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Quantum Mechanical Manifestation of Cantori: Wave-Packet Localization in Stochastic Regions

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Numerical calculations for a model anharmonic system interacting with a laser are used to analyze the quantum mechanical implications of classical structure in stochastic regions due to cantori [associated with the breakup of invariant Kolmogorov-Arnol'd-Moser surfaces). The numerical results show that a quantum wave packet may remain localized, even though classical orbits are strongly chaotic. Consequently, the quantum dynamics continues to exhibit "tunnelinglike" behavior even when diffusion is not classically forbidden.

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Over the past several years, theoretical work on nonlinear systems has shown that "chaotic orbits" can exhibit *nonrandom* behavior, even in strongly stochastic regions.[1-10] This behavior is attributable to the remnants of Kolmogorov-Arnol'd-Moser (KAM) tori, resonance islands, and separatrices which have broken up under the nonlinear coupling and are no longer invariant, yet they continue to influence the dynamics on an intermediate time scale. Coupled to these observations concerning classical stochasticity, comparisons between classical and quantum dynamics indicates that quantum mechanics tends to suppress the appearance of chaos.[11-19] These two sets of observations have lead to the suggestion that some large quantum effects may be associated with the classical structure that gives rise to the nonrandom behavior that can occur in chaotic regions.[6,8,10]

Significant progress has been recently made in the analysis of the classical structure in chaotic regions.[3-6] The recent work by MacKay, Meiss, and Percival[5] (Paper I) and Bensimon and Kadanoff[6] (Paper II) on the barriers due to cantori associated with the breakup of invariant KAM tori having irrational frequency ratios is of particular importance to the present study. (The dynamical barriers associated with the breakup of resonance zones and separatrices are treated in Refs. 4 and 5.) These papers present a classical theory for diffusion in two-degree-of-freedom systems which (1) suggests the location of the strongest barriers to diffusion, and (2) provides a methodology for constructing a phase-space representation of the barriers and calculating the fluxes and rates for transport across them.

In this Letter, we examine the response of quantum dynamics to the phase-space structure associated with cantori. Our primary purpose is to present numerical evidence that quantum mechanics may sense this underlying structure, and that its response is manifested in stronger localization and slower diffusion rates than predicted by classical dynamics. Thus, for strongly quantum mechanical systems these effects can be particularly strong since the quantum uncertainty would require a significant breakup of a KAM torus *before* the corresponding flux would be on the order of Planck's constant.

In this analysis, we will treat multiphoton dissociation in a model for vibrational anharmonic motion (a model for the HF diatomic molecule). The results should be qualitatively indicative, however, of the types of effects that may occur in other dynamical processes.[8] In addition, we will at this time focus on only one field frequency and intensity. How the effects reported in this paper depend on these two parameters will be treated elsewhere.[20]

The classical Hamiltonian for a nonlinear oscillator interacting with a monochromatic radiation field can

be written[21]

$$H_C = H_M + H_F + \lambda H_I, \tag{1}$$

where H_F and H_M are integrable Hamiltonians for the radiation field and the oscillator, respectively, and H_I is the field-molecule nonlinear, dipole interaction. These are given in atomic units (a.u.) as

$$H_M = p^2/2\mu + 0.225[1 - e^{-1.174(x-1.7329)}]^2, \tag{2a}$$

$$H_F = 1/2[P_F^2 + \omega_F^2 X_F^2], \tag{2b}$$

$$H_I = -X_F D(x), \tag{2c}$$

with

$$D(x) = -0.4541 x \exp(-0.0064x^2), \tag{2d}$$

where μ is the reduced mass ($\mu = 1744.59$ a.u.) and ω_F is the radiation-field frequency. The Hamiltonian H_C thus describes a two-degree-of-freedom, nonlinear system with conserved total energy, and is amenable to the classical theory in I and II. This system has been previously used in several classical studies of multiphoton excitation.[22-24]

It was found advantageous to use a time-dependent semiclassical Hamiltonian of the form

$$\mathbf{H}_Q = \mathbf{H}_M - E_0 \cos(\omega_F t)\mathbf{D}(x) \tag{3}$$

to describe the quantum dynamics. In Eq. (3), $\mathbf{H}_M$ and $\mathbf{D}(x)$ are quantum mechanical operators for the molecular Hamiltonian and dipole moment defined in Eq. (2). The molecule-field interaction is then incorporated as a periodically time varying dipole coupling between field-free Morse-oscillator eigenstates $|\chi_k\rangle$. The parameters E_0 and ω_F represent the radiation-field strength and frequency, respectively. The Hamiltonian $\mathbf{H}_Q$ has been shown to be equivalent to H_C in the limit of high-intensity fields, when the nonlinear coupling parameter λ is related to the field strength and intensity I by

$$\lambda = \omega_F E_0/(2E)^{1/2} = \omega_F (I/c\epsilon_0)^{1/2}, \tag{4}$$

where c is the speed of light, ϵ_0 is the permittivity of free space, and E ($4.13\times10^{+7}$ a.u.) is the conserved total energy of the laser-oscillator system. For the calculations reported here, $I = 2.9\times10^{+13}$ W/cm^2 ($\lambda/\omega_F = 1.00\times10^{-5}$ a.u.) and ω_F is 3922 cm^{-1} (0.00178 a.u.), shifted slightly to the red from the HF fundamental of 4138 cm^{-1}.

Dissociation is treated by[25] (1) discretizing (constructing an infinite wall at $x = x_* = 16.75a_0$) the continuum energy levels, and (2) associating a decay width γ_k with each discretized state $|\chi_k\rangle$ whose energy E_k is greater than the dissociation energy D_0, by defining the complex energy

$$E_k' = E_k - i\gamma_k. \tag{5}$$

The decay widths are defined by diagonal matrix elements of the optical potential

$$\gamma_k = \langle \chi_k | V_{\rm op} | \chi_k \rangle, \tag{6}$$

where

$$V_{\rm op}(x) = 0.02\{1 + \exp[-(x - x_*)/0.35]\}^{-1}. \tag{7}$$

The optical potential has been shown[25] to give good agreement with dissociation probabilities obtained by numerical integration of the time-dependent Schrödinger equation over a grid in two variables, x and t.

There are two advantages in formulating the quantum system as described above. First, because the nonlinear interaction is periodic in time, Floquet theory[26] can be used to provide a straightforward, nonperturbative method for following the time evolution of the molecular wave function, $\Psi_i(x,t)$ with $\Psi_i(x,t=0) = \chi_i(x)$. Second, since Ψ_i depends only on the diatomic internuclear distance and time, the Wigner transform,[27-29]

$$\Gamma_W^i(x,p,t) = (1/\pi h)\int e^{(2ips/\hbar)} \langle x - s/2|\Psi_i\rangle \times \langle \Psi_i | x + s/2\rangle \, ds, \tag{8}$$

can be unambiguously displayed on the x-p surface of section. Thus, H_C will first be used in describing the phase space and locating the strongest barriers to classical dissociation. Then, with use of $\mathbf{H}_Q$, the time development of the Wigner transform of a nonstationary state can be viewed relative to the topology of the classical barriers.

Figure 1(a) shows the Poincaré surface of section for the molecule-field Hamiltonian H_C, Eq. (1). The phase space is characterized by two distinct regions, with the boundary between them indicated by the dense set of points lying just exterior to the islands of a 3:2 resonance. This includes a predominantly regular (interior) region corresponding to bound motion and quasiperiodic energy exchange between the field and molecular degree of freedom; and a stochastic (exterior) region in which classical orbits will eventually dissociate. Note, in Fig. 1(a), that dissociation is indicated by the splatter of points extending out toward large positive values of x and tending to have negative momentum.

The work in I and II indicates that the strongest barriers to classical diffusion in two-degree-of-freedom systems arise from cantori having noble frequency ratios. These are cantori associated with the breakup of KAM tori with an irrational winding number, $\alpha = \omega_1/\omega_2$, where ω_1 and ω_2 are the two fundamental frequencies (in the present case, $\omega_1 = \omega_F$ and ω_2 is a dynamical frequency for the oscillator), whose continued-fraction representation,

$$\alpha = n + (a_1, a_2, \ldots, a_k, \ldots) = n + \cfrac{1}{a_1 + \cfrac{1}{a_2 + \ldots}}, \tag{9}$$

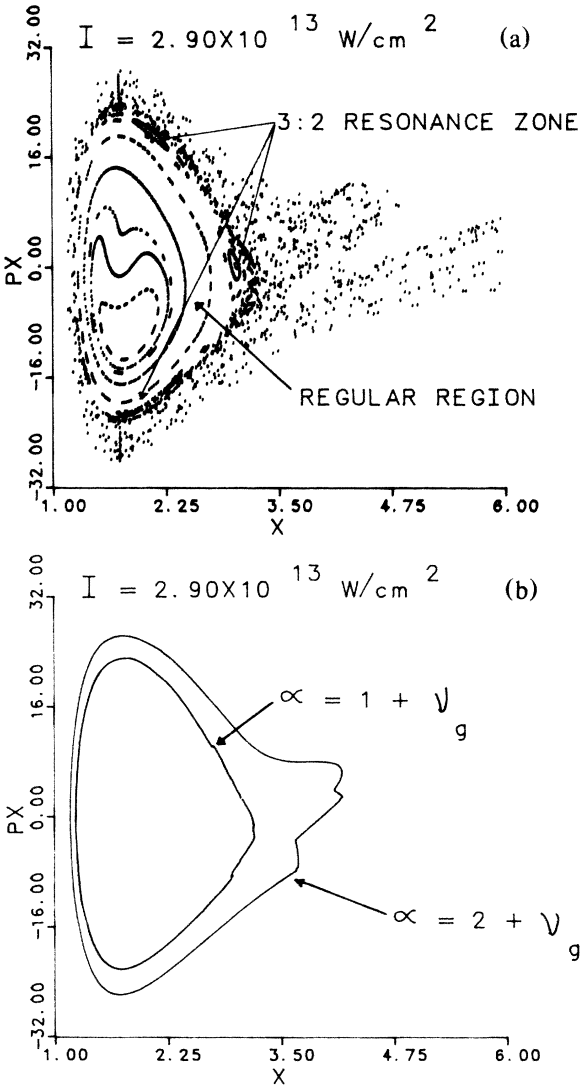

FIG. 1. (a) The x-p surface of section for the Hamiltonian describing the interaction between an anharmonic oscillator and an intense radiation field. (b) Phase-space representation of the cantori resulting from the breakup of tori with winding numbers equal to 1 and 2 plus the golden mean.

has $a_k = 1$ for all $k >$ a positive integer N. This is based on numerical evidence[1,2] which suggests that the flux through these barriers is a relative minimum. Since noble winding numbers are the most difficult irrationals to approximate by rationals (many terms are required in the truncated continued fraction), the strongest barriers to field-induced diffusion can be attributed to those regions of phase space in which the field and molecular frequencies are most out of resonance. In this sense, the strongest barriers will be associated with cantori in which $\alpha = n + v_g$, where $v_g(5^{1/2}-1)/2 = (1,1,1,1,1,\ldots) = 0.618\ldots$ is the venerable *golden mean*, and n is an integer.

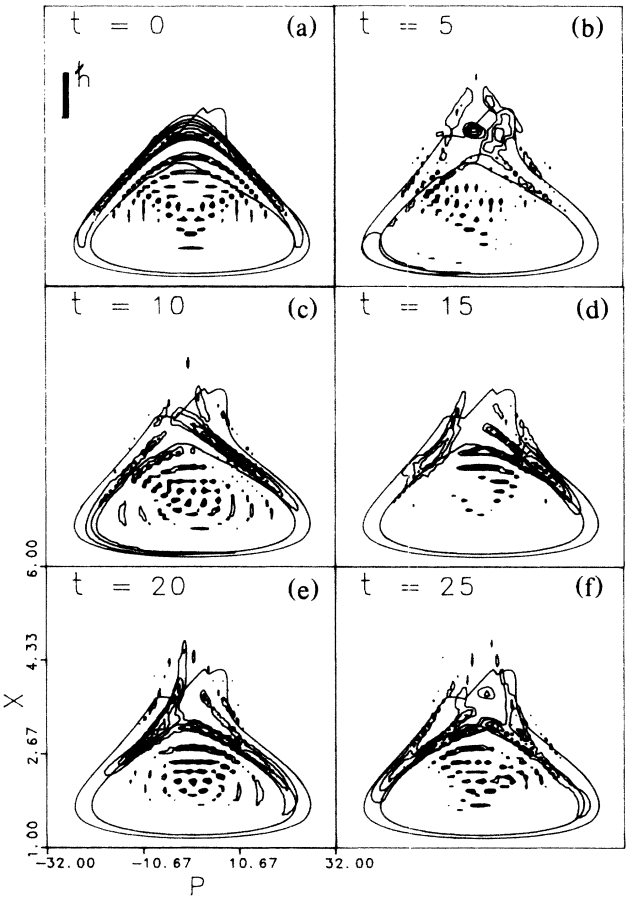

FIG. 2. Contour plots (positive contours only) for the Wigner transform of a nonstationary state for an anharmonic oscillator evolving in response to the the radiation field after (a) 0, (b) 5, (c) 10, (d) 15, (e) 20, and (f) 25 field periods ($\tau = 0.054$ ps). The cantori shown in Fig. 1(b) are also shown in each of the diagrams (a)–(f). A box indicating the size of $\hbar$ is shown in (a).

Using the techniques described in I and II, we can construct a representation, on the surface of section, of the cantori which tend to be the strongest barriers. These are shown in Fig. 1(b) and correspond to the breakup of KAM tori with rotation numbers equal to $1+v_g$ and $2+v_g$. (We were unable to generate cantori with $\alpha = n + v_g$ with $n > 2$ because the phase space in these regions is too unstable.) The classical flux (per mapping on the surface of section) across $1+v_g$ was 0.013 a.u., and the flux across $2+v_g$ was 2 orders of magnitude larger, 1.163 a.u. (Recall that $\hbar = 1$ in atomic units.)

Figure 2 shows several contour plots of the Wigner transform of the time-dependent wave function Ψ_i at selected times during the first 25 field periods ($\tau = 0.054$ ps). For clarity, only positive contours are shown. Contours were taken in fixed increments, with the smallest value corresponding to between $\frac{1}{5}$ and $\frac{1}{6}$

of the maximum.

The system initially was in the fifteenth field-free oscillator eigenstate, Fig. 2(a), which is localized predominantly in the region of $2+v_g$. The subsequent time development, indicated in Figs. 2(b)–2(f), indicates that the dynamics is dominated by an oscillation in the classically stochastic region localized *between* 1 and 2 plus the golden-mean cantori. Examination of state-to-state transition probabilities indicates that this oscillation corresponds to a Rabi cycling of amplitude between the eleventh, thirteenth, and fifteenth oscillator eigenstates, with the primary contribution arising from states 11 (which is localized in the region of the $1+v_g$ cantorus) and 15.[30] During the 100 field periods for which the dynamics was followed, there was never any buildup in probability outside the $2+v_g$ cantorus, except for the type of small extensions to positive x in the Wigner transform that are exhibited in Fig. 2. Dissociation results from leakage of probability from these extensions. The wave packet thus senses the underlying $2+v_g$ cantorus, which blocks its dissociation, even though classical trajectories easily penetrate it!

In summary, cantori in classical stochastic regions of phase space, while permitting extensive classical flow, may effectively act as barriers to quantum wave packet evolution. As a result, the wave packet remains well localized for long periods of time in the classically stochastic region.

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