optical materials.[1] An important feature of these NCs is the very large surface-to-volume ratio. For example, CdSe nanocrystals have 10%–50% of their total atoms exposed on the surfaces.[2] Therefore, various passivation ligands have to be used during synthesizing the colloidal NCs to neutralize the surface dangling bonds and to sterically prevent the nanocrystals from touching each other and permanently fusing together.[3] The surface of the resulting NCs might then be considered as an ensemble of dangling and terminated bonds and various point defects.[4] As a result, the complex surfaces of the obtained colloidal semiconductor NCs play an important role in the photoluminescence (PL) quantum yield (QY) and the stability of the emission. An organometallic approach using dimethylcadmium as the cadmium precursor has been well developed during the past ten years to fabricate CdSe NCs.[5] Traditionally, the initial Se:Cd ratio of the precursors in the solution is set close to unity. The best PL QY reported for the as-grown NCs is around 20% in the wavelength range between 520 and 600 nm, and the PL QY can be boosted to 50% with some inorganic/organic surface passivations.[6] Recently, an alternative route with safe and inexpensive cadmium precursors has been introduced, which adopts a large initial Se:Cd ratio of the precursors (in the range of 5–10).[7,8] The resulting CdSe NCs have high PL QY values (as high as 80% in the orange–red range) and long stability period upon aging at least for several months. Previous work proposed that the high PL efficiency is not only from surface passivation by ligands, but also from surface optimization/reconstruction during growth.[8] In the letter, we further explore and experimentally confirm such surface optimization/reconstruction by taking Raman and x-ray diffraction (XRD) measurements of these newly synthesized NCs.

The fabrication of the colloidal CdSe NCs was described with detail in previous publications:[7,8] The initial Se:Cd ratio was selected to be 10 to fabricate these CdSe NCs used in this letter. A series of samples was taken from the solution during growth and washed to remove excess surfactants. The size of the NCs, which was controlled by the growth time, ranged between 2.5–6 nm. Transmission electron microscopy (TEM) images indicated that these obtained NC dots are spherical. The PL, absorption, and PL excitation (PLE) measurements were taken for these NCs in toluene solution, but the Raman and XRD measurements were done with these NCs on silicon wafers. The PL QY values were obtained by comparing the integrated PL intensities of the NCs to the well-characterized dye (R6G). Figure 1 shows the dependence of PL QY on the size of the NCs. With the increase of the size of the NCs (R), i.e., the growth time, the PL QY increases to a maximum value at R around 4 nm and then decreases. The point was defined as the PL bright point in the previous article.[8] The typical PL, absorption, and PLE spectra are also shown in Fig. 1 (inset). The narrow PL spectral band and the fine features shown in both the absorption and PLE spectra indicate that the obtained CdSe NCs have a narrow size distribution. The size distribution, as determined by measuring TEM images of more than 500 particles for each sample, was about 5%–10%. Hexadecylamine (HDA) was used as the surface ligand for the NCs used in the ex-

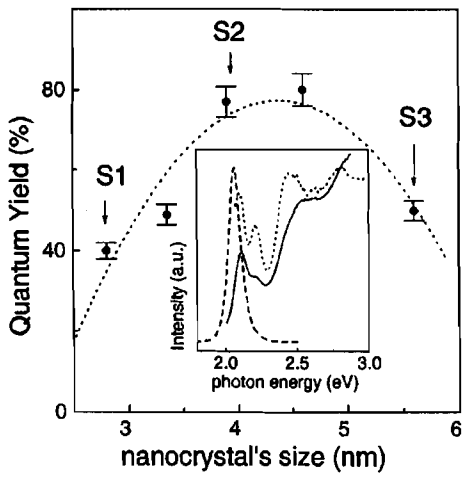

FIG. 1. The dependence of PL QY on the size of the CdSe nanocrystals. Three samples used for XRD and Raman measurements are labeled as S1, S2, and S3. The inset shows the typical PL (dashed), absorption (solid), and PLE (dotted) spectra.

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Polarization spectroscopy of single CdSe quantum rods

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Excitation and emission spectra of CdSe single quantum rods with aspect ratios of 2 and 4 were investigated by far-field microscopic technique. Typical spectral linewidths of the rods are less than 60 meV at room temperature. Both excitation and emission of the single rods exhibit strong polarization dependence, indicating that dipole moment exists along the long axis of the rods, e.g., the unique axis c of the wurtzite structure.

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Colloidal CdSe quantum dots have received much attention in recent years.[1–4] Their relatively reproducible and controllable synthetic chemistry and their strong size-dependent optical properties with band edge tunable through the visible wavelength range make them of particular interest for fundamental studies as well as potential applications. The spectroscopy of single quantum dots has revealed many interesting physical phenomena such as ultranarrow line shapes, luminescence intermittence, shifting of emission spectra, and a highly polarizable emitting state in the presence of strong local electric fields.[5]

Wurtzite CdSe nanocrystal is intrinsically an anisotropic material. Measurements of the dielectric dispersion of CdSe nanocrystals have shown the existence of large dipole moments along the unique c axis of the wurtzite structure.[6] Quantum-confined Stark effect measurements in single CdSe dots also implied the presence of a permanent dipole.[7] More direct measurements using electrostatic force microscopy (EFM) determined that the static dielectric constant of single CdSe nanocrystals is uniform, although the charge per nanocrystal is nonuniform.[8] Due to the prolate shape and wurtzite crystal structure, calculations have predicted two potential dipole orientations for the CdSe nanocrystals.[9] One is oriented parallel to the wurtzite c axis, and the other is in the plane perpendicular to the c axis. Polarized emission of quantum dots suggested that degenerated dipoles are only formed in the plane.[10] Recent theoretical treatments have found very large dipole moments along the c axis, which strongly depend on the exact structure of the nanocrystals and may vary significantly with small structural changes.[11] However, polarized absorption was also observed in single CdS nanocrystals, which is unexpected since CdS nanocrystals have a tetrahetral structure.[12] All these previous results indicate the importance of polarization studies for understanding the basic electronic and optical properties of nanocrystals. Consequently, studies of polarization properties will explore another dimension for device applications of colloidal nanocrystals.

Recently, it has been demonstrated that the shape of CdSe nanocrystals can be manipulated by controlling the growth kinetics.[13,14] The resulting nanoparticles can be from a nearly spherical morphology to a rodlike one. The successful growth of these rod- and wire-like structures provides a testing ground for studying the shape-dependent electronic and optical properties of nanocrystals and the quantum confinement effects in one-dimensional (1D) and quasi-1D regimes. CdSe quantum rods have well-defined shape: their long axis is preferably grown along the unique c axis.[13,14] This unique feature is helpful to determine nanocrystal orientation and make them particularly interesting for studying dipole formation and polarization effects. Polarization-dependent emissions have been observed in an ensemble multirod system, in which a certain degree of alignment of being parallel along their long axis of the rods is obtained by stretching PVB polymer embedded with the rods. A stronger polarized emission was observed along the long axis of the rods.[13] This was tentatively assigned to the level crossover of the two highest occupied electronic states due to the elongation of the nanocrystals along their c axis. A recent empirical pseudopotential calculation has shown that, due to energy level crossover, elongated dots will emit linearly polarized light instead of plane-polarized light as the aspect ratio of the elongated dots (or rods) becomes larger than 2.[15]

In this paper, we report our experimental studies of the polarization dependence of both emission and excitation (or absorption) photoluminescence (PL) spectra of single quantum rods with aspect ratios of 2 and 4. Such studies can eliminate uncertainties in ensemble multirod measurements and be used to make comparisons with theoretical predictions. Measurements of the excitation polarization dependence in single quantum rods are particularly significant since they are fundamentally different from measurements done with single quantum dots.[10]

Colloidal CdSe quantum rods were prepared using a method as described by Peng and Peng.[14] Briefly, trioctylphosphine oxide (TOPO) and a strong Cd ligand, either hexylphosphonic acid (HPA) or tetradecylphosphonic acid (TDPA), were heated to 320–360 °C. Various stock solutions of Cd $(CH_3)_2$ and Se in TBP were prepared and rapidly injected into the hot solution of HPA or TDPA in TOPO. These colloidal rods were examined by transmission electron microscopy (TEM) to determine their sizes and to analyze the size and shape distributions. Figure 1 shows the TEM pictures of two quantum rod samples. The first rod sample, shown in Fig. 1(a), has a length of about 8 nm with an average aspect ratio of about 2. The second sample, shown in

PHYSICAL REVIEW A, VOLUME 65, 011801(R)

Bistability and instability of three-level atoms inside an optical cavity

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Optical bistability and dynamic instability are experimentally observed and studied in a system consisting of three-level Λ-type rubidium atoms in an optical ring cavity. The bistable behavior and self-pulsing frequency are experimentally manipulated by changing the controlling and cavity field parameters (power and frequency). These nonlinear effects are influenced by the enhanced Kerr nonlinearity due to atomic coherence in such a system.

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When two-level atoms couple with an optical cavity, many interesting effects occur. For example, bistable states appear in the cavity output intensity for a certain range of input intensities [1] and the output field can become unstable in the upper branch of the bistable curve under certain conditions [2]. Bistability and instability were also observed in cold clouds of cesium atoms inside an optical cavity, where degenerate Zeeman sublevels participate in the dynamic processes [3]. In that experiment, one circularly polarized laser beam was injected into the optical cavity, which interacts with all the Zeeman sublevels of the $6S_{1/2}$ $F=4$ and $6P_{3/2}$ $F=5$ states in cesium atoms. The dynamic instability was attributed to the competition between optical pumping to state $6S_{1/2}$ $F=4$, $m_F=4$ from all other Zeeman sublevels and nonlinearities due to saturation of the optical transition (from $6S_{1/2}$ $F=4$, $m_F=4$ to $6P_{2/3}$ $F=5$, $m_F=5$). However, since both optical pumping and saturation inside the optical cavity were generated by the same input laser beam, the two competing dynamic processes could not be separately controlled. Also, the trapping beams and the repumping beam in that experiment for creating the cold atomic clouds bring more complications and limitations to the understanding of these bistable and dynamic instability behaviors.

In recent years, electromagnetically induced transparency and related effects in three-level atomic systems have been studied extensively [4–6]. Many experimental demonstrations of enhancing nonlinear optical processes (such as four-wave mixing, harmonic generations, and two-photon absorption) were reported in the literature [7–11]. Recently, we have studied cavity linewidth narrowing due to the sharp dispersion change in three-level Λ-type rubidium atoms inside an optical ring cavity [12] and directly measured the Kerr-nonlinear coefficient near resonance [13]. In this paper, we report our recent experimental observations of optical bistability and dynamic instability with three-level rubidium atoms inside an optical ring cavity. Due to the exact knowledge of the Kerr-nonlinear coefficient at different controlling (or coupling) and cavity (or probe) field parameters, such as optical powers and frequency detunings [13], we can control and manipulate the bistable and unstable behaviors in such a system by simply tuning these experimental parameters.

Our basic experimental setup is shown in Fig. 1. The optical ring cavity is about 37 cm long and is composed of three mirrors. The flat mirror $M1$ and the concave mirror $M2$ ($R=10$ cm) have about a 1 and 3 % transmissivity, respectively, while the third mirror is concave ($R=10$ cm) with a reflectivity larger than 99.5% and is mounted on a piezoelectric transducer (PZT). The empty cavity finesse was measured to be about 100 with a free spectral range of 822 MHz. The rubidium vapor cell is 5 cm long with Brewster windows and is wrapped in μ metal for magnetic shielding and heat tape for atomic density control. The experiment was conducted at a vapor cell temperature of about 70 °C. One laser beam enters the cavity through mirror $M2$ and circulates inside the cavity. Another beam, i.e., the controlling field, is introduced through the polarizing beam splitter (PBS) with an orthogonal polarization to the first (cavity field) beam. The controlling beam is misaligned from the cavity field beam by a 2° angle and does not circulate inside the ring cavity. The radii of the controlling and cavity field beams at the center of the Rb vapor cell are estimated to be 700 and 80 μm, respectively. With insertion losses of the PBS and reflection losses from the vapor cell windows, we find the cavity finesse to be degraded to about 50 at a frequency far from atomic resonance.

Both lasers are extended cavity diode lasers. The controlling field is tuned to the $5S_{1/2}$ $F=2 \leftrightarrow 5P_{1/2}$ $F=2$ transition, while the cavity field is tuned to the $5S_{1/2}$ $F=1 \leftrightarrow 5P_{1/2}$ $F=2$ transition in ^{87}Rb, as in Ref. [12]. Both lasers are frequency locked to Fabry-Perot cavities. Their frequency de-

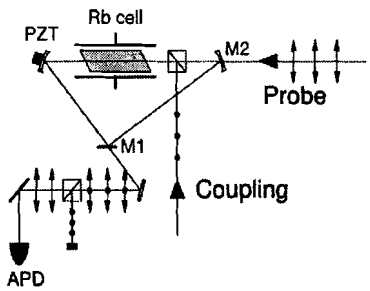

FIG. 1. Experimental setup. $M1$ and $M2$ are mirrors; PZT is a mirror mounted on a piezoelectric transducer; APD is an avalanche photodiode detector.

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Wide-bandwidth high-frequency electro-optic modulator based on periodically poled $LiNbO_3$

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We propose a high-frequency traveling-wave integrated electro-optic modulator based on a periodically poled $LiNbO_3$. The traveling velocity of the optical wave and the electrical wave velocity in the waveguide can be quasimatched due to the periodic structure. Using this design, a modulation frequency of several hundred GHz can be realized. Wide-bandwidth modulation is also achievable by employing an aperiodic domain grating. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1350426]

As an outstanding nonlinear optical material, periodically poled $LiNbO_3$ (PPLN) is attracting more and more attention.[1–5] In PPLN, the spontaneous polarization is periodically reversed, resulting in nonlinear optical properties. For example, the quasi-phase-matching (QPM) technique can be used instead of birefringence phase matching for nonlinear frequency conversions.[1–5] The physical mechanism of QPM is that the nonlinear optical coefficient periodically changes its sign due to the periodic domains. As a result, the excited parametric wave will have a π phase shift when passing through the domain boundary. If each domain thickness is equal to the coherence length, the excited parametric wave from each domain will interfere constructively. In addition to frequency conversion, $LiNbO_3$ (LN) is also widely used in pizeoelectric and electro-optic (EO) processes. The piezoelectric and EO coefficients also change their signs periodically in a PPLN and yield interesting effects.[6–9]

A popular application of the EO effects of LN is the integrated EO modulator (EOM) that has important applications in signal processing and optical communications.[10–12] Driven by the demands of high-speed devices, the modulation frequency of EOM has been increasing. However, high-speed modulation is limited due to the velocity mismatch between the electrical wave and the optical wave.[10,11] To increase the modulation speed, several effective methods have been proposed.[12–14] Since PPLN has enhanced EO properties,[8] and PPLN wafers are now commercially available, the application of PPLN for high frequency EOM is a very attractive option.

Before studying EOM in PPLN, let us look back to the ordinary traveling wave modulator. Since the Mach–Zehnder modulator is based on phase modulation. We can study the phase modulating properties first. For simplicity, we assume the electrode to be impedance matched to the drive cable and termination. The microwave loss is also ignored.

We consider a single frequency (ω_m) drive electrical signal, which propagates in the waveguide along the x direction with the velocity $v_m = c/n_m$. c is the light velocity in vacuum and n_m is the refractive index of the waveguide at the drive frequency. The voltage of the drive signal in the waveguide then could be written as:

$$u(x,t) = u_0 \sin(k_m x - \omega_m t) \quad (0 \leqslant x \leqslant L), \tag{1}$$

where k_m is the wave vector of the electrical wave; L is the total interaction length. The optical wave that enters the interaction zone ($x=0$) at $t=t_0$, meets the drive voltage $u(x,t_0)$. When $t=t_0+\Delta t$, the optical wave has traveled a distance x with the velocity of $v_o = c/n_o$, which takes the time $\Delta t = x/v_o$, where n_o is the refractive index for the optical wave. Thus, the applied voltage that the optical wave actually sees is

$$u(x,t) = u_0 \sin[k_m \alpha x - \omega_m t_0], \tag{2}$$

where $\alpha = 1 - v_m/v_o$. For an ordinary medium, due to the velocity difference between the optical wave and the electrical wave, the voltage changes along the waveguide. We can easily build this physical image with the help of the Fig. 1.

From Fig. 1, the wavefront of the optical wave meets a different drive voltage at a different point. The changing period Λ of the actual voltage is the distance for the optical wave to catch up with the electrical wave with 2π phase difference, where Λ is given by:

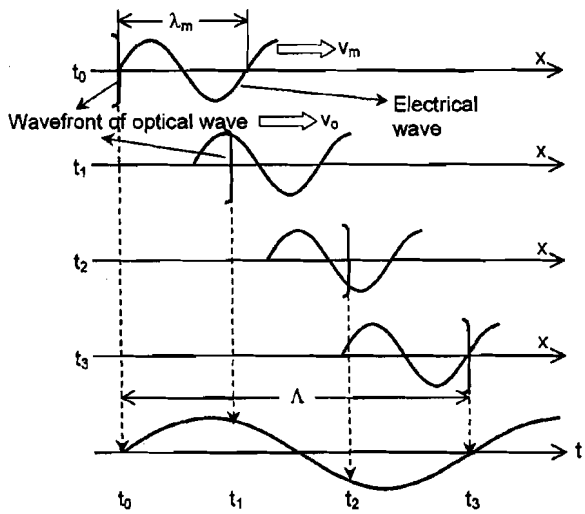

FIG. 1. Velocity mismatch between input optical wave and drive electrical signal.

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