Frame transformation relations for fluxional symmetric rotor dimers

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The theory of frame transformation relation connecting body oriented angular momentum states and lab weakly coupled momentum states have been extended from rotor-electron to rotor-dimer systems. Coupling schemes are analyzed for weak and strong cases of correlation between lab and two different rotor body frames. It is shown that the frame transformation relation is a purely quantum effect at low angular momentum but an approach to a classical limit for high J. Symmetry analysis of frame transformation is compared to eigensolutions of model coupling Hamiltonian.
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I. INTRODUCTION

The frame transformation of diatomic rotor-electron states was first derived by Chang and Fano[1] and was extended to polyatomic rovibronic states by Harter, Patterson, and de Paixao.[2] Frame transformations allow one to connect lab weakly coupled (LWC) momentum states with the opposite situation, known as body oriented angular (BOA) momentum states. Chang and Fano frame transformations take account of coulomb fields in their wave function development whereas Harter[20] and Patterson[17,18] consider relativity of angular quantities. The two approaches are closely related if the Coulomb potential is treated using multipole expansion.

Jungen and co-workers[3] extended Chang and Fano's approach to develop a multi-channel quantum defect theory (MCQDT) for studying highly excited H_2 Rydberg states in region where Born-Oppenheimer approximations break down. In a more recent work Jungen[4] used MCQDT along with *ab initio* theory to calculate hyperfine splitting in highly excited Rydberg states. This reproduced the experimental transitions and predicted transition moments in a region not yet explored.

Chang and Fano's idea was originally intended only for a diatomic molecule and an electron. In Ref. 2 this is extended to polyatomic molecular rotor-electron models by taking molecular symmetry operations into account. The present work[23] extends that of Ref. 2 to rotor-rotor transformation relations that are more general than those for a rotor-electron model. Relations between two molecular frames and the laboratory frame are developed so as to visualize one rotor's waves as passengers "boarding," a second frame as the two rotor frames go from weakly coupled to strongly coupled states in a single composite rotor. Often theories based on the body frame become confused by noninertial (centrifugal and coriolis) effects. These may be understood better by modeling the dynamics of two or more molecular frames. The strongly coupled bases used by Chang and Fano were achieved using the Born-Oppenheimer approximation (BOA). In our work the acronym BOA may also refer to Born-Oppenheimer approximation as well as body oriented angular momentum coupling as will be explained.

More recently Hougen and Kleiner[5,6] considered torsional-rotor molecule and showed that there is K-rotation labeling problem for eigenvectors as the molecule moves across the barrier. Gronier[7] developed an effective Hamiltonian starting from a general Hamiltonian for a molecule with two periodic larger amplitude motion. This work considered di-methyl group of previous work[5,6] considering only a single methyl group. Our goal is to more clearly understand the effects of rotor-rotor behavior above and below the torsional barriers. The subject of internal rotation goes back before the 1950s and good description of this subject is given by Lin and Swalem.[8] Then, the interest was microwave spectroscopy[22,24] of hindered internal rotations. Fifty years

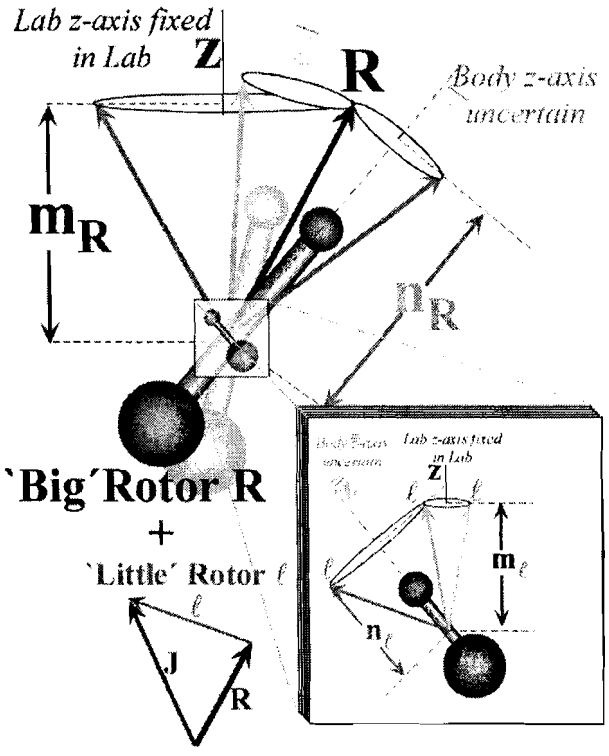

FIG. 1. Schematic of coupled rotor system. The angular momentum J, R, and ℓ are shown along with their projection the lab and body axis.

Quantum-fractal revival structure in C_N quadratic spectra: Base-N quantum computer registers

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A closer examination of Eberly "revivals," Berry "quantum fractals," and Schleich "carpets" exposes a simple unitary-cyclic group structure in the dynamics of wave packets for rotorlike systems with quadratic spectra. Quantum quasifractal patterns are revealed to be permeated by self-similar "X-braid" structures seen by Hofstadter in Zeeman-perturbed band structures and in other spectra related to circle maps. The patterns are shown to correspond to revival dynamics in the flow of energy or information through these N-level systems with X-braids being directly related to quantum beats or elementary U(2) resonance transfer. C_N-group analysis and Farey arithmetic discretely catalog X-braid hierarchy and suggest the possibility of homocyclic nanostructures or C_N circuits that are base-N digital quantum computing registers. The phase dynamics corresponds to a fairly sophisticated integer arithmetic that includes an output of all rational numbers resolvable by a given energy input. C_N circuits automatically give all factors of N. The dynamics of a C_6 device is discussed in detail.

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I. INTRODUCTION

Correspondence and contrasts between classical and quantum dynamics have been a part of physical theory since the introduction of Bohr's first atomic models. At a deeper level, this physical correspondence is mirrored by mathematical relations between the continuous real line and the discrete sets of integral fractions or rational numbers.

Here we will show how the very simplest of Bohr models, a particle confined to a ring, exhibits and elucidates fundamental quantum-classical relationships while it cuts a space-time continuum into arbitrarily fine rational subsets. Furthermore, a possible practical application emerges in the form of an electronic or photonic computing element that might serve as base-N quantum registers and integer factoring circuits.

The model dynamics described below are each an example of *revival* phenomena. Revival is a somewhat nonsecular term coined by Eberly and co-workers [1,2] to describe constructive rephasing observed in computer studies of Jaynes-Cummings quantum-field-atom dynamics. Subsequently, revival phenomena were noted in computer studies of other systems such as Morse oscillators [3] and atomic Rydberg states [4], and more recently, diverse revival phenomena have been observed in computer and laboratory experiments [5–9].

Recent theory has distinguished between *perfect* and *imperfect* revival phenomena according to values of parameters $a_1, a_2, a_3, \ldots$, that determine uniformity of an energy level spectrum ε_0, ε_1, ε_2, of a quantum system.

$$\varepsilon_n = \varepsilon_0 + a_1(n-n_0) + a_2(n-n_0)^2 + \cdots . \tag{1}$$

Revival theories skip the linear or harmonic case of $a_k = \hbar\omega_1\delta_{k,1}$ and $n_0 = 0$ that has a perfectly uniform spectrum of a harmonic oscillator and "revives" monotonously at a single frequency ω_1. (Such a system is the only one that supports perfectly *coherent* states.) Instead, attention has been directed to perfectly *quadratic* spectra with $a_k = \hbar\omega_2\delta_{k,2}$ and $n_0 = 0$ whose levels are a subset of a harmonic spectrum of frequency ω_2,

$$\varepsilon_n = \varepsilon_0 + \hbar\omega_2 n^2 = \varepsilon_0 + \hbar\omega_2(0^2, 1^2, 2^2, 3^2, \ldots). \tag{2}$$

Such model systems include a three dimensional (3D) rigid diatomic or polyatomic rotor [10], a particle in a box [11], and a simple Bohr single-coordinate angle rotor, which is the subject of the first part of this exposition.

Because spectrum (2) is a subset of a harmonic one it is required to repeat perfectly with a fundamental frequency ω_2. In other words, it must have perfect revivals with time period $\tau = 2\pi/\omega_2$. However, as found by the earlier studies [10,11], there are quasidiscrete fractional revivals occurring at fractional periods $\tau/2$, $\tau/3$, $\tau/4$, and times $2\tau/3$, $3\tau/4$,.., each consisting of arrays of 2, 3, 4,.., "kaleidoscopic" copies of the initial probability distribution. Much of this structure was hilighted by Berry [12] in quantum fractal landscapes of wave probability and by Schleich and co-workers [13–16] in quantum carpets made using colored and shaded space-time plots of particle-in-a-box wavepacket probability.

A closer and modified study of Schleich carpets, such as the ones shown in Fig. 1, shows a simple underlying group structure in wave amplitude *and phase*. This analysis suggests a new kind of elementary quantum computing devices that automatically do a kind of integer arithmetic. The study reported here elucidates a quantum fractal amplitude and phase structure in space-time wave patterns. The amplitude patterns are reminiscent of "Hofstadter X-braids" [17–19] as well as the more recently discovered "chaotic" structure associated with problems involving circle maps [21]. Revival structure will be explained here using simple number-theoretic tools such as the Farey sum (used in classical chaos) [20–22] and a fundamental C_N group analysis [23] of coupled oscillators.

An overall mathematical tool for this development is a unitary-homocyclic $[U(n) < C_n]$ symmetry approach to Fourier wave mechanics [23]. Simply put, an artificial "lattice-gauge" discretization of space time is done to match the

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Wave Node Dynamics and Revival Symmetry in Quantum Rotors

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Symmetries and dynamics of wave nodes in space and time expose principles of quantum theory and its relativistic underpinning. Among these are key principles behind recently discovered dephasing and rephasing phenomena known as *revivals*. A reexamination of basic Eberly revivals, Berry "quantum fractal" landscapes, and the "quantum carpets" of Schleich and co-workers reveals a simple Farey arithmetic and C_n-group revival structure in one of the earliest quantum wave models, the Bohr rotor. These principles may be useful for interpreting modern time-dependent rovibrational spectra. The nodal dynamics of the Bohr rotor is seen to have a quasi-fractal structure similar to that of earlier systems involving chaotic circle maps. The fractal structure is an overlay of discrete versions of Bohr's rotor model. Each N-point Bohr rotor acts like a base-N quantum "odometer" which performs rational fraction arithmetic. Such systems may have applications for optical information technology and quantum computing. © 2001 Elsevier Science

Wave phase behavior in space and time is fundamental to the understanding of physics and optics. Phase coherence has been an important idea since Huygens developed the principles of refraction in the late 1600s and it continues to be a basis of the quantum theory of atomic, molecular, or optical nanostructures. Newton's classical dynamics of the 1700s and the related Hamilton–Jacobi least-action principles of the 1800s were, in the mid-1900's, seen by Dirac and Feynman to result from the need for a stationary quantum phase. Only "right" paths which "agree" on an extremal phase value are able to win a kind of quantum lottery to exist while a vast majority of "wrong" paths cancel each other out in a cacophonous mishmash of mismatched phases.

Recently, the phenomena of *rephasing* or *revival* has arisen. Quantum revivals, like classical chaos and fractals, are a class of phenomena that many saw but few observed. Each has required modern computer simulations and graphics to make a convincing case for its existence, but each was thought, at first, to be an artifact of numerical code gone awry. Also, each of these phenomena is inadequately described by Newtonian differential continuum analysis but yields to discrete algebraic or number-theoretic approaches. Finally, revivals are relevant to a field entitled "quantum chaos," thought by some to be an oxymoron.

The term *revival* is a coinage by Eberly (*1*) to describe unexpected rephasing that appeared in 1976 computer studies of atom–quantum electrodynamics. For the next two decades there were sporadic reports of revival phenomena including *fractional revivals* in quantum treatments of simpler systems such as rotors (*2*), anharmonic vibrators (*3*), Rydberg orbitals (*4*), or an infinite square well or "particle-in-a-box" (*5*). A theory (*6*) of revivals in 1989 was based on box waves. Finally, in 1996 and 1997, Berry (*7*) and Schleich and co-workers (*8, 9*) used box wave simulations to plot "quantum fractal" landscapes and "quantum carpets" and thereby showed convincingly that revivals are a phenomenon whose theory and applications needs to be more deeply explored.

This work analyzes revival phenomena using symmetry group theory and a Farey arithmetic adapted from classical chaos theory. This new approach, which utilizes space–time behavior of wave phase-zeros or nodes, also suggests a kind of nanostructure that may be a base-N quantum computer of rational numbers. It also provides a wave-based derivation of special relativity that has extraordinary simplicity and clarity.

TWO-COMPONENT NODAL DYNAMICS

To introduce wave zero behavior in space–time, consider an interference of electromagnetic waves (photon or other mass-0 particle waves) such as is shown in Fig. 1a. Here a left-to-right-moving wave of amplitude $A_{\rightarrow} = 0.7$ collides with a right-to-left-moving wave of greater amplitude $A_{\leftarrow} = -0.9$. The result is a fixed wave envelope with standing wave ratio SWR of $valley:peak = (|A_{\leftarrow}| - |A_{\rightarrow}|)/(|A_{\leftarrow}| + |A_{\rightarrow}|) = -1:8$ for the wave magnitude $|\Psi| = \sqrt{\Psi * \Psi}$ which envelopes the real and imaginary parts $\mathrm{Re}(\Psi)$ and $\mathrm{Im}(\Psi)$. Inside $\pm|\Psi|$ is a "galloping" motion (*10*) of $\mathrm{Re}(\Psi)$ and $\mathrm{Im}(\Psi)$ which periodically exceeds and falls below the speed c of light by factors of SWR^{-1} and SWR. In the top frame of Fig. 1a, $\mathrm{Re}(\Psi)$ is a wave galloping at $-8c$ through a narrow opening of the envelope. Galloping motion is related to Kepler's law where orbiting particles slow at apogee and quicken at perigee (*10*).

Zeros of $\mathrm{Re}(\Psi)$ are open or solid dots which zigzag in the center space–time plot of Fig. 1a. First, $\mathrm{Re}(\Psi)$ shrinks to go

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Monte Carlo derived diffusion parameters for Ga on the GaAs(001)-(2×4) surface: A molecular beam epitaxy–scanning tunneling microscopy study

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The migration of individual Ga atoms on the technologically important GaAs(001)-(2×4) reconstructed surface has been studied as a function of substrate temperature and As_4 pressure using a combined molecular beam epitaxy and scanning tunneling microscope ultrahigh vacuum multichamber facility. We have deposited 10% of a plane of Ga onto a GaAs(001) surface with a low defect density (<1%) and with large terraces (>0.5 μm) to avoid the influence of surface defects like step edges and vacancies. Both the island number density and the geometry are measured and compared to Monte Carlo solid-on-solid simulations. Basic diffusion parameters, such as the activation energy, directional hopping-rate ratio, directional sticking-probability ratio, etc., are reported. © *2000 American Vacuum Society.* [S0734-2101(00)08204-X]

I. INTRODUCTION

The optical properties of III–V compound semiconductors are making high-speed global, wireless communications possible.[1] This application has created a demand for higher performance device structures, which are more complex and more difficult to realize. Unlike silicon-based devices, which are primarily fabricated using ion implantation,[2] III–V structures are formed at the surface by depositing one plane of atoms on top of another until the entire structure is formed. A more accurate atomic-scale understanding of the fundamental physics governing the motion of group III and group V atoms on III–V crystal surfaces would aid the fabrication process.

To date, several macroscopic methods, primarily reflection high-energy electron diffraction (RHEED), have been used to study Ga diffusion on the GaAs(001) surface.[3–9] In some of these studies, an estimate of the surface adatom diffusion length at one temperature is obtained from knowing the average terrace width at which the growth mode changes from two-dimensional island nucleation to step-flow mode. Interpretation of these experiments can be ambiguous because the influence of surface defects such as vacancies, step bunching, and interactions between adatoms themselves cannot be taken into account. Furthermore, since the electron beam averages over the entire substrate, temperature gradients and morphological variations across the wafer will effect this type of measurement. Some of these RHEED studies have utilized scanning tunneling microscopy (STM) to image the morphology as a function of multiple monolayers of coverage.[5,10] However, growing more than about 10% of a monolayer on a surface that is not flat, makes it difficult to separate the basic diffusion process from the large number of other interactions that can occur.

First-principles total-energy calculations of the pure migration of Ga atoms on the GaAs(001)-(2×4) reconstructed surface have been performed.[11] These calculations predict an activation barrier of 1.2 eV along the As dimer rows (i.e., along the $[1\bar{1}0]$ direction) and 1.5 eV across them (i.e., along the [110] direction). These state-of-the-art calculations are only true for the pure migration of individual Ga atoms on an otherwise perfect (2×4)-reconstructed surface in the absence of defects and an arsenic flux. These predictions have not been tested experimentally to date. The recent application of STM to the study of atomic diffusion on various elemental single crystal surfaces (e.g., Si and Fe) has significantly broadened the fundamental knowledge of the motion of atoms on these surfaces.[12–15] STM studies of diffusion on the GaAs(001) surface have also been carried out under metalorganic vapor-phase epitaxy conditions.[16,17] Unfortunately, these GaAs(001) diffusion studies do not provide an accurate test for the above theoretical predictions, since many other processes, like organic chemical reactions, are involved in the growth. Nucleation and growth on the molecular beam epitaxy (MBE)-prepared GaAs(001) surface has been experimentally investigated using STM and kinetic Monte Carlo simulations.[18] This study deposits a submonolayer amount of material and fits the island's geometry to simulations. Unfortunately, the study is only done at one temperature, making it difficult to address the diffusion properties.

Motivated by these issues, the activation energy for diffusion of gallium on the GaAs(001)-(2×4) reconstructed surface has been measured under an ultrahigh vacuum molecular beam epitaxy growth condition. Both the GaAs island number density and island geometry are measured from STM images for samples after depositing 10% of a plane of Ga atoms onto the GaAs(001)-(2×4) reconstructed surface held at various temperatures and exposed to different As_4 fluxes. This measurement is performed with atomic-scale resolution, naturally allowing for the influence of steps, vacancies, and other defects to be accounted for, further isolating the pure activation energy of diffusion. One could apply rate equation analysis to this data to determine the diffusion coefficient and activation barrier for isotropic two dimen-

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Some Portraits of a Hecht Hamiltonian: Molecular Rotations and Symmetry

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ABSTRACT

The development of lasers for spectroscopy of molecules has caused a renaissance of the theory of molecular symmetry and dynamics. Extensions of Hecht's original 1960 theory for the spectra of methane has been used to study ever more complex spectral structure of high symmetry molecules such as SF_6, C_8H_8, and C_{60}. A graphical description of eigensolutions to Hecht's Hamiltonian helps to understand complex rotational spectra and to probe the idea of spontaneous symmetry breaking. This provides a connection between Hecht's earlier work and nuclear physics and shows how nuclear spins can have extraordinary effects on molecular wavefunctions.

1. Introduction

In a 1960 paper Hecht[1] developed the theory of infrared spectral fine structure for the fundamental rotation-vibrational resonances of the tetrahedral methane molecule CH_4. This defining work has received hundreds of citations and continues to be a valuable reference. Because Hecht's theory was based upon quite general symmetry principles and Racah tensor analysis it has been applied to many other spherical top molecules including CF_4, SiF_4, C_8H_8 and the octahedral sulfur hexafluoride (SF_6) molecule[2-4]. The optical properties of SF_6 have probably been studied more than any other polyatomic molecule. It has been the subject of the highest resolution infrared spectra taken so far; less than ± 100 Hz at $\lambda = 10\mu$m or $\nu = 30$ THz[5-6].

The invention of the laser diode around 1972 made it possible to begin to resolve the rotation-vibration fine structure of heavy spherical top molecules. High resolution was necessary to help solve problems connected with molecular laser isotope separation. At that time there were considerable economic incentives to develop cheaper ways to obtain nuclear reactor grade uranium fuel. One scheme used tiny spectral shifts between vibrational modes of octahedral molecules $U^{235}F_6$ and $U^{238}F_6$. Also, the cold war was providing an incentive to produce weapons grade plutonium by a similar selection in PuF_6. The latter bit of history is not mentioned as often, but one must realize that no good theory, even Hecht's, is immune to having dubious applications. Fortunately, it now appears to have been too sophisticated for application in the Mesopotamian regions, so far.

We now have the hope that the end of the cold war might also end the demand for nuclear weapons. Peaceful applications of nuclear energy are still being developed, however a public distrust of nuclear power has temporarily reduced the demand for reactor fuel, as well. Therefore the economic incentive for separating heavy isotopes has gone away at least for the time being. Nevertheless, this program has yielded many advances in physical understanding, spawned new fields such as the study of intramolecular energy redistribution, and stimulated new developments of

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Rotation–vibration spectra of icosahedral molecules. III. Rotation energy level spectra for half-integral angular momentum icosahedral molecules

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Icosahedral spinor representation theory is developed for analyzing half-integral J eigenlevels of rotational tensor Hamiltonians for molecules such $B_{12}H_{12}^{-2}$, $C_{20}H_{20}$, and C_{60}. Large half-integral J eigenlevels for general sixth rank icosahedral tensors are discussed using different approximations and quantization schemes. Interpretations are made concerning some of the qualitative and semiquantitative spectral structure that is revealed.

I. INTRODUCTION

Mass spectroscopy observations by Curl and Smalley[1] and Kroto[2] of C_{60} clusters has stimulated considerable interest in the possibility of icosahedrally symmetric "Buckminsterfullerene" structures being generated during laser ablation of graphite. There is the related possibility that such highly symmetric ions and molecules occur in carbon-rich stellar nebulae and in some soot producing combustion processes. Recent infrared spectra of carbon dust particles deposited on Ge substrates by Kraetschmer, Fostiropoulos, and Huffman[3(a)] indicated the presence of icosahedrally symmetric C_{60} structures. Subsequently, their claimed observation of macroscopic solid C_{60} has received considerable attention.[3(b),3(c)] Also, the potential exists for spectroscopic studies of other icosahedrally symmetric structure such $C_{20}H_{20}$, which was synthesized by Pacquete *et al.*[4] the borohydride ion $B_{12}H_{12}$ (Ref. 5), and a C_{80} structure recently proposed by Chapman.[6] A recent paper by Ceulemans and Fowler[7] carried out an analysis of Jahn–Teller distortion for the fivefold degenerate state of a neutral icosahedral molecule.

All this has increased our need to understand the spectroscopic consequences of this extraordinary symmetry Y and its representations. (Here we use the conventional notation Y for the group.) The C_{80} structure is particularly intriguing since it is closely related to that of C_{60}; as seen in Fig. 1, both are composed of interlocking pentagons and hexagons. Each carbon in the C_{60} structure sits in an identical environment of C_v symmetry while C_{80} contains twenty "extra" carbon atoms which sit at points of C_{3v} local symmetry.

It is possible that spectroscopic investigation of these highly symmetric cage molecules might first be done using their positive or negative ions, which have lost or gained, respectively, a single electron. The mass spectroscopy[8] which indicates the existence of C_{60}, involves the positive ionic state, and it is well known that many cage molecules form stable negative ions. Recent developments in ion trap technology and ion spectroscopy raise the possibility for identifying and investigating high resolution ionic spectra of these structures. However, the symmetry analysis of singly charged ions is quite different from that of the molecules, which was treated in the preceding articles I and II.[9,10] One must deal with half-integer angular momentum which has very different symmetry properties. For example, the uncoupled half-integer spin of the electron changes sign under a rotation of 2π. This necessitates the use of multiplier or ray representations of the symmetry algebra. Conventional treatments of finite symmetry and crystal field splitting use the faithful representations of the point double groups in the case of half-integral J values. For larger groups, such as the icosahedral group, it helps to reduce computational labor by using a ray algebra,[11,12] in which the operations addition and subtraction are allowed. This has the effect of halving the size of the class algebra rather than doubling it. This symmetry analysis will be useful in analyzing combined spin–orbit-rotation and Jahn–Teller effects in icosahedral molecules with an odd number of electrons.

Nevertheless, there are quite a number of similarities between the symmetry and spectral analysis of half-integral J values and that of integral J values, particularly for the high J limit. In the classical limit the distinction of the two types is diminished. As will be shown here, many of the ap-

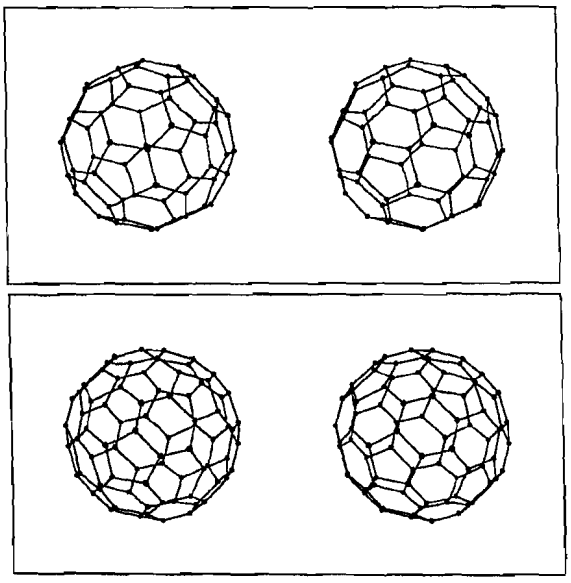

FIG. 1. Stereoscopic views of C_{60} and C_{80}.

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PREPARATION AND CHARACTERIZATION OF UNUSUAL LEVITATION Tl–Ba–Ca–Cu–O SAMPLES

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A preparation procedure for the Tl–Ba–Ca–Cu–O superconducting samples which can be levitated above or beneath a magnet is described. X-ray powder diffraction data for these samples are presented and discussed. Their electronic and magnetic properties are presented. The explanation for the unusual levitation is briefly discussed.

Discoveries of the 90 K Tl–Ba–Cu–O system[1,2] and the 120 K Tl–Ba–Ca–Cu–O superconducting system[3] have led to the identification of various superconducting compounds[4–8] and the observation of superconductivity in new Tl-based superconducting systems: Tl–Sr–Ca–Cu–O,[9–11] Tl–Pb–Sr–Ca–Cu–O[12] and Tl–*R*–Sr–Ca–Cu–O with *R* = rare-earths.[13] In addition to this progress, which has contributed to the understanding of the mechanism of high temperature oxide superconductivity and to the search for higher temperature superconductors, an interesting levitation phenomenon — a superconductor may be levitated above, to the side of, or beneath a magnet — has been observed for some Tl–Ba–Ca–Cu–O samples.[14] This kind of levitation is denoted as unusual in order to distinguish it from the normal levitation (only above a magnet). Previous examples of the unusual levitation were achieved for some AgO-doped $YBa_2Cu_3O_x$ samples.[15] While the rare-earth based-superconductors which display this unusual levitation must be doped with silver oxide, the Tl-based superconductors showing this behavior need to be specially prepared. In this letter, we describe the preparation procedure of the unusual Tl-based samples. According to our knowledge, this is the first description of a preparation procedure for high-quality unusual Tl-based samples which can be fabricated with great consistency

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COMPUTER GRAPHICAL AND SEMICLASSICAL APPROACHES TO MOLECULAR ROTATIONS AND VIBRATIONS

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Rotation–vibration dynamics and eigensolutions for several models are visualized using computer generated rotational energy surface and semiclassically quantized trajectories. Several numerical approximations to rotation–vibration eigensolutions are developed which use information gained from graphical presentations. Techniques are given for finding essential physical phenomena in complex numerical rovibronic eigenvalue problems. Possible alternatives to large scale numerical diagonalizations are introduced.

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Levitation effects involving high T_c thallium based superconductors

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The thallium based superconductor $Tl_2Ca_2Ba_2Cu_3O_{10+y}$ has been shown to exhibit very stable and unusual levitation equilibria in various arrangements involving this material and permanent magnets. Attractive and repulsive forces are evident in experiments in which samples are levitated above and below magnets. Photographs of these experiments and approximate quantitative discussions of the results are given.

The recent discovery of thallium based superconductors[1-4] with critical temperatures greater than 100 K has stimulated renewed interest in applications of materials with unusual magnetic properties. Of particular interest is the ability of these materials to be suspended or levitated in the presence of permanent magnets or to suspend magnets themselves. Levitation of rectangular magnets over rare-earth based superconductors has been discussed by Hellman *et al.*[5] and by Williams and Matey.[6] Hellman *et al.*[5] analyzed and measured the point of equilibrium in the vertical direction for a magnet over a flat rare-earth based superconductor, and they gave a qualitative discussion of the curious horizontal stability. Williams and Matey[6] also noted the curious horizontal stability and measured the frequency of torsional and translational motions of a magnet around its horizontal equilibrium above a flat superconductor.

For the thallium based superconductors, even more striking examples of stable levitation occur when either the magnet or the superconductor are suspended. There appears to be a continuous range of stable levitation positions for a magnet above a thallium based superconducting disk, and these disks may themselves be levitated above, to the side of, or *below* a ring magnet as shown in photographs given in the following. Previous examples of inverted levitation were achieved by Peters *et al.*[7] using a rare-earth based material, but in that case it was necessary to dope the material with silver oxide.

A spinning cylindrical Co-Sm permanent magnet is shown in Fig. 1 suspended above a cylindrical sample of a $Tl_2Ca_2Ba_2Cu_3O_{10+y}$ superconductor. The sample was prepared according to Ref. 1. The magnet is magnetized along its axis. A small amount of liquid nitrogen (LN) is maintained in an inverted styrofoam container which supports the sample. The magnet has a diameter and height of 0.3 cm and a mass of 0.167 g. The superconducting sample has a diameter of 0.8 cm and a thickness of 0.1 cm.

The magnet has a range of stable positions which can be achieved by simply forcing the magnet into them. The range of stable points for the center of the magnet comprises a roughly cylindrical volume which begins at a height of about 0.2 cm above the sample, extends up to about 0.4 cm, and includes all points within about 0.15 cm of the edge of the sample. In other words, the magnet can be placed so that it rotates stably with one end very near the edge of the sample or anywhere in between. In addition, the orientation of the magnet may be set somewhat arbitrarily. At each point it can be tilted to any angle within about 40° of the horizontal and it will usually stay there.

The stability of each position and orientation can be roughly tested by simply picking up the sample while it levitates the magnet and tilting or shaking the sample. Surprisingly, for some positions, the magnet remains near its equilibrium point even when the sample is tilted by more than 45°. The critical angle of repose depends on the orientation and position of the magnet, and provides an easy way to find the horizontal components of force needed to change the equilibrium position given the mass (m) or weight (mg) of the magnet.

$$f^{\text{critical}} = mg \sin \phi^{\text{critical}} . \tag{1}$$

The critical angle and force are greatest when the magnet is centered parallel to the sample and is tilted along its axis. It is also greatest when the point of equilibrium is as close as possible to the surface of the magnet.

The levitation of two disks of the thallium based superconductor by a ring magnet is shown in Figs. 2 and 3. The orientation in each figure can be verified by the falling of the cooled vapor which is visible in the lower part of the photographs. In each case the magnet is gripped in a pair of hand-held insulating pliers and is viewed edge-on and the samples appear to be suspended in magnetic pockets. The Co-Sm ring magnet has an outer diameter of 1.9 cm, an inner diameter of 1.0 cm, and it is 0.6 cm thick. The suspended thallium based

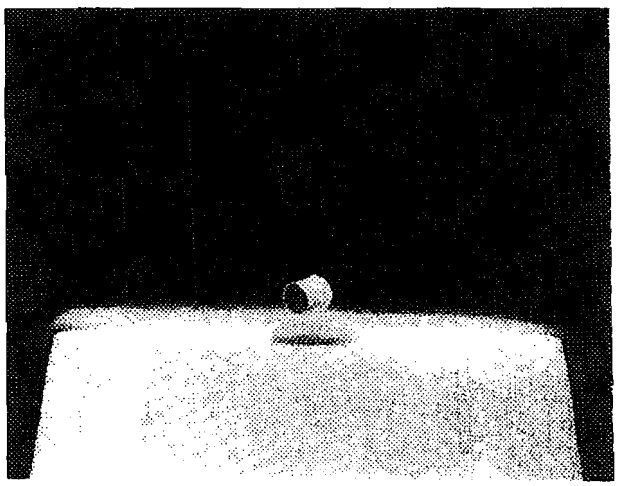

FIG. 1. Rotating magnet suspended above a thallium based superconductor behaves like a frictionless bearing.

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KRONIG-PENNEY BAND STRUCTURES FOR ONE DIMENSIONAL SUPERLATTICES

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ABSTRACT

Detailed calculations of high order band-gap boundaries are given as functions of potential barrier heights and widths for a range of superlattice structures. Prominent features of the structures include a multitude of band-gaps above the barrier as well as many band crossings. An interpretation of the crossings provides an approximate formula for estimating the band structure.

1. INTRODUCTION

The advent of molecular beam epitaxy[1] and plasma enhanced chemical vapor deposition[2] has resulted in the fabrication of high quality crystalline and amorphous multilayer superlattice structures. The potential energy distribution of these structures has been modeled by an infinite number of periodic square wells (Kronig-Penney model)[3-6] perpendicular to the superlattice layers (Figure 1). Experiments in photon absorption, carrier transport and resistivity have been performed on these structures with the results strongly supporting the Kronig-Penney model[4-10].

$SU(2)$ coordinate geometry for semiclassical theory of rotors and oscillators

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A spinor coordinate geometry is described which unifies a number of theoretical approaches to rotational and vibrational dynamics. Visualization aids previously developed for describing complex rotor spectra are extended and related to vibrational models. The relation is based upon analogies with optical polarization ellipsometry and spin rotation or precession. Rotational energy (RE) surfaces are used to describe a transition between normal and local modes in the presence of simple forms of Coriolis coupling and anharmonicity.

I. INTRODUCTION

During the past few years there have been a number of new approaches to the theory of molecular vibrations and rotations which complement those used in more conventional spectroscopic theory. The new approaches are partly a result of more revealing spectroscopic experiments which probe highly excited rovibronic states and dynamics. The new approaches provide ways to analytically model selected spectroscopic effects and to visualize more easily the underlying classical or quantum dynamics which may be concealed by a conventional numerical fitting of a molecular spectrum.

The purpose of this paper is to describe coordinate systems and geometrical methods which may help to connect and improve some of the new theoretical approaches. The main connection to be shown here is one which exists between the theory of two coupled vibrational degrees of freedom on one hand, and the theory of deformable quantum rotors on the other. Methods for visualizing rotor spectra in terms of quantizing trajectories on a rotational energy (RE) surface have been previously discussed,[1,2] and it will be shown how the same methods can be used to help describe certain vibrational models and effects.

One such effect is "dynamical tunneling" between different vibrational phase-space tori. An example of this was described by Davis and Heller[3,4] in connection with dynamics and spectra for a Barbannis vibrational potential. More recently, similar tunneling effects were treated in detail for the Henon–Heiles vibrational potential by Jaffe and Reinhardt.[5] An explicit connection will be shown between vibrational tunneling on one hand and rotor momentum space tunneling responsible for rotational superfine structure on the other.

The connection between rotational and vibrational models is based upon the relation between the three-dimensional rotation group $R(3)$ and the special (unimolecular) two-dimensional unitary group $SU(2)$ or spinor group. The use of $SU(2)$ and spinor operators for vibrational models has been developed by Kellman[6,7] and Lehmann[8] in treating normal to local mode transitions. A related discussion from a different point of view was made using the vibron model by van Roosmalen, Benjamin, and Levine[9] and van Roosmalen, Iachello, Levine, and Dieparink.[10] As will be shown in the present work, the rotational–vibrational connection provides a simple geometrical way to visualize the spectra and dynamics for a variety of models.

The first physical applications of the $R(3) \sim SU(2)$ or vector–spinor relation were made more than a century ago by Poincare[11] and Stokes[12] in their development of optical polarization ellipsometry. Ellipsometry is relevant to the coupled oscillator problem since a precise and convenient characterization for the evolution of a polarization ellipse can also be used to conveniently describe trajectories in oscillator phase space. The rotational description of oscillator dynamics given here is based partly on the polarization spinor geometry[13] of a commercially available ellipsometric instrument.[14] The theory behind spinor geometry has been traced back to Poincare, Stokes, and Hamilton[15] by Biedenhan and Louck.[16]

The spinor geometry and algebra will be reviewed using modern notation in Sec. II. The general form of coordinates for spinor states and operators will be described in Secs. II B and II C, respectively. The use of these coordinates will be introduced for coupled harmonic oscillators in Sec. III and physically interpreted in terms of polarization ellipsometry in Sec. IV. The connection between the ellipsometric description and the oscillator tori will be exhibited in Sec. V using stereo views into phase space. Finally, a rotational energy surface description of the dynamics of anharmonically coupled oscillators is given in Sec. VI where the analogy with deformable rotor dynamics is shown.

II. COORDINATES FOR DESCRIBING SPINORS

In this section general sets of coordinates will be developed for two-state systems and their operators. Probably, the most widely known two-state system is that of spin-$\frac{1}{2}$ or electron spin. The fundamental description of this is based upon a complex two-dimensional (spin-up, spin-down) space which is sketched in the upper left-hand corner of Fig. 1. A well-known representation of a spinor state is provided by a real three-dimensional spin vector space which is indicated in the upper right-hand corner of Fig. 1. One may prefer the vector space over the spinor space since the 3-space is real and more like the one in which we live. However, the 3-space only provides half the picture since a 180° rotation of spin-up into spin-down in the 3-space is only a 90° rotation in the original 2-space. The spinor state $|up\rangle$ and $-|up\rangle$ are represented by the same *up* vector in the 3-space.

Therefore it is helpful to find an intuitive way to view the spinor space directly. Such a viewpoint is suggested by the application of $SU(2)$ and $R(3)$ labeling to photon spin po-

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ABSTRACT

The development of lasers for spectroscopy of molecules has caused a renaissance of the theory of molecular symmetry and dynamics. Extensions of Hecht's original 1960 theory for the spectra of methane has been used to study ever more complex spectral structure of high symmetry molecules such as SF_6, C_8H_8, and C_{60}. A graphical description of eigensolutions to Hecht's Hamiltonian helps to understand complex rotational spectra and to probe the idea of spontaneous symmetry breaking. This provides a connection between Hecht's earlier work and nuclear physics and shows how nuclear spins can have extraordinary effects on molecular wavefunctions.

1. Introduction

In a 1960 paper Hecht[1] developed the theory of infrared spectral fine structure for the fundamental rotation-vibrational resonances of the tetrahedral methane molecule CH_4. This defining work has received hundreds of citations and continues to be a valuable reference. Because Hecht's theory was based upon quite general symmetry principles and Racah tensor analysis it has been applied to many other spherical top molecules including CF_4, SiF_4, C_8H_8 and the octahedral sulfur hexafluoride (SF_6) molecule[2-4]. The optical properties of SF_6 have probably been studied more than any other polyatomic molecule. It has been the subject of the highest resolution infrared spectra taken so far; less than ±100 Hz at $\lambda = 10\mu m$ or $\nu = 30$ THz[5-6].

The invention of the laser diode around 1972 made it possible to begin to resolve the rotation-vibration fine structure of heavy spherical top molecules. High resolution was necessary to help solve problems connected with molecular laser isotope separation. At that time there were considerable economic incentives to develop cheaper ways to obtain nuclear reactor grade uranium fuel. One scheme used tiny spectral shifts between vibrational modes of octahedral molecules $U^{235}F_6$ and $U^{238}F_6$. Also, the cold war was providing an incentive to produce weapons grade plutonium by a similar selection in PuF_6. The latter bit of history is not mentioned as often, but one must realize that no good theory, even Hecht's, is immune to having dubious applications. Fortunately, it now appears to have been too sophisticated for application in the Mesopotamian regions, so far.