High-harmonic generation in a driven triangular well: The implications of chaos

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We compute the spectrum of emitted radiation for a particle in a triangular potential well and driven by an electromagnetic field. For intense fields, the spectrum exhibits a plateau structure, a series of harmonic peaks of comparable strength up to some high-harmonic order, similar to that found in experiments and similar theoretical models. By comparing the quantum and classical dynamics, we see that the onset of high-harmonic generation in the quantum system appears to be correlated to the onset of chaos in the classical system. We also consider the effect of a slow turn on of the field and show why the acceleration should be used instead of the induced dipole moment to compute the spectrum.

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I. INTRODUCTION

Recent experiments by Ferray *et al.* [1] have demonstrated the production of very high harmonics in laser irradiation of rare gases using intense 1064 nm 30 ps pulses. More recently, harmonics of order 135 have been reported [2]. The harmonics exhibit a plateau structure: a series of odd harmonics of comparable strengths up to some high harmonic order, after which they fall off rapidly. The harmonic peaks do not fall off as some simple power law of the intensity equal to the order of the process, so perturbation theory fails to explain high-harmonic generation (HHG). Simple theoretical single-atom models have been shown to exhibit HHG very similar to that found in experiments [3,4]. The success of these models leads us to study further the HHG of simple quantum wells subject to a strong external time-periodic field. In this paper we compute the radiation spectrum for a particle in a driven triangular potential well. The triangular potential well is convenient in that many calculations of both the quantum and classical dynamics can be done analytically or with a minimum of numerical computation. We briefly explain why we use the acceleration spectrum rather than the more commonly used induced dipole moment as the spectrum of emitted radiation. We include the effect of a *pulse rise time* on the spectrum. An extremely rapid or sudden turn on of the external field can produce many intermediate peaks, or *shifted harmonics*, of comparable strength to the harmonic peaks. These peaks are of the same type predicted by Bavli and Metiu [5] and Cocke [6] in similar models. The triangular potential well has been shown [7] to exhibit both dynamic localization and delocalization as parameters are varied. We compare the spectra generated in both cases. The dynamic localization of the particle wave packet appears to have a dramatic effect on the emitted spectrum, as we shall see. Finally, we compare the dynamics of the quantum and corresponding classical system and find that HHG in the quantum system appears to be associated with the onset of chaos in the classical system. We believe these results to be fairly general, even though this model does not represent all aspects of a real atom: it has no continuous spectrum, (i.e., ionization is not possible), and it is asymmetrical and thus even harmonics are not forbidden. Nevertheless, the model may be relevant to atomic systems in which the bound states play a dominant role in the dynamics. In particular, the model may have relevance to systems in which the field intensities are strong enough to lead to strong HHG generation, but too weak to rapidly ionize the system.

II. MODEL

Let us consider a classical system consisting of a time-periodically driven particle in a triangular-shaped well formed by linear potential (constant force) with an infinite barrier (wall) at one end. The Hamiltonian for this system is

$$H=\frac{p^2}{2m}-\epsilon_0 x-\epsilon x\,\cos(\omega t+\phi_0)+V_L(x), \tag{1}$$

where ϵ, ω, and ϕ_0 are the field strength, frequency, and phase of the electromagnetic driving field, respectively, ϵ_0 is the strength of linear potential, and L is the location of the wall. The potential $V_L(x)=0$ for $x<L$ and $V_L(x)=\infty$ for $x>L$. The presence of the electromagnetic field will create an infinite array of nonlinear resonances. A nonlinear resonance occurs when a multiple of the field frequency is equal to a multiple of the orbit frequency of the particle in the triangular well. This can be seen by writing the Hamiltonian, Eq. (1), in terms of action-angle variables, $[J,\theta]$ (with $\phi_0=0$) [8],

$$H=E_0(J)-\epsilon\sum_{l=-\infty}^{\infty} x_l(J)\cos(l\theta-\omega t), \tag{2}$$

where $J=1/2\pi\oint p\,dx=[(2\sqrt{2m})/3\pi\epsilon_0]\,(E_0+\epsilon_0 L)^{3/2}$, E_0 is the unperturbed Hamiltonian, and $x_l\propto J^{2/3}$. The external field can now be seen to induce an infinite series of traveling waves into the phase space of the system. Each traveling wave may trap a phase space trajectory and gives rise to a nonlinear resonance. The condition for resonance is $l\dot{\theta}=s\omega$, s integer. If we increase the field intensity, the resonances, which appear as localized pendulumlike topological changes in the phase space, will "grow" larger and overlap, leading to chaos. We shall show an example in Sec. V.

Let us now write the quantum version of the model. For the unperturbed case ($\epsilon=0$), the Schrödinger equation for stationary states, $\Psi_i(x)=\langle x|E_i\rangle$, with energy E_i, is given by

$$-\frac{1}{2m}\frac{\partial^2\Psi_i}{\partial x^2}=(E_i+\epsilon_0 x)\Psi_i \quad \text{for } x\leq L. \tag{3}$$

The solution is given in terms of Airy functions:

$$\Psi_i(x)=N_i\mathrm{Ai}[-\{(2m)^{1/3}\epsilon_0^{-2/3}E_i+(2m\epsilon_0)^{1/3}x\}], \tag{4}$$

where N_i is the normalization constant. The energy eigenvalues E_i can be found from the condition that $\Psi_i(L)=0$. The eigenspectrum is purely discrete and the eigenvalues E_i are proportional to $i^{2/3}$, in correspondence to the classical case where $E_0(J)\propto J^{2/3}$. We compute the acceleration and induced dipole moment of the perturbed system ($\epsilon\neq 0$) by integrating the Schrödinger equation in the unperturbed energy basis,

$$i\frac{dc_j}{dt}=E_j c_j+\sum_k c_k x_{jk}\xi(t)\cos(\omega t), \tag{5}$$

where $|\psi(t)\rangle$ is the probability amplitude of the particle at time t, $c_j(t)=\langle E_j|\psi(t)\rangle$, and the turn-on function $\xi(t)=\epsilon\sin^2(\omega t/4n)$ for $t<2\pi n/\omega$ and $\xi(t)=\epsilon$ for $t>2\pi n/\omega$. The turn-on function is used to simulate the pulse rise time typical of experiments. In the turn-on function $\xi(t)$, n is the number of cycles for the field to rise to maximum strength. The expectation value of the acceleration at time t is given by

$$\langle\psi(t)|\ddot{x}|\psi(t)\rangle=\sum_{ij}\langle\psi(t)|E_i\rangle\langle E_i|\ddot{x}|E_j\rangle\langle E_j|\psi(t)\rangle$$
$$=\sum_{ij}c_i c_j^*\ddot{x}_{ij}, \tag{6}$$

where

$$\ddot{x}_{ij}=-(E_j-E_i)^2 x_{ij}-\sum_k[2E_k-E_j-E_i]x_{ik}x_{kj}\epsilon\cos(\omega t) \tag{7}$$

and the dipole matrix elements are given by [7]

$$x_{ij}=\begin{cases}\dfrac{-\epsilon_0}{m(E_i-E_j)^2} & \text{if } i\neq j\\[2ex] \dfrac{2E_i}{3\epsilon_0} & \text{if } i=j.\end{cases} \tag{8}$$

The *induced* dipole moment is just given by

$$\langle\psi(t)|x|\psi(t)\rangle=\sum_{ij}c_i c_j^* x_{ij}. \tag{9}$$

In order to obtain the power spectrum of the acceleration, we compute the time series of the expectation value of the acceleration and then take the modulus squared of its Fourier transform.

In the strongly driven classical system we have overlap of nonlinear resonances leading to chaos. In that case, the particle can sample a wide range of energies, and its subsequent motion can be described by a diffusion process. In quantum systems we have instead a significant broadening of the Floquet eigenstates [9] (these are sometimes called quasienergy eigenstates). This broadening generally falls into two regimes: localized (due to dynamic Anderson localization), and delocalized or extended. Localized Floquet states are distributed over a relatively few unperturbed basis states and fall off exponentially, whereas delocalized Floquet states extend over nearly the entire set of unperturbed basis states (thus making computations difficult when a finite basis is used). When delocalization occurs, the diffusion of the energy of the quantum particle follows that of the corresponding classical particle. For localized systems this diffusion is suppressed. An approximate condition for delocalization of a driven quantum particle in a triangular potential well has been given by Benvenuto *et al.* [7]. They found that delocalization occurs when $\epsilon>\frac{1}{2}\omega^{3/2}$. This criterion agreed qualitatively with numerical checks that they made.

III. ACCELERATION VERSUS DIPOLE MOMENT

While most researchers compute the modulus squared of the Fourier transform of the induced dipole moment as the power spectrum for single atomic systems or quantum wells, Burnett *et al.* [10] and Sundaram and Milonni [3] have pointed out that the spectrum of emitted radiation is obtained directly from the expectation value of the acceleration, and the power spectrum of the induced dipole moment can be misleading. In principle, one could obtain the acceleration from the dipole moment: the Fourier transform $X(\omega)$ of the acceleration is related to the Fourier transform of the dipole moment as follows:

$$X(\omega)=\frac{1}{\sqrt{2\pi}}\int_0^T dt e^{-i\omega t}\ddot{x}(t)$$
$$=\frac{1}{\sqrt{2\pi}}\left(e^{-i\omega T}\dot{x}(T)+i\omega e^{-i\omega T}x(T)-\omega^2\int_0^T dt e^{-i\omega t}x(t)\right), \tag{10}$$

where T is the pulse length [the initial position and velocity are assumed to be zero in Eq. (10)]. If the final position and velocity of the electron is zero then the power spectrum of the acceleration can be obtained by multiplying the power spectrum of the dipole moment by ω^4. However, for strongly perturbed or ionizing systems, this is not the case. Burnett *et al.* [10] and Tsin-Fu Jiang and Shih-I Chu [11] give examples where there are large discrepancies in the computed spectra when there is ionization present. In these cases the final displacement of the ionized electron can be quite large. Thus it may be numerically difficult, if not impossible, to obtain the acceleration spectrum from the Fourier transform of the dipole moment. For the model considered in this paper, there is no possibility of ionization. Nevertheless, the final position and velocity can be very large. In Figs. 1(a) and 1(b), we show the power spectra computed from the

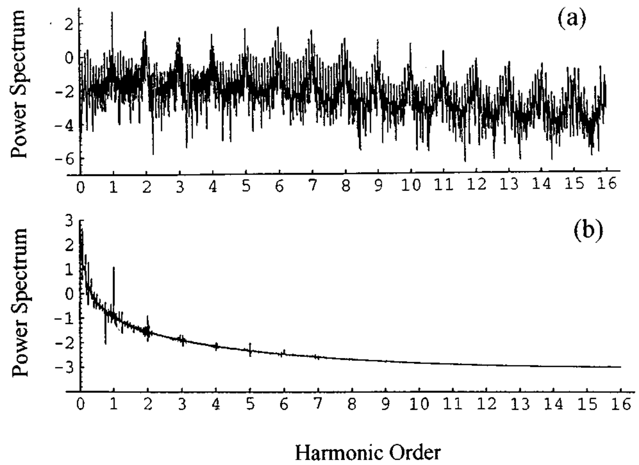

FIG. 1. Power spectrum for the system for the parameters $\omega=2.52$, $\epsilon=0.5$, $\epsilon_0=0.4$, and $T=256$ optical cycles (sudden turn on) with the initial state $n_0=60$ using the expectation value of (a) the acceleration and (b) the induced dipole moment.

acceleration and dipole moment, respectively, for the parameters $\omega=2.52$, $\epsilon=0.5$, $\epsilon_0=0.4$, and $T=256$ optical cycles (sudden turn on) with the initial state $n_0=60$. The initial state was chosen for comparison with the results of Benvenuto *et al.* [7]. In Fig. 1(b) there is a spurious background due to the first two terms of Eq. (10) being nonzero. Also, the peak heights are diminished, especially for the higher harmonics, because of the absence of the ω^4 factor.

IV. EFFECT OF PULSE RISE TIME ON SHIFTED HARMONIC GENERATION

It has been shown, for systems similar to that presented here, that a sudden turn on of the driving field can produce *shifted* harmonic peaks of the same order of magnitude as the purely harmonic ones [5,6]. These shifted harmonic peaks are located at frequencies $q_\alpha - q_\beta + m\omega$ (m integer, $q_\alpha, q_\beta \in [0,\omega]$) where q_α and q_β are quasienergies (Floquet eigenvalues) of the system. The shifted harmonics occur at intermediate frequencies to the pure harmonics and may be of comparable order of magnitude in strength. In Fig. 2(a) we show the power spectrum for the parameters $\omega=2.52$, $\epsilon=0.5$, $\epsilon_0=0.4$ with sudden turn on [$n=0$ in Eq. (5)] of the field for a pulse length of 256 cycles. For these parameters, the quantum states are localized. We see many shifted harmonics, particularly about a dozen between the fifth and sixth pure harmonics, suggesting that about a half dozen or so quasieigenstates significantly comprise the initial state, $n_0=60$ [5 quasienergies can produce up to $5!/(3!2!)=10$ quasienergy differences]. In Fig. 2(b), we show the power spectrum for the system in the delocalized regime, where now $\omega=0.6$. As expected, many more quasieigenstates are relevant to the evolution and thus there are many more peaks. In Fig. 3, we show the power spectrum for the system with the same parameters as in Fig. 2(a), but with pulse rise time of 3(a) 4 and 3(b) 12 optical cycles, respectively. The shifted harmonic peaks are diminished considerably for the slower rise time, while the purely harmonic ones are relatively unaffected. Thus the absence of shifted harmonics suggests that the turn on adiabatically moves the system into a single Floquet eigenstate. Twelve optical cycles is a relatively short time by experimental standards; thus it will be very difficult to observe shifted harmonics in the laboratory. However, we have not considered other pulse shapes or other frequency regimes such as the on-resonance case (the driving frequency equal to the transition frequency, $E_i - E_j$) where Rabi oscillations would be stronger.

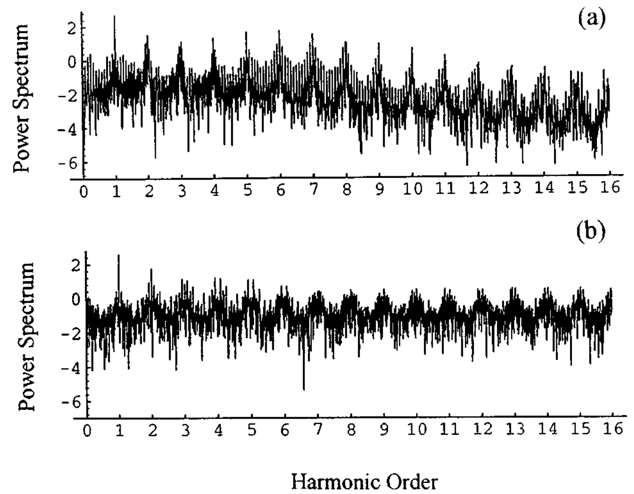

FIG. 2. The power spectrum of the acceleration. (a) The localized regime with $\omega=2.52$, $\epsilon=0.5$, $\epsilon_0=0.4$ with sudden turn on of the field for pulse length $T=256$ optical cycles. (b) The delocalized regime with parameters $\omega=0.6$, $\epsilon=0.5$, $\epsilon_0=0.4$ with sudden turn on of the field for pulse length $T=256$ optical cycles.

V. HIGH HARMONIC GENERATION AND VARIATION OF FIELD INTENSITY

We now look at the power spectrum of the acceleration as the intensity is varied for constant frequency. In Fig. 4, we show the power spectra for the parameters $\omega=2.52$, $n_0=60$, $\epsilon_0=0.4$ and for the field strengths $\epsilon=0.01$, 0.1, and 0.25 and a sudden turn on of the field. It appears that at some critical field strength between $\epsilon=0.1$ and 0.25 there is sud-

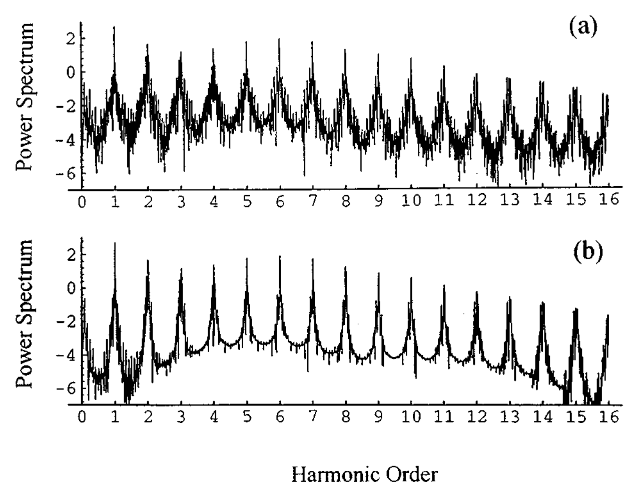

FIG. 3. Power spectra of the system for the parameters $\omega=2.52$, $\epsilon=0.5$, $\epsilon_0=0.4$ with pulse rise time of (a) 4 and (b) 12 optical cycles.

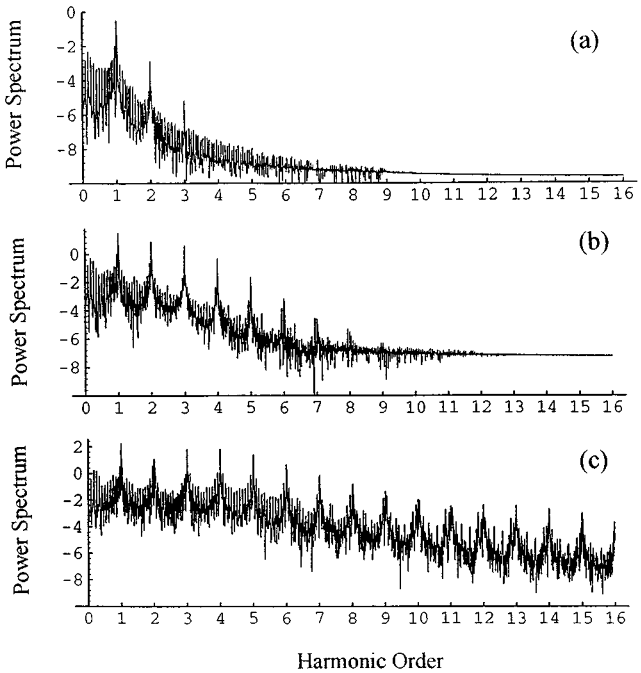

FIG. 4. Power spectra of the system for the parameters $\omega=2.52$ and (a) $\epsilon=0.01$, (b) $\epsilon=0.1$, (c) $\epsilon=0.25$.

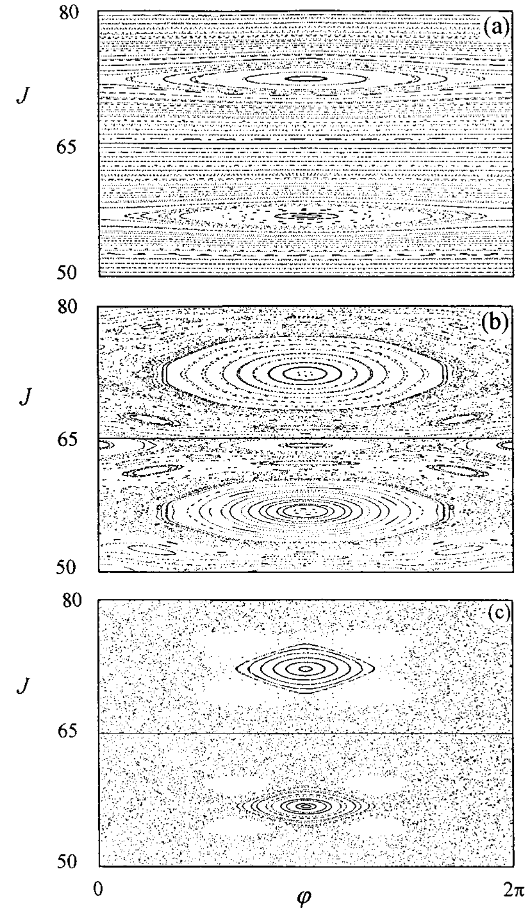

FIG. 5. Strobe plots of the classical system for the same parameters as in Fig. 4.

den generation of very high harmonics. (Note only 16 are shown.) In order to gain some insight to this sudden increase in harmonics, let us look at the corresponding classical dynamics of the system. In Fig. 5 we show classical phase space strobe plots of the action J versus phase angle ϕ of the external field (not the canonical angle θ) after each orbit for an ensemble of initial conditions near $J=60$ for the same parameters as in Fig. 4. In Fig. 5(a), the system is relatively unaffected by the external field except in the vicinity of the two resonances shown. The resonances are separated by KAM tori, and the particle motion is stable throughout the phase space shown. In Fig. 5(b) the resonances have increased in size, but are still separated by KAM tori. In Fig. 5(c) the resonances have overlapped, and all KAM tori separating the resonances are destroyed. The motion is then chaotic throughout most of the phase space. Comparing Figs. 4 and 5 it seems that HHG occurs when the classical system becomes fully chaotic. It has been shown for many systems [9] (microwave driven hydrogen, for example), multiphoton ionization is associated with the onset of chaos in the corresponding classical system. Thus for atomic systems, HHG may generally be expected to occur near the ionization threshhold. Indeed, Ferray *et al.* point out that "...harmonics are only generated in a laser intensity range very close to that required for multiphoton ionization of the medium [1]." However, in general, ionization is not necessary for HHG to occur. For example, HHG also occurs in a two-level model [3]. HHG seems particularly efficient when there is localization or quantum suppression of classical chaos. Therefore higher frequencies may lead to more efficient harmonic generation, since the delocalization threshhold is higher. This is the same conclusion reached (for perhaps different reasons) by Lewenstein *et al.* [12] and Corkum [13] in their atomic models, though the connection between our model and theirs requires more study.

VI. CONCLUSION

We have presented a very simple model that exhibits HHG very similar to that found in recent experiments. While our model is not directly applicable to the experiments of Ferray *et al.*, the model may nevertheless provide some insights. For example, it may lead to an explanation of why HHG occurs near the multiphoton ionization as reported by Ferry *et al.* An understanding of the corresponding classical dynamics may lead to a deeper understanding of HHG, much as it has for multiphoton ionization. While classical and quantum systems do not have the same spectra, the radiation laws governing them are nonetheless very similar. Thus comparing the quantum and classical dynamics may be insightful. Also of interest is when the classical and quantum dynamics are very different, as in the case of dynamic localization. Here we may find new regimes of efficient HHG. The triangular well is particularly useful because it is one of the simplest models that exhibit both dynamic localization and delocalization.

We should point out that with regard to the experiments mentioned above, a fairly developed theory has emerged [12,13]. We think the approach provided here could provide a complementary view. We think the results presented here are fairly general and may apply to a wide variety of quantum systems.

As a final comment, we note that in any laboratory experiment, there will usually be an ensemble of quantum wells or atoms, and one has to be concerned about the matching of the phases of the emitted radiation from all of the wells or atoms. For the conditions of the experiments mentioned above, a tightly focused beam and low pressure, L'Huillier *et al.* [14] have found that the phased-matched results do not differ significantly from the single-atom response. Therefore, the study of the spectra of single atoms or wells may be very useful in furthering our understanding of actual experimental results.

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