

# Classical versus quantum harmonic-generation spectrum of a driven anharmonic oscillator in the high-frequency regime

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The harmonic-generation spectrum of an anharmonic oscillator perturbed by a high-frequency, high-intensity external field is discussed from both classical and quantum mechanical points of view. The classical theory of harmonic generation in this regime is shown to provide a simple analytical expression for the probability to emit the  $k$ th harmonic of the external field frequency. The theory is applied to the harmonic generation by a charged particle moving in Morse and “soft-core” Coulomb model potentials and interacting with a time-periodic electric field. For both models the probability to emit the  $k$ th harmonic attains a maximal value at some  $k_{\max}$  so that a single harmonic peak dominates the spectrum.  $k_{\max}$  can be controlled by properly choosing the field and oscillator parameters. The quantum mechanical numerical results are shown to be in excellent agreement with the classical predictions due to the localization of the wave function around either stable (in the case of Morse potential) or unstable (in the case of “soft-core” Coulomb potential) equilibrium position of the corresponding effective Kramers-Henneberger potential. [S1050-2947(98)04202-4]

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## I. INTRODUCTION

Atoms being irradiated by strong ac fields emit odd harmonics of the incident radiation frequency. This phenomenon, called harmonic generation (HG), has recently attracted a lot of attention from both experimentalists and theorists. The probability  $\sigma_k$  to emit the  $k$ th harmonic is associated with the Fourier transform of the dipole moment acceleration:

$$\sigma_k \propto \lim_{T \rightarrow \infty} \left| \int_0^T \langle \ddot{\mu} \rangle(t) e^{-ik\omega t} dt \right|^2. \quad (1)$$

It has been found [1] that at field intensities beyond the perturbative regime the HG spectrum exhibits a plateau extending to some high harmonic order, which ends by an abrupt cutoff. The typical experimental situation is that the radiation frequency is much lower than the transition frequency between the adjacent atomic states. The quantum mechanical (QM) theory of HG in the low-frequency-high-intensity regime has been developed by Lewenstein and co-workers [2,3] following the earlier numerical studies, e.g., [4] and theoretical works, e.g., [5]. A simple quasiclassical description of HG in this limit due to Kulander, Schafer, and Krause [6] and Corkum [7] represents it as a three-stage process. In the course of this process an electron first tunnels through an adiabatically oscillating (“quasistatic”) potential barrier. The electron appears then in the continuum at some phase of the low-frequency, high-intensity field. Its further motion is affected to a good approximation by the field only. The initial field phase can be such that the electron returns to the vicinity of the ionic core and radiatively recombines to produce the neutral atom in its ground state. The highest harmonic emitted is thus determined by the maximal kinetic energy of the electron at the moment of its encounter with the core and by the atom’s ionization potential:

$$\Omega_{\max} \approx \frac{I_p + 3.17U_p}{\hbar}, \quad (2)$$

where  $U_p = e^2 f_0^2 / 4m\omega_f^2$  is a ponderomotive potential,  $I_p$  is the ionization potential,  $e$  is the electron’s charge,  $f_0$  is the ac field amplitude,  $m$  is the electron’s mass,  $\omega_f$  is the field frequency, and 3.17 is a numerical factor determined in the classical simulations [6,7]. Its value has been confirmed by the fully QM treatment [2].

HG, however, is not a purely QM phenomenon. As a matter of fact,  $\sigma_k$  of Eq. (1) can be calculated classically if the expectation value of the dipole moment acceleration is replaced by its average over classical trajectories. The HG spectra obtained in classical trajectory calculations [8–10] exhibit several characteristic features of the experimental ones, but do not match them quantitatively. The classical theory of HG in the low-frequency regime has been developed by Leopold and Richards [11]. It was found that the classical HG spectrum consists of bands of harmonic peaks centered around the multiples of the mean atomic frequency. The theoretical results for a one-dimensional hydrogen model were found to be consistent with the three-dimensional classical simulations of Bandarage *et al.* [8,9] (see also [10]). As discussed by Leopold and Richards, the theory is limited to the regular classical dynamics. It is not applicable to the high intensities, at which the motion becomes largely chaotic. The attempts to trace the implications of classical chaos on the HG spectra of quantum systems have been made recently by Averbukh and Moiseyev [12] and Cocke and Reichl [13].

The main purpose of this article is to present a simple classical theory of HG in the high-frequency limit. The general discussion is presented in Sec. II. Newton’s equation in the Kramers-Henneberger (KH) frame [14,15] is shown to provide a direct route to calculation of the acceleration Fourier transform. We employ Kapitza’s idea [16] of separating

the motion under the influence of a high-frequency field into large amplitude slow motion and fast oscillations of small amplitude. Simple expressions for  $\sigma_k$  are derived within the framework of the first-order perturbation theory for both continuous wave (cw) and pulse cases. The general theory is first applied to the model of the Morse oscillator in the high-frequency, high-intensity cw laser field, studied previously by Jiang [17]. The  $k$ th harmonic intensity is shown to go through a maximum,  $k_{\max}$  being field controllable. The results, presented in Sec. III, are shown to be in excellent agreement with both classical and QM (Sec. IV) numerical calculations. In Sec. V we show that even in the problem with largely chaotic phase space, such as the one-dimensional ‘‘soft-core’’ Coulomb model in the high-frequency, high-intensity external field, the simple classical theory still applies due to the existence of the ‘‘scarred’’ quasienergy states. In Sec. VI the limits of applicability of the classical theory are discussed and the conclusions are drawn.

## II. A CLASSICAL THEORY OF HARMONIC GENERATION IN THE HIGH-FREQUENCY REGIME

Consider an anharmonic oscillator in an external time-dependent field. The Hamilton function of the system has the following general form:

$$H(X, P, t) = \frac{P^2}{2m} + V(X) + \mu(X)f(t), \quad (3)$$

where  $m$  is the mass (reduced mass) of the moving particle(s),  $\mu(X)$  is the coupling, say, a dipole moment, and  $f(t)$  is the field.

Let us assume that the coupling term in Eq. (3) is linear in coordinate:  $\mu(X) = \mu_1 X$  (which is usually a reasonable approximation). Then the Hamilton function reduces to the so-called length gauge Hamilton function  $H_L(X, P, t)$ :

$$H_L(X, P, t) = \frac{P^2}{2m} + V(X) + Xf(t), \quad (4)$$

where the coupling strength constant  $\mu_1$  is incorporated into  $f(t)$ . The specific case of interest in the present work is when the frequency of the time-dependent field is much higher than that of the unperturbed motion. The KH frame has been shown to be an effective theoretical tool for investigation of both classical and quantum dynamics of ionizing (dissociative) systems in high-frequency fields [18,19]. In classical mechanics one can obtain the Hamilton function in the KH frame by means of a canonical transformation. The corresponding generating function of the second kind,  $F_2(X, p, t)$ , is given by

$$F_2(X, p, t) = Xp + h(t)p + g(t)X, \quad (5)$$

where

$$g(t) = -\int^t f(t') dt' + C_1, \\ h(t) = (1/m) \int^t \int^{t'} f(t'') dt'' dt' - (C_1/m)t + C_2.$$

The resulting Hamilton function in the KH frame,  $H_{\text{KH}}(x, p, t)$ , is

$$H_{\text{KH}}(x, p, t) = \frac{p^2}{2m} + V(x - h(t)) \quad (6)$$

(here we dropped the terms which depend on time only). The advantage of such transformation can be understood by comparing Newton’s equations of motion in the length gauge and in the KH frame. The former looks like

$$m\ddot{X}(t) = -\frac{\partial V(X)}{\partial X} - f(t). \quad (7)$$

It is well known [16] that the motion under the influence of a high-frequency field consists of a slow motion in an effective potential and rapid oscillations superimposed over it. Therefore representing the trajectory  $X(t)$  as

$$X(t) = x(t) + x_f(t), \quad m\ddot{x}_f(t) = -f(t) \quad (8)$$

( $x_f$  is the trajectory as it would be under the influence of the time-dependent field only) and deriving the equation for  $x(t)$ , one gets

$$m\ddot{x}(t) = -\frac{\partial V(x + x_f(t))}{\partial x}, \quad (9)$$

which is Newton’s equation in the KH frame. It is easy to see now that  $x_f$  corresponds to  $-h(t)$  of Eq. (5). The constant  $C_1$  is associated with the drift momentum of  $x_f$  and  $C_2$ —with its mean deviation from the smooth trajectory. Apparently, these constants can be chosen in such a way that both the drift momentum (representing dissociation or ionization) and the mean deviation are zero. This simple derivation shows that by using the KH frame one takes into account the ‘‘trivial’’ part of the trajectory, i.e.,  $x_f(t)$ .

The further general discussion is based on the assumption that the classical motion is regular. In this case one expects the quantum mechanical expectation values to behave like classical quantities, provided the action is much larger than  $\hbar$ . Moreover, regular motion implies that the trajectories are multiperiodic functions of time (no dissociation or ionization takes place). A very important specific example in which this assumption does not hold will be discussed in Sec. V.

Consider now the expression (1) for the probability of harmonic emission. The key quantity one has to calculate in order to obtain  $\sigma_k$  is the dipole moment acceleration as a function of time:

$$\ddot{\mu}(t) = \mu_1 [\ddot{x}(t) + \ddot{x}_f(t)] = \mu_1 \left( \ddot{x}(t) - \frac{1}{m} f(t) \right). \quad (10)$$

Newton’s equation (9) provides a direct root to the derivation of a perturbation expansion for acceleration. The shape of the field is important for such analysis, so let us first consider the cw case, e.g.,

$$f(t) = f_0 \mu_1 \sin(\omega_f t), \quad x_f = \frac{f_0 \mu_1}{m \omega_f^2} \sin(\omega_f t) \equiv \alpha_0 \sin(\omega_f t). \quad (11)$$

In such a case the time-dependent potential of Eq. (6) can be expanded in Fourier series:

$$\begin{aligned} H_{\text{KH}}(x, p, t) &= \frac{p^2}{2m} + V(x + \alpha_0 \sin(\omega_f t)) \\ &= \frac{p^2}{2m} + \sum_{n=-\infty}^{+\infty} V_n(x, \alpha_0) e^{in\omega_f t}, \end{aligned} \quad (12)$$

where

$$V_n(x, \alpha_0) = (\omega_f/2\pi) \int_0^{2\pi/\omega_f} V(x + \alpha_0 \sin(\omega_f t)) e^{-in\omega_f t} dt.$$

The corresponding Newton equation (9) is

$$m\ddot{x}(t) = -\frac{\partial V_0(x, \alpha_0)}{\partial x} + \sum_{n \neq 0} F_n(x, \alpha_0) e^{in\omega_f t}, \quad (13)$$

where  $F_n(x, \alpha_0) \equiv -\partial V_n(x, \alpha_0)/\partial x$ .

The time-averaged potential  $V_0(x, \alpha_0)$  generates a slow motion of large amplitude (the smooth part of a trajectory) while the time-dependent forces  $F_n(x, \alpha_0) e^{in\omega_f t}$  are responsible for fast, small amplitude oscillations. Consequently, the zero-order approximation for  $x(t)$  is the motion in the effective  $V_0$  potential:

$$m\ddot{x}^0(t) = -\left. \frac{\partial V_0(x, \alpha_0)}{\partial x} \right|_{x=x^0(t)}. \quad (14)$$

Moreover, the first-order approximation for the acceleration,  $\ddot{x}(t)$ , is

$$\ddot{x}^1(t) = \ddot{x}^0(t) + \frac{1}{m} \sum_{n \neq 0} F_n(x, \alpha_0) \Big|_{x=x^0(t)} e^{in\omega_f t}. \quad (15)$$

In order to obtain the harmonic emission probability, we have to consider the Fourier transform of  $\ddot{x}(t)$ , where the acceleration, as given by Eq. (15), is a multiperiodic function of time. The largest period is clearly that of the  $x^0(t)$  motion,  $2\pi/\omega_{\text{KH}}$ . Thus, computing the Fourier transform of acceleration, it is natural to take this period as the interval of integration. The result, up to a common factor, reads

$$\begin{aligned} \dot{\mu}(k\omega_f) &\propto \int_0^{2\pi/\omega_{\text{KH}}} \left( \sum_{n=-\infty}^{+\infty} F_n(x^0(t), \alpha_0) e^{i(n-k)\omega_f t} \right. \\ &\quad \left. + \frac{f_0 \mu_1}{2i} e^{i(1-k)\omega_f t} - \frac{f_0 \mu_1}{2i} e^{i(-1-k)\omega_f t} \right) dt, \\ k &= 1, 2, \dots \end{aligned} \quad (16)$$

However, the functions  $e^{i(n-k)\omega_f t}$ ,  $n \neq k$  oscillate much more rapidly than  $F_n(x^0(t), \alpha_0)$  do. Consequently, the main contribution to the integral (16) comes from the ‘‘diagonal’’ terms ( $n=k$ ). Confining ourselves to these terms only, we obtain the following expression for the probability to emit the  $k$ th harmonic:

$$\sigma_k \propto |\mu(k\omega_f)|^2 \propto \left| \int_0^{2\pi/\omega_{\text{KH}}} F_k(x^0(t), \alpha_0) dt + \delta_{k,1} \frac{f_0 \mu_1}{2i} \right|^2. \quad (17)$$

The functional form of  $x^0(t)$  can be quite complicated, but, whenever the harmonic approximation to the effective potential holds, one can use a simple sine function instead of the exact zero-order trajectory.

It is easy to see also that HG depends only on the action of the zero-order trajectory. Indeed,  $x^0(t)$  is a periodic function of angle, since it corresponds to the motion induced by a time-independent Hamilton function. Hence, the initial value of the angle variable plays no role within the diagonal approximation and the result (17) can be considered as an ensemble average over a microcanonical distribution of initial conditions. This distribution corresponds, however, to a specific value of the effective Hamilton function

$$H_{\text{KH}}^0(x^0, p^0) = \frac{(p^0)^2}{2m} + V_0(x^0, \alpha_0) \quad (18)$$

and not to the field-free Hamilton function. This is consistent with the situation in which the cw field is switched on not suddenly, but rather adiabatically.

Since  $x^0(t)$  represents small amplitude oscillations around the equilibrium position,  $x_e$ , of the effective  $V_0(x, \alpha_0)$  potential, it is clear from Eq. (17) that the dominant harmonics will be those for which  $|F_k(x_e, \alpha_0)|^2$  get large values. Moreover, in our model calculations (Secs. III, V) we find that the spectral distribution of harmonics is peaked around a single frequency. We call this ‘‘an approximative selection rule.’’

Finally, the implications of the pulse shape on the above treatment need to be clarified. First, let us consider the smooth part of the trajectory in the case of pulse excitation. (In the subsequent derivation the idea of adiabatic invariance is exploited in the same way as in the treatment of low-frequency limit by Leopold and Richards [11].) Let us assume that the pulse duration time  $\tau_p$  is much larger than the period of the unperturbed motion,  $\tau^0$ :

$$\tau_p \gg \tau^0 \gg \frac{2\pi}{\omega_f}. \quad (19)$$

In such a case the Hamilton function (18) can be regarded as adiabatically dependent on the field strength parameter  $\alpha_0$ :

$$H_{\text{KH}}^0(x, p, t) = \frac{(p^0)^2}{2m} + V_0(x^0, \alpha_0(t)), \quad (20)$$

where the time dependence of  $\alpha_0(t)$  is determined by the pulse shape. Suppose the time-independent generating function  $\tilde{F}_2(J, x, \alpha_0)$  defines the canonical transformation of Eq. (18) to the action-angle variables. Then the time-dependent function  $F_2(J, x, \alpha_0(t))$  leads to the following Hamilton function:

$$K(J, \theta, t) = H_a(J, \alpha_0(t)) + \frac{\partial F_2(J, x, \alpha_0(t))}{\partial t}, \quad (21)$$

where  $H_a(J, \alpha_0(t))$  is the adiabatic Hamilton function written in action-angle variables. The last term of Eq. (21) can be neglected in the limit  $\tau_p \gg \tau^0$ , Eq. (19). Therefore Hamilton's equations of motion in the adiabatic approximation are given by

$$\begin{aligned} \dot{J} &= -\frac{\partial H_a(J, \alpha_0(t))}{\partial \theta} = 0, \\ \dot{\theta} &\equiv \omega_a(J, t) = \frac{\partial H_a(J, \alpha_0(t))}{\partial J}, \end{aligned} \quad (22)$$

and

$$\begin{aligned} J(t) &= J_0, \\ \theta(t) &= \theta_0 + \int_0^t \omega_a(J_0, t') dt'. \end{aligned} \quad (23)$$

The zero-order trajectory can be obtained by an inverse transformation from the  $(J, \theta)$  to the  $(p, x)$  coordinates:

$$x^0(J, \theta) = x^0(J_0, \theta_0, t). \quad (24)$$

Unlike in the cw case,  $x^0$  of Eq. (24) is no longer a periodic function of either time or angle.

Having estimated the smooth part of a trajectory, one would like next to write down Newton's equation of motion from which the first-order approximation to the acceleration can be obtained. This equation can be derived easily from the Hamilton function (6). Note that  $-h(t)$  of Eq. (6) represents now a motion of a "free" particle affected by the radiation pulse only. Consequently, the KH potential  $V(x-h(t))$  is, strictly speaking, no longer a periodic function of time. However, the pulse can be imagined to repeat itself infinitely many times after it has effectively decayed. In this case, the potential of Eq. (6) is a time-periodic function with the period equal to the pulse duration  $\tau_p$ . Suppose, for simplicity, that  $\tau_p$  is equal to an integer number of optical cycles of the high-frequency field:

$$\tau_p = N_p \frac{2\pi}{\omega_f}, \quad N_p \gg 1. \quad (25)$$

Then Eq. (6) can be rewritten as

$$H_{\text{KH}}(x, p, t) = \frac{p^2}{2m} + \sum_{n=-\infty}^{+\infty} V_n(x) e^{in(\omega_f/N_p)t}, \quad (26)$$

where  $V_n(x) = (1/\tau_p) \int_0^{\tau_p} e^{-in(\omega_f/N_p)t} V(x-h(t)) dt$ .

The corresponding Newton equation reads

$$m\ddot{x}(t) = \sum_{n=-\infty}^{\infty} F_n(x) e^{in(\omega_f/N_p)t}, \quad (27)$$

where  $F_n(x) \equiv -\partial V_n(x)/\partial x$ .

The first-order approximation to the acceleration can be obtained by substitution of the zero-order trajectory (24) into Eq. (27):

$$\ddot{x}^1(t) = \frac{1}{m} \sum_{n=-\infty}^{+\infty} F_n(x) \Big|_{x=x^0(J_0, \theta_0, t)} e^{in(\omega_f/N_p)t}. \quad (28)$$

The dominant  $F_n$ 's are those corresponding to the  $\omega_f$  harmonics, since  $\omega_f$  itself is the dominant frequency in the Fourier decomposition of  $-h(t)$ . Therefore only these dominant

terms will be taken into account in the evaluation of the Fourier transform of acceleration. Naturally, the time interval over which the integration should be performed is the pulse duration time. Using the diagonal approximation, just as in the cw case, and taking into account the acceleration of the  $x_f(t)$  trajectory one arrives at the following expression for the dipole moment acceleration Fourier components:

$$\begin{aligned} |\mu(k\omega_f)|^2 &\propto \left| \int_0^{\tau_p} F_{k \times N_p}(x^0(J_0, \theta_0, t)) dt \right. \\ &\quad \left. - \int_0^{\tau_p} f(t) e^{-ik\omega_f t} dt \right|^2. \end{aligned} \quad (29)$$

The last expression, unlike its cw analog (17), still depends on the  $\theta_0$  initial condition. In order to get the  $k$ th harmonic emission probability as the square of the absolute value of the *mean* acceleration Fourier component [see Eq. (1)] one has to average  $F_{k \times N_p}(x^0(J_0, \theta_0, t))$  over  $\theta_0$ :

$$\begin{aligned} \sigma_k &\propto \left| \frac{1}{2\pi} \int_0^{\tau_p} \int_0^{2\pi} F_{k \times N_p}(x^0(J_0, \theta_0, t)) d\theta_0 dt \right. \\ &\quad \left. - \int_0^{\tau_p} f(t) e^{-ik\omega_f t} dt \right|^2. \end{aligned} \quad (30)$$

Note that the force Fourier components in the right-hand side of Eq. (30) are averaged over the microcanonical ensemble of initial conditions, which corresponds to a specific value of the field-free Hamilton function.

An analytical evaluation of the last expression is a hard task. Nevertheless, it provides some insight into the shape of the HG spectrum as a function of the pulse shape. Suppose the pulse decays immediately after it has been switched on (e.g., a Gaussian or a  $\sin^2$  profile). In such a case, the zero-order trajectory is not localized, since the equilibrium position  $x_e$  of the effective potential changes adiabatically. So, unlike the cw case, there are no approximative selection rules for harmonic emission. The Fourier components of force, being averaged over a large interval, are expected to produce monotonously decreasing harmonic amplitudes. However, the rate of decrease is expected to be lower than in the cw case. Indeed, the HG spectrum of the Morse oscillator excited by a pulse of high-frequency radiation shows this kind of behavior [17]. If, on the other hand, the rise of the pulse is followed by a constant value of the envelope during a lot of optical cycles, the selectivity is regained (see the discussion in Sec. IV).

### III. CLASSICAL HG SPECTRUM OF MORSE OSCILLATOR DRIVEN BY A HIGH-FREQUENCY cw LASER FIELD

Consider Morse oscillator linearly coupled to the periodic external field. This system is described by the following Hamiltonian:

$$\hat{H}_L = \frac{\hat{p}^2}{2m} + D_0(e^{-2\alpha(X-X_0)} - 2e^{-\alpha(X-X_0)}) + \mu_1 X f_0 \sin(\omega_f t). \quad (31)$$

In the subsequent calculations we choose to use the parameters corresponding to the nuclear motion of the HF molecule being in the ground electronic state: the reduced mass,  $m = 1744.7$  a.u.,  $D_0 = 0.225$  a.u.,  $\alpha = 1.1741$  a.u.,  $X_0 = 1.7329$  a.u.,  $\mu_1 = 0.31$  a.u. The model Hamiltonian (31) and its classical counterpart,  $H_L(P, X, t)$  [see Eq. (4)], have been used to study the multiphoton transitions [20], classical chaotic dynamics, and photodissociation [21] and, more recently, harmonic generation [17]. The Hamiltonian (31) is not applicable to the description of the interaction of the molecule with the high-intensity fields considered in this work. Nevertheless, it can be used as a simple mathematical model in order to test the predictions of the general theory of Sec. II.

Transforming the length gauge Hamilton function to the KH frame one gets

$$H_{\text{KH}}(p, x, t) = \frac{p^2}{2m} + D_0(e^{-2\alpha[x + \alpha_0 \sin(\omega_f t) - X_0]} - 2e^{-\alpha[x + \alpha_0 \sin(\omega_f t) - X_0]}). \quad (32)$$

The Fourier expansion of the KH potential (32) produces

$$H_{\text{KH}}(p, x, t) = \frac{p^2}{2m} + \sum_{n=-\infty}^{+\infty} i^n D_0 [I_n(2\alpha\alpha_0) e^{-2\alpha(x-X_0)} - 2I_n(\alpha\alpha_0) e^{-\alpha(x-X_0)}] e^{in\omega_f t}, \quad (33)$$

where  $I_n$  is the  $n$ th-order modified Bessel function of the first kind [22]. Note that the averaged KH potential  $V_0(x, \alpha_0)$  is nothing but the generalized Morse potential [23].

In order to apply the general result (17) to this model, one should first verify the applicability of the classical theory. The latter is presumably correct if the classical motion is regular and the corresponding value of action is much higher than  $\hbar$ . Poincaré surface of section corresponding to Hamilton's function (32) for the field parameters  $\alpha_0 = 3.1$  a.u.,  $\omega_f = 0.0858$  a.u.  $\approx 8\omega_{\text{KH}}$  is represented in Fig. 1. The regular or chaotic character of motion is determined by the overlap of classical resonances (see Goggin and Milonni [21] for Chirikov's resonance overlap criterion [24] applied to the Morse oscillator in the time-periodic field). As the field frequency grows, the resonances of higher order (chains of regular islands in Fig. 1) affect the motion and the measure of regular trajectories inside the  $V_0(x, \alpha_0)$  separatrix becomes larger. Moreover,  $V_0(x, \alpha_0)$  can be approximated by a harmonic potential in the vicinity of its equilibrium position  $x_e$ ,

$$x_e = X_0 + \frac{1}{\alpha} \ln\left(\frac{I_0(2\alpha\alpha_0)}{I_0(\alpha\alpha_0)}\right) \quad (34)$$

and the couplings  $V_n(x, \alpha_0)$ ,  $n \neq 0$  can be linearized around  $x_e$ . Therefore in this region of the phase space the problem

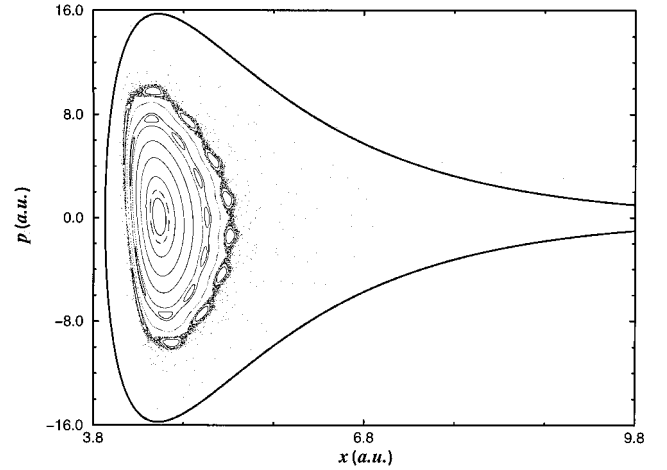


FIG. 1. Poincaré surface of section corresponding to Hamilton's function (32). The field parameters are  $\alpha_0 = 3.1$  a.u.,  $\omega_f = 0.0858$  a.u.  $\approx 8\omega_{\text{KH}}$ . The points in the chaotic region result from a single dissociative trajectory. The separatrix of the time-averaged KH Hamilton function [Eq. (18)] is shown by a line.

reduces to that of a harmonic oscillator linearly coupled to the time-dependent field. The quantum mechanical solution of such a problem is known to reproduce the classical one exactly [25].

The zero-order trajectory in the harmonic approximation is simply

$$x^0(t) = x_m^0 \sin(\omega_{\text{KH}}^0 t + \theta_0) + x_e, \quad (35)$$

where  $\omega_{\text{KH}}^0$  is the harmonic frequency of the averaged KH potential and  $x_m^0$  is the amplitude of small oscillations around  $x_e$ .

It has been shown in Sec. II that one can estimate which harmonics will be dominant in the spectrum considering the magnitudes of the force Fourier components,  $F_n$ , Eq. (13), in the vicinity of the equilibrium position of the effective potential. For example, comparing the absolute values of these components for  $\alpha_0 = 3.1$  a.u. (see Fig. 2), one can predict that the third harmonic is going to be the dominant one in the spectrum. The direct evaluation of Eq. (17) produces the following expression for  $\sigma_k$ :

$$\sigma_k \propto \left| -2\alpha D_0 i^n \frac{I_0(\alpha\alpha_0)}{I_0(2\alpha\alpha_0)} \left( \frac{I_0(\alpha\alpha_0) I_0(2\alpha x_m^0)}{I_0(2\alpha\alpha_0)} I_k(2\alpha\alpha_0) - I_0(\alpha x_m^0) I_k(\alpha\alpha_0) \right) + \delta_{k,1} \frac{f_0 \mu_1}{2i} \right|^2. \quad (36)$$

The oscillation amplitude  $x_m^0$  is small on the scale of the  $V_0(x, \alpha_0)$  potential range, so  $\alpha x_m^0$  is a small parameter. It implies that the HG spectrum depends weakly on the initial condition:

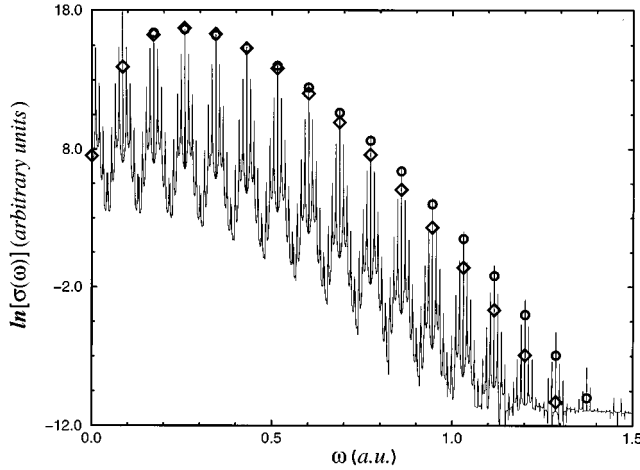


FIG. 2. The classical HG spectrum resulting from the numerical integration of Hamilton's equations of motion corresponding to Eq. (32) (continuous curve), the analytical result (37) (circles), and the absolute values of the Fourier components of the time-dependent force in the KH frame,  $\ln|F_n(x_e, \alpha_0)|$ , Eq. (13), evaluated at the equilibrium position of the effective KH potential (diamonds). The field parameters are  $\alpha_0 = 3.1$  a.u.,  $\omega_f = 0.0858$  a.u.,  $x_m^0 \approx 0.236$  a.u., which corresponds to the harmonic approximation to the ground state of the time-averaged KH potential. The field frequency peak is out of scale. The spectra are normalized in such a way that the third harmonic intensity is the same in all of them. Note the third harmonic dominance in all the spectra.

$$\sigma_{k>1} \approx \left| \frac{I_0(\alpha\alpha_0)[1 + (\alpha x_m^0)^2]}{I_0(2\alpha\alpha_0)} I_k(2\alpha\alpha_0) - \left(1 + \frac{1}{4}(\alpha x_m^0)^2\right) I_k(\alpha\alpha_0) \right|^2 \quad (37)$$

and the only important parameter of the problem is  $\alpha\alpha_0$ , namely, the amplitude of the field-induced motion,  $\alpha_0$  on the scale of a potential range,  $1/\alpha$ . The parameter  $\alpha\alpha_0$  defines actually which peak will be the most prominent one in the HG spectrum. It is easy to see that by changing it one can achieve the enhancement of a specific harmonic. However, it takes totally unrealistic field intensities to get very high harmonics enhanced.

The relative harmonic intensities as predicted by Eq. (37) have been found to be in excellent agreement with the classical numerical results (see Fig. 2). The latter were obtained by the numerical solution of Hamilton's equations of motion in the KH frame. The single trajectory HG spectrum was found to be practically independent of the initial phase  $\theta_0$ , while  $x_m^0$  was chosen to match approximately the energy of the  $V_0(x, \alpha_0)$  ground state. The following calculation parameters were chosen:  $\alpha_0 = 3.1$  a.u.,  $\omega = 0.0858$  a.u. Such a value of  $\alpha_0$  corresponds to the enhancement of the third harmonic peak.

#### IV. HG SPECTRUM OF MORSE OSCILLATOR DRIVEN BY A HIGH-FREQUENCY cw LASER FIELD—QM NUMERICAL RESULTS

Quantum mechanically, the problem can be approached in two different ways. One can either perform a wave packet

calculation, or deal with the quasienergy (QE) states. The QE states are quasistationary solutions of Schrödinger's equation:

$$\Psi_\beta(x, t) = e^{-i(\epsilon_\beta t/\hbar)} \Phi_\beta(x, t), \quad (38)$$

$$\Phi_\beta(x, t) = \Phi_\beta\left(x, t + \frac{2\pi}{\omega_f}\right).$$

It has been shown previously that under certain conditions the HG spectrum can be associated with a single QE state (see [26], and references therein). The expectation value of dipole moment, entering Eq. (1), should be taken with the length gauge wave function. However, the length gauge is not suitable for the treatment of high-frequency external fields from the numerical point of view. So using Ehrenfest's theorem and the operators of the unitary transformations from length to reduced momentum gauge and from reduced momentum gauge to KH frame (see, for example, [27]) one easily obtains

$$\begin{aligned} \frac{\partial^2}{\partial t^2} \langle \Psi^L | \mu_1 \hat{x} | \Psi^L \rangle &= \frac{\mu_1}{m} \frac{\partial}{\partial t} \langle \Psi^L | \hat{p} | \Psi^L \rangle \\ &= \frac{\mu_1}{m} \frac{\partial}{\partial t} \langle \Psi^P | \hat{p} | \Psi^P \rangle - \mu_1 \alpha_0 \omega_f^2 \sin(\omega_f t) \\ &= \frac{\mu_1}{m} \frac{\partial}{\partial t} \langle \Psi^{\text{KH}} | \hat{p} | \Psi^{\text{KH}} \rangle \\ &\quad - \mu_1 \alpha_0 \omega_f^2 \sin(\omega_f t), \end{aligned} \quad (39)$$

where  $\Psi^L$ ,  $\Psi^P$ , and  $\Psi^{\text{KH}}$  are the wave functions in the length gauge, reduced momentum gauge, and KH frame, respectively. Fourier decomposition of the time-periodic part of a KH frame QE function reads

$$\Psi_\beta^{\text{KH}}(x, t) = e^{-i(\epsilon_\beta t/\hbar)} \sum_{n=-\infty}^{+\infty} \varphi_n(x) e^{in\omega_f t}. \quad (40)$$

Substitution of Eq. (40) into Eq. (39) and subsequent Fourier transformation result in the following expression for the probability to emit the  $k$ th harmonic by the system being in the  $\beta$ th QE state:

$$\sigma_k \propto \left| k \sum_{n=-\infty}^{+\infty} \langle \varphi_n(x) | \hat{p} | \varphi_{n+k}(x) \rangle - \frac{\alpha_0 m \omega_f}{2} \delta_{k,1} \right|^2. \quad (41)$$

Clearly, the HG spectrum of a quasiperiodic  $\psi_\beta^{\text{KH}}$  state consists only of the harmonics of the radiation frequency.

In the problem with dissociation or ionization, such as a Morse oscillator in the external cw field, only the resonance QE states, which live sufficiently long in the interaction region, should be considered:

$$\epsilon_\beta = \text{Re}(\epsilon_\beta) - i \frac{\Gamma}{2}, \quad (42)$$

where  $\Gamma/\hbar$  is the dissociation rate. These states are being routinely obtained by the complex scaling (CS) technique [28]. According to the CS procedure, the coordinate is rotated into the complex plane by an angle  $\theta$ :

$$x \mapsto (x - x_{\text{CS}})e^{i\theta} + x_e. \quad (43)$$

The most convenient choice of the point  $x_{\text{CS}}$ , about which the rotation to the complex plane occurs, is the center of the interaction region,  $x_{\text{CS}} = x_e$ .

The complex-scaled QE functions have been calculated in KH frame for the field parameters  $\alpha_0 = 3.1$  a.u.,  $\omega_f = 0.0858$  a.u. by diagonalization of the one optical cycle complex scaled propagator matrix:

$$\hat{U}(T,0)\Psi_\beta^{\text{KH}}(x,0) = e^{-i(\epsilon_\beta T/\hbar)}\Psi_\beta^{\text{KH}}(x,0). \quad (44)$$

The propagator was calculated by the recently developed  $(t,t')$  algorithm [29]. Eleven Fourier basis functions were used to represent the time dependence of the Floquet Hamiltonian:

$$\hat{\mathcal{H}}_{\text{KH}}(x,t) \equiv \frac{\hbar}{i} \frac{\partial}{\partial t} + \hat{H}_{\text{KH}}(x,t). \quad (45)$$

One hundred eigenfunctions of the zero-order KH Hamiltonian,

$$\hat{H}_{\text{KH}}^0(x) = \frac{\hat{p}^2}{2m} + \hat{V}_0(x, \alpha_0) \quad (46)$$

have been used to represent the coordinate dependence of  $\hat{\mathcal{H}}_{\text{KH}}$ . Twenty-one Floquet channels and 1000 time steps were used in  $(t,t')$  calculation of  $\hat{U}(T,0)$ . The number of resonance QE states was found to be equal to the number of bound states of the  $\hat{H}_{\text{KH}}^0$  Hamiltonian. The lifetime of the narrowest resonance QE state, closely corresponding to the  $\hat{H}_{\text{KH}}^0$  ground state, is  $\Gamma \approx 1.22435 \times 10^{-6}$  a.u. The complex QE's are stationary over a wide range of rotation angles. The calculations described below have been performed at  $\theta = 0.1$  rad.

In order to calculate the HG spectrum, we should generalize the expression (41) for the complex rotated QE states. This implies two changes: first, the generalized inner product,  $\langle\langle \Psi^\theta | \Psi^\theta \rangle\rangle$  [30], should be used instead of the regular one,  $\langle \Psi | \Psi \rangle$ , second, the momentum operator  $\hat{p} \equiv (\hbar/i) \partial / \partial x$  should be substituted by its complex rotated counterpart:

$$\hat{p}^\theta = e^{-i\theta} \frac{\hbar}{i} \frac{\partial}{\partial x}. \quad (47)$$

The generalized inner product ( $c$  product of Ref. [30]) differs from the regular one by the lack of complex conjugation of those parts of bra and ket which have become complex only due to the CS. Being applied to the result (41) the above prescription gives

$$\sigma_k \propto \left| k \sum_{n=-\infty}^{+\infty} \langle\langle \varphi_n(x) | \hat{p}^\theta | \varphi_{n+k}(x) \rangle\rangle - \frac{\alpha_0 m \omega_f}{2} \delta_{k,1} \right|^2, \quad (48)$$

where  $\langle\langle \varphi_n(x) |$  and  $| \varphi_{n+k}(x) \rangle\rangle$  are Fourier components of the left and the right eigenvectors of the complex-scaled Floquet Hamiltonian correspondingly. The HG spectrum of the

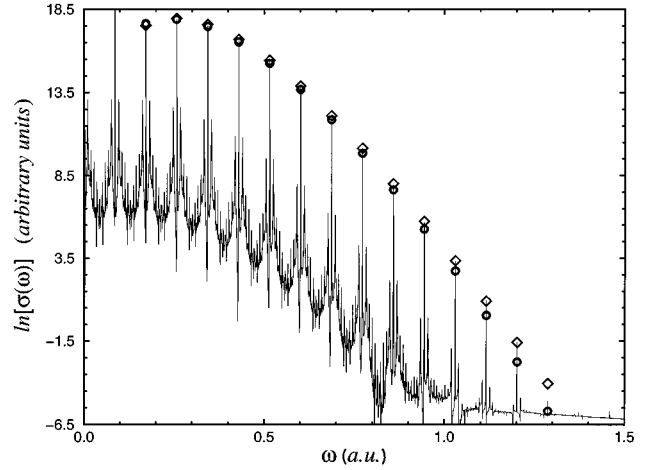


FIG. 3. The QM HG spectrum of the narrowest resonance Floquet state (diamonds), QM HG spectrum of a wave packet placed in the harmonic region (see Fig. 1) (continuous line), and the classical prediction of Eq. (37) (circles). The relative intensities of the second and higher harmonics are shown. The field parameters are those of Fig. 1. The spectra are normalized in such a way that the third harmonic intensity is the same in all of them.

longest-living QE state is presented in Fig. 3. As one can see, the single QE state QM HG spectrum is in excellent agreement with the classical analytical result. The HG spectra of all the resonance QE states exhibit strong similarity. All of them show the enhancement of the third harmonic peak.

In order to explore the properties of the QM HG spectrum as a function of phase space variables, wave packet calculations have been performed. Schrödinger's equation in the KH frame,

$$i\hbar \frac{\partial}{\partial t} \Psi(x,t) = \hat{H}_{\text{KH}}(x,t) \Psi(x,t), \quad (49)$$

$$\hat{H}_{\text{KH}}(x,t) = \frac{\hat{p}^2}{2m} + D_0(e^{-2\alpha[x + \alpha_0 \sin(\omega_f t) - X_0]} - 2e^{-\alpha[x + \alpha_0 \sin(\omega_f t) - X_0]}),$$

was solved numerically for the initial condition of the form

$$\Psi(x,t=0) = \left( \frac{m\omega_{\text{KH}}}{\pi\hbar} \right)^{1/2} e^{-(m\omega_{\text{KH}}/2\hbar)(x-x_{\text{in}})^2} e^{ip_{\text{in}}x/\hbar}, \quad (50)$$

where  $x_{\text{in}}$  and  $p_{\text{in}}$  are the initial mean values of the coordinate and the momentum of the minimum uncertainty wave packet. The expectation value of the dipole moment acceleration as a function of time can be expressed, again using Ehrenfest's theorem and the operator of transformation from the length gauge to KH frame as

$$\begin{aligned}
\frac{d^2}{dt^2} \langle \Psi^L | \mu_1 x | \Psi^L \rangle &= \frac{2\alpha D_0 \mu_1}{m} \langle \Psi^L | e^{-2\alpha(x-X_0)} \\
&\quad - e^{-\alpha(x-X_0)} | \Psi^L \rangle - \alpha_0 \omega_f^2 \mu_1 \sin(\omega_f t) \\
&= \frac{2\alpha D_0 \mu_1}{m} \langle \Psi^{KH} | e^{-2\alpha[x+\alpha_0 \sin(\omega_f t)-X_0]} \\
&\quad - e^{-\alpha[x+\alpha_0 \sin(\omega_f t)-X_0]} | \Psi^{KH} \rangle \\
&\quad - \alpha_0 \omega_f^2 \mu_1 \sin(\omega_f t). \tag{51}
\end{aligned}$$

A Gaussian wave packet centered at the equilibrium position of the averaged KH potential ( $x_{\text{in}}=x_e$ ,  $p_{\text{in}}=0$ ) has been propagated in time by the split operator algorithm [31]. 512 equally spaced grid points have been used to represent the Hamiltonian  $\hat{H}_{\text{KH}}$ . The box length has been chosen to be 13 a.u. and the absorbing boundary conditions were introduced at  $x_{\text{opt}}=12$  a.u. by means of the following optical potential:

$$V_{\text{opt}}(x) = -i \frac{\epsilon_{\text{opt}}}{1 + e^{(x_{\text{opt}}-x)/\eta_{\text{opt}}}}, \tag{52}$$

where  $\epsilon_{\text{opt}}=10$  a.u.,  $\eta_{\text{opt}}=0.25$ . The resulting quantum mechanical HG spectrum (1) corresponding to the field parameters of Fig. 2 is represented in Fig. 3. The result is in excellent agreement with the classical prediction (37) and with the QE calculation.

The similar calculations have been performed with the initial wave packet located at one of the elliptical fixed points in the chain of the ten islands of regularity (see Fig. 1). Note that the measure of chaotic initial conditions close to a hyperbolic fixed point is substantially less than  $\hbar$ , so one cannot locate the least uncertainty wave packet in the chaotic region only. The HG spectrum obtained by propagation of the wave packet centered around the point  $x_{\text{in}}=4.46$  a.u.,  $p_{\text{in}}=9.8$  a.u. has been found to be almost identical to that of the wave packet placed in the harmonic region, in accordance with the results of the QE calculations.

Finally, one has to consider the effect that the switching of the cw laser field may have on the HG spectrum. Apparently, if driving the initial state (which is localized in the field-free potential well,  $x \approx 1.7$  a.u.) to the interaction region (namely, the vicinity of the effective KH potential,  $x \approx 4.5$  a.u.) takes a switching time comparable to the lifetime of the resonance QE state, the cw theory can hardly reflect the real physical situation. In order to check the implications of switching on the HG, a series of numerical calculations has been performed. The eigenstates of the field-free Hamiltonian have been propagated in time under the influence of the following time-dependent field:

$$f(t) = \begin{cases} \alpha_0 \omega_f^2 m \sin^2\left(\frac{\omega_f t}{4N}\right) \sin(\omega_f t), & t \leq \frac{2\pi N}{\omega_f} \\ \alpha_0 \omega_f^2 m \sin(\omega_f t), & t > \frac{2\pi N}{\omega_f} \end{cases} \tag{53}$$

where  $N$  is the number of optical cycles it takes to switch the field on. The propagation has been performed in the KH frame by the split operator method [31]. The wave function has been represented on a grid of 512 equally spaced points

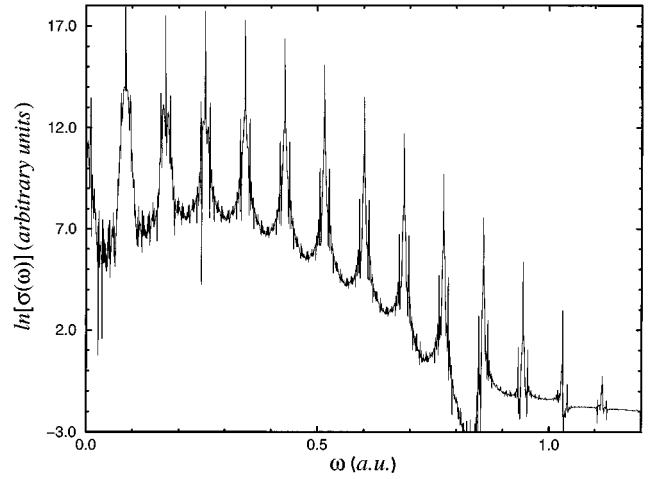


FIG. 4. The QM HG spectrum of a ground state of the field-free Hamiltonian. Contrary to the calculations presented in Fig. 3, the field is turned on here according to Eq. (53),  $\alpha_0=3.1$  a.u.,  $N=20$ . The field frequency peak is out of scale.

covering the interval from 0 to 15 a.u. The fast Fourier transform (FFT) procedure has been used to allow the operation of the kinetic term on the wave function. The optical potential (52) has been used to prevent the reflection from the edge of the interval.

The resulting HG spectrum corresponding to the ground state of the field-free Hamiltonian is represented in Fig. 4. The switching time was taken to be 20 optical cycles. The spectrum agrees very well with the results of the pure cw treatment (Figs. 2, 3). If, however, one chooses a higher excited eigenstate as an initial condition, the longer switching time is required to avoid a significant dissociation during the rising of the field. Consequently, the switching part of the dipole moment time dependence begins to play a role. Indeed, starting with the tenth eigenstate of the Morse oscillator and allowing a 40 optical cycle switching time, we have obtained a HG spectrum with less pronounced third harmonic dominance effect.

## V. HG SPECTRUM OF “SOFT-CORE” COULOMB POTENTIAL DRIVEN BY A HIGH-FREQUENCY cw LASER FIELD—QM NUMERICAL RESULTS

The example discussed in the previous two sections is, perhaps, the simplest case on which the predictions of the general theory (Sec. II) can be tested. The Morse potential can be considered as a generic one for the molecular vibrational motion. However, at the field strengths causing the enhancement of high harmonics, the multiphoton transitions to the excited electronic states and the disintegration of a molecule would occur. Moreover, the harmonic generation by electronic motion will always be more effective just due to the electron-proton mass ratio and different magnitudes of transition matrix elements between the electronic and nuclear eigenstates. Therefore the general theory needs to be applied to the Coulomb potential. This section will be devoted to the study of the HG spectrum of a one-dimensional “soft-core” Coulomb potential model in the high-frequency, high-intensity regime. Before reporting our QM numerical results,

let us discuss several questions concerning the applicability of the classical theory to the Coulombic case.

If the zero field potential is a symmetric function (of the coordinate defining the polarization direction of the external field), then its averaged KH counterpart at high enough field strengths is a double well. Consequently, the zero-order classical motion does not take into account the tunneling phenomenon. The symmetry considerations, however, apply to the force Fourier components as well, therefore their values in each one of the wells can differ only by a phase and the tunneling is not expected to affect the spectrum drastically.

What can be more important, however, is the measure of the regular classical motion in the vicinity of the equilibrium points of the zero-order KH potential on the scale of  $\hbar$ . Our numerical experience with the Morse potential model shows though that the QM resonance states are not necessarily associated with the surviving tori. As a matter of fact, the resonances living in the chaotic region of the phase space can possess longer lifetimes than those associated with the regular islands [32]. Jensen and Sundaram [33] have pointed out that the suppression of ionization in high-frequency fields is related to the existence of scarred QE states. The authors studied a one-dimensional ‘‘soft-core’’ Coulomb potential model [34]:

$$V(X) = \frac{1}{\sqrt{1+X^2}} \quad (54)$$

under the influence of the high-frequency, high-intensity cw field. The two stable fixed points in the phase space of the model are associated with the two minima of the double well effective potential, while the unstable fixed point is situated on the saddle between the wells ( $x=0$ ,  $p=p_0$ ). It has been suggested by Jensen and Sundaram that the quantum dynamics is governed by the QE states localized in the vicinity of the unstable periodic orbit, ‘‘scars’’ (see also Leopold and Richards [35] on the role of scars in the suppression of ionization). In order to check the implications of this phenomenon on the HG of the system, we have compared the QM HG spectrum with the prediction of the approximative classical selection rule. Schrödinger’s equation in the KH frame has been solved numerically by the split operator method [31]. The ground state of the field-free potential (54) was chosen as the initial condition and ten optical cycle long switching time was allowed [see Eq. (53)]. The calculation has been performed at the field parameters  $\omega_f=1$  a.u.,  $\alpha_0=10$  a.u. The Hamiltonian was represented on a grid of 4096 equally spaced points in the interval from  $x=-100$  to 100 a.u. The symmetric optical potential of the type (53) ( $x_{\text{opt}}=\pm 98$  a.u.,  $\epsilon_{\text{opt}}=5$  a.u.,  $\eta_{\text{opt}}=0.5$  a.u.) was introduced to prevent the reflections from both edges of the grid. The resulting HG spectrum presented in Fig. 5 has been found to be in remarkable agreement with the simple classical prediction:

$$\sigma_{k>1} \approx |F_x(x=0, \alpha_0)|^2. \quad (55)$$

The last equation is valid under the assumption that the deviation of the unstable periodic orbit,  $x_{\text{per}}(t)$ , from the unstable equilibrium position of the effective KH potential is small on the scale on which the force Fourier components

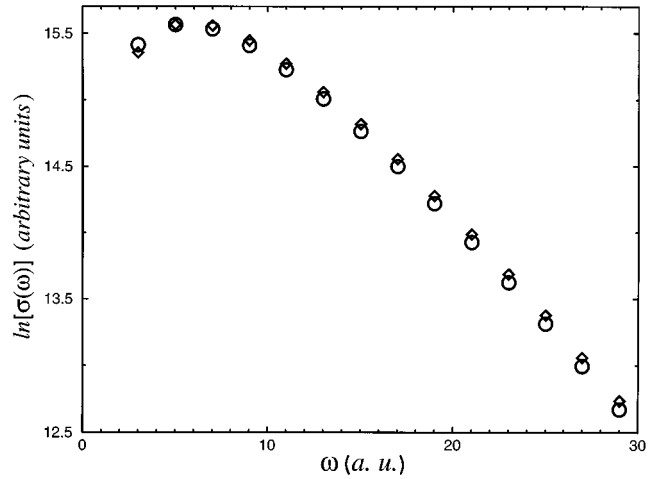


FIG. 5. The QM HG spectrum of a ground state of the soft-core Coulomb potential (circles) and the classical prediction (55) (diamonds). The field is turned on according to Eq. (53),  $\alpha_0=10$  a.u.,  $N=10$ . The field frequency peak is out of scale. The spectrum is normalized in such a way that the fifth harmonic intensity is equal to  $\sigma_5$  of Eq. (55).

change:  $F_k(x_{\text{per}}(t), \alpha_0) \approx F_k(x=0, \alpha_0)$ . Note that due to the symmetry of the problem only odd harmonics appear in the spectrum. As in the case of Morse oscillator, a single (fifth) harmonic is the dominant one in the spectrum. The behavior of the Fourier components of the force at the unstable fixed point suggests that the higher the field intensity, the higher harmonic signal is enhanced. It is remarkable that the HG spectrum bears purely classical character even in the case when the measure of the regular trajectories is much smaller than  $\hbar$  and tunneling takes place.

## VI. CONCLUSIONS

It has been shown that classical mechanics provides an adequate description of the HG phenomenon in the high-frequency regime. Moreover, it provides an approximative selection rule for the relative strengths of different harmonics. This gives a possibility to adjust the system and the laser field parameters in such a way that a specific single harmonic dominates the spectrum.

The classical theory is applicable if the wave function is localized in space. In such a case the Newton equation used in the derivation of the harmonic emission probabilities is a good approximation to the quantum equations of motion by virtue of Ehrenfest’s theorem:

$$\begin{aligned} \langle \ddot{x} \rangle(t) &= \frac{1}{m} \left\langle - \frac{\partial V(x+x_f(t))}{\partial x} \right\rangle \approx - \frac{1}{m} \frac{\partial V(x+x_f(t))}{\partial x} \Big|_{x=x_e} \\ &\approx \ddot{x}_{\text{per}}(t). \end{aligned} \quad (56)$$

The wave function implicit in Eq. (56) can be localized around either stable or unstable periodic trajectory,  $x_{\text{per}}(t)$ . These trajectories, in their turn, deviate only slightly from the corresponding (stable or unstable) equilibrium position of the averaged KH Hamiltonian. Indeed, such deviation is due to the high-frequency forces  $F_k(x)e^{ik\omega_f t}$ ,  $\omega_f \gg \omega_{\text{KH}}$  [see Eq. (13)] and thus it is proportional to  $1/(\omega_f k)^2$ . For frequencies

high enough the deviation of the periodic trajectory from  $x_e$  becomes small on the characteristic scale on which the  $F_k(x)$ 's change. If the wave function localization width is small on this scale as well, the characteristic pattern of the force Fourier components is translated directly into the HG spectrum [see Eq. (55)]. It turns out that long-living quantum states of various nonlinear oscillators, such as Morse, "soft-core" Coulomb, and hydrogen atom [33,36] perturbed by the high-frequency external fields exhibit this kind of localization. This fact lets us think that the classical approach to HG in this regime and the result (55) are quite general.

Besides the specificity of harmonic emission, the characteristic shape of the HG spectrum can be used to detect the high-frequency stabilized states, as proposed by Kulander, Schafer, and Krause [36]. It still has to be shown, however, that the approximative selection rules for harmonic emission derived here have an analog in the three-dimensional case.

The validity of the adiabatic approach to the radiation pulses, as described in Sec. II, is not guaranteed in the case of two- and more-dimensional effective KH potentials. In such problems even the zero-order classical motion can be nonintegrable.

The questions raised above outline the directions of future studies. Research along these directions is currently in progress.

#### ACKNOWLEDGMENTS

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