# Cutoff in molecular harmonic-generation spectra resulting from classical chaotic dynamics

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The plateau in the harmonic-generation (HG) spectrum of a bound quantum system is first associated with classical chaotic dynamics. The molecular cutoff in the HG spectra was obtained at $\Omega = m\omega$, where $\omega$ is the frequency of the time-periodic field and $m$ is the area of the bound chaotic region in the phase space divided by $2\pi\hbar$.

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The spectra emitted from strongly irradiated low-pressure rare gas were found to consist of high harmonics of the fundamental frequency $\omega$ of the irradiative field [1]. A characteristic harmonic-generation (HG) spectrum exhibits an exponential decay of the intensity of the first few peaks relative to the intensity of the fundamental frequency $\omega$. This decay is followed by a plateau, which ends in an exponential decay. The cutoff in the atomic HG spectra was associated by Krause, Schafer, and Kulander with the minimal energy that is required for ionization [2]. A cutoff of the plateau in the HG spectra can be obtained, however, for bound systems as well. The high-order HG spectra were studied by Sundaram and Milonni [3] and, more recently, by Kaplan and Shkolnikov [4], Roso and Plaja [5] for two-level systems, and by Zuo, Chelkowski, and Bandrauk [6] for a three-level system. The quantum vs classical chaotic dynamics of periodically driven bound systems was extensively studied over the last decade. (See, for example, the book by Haake [7] and the papers on quantum vs classical dynamics of periodically driven systems [8].) To the best of our knowledge, the plateau in the HG spectra and the cutoff phenomenon have never been associated so far with the chaotic dynamics of a classical system. Our study shows that the "longest" plateau in the HG spectra is obtained when the infinite-level driven system (i.e., a driven rotor in our case) exhibits a chaotic behavior.

The Hamiltonian of a rigid rotor in high fields in dimensionless units is given by

$$H(\phi,t)=\frac{p_\phi^2}{2}-\cos(\phi)f(t)\cos(\omega t)\ , \tag{1}$$

where $f(t)$ describes the amplitude of the cw laser as a function of time. Like the kicked-rotor model studied by Blümel, Fishman, and Smilansky [9], the continuously driven rotor presented in Eq. (1) describes the dynamics of a heteronuclear diatom, such as CsI, in high-intensity laser fields. The relevant experiment, however, is much easier to perform with the cw laser. When the cw laser is turned on sufficiently slowly, the solution of the time-dependent Schrödinger equation, $\chi(t)$, for the initially given free-rotor state is approximately the quasienergy solution (i.e., Floquet solution) of the time-periodic Hamiltonian,

$$H(\phi,t)=\frac{p_\phi^2}{2}-\epsilon_0\cos(\phi)\cos(\omega t)\ , \tag{2}$$

where $\epsilon_0$ is the maximum field amplitude (taken here to be unity). That is, within the framework of adiabatic approximation,

$$\chi(t)\cong\psi_\alpha(\phi,t),\quad t>0\ , \tag{3}$$

where for initially excited free-rotor states, $m\neq 0$:

$$\chi(t=0)=\pi^{-1/2}\sin(m\phi)\ , \tag{4}$$

$$\chi(t=0)=\pi^{-1/2}\cos(m\phi)\ . \tag{5}$$

For the free-rotor ground state, $m=0$, $\chi(t=0)=(2\pi)^{-1/2}$. A quasienergy state is given by

$$\psi_\alpha(\phi,t)=e^{-i\epsilon_\alpha t/\hbar}\Phi_\alpha(\phi,t)\ , \tag{6}$$

where

$$\Phi_\alpha(\phi,t)=\Phi_\alpha(\phi,t+T)=\sum_{k=-\infty}^{+\infty}\varphi_{k,\alpha}(\phi)e^{i\omega kt}\ . \tag{7}$$

$\Phi_\alpha(\phi,t)$ and $\varepsilon_\alpha$ are, respectively, an eigenfunction and an eigenvalue of the Floquet Hamiltonian,

$$\mathcal{H}_F(\phi,t)=-i\hbar\frac{\partial}{\partial t}+H(\phi,t)\ . \tag{8}$$

As shown by Moiseyev, Korsch, and Mirbach [10] the continuously driven rotor [Eq. (2)] exhibits a bounded chaotic motion, which is presented in Fig. 1 for $\omega=1$. For $\hbar=0.02$, it is expected to find 4 symmetry-adapted regular quasienergy states (the prediction is made by dividing the area of the inner-regular island by $2\pi\hbar$) and 92 symmetry-adapted chaotic states. The HG spectra is associated with the Fourier transform of the time-dependent dipole moment amplitude

$$\sigma_{\mathrm{HG}}(\Omega)=\left|\int_{-\infty}^{\infty}e^{-i\Omega t}D(t)\right|^2\ , \tag{9}$$

where

$$D(t)=\langle\chi(t)|\hat{\mu}|\chi(t)\rangle \tag{10}$$

and in our case

$$\hat{\mu}=\cos\phi\ , \tag{11}$$

 

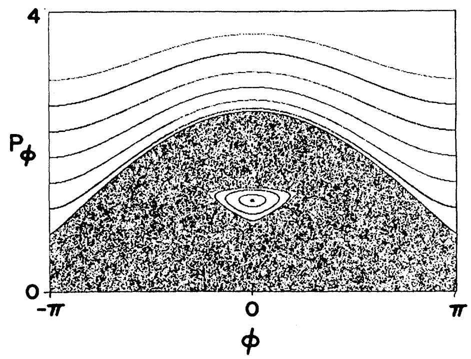


FIG. 1. Poincaré section of classical phase space at $t=nT$, ($n=0,1,2,\ldots,$) taken from Ref. [10]. Only the upper region, $p_\phi \geq 0$, is shown because of symmetry. All points in the chaotic region result from a single classical trajectory.

when

$$\Omega = n\omega \ .$$

When the field is turned on sufficiently slowly as discussed above, then

$$\sigma_{\mathrm{HG}}(\Omega=n\omega)\cong|D_\alpha^{\mathrm{QE}}(n\omega)|^2 \ , \tag{12}$$

where

$$D_\alpha^{\mathrm{QE}}(n\omega)=\int_{-\infty}^{\infty} e^{-i\Omega t}\langle \Phi_\alpha(t)|\hat{\mu}|\Phi_\alpha(t)\rangle dt$$
$$=\sum_{k=-\infty}^{+\infty} \langle \varphi_{k-n,\alpha}|\hat{\mu}|\varphi_{k,\alpha}\rangle \tag{13}$$

and $\alpha$ indicates the quasienergy solution, which is associated following adiabatic theorem with the given initial free-rotor state $\chi(t=0)$. In the absence of quantum interferences (the mechanism that "kills" the quantum-mechanical (QM) interferences will be discussed later) the HG spectra are given by

$$\tilde{\sigma}_{\mathrm{HG}}(\Omega=n\omega)\cong\tilde{\sigma}_{\mathrm{HG}}^{\mathrm{QE},\alpha} \ , \tag{14}$$

$$\tilde{\sigma}_{\mathrm{HG}}^{\mathrm{QE},\alpha}(\Omega=n\omega)=\sum_{k=-\infty}^{\infty} |\langle \varphi_{k-n,\alpha}|\hat{\mu}|\varphi_{k,\alpha}\rangle|^2 \ . \tag{15}$$

We first shall show that the QE-HG spectra defined as

$$\sigma_{\mathrm{HG}}^{\mathrm{QW},\alpha}(\Omega=n\omega)=|D_\alpha^{\mathrm{QE}}(n\omega)|^2 \tag{16}$$

[and, therefore, $\sigma_{\mathrm{HG}}$ [Eq. (12)] in the more generalized case] is exponentially localized at $n=1$ (i.e., the fundamental laser frequency) and is not followed by a plateau.

It implies that the quantum-mechanical molecular HG spectra, $\sigma_{\mathrm{HG}}(n\omega)$, consists almost uniquely of the fundamental frequency of the irradiative laser $\omega$ regardless of the nature (i.e., regular or chaotic) of the quasienergy states of the driven rotor. These results are presented in Figs. 2–4. In Fig. 2(c), the HG spectra for one out of the four QE states that are located in the inner-regular island in the classical chaotic "sea" [see Fig. 2(b)] is presented. As shown in Fig. 2(a) the inner-regular states are exponentially localized in the free-rotor basis set [$|\langle\chi(0)|\Phi_\alpha(0)\rangle|^2$ is the projection of the $\alpha$th quasienergy state on the $m$th free-rotor state.] Figure 3(c) shows the HG spectra obtained for 1 out of the 92 QE states, which are located in the chaotic bounded region in the classical phase space. [See the Husimi distribution plot presented in Fig. 3(b).] This QE state, like all the other 92 chaotic states, is dominated by 92 free-rotor basis functions [see Fig. 3(a)]. In Fig. 4(c), a typical HG spectra of one of the regular states, which are embedded in the quasiperiodic region in phase space, which is outside of the chaotic bounded region [see Fig. 4(b)], is shown. As one can see

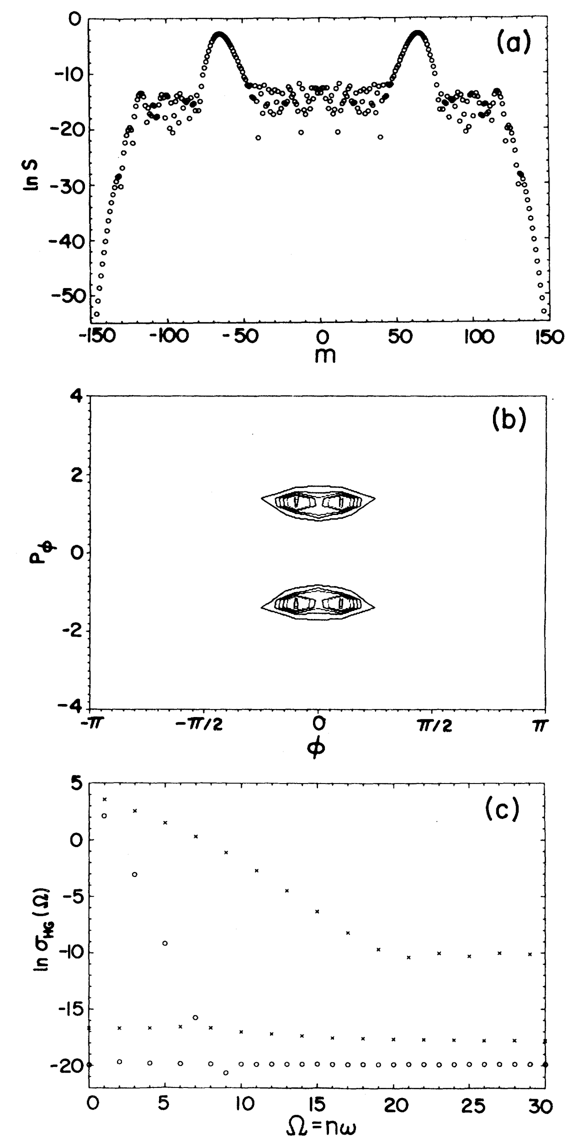


FIG. 2. The inner-regular quasienergy state, its Husimi distribution, and the corresponding harmonic-generation spectra. (a) The projection of the quasienergy solution, $\Phi_\alpha(t=0)$, as defined in Eq. (7) on the free-rotor states $\chi(0)=\exp(im\phi)/\sqrt{2\pi}$, which are linear combinations of the states defined in Eqs. (4) and (5). $S=|\langle\Phi_\alpha(0)|\chi(0)\rangle|^2$. (b) The Husimi distribution as obtained in Ref. (10). (c) The quasienergy harmonic-generation spectra as defined in Eq. (16); $\sigma_{\mathrm{HG}}^{\mathrm{QE},\alpha}$, denoted by $o$, and the HG spectra obtained in the absence of the quantum-mechanical interferences, $\tilde{\sigma}_{\mathrm{HG}}^{\mathrm{QE},\alpha}$ as defined in Eq. (15) denoted by $x$.

from Fig. 4(a), those states are dominated by several numbers of free-rotor states. As the outer-regular QE state is located closer to the boundaries of the chaotic sea, more free-rotor states dominate in the basis-set expansion of that QE state.

The HG molecular spectra is dramatically changed in the absence of the quantum interference. In such a case, the molecular HG spectra as calculated from Eq. (15) exhibit the following phenomena.

(1) In all cases the even harmonics were suppressed, and only the odd multiplies of the original frequency were produced. This phenomenon was attributed to the symmetry property of the studied model Hamiltonian [11].

(2) For a regular QE state, which is located in the inner-regular island in the chaotic sea and is exponentially localized in the free-rotor basis set, *no* plateau in the HG spectra is obtained; this is exactly as before when the quantum interferences were taken into consideration.

(3) For the QE state, which is located in the outer-regular region in the classical phase space, however, a plateau that ends in an exponential decay is obtained [see Fig. 3(c)]. The number of the harmonics in the plateau are equal to the number of basis functions, which dominate in the free-rotor expansion of $\Phi(\phi,t)$.

(4) The highest harmonics in the HG spectra are obtained for chaotic QE states [see a typical example in Fig. 4(c)]. All the 92 chaotic QE states produce a plateau in the HG spectra with a cutoff at $\Omega=92\omega$.

The fact that the HG spectra of the quasienergy states, which are localized in the inner-regular islands, show an exponential localization at the fundamental frequency $\omega=1$ can be explained by simple classical arguments.

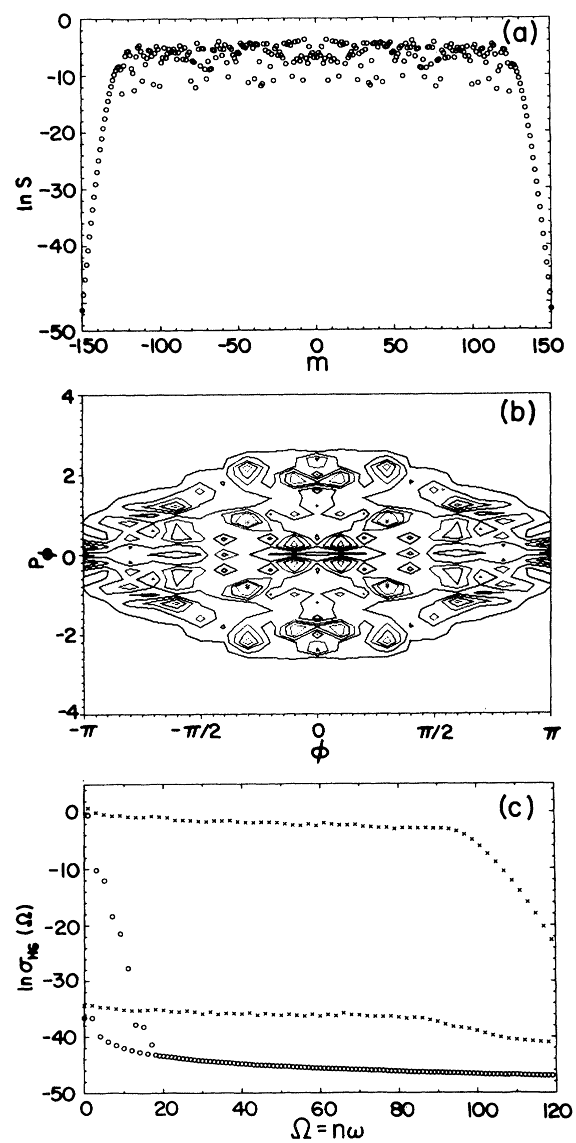


FIG. 3. The same as Fig. 2 for a chaotic quasienergy state.

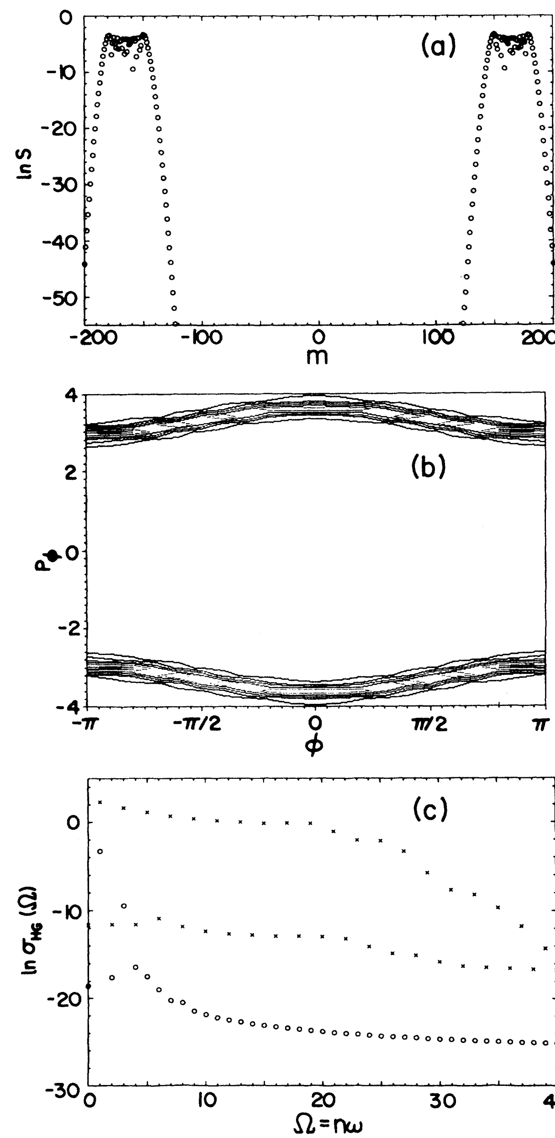


FIG. 4. The same as Fig. 2 for an outer-regular quasienergy state.

The inner-regular islands in the stroboscopic map shown in Fig. 1 are associated with the classical resonances $p_\phi = \pm\omega$. Therefore, the HG spectra should be dominated by the fundamental frequency as obtained in the quantum-mechanical calculations [Fig. 2(c)]. The cutoff in the HG spectra of the chaotic quasienergy states is found to be given by

$$\Omega_{\max} = n_{\max}\omega \quad (n_{\max}=92) , \tag{17}$$

where

$$\eta_{\max} = \frac{S}{2\pi\hbar} \quad (\hbar=0.02) , \tag{18}$$

where $S$ is the area of the bounded chaotic region and the inner-regular regions in phase space. A simple classical explanation for this is as follows: Since a classical particle exhibits a random walk in the bounded chaotic region, it may happen that during one optical cycle it changes its momentum from $P_{\max}$ to $O$; therefore, its energy changes from $P_{\max}^2/2$ to $O$. The largest frequency of the radiated photon is $\Omega_{\max}$, where

$$\hbar\Omega_{\max} = \frac{P_{\max}^2}{2} . \tag{19}$$

$P_{\max}$ is the averaged value of the boundary $p_\phi(\phi)$ between the chaotic region and the regular region, which is given by

$$P_{\max} = \frac{S}{2\pi} . \tag{20}$$

According to semiclassical quantization,

$$S = 2\pi\hbar N , \tag{21}$$

where $N$ is the number of the chaotic (92) and inner-regular (4) states (for $\hbar=0.02$, $N=96$). Consequently,

$$\hbar\Omega_{\max} = \frac{1}{2}\left[\frac{2\pi\hbar N}{2\pi}\right]^2 = \frac{\hbar^2 N^2}{2} \tag{22}$$

and

$$\Omega_{\max} = \frac{\hbar N^2}{2} = \frac{0.02}{2} \times 92^2 \cong 92 \tag{23}$$

exactly as obtained in the numerical calculation [see Fig. 3(c)].

It is a point of interest that the cutoff in the HG spectra is obtained in quantum-mechanical calculation if, and only if, the quantum-interference effects are neglected. The exact QM HG spectra show an exponential localization at the fundamental frequency $\omega=1$ [see Fig. 3(c)] even in the semiclassical limit of $\hbar \to 0$! Two different mechanisms that may destroy the quantum interferences can be considered. The first one is the destruction of the quantum interferences due to a random noise, which is provided by arbitrarily varying the maximum field amplitude within an interval of $\pm 5\%$ of its maximal value [12]. The second mechanism is a more "natural" one. The vibrational modes of the heteronuclear diatom (neglected in the present study) are coupled to the rotational mode. The vibrational-rotational coupling may introduce a natural noise into the calculations, which will destroy the quantum interferences and a finite plateau in the HG spectra will be produced.

The fact that the plateau in the HG spectra due to the classical chaotic dynamics requires high-intensity fields raises the question whether the system breaks up as the field intensity is increased beyond a certain value. It was shown recently by Yao and Chu that when a system is subjected to a monochromatic laser field, an increase in the laser intensity does not necessarily lead to an increase of ionization or dissociation rate [13]. (See also on the "breathing" above-threshold-ionization and above-threshold-dissociation spectra in Ref. [14]). In the extreme case for a given frequency and for a strong field, an almost zero resonance width is obtained. On the basis of this phenomenon, for example, it was suggested to separate the isotopes of $H_2^+$ [15].

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[1] X. F. Li, A. L'Huillier, M. Ferray, L. A. Lompre, and G. Mainfray, Phys. Rev. A **39**, 5751 (1989).
[2] J. L. Krause, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).
[3] Bala Sundaram and P. W. Milonni, Phys. Rev. A **41**, 6571 (1990).
[4] A. E. Kaplan and P. L. Shkolnikov (unpublished).
[5] L. Roso and L. Plaja, J. Opt. Soc. B **9**, 2210 (1992).
[6] T. Zuo, S. Chelkowski, and A. D. Bandrauk, Phys. Rev. A (to be published).
[7] F. Haake, *Quantum Signatures of Chaos* (Springer, New York, 1991).
[8] B. V. Chirikov, I. Guarneri, and D. L. Shepelyansky, Phys. Rev. Lett. **56**, 2437 (1988); R. Blümel and A. Smilansky, *ibid.* **69**, 217 (1992); S. Fishman, D. R. Grempel, and R. E. Prange, *ibid.* **49**, 509 (1982); E. Doron and S. Fishman, *ibid.* **60**, 867 (1988); R. E. Prange and S. Fishman, *ibid.* **63**, 704 (1989); G. Casati, L. Molinari, and F. Izrailev, *ibid.* **64**, 1851 (1990); G. Jona-Lasinio, C. Presilla, and F. Capasso, *ibid.* **68**, 2269 (1992); L. Bonci, R. Roncaglia, B. J. West, and P. Grigolini, *ibid.* **67**, 2593 (1991); G. Casati, B. V. Chirikov, and D. L. Shepelyansky, *ibid.* **53**, 2525 (1984); E. J. Galvez, B. E. Sauer, L. Moorman, P. M. Koch, and D. Richards, *ibid.* **61**, 2011 (1988); J. E. Bayfield, G. Casati, I. Guarneri, and D. W. Sokol, *ibid.* **63**, 364 (1989).
[9] R. Blümel, S. Fishman, and U. Smilansky, J. Chem. Phys. **84**, 2604 (1986).
[10] N. Moiseyev, M. J. Korsch, and B. Mirbach, Z. Phys. D **29**, 125 (1994).

[11] N. Ben-Tal, N. Moiseyev, and A. Beswick, J. Phys. B **26**, 3017 (1993).
[12] N. Ben-Tal, N. Moiseyev, S. Fishman, F. Bench, and H. J. Korsch, Phys. Rev. E **47**, 1646 (1993).
[13] G. Yao and S.-I. Chu, Phys. Rev. A **45**, 6735 (1992).
[14] Uri Peskin and Nimrod Moiseyev, J. Chem. Phys. **99**, 4590 (1993).
[15] O. Atabek, M. Chrysos, and R. Lefebvre, Phys. Rev. A **49**, R8 (1994).