Quantum theory of spontaneous emission and excitation near a phase-conjugating mirror

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Received February 15, 1989; accepted May 18, 1989

We present a quantum-electrodynamic theory of spontaneous emission in the presence of a phase-conjugating mirror (PCM) and show that the radiative relaxation rate is increased owing to the amplification of quantum noise by the PCM. For any finite value of the PCM reflectivity an atom in any initial state will relax to a steady state with a finite probability of being excited. In particular, there is a finite probability of spontaneous excitation of a ground-state atom placed near the PCM. The fluorescence spectrum of an atom near a PCM is generally described by two Lorentzian functions.

1. INTRODUCTION

It has long been recognized that spontaneous emission does not always proceed at the rate given by the Einstein coefficient

$$A = 4|\mu|^2\omega_0^3/3\hbar c^3, \tag{1.1}$$

where ω_0 is the transition (angular) frequency and μ is the electric dipole matrix element for the transition. Purcell in 1946 argued that when an atom is placed in a lossy (overdamped) cavity the spontaneous-emission rate is increased by the factor Q, the cavity quality factor[1]; this result has been derived from various points of view.[2,3] More interesting perhaps are experiments in which the spontaneous-emission rate is found to vary with the distance of the emitter from a mirror surface.[4] This effect is well understood theoretically: the emission rate depends on the modal properties of the field, and in the presence of mirrors the appropriate modes are not the free-space modes giving rise to Eq. (1.1). At precisely a nodal point of the field for a particular mode, for instance, there is no spontaneous emission into that mode. We can understand these effects in terms of the reflection of the atomic field back to the atom or, for some purposes, as a consequence of a cooperative emission by the atom and its image.[5] Various aspects of cavity quantum electrodynamics have been the subject of some elegant experiments involving the enhancement or inhibition of spontaneous emission.[4,6,7]

In this paper we discuss the rather different situation in which the reflecting surfaces are phase conjugating. Phase conjugation is of interest in a variety of applications in which it is desirable to remove the phase distortions of a wave front and has been studied extensively both theoretically and experimentally.[8] The case of an atom near a phase-conjugating mirror (PCM) is the simplest example of the interaction of an atomic medium with a PCM. Intuitively, it is expected that a PCM could affect spontaneous emission more dramatically than could an ordinary mirror. Such an expectation might follow from Fig. 1, which compares the effects of PCM's and ordinary mirrors on the spherical wave produced by a point source. Unlike an ordinary mirror, a PCM converts a diverging spherical wave into a converging spherical wave focused back onto the source.

Such an effect was considered by Agarwal.[9] He considered a classical oscillating dipole in the presence of an infinite PCM and found that the PCM changes the radiative decay rate from the free-space value β to $\beta_{\rm PCM} = [1 - {\rm Re}(\tilde{a})]\beta$, where $\tilde{a} = |a|e^{i\theta}$ is the complex gain factor of the PCM. Agarwal emphasizes that the change in the decay rate is independent of the distance of the dipole from the PCM. Recently we showed how such results are modified by a PCM of finite extent and considered the spontaneous decay of a localized collection of N atoms in the presence of a PCM.[10] We argued that, for $N \gg 1$, semiclassical radiation theory is adequate for an understanding of some prominent features of the radiative decay. Using the semiclassical approach, we showed that the ground state of the atomic sample is unstable when $|m| \equiv 3|a|\Omega/4\pi > 1$, where Ω is the solid angle subtended by the PCM at the source. In particular, we inferred that any small disturbance would cause the atoms to become excited (spontaneous excitation) and begin fluorescing, with the fluorescence taking the form of an irregular sequence of superradiant pulses.

In the present work we will use a quantum-electrodynamic approach to the case of a single atom near a PCM, a problem for which semiclassical theory is inadequate. We assume

0740-3224/89/101932-10$02.00

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ADVANCES IN ATOMIC, MOLECULAR, AND OPTICAL PHYSICS, VOL 28

SOME RECENT DEVELOPMENTS IN THE FUNDAMENTAL THEORY OF LIGHT

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ISBN 0-12-003828-5

PHYSICAL REVIEW A VOLUME 40, NUMBER 7 OCTOBER 1, 1989

Natural line shape

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For many years there have been two expressions for the natural radiative line shape, depending on the form of the Hamiltonian assumed for the interaction of an atom with the radiation field. It is shown that this difference arises from artificial assumptions about the atom-field interaction, and that the same line shape is obtained from either form under the appropriate physical assumptions.

I. INTRODUCTION

In 1952 Lamb noted that two different expressions could be obtained for the natural radiative line shape, depending on whether a bound electron is assumed to couple to the radiation field through the vector potential $\mathbf{A}$ or the electric field $\mathbf{E}$.[1] If an interaction $-(e/mc)\mathbf{A}\cdot\mathbf{p}$ was assumed, the line shape was

$$S_A(\omega)=\frac{\beta}{\pi}\frac{\omega/\omega_0}{(\omega-\omega_0)^2+\beta^2}\ , \tag{1.1}$$

where $S_A(\omega)d\omega$ is the probability of finding the emitted photon in the (angular) frequency interval $[\omega,\omega+d\omega]$, ω_0 is the transition frequency, and β is half the Einstein A coefficient. If instead the interaction was taken to be $-e\mathbf{r}\cdot\mathbf{E}$, then the line-shape function was found to be

$$S_E(\omega)=\frac{\beta}{\pi}\frac{\omega^3/\omega_0^3}{(\omega-\omega_0)^2+\beta^2}\ . \tag{1.2}$$

The difference between $S_A(\omega)$ and $S_E(\omega)$ arises from the fact that $\mathbf{E}=-(1/c)\partial\mathbf{A}/\partial t$, and so $\mathbf{E}$ has an extra factor of ω, compared with $\mathbf{A}$, in the mode expansion of the field. Lamb remarked that "the usual interpretation of probability amplitudes is valid only [in the $-e\mathbf{r}\cdot\mathbf{E}$ interaction], and no additional factor $[\omega_0^2/\omega^2]$ actually occurs." The line shape $S_E(\omega)$ was found to agree better with experiment than $S_A(\omega)$.

Power and Zienau[2] later discussed the two forms of the interaction Hamiltonian for nonrelativistic quantum electrodynamics, using a unitary transformation to obtain the $-e\mathbf{r}\cdot\mathbf{E}$ form from the fundamental minimal coupling form. They attributed the difference between $S_A(\omega)$ and $S_E(\omega)$ to the unphysical, nonretarded "precursor" field associated with the use of the vector potential in the Coulomb gauge.[3] The problem was later taken up by Fried,[4] who argued that the $\mathbf{A}\cdot\mathbf{p}$ line shape could be brought into better agreement with $S_E(\omega)$ by including nonresonant background terms which are usually ignored. Davidovich and Nussenzveig[5] have argued that the line shape cannot be unambiguously defined without taking account of the excitation process, and that the two interactions do give different line shapes for the usual "Weisskopf-Wigner initial condition."

Although the two forms of interaction Hamiltonian are unitarily related, there are certain subtleties that can arise when comparing results of calculations. In particular, operators that are unitarily related by the Power-Zienau transformation will not necessarily have the same physical interpretation.[6] Nevertheless, it is usually not difficult to show by explicit calculation that the same physical predictions are obtained regardless of which of the two forms of interaction is used. Thus the Kramers-Heisenberg dispersion formula, for example, may be obtained from either interaction, provided the $\mathbf{A}^2$ term is included in the minimal coupling form.

In this paper we give a simple explanation for the origin of the two different line shapes, and show that the "correct" line shape $S_E(\omega)$ can, in fact, be obtained in a straightforward way from the minimal coupling form of interaction. For this purpose it is necessary to pay attention to assumptions about the sudden switching on of interactions. Furthermore, we show that the "incorrect" line shape $S_A(\omega)$ may also be obtained from the $-e\mathbf{r}\cdot\mathbf{E}$ interaction if we take sudden switchings on seriously.

Consider the minimal coupling Hamiltonian

$$H=p^2/2m+V(r)+H_{\rm field}-(e/mc)\mathbf{A}\cdot\mathbf{p}+(e^2/2mc^2)\mathbf{A}^2\ , \tag{1.3}$$

with

$$\mathbf{A}=\sum_{\mathbf{k},\lambda}\left[\frac{2\pi\hbar c^2}{\omega_k V}\right]^{1/2}(a_{\mathbf{k}\lambda}+a_{\mathbf{k}\lambda}^{\dagger})\hat{\mathbf{e}}_{\mathbf{k}\lambda} \tag{1.4}$$

in the standard notation in which $a_{\mathbf{k}\lambda}$ is the photon annihilation operator associated with the free-space mode with wave vector $\mathbf{k}$ and polarization index λ, and where V is the quantization volume. From the Heisenberg equation of motion for $a_{\mathbf{k}\lambda}(t)$ we obtain the formal solution

PHYSICAL REVIEW A VOLUME 39, NUMBER 3 FEBRUARY 1, 1989

Keldysh approximation, A^2, and strong-field ionization

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The Keldysh approximation as applied to above-threshold ionization (ATI) is compared with conventional (weak-field) perturbation theory and the Kroll-Watson theory of electron scattering in intense fields. Unlike the latter "nonperturbative" theory, the Keldysh approximation treats the effects of the field on the initial and final states differently, and this leads to a spurious contribution from the $\mathbf{A}^2$ part of the Hamiltonian in the $\mathbf{A}\cdot\mathbf{p}$ gauge. We attempt to clarify the role of the ponderomotive potential in ATI and the influence of the $\mathbf{A}^2$ term on the polarization dependence of the transition amplitude. The fundamental defect of the Keldysh approximation with regard to $\mathbf{A}^2$ is shown to be of little practical consequence for the experiments carried out *thus far*. However, it is shown that the Keldysh amplitude is not "exact" in the applied field, as sometimes claimed, and is best regarded as an ansatz rather than a leading term in a gauge-invariant perturbation expansion. A simple modification of the Keldysh theory is proposed.

I. INTRODUCTION

The Keldysh approximation[1-3] remains perhaps the principal theoretical model against which experimental data on above-threshold ionization (ATI) are compared.[4,5] Yet there is still controversy over the extent to which the Keldysh theory is "nonperturbative," and there do not appear to be any workable extensions of the usual lowest-order theory.

One purpose of this paper is to derive the Keldysh approximation in an elementary way based on the time-dependent Schrödinger equation in the form of coupled equations for probability amplitudes. It is shown that the usual lowest-order Keldysh approximation rests on the assumptions that (a) the probability amplitude of the initial bound state remains at all times near unity, and (b) the binding potential is sufficiently short ranged that its effect on the ionized electron is negligible.

We also attempt to clarify certain aspects of the Keldysh approximation connected with the "ponderomotive" or $\mathbf{A}^2$ term in the Hamiltonian, and with comparisons of above-threshold ionization in linearly and circularly polarized fields.

We compare the Keldysh approximation with the standard perturbation theory of multiphoton processes, and with the Kroll-Watson theory of multiphoton stimulated bremsstrahlung. A simple modification of the lowest-order theory is derived and applied in two model examples.

It is important to make clear just what we mean by the "Keldysh approximation." Reiss[3] has modified the original Keldysh approach by avoiding a high-multiphoton-order approximation made at the outset by Keldysh. In the "Keldysh-Reiss theory" there appears, in addition to the Keldysh parameter γ, an intensity parameter which Reiss denotes by z. Reiss uses the "velocity" $(\mathbf{A}\cdot\mathbf{p}+\mathbf{A}^2)$ rather than "displacement" $(\mathbf{r}\cdot\mathbf{E})$ form of the Hamiltonian used by Keldysh, with the consequence that the ponderomotive potential appears explicitly in various expressions, as discussed in this paper. The most tractable form of the modified theory, and the one that has been compared by Reiss and others with experiments, is the lowest-order approximation. Following what appears to have become a somewhat standard practice, we will refer to this lowest-order form of Reiss's modification of the Keldysh approach as "the Keldysh approximation."

In Sec. II a simple derivation of the Keldysh approximation to the transition amplitude is given. Section III is an attempt to clarify the role of the so-called ponderomotive shift in above-threshold ionization. In the present authors' opinion some of the confusion that has surrounded the ponderomotive shift is connected with an incorrect treatment of the $\mathbf{A}^2$ term in the Hamiltonian. It is shown explicitly that this term has no effect on transition amplitudes when semiclassical radiation theory and the dipole approximation are applicable, and that any shifts in the ATI peaks are simply Stark shifts. This agrees with results obtained by working from the outset with the $\mathbf{r}\cdot\mathbf{E}$ Hamiltonian. The derivation of the Keldysh approximation in Sec. II suggests a simple modification used in two model examples in Sec. IV. In Sec. V we compare the Keldysh approximation with standard perturbation theory and with the Kroll-Watson theory of electron scattering in intense fields. Section VI considers again the $\mathbf{A}^2$ term and the interpretation of ATI experiments with linearly and circularly polarized fields. Section VII is a summary of our conclusions.

II. DERIVATION OF KELDYSH TRANSITION AMPLITUDE

We begin by writing the state vector at time t as an expansion in eigenstates $|\phi\rangle$ of the unperturbed Hamiltonian $H_A = p^2/2m + V(r)$:

$$|\psi(t)\rangle = \sum_n a_n(t)|\phi_n\rangle + \int d^3k\, a_{\mathbf{k}}(t)|\phi_{\mathbf{k}}\rangle , \qquad (2.1)$$

where n and $\mathbf{k}$ label bound and continuum eigenstates, re-

RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 38, NUMBER 5 SEPTEMBER 1, 1988

Shortcomings of the Keldysh approximation

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A simple derivation of the Keldysh amplitude for the ionization of an atom by a strong field is presented using a perturbation expansion in the binding potential. It is shown that the "Keldysh approximation" is questionable under conditions of strong ionization, when there is substantial probability of removing the electron from its initial bound state. This is consistent with published numerical studies based on model binding potentials. It is argued that the Keldysh amplitude is not gauge invariant in the usual sense, and that it is effectively canceled in a conventional gauge-invariant formulation of strong-field perturbation theory.

The Keldysh theory of ionization by a strong oscillating field is characterized by the treatment of the detached electron as an otherwise free particle in the field.[1] The photoelectron is therefore described by a Volkov wave function, i.e., a solution of the Schrödinger equation for a charged particle in an external field.[2] Apart from its neglect of the Coulomb interaction of the electron with the residual ion, the Keldysh approximation is often regarded as a "nonperturbative" approach to strong-field atomic ionization.

In this paper the well-known form of the Keldysh transition amplitude involving an initial bound state and a final Volkov state is derived very simply using a perturbation expansion of the time evolution operator in the atomic binding potential $V(\mathbf{r})$. (As in much of the recent literature, the "Keldysh approximation" here refers to the lowest-order approximation to the formulation originally outlined by Keldysh.) In order to arrive at the Keldysh amplitude it is necessary to assume that the probability of the electron being removed from its initial state is small. The Keldysh approximation is therefore of questionable validity under conditions of strong ionization, when this probability is not small. This observation is consistent with *ab initio* numerical studies in which the Keldysh approximation has been found to be in serious disagreement, sometimes even qualitatively, with exact results.

Another difficulty with the Keldysh approximation is that it produces gauge-dependent results. This problem is discussed below in connection with a more conventional, *gauge-invariant* perturbation theory advocated by Antunes Neto and Davidovich.[3] This gauge-invariant approach is formally equivalent to that employed by Kroll and Watson[4] for the scattering of an electron in the presence of a strong field.

The Hamiltonian of interest has the form

$$H(t) = H_0(t) + V(\mathbf{r}) \tag{1a}$$

$$= p^2/2m + H_I(t) + V(\mathbf{r}) \tag{1b}$$

$$= H_A + H_I(t) , \tag{1c}$$

where $H_0(t)$ is the Hamiltonian for an electron in the applied field alone, V is the atomic binding potential, $H_I(t)$ is the interaction Hamiltonian for the coupling of the electron to the field [e.g., $H_I(t) = -e\mathbf{r}\cdot\mathbf{E}(t)$ or $H_I(t) = -(e/mc)\mathbf{A}(t)\cdot\mathbf{p} + (e^2/2mc^2)\mathbf{A}^2(t)$], and $H_A = p^2/2m + V$ is the unperturbed Hamiltonian for the bound electron in the absence of the applied field.

The transition amplitude of interest is

$$A_{fi}(t) = \langle f | U(t) | i \rangle , \tag{2}$$

where $|i\rangle$ is the initial (bound) state and $|f\rangle$ is a plane-wave state associated with a *free* electron of momentum $\mathbf{p}$. The time evolution operator $U(t)$, satisfying $i\partial U/\partial t = HU$, is given by

$$U(t) = U_0(t)u(t) , \tag{3}$$

where

$$i\partial U_0/\partial t = H_0 U_0 , \tag{4a}$$

$$i\partial u/\partial t = U_0^\dagger V U_0 u = U_0^\dagger V U , \tag{4b}$$

and $U_0(0) = u(0) = 1$. Thus

$$A_{fi}(t) = \langle f | U_0(t) | i \rangle - i\int_0^t dt' \langle f | U_0(t) U_0^\dagger(t') V U(t') | i \rangle , \tag{5}$$

$U(t')|i\rangle$ is the state to which $|i\rangle$ evolves after a time t'. If we assume that

$$U(t')|i\rangle \cong e^{iI_0 t'}|i\rangle \equiv |\psi_i(t')\rangle , \tag{6}$$

where I_0 is the ionization potential associated with state $|i\rangle$, then

$$A_{fi}(t) \cong \langle f | U_0(t) | i \rangle - i\int_0^t dt' \langle f | U_0(t) U_0^\dagger(t') V | \psi_i(t') \rangle . \tag{7}$$

The approximation (6) assumes that the probability of the electron leaving its initial bound state is negligible, and also that the level shift due to the applied field may be ignored.

Now since

$$i\frac{\partial}{\partial t'}|\psi_i(t')\rangle = [p^2/2m + V]|\psi_i(t')\rangle , \tag{8}$$

we have

Classical stimulated emission

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Received June 9, 1986; accepted September 25, 1986

Stimulated emission is formulated in completely classical terms and is shown to occur in general only in nonlinear systems. Our approach is based on the frequency-dependent susceptibility, which in both the classical and quantum-mechanical descriptions is the main characteristic determining whether there is absorption or stimulated emission. By using Born's correspondence rule, we derive an expression for the lowest-order quantum correction to the classical susceptibility.

1. INTRODUCTION

The concept of stimulated emission was introduced by Einstein in his derivation of the blackbody spectrum.[1] Because stimulated emission is crucial to the derivation of the Planck spectrum and because it is usually described in terms of a transition rate between quantum states, it is sometimes regarded as a distinctly quantum-mechanical effect. One purpose of this paper is to show that stimulated emission arises naturally also in purely classical systems. This point was made some time ago by Gaponov *et al.*,[2] and one of the present authors formulated the problem in semiclassical terms.[3] Partly because of the great current interest in various aspects of nonlinear dynamics, we have reexamined the role of stimulated emission in classical nonlinear systems.

We should emphasize straightaway just what we mean by stimulated emission. We consider a system in some force field and consider the change in energy of the driven system. We define stimulated emission simply as a decrease in the energy of the driven system, and we define absorption as an increase in the system's energy. We do not specify what physical field is being amplified or absorbed. In the familiar case of stimulated emission of radiation, the stimulated emission can be understood physically as a constructive interference between the incident field and the field scattered by the atom.[4]

For our purposes it is convenient to deal with the frequency-dependent susceptibility, which provides a framework for both classical and quantum calculations of stimulated emission. Consider, for instance, some system subjected to an electric field $E(t)$, or some other external force, such that the Hamiltonian for the system takes the form

$$\hat{H} = \hat{H}_0(\hat{x}, \hat{p}) - e\hat{x}E(t). \tag{1.1}$$

[We write quantum-mechanical operators with a caret (^) in order to distinguish them from classical variables.] To find the change of internal energy of the system we use the relation

$$\frac{d\hat{H}_0}{dt} = (i/\hbar)[\hat{H}, \hat{H}_0] = (ie/\hbar)[\hat{H}_0, \hat{x}]E(t) = e\dot{\hat{x}}E(t) \tag{1.2}$$

for the quantum-mechanical operators and

$$\frac{dH_0}{dt} = |H, H_0| = e\dot{x}E(t) \tag{1.3}$$

in the classical case, where $|H, H_0|$ is a Poisson bracket. Equation (1.2) leads to an expectation value that has exactly the same form as the classical equation [Eq. (1.3)], because the interaction of the system with the external force has been taken to be linear in the system variables. (The force itself is treated as classically prescribed.) Therefore it is convenient for the present to let x and H_0 denote either quantum-mechanical operators or purely classical quantities.

The velocity $\dot{x}$ may be divided into two parts,

$$\dot{x} = \dot{x}_0 + \dot{x}_1, \tag{1.4}$$

where $\dot{x}_1$ is the part induced by the external force

$$e\dot{x}_1 = \frac{d}{dt}[\chi(\omega)E(\omega)e^{-i\omega t} + \chi(-\omega)E(-\omega)e^{i\omega t}]. \tag{1.5}$$

Here $\chi(\omega)$ is a susceptibility describing the response of the system to the external force:

$$x(t) = x(\omega)e^{-i\omega t} + x(-\omega)e^{i\omega t}, \tag{1.6a}$$

$$ex(\omega) = \chi(\omega)E(\omega). \tag{1.6b}$$

(In general, of course, there will be a summation or integration over frequencies ω, as in the following sections.) The sign of the right-hand side of Eq. (1.2) or (1.3) determines whether there is absorption of energy from the applied field or stimulated emission, in which case the system loses energy; that is, if $dH_0/dt > 0$, there is absorption, whereas if $dH_0/dt < 0$, there is stimulated emission.

Consider the work associated with the free part of the velocity. For simplicity we suppose that the unperturbed motion of the system is just a sinusoidal oscillation at the frequency ω_0, i.e.,

$$x_0(t) = x_0e^{-i\omega_0 t} + x_0^*e^{i\omega_0 t}, \tag{1.7}$$

so that

0740-3224/87/010078-08$02.00

PHYSICAL REVIEW A VOLUME 33, NUMBER 5 MAY 1986

Solitons and four-wave mixing

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We develop the formalism for studying transient four-wave-mixing phenomena in a Raman medium, and apply it to the study of solitary-wave propagation. We consider the problems of second-Stokes generation, anti-Stokes scattering, and two-pump Raman scattering.

I. INTRODUCTION

Over the past few years the field of Raman solitons has received much attention following the observation of these solitons in a nondispersive medium by Carlsten, Wenzel, and Drühl in 1983.[1] Raman solitons were first discussed by Chu and Scott, Tan-no *et al.*, and Makhviladze *et al.* in 1975 and more recently by Kaup, Steudel, and Meinel.[2–4] Other work on Raman solitons has been done by Makhviladze and Sarychev,[5] who looked at soliton stability and dispersive effects, and by Hasegawa,[6] who considered propagation in optical fibers where dispersive effects are very strong.

Historically, these solitons were first observed to appear at random, roughly every twentieth shot, during Raman scattering experiments performed by Wenzel and Carlsten at Los Alamos.[7] In discussing this phenomenon with Drühl they learned about the work of Chu and Scott,[2] which showed that the pump was a hyperbolic secant function and the first-Stokes a hyperbolic tangent function. Since the hyperbolic tangent function is reminiscent of a function which simply changes sign or undergoes a π phase flip, Drühl in his computer model simply inserted this phase flip in the electric field envelope of the first-Stokes seed. The output of computer simulations showed that a soliton could be generated and located temporally so as to reproduce the experimental results. The next question asked was "Why did the soliton appear in the experiments at all?," since no effort was made to induce the soliton's formation. While the answer to this question is still under speculation, these solitons are now consistently generated experimentally by introducing a π phase shift in the first-Stokes seed pulse's temporal profile using a Pockels cell. Now that a reliable technique for generating these Raman solitons exists, new studies of the subtle details of soliton formation are being made both theoretically and experimentally.[8]

In this paper we will generalize the theoretical work on Raman solitons to include all four-wave-mixing phenomena which occur in a Raman medium. We plan to study three-field processes, i.e., second-Stokes generation and anti-Stokes scattering, and four-field processes, i.e., two-pump conversion. In Sec. II the equations of motion will be derived. In Sec. III we will discuss the essential physics of the four-wave-mixing process and the analytic calculation of mathematical soliton solutions. In Sec. IV computer generated simulations of these soliton processes will be shown. A summary will be presented in Sec. V.

There is one caveat to this work which must be understood before reading the paper. We are interested in the essential physics of four-wave-mixing processes in a Raman medium and soliton formation. Because of this goal we will not pursue fine details of the physical processes involved which are or may be relevant to the real world, but do not affect the fundamental physics of the process. Some examples of these details are complex field envelopes, gain parameters, field frequencies, bandwidths, field intensities, transverse beam profiles, dispersion, off-resonant detuning, etc.

II. EQUATIONS OF MOTION

A. Material medium

The essential physics of a Raman medium requires the existence of at least three energy states (0,1,2) where the highest-energy state (1) is electric-dipole coupled to the remaining two states (0,2). Only the ground state (0) is initially populated. For simplicity, we will assume the two transition dipole moments are equal ($\mu_{01}=\mu_{12}$) and equal to μ. The energy of state (1) (in frequency units) is ω_{01}. The energy of state (2) is Δ, and state (0), zero. In Fig. 1 we show this three-state system.

Since all the electric fields in the four-wave-mixing problems under consideration, i.e., pump, first-Stokes, second-Stokes, ..., first-anti-Stokes, etc., are potentially capable of coupling states (0-1) and (1-2), we will represent all of them generically by a single electric field $\mathbf{E}(z,t)$, where we are considering plane-wave propagation in the $\hat{\mathbf{z}}$ direction. Using the notation that n refers to a specific field, i.e., first-anti-Stokes, pump, first-Stokes, etc., we can define

$$\mathbf{E}(z,t)\equiv\tfrac{1}{2}\sum_n [\mathscr{E}_n(z,t)e^{i(k_n z-\omega_n t)} + \mathscr{E}_n^*(z,t)e^{-i(k_n z-\omega_n t)}] . \tag{2.1}$$

The quantities $\mathscr{E}_n$, k_n, and ω_n are the electric field en-

NONLINEAR OSCILLATIONS AND CHAOS IN n-InSb

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Received 18 January 1985; accepted for publication 13 February 1985

We present evidence for chaotic behavior in n-InSb. The Hall voltage exhibits a period-doubling route to chaos as the (non-ohmic) dc current is increased. The nonlinear oscillation and bifurcation processes are strongly influenced by irradiation with CO_2 laser radiation.

There is much interest in deterministic chaos, especially with the relatively recent realization that (a) there are certain well-defined and "universal" routes to chaos in dissipative systems, and (b) chaos can appear in low-dimensional dynamical systems (systems of few first-order autonomous differential equations). The signature of chaotic behavior is the property of very sensitive dependence on initial conditions, a property associated with a positive Lyapunov exponent. Chaotic dynamical systems also have a broadband component in the power spectra of their dynamical variables, reflecting their aperiodic evolution in time. Chaos has been observed in a variety of physical systems, including fluids [1], lasers [2], Josephson junctions [3], and semiconductors [4–6]. In this letter we report evidence of chaos in the electrical properties of n-InSb [‡1], a well-known narrow gap semiconductor.

High-purity tellurium-doped n-InSb is known to exhibit nonlinear electrical characteristics near 4 K with a small applied magnetic field [7]. This nonlinear behavior is a consequence of the small donor activation energy, which leads to impurity impact ionization at quite small currents. Under these conditions, both the Hall coefficient and resistivity exhibit an oscillatory behavior as the dc current is increased. In fig. 1 we

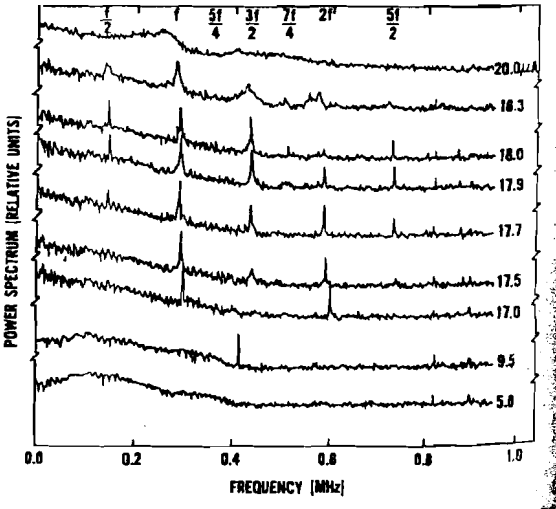

Fig. 1. Power spectra of the Hall voltage at 10.7 kG and 1.9 K. Two period doublings are exhibited, one at ≈17.5 μA and the other at ≈17.9–18.0 μA. Chaotic behavior at ≈18.3 μA is indicated by the increased level of broadband noise and the increasing amount of aperiodicity. The structure at 0.82 MHz arises from a Fort Worth radio station.

‡1 A brief account of this work was presented at the XVII International Conference on the Physics of Semiconductors, San Francisco, California, 6–10 August 1984.

0.375-9601/85/$ 03.30

PHYSICAL REVIEW A VOLUME 31, NUMBER 6 JUNE 1985

Time dependence of photon correlations in a three-level atomic cascade

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(Received 22 August 1984)

The question of the temporal correlation of photons arising from a three-level atomic cascade is considered by calculating the joint probability of photoelectric detection for two detectors placed in the far field of the atomic emission. The analysis is carried out in the Heisenberg picture and involves the use of dipole, adiabatic (Weisskopf-Wigner), and rotating-wave approximations. Except for a small interval about zero time delay, which is however too short to be detected, it is found that the temporal correlation between emitted photons is one of sequential but otherwise statistically independent events, even when Landau's generalized decay and frequency-shift terms are included. This result is compared with a well-known expression for the joint spectral density of the emitted photons.

I. INTRODUCTION

The measurement of the correlation of photons emitted in an atomic cascade is an accepted technique for determining lifetimes of excited atomic levels.[1–4] For an arrangement of levels such as shown in Fig. 1, with level 3 initially excited, the lifetime of level 2 is found by recording the distribution of time delays between an initial detection arising from the $3\rightarrow2$ transition and a subsequent detection from the $2\rightarrow1$ decay. Implicit in this scheme is the assumption that the emission of the first photon projects the atom into level 2 with absolute certainty and that there are no residual correlations between successive photons. While these assumptions are certainly plausible, there appears to be in the literature no corresponding calculation of the actual joint probability for photoelectric detection.

Another important application of the measurement of photon correlations is found in tests of Bell inequalities in studies of polarization correlations for photons emitted in an atomic cascade. Extensive reviews of this work are available.[5–7] Generally one is interested in the coincidence rate as a function of the relative polarization of emitted pairs of photons from a cascade. The actual time dependence of the joint detection probability may be recorded, or more simply the number of coincidences may be obtained by employing a relatively wide time window triggered by the initial event. In the former case the joint detection probability as a function of time delay between events is well described by a decaying exponential of time constant given by the lifetime of the intermediate state.[8] In either case quantum-mechanical predictions violating Bell inequalities do not depend upon the dynamics of the photon emissions but rather upon consideration of the angular momenta of initial and final states, and on the prescription chosen for extracting probabilities from a two-particle state.

Treatments that do yield dynamical information about the intensity correlation of the field emitted in a cascade have been given by Loudon[9] and by Aspect *et al.*[6(b)] For the case of continuous but incoherent excitation of level 3, Loudon[9] uses rate equations together with the quantum regression theorem to show that the decay $3\rightarrow2\rightarrow1$ proceeds in a sequential but otherwise uncorrelated fashion, in agreement with the numerous observations quoted above. Scattering theory has been employed by Aspect *et al.*[6(b)] to analyze the photon correlations in the excitation and decay of a cascade, including the role of Zeeman levels. Agarwal and Jha[11] treat the related problem of resonant Raman scattering.

A different perspective on the question of photon correlations is provided by a calculation of the joint spectral density $J(\omega,\omega')$ from the decay sequence $3\rightarrow2\rightarrow1$ for an atom initially excited to state 3. Weisskopf and Wigner[10] and others[11,12] have considered this problem and find

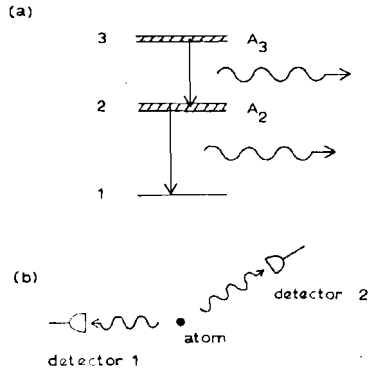

FIG. 1. (a) Level scheme for the atomic cascade $3\rightarrow2\rightarrow1$. The transition $3\rightarrow1$ is forbidden; A_2 and A_3 are the Einstein A coefficients for the levels 3 and 2, respectively. (b) Detectors in the far field of the atomic radiation for registering the joint probability of photoelectric detection at points (r_1,t_1) and (r_2,t_2).

notoJournalTitle: PHYSICS REPORTS-REVIEW SECTION OF PHYSICS LETTERS
notoJournalVolume: 128
notoJournalIssue: 4-5
onth:
ear: 1985
ages: 205 only
t e Author: ACKERHALT,
t e Title: CHAOS IN QUANTUM OPTICS

CHAOS IN QUANTUM OPTICS

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NORTH-HOLLAND-AMSTERDAM

130 J. Opt. Soc. Am. B/Vol. 2, No. 1/January 1985

Modeling laser instabilities and chaos

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Received June 28, 1984; accepted August 17, 1984

We summarize some previous work on the analysis of laser instabilities and chaos and present new results on routes to chaos and field-correlation functions.

1. INTRODUCTION

In 1963, Lorenz[1] presented a detailed study of the set of three differential equations now known as the Lorenz model. For certain ranges of parameters, the Lorenz system is chaotic, i.e., it has nonperiodic time evolution with sensitive dependence on initial conditions.[2] In 1975, Haken[3] showed that the Lorenz system has the same mathematical form as the Maxwell–Bloch equations describing a homogeneously broadened, single-mode, on-resonance laser medium[4]:

$$\dot{v} = -\beta v + \frac{d}{\hbar}\epsilon w, \tag{1.1a}$$

$$\dot{w} = -\gamma(w - w_0) - \frac{d}{\hbar}\epsilon v, \tag{1.1b}$$

$$\dot{\epsilon} = -\gamma_c \epsilon + (2\pi N d\omega) v. \tag{1.1c}$$

This observation showed that chaotic behavior might occur in lasers. Indeed, at about the same time as Lorenz, Buley and Cummings[5] made a numerical study of essentially the same system of equations and remarked that "A case has ... been run ... in which the output [intensity] ... appears as a series of almost random spikes." It seems likely that they were seeing Lorenz-model chaos.

Shirley[6] in 1968 noted that the rate-equation approximation to Eq. (1.1), i.e.,[4]

$$\dot{w} = -\gamma(w - w_0) - \frac{8\pi d^2}{\beta c \hbar^2} I w, \tag{1.2a}$$

$$\dot{I} = -2\gamma_c I + \frac{4\pi N d^2 \omega}{\hbar} I w, \qquad I = \frac{c}{8\pi}\epsilon^2, \tag{1.2b}$$

predicts only stable steady-state solutions, whereas Eq. (1.1) permits undamped spiking of the field. He derived the same conditions for instability that are now well known for the Lorenz model. These conditions are possible to meet in real lasers, but they are atypical, and, in particular, they require not only a bad cavity ($\gamma_c > \beta$) but also a pumping level at least nine times above threshold.[7–9]

Recently, an important advance was made by Casperson,[10] who showed that instability of the steady-state solution of the single-mode laser equations is much easier to realize in the case of *inhomogeneous* broadening. The Casperson instability still requires a bad cavity but not the high pumping level needed for single-mode instability in the homogeneous case. Although an instability (of a constant steady-state solution) does not necessarily imply chaos, Casperson found in both numerical and actual experiments with bad cavities that in some cases there were chaotic pulsations of the intensity. As far as we know, he did not characterize this chaos further or look for a well-defined route to it; in at least one case, however, he did observe one period doubling of orderly pulsations.

We consider in Section 2 the Maxwell–Bloch equations [Eqs. (1.1)] but generalized to permit inhomogeneous (Doppler) broadening, off-line operation, and different relaxation rates of upper and lower levels of the laser transition. We summarize the results of extensive numerical experiments[11] and discuss their comparison with the actual single-mode experiments of Abraham *et al.*[12,13] In Section 3, we consider a coupled-mode model with homogeneous broadening and conclude that chaos can occur as a result of spatial inhomogeneity of the pump[13]; we note that there is a period-doubling route to chaos. In Section 4, we consider (classical) first- and second-order field-correlation functions, and, in Section 5, we summarize our conclusions.

2. CHAOS IN THE MAXWELL–BLOCH SYSTEM

Consider the following Maxwell–Bloch equations describing a single-mode unidirectional ring laser:

$$\dot{u} = -(\Delta - \dot{\phi} - ks)v - \beta u, \tag{2.1a}$$

$$\dot{v} = (\Delta - \dot{\phi} - ks)u - \beta v + \Omega(z_2 - z_1), \tag{2.1b}$$

$$\dot{z}_2 = R_2 - \gamma_2 z_2 - \tfrac{1}{2}\Omega v, \tag{2.1c}$$

$$\dot{z}_1 = R_1 - \gamma_1 z_1 + \tfrac{1}{2}\Omega v, \tag{2.1d}$$

$$\dot{\Omega} = -\gamma_c \Omega + A\int_{-\infty}^{\infty} ds W(s) v(s,t), \tag{2.1e}$$

$$\dot{\phi}\Omega = -A\int_{-\infty}^{\infty} ds W(s) u(s,t). \tag{2.1f}$$

We have written these equations in terms of the Rabi frequency $\Omega = d\epsilon/\hbar$, with the electric field given by

$$E(z,t) = \epsilon(t)\exp\{i[kz - \omega t + \phi(t)]\}, \tag{2.2}$$

0740-3224/85/010130-07$02.00

ROUTES TO CHAOS IN THE MAXWELL-BLOCH EQUATIONS

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Received 22 August 1984
Revised manuscript received 6 December 1984

We consider the single-mode Maxwell-Bloch equations describing a Doppler-broadening ring laser. For different parameter ranges this system undergoes period-doubling, intermittency, and two-frequency routes to chaos as a single parameter (e.g., detuning) is varied, as has been observed in recent experiments with He-Ne and He-Xe lasers.

The Maxwell-Bloch equations describe the resonant interaction of light with matter [1]. In various forms they describe a large variety of interesting nonlinear phenomena, such as soliton propagation (self-induced transparency) [1–3], photon echoes [1], and optical bistability [4]. They also form the basis of the semiclassical theory of the laser [5]. In this letter we present evidence showing that under certain conditions the Maxwell-Bloch system is deterministically chaotic, and displays "universal" routes to chaos that are of current interest in dynamical systems theory. In addition to the rich variety of behavior already known for the Maxwell-Bloch equations, these results are of interest in connection with recent observations of universal routes to chaos in He-Ne and He-Xe lasers [6,7]. We believe this is the first instance where a physically relevant set of equations for a realistic number of parameters is both tractable and exhibits all the "universal" routes to chaos. In fluid flow all of these routes to chaos have also been observed; however, the dynamical system of equations is practically intractable.

We consider Maxwell-Bloch equations under the simplifying assumption that the spatial dependence of the atomic and field variables may be ignored. The resulting equations describe, within certain approximations (e.g., slowly-varying amplitudes, plane-wave field) a unidirectional ring laser:

$$\dot{x} = -(\Delta - \dot{\phi} - kv)y - \beta x \,, \tag{1}$$

$$\dot{y} = -(\Delta - \dot{\phi} - kv)x - \beta y + \Omega(z_2 - z_1) \,, \tag{2}$$

$$\dot{z}_2 = R_2 - \gamma_2 z_2 - \tfrac{1}{2}\Omega y \,, \tag{3}$$

$$\dot{z}_1 = R_1 - \gamma_1 z_1 + \tfrac{1}{2}\Omega y \,, \tag{4}$$

$$\dot{\Omega} = -\gamma_c \Omega + \frac{2\pi N d^2 \omega}{\hbar} \int_{-\infty}^{\infty} \mathrm{d}v\, W(v) y(v,t) \,, \tag{5}$$

$$\dot{\phi}\Omega = -\frac{2\pi N d^2 \omega}{\hbar} \int_{-\infty}^{\infty} \mathrm{d}v\, W(v) x(v,t) \,. \tag{6}$$

Here $\Omega = d\epsilon/\hbar$ is the Rabi frequency [1]: z_2 and z_1 are the upper- and lower-state occupation probabilities; x and y are the in-phase and in-quadrature (with the field) parts of the expectation value of the induced dipole moment; kv is the Doppler shift for an atom with velocity v along the optical axis, with $W(v)$ the

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PHYSICAL REVIEW D VOLUME 32, NUMBER 4 15 AUGUST 1985

Comment on "How to make quantum mechanics look like a hidden-variable theory and vice versa"

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The hidden-variable theory considered by Scully is discussed in the context of photon polarization correlations.

Bell's work shows that no local hidden-variable theory can be expected to be in full agreement with quantum mechanics, even if the hidden variables are stochastic.[1] It leaves open, however, the possibility of a nonlocal hidden-variable theory. A nonlocal hidden-variable theory requires action at a distance, or some other drastic departure from present concepts of space-time. It would admit an objective physical reality "in the way which Einstein would have liked least!"[2]

It can perhaps be argued that nonlocal hidden-variable theories should not yet be abandoned altogether. Scully[3] has considered a "new type" of nonlocal hidden-variable theory, partly to "demonstrate a much closer correspondence between quantum mechanics and certain hidden-variable theories than was previously appreciated." The theory, motivated by a quantum distribution function, is put forth for the Einstein-Podolsky-Rosen-type, two-particle spin-$\frac{1}{2}$ correlation.[4] Here this theory is considered in the context of experimentally testable photon-polarization correlations.

To paraphrase Scully's argument for the case of photon polarization, we assume there is a hidden variable α that determines the polarization, such that $\cos^2(\theta-\alpha)$ is the "likelihood"[3] that the photon will pass through a polarizer oriented at an angle θ to the vertical. In a $J=0\rightarrow 1\rightarrow 0$ atomic cascade decay, we are interested in the probability $\tilde{P}(\theta,\phi)$ of counting two photons when the two polarizers are at angles θ and ϕ,[5] and write

$$\tilde{P}(\theta,\phi)=\int d\alpha \int d\beta \cos^2(\theta-\alpha)\cos^2(\phi-\beta)P(\alpha,\beta)\ , \quad (1)$$

where $P(\alpha,\beta)$ is the distribution function for the hidden variables imagined for the two photons. By analogy with Scully's Eq. (4.5) for the spin-$\frac{1}{2}$ case, and knowing the corresponding "quantum distribution function," we are motivated to consider

$$P(\alpha,\beta)=\delta(\alpha-\beta)\delta(\alpha-\theta)\ , \quad (2)$$

which gives

$$\tilde{P}(\theta,\phi)=\cos^2(\theta-\phi)\ , \quad (3)$$

in agreement with quantum mechanics.[6]

Let us emphasize in simple terms the meaning of (1) and (2). In writing (1) we are assuming that the photon polarizations have objectively real values "out there," determined by the hidden variables α and β, and $\tilde{P}(\theta,\phi)$ depends on how these hidden variables are distributed. Equation (2) asserts that these hidden variables are correlated in a particular way, namely, that the two photons are linearly polarized in the same direction; this is the meaning of the factor $\delta(\alpha-\beta)$. The second factor, $\delta(\alpha-\beta)$, says that one of the photons is polarized in exactly the direction in which the experimenter orients his polarizer. The atom somehow emits two photons that happen to be polarized in precisely the direction of one of the experimenter's analyzers.

The model thus asserts that the emitter produces two photons with polarization determined by the whim of the experimenter. Unless this assertion can be made plausible, the model would appear to be nothing more than a concoction for obtaining the quantum-mechanical prediction (3).

It might be supposed that the atom is only *allowed* to emit photons polarized along the direction of one of the polarizers, based on the knowledge that an atom in a closed cavity can only emit into one of the cavity modes. Aside from the weakness of the analogy, this supposition is testable, for the polarizers cannot be expected to affect the emission if they are farther from the atom than c times a radiative lifetime τ. Experimentally, we would expect (3) to become inapplicable, based on the hidden-variable model, if the atom-polarizer separation is considerably larger than the coherence length $c\tau$ of, say, the lower transition of the cascade. In the experiments of Aspect, Grangier, and Roger[7] the polarizers were as much as 6.5 m from the emitting atoms, compared with a coherence length $c\tau$ of only 1.5 m. The joint detection rate was nevertheless in excellent agreement with quantum mechanics.

In another experiment of Aspect, Dalibard, and Roger[8] furthermore, the polarizer orientations were effectively varying rapidly in time while the photons were "in flight." Again the results were in good agreement with quantum mechanics.

While polarizer-determined emission is not the only conceivable mechanism for fixing the polarizations in Scully's "HV_3", it seems, at least to the present author, that other possibilities are even less palatable.

It may be worthwhile to make a connection with a local hidden-variable model in which the two photons have the same polarization, but this being a uniformly distributed random variable:

$$P(\alpha,\beta)=\frac{1}{2\pi}\delta(\alpha-\beta)\ . \quad (4)$$

In this case (1) yields

$$\tilde{P}(\theta,\alpha)=\tfrac{1}{4}[\tfrac{1}{2}+\cos^2(\theta-\phi)]\ . \quad (5)$$

Equation (5) implies a minimum-to-maximum counting ratio of $\frac{1}{3}$ as $\theta-\phi$ is varied. This agrees with the prediction of semiclassical radiation theory derived by Clauser.[6]

Scully's calculations seem useful because they show how strange a hidden-variable theory would have to be to agree, even partially, with quantum mechanics.

Interaction Hamiltonian of quantum optics

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We consider the fully quantum-mechanical Hamiltonian for the interaction of light with bound electrons. There are, roughly speaking, two different viewpoints relating the minimal-coupling and electric-dipole forms of the Hamiltonian. We attempt to clarify the situation by showing that either viewpoint is justified. As an example we discuss the Maxwell–Bloch equations and show that the usual derivation of these equations is somewhat misleading. In addition, we find that the Maxwell–Bloch equations appearing in standard texts are inconsistent.

1. INTRODUCTION

The fundamental minimal-coupling form of the Hamiltonian for the interaction of light with a bound electron is

$$H = \frac{1}{2m}\left(\mathbf{p} - \frac{e}{c}\mathbf{A}\right)^2 + V(r) + \frac{1}{8\pi}\int d^3x(\mathbf{E}^{\perp 2} + \mathbf{B}^2)$$
$$= H_{\mathrm{ATOM}} + H_{\mathrm{FIELD}} - \frac{e}{2mc}(\mathbf{A}\cdot\mathbf{p} + \mathbf{p}\cdot\mathbf{A}) + \frac{e^2}{2mc^2}\mathbf{A}^2. \tag{1.1}$$

$\mathbf{E}^{\perp}$ denotes the transverse part of the electric field:

$$\mathbf{E}^{\perp} = -\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t}. \tag{1.2}$$

(We will use the Coulomb gauge, $\nabla\cdot\mathbf{A} = 0$, throughout this paper.) $V(r)$ is the potential binding the electron, the electromagnetic part of which arises from the longitudinal part of $\mathbf{E}$, e.g., the Coulomb field of the nucleus. Since all the explicit field variables are transverse, we will henceforth drop the $\perp$ label, except on specified atomic distributions.

The Hamiltonian [Eq. (1.1)] is used because it gives the correct Heaviside–Lorentz force law for the electron, together with the correct Maxwell equations for the field in the presence of an electron. These equations of motion are valid not only as classical equations but also as Heisenberg operator equations. All electron and field variables in this paper are quantum-mechanical operators in the Heisenberg picture. For simplicity we work within the electric-dipole approximation.

The electric-dipole Hamiltonian

$$H = H_{\mathrm{ATOM}} + H_{\mathrm{FIELD}} - e\mathbf{r}\cdot\mathbf{E} \tag{1.3}$$

is frequently used instead of Eq. (1.1) because it is more convenient in some respects.[1-3] During the past decade there has been considerable controversy regarding the relation between Eqs. (1.1) and (1.3). There appear to be two principal schools of thought on this question, and in this paper we argue that both viewpoints are justified, neither being more nearly correct than the other. However, Eq. (1.3) as it stands is not complete.

In Sections 2 and 3 we derive from Eq. (1.1) two *different* Hamiltonians of the electric-dipole type [Eq. (1.3)]. These Hamiltonians appear at first glance to be the same, but they are in fact different, and this, in our view, is responsible for some of the confusion and controversy surrounding Eq. (1.3). Both Hamiltonians are valid and give rise to precisely the same physical predictions when treated correctly. In Section 4 we discuss the Hamiltonian [Eq. (1.3)] and its validity. In Section 5 we derive Maxwell–Bloch equations from Eq. (1.1) and discuss the analogous set of equations obtained from the electric-dipole Hamiltonian. We summarize our conclusions in Section 6.

2. TRANSFORMATION OF THE MINIMAL-COUPLING HAMILTONIAN

Let us denote the minimal-coupling Hamiltonian [Eq. (1.1)] and wave functions by H_{OLD} and $|\psi_{\mathrm{OLD}}\rangle$ and write the Schrödinger equation represented by Eq. (1.1) as

$$i\hbar\frac{\partial}{\partial t}|\psi_{\mathrm{OLD}}\rangle = H_{\mathrm{OLD}}|\psi_{\mathrm{OLD}}\rangle. \tag{2.1}$$

We can obtain equivalent dynamics for this physical system by applying a unitary transformation U and making the identifications

$$|\psi_{\mathrm{NEW}}\rangle = U^{+}|\psi_{\mathrm{OLD}}\rangle \tag{2.2}$$

and

$$H_{\mathrm{NEW}} = U^{+}H_{\mathrm{OLD}}U. \tag{2.3}$$

We obtain

$$i\hbar\frac{\partial}{\partial t}(U|\psi_{\mathrm{NEW}}\rangle) = UH_{\mathrm{NEW}}|\psi_{\mathrm{NEW}}\rangle, \tag{2.4}$$

which, by simple manipulation, becomes

$$i\hbar\frac{\partial}{\partial t}|\psi_{\mathrm{NEW}}\rangle = \left(H_{\mathrm{NEW}} - i\hbar U^{+}\frac{\partial}{\partial t}U\right)|\psi_{\mathrm{NEW}}\rangle. \tag{2.5}$$

0740-3224/84/010116-05$02.00

IL NUOVO CIMENTO Vol. 83 B, N. 1 11 Settembre 1984

Are Quantum Waves Observable?

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(ricevuto il 2 Novembre 1983; manoscritto revisionato ricevuto il 30 Maggio 1984)

Summary. — An experiment has been proposed to test whether an objective physical reality can be granted to probability amplitudes. We consider an idealized version of the experiment and show that quantum mechanics predicts a null result.

PACS. 03.65. – Quantum theory; quantum mechanics.

1. – Introduction.

The probability amplitude represents perhaps the most striking departure of quantum theory from classical physics. This is seen convincingly in the two-slit experiment, which in a sense « contains the *only* mystery » of quantum mechanics (1). The two-slit experiment shows how the concept of a probability amplitude resolves the paradox of wave-particle duality.

In what sense, if any, can an objective physical reality be attached to the probability amplitude? This question has recently been raised by SELLERI (2),

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(1) R. P. FEYNMAN, R. B. LEIGHTON and M. SANDS: *The Feynamn Lectures on Physics*, Vol. 3 (Addison-Wesley, Reading, Mass., 1965), p. 1.

(2) F. SELLERI: *Ann. Fond. L. de Broglie*, 7, 45 (1982); F. SELLERI: *Found. Phys.*, 12, 1087 (1982); *Wave-Particle Dualism*, edited by S. DINER, D. FARGUE, G. LOCHAK and F. SELLERI (Reidel, Dordrecht, Holland, 1983).

Concentrated and illuminating: 25 years of lasers

Masers and Lasers: An Historical Approach

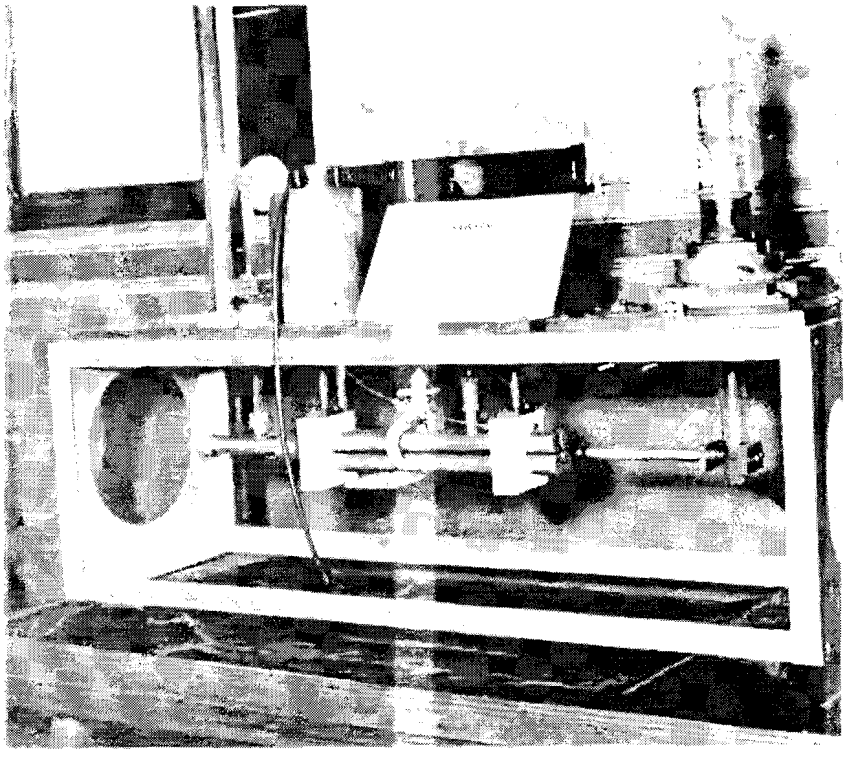
M. Bertolotti
258 pp. Hilger (US dist. Heyden), Philadelphia), 1983. $30.00

Reviewed by Peter W. Milonni

The construction of the first laser nearly a quarter of a century ago marked a turning point in the science and use of light. Mario Bertolotti's book traces the conceptual developments leading to masers and lasers. It is partly history, partly an introduction to the theory of operation of these devices.

Optical dispersion theory and the understanding of stimulated emission as "negative dispersion," played an important role in the formulation of quantum theory, especially in connection with the correspondence principle and matrix mechanics. A brief but lucid chapter ("Stimulated emission. Could the laser have been built more than 50 years ago?") is devoted to elementary dispersion theory and the experiments of Rudolf Ladenburg and others in the 1920s and early 1930s. It is followed by an intermezzo on magnetic resonance and optical pumping—Bloch equations, spin echoes and the Overhauser effect—and experimental evidence of population inversion. This work was concerned largely with the manipulation of energy-level populations, and in a sense the laser extends such manipulations to shorter wavelengths.

Research on the interaction of microwave radiation with matter in the 1940s and 1950s grew out of wartime radar research. A chapter on the maser begins with "The first published description of the maser principle without a working device" by Joseph Weber in 1952. In the spring of 1951 the operating principles of practical masers were worked out one morning by Charles Townes on a park bench in Washington. Townes was perhaps the first to understand the need for a resonant cavity. By 1953 the ammonia maser was built at Columbia by Townes's group; the acronym came with help from students when the group was unsuccessful in inventing a Latin or Greek name for the device. ("Skeptics read it as a Means of Acquiring Support for Expensive Research.") Independently Nikolai Basov and Alexander Prokhorov were at work in the Soviet Union, and just a few months later Basov in his doctoral research built the first Russian maser.



The original maser, built by Charles H. Townes (on display at the Franklin Institute), began a new epoch in quantum electronics. (Photo by J. Barton, courtesy AIP Niels Bohr Library.)

One of the major problems in extending the maser principle to the visible was that a closed cavity of reasonable size would permit far too many modes to oscillate simultaneously. Arthur Schawlow had used a Fabry–Perot interferometer in his graduate studies in spectroscopy, and it occurred to him around the end of 1957 that a Fabry–Perot could be used as a mode-selective laser resonator. Earlier (1956) Robert Dicke had also suggested a Fabry–Perot, and he obtained a patent on the resonator in 1958. Schawlow and Townes described the basic theory of laser operation, and some possible pumping schemes, in an article published in December of 1958. (Townes at first could not arouse the interest of the patent office at Bell Laboratories, because "the invention had little bearing on Bell System interests.") Gordon Gould, a graduate student at Columbia, was thinking along somewhat similar lines but did not publish. Bertolotti also discusses Dicke's idea for a "coherence-brightened laser" based on cooperative spontaneous emission and the historically interesting work of V. A. Fabrikant in Russia, going back to as early as 1939.

A chapter on "The Laser. Further

Peter W. Milonni, associate professor of physics at the University of Arkansas, does work in quantum optics and electrodynamics.

WAVE-PARTICLE DUALITY OF LIGHT: A CURRENT PERSPECTIVE

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ABSTRACT.

Ideas of wave-particle duality of light are traced through various stages, from Einstein to quantum optics and recent developments in the conceptual foundations of quantum mechanics.

I. INTRODUCTION

One of the remarkable things about quantum mechanics is the extent to which people disagree on what the theory says about physical reality. Even the founding fathers of wave-particle duality, Albert Einstein and Louis de Broglie, ultimately rejected the prevalent (Copenhagen) interpretation of quantum mechanics, as did Schrödinger. Perhaps "if one percent of the effort spent on physics were devoted to clarification, we could soon teach the basic concepts of quantum mechanics to the general run of nine-year-olds!"(1) At the present time, however, it seems safe to say, without too much exaggeration, that "...nobody understands quantum mechanics." (2)

The most positivistic advocates of the Copenhagen interpretation find the notion of an objective physical reality to be irrelevant and obsolete. Those of us of an Einsteinian mind-set are of course encouraged by the fact that such great scientists as de Broglie reject the Copenhagen philosophy. However, we must admit that recent evidence points against a "naive realism." In any case these questions will be debated for a long time to come, and new ideas may be encouraged by recording the thoughts of many people. In this vein I hope this survey will not be entirely superfluous.

S. Diner et al. (eds.), The Wave-Particle Dualism, 27–67.

CHAOTIC TWO-MODE LASING

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Received 5 September 1983
Revised manuscript received 24 November 1983

We consider a homogeneously broadened, two-mode laser under conditions of rapid relaxation of off-diagonal density-matrix elements. Treating as an example the case of two transverse modes, we show that a spatially inhomogeneous pumping rate in such a laser can give rise to chaotic emission.

The hallmark of (deterministic) chaotic behavior is very sensitive dependence on initial conditions, i.e., an exponential separation on average of initially close trajectories. Chaotic systems have broadband power spectra, even without a stochastic driving force. It is now evident from both theoretical and experimental studies that chaotic dynamics appear in a wide variety of physical systems.

Quantum optics and laser physics offer many interesting possibilities for the study of chaos [1]. For instance, the "simple" problem of a single classical field mode interacting with a collection of two-level atoms is known to admit chaotic time evolution [2]. It has recently been proposed that the appearance of chaos in a model of molecular vibrational photoexcitation may explain the incoherent absorption observed for many molecules [3]. Along somewhat different lines, there is considerable interest in "optical turbulence" in optically bistable devices [4–7].

Lasers can also in some instances oscillate chaotically. Haken [8] noted that a set of three equations modeling a single-mode, homogeneously broadened laser (SMHBL) is of the same form as the Lorenz equations; however, the parameter range for which chaos is expected is highly atypical. Yamada and Graham [9] found chaos in a driven van der Pol model of a SMHBL. Using a CO_2 laser with a modulated cavity loss, Arecchi et al. [10] have observed period doubling and chaos that is well described by a rate-equation model of a SMHBL. Evidence of chaos in a single-mode, highly inhomogeneously broadened He-Xe laser has been reported by Abraham et al. [11]. Weiss et al. [12] have observed chaotic emission from a 3.39 μm HeNe laser, and this appears to be a consequence of a three-mode instability. Some time ago Graham [13] showed that the three-mode instability in homogeneously broadened media will have the chaotic features of the Lorenz model.

The literature on multimode laser operation and instabilities is quite extensive [14]. Recent interest in chaotic dynamics has revitalized interest in laser instabilities including multimode effects [15]. In this note we show that chaotic emission is possible in a two-mode homogeneously broadened laser. More specifically, we show that a certain degree of spatial inhomogeneity in the pumping mechanism can give rise to chaotic emission. This inhomogeneity alone can lead to chaos; there is no need to "force" the chaos by cavity modulation, for instance. We will here summarize our model and conclusions, and present representative numerical results. A more detailed discussion of the model, possible experiments, and routes to chaos, is in preparation.

The model derives from the coupled Maxwell–Bloch equations, assuming a scalar electric field expanded in resonator mode functions $E_\lambda(r)$. The off-diagonal Bloch variables are assumed to follow adiabatically the population difference, an approximation expected to be valid if the homogeneous linewidth is much greater than population decay rates.

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ERRATUM

Mei-Li Shih and Peter W. Milonni, Chaotic two-mode lasing, Optics Comm. 49 (1984) 155.

The above paper was printed with figures from an earlier version of the manuscript. At the suggestion of a referee the authors had changed some parameters and produced new figures. They wish to note that none of the basic conclusions of the paper need modification, and would be happy to supply corrected figures to interested readers.

PHYSICAL REVIEW A VOLUME 28, NUMBER 1 JULY 1983

Exponential decay, recurrences, and quantum-mechanical spreading in a quasicontinuum model

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(Received 20 December 1982)

We consider a single quantum state coupled equally to each of a set of evenly spaced quasicontinuum (QC) states. We obtain a delay differential equation for the initial-state probability amplitude, and this equation is solved analytically. When the QC-level spacing goes to zero, the initial-state probability decays exactly exponentially. For finite QC-level spacings, however, there are recurrences of initial-state probability. We discuss Tolman's "quantum-mechanical spreading" of probability and also a classical analog of our model.

I. INTRODUCTION

One of the best-known general problems in physics is the decay of a single excited quantum state into a background of states. Examples may be found in spontaneous emission from an atom, decay of a radioactive nucleus, radiationless transitions in polyatomic molecules, autoionization, and, more generally, the problem of time asymmetry in physics.[1]

The problem of interest to us in this paper involves a discrete background or "quasicontinuum" (QC) of states. These background states are all coupled to a single, initially excited quantum state, but not directly to each other. Whereas the details of the distributions of coupling strengths and energy levels play a role in the dynamics of this system, some interesting general features may be obtained from specific models. In particular, we will consider the QC states to be equally spaced in energy and to have the same coupling to the initially excited state (Fig. 1).

Various researchers have found this model useful in their fields of interest. Davies,[1] for example, uses it to discuss the physics of time asymmetry. Bixon and Jortner[2] applied it to intramolecular radiationless transitions, while Stey and Gibberd[3] used it and other solvable models to discuss the decay of the initial state in the limit of a background continuum of states. Similar work was reported by Lefebvre and Savolainen.[4] More recently, Eberly *et al.*[5,6] have applied a somewhat more general model to a study of laser excitation of a molecular quasicontinuum. On the basis of numerical computations, they emphasized that the system has a characteristic "recurrence time" that is directly proportional to the QC density of states.

This problem in elementary quantum mechanics has a certain richness that deserves a simple and general treatment outside the specialized contexts in which it has appeared. It is our purpose here to give such a treatment, which we feel to be of considerable pedagogical value. The model illustrated in Fig. 1, while exactly solvable, is far from trivial. It can be used not only as a paradigm for the problem of dissipation and exponential decay in quantum mechanics, but also to elucidate Fermi's Golden Rule and the general phenomenon of "quantum-mechanical spreading."[7] We discuss these and other aspects of the model that are not evident in the cited research literature.[1-6]

II. THE SCHRÖDINGER EQUATION AND A DELAY EQUATION

The time-dependent Schrödinger equation for the QC model of Fig. 1 may be written in terms of the state amplitudes as follows:

$$\dot{a}(t) = -i\beta \sum_{n=-\infty}^{\infty} b_n(t) , \tag{2.1a}$$

$$\dot{b}_n(t) = -i(\Delta_0 + n\rho^{-1})b_n(t) - i\beta a(t) , \tag{2.1b}$$

where a and b_n are the amplitudes for the initially excited state $[a(0)=1]$ and the background states, respectively.

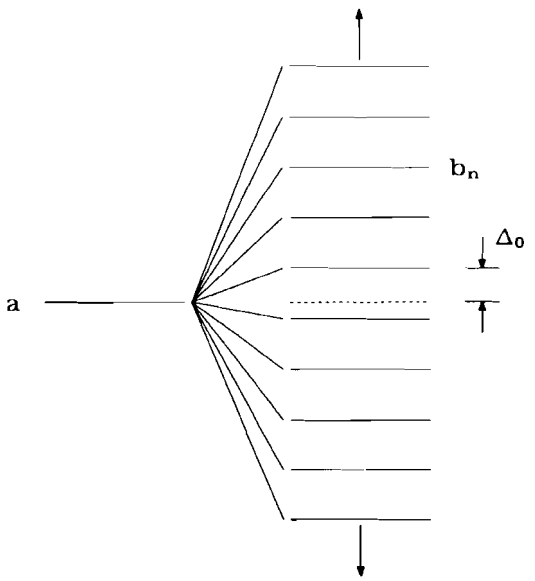

FIG. 1. Diagram of model shows coupling of initially excited state $|0\rangle$ with infinite number of equally spaced background states $|n\rangle$ with spacing ρ^{-1}.

Chaos in the multiple photon excitation of molecules due to vibration-rotation coupling at lowest order

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We study the multiple photon excitation of a molecule with a triply degenerate near-resonant IR-active mode. The molecule is described by its zeroth-order Hamiltonian as an uncoupled harmonic oscillator-rigid rotor. The molecule-field interaction is dipolar with the field treated semiclassically. The Hiesenberg equations of motion are solved in the classical approximation and chaos is found when the photon absorption exceeds the $t = 0$ angular momentum. The origin of the chaos is due to the nonconservation of the molecule angular momentum.

I. INTRODUCTION

One of the salient empirical features of multiple photon absorption is the general lack of coherence between the laser driving and the molecular excitation.[1] This lack of coherence, found in all but the very most intense laser fields, is manifest as a dependence of the absorption on laser fluence (or pulse energy) rather than field intensity. Quantum mechanically, incoherence occurs mainly in damped systems or in broadened transitions.[2] We have previously investigated models in which the pumped mode of a molecule is coupled to N-harmonic background modes (IR-inactive). When the pumped mode is anharmonic, chaotic classical motions are found which give the classical analog of incoherent, fluence dependent absorption.[3] However, in these calculations the rate of absorption is found to depend upon detailed characteristics of the specific molecule under study, in contrast with the data. Blame is therefore cast upon the rotational effects which at first sight appear to be treatable as simple inhomogeneous broadening[4] (treated analogously with the Doppler effect). One is then led to the conclusion that in very cold experiments (where we "freeze out" the rotational Boltzmann distribution) we should still observe incoherent absorption but absorption nonetheless characteristic of the specific molecule being excited.

In this paper we consider (for clarity in separating effects) the simplest possible molecular model, that of a (degenerate) harmonic oscillator-rigid rotor.[5] The spectrum of such a model consists of equally spaced vibration-rotation bands. The molecule-field interaction is electric dipolar, however herein lies a nonlinear coupling which causes the nonconservation of the molecular angular momentum[6] (quantum mechanically $\Delta J = 0, \pm 1$ for each transition). As we shall see by direct integration of the classical equations of motion, complicated behavior can occur under strong driving such that: (1) the absorption is incoherent, and (2) rotational effects cannot be treated as simple incoherent broadening.

II. MODEL AND EQUATIONS OF MOTION

Our model Hamiltonian is

$$H = \Delta \mathbf{a}^* \cdot \mathbf{a} + B_0 \mathbf{J}^2 + \Omega \boldsymbol{\epsilon} \cdot C \cdot (\mathbf{a}^* + \mathbf{a}) , \tag{II 1}$$

where $\mathbf{a}^*$ ($\mathbf{a}$) is the vector creation (annihilation) operator for the IR-active normal mode of the molecule with components referred to body-fixed axes, J is the molecular angular momentum operator, C is the 3×3 orthogonal matrix relating the lab-body reference frames, $\boldsymbol{\epsilon}$ is the laser's polarization vector whose components are referred to the laboratory frame, Δ, B_0, and Ω are parameters representing the laser detuning, the inverse moment of inertia of the molecule in its equilibrium configuration and the laser-molecule Rabi frequency, respectively. If $\{\hat{l}_1, \hat{l}_2, \hat{l}_3\}$ are the lab-frame unit vectors and $\{\hat{f}_1, \hat{f}_2, \hat{f}_3\}$ are those of the body frame, then

$$C_{ij} = \hat{l}_i \cdot \hat{f}_j \tag{II 2}$$

and the body-fixed and lab-fixed components of the molecular angular momentum[7] are, respectively,

$$J_i = i \sum_{s=1}^{3} \left(C_{sj} \frac{\partial}{\partial C_{sk}} - C_{sk} \frac{\partial}{\partial C_{sj}} \right) , \tag{II 3}$$

and

$$J_I = -i \sum_{s=1}^{3} \left(C_{Js} \frac{\partial}{\partial C_{Ks}} - C_{Ks} \frac{\partial}{\partial C_{Js}} \right) , \tag{II 4}$$

with $i, j, k (I, J, K)$ cyclic in $\{1, 2, 3\}$. We then have the well known relations

$$[J_I, J_J] = i \epsilon_{IJK} J_K , \tag{II 5}$$

$$[J_i, J_j] = -i \epsilon_{ijk} J_k , \tag{II 6}$$

$$[J_i, J_I] = 0; \quad i, I \in \{1, 2, 3\} , \tag{II 7}$$

and

$$\mathbf{J} \cdot \mathbf{J} = \sum_{I=1}^{3} J_I^2 = \sum_{i=1}^{3} J_i^2 . \tag{II 8}$$

These equations show the basic $0(3)\times0(3)$ structure of

 0021-9606/83/235345-06$02.10

Why spontaneous emission?

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(Received 3 March 1983; accepted for publication 24 June 1983)

This paper is a discussion of that perennial question, "Why does an excited atom radiate?" A satisfactory physical picture emerges when proper account is taken of the interplay between radiation reaction and the (quantum-mechanical) zero-point fluctuations of the radiation field. The fluctuation-dissipation connection between these two effects is therefore emphasized.

The intense atom glows
A moment, then is quenched in a most
cold repose.
—P. B. Shelley, *Adonais*

I. THE QUESTION

Professor Feynman has described an experience familiar to many of us[1]:

> You might wonder what [my father] got out of it all. I went to MIT. I went to Princeton. I came home, and he said, "Now you've got a science education. I always wanted to know something that I have never understood; and so, my son, I want you to explain it to me." I said, "Yes."
>
> He said, "I understand that they say that light is emitted from an atom when it goes from one state to another, from an excited state to a state of lower energy."
>
> I said, "That's right."
>
> "And light is a kind of particle, a photon, I think they call it."
>
> "Yes."
>
> "So if the photon comes out of the atom when it goes from the excited to the lower state, the photon must have been in the atom in the excited state."
>
> I said, "Well, no."
>
> He said, "Well, how do you look at it so you can think of a particle photon coming out without it having been there in the excited state?"
>
> I thought a few minutes, and I said, "I'm sorry; I don't know. I can't explain it to you."
>
> He was very disappointed after all these years and years of trying to teach me something, that it came out with such poor results.

The question of why or how an atom radiates would likely elicit a similar response in most physicists even today. But some progress has been made in the past ten years. In this article I will describe recent developments in the simplest way I can. To set a proper context for the discussion I will begin in Sec. II by emphasizing the "relevance" of the subject, and follow in Sec. III with some of its history. Sections IV and V are devoted to two possible interpretations of spontaneous emission, and in Sec. VI the two are, to some extent, made one. We close in Sec. VII with some details.

II. THE RELEVANCE

Spontaneous emission is ultimately responsible for most of the light around us. We would not be here without it.

Consider a thermal source of radiation. The atoms in such a source radiate by both spontaneous and stimulated emission. The rates of spontaneous and stimulated emission are A and $B\rho(\nu)$, respectively, where A and B are the Einstein coefficients for spontaneous and stimulated emission and $\rho(\nu)$ is the Planck spectral energy density. The ratio of these rates for a transition with Bohr frequency ν is

$$\frac{A}{B\rho(\nu)} = e^{h\nu/kT} - 1 \tag{2.1}$$

since $\rho(\nu) = (A/B)(e^{h\nu/kT} - 1)^{-1}$.[2] If the Sun is regarded as a thermal source at $T = 6000$ K, this ratio is about 400 at $\lambda = 4000$ Å and about 30 at $\lambda = 7000$ Å. Thus, to the extent that the Sun is an ideal blackbody radiator, most of its visible output is due to spontaneous emission.

Spontaneous emission is so ubiquitous that there are many names associated with what is basically the same thing. If the atoms (or molecules) are excited by some means other than by heating, the spontaneous emission is called luminescence. Fireflies are luminescent. And there are different names associated with luminescence, depending specifically on how excited atoms are produced (electroluminescence, chemiluminescence, etc.) If the excitation is effected by the absorption of radiation, the spontaneous emission is called fluorescence. Sometimes the molecules have a metastable level and continue to fluoresce long after the exciting radiation is turned off. This is called phosphorescence. Figurines that magically glow in the dark are phosphorescent.

Lasers, of course, produce light by stimulated emission. However, when a laser is turned on the photons that first do the stimulating are themselves the result of spontaneous emission.

III. SOME HISTORY

Hertz's experiments of 1887 confirmed that oscillating charges radiate. In Lorentz's theory of light and matter[3] atomic radiation was attributed to the oscillation of atomic electrons. There was no way to understand why they would radiate at only certain frequencies. The emission and absorption frequencies of an atom were simply inserted into the theory through "spring constants" associated with the (unexplained) binding of electrons. All this changed in 1913 with the advent of the Bohr theory of the hydrogen atom.

Bohr recognized a nonclassical element in spontaneous emission, for to him "spontaneous" meant "acausal." It was (and is) impossible to predict exactly when an excited atom will make a quantum jump and emit a photon.

Further nonclassical aspects of spontaneous emission were uncovered by Einstein in 1917. In particular, Einstein inferred that an atom must recoil upon spontaneous emission. This recoil cannot be understood classically, because

Chaos in Multiple-Photon Excitation of Molecules

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(Recieved 11 July 1983)

With use of a Hamiltonian model system to represent ir laser excitation of a molecule it is shown that chaos is a fundamental aspect of this physical process. On average the chaos leads to a fluence-dependent absorption which previously was attributed to rapid statistical equilibration of energy in the quasicontinuum and was modeled by population rate equations. The origin of the chaos is the interplay of the pump mode's anharmonicity and intramolecular energy transfer.

PACS numbers: 33.80.Kn

During the past 7–10 years multiple-photon excitation (MPE) of molecules has been a major field of research both experimentally and theoretically.[1] The major interest was initiated by the unexpectedly easy dissociation of SiF_4 where many ir photons of like frequency were absorbed per molecule.[2] Conventional wisdom viewed the pump mode of the molecule as a triply degenerate anharmonic oscillator which after the absorption of a few photons would be off resonant, eliminating further photon absorption. This surprising experimental result inspired many researchers to follow up this work in SF_6 and in many other molecules with the hope of achieving bond-selective photochemistry and laser isotope separation. By the early 1980's it was clear that most molecules act like a sponge where intramolecular relaxation from the pump mode into the background modes redistributes the laser energy very quickly removing the potential for easily achieving these goals. In fact the redistribution of laser energy was apparently so rapid that a statistical energy distribution in the background modes was a basic assumption in almost all theoretical models.[3] This background became known as the quasicontinuum, and its properties typically were modeled with rate equations giving a strong fluence-dependent absorption consistent with experiment.

Using a simplified model of MPE we will show that our present conventional wisdom may again be wrong in that fluence-dependent absorption can result from chaotic dynamics in the molecule-photon interaction even when the density of background states is too small to justify a rate-equation treatment. We believe the generic nature of our model will imply that chaos is a fundamental part of the fluence-dependent physics of MPE.

Our basic Hamiltonian describing the physics of MPE in the rotating-wave approximation consists of an anharmonic pump mode, intramolecular coupling to harmonic background modes, and the laser–pump-mode interaction:

$$H=\Delta a^{\dagger}a-\chi(a^{\dagger}a)^2+\sum_{m}^{N}(\Delta+\epsilon_m)b_m^{\dagger}b_m+\sum_{m}^{N}\beta_m(a^{\dagger}b_m+b_m^{\dagger}a)+(\Omega/\sqrt{n})(ac^{\dagger}+ca^{\dagger}). \quad (1)$$

The pump-mode oscillator described by the annihilation operator a has a laser-molecule detuning Δ and anharmonicity χ. The monochromatic laser is represented by the creation and annihilation operators $c^{\dagger}$ and c. The photon number operator $c^{\dagger}c$ has been removed from the Hamiltonian by making the rotating-wave approximation. The N background modes described by the annihilation operator b_m have their fundamental frequencies determined by ϵ_m. The intramolecular coupling is characterized by β_m. The laser has initially n photons on average, and the Rabi frequency is given by Ω. This Hamiltonian conserves excitation number.

We will solve the dynamics implied by (1) by making several approximations: (a) $\beta_m \equiv \beta$; (b) $\epsilon_m \equiv \Delta_0 + m\rho^{-1}$ (where Δ_0 is the separation between band origins of the nearest background oscillator and the pump mode and where ρ is the density of background origins which are now assumed equally spaced); (c) $N \to \infty$ (this approximation has been justified by researchers in the field of radiationless transition theory)[4,5]; (d) $n \gg$ the number of

International Journal of Theoretical Physics, Vol. 22, No. 4, 1983

Does the Electromagnetic Mass of an Electron Depend on Where It Is?[1]

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The question of whether the electromagnetic mass of the electron depends upon its electromagnetic environment is discussed in connection with previous theoretical and experimental work. When the quantization of the field in other than free space is properly understood, it is evident that the true electromagnetic mass of the electron is unaltered.

Orthodox radiation theory leads to the conclusion that a part δm of the mass of an electron is of electromagnetic origin. It is also asserted that no experiment can differentiate between δm and the remaining, bare mass of the electron (see, for example, Schweber, 1962). Other radiation theories, in which the electron experiences no self-interaction and therefore has no electromagnetic mass, have been advanced (Wheeler and Feynman, 1945). It is therefore of interest to consider the possibility that δm may be affected by the presence of conducting plates (Power, 1966; Guttrich and Billman, 1967; Golub and Guttrich, 1967).

I recently considered the possibility, in light of a previous interest in the modification of spontaneous emission rates by the presence of conducting plates (Milonni and Knight, 1973), that this modification of δm might provide a test of the reality of electromagnetic mass. Barton's careful work on quantum electrodynamics near conducting plates (Barton, 1974) brought the work of Power (1966) to my attention, and so I learned that the modification of δm by conducting plates was not a new idea. However, I now believe that the proposed experiment does not actually involve a modification of δm *per se*, so that the orthodox view of the unobservability of electromagnetic mass is not in question.

[1]Research supported in part by dual funding by NSF-EPSCOR Grant ISP 8011447 and the State of Arkansas.

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0020-7748/83/0400-0323$03.00/0

PHYSICAL REVIEW A VOLUME 26, NUMBER 3 SEPTEMBER 1982

Theory of gas lasers operating on two coupled transitions

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The semiclassical Lamb theory of gas lasers is extended to the case of lasers operating simultaneously on two coupled transitions. The theory is formulated in terms of the density-matrix equations for a three-level gaseous laser medium interacting with the field through electric-dipole transitions, so that there are two allowed transitions and one two-photon transition. It is assumed that there is a single resonator mode near each of the transition frequencies. Previous treatments of such three-level gas lasers either have used simple rate equations or have solved the density-matrix equations in an approximate fashion valid only for low intensities. Thus the numerical approach developed herein represents, to our knowledge, the first semiclassical theory of three-level gas lasers which is accurate for arbitrary detuning and intensity. Significant deviations from earlier, approximate treatments are found.

I. INTRODUCTION

In describing the resonant interaction of light with an atom or molecule, it is often a very accurate approximation to include in the analysis only those discrete energy levels that define the resonant transitions. If the resonant radiation is nearly monochromatic, for instance, the model of the "two-level" atom is extremely useful.[1,2] The well known and highly successful Lamb theory of the gas laser is based on the treatment of the laser medium as a collection of two-level atoms.[1,3]

There has recently been considerable interest in the resonant interaction of light with three-level atoms or molecules. (See Refs. 4–8 and the references cited therein.) The analysis of the three-level system is more complicated than the two-level case, and certain aspects of the problem have consequently not been carefully analyzed. This is especially true for the case of the three-level laser operating simultaneously on two coupled transitions of a Doppler-broadened gaseous gain medium. Beterov and Chebotaev noted in 1975 that "In a three-level gas oscillator, the picture of saturation effects becomes considerably more complicated (than in a two-level system) and at present there is no theory of a three-level gas laser."[4] A low-intensity theory of the three-level gas laser was published in 1975.[9] It is our purpose to present a more general theory.

The motivation for this work has been to develop a more complete understanding of lasers which operate on multiple transitions. A number of gas-laser gain media consist of atoms or molecules with three or more states which are all coupled by allowed transitions in a cascading configuration. Some examples are CO, He-Ne, DF, and HF. The existence of several coupled transitions will influence the laser gain and index of refraction by causing the level population and the induced polarization oscillations to have complicated spatial variations along the direction of propagation of the light (the z axis). The spatial variation of any of the level populations will contain harmonics of the wave numbers of all the laser modes.

In the two-transition laser there are effects associated with the direct coupling of the laser modes by the density-matrix element connecting the two levels between which there is no allowed transition. The third-order semiclassical theory of the response of a three-level gain medium indicated that these coupling effects are significant.[10] The analysis of a three-level gas with a high-intensity laser beam saturating one transition has also shown that these coherence effects qualitatively change the line shape.[11–14] The third-order laser theory of Najmabadi *et al.*[9] showed that peaks in plots of mode intensity as a function of laser tuning near line center were caused by these coupling effects. This treat-

Stimulated emission, absorption, and interference

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(Received 22 July 1981; accepted for publication 20 January 1982)

It is shown how stimulated emission and absorption may be viewed in terms of the interference of the incoming field and the field radiated by the atom.

I. INTRODUCTION

The concept of stimulated emission was introduced by Einstein in his remarkably simple derivation of the blackbody radiation spectrum.[1] Einstein set forth his hypotheses concerning the absorption and emission of radiation and then showed that the Planck spectrum follows from the condition of thermal equilibrium between radiation and two-level atoms. The A and B coefficients for the spontaneous and stimulated processes could not be computed directly. Their computation required "an electrodynamics and mechanics modified in the sense of the quantum hypothesis,"[2] a requirement that was met a decade later with the advent of quantum mechanics.

The derivation of the Einstein A and B coefficients is not an especially difficult exercise in quantum mechanics.[3] In particular, stimulated emission and absorption may be understood in the "semiclassical" approach of treating the atom quantum mechanically and the field classically.[4] The usual semiclassical treatment includes only one mode of the electromagnetic field.[5] A transition rate is calculated for emission or absorption, depending on whether the atom is initially in the upper or lower state, respectively, of the resonant transition. This transition rate is proportional to the intensity of radiation at the Bohr frequency of the transition.

According to the superposition principle for electromagnetic waves, the total electric (or magnetic) field is the sum of fields from all the sources in the universe. In particular, the field from the atom has a multipole character, and it is usually sufficient to consider the dipole case. One is then led to imagine, in addition to the applied field, a dipole field emanating from the atom. There are three contributions to the total intensity of the electromagnetic field: (1) The intensity of the incident wave. (2) The intensity of the field radiated by the atom. This term describes Rayleigh scattering or absorption followed by spontaneous emission. (3) The interference between the incident wave and the radiated wave. Evidently only the third contribution can be associated with a change in electromagnetic field energy due to induced emission or absorption. According to Lamb:

> When stimulated emission by an excited atom is treated, either quantum mechanically or by a suitable classical model, one finds that the numbers of photons in those modes of the radiation field which were initially excited are increased by the interaction. On the other hand, the electromagnetic field radiated by such an atom is found to have the appropriate multipole character and shows no trace of the above augmentation of the incident wave. In order to get amplification of the incident wave it is necessary to consider the interference of the incident and radiated waves.[6]

The standard textbook treatments of stimulated emission and absorption make no mention of interference. Instead they focus on the atom and the transition rate associated with these induced processes. In this paper we consider a single-mode incident field and an atom with a near-resonant electric dipole transition. We show explicitly how stimulated emission and absorption arise from the interference of the incident field and the field radiated by the atom.

It is convenient to work as closely as possible along the lines of a classical electron oscillator model of the atom. As we show in Sec. II, this model can be justified as an approximation to the quantum theory, and in fact the approximation is essentially equivalent to that inherent in the standard perturbation–theoretical approach to absorption and induced emission.[5] We find it convenient to use a certain expansion formula for the dipole field, which is derived in Sec. III. In Sec. IV we consider the total electromagnetic field energy, and we show that stimulated emission and absorption are indeed associated with the interference between the incident and radiated fields. We conclude in Sec. V with a discussion of our results.

II. ELECTRON OSCILLATOR MODEL

Before the advent of quantum theory, the interaction between light and matter was often described in terms of a classical oscillator system. In this model, frequently called the Lorentz model, each electron in an atom is bound by a springlike elastic force. When an electron is displaced from its equilibrium position a restoring force arises tending to re-establish the original unperturbed state. Although this model is of course unrealistic from the modern viewpoint, consideration of light as an electromagnetic wave acting on an "electron on a spring" produces remarkably accurate results, accounting for a host of optical effects including absorption, dispersion, and reflection.

The electron oscillator model, because of its general applicability and inherent simplicity, is cited frequently. Weisskopf, for instance, states that,

> ... the interaction of light with atoms can be described rather simply. One obtains the essential features of that interaction ... by replacing the atom with electron oscillators ... .[7]

In his *Lectures on Physics* Feynman writes

> To find what motion we expect for the electrons, we will assume that the atoms are little oscillators, that is, that the electrons are fastened elastically to the atoms, which means that if a force is applied to an electron its displacement from its normal position will be proportional to the force.

PHOTONS CANNOT ALWAYS BE REPLICATED

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Received 5 August 1982
Revised manuscript received 1 October 1982

Perfect and certain replication of any single photon is impossible.

Recent experimental tests of Bell's inequalities [1, 2] have stimulated interest in the possibility of carrying out other experiments with implications for the conceptual foundations of quantum mechanics. One type of experiment, already is progress, involves the amplification of a beam of light so attenuated that on average only a single photon passes through the gain tube [‡1]. The experiments in question, like the previous ones [1,2], make use of the two-state nature of photon polarization.

For such experiments the excitation scheme of fig. 1 is being considered [3]. It is implied that, because the amplification of the signal field of frequency ν_s is not associated with a real "allowed" transition, there is little spontaneous emission of photons into the signal mode. If this were true, it should be possible to clone any single photon with practically no fundamental noise limitation.

We will argue that it is not possible to replicate with certainty a single photon of unknown polarization. This impossibility has deleterious implications for a recent proposal for superluminal communication [4], which will be discussed elsewhere.

Consider first the specific excitation scheme of

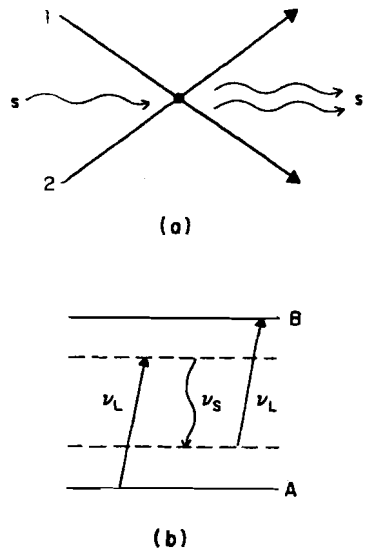

Fig. 1. (a) Excitation scheme using two crossed laser beams of frequency ν_L and a signal field of frequency ν_S. (b) Three-photon process using atomic levels A and B.

fig. 1. The interaction hamiltonian describing the coupling of the two laser fields 1 and 2 with the signal mode is

$$H_I = Ca_1^\dagger a_2^\dagger a_s + C^* a_1 a_2 a_s^\dagger , \qquad (1)$$

where C is a coupling constant and the a's and $a^\dagger$'s are annihilation and creation operators, respectively.

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[‡1] Such experiments were discussed by several lecturers at the Symposium on Wave–particle dualism (in honor of the 90th birthday of Louis de Broglie), ref. [3].

PHYSICAL REVIEW A VOLUME 25, NUMBER 3 MARCH 1982

Casimir forces without the vacuum radiation field

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(Received 12 March 1981)

Although the electromagnetic Casimir forces are customarily explained in terms of the vacuum radiation field, they may also be understood in terms of radiation reaction, without explicit reference to the vacuum field. In particular, the Casimir forces may be obtained if one recognizes that the radiation reaction field of a dipole on itself depends on where the dipole is located, for this field is determined by the modal characteristics of the surrounding space. Only in infinite free space does the field of radiation reaction have a spatially invariant meaning. The fact that the Casimir forces may be understood in terms of either the vacuum electromagnetic field or the field of radiation reaction is an example of the general fluctuation-dissipation relation between the two concepts. These ideas are illustrated with derivations of several "vacuum-field effects," including the Casimir-Polder attraction of an atom to a conducting wall and the van der Waals force between two atoms.

I. INTRODUCTION

The quantum theory of radiation predicts a zero-point energy $\frac{1}{2}\hbar\omega$ per normal mode of the field. This is a consequence of the fact that each mode is canonically equivalent to a harmonic oscillator. Summation over all modes leads to an infinite zero-point field energy, a result occasionally considered embarrassing.[1] Consideration of the zero-point electromagnetic energy may be sidestepped, general relativity notwithstanding, on the grounds that a constant additive term in the Hamiltonian has no effect on commutators and equations of motion. At the same time, it is generally believed that there are indeed real physical effects attributable to the zero-point field. Such effects include the Casimir-Polder attraction of a neutral polarizable particle to a conducting wall, the Casimir attraction between two parallel conducting plates, and the van der Waals interaction between polarizable particles.[2]

In 1948 Welton[3] showed that the (nonrelativistic) radiative level shift obtained by Bethe[4] could be attributed to the effect on the atomic electron of the zero-point field. Welton's elegant derivation seems to have influenced many physicists to believe that the ubiquitous phenomenon of spontaneous emission is also a consequence of the quantum-mechanical zero-point field.[5] An atom in a pure stationary state has no dipole moment, i.e., the expectation value of the dipole-moment operator vanishes, so that the process of spontaneous emission cannot be simply understood on the basis of classical radiation theory. But a fluctuating zero-point field, present even when there are no other sources of radiation but the excited atom, can perturb the electron motion and induce a transition to a state of lower energy. The coupling of the electron to the zero-point field thus provides an intuitive explanation of spontaneous emission. But this interpretation is hardly satisfying. In particular, it offers no good explanation of the fact that atoms do not absorb energy from the zero-point field.

In classical electrodynamics the radiation by an accelerating charge is associated with the force of radiation reaction. It seems that the early workers in the quantum theory considered spontaneous emission to be a result of this "radiation force."[6] Of course, radiation reaction also appears in quantum electrodynamics. As in classical theory, the radiation reaction field is just the solution of the inhomogeneous Maxwell equation for the electric field, evaluated at the source. But in quantum theory there is also the zero-point field, i.e., the solution of the homogeneous Maxwell equation, acting on the source. What does the quantum theory of radiation have to say about spontaneous emission, and in particular about the radiative corrections to the energy levels?

Ackerhalt *et al.*[7] have given a Heisenberg-picture treatment of spontaneous emission in which the usual procedure of normal ordering is adopted. They show that spontaneous emission, and the associated level width and (nonrelativistic) shift, can be understood solely on the basis of radiation reaction. As a result of normal ordering, there are no explicit contributions from the zero-point field. In particular, it was interesting that they could obtain essentially the same result as Welton for the level shift, even though Welton dealt directly with the zero-point field. The two approaches were tied together by Senitzky[8] and Milonni *et al.*,[9] who performed Heisenberg-picture calculations in which operator orderings other than normal were used. If annihilation and creation operators were symmetrically ordered, for in-

RADIATION REACTION AND THE NONRELATIVISTIC THEORY OF THE ELECTRON

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Received 25 November 1980
Revised manuscript received 5 February 1981

Radiation reaction must be included in the nonrelativistic Heisenberg equation of motion of the point electron in order to preserve the position–momentum commutation rule.

The nonrelativistic theory of the point electron interacting with the vacuum electromagnetic field provides a useful intuitive picture of certain quantum-electrodynamical effects [1,2]. The Heisenberg equations of motion for the electron and the field are, of course, formally identical to their classical counterparts. In particular, for the x-component of the electron position vector we have the equation

$$m\ddot{x}(t) = eE(t)\,, \tag{1}$$

where $E(t)$, the x-component of the electric field operator, is the vacuum field $E_0(t)$ plus the field of radiation reaction. The latter may be calculated along precisely the same lines as the classical theory of Lorentz, so that eq. (1) takes the familiar form

$$\ddot{x}(t) - \gamma\dddot{x}(t) = (e/m)E_0(t)\,, \tag{2}$$

where $\gamma = 2e^2/3mc^3$ and m is now the renormalized mass.

Physically meaningful results can be obtained when radiation reaction is ignored [1,2]. It is shown below, however, that the theory is fundamentally inconsistent unless both radiation reaction and the vacuum field are allowed in the theory to act on the electron. In particular, it will be shown that the canonical commutation rule $[x, p] = i\hbar$ for the electron is violated if radiation reaction is ignored.

Eq. (2) follows simply from the hamiltonian and the canonical commutation rule for the electron position and momentum; the commutation properties of the field operators are not explicitly required for its derivation. An interesting question is whether the commutation relation $[x, p] = i\hbar$ for the electron is implied by the equation of motion (2) when one invokes the commutation rules for the vacuum field. The answer will be shown to be in the affirmative. The question is reminiscent of the general one posed by Wigner ("Do the equations of motion determine the quantum-mechanical commutation relations?"), who demonstrated that even for the simple harmonic oscillator the equations of motion do not uniquely determine the commutation relations [3].

It is consistent within the nonrelativistic approximation to neglect any spatial dependence of E_0 in eq. (2). The quantization of the free field in the usual fashion then leads to the equation of motion

$$\ddot{x}(t) - \gamma\dddot{x}(t) = \frac{ie}{m}\sum_{k,s}\left(\frac{2\pi\hbar\omega_k}{V}\right)^{1/2}\epsilon_{ks} \times (a_{ks}e^{-i\omega_k t} - a_{ks}^{\dagger}e^{i\omega_k t})\,, \tag{3}$$

where ϵ_{ks} is the x-component of the plane-wave mode with wave vector k and polarization s, $\omega_k = |k|c$, V is a quantization volume, and a_{ks} is the photon annihilation operator for mode (k, s). If we assume that equal-time electron and field operators commute, we can use eq. (3) to calculate $[x, p]$; in particular, $[x, p] = [x, m\dot{x}]$ since $[x(t), A(t)] = 0$. From eq. (3), therefore,

$$[x(t), p(t)] = 4i\gamma\pi^2c^3\int_0^{\infty}\frac{d\omega\,\rho(\omega)}{\omega^3(1+\gamma^2\omega^2)} = i\hbar\,, \tag{4}$$