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Superdressed two-level atom: Very high harmonic generation and multiresonances

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We show that a simple nonperturbative two-level model of an atom driven by a very strong periodic field results in a rich picture of very high harmonic generation and related phenomena. It reproduces the experimentally observed plateau, yields simple analytic formulas for the plateau cutoff frequency, critical driving intensity, and saturation, and predicts intensity-induced multiresonances.

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One of the most fascinating phenomena discovered recently in nonlinear interaction of light with atoms and ions is very-high-order (up to 135) odd harmonic generation (HHG) by intense ($\sim 10^{13}$ W/cm^2 and higher) optical laser radiation in rare gases and some ions [1]. The spectra of generated harmonics drastically deviate from the perturbation-theory predictions [2]. In particular, intensity of harmonics, falling monotonically with their orders only up to a certain point, levels off, forming a so-called "plateau," and falls monotonically again beyond it. Generally, harmonic generation depends on phase-matching conditions and nonlinear response of individual atoms. It has recently become clear, however [2–4], that the major features of HHG, in particular the plateau, result mainly from general properties of atomic nonlinear response (moreover, the plateau also appears to be a generic feature of many nonlinear models, including classical nonlinear systems [2(c)]). The most direct and apparently successful way so far to approach the problem theoretically has been numerical simulation of the Schrödinger equations [2,5–7] (including an empiric rule [6]) for many-electron atoms using Hartree-Slater approximation. This approach requires, however, a tremendous number of calculations and involves many processes, making it difficult to gain simple insights. An interesting simplified model [8] based on a three-dimensional δ potential with a single (ground) level [9] produces results in the form of integrals. The idea [9] of retaining a single-energy scale (ionization energy) brings one close to an even simpler system: a two-level model atom. A two-level model of HHG [10], however, due to various complications introduced into it in order to take into account some experimental factors, did not attempt to generate simple results, whereas an analytic solution [11] holds for a virtually degenerate two-level model only (see below) which is inapplicable to rare-gas atoms used in experiment [1–4,7,9]. A very interesting and detailed earlier work [12] holds only for quasiadiabatic approximation. Besides, no relaxation was considered in [10–12].

In this article we present a two-level model (nondegenerate and with relaxation) that not only reproduces experimentally observed plateau, but also allows one to evaluate its characteristics in explicit analytic form and predicts dramatic effects [13]. Our very simple analytic formulas for the cutoff frequency of the plateau, critical intensity for the plateau formation, and saturation at each individual harmonics relate all these quantities to the energy of the first excited level, E_1 (rather than to the ionization limit, E_{ion}, to which they are usually attributed), and are consistent with the available experimental data, at least for some gasses. We also predict well-ordered multiresonances in harmonic intensity and in population difference vs driving intensity when the energy of interaction becomes comparable with E_1. Due to its simplicity, the model offers new insights into the HHG and points to new situations in which this phenomenon can be observed.

A two-level atom with relaxation is described by a density matrix whose diagonal elements are related to the population per atom at both the ground (ρ_{11}) and excited (ρ_{22}) levels ($\rho_{11}+\rho_{22}=1$), and whose nondiagonal elements, $\rho_{12}=\rho_{21}^*$, are related to the induced polarization $\mathbf{P}=\mathbf{d}_{12}(\rho_{12}+\rho_{21})$, where $\mathbf{d}_{12}$ is a dipole moment. The population difference, $\eta\equiv\rho_{11}-\rho_{22}$, is maximal, $\eta=\eta_0$, at the thermal equilibrium determined by the Boltzmann distribution; $\eta_0=\tanh(-kT_K/2\hbar\omega_0)$, where T_K is the temperature, k is the Boltzmann constant and ω_0 is a resonant frequency of the two-level atom (in most of the cases of interest, $\eta_0\approx 1$). In terms of normalized variables $x\equiv\eta/\eta_0$, $y\equiv\rho_{21}/\eta_0$, the dynamics of an atom driven by a periodic field $\mathbf{E}\sin(\omega t)$ is governed by the equations [14]:

$$\dot{x}+\tau^{-1}(x-1)=2i\Omega_R(y-y^*)\sin(\omega t)\ , \tag{1}$$

$$\dot{y}+(T^{-1}+i\omega_0)y=i\Omega_R x\sin(\omega t)\ , \tag{2}$$

where the Rabi frequency $\Omega_R\equiv\mathbf{d}_{12}\cdot\mathbf{E}/\hbar$ is a measure of the dipole interaction energy, and T and τ are longitudinal and transverse relaxation times, respectively. Equations (1) and (2) can be reduced to a differential equation for a single variable (e.g., x), which allows one to directly obtain analytic solutions in many cases [15]. They can also be solved simultaneously in the form of Fourier extensions,

$$x=x_0+\sum_{n=1}^{\infty}(x_n e^{2in\omega t}+\text{c.c.})\ ,$$
$$y=\sum_{n=-\infty}^{\infty}y_n e^{i(2n-1)\omega t}\ . \tag{3}$$

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By solving first Eq. (2) for y,

$$y_n = \frac{-(i/2)\mu R(x_{n-1} - x_n)}{2n - 1 + R(1 - i\Gamma)} , \tag{4}$$

and using it in Eq. (1), one arrives at a recurrent formula for the ratio $w_n \equiv x_n / x_{n-1}$:

$$f_n(1 - w_n^{-1}) + f_{n+1}(1 - w_{n+1}) = 2\tilde{n} / R^2\mu^2 , \quad n \ge 1 , \tag{5}$$

where $f_n \equiv \tilde{N} / (\tilde{N}^2 - R^2)$, $\tilde{N} \equiv N - iR\Gamma$, $N = 2n - 1$ is a harmonic number, $\tilde{n} \equiv n - i\theta R\Gamma$, $R \equiv \omega_0/\omega$, $\Gamma \equiv (T\omega_0)^{-1}$, $\theta \equiv T/2\tau$ ($\theta = 1$ for purely radiation relaxation), and $\mu \equiv \Omega_R / \omega_0$ is a dimensionless driving amplitude; thus

$$x_n = x_0 w_1 w_2 \cdots w_n . \tag{6}$$

The Fourier coefficients p_n of normalized polarization

$$p \equiv P / d_{12}\eta_0 = y + y^* = \sum_{n=1}^{\infty} [p_n e^{i(2n-1)\omega t} + \text{c.c.}] \tag{7}$$

are obtained then as

$$p_n = i\mu R^2 x_{n-1}(1 - w_n) f_n / \tilde{N} . \tag{8}$$

A recurrent formula for the ratio $v_n \equiv p_{n+1}/p_n$ is

$$\frac{g_n/v_n - 1}{\tilde{n}} + \frac{v_{n+1} - g_{n+1}}{\tilde{n} + 1} + \frac{2}{\mu^2 R^2 f_{n+1}} = 0 , \quad n \ge 1 , \tag{9}$$

where $g_n \equiv \tilde{N} / (\tilde{N} + 2)$, so that [16,17]

$$p_{n+1} = p_1 v_1 v_2 \cdots v_n . \tag{10}$$

The equation for x_0 is

$$x_0^{-1} - 1 = \mu^2 R \, \text{Im}[f_1(1 - w_1)] / \theta\Gamma ; \tag{11}$$

it coincides with a familiar formula for resonant saturation when $R \approx 1$, $\mu \ll 1$, and $w_1 = 0$. Physical solutions w_n and v_n of Eqs. (5), (9), and (11) are those vanishing as $n \to \infty$. Figure 1 shows the ($|p_n|^2$) spectra calculated by setting $w_{500} = 0$ in Eq. (5) for $\theta = 1$, $\Gamma = 10^{-3}$, and $R = 7.24$ [assuming $\omega_0 = E_1/\hbar$ and ω as in a Xe atom driven by a neodymium-doped yttrium-aluminum-garnet (Nd:YAG) laser]. Formation of plateau and "irregular" behavior inside it as μ increases are evident. In the small-perturbation approximation [$\mu^2 \ll |(N/R)^2 - 1|$], Eqs. (5) and (9) yield

$$w_n \approx f_n / (f_n + f_{n+1} - 2\tilde{n} / \mu^2 R^2) ,$$
$$v_n \approx g_n / [1 + \tilde{n} g_{n+1} / (\tilde{n} + 1) - 2\tilde{n} / \mu^2 R^2 f_{n+1}] , \tag{12}$$

i.e., $w_n \approx v_n \approx -(\mu R / 2n)^2$ as $n \to \infty$. In the limits of either $R \ll 1$ or $R \gg 1$ (and $\Gamma = 0$) approximate analytic solutions can be obtained for arbitrary μ. We now rewrite Eqs. (1) and (2) as

$$\dot{x} = -2\mu \dot{p} \sin(\omega t) , \quad \ddot{p} / \omega_0^2 + p = 2\mu x \sin(\omega t) . \tag{13}$$

If $R \ll 1$ (degeneration into a virtually one-level system), one can neglect the term p in these equations, thus making them isomorphic to a rotation wave approximation for exact resonance [14,15] ($\mu \ll 1$, $R = 1$). The solution

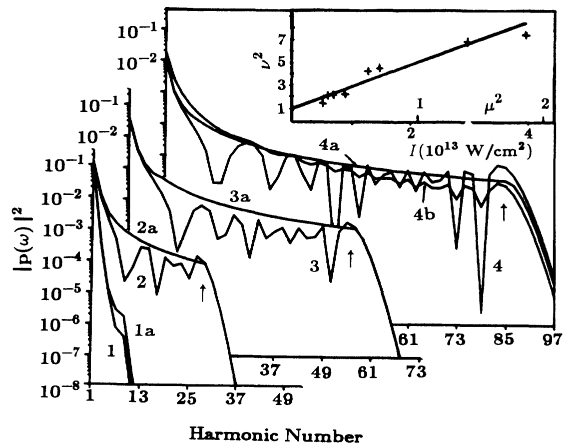

FIG. 1. HHG polarization spectra, $|p_n|^2$ ($N = 2n - 1$ is harmonic order), for various dimensionless driving intensities μ^2, and $R = 7.24$ and $\Gamma = 10^{-3}$ (except 4b, where $\Gamma = 0.1$). Curves: 1, $\mu = 0.4$; 2, $\mu = 2$; 3, $\mu = 4$; 4, $\mu = 6$; 1a–4a, respective envelope approximations. Arrows indicate plateau cutoff points. Inset: cutoff ratio square v^2 vs μ^2 for Xe. Crosses: experiment [2(a),4]; solid line: linear fit.

[11,15] is then

$$x \approx \cos[2\mu R \cos(\omega t)] ,$$
$$p \approx -R \int \sin[2\mu R \cos(\omega t)] d(\omega t) . \tag{14}$$

i.e.,

$$x_n \approx (-1)^n J_{2n}(2\mu R) ,$$
$$p_n \approx R(-1)^n J_N(2\mu R) / N , \tag{15}$$

where $N = 2n - 1$, and $J_m(z)$ is an ordinary Bessel function of mth order. (Note that $p \to 0$ as $R \to 0$.) Equations (5) and (9) show that spectrumwise, this approximation holds for arbitrary R to some factor $C = C(R, \mu)$ and only for $N^2 \gg R^2$ [18]. If now $R \gg 1$ (a typical situation for the HHG experiments [2]), neglecting the term $\ddot{p}/\omega_0^2$ in the above equations, one obtains [12]

$$x \propto \tilde{x} \approx [1 + 4\mu^2 \sin^2(\omega t)]^{-1/2} . \tag{16}$$

Fourier components $\tilde{x}_n$ and $\tilde{p}_n$ are expressed then in terms of elliptic integrals. Spectrumwise, this approximation holds only for $N^2 \ll R^2$.

To explore HHG *spectrum* in the *entire* range of N's and μ's, one has to deal directly with Eqs. (5), (9), and (11). They show that for "end" areas, i.e., for $N \to \infty$ and $N \lesssim R$, w_n and v_n are *slowly* varying functions of n. For $N < R$ this is consistent with $\tilde{x}_n$ and $\tilde{p}_n$ being smooth functions of n, and for $n \to \infty$, with similar behavior of $J_n(z)$ for sufficiently large n (see below). By assuming, e.g., in Eq. (5) that $w_{n+1} \approx w_n$ and $f_{n+1} \approx f_n$, Eq. (5) is reduced to a quadratic equation for w_n,

$$(w_n - 1)^2 + 2n w_n / (f_n \mu^2 R^2) \approx 0 .$$

Its solution,

$$\bar{w}_n^{(\text{out})} = A \pm \sqrt{A^2 - 1}\ , \quad A \equiv 1 - n/(f_n \mu^2 R^2)\ ,$$

is *real* (note that w_n and v_n must be real if Γ is neglected) only if either

$$N \le R \quad \text{or} \quad N \ge N_{\text{cut}} \approx R\sqrt{1+4\mu^2}\ . \tag{17}$$

(The upper sign in the formula for $\bar{w}_n^{(\text{out})}$ corresponds to $N \ge N_{\text{cut}}$, and the lower sign to $N \le R$.) Between these two points there lies an "inside" area, $R < N < N_{\text{cut}}$, with distinct irregular spectra, Fig. 1, consistent with other models [2,6]. These chaoslike (with respect to the order or number of harmonic) fluctuations are originated by Eqs. (5) and (9) in the "inside" area, where these equations are to the extent similar to the well-known iterative equations giving rise to a strange attractor (although the former are more complicated, especially in that they have "number-dependent" parameters). In the limit $\mu R \gg 1$ and $N^2 \gg R^2$, this behavior can be explained by the properties of the zeros of $J_N(2\mu R)$ approximating the solution in this limit. It is well known [19] that none of zeros of $J_N(\zeta)$ (except for $\zeta = 0$) coincides with zeros of $J_{N+1}(\zeta)$. The lowest positive zeros, z_N, of $J_N(\zeta)$ are given by the Tricomi formula [19] $z_N \approx N + 2N^{1/3} + N^{-1/3} + O(N^{-1})$. When $N < \zeta$, $J_N(\zeta)$ is an almost oscillatory function of ζ with a slowly accumulating dependence of its phase on integer N; this dependence is reflected in the Tricomi formula, in the terms nonlinear in N. This makes the chaoslike behavior of harmonic amplitudes as function of their order, similar in nature to the ball dynamics in "round billiards" in the case when the single angular "hop" of the ball is incommensurate with 2π. The smooth envelopes of these spectra, $\bar{w}_n^{(\text{in})}$ and $\bar{v}_n^{(\text{in})}$ [they also approximate monotonic spectra for $N \le R$ for large μ and away from resonance; Eq. (23) below] are obtained by assuming $\mu \to \infty$ in Eqs. (5) and (9), which results in $\bar{w}_n^{(\text{in})} \approx 1$, and $\bar{v}_n^{(\text{in})} \approx g_n \approx N/(N+2)$, i.e.,

$$\bar{x}_n^{(\text{in})}(N) \approx \text{const} \quad \text{and} \quad \bar{p}_n^{(\text{in})}(N) \approx \text{const}/N\ . \tag{18}$$

The analytic envelopes for p_n based on $\bar{v}_n^{(\text{out})}$ and $\bar{v}_n^{(\text{in})}$ are shown at Fig. 1, curves 1a–4a; they clearly show that the inside area is a plateau. A good fit between these envelopes and exact solutions is evident; the best predicted is the plateau cutoff, N_{cut}. The simple "cutoff" formula can be written in a universal form:

$$\nu \equiv N_{\text{cut}}/R \approx (1+4\mu^2)^{1/2} \quad \text{or} \quad \omega_{\text{cut}} \approx (\omega_0^2 + 4\Omega_R^2)^{1/2}\ , \tag{19}$$

which allows for comparison of different atoms driven by different lasers. The plateau width scales almost quadratically with μ (or with the driving *intensity*) when $\mu \ll 1$ (similar to [6]), and linearly with μ [10], $N_{\text{cut}} \approx 2\Omega_R/\omega$, when $\mu \gg 1$. From the mathematical point of view, the latter result is understood by noting that in such a limit, $J_N(2\mu R)$ in p_n in Eq. (15) becomes a monotonic function of N at $N \ge N_{\text{cut}} \approx 2\mu R$, and rapidly vanishes as $N \to \infty$. The physical interpretation of this result is that for sufficiently large excitation, $\mu \gg 1$ or $\Omega_R \gg \omega_0$, the maximum energy of the electron in the two-level atom is $2\hbar\Omega_R$, which coincides with the maximum (cutoff) energy of generated photons. It is worth noting also that the cutoff point is insensitive to the relaxation; see curve 4b for $\Gamma = 0.1$ in Fig. 1.

The analytic formula, Eq. (19), that suggests the scaling law linear with the driving *amplitude* (or with Ω_R) contradicts the numerical result [6], the latter suggesting that ω_{cut} increases linearly with the driving intensity (or Ω_R^2 in our notations), or, more specifically, that the cutoff energy increases as $\sim 3U_p$, where U_p is the ponderomotive potential proportional to the driving intensity. While the experimental confirmation of the "$3U_p$ rule" is still at a dynamic stage, with the coefficient at U_p varying between 2 and 3+, we show here (see below) that, at least for Xe atoms, our analytic formula, Eq. (19), is consistent with the experimental data at the intensities up to $\sim 10^{14}$ W/cm^2. The common point for both of these results is that they both relate the plateau cutoff with the maximum energy of the driven electron. Indeed, as shown by Corkum in his "almost-classical" 3D model [20], the coefficient 3.17 at U_p corresponds to the *maximum* energy gained by an electron from the optical field if the atomic potential is neglected, while $2\hbar\Omega_R$ is the maximum energy of the electron bound within a two-level system.

If $R \gg 1$, the critical μ required for the plateau to appear at all is evaluated as

$$\mu_{\text{cr}}^2 \approx 1/R \quad \text{or} \quad (\Omega_R)_{\text{cr}}^2 \approx \omega_0 \omega\ . \tag{20}$$

Since $\Omega_R = \mathbf{d}_{12} \cdot \mathbf{E}$, $d_{12} \propto \omega_0^{-1/2}$, and $E^2 \propto n_{\text{ph}}\omega$, where n_{ph} is the flux of the driving photons, Eq. (20) suggests that for $\omega_0 \gg \omega$, $(n_{\text{ph}})_{\text{cr}} \propto \omega_0^2$ and does not depend on ω; this indicates the potential for HHG at lower frequencies. Figure 1 shows that, consistently with experiment [3], the harmonic intensities as function of their order make deep minima, which could be substantially below the envelope, Eq. (18), if $\mu \gg \mu_{\text{cr}}$. The position N_1 of this first minimum for relatively small μ is $N_1 \sim R + \mu/\mu_{\text{cr}}$. For $\mu \gg 1$, N_1 fluctuates considerably with μ in a periodic fashion [see also Eq. (23)] [below; however, $(N_1)_{\max} = O[(\mu R)^{2/3}]$. As μ increases, the chaoslike spectrum inside the plateau becomes more "regular," with the number and amplitude of the dips increasing; the spacing s between them is maximal near both ends of the plateau [$s_{\text{cut}} \sim 2(2\mu R)^{1/3}$ for $\mu \gg 1$] and very small in the middle of it. As Γ increases (or μ rapidly varies in time), these jumps become significantly inhibited, and the spectrum smoothes out; see curve 4b in Fig. 1. [Note that in the limit $N^2 \gg R^2$ and $\theta = 1$, Eq. (9) results in $p_n \propto J_{\tilde{N}}(2\mu R)/\tilde{N}$, with complex index $\tilde{N} = N + iR\Gamma$.]

Although the real systems (rare gases and rare-gas-like ions) in which HHG has been observed so far are much more complex than a two-level atom, they may not be inconsistent with the two-level description as far as HHG is concerned. Indeed, the energy E_1 of a rare-gas atom is fairly large ($R = E_1/\hbar\omega > > 1$) and very close to E_{ion}, so that $\Delta E \equiv E_{\text{ion}} - E_1 \ll E_{\text{ion}}$, and $\Delta E \sim \hbar\omega$. Thus virtually *all* the higher harmonics, in particular, most of those inside the plateau (except for very few of them), may "see" a cluster of atomic levels between E_1 and E_{ion} as a *single* level; its effective parameters are determined by all the contributing levels, with a strong dominance of the first excited level. Rabi splitting of each individual level in that cluster and multiple- (most of them avoided) level crossings may also be instrumental in forming a bandlike single level. Comparison of the model with experimental data is encouraging. The inset in Fig. 1 shows a theoretical straight line [see Eq. (19)] in the space of parameters ν^2 and $|E|^2 \propto \mu^2$, and available experimental points for Xe [2(a),4]. The fit is good considering that no other of many contributing effects was accounted for. Writing now $\mu^2=\beta I$, where I is the driving intensity in 10^{13} W/cm^2 and $\beta \approx 0.47$ is the coefficient found by us from this plot, we evaluate critical intensity for plateau formation, Eq. (20), for Xe as 5.5×10^{12} W/cm, which compares well with experiment [2], 5.0×10^{12} W/cm^2.

Ions produced by the ionization of rare-gas atoms have an electronic structure significantly different from that of an original rare-gas atom, such that the two-level model may not apply to them. For example, a single-ionized He atom is a H-like ion, whose energy levels are distributed much more evenly (and therefore are less bandlike) than those of He. Because of the significant increase of the transition frequencies in ions, however, the harmonic intensities at the "ionic" plateau are expected to be much lower than those for the main, "atomic" plateau, so that Eqs. (17) and (19) are expected to be valid for the main plateau.

It is known [2(a)] that before the saturation sets in, the slope α_N of $\log_{10}(|p_n|)$ vs $\log_{10}(\mu)$ for $N=2n-1$ inside the plateau is less than N, as opposed to the perturbation-theory predictions. Consistently with [2(a)], we found that for the harmonics above the resonance ($N=13-21$), α_N is insensitive to N, $\alpha_N \sim 12-13$. Furthermore, again using $\beta=0.47$, we were able to closely fit (Fig. 2) experimental [7] and theoretical data on the Nth harmonic intensity (for $N=17,19,21$) vs μ^2, assuming [2] that the harmonic amplitude reflects atomic response, i.e., polarization. We chose $\Gamma=0.1$ for theoretical curves for all the harmonics since for larger Γ's theoretical curves are close to their envelopes measured in short-pulse experiments [for comparison, for $N=17$ we show a curve, 1(b), for $\Gamma=0.01$]. Our calculations predict that (at least for $N>R$) the onset of saturation of p_n (as μ increases) occurs at different intensities for each individual harmonic N:

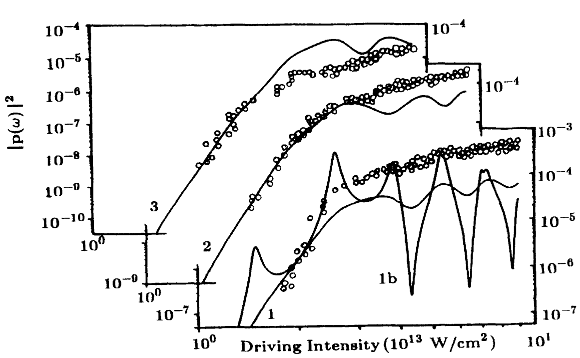

FIG. 2. ρ_n^2 vs μ^2 for $R=7.24$ (Xe+Nd:YAG laser) and various N. Solid lines: theory ($\Gamma=0.1$, except 1b, where $\Gamma=0.01$), circles: experiment [7]. Curves: 1, $N=17$; 2, $N=19$; 3, $N=21$.

$$\mu_{\rm sat}(N)\approx N/2R, \quad (\Omega_R)_{\rm sat}\approx N\omega/2 , \tag{21}$$

which offers a very simple saturation formula. Figure 2 shows that Eq. (21) is consistent with experiment [7] for $N=17,19,21$. We found that a similar good fit could be attained for harmonics 15–9 by reducing $\beta^{1/2}\propto d_{12}$ by a factor of 1.2–1.5, in amazing agreement with a similar adjustment in Ref. [9].

One of the factors believed to be a major reason for saturation of individual harmonics [2] is the depletion of neutral atoms due to ionization, although neither theoretical nor experimental proof has been offered for this conjecture. The depletion of neutrals due to ionization must certainly be a factor in the HHG phenomenon, to the extent "external" to any model. It is hard to believe, however, that the ionization makes a drastic impact on the saturation of individual harmonics. First of all, while ionization may deplete the population of neutral atoms by one or two orders of magnitude, the amplitude of very high harmonics are affected by other factors by many (often more than ten) orders of magnitude. Second, calculations based on the standard Hartree-Slater approach [7] show that ". . . the saturation seems to also be present in the single-atom response, without invoking. . . the depletion of the neutral-atom population. . . ." Third, there are experimental observations of saturation [9] indicating almost negligible contribution of the ionization to HHG (for subpicosecond pulses). Finally, one can argue that if the ionization were the major factor, all the harmonics would undergo the saturation at the same driving intensity, contrary to the experimental observations that the saturation intensity significantly increases with the harmonic number. Amazingly enough, Eq. (21) shows a good agreement with experiment [7,9].

A significant feature in Fig. 2, curve 1(b), consistent with experimental data [7,9,21] is intensity-induced resonances in HHG when $\Gamma \ll 1$. The most pronounced and "ordered," though, are very large multiresonances in the population difference x_0 [see Fig. 3(a), curve 1, for $\Gamma=10^{-3}$]. They appear almost periodically with the amplitude μ as it increases. For the sake of demonstration, we chose relatively low resonant frequency, $R=4.25$, and $\Gamma=10^{-3}$; for larger R the resonances become too sharp. [Due to complex $\tilde{N}$ in Eqs. (5) and (9), the resonances are inhibited as Γ increases; see Fig. 3(a), curve 2, for $\Gamma=0.1$]. The smooth saturation envelope, curve 3 in Fig. 3(a), is obtained analytically from Eq. (9) with w_1 approximated by Eq. (12) for $n=1$.

We found that all these resonances are directly related to the structure of the intensity-induced eigenfrequencies (or quasienergies) $\lambda(\mu)$ (essentially "super-Rabi" frequencies) of a hard-driven two-level atom. We use the term "super" here to emphasize the fact that the Rabi frequency becomes larger than the initial resonant frequency of the system, thus marking a very large perturbation in the system. Due to Floquet theory, the "eigensolution" (i.e., the one with frequencies different from the driving frequency or its higher harmonics) of Eqs. (1) and (2) can be sought in the form

$$\Delta x = e^{-i\lambda\omega t}\sum_{n=-\infty}^{\infty} a_n e^{i(2n+1)\omega t} + \text{c.c.} , \tag{22}$$

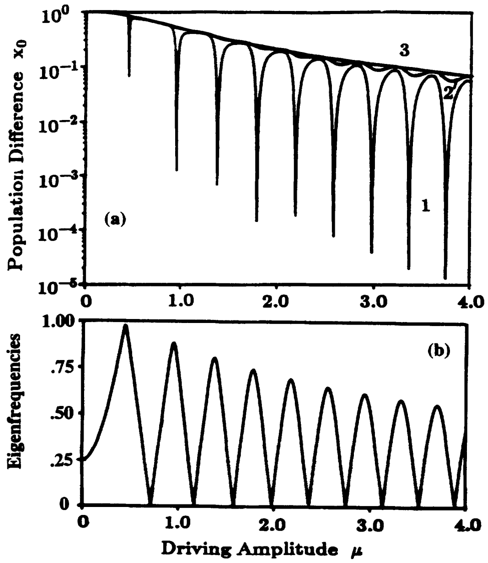

FIG. 3. Multiresonances in population difference x_0 (a) and eigenfrequency λ_0 in the (0,1) domain (b) vs amplitude μ, for $R=4.25$. Curves in (a): 1, $\Gamma=10^{-3}$; 2, $\Gamma=0.1$; 3, envelope approximation.

leading to the infinite set of linear algebraic equations for a_n. These equations are obtained from Eqs. (1) and (2) by assuming that the total solution is $x+\Delta x$ (and $y+\Delta y$, with Δy found through Δx) where x and y are the driven solutions, Eq. (3). Each of these linear equations couples three neighboring coefficients a_{n-1}, a_n, and a_{n+1}. The eigenfrequencies λ are found by setting the determinant Δ_λ of that infinite set of linear equations for a_n to zero. Thus there is an infinite set of the solutions for the super-Rabi frequencies λ; the most important contribution is provided by those which lie below the cutoff. In the superdriven two-level atom the contribution of *many* eigenfrequencies (or quasienergies) λ to the oscillation in the system becomes significant; by contrast, a familiar weakly perturbed two-level dressed atom [22] is comprehensively described by the splitting of just two original energy levels.

A property of Floquet solutions, Eq. (22), is that if some $\lambda_0 \in (0,1)$ is an eigenvalue, then any $\lambda=2n\pm\lambda_0$, $n\in(-\infty,\infty)$ is an eigenvalue too. In our case, all the solutions $\lambda(\mu)$ are developed by the interplay of two families of curves (with avoided crossings between some of them), $(\Lambda_n)_{1,2}=2n\pm[R+F(\mu)]$, where $F(0)=dF(0)/d\mu=0$ and $dF/d\mu\rightarrow$const as $\mu\rightarrow\infty$; there is only one μ-dependent eigenvalue $\lambda_0\in(0,1)$ for each μ. Note that $(-1,0)\ni\lambda=-\lambda_0$. Because of asymptotic properties of $F(\mu)$, $\lambda_0(\mu)$ is an almost periodic function, with a slowly decaying amplitude due to increasingly widening gaps at the points of avoided crossings of $(\Lambda)_1$ and $(\Lambda)_2$ branches around $\lambda=2n-1$ (the crossings at $\lambda=2n$ are not forbidden). $\lambda_0(\mu)\in(0,1)$ for $R=4.25$, as a numerical solution of the algebraic equation $\Delta_\lambda=0$ for Δ_λ of the 44th order, with $T^{-1}=\tau^{-1}=0$, is depicted in Fig. 3(b), and demonstrates a close resemblance between our numerical results and that of Ref. [12]. The multiresonances of population difference occur almost exactly at the maxima of the function $\lambda_0(\mu)$, i.e., near avoided crossings, with all the harmonics simultaneously resonant to the intensity-induced frequencies $(\Lambda)_{1,2}$. This behavior is universal; for $\mu^2R^2\gg1$ the resonances occur at the zeros of the analytic solution for such a case, $x_0=J_0(2\mu R)$, or, in the limit $\mu\gg1$, at the zeros of $\sin(2\mu R+\phi)$, where ϕ is a constant. Thus the spacing between any two adjacent resonances is

$$\Delta\mu\approx\pi/2R,\quad \Delta(\Omega_R)\approx\pi\omega/2\ . \tag{23}$$

Some distinct resonant features have been observed in many experiments [21,23], and have been attributed to some unspecified or rather conjectured atomic resonances. Although this may be true, the resonances predicted by us may constitute substantial if not dominant, contribution to this phenomenon. The major manifestation of the new, intensity-induced resonances that set them aside from the other resonances is their near-periodical dependence on the driving amplitude. It is worth noting that according to the computer simulation in Ref. [9] based on model [8], "the phase of dipole . . . appears to sweep through resonances at some integer value of η" (η in [9] is related to the driving intensity). Since the theoretical model [8] assumes a δ-function potential, this may indicate some profound underlying similarities between the behavior of such a system and the two-level atom. The resonances disucssed here may be observed using time-resolved spectroscopy with longer driving pulses in order not to smear out the resonances by rapidly varying amplitude $\mu(t)$. Also, while at the multiresonances the dc population difference is inhibited, the population *oscillations,* in particular, at the frequency 2ω ($|w_1|\gg1$), substantially increase, which may be observed using a probe field resonant to some isolated quantum level.

In conclusion, a simple nonperturbative two-level model of a hard-driven atom reproduces major HHG features, analytically describes their characteristics, compares well with the experiment for some gasses, and predicts new intensity-induced multiresonances. Aside from a possible HHG mechanism in rare-gas atoms or ions by optical lasers, our results suggest that (in addition to the known nonresonance effects, such as in plasma at surfaces), HHG based on *two-level* systems may also be produced in other media and frequency domains, e.g., by microwave sources (such as gyrotrons) in an electron gas [24] or plasma in a magnetic dc field, or by IR lasers (e.g., CO and CO_2 lasers) in gasses or even semiconductors.

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