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NONLINEAR OPTICAL EFFECTS IN A SUPER-DRESSED TWO-LEVEL ATOM

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Abstract

A simple nonperturbative two-level model of an atom driven by a very strong EM periodic field, with a Rabi frequency exceeding a resonant frequency, results in a rich picture of nonlinear optical effects. The most pronounced of them are very high harmonic generation (HHG) and intensity-induced multi-transparency resonances (IDMT). The model reproduces the experimentally observed plateau in HHG and yields for the first time simple analytic formulas for the plateau cutoff frequency, critical driving intensity, and saturation. It also predicts that IDMT resonances should occur approximately at the subharmonics of the time-averaged instantaneous Rabi frequency.

One of the most fascinating phenomenon 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/cm² and higher) optical laser radiation in rare gasses 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 appears to be also a generic feature of many nonlinear models, including classical nonlinear systems [2c]). 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, tremendous amount of calculations and involves many processes, making it difficult to gain simple insights. An interesting simplified model [8] based on a 3-D delta-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 unapplicable to rare-gas atoms used in experiment [1-4,7,9]. A very interesting and detailed earlier work [12] holds only for quasi-adiabatic approximation. Besides, no relaxation was considered in [10-12].

In our work [13] we developed 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 new dramatic effects. In this model, we completely abandon rotation approximation and consider fully non-perturbative theory of the phenomenon, whereby the Rabi frequency may not only be much smaller than the resonant

frequency of two-level system (as in the rotation approximation), but may become of the same order as or even significantly greater than that resonant frequency. 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 , and are consistent with the available experimental data for some rare gases. We also predict well-ordered multi-resonances in HHG and in population difference vs driving intensity when the energy of interaction becomes comparable with or substantially exceeds energy E_1 (i. e. when Rabi frequency exceeds the resonant frequency). These resonances are indication of intensity-induced eigenfrequencies (or eigenenergies) in the super-driven two-level system and are obtained using Floquet theory. The dramatic manifestation of these resonances is multi-transparency, whereby at certain driving intensities, the "Rabi-sphere" of the super-driven two-level system collapses, resulting in almost full suppression of population difference and polarization at all the harmonics (including polarization at the driving frequency). 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 nondiagonal elements, $\rho_{12} = \rho_{21}^*$, -- to the induced polarization $\vec{P} = \vec{d}_{12}(\rho_{12} + \rho_{21})$, where \vec{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 Boltzman distribution; $\eta_0 = \tanh(-kT_R/2\hbar\omega_0)$, where k is Boltzman 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 $\vec{E}(t) = \vec{E} \sin\omega t$, is governed by equations [14]:

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

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

where Rabi frequency $\Omega_R \equiv \vec{d}_{12} \vec{E}/\hbar$ is a measure of the dipole interaction energy, T and τ are longitudinal and transverse relaxation times respectively. Eqs. (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 series:

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

By solving first Eq.(2) for y :

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

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

$$f_n (1 - w_n^{-1}) + f_{n+1} (1 - w_{n+1}) = 2\bar{n}/R^2 \mu^2; \quad n \geq 1, \quad (5)$$

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

$$\mu \equiv \Omega_R/\omega_0 = \vec{d}_{12} \vec{E}/\hbar\omega_0$$

is a dimensionless driving amplitude; thus

$$x_n = x_0 w_1 w_2 \dots w_n. \quad (6)$$

The Fourier coefficients p_n of normalized polarization

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

are obtained then as

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

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

$$(g_n/v_n - 1)/\bar{n} + (v_{n+1} - g_{n+1})/(\bar{n} + 1) + 2/(\mu^2 R^2 f_{n+1}) = 0; \quad n \geq 1, \quad (9)$$

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

$$p_{n+1} = p_1 v_1 v_2 \dots v_n. \quad (10)$$

The equation for x_0 is:

$$x_0^I - I = \mu^2 R \operatorname{Im}[f_1(I - w_1)] / \theta \Gamma; \quad (11)$$

It coincides with a familiar formula for resonant saturation when $R = I$, $\mu \ll I$, and $w_1 = 0$. Physical solutions w_n , v_n , of Eqs. (5), (9) and (11) are those vanishing as $n \rightarrow \infty$. Fig. 1 shows the $(|p_n|^2)$ spectra calculated by setting $w_{500} = 0$ in Eq. (5) for $\theta = I$, $\Gamma = I0^{-3}$, and $R = 7.24$ (assuming $\omega_0 = E_I/\hbar$ and ω as in a Xe atom driven by a Nd-YAG laser). Formation of plateau and "irregular" behavior inside it as μ increases, are evident. In the small perturbation approximation ($\mu^2 \ll |(N/R)^2 - I|$), Eqs. (5) and (9) yield:

$$w_n = f_n / (f_n + f_{n+1} - 2\bar{n}/\mu^2 R^2); \quad v_n = g_n / [I + \bar{n}g_{n+1}/(\bar{n} + 1) - 2\bar{n}/\mu^2 R^2 f_{n+1}], \quad (12)$$

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

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

If $R \ll I$ (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] ($\mu \ll I$, $R = I$). The solution [11,15] is then

$$x = \cos(2\mu R \cos \omega t); \quad p = -R \int \sin(2\mu R \cos \omega t) d(\omega t), \quad (14)$$

i. e.

$$x_n = (-I)^n J_{2n}(2\mu R), \quad p_n = R(-I)^n J_N(2\mu R)/N, \quad (15)$$

where $N = 2n - I$, and $J_m(z)$ is an ordinary Bessel function of the m -th order. (Note that $p \rightarrow 0$ as $R \rightarrow 0$). Eqs. (5) and (9) show that spectrum-wise, this approximation holds for arbitrary R to some factor $C = C(R, \mu)$ and only for $N^2 \gg R^2$ [18]. If now $R \gg I$ (a typical situation for the HHG experiments [2]), neglecting the term \ddot{p}/ω_0^2 in the above equations, one obtains [12]

$$x \approx \bar{x} = (I + 4\mu^2 \sin^2 \omega t)^{-1/2}; \quad (16)$$

Fourier components \bar{x}_n and \bar{p}_n are expressed then in terms of elliptic integrals. Spectrum-wise, 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 $N \rightarrow \infty$ and $N \leq R$, 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 $n \rightarrow \infty$ -- with similar behavior of $J_n(z)$ for sufficiently large n (see below). By assuming e. g. in Eq. (5) that $w_{n+1} = w_n$ and $f_{n+1} = f_n$, Eq. (5) is reduced to a quadratic equation for w_n ,

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

Its solution,

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

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

$$\text{either } N \leq R, \text{ or } N \geq N_{cut} = R \sqrt{1 + 4\mu^2}. \tag{17}$$

(The upper sign in the formula for $\bar{w}_n^{(out)}$ corresponds to $N \geq N_{cut}$, and the lower sign -- to $N \leq R$.) Between these two points there lays an "inside" area, $R < N < N_{cut}$, with distinct irregular spectra, Fig. 1, consistent with other models [2,6]. These chaos-like, 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] $z_N = N + 2N^{1/3} + N^{-1/3} + O(N^{-1})$. 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 chaos-like behavior of harmonic amplitudes as function of their order, similar in nature to the ball dynamics in "round billiard" in the case when the single angular "hop" of the ball is incommensurate with 2π . The smooth envelopes of these spectra, $\bar{w}_n^{(in)}$ and $\bar{v}_n^{(in)}$ (they also approximate monotonic spectra

for $N \leq R$ for large μ and away from resonances, Eq. (24) below) are obtained by assuming $\mu \rightarrow \infty$ in Eqs. (5) and (9), which results in $\bar{w}_n^{(in)} = I$, and $\bar{v}_n^{(in)} = g_n = N/(N+2)$, i. e.

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

The analytic envelopes for p_n based on $\bar{v}_n^{(out)}$ and $\bar{v}_n^{(in)}$, are shown at Fig. 1, curves (1-4)a; 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:

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

which allows for comparison of different atoms driven by different lasers. The plateau width scales almost quadratically with μ (or with the driving *amplitude*) when $\mu \ll 1$ (similarly to [6]), and linearly [10], $N_{cut} = 2\Omega_R/\omega$, -- 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 $N \geq N_{cut} = 2\mu R$, and rapidly vanishes as $N \rightarrow \infty$. 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 (cut-off) 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 results [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 $-3U_p$, 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 -10^{14} W/cm². 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 is shown by Corkum in his "almost-classical" 3-D 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 $R \gg I$, the critical μ required for the plateau to appear at all is evaluated as:

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

Since $\Omega_R \approx \vec{d}_{12} \vec{E}$, $d_{12} \approx \omega_0^{-1/2}$, and $E^2 \approx n_{ph} \omega$, where n_{ph} is flux of driving photons, Eq. (20) suggests that for $\omega_0 \gg \omega$, $(n_{ph})_{cr} \approx \omega_0^2$ and does not depend on ω ; this indicates the potential for HHG at lower frequencies. Fig. 1 shows that consistently with experiment [3], the harmonic intensities as function of their order, make a dip minimum which could be substantially below the envelope, Eq. (18), if $\mu \gg \mu_{cr}$. The position, N_I , of this first minimum for relatively small μ is $N_I \approx R + \mu/\mu_{cr}$. For $\mu \gg I$ N_I fluctuates considerably with μ in a periodic fashion (see also below, Eq. (24)); however $(N_I)_{max} \approx O[(\mu R)^{2/3}]$. As μ increases, the chaos-like 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 ($s_{cut} \approx 2(2\mu R)^{1/3}$ for $\mu \gg I$) 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 $\theta = I$, Eq. (9) results in $p_N \approx J_{\bar{N}}(2\mu R)/\bar{N}$, with complex index $\bar{N} = N + iR\Gamma$.)

Although the real systems (rare gasses 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 two-level description as far as HHG is concerned. Indeed, the energy E_I of a rare-gas atom is fairly large ($R = E_I/\hbar\omega \gg I$) and very close to E_{ion} , so that $\Delta E \equiv E_{ion} - E_I \ll E_{ion}$, and $\Delta E \approx \hbar\omega$. 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_I 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 further be instrumental in forming a band-like 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 v^2 and

$|E|^2 \propto \mu^2$, and available experimental points for Xe [2a,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 $\beta = 0.47$ is the coefficient found by us from this plot, we evaluate critical intensity for plateau formation, Eq. (20), for Xe as $5.5 \times 10^{12} \text{ W/cm}^2$ which compares well with the experiment [2], $5.0 \times 10^{12} \text{ W/cm}^2$. We note here that $\beta = 0.47$ corresponds to the effective size of the dipole moment, $d_{12}/e = 7\text{\AA}$ which is not inconsistent with the notion of the effective two-level system incorporating the properties of the group of quantum levels (most of them -- Rydberg states) laying between the first excited level of a rare-gas atom and ionization limit.

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 "band-like") than in 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 [2a] that before the saturation sets in, the slope, α_N , of $\log(|p_N|)$ vs $\log(\mu)$ for $N = 2n - 1$ inside the plateau is less than N , as opposed to the perturbation theory predictions. Consistently with [2a] we found that for the harmonics above the resonance ($N = 13 - 21$), α_N is insensitive to N , $\alpha_N \approx 12 - 13$. Furthermore, using again $\beta = 0.47$, we were able to closely fit, Fig. 2, experimental [7] and theoretical data on N -th 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, 1b, 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 :

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

which offers a very simple saturation formula. Fig. 2 shows that Eq. (21) is consistent with

experiment [7] for $N=17, 19, 21$. We found that similarly good fit could be attained for harmonics 15 through 9, by reducing $\beta^{1/2} \propto d_{12}$ by factor of 1.2 to 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 conjuncture. 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 population of neutral atoms by one or two orders of magnitude, the amplitudes of very high harmonics are affected by other factors, that may change them by many orders of magnitude. Second, calculations based on 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 pretty good agreement with experiment [7,9].

A significant feature in Fig. 2, curve 1b, 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 multi-resonances in the population difference, x_0 (see curve 1, Fig. 3a, 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 \bar{N} in Eqs. (5) and (9), the resonances are inhibited as Γ increases, see curve 2, Fig. 3a, for $\Gamma=0.1$.) The smooth saturation envelope, curve 3 in Fig.3a, is obtained analytically from Eq. (9) with w_I approximated by Eq. (12) for $n=1$. We also found that at these resonances, not only the zero-frequency component

of population difference is suppressed, but all the higher harmonics of population difference, as well as all the harmonics of polarization (including the component at the driving frequency), are suppressed as well. Essentially, the entire dynamics of the two-level system at these resonances is almost fully suppressed. This effect therefore may be regarded as multiple intensity-induced transparency, reminiscent of the effect of the intensity-induced inhibited dynamics in two-level system excited by a periodically modulated driving resonant to ω_0 , in the *perturbative* limit (i. e. in the rotating wave approximation). It was shown recently [25] that at certain (multiple) resonant conditions, a periodically modulated field driving a two-level system, can suppress the dynamics of both its population and polarization and induce self-transparency. Outside of these narrow resonances, the system exhibits a high-order frequency mixing (analogous therefore to the very high order harmonics generation in a super-dressed atom) with the spectral plateau giving rise to a soliton train as the field propagates. Following the approach [25], these resonances can be regarded as a collapse of Rabi-sphere of the system resulting in a driving-induced transparency. In the no-relaxation case, Eq. (13) yields:

$$x^2 + p^2 + (\dot{p}/\omega_0)^2 = \text{const} = r_{Rabi}^2, \quad (22)$$

where r_{Rabi} is a Rabi-sphere radius; in the ideal, no-relaxation case, $r_{Rabi} = I$. One can prove [25] that when the relaxation times are the same, $T = \tau$, and sufficiently long, $\Gamma \ll I$, Eq. (22) is still valid in the sense that its lhs remains almost constant in time, however this constant, r_{Rabi}^2 , is now smaller than I and in general depends on the driving intensity and on the frequencies ω^{-I} and ω_0^{-I} . At certain (multiple) intensities (see below), r_{Rabi} vanishes resulting in the intensity-induced transparency resonances.

We found that all these resonances are directly related to the structure of the intensity-induced eigenfrequencies (or quasi-energies), $\lambda(\mu)$ (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

$$\Delta x = e^{-i\lambda\omega t} \sum_{n=-\infty}^{\infty} a_n e^{i(2n+1)\omega t} + c. c., \quad (23)$$

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 infinite number of the solutions for the "super-Rabi" frequencies λ 's; the most important contribution is provided by those of them which lie below the cutoff. In the super-driven two-level atom the contribution of *many* eigenfrequencies (or quasi-energies), λ 's, to the oscillation in the system becomes significant; by contrast, a familiar weakly perturbed two-level dressed atom [22] is comprehensively described by splitting of just two original energy levels.

A property of Floquet solutions, Eq. (23), is that if some $\lambda_0 \in (0, I)$ 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 $F(\mu)$ is some monotonically increasing function of μ with $F(0) = dF(0)/d\mu = 0$ and with $dF/d\mu \rightarrow \text{const}$ as $\mu \rightarrow \infty$; there is only one μ -dependent eigenvalue $\lambda_0 \in (0, I)$ for each μ . Note that $(-I, 0) \ni \lambda = -\lambda_0$. Because of asymptotic properties of $F(\mu)$, $\lambda_0(\mu)$ is an almost periodic function, with slowly decaying amplitude due to increasingly widening gaps at the points of avoided crossing of (Λ_1) and (Λ_2) branches around $\lambda = 2n - I$ (the crossings at $\lambda = 2n$ are not forbidden.) $\lambda_0(\mu) \in (0, I)$ for $R = 4.25$, as a numerical solution of the algebraic equation $\Delta_\lambda = 0$ for Δ_λ of the 44-th order, with $T^{-I} = \tau^{-I} = 0$, is depicted in Fig. 3b, and demonstrates a close resemblance between our numerical results and that of Ref. [12]. The multi-resonances of population difference occur almost exactly at the maxima of the function $\lambda_0(\mu)$, i. e. near avoided crossing, with all the harmonics simultaneously resonant to the intensity-induced frequencies $(\Lambda_1)_{1,2}$. This behavior is universal; for $\mu^2 R^2 \gg I$ 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 $\mu \gg I$, at the zeros of $\sin(2\mu R + \phi)$, where ϕ is a constant. Thus, the spacing between any two adjacent resonances is:

$$\Delta\mu = \pi/2R, \quad \Delta(\Omega_R) = \pi\omega/2. \quad (24)$$

Writing this formula as

$$\omega + \omega_R = (2/\pi) \cdot (\Omega_R/n), \quad (n \gg 1) \quad (25)$$

where n is integer, and ω_R is some constant ($\omega_R/\omega = O(1)$) determined by the ratio ω/ω_0 , we note that the Rabi frequency used by us, $\Omega_R = d_{12}E/\hbar$, reflects *amplitude* E of the field. Ω_R could be related to the *instantaneous* Rabi frequency, $\bar{\Omega}_R(t) = d_{12}|\vec{E}(t)|/\hbar$ where $\vec{E}(t) = \vec{E} \sin \omega t$ is an instantaneous driving field, as $\Omega_R = \bar{\Omega}_R \cdot \pi/2$ with $\bar{\Omega}_R = (\omega/2\pi) \int_0^{2\pi/\omega} \bar{\Omega}_R dt$, where $\bar{\Omega}_R$ is a *time-averaged* instantaneous Rabi frequency. Thus, Eq. (25) can be rewritten now as

$$\omega + \omega_R = \bar{\Omega}_R/n, \quad (26)$$

which can be interpreted as resonances at "subharmonics" of the time-averaged Rabi frequency. This property makes the discussed phenomenon reminiscent of the so called subharmonic resonances occurring in the slightly-perturbed two-level system driven by relatively weak ($d_{12}\vec{E} \ll \hbar\omega_0$) resonant field whose amplitude is modulated, when the frequency of amplitude modulation is close to a subharmonic of the averaged Rabi frequency, [26] (see also references therein to earlier work). This analogy was explored in [25].

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 sets 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 delta-function potential, this may indicate some profound underlying similarities between behavior of such a system and the two-level atom. The resonances discussed here may be observed using time-resolved spectroscopy with longer driving pulses in order not to smear out the resonances by rapidly varying amplitude $\mu(t)$.

It is worth mentioning here that strongly-driven atoms with Raman-active transitions (which may be described by a generalized two-level system with allowed two-photon transitions) can also exhibit strongly nonlinear-optical properties. It was shown recently [27] that in the process of multi-frequency Cascade Stimulated Raman Scattering (CSRS), Raman active materials (with the transition frequency ω_0) can support solitons consisting of pump laser wave (with the frequency ω) and many Stokes and anti-Stokes components with their frequencies $\omega_j = \omega \pm j\omega_0, j = 1, 2, 3, \dots$. These multi-frequency waves form 2π -solitons mode-locked in their propagation by the dynamics of population at Raman quantum transition. Soliton components at each individual frequency are "bright" pulses of the same, Lorentzian, shape (in contrast to the familiar bright+dark soliton pair in a regular SRS). The most striking result is that the coherent interference of the multiple CSRS components may give rise to the train of well resolved *subfemtosecond* pulses (with the duration down to $\sim 10^{-16}$ s) of high intensity (up to $\sim 10^{14} - 10^{15}$ W/cm²).

In conclusion, a simple nonperturbative two-level model of a hard-driven atom demonstrates capability of the system to generate very high harmonics, reproduces major HHG features, analytically describes their characteristics, compares well with the experiment, and predicts new intensity-induced multi-transparency resonances. Aside from a possible HHG mechanism in rare-gas atoms or ions by optical lasers, our results suggest that (in addition to the known non-resonance effects, such as in plasma at surfaces), HHG based on *two-level* systems may also be feasible in other media and frequency domains, e. g. by microwave sources (such as gyrotrons) in electron gas [24] or plasma in magnetic *dc* field, or by IR-lasers (e. g. CO and CO₂ lasers) in gasses or even semiconductors.

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Figure Captions

- 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 -- $\mu=6$; (1-4)a -- respective envelop approximations. Arrows indicate plateau cutoff points. Inset: Cutoff ratio square, v^2 , vs μ^2 for Xe. Crosses -- experiment [2a,4], solid line -- linear fit.
- Fig.2 p_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 -- $N=21$.
- Fig.3 Multi-resonances in population difference, x_0 , (a), and eigen-frequency 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 -- envelop approximation.

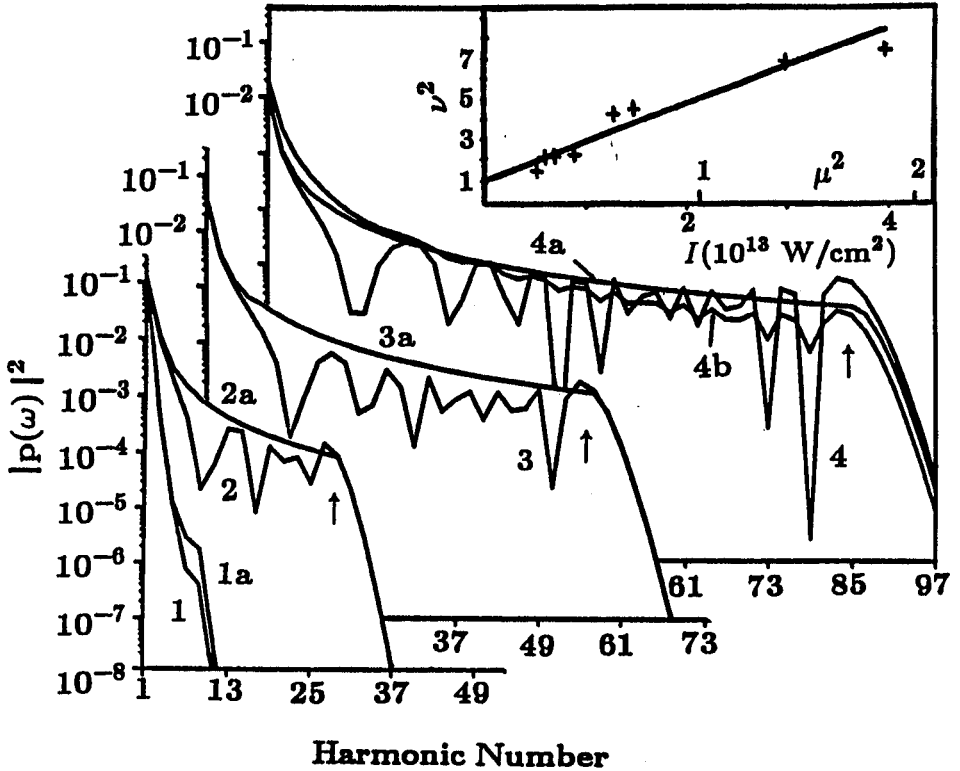


Fig.1

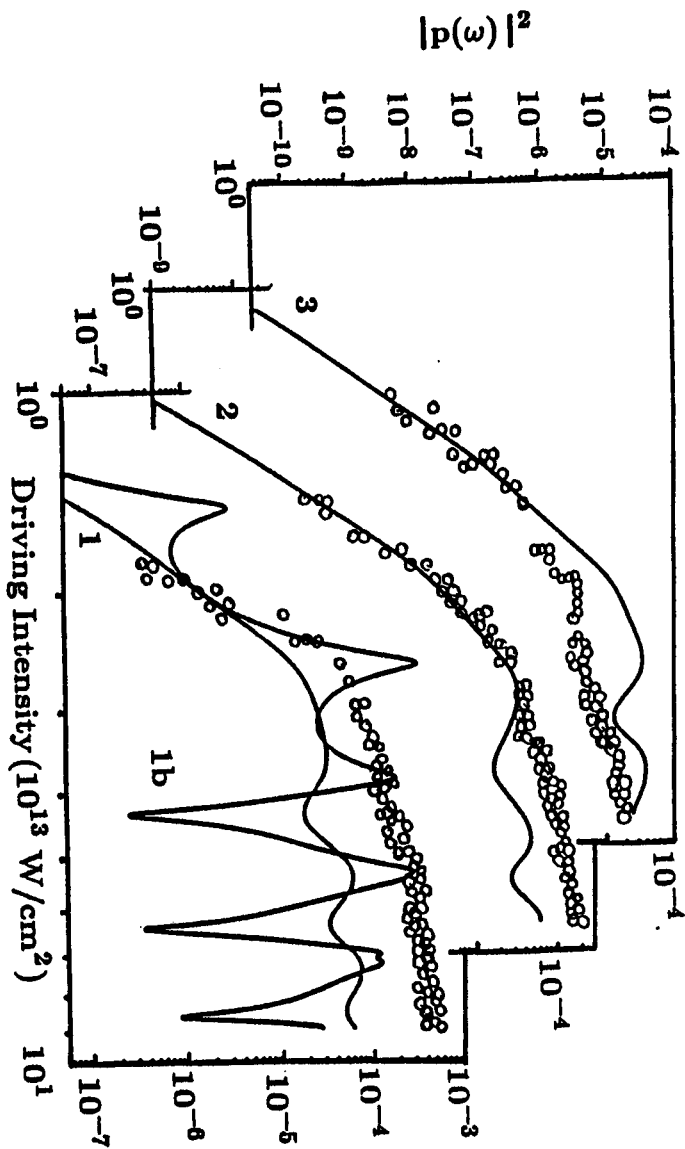


Fig. 2

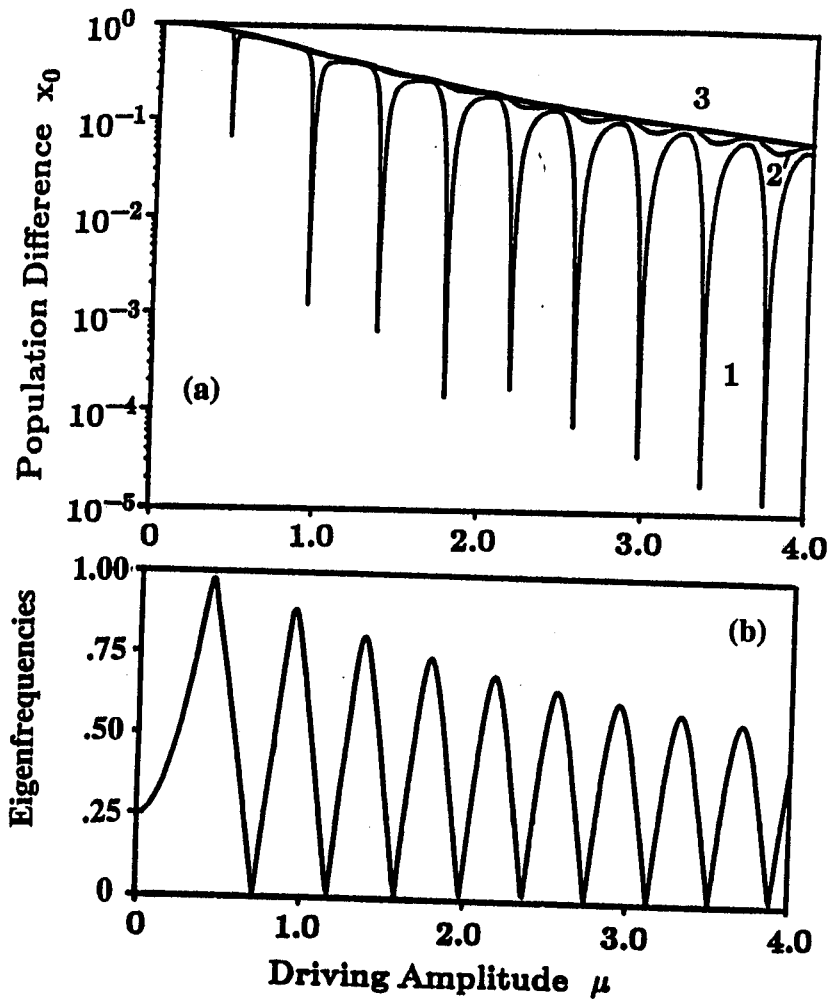


Fig.3