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High-order harmonic generation: Simplified model and relevance of single-atom theories to experiment

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We present a simple two-state model illustrating the fact that the most prominent features observed in high-order harmonic generation are generic to strongly driven systems. We also address two important questions that arise in the comparison of theory and experiment.

In experiments employing very intense laser fields ($I > 10^{13}$ W/cm^2) in gaseous media, high-order harmonics of the fundamental driving frequency have been observed. For instance, the thirty-third harmonic has been obtained in argon with 1.06 μm irradiation,[1] and with irradiation by a 248-nm excimer laser the seventeenth harmonic has been reported, corresponding to a wavelength of 14.6 nm.[2] Thus far the most successful computational approach to this harmonic generation involves direct numerical integration of the time-dependent Schrödinger equation.[3,4] The numerical results display some of the prominent features observed experimentally: (1) the spectrum of scattered radiation consists of peaks at the odd harmonics of the fundamental driving frequency; (2) the spectrum has a plateau region in which harmonic peaks are of similar strength; and (3) there is a rapid cutoff at the highest harmonics.

In this Rapid Communication we first describe a simple model for this high-order harmonic generation. Our purpose in doing so is mainly to point out that the same qualitative features (1)–(3) just mentioned are generic to strongly driven systems, and are not peculiar to atoms in quasimonochromatic fields. Furthermore, such a simple model allows us to study some aspects of harmonic generation with a minimum of computational effort.

A second purpose of this paper is to address two questions that, to our knowledge, have not previously been discussed in the literature: (1) Should the calculation of the spectrum be based on the power spectrum of the expectation value of the induced dipole moment, or of the dipole correlation function? We show that it should be based on the latter. However, we also suggest why the existing theories, which employ only the dipole expectation value, may in fact be relevant to the experiments. (2) What is the appropriate power of the frequency that should multiply the square of the Fourier transform of the dipole correlation function (or expectation value)? We show that the answer depends critically on whether one is considering single-atom scattering or the actual experimental situation in which multiatom scattering and propagation effects must be addressed. We suggest, therefore, that spectra measured from single atoms or atomic beams would be different from those measured in the experiments cited above.

The model we consider is quantum mechanical but involves only two atomic states. Thus we consider the following optical Bloch equations[5] for a two-state atom in a quasimonochromatic field:

$$\dot{x} = -\omega_0 y , \tag{1a}$$

$$\dot{y} = \omega_0 x + \Omega \sin(\omega t) z , \tag{1b}$$

$$\dot{z} = -\Omega \sin(\omega t) y , \tag{1c}$$

where ω_0 is the transition frequency, z is the difference between the upper- and lower-state probabilities, and x and y involve cross products of the upper- and lower-state probability amplitudes.[5] $\Omega = (2d/\hbar)E_0$, where E_0 is the electric-field amplitude and d is the transition dipole moment. Note that we do not make the rotating-wave approximation (RWA).

We compute using Eqs. (1) the squared modulus of the Fourier transform of the induced dipole moment $ex(t)$. As in previous work[3,4] we temporarily associate this quantity with the spectrum of scattered radiation. In our computations the field is ramped on according to the algorithm

$$E = \begin{cases} E_0 \sin^2(\omega t/4a)\sin(\omega t), & 0 < t < 2\pi a/\omega, \\ E_0 \sin(\omega t), & t \geq 2\pi a/\omega, \end{cases} \tag{2}$$

where a, the number of cycles required to "turn on" the field, is typically set equal to five. For fairly weak fields the computed spectrum shows peaks at the first few odd harmonics of the applied frequency, but the peak amplitudes fall off monotonically with increasing harmonic order, as shown, for example, in Fig. 1. The appearance of

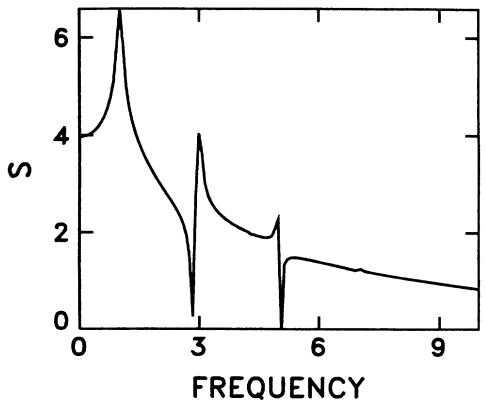

FIG. 1. Log spectrum $S(\omega)$ for a two-level model with $\Omega/\omega = 0.5$ and $\omega/\omega_0 = 0.25$.

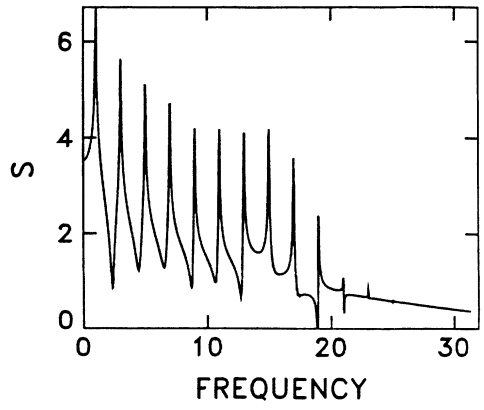

FIG. 2. As in Fig. 1 with $\Omega/\omega=4.0$ and $\omega/\omega_0=0.25$.

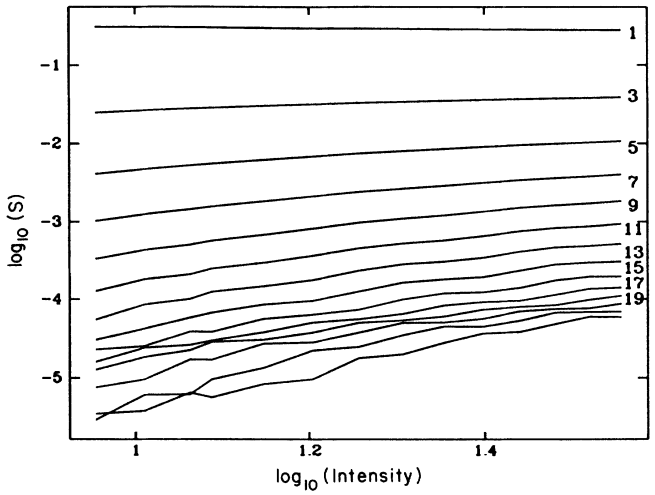

FIG. 3. Intensity dependence of different harmonics obtained with a two-level model, showing that higher harmonics have approximately the same intensity dependence. The integer labels for the different curves indicate the harmonic orders.

only odd harmonics follows automatically from such an approach, without direct recourse to dipole selection rules.

For larger values of Ω we observe qualitatively the same plateau and cutoff structure obtained by numerical integration of the full Schrödinger equation. The consequences of a more sudden turn on of the field than occurs with $a=5$ are that the peaks are less sharply defined and there is a high-frequency continuum component in the spectrum. Effects associated with field turn on may also be avoided in our model by introducing small damping terms in Eqs. (1).

Figure 2, for instance, shows the log spectrum obtained with $\Omega=4.0$ and $\omega/\omega_0=0.25$. This figure illustrates that, for field strengths at which perturbation theory breaks down, successive harmonic peaks at intermediate orders do not fall off sharply but rather display a plateau structure. However, the falloff is quite sharp at the highest orders. This is qualitatively the same kind of behavior that has been observed experimentally and in computations of the spectrum based on space-time integration of the Schrödinger equation for one- and three-dimensional models.[3,4] In fact, results quite similar to those obtained by integration of the Schrödinger equation may be obtained with our grossly simplified model, as can be seen by comparing Fig. 2 with Fig. 1 of the paper by Kulander and Shore.[3]

In our simple model the cutoff in the harmonic generation spectrum is linearly proportional to the ratio of the Rabi frequency Ω to the external field frequency; such a linear dependence of the cutoff on the field strength is, in fact, found experimentally.[6] In the present context of the Bloch equations, this is easily understood: In a strong field the dipole moment induced in a two-level atom is amplitude modulated at the Rabi frequency Ω, so that Ω determines the envelope of the oscillations induced by the field of frequency ω. The quantity Ω/ω is therefore the ratio of the modulation frequency to the driving frequency, and gives the number of harmonics of ω over which the atom can substantially respond to the field.

It is also interesting to note that other, more quantitative features seen in the space-time integration approach are also reproduced by the simple two-level model. For example, Kulander and Shore report that on considering the intensity dependence of the harmonic generation spectra, the higher-order peaks all exhibit the same *effective order of nonlinearity,* a simple measure of power-law scaling with intensity. The intensity dependence obtained from our two-level model is shown in Fig. 3, and it is also clear that in our model the higher harmonics all have approximately the same intensity dependence.

We now turn to the question of the relevance of any single-atom model or theory to the experiments. Consider the formal expression for the expectation value of the number of photons in mode $(\mathbf{k},\lambda)$ at time t:[7]

$$\langle a^\dagger_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle = \langle a^\dagger_{\mathbf{k}\lambda}(0)a_{\mathbf{k}\lambda}(0)\rangle + 2C_{\mathbf{k}\lambda}\mathrm{Re}\int_0^t dt'\langle x(t')a_{\mathbf{k}\lambda}(0)\rangle e^{-i\omega_k t'} + C^2_{\mathbf{k}\lambda}\int_0^t dt'\int_0^t dt''\langle x(t'')x(t')\rangle e^{i\omega_k(t'-t'')}, \tag{3}$$

where $C_{\mathbf{k}\lambda}$ is a coupling constant and x is the component of the electron coordinate along the polarization of mode $(\mathbf{k},\lambda)$. The first term, $\langle a^\dagger_{\mathbf{k}\lambda}(0)a_{\mathbf{k}\lambda}(0)\rangle$, is simply the number of photons in mode $(\mathbf{k},\lambda)$ at time $t=0$. The second term corresponds to absorption from, or stimulated emission into, mode $(\mathbf{k},\lambda)$. If mode $(\mathbf{k},\lambda)$ is initially described by a coherent state $|a_{\mathbf{k}\lambda}\rangle$, so that $a_{\mathbf{k}\lambda}|a_{\mathbf{k}\lambda}\rangle = a_{\mathbf{k}\lambda}|a_{\mathbf{k}\lambda}\rangle$, then

$$2C_{\mathbf{k}\lambda}\mathrm{Re}\int_0^t dt'\langle x(t')a_{\mathbf{k}\lambda}(0)\rangle e^{-i\omega_k t'} \equiv \langle a^\dagger_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle_{\mathrm{abs}} = 2C_{\mathbf{k}\lambda}\mathrm{Re}\left[a_{\mathbf{k}}\int_0^t dt'\langle x(t')\rangle e^{-i\omega_k t'}\right]. \tag{4}$$

The dipole expectation value $\langle e\mathbf{r}(t)\rangle$ thus gives the absorption (or stimulated emission) spectrum.

The last term in (3) is the contribution to the photon number expectation value of mode $(\mathbf{k},\lambda)$ from scattering and spontaneous emission. If the mode is initially unexcited, this term is the only one contributing to $\langle a^{\dagger}_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle$:

$$\langle a^{\dagger}_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle = C_k^2\int_0^t dt'\int_0^t dt''\langle x(t'')x(t')\rangle e^{i\omega_k(t'-t'')}. \tag{5}$$

Thus, whereas the absorption spectrum is determined by the dipole moment expectation value, the spectrum of scattered light is determined by the dipole correlation function. If we make the replacement $\langle x(t'')x(t')\rangle \rightarrow \langle x(t'')\rangle\langle x(t')\rangle$, which of course is in general unjustified, then (5) becomes

$$\langle a^{\dagger}_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle \rightarrow \langle a^{\dagger}_{\mathbf{k}\lambda}(t)\rangle\langle a_{\mathbf{k}\lambda}(t)\rangle = C_k^2\left|\int_0^t dt'\langle x(t')\rangle e^{i\omega_k t'}\right|^2. \tag{6}$$

This decorrelation *Ansatz* is implicit in the computational approaches of Kulander and Shore,[3] Eberly and co-workers,[4,8,9] and Potvliege and Shakeshaft.[10]

These approaches to harmonic generation rely on the assumption that a single-atom theory is relevant to the experiments. Although it is well known that serious errors can result from the use of the dipole expectation value instead of the correlation function in the computation of scattered light spectra,[7] the single-atom theories based on power spectra of dipole expectation values may in fact be applicable to the experiments under consideration. Consider the scattering of light by N atoms, in which case (5) is replaced by

$$\langle a^{\dagger}_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle = C_k^2\sum_{i=1}^{N}\sum_{j=1}^{N}\int_0^t dt'\int_0^t dt''\langle x_i(t'')x_j(t')\rangle \times e^{i\omega_k(t'-t'')}. \tag{7}$$

If the different atomic dipoles are uncorrelated, so that $\langle x_i(t'')x_j(t')\rangle = \langle x_i(t'')\rangle\langle x_j(t')\rangle$ for $i\neq j$, and if $N\gg 1$, then it follows from (7) that

$$\langle a^{\dagger}_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle \cong C_k^2\left|\sum_{i=1}^{N}\int_0^t dt'\langle x_i(t')\rangle e^{i\omega_k t'}\right|^2 = C_k^2\left|\sum_{i=1}^{N}\int_0^t dt'\langle x(t')\rangle e^{i\omega_k t'}\right|^2 \tag{8}$$

if all the atoms are assumed to see the same field. In other words, the single-atom theories based solely on the dipole expectation value are applicable under the assumption that there are no interatomic correlations.

Regardless of whether the dipole expectation value or the correlation function is employed, there is another important consideration involved in the comparison with experimental spectra. Consider the expectation value of the power radiated (scattered) by a single atomic dipole. As is well known, this expression has the same form as the classical Larmor formula:[11]

$$\frac{d}{dt}\sum_{\mathbf{k},\lambda}\hbar\omega_k\langle a^{\dagger}_{\mathbf{k}\lambda}(t)a_{\mathbf{k}\lambda}(t)\rangle = (2e^2/3c^3)\left\langle\left|\frac{d^2\mathbf{r}(t)}{dt^2}\right|^2\right\rangle. \tag{9}$$

Because of the appearance of the square of the second derivative of $\mathbf{r}(t)$ in this expression, each frequency component ω in the spectrum of the scattered light is therefore weighted by $(\omega^2)^2=\omega^4$, exactly as in Rayleigh scattering. The rate of emission of photons of energy $\hbar\omega$ therefore has a factor ω^3. This factor also follows from (5), (6), or (7), since C_k^2 is proportional to ω_k and the summation over modes brings in another factor ω_k^2, as is well known. This factor, which has been included by Potvliege and Shakeshaft,[10] but not by Kulander and Shore[3] and Eberly and co-workers,[4,8,9] obviously has a large effect on the peak height ratios predicted for high-order harmonic generation. An accurate assessment of such ω-dependent prefactors demands, of course, that attention be paid to field propagation and phase-matching effects. Work in this direction is in progress.

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[1]M. Ferray, A. L'Huillier, X. F. Li, L. A. Lompré, G. Mainfray, and C. Manus, J. Phys. B **21**, L31 (1988).

[2]C. K. Rhodes, Phys. Scr. **T17**, 193 (1987).

[3]K. C. Kulander and B. W. Shore, Phys. Rev. Lett. **62**, 524 (1989).

[4]J. H. Eberly, Q. Su, and J. Javanainen, Phys. Rev. Lett. **62**, 881 (1989).

[5]L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

[6]C. K. Rhodes (unpublished).

[7]P. L. Knight and P. W. Milonni, Phys. Rep. **66**, 21 (1980).

[8]J. H. Eberly, Q. Su, J. Javanainen, K. C. Kulander, B. W. Shore, and L. Roso-Franco, J. Mod. Opt. **36**, 829 (1989).

[9]J. H. Eberly, Q. Su, and J. Javanainen, J. Opt. Soc. Am. B (to be published).

[10]R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **40**, 3061 (1989).

[11]See, for instance, B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990), Vol. 1, p. 667.