

Nonperturbative Behavior of Higher-Harmonic Production in a Model Atom*

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The emission of very high harmonics of a laser field irradiating a model atom defined by a three-dimensional delta function is considered and several nonperturbative features of the harmonic spectrum are discussed.

The interaction of atoms and ions with intense laser fields has become a subject of growing interest. In particular, two phenomena have received a great deal of attention recently [1]. One is the process dubbed above-threshold ionization (ATI), where the atom absorbs more photons than required for ionization. This is revealed by the energy spectra of the emitted electrons which consist of a sequence of equally spaced peaks separated by the energy of a laser photon. With increasing intensity, the strongest peak corresponds to the absorption of many more photons than the minimum number required for ionization. The other effect concerns the production of surprisingly high harmonics in an atomic vapor irradiated by an intense laser beam. Up to the 53rd harmonic has been observed [2] in neon. A most unexpected feature is the existence of a kind of plateau in the emitted harmonic intensities, cp. Figure 1. These effects have raised so much interest because they cannot be explained by lowest-order perturbation theory (LOPT) with respect to the coupling between the laser field and the electron. Apparently, neither the existence of this plateau nor the fact that in ATI the lowest peak is not the dominant one, are compatible with LOPT. The fact that LOPT, which had been the backbone of the theoretical description of multiphoton ionization phenomena previously, is no longer applicable, is a challenge to theory: in the simplest case one has to deal with one electron interacting with both the binding potential and the laser

field such that neither can be treated as weak compared with the other.

Even though these phenomena immediately suggest an interpretation in terms of emission and absorption of photons, the theoretical description relies on a classical field, which is an excellent approximation for the high intensities involved. Photons do show up in this semiclassical description since energy/ \hbar is in the presence of the external laser field only conserved modulo an integer multiple of the laser frequency, and each power of the interaction Hamiltonian $-e\mathbf{r} \cdot \mathbf{E}(t)$ (or $e\mathbf{p} \cdot \mathbf{A}(t)$) changes the energy by $\pm \hbar\omega$. This is in close analogy with the semiclassical description of photoelectronic detection by Mandel, Sudarshan, and Wolf [3].

In this paper we concentrate on the emission of higher harmonics of the incident laser frequency by single atoms. Moreover, we model the atom by a three-dimensional delta-function potential which has just one bound state and a largely structureless continuum. In spite of the simplicity of this bare-bones atom, the harmonic intensities will show a surprisingly involved structure.

Owing to the low efficiency of higher-harmonic emission (HHE), experiments have to be carried out at quite high pressure (about 10 Torr). Because emission is found to be strongly peaked in the forward direction and proportional to the square of the pressure, it is clear that the observed response is strongly affected by collective effects. Their description therefore poses another high-intensity problem in addition to the emission by the single atom. However, it has been shown [4] that at higher intensities the collective response is quite close to the response of the single atom, in particular a plateau in the single-atom spectrum tends to be preserved in the collective spectrum.

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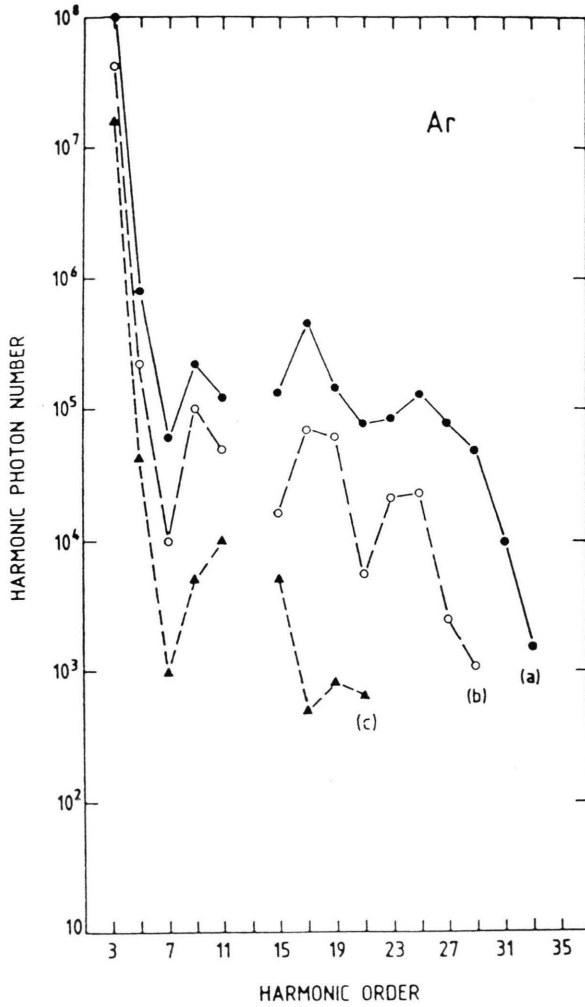


Fig. 1. Measured harmonic-intensity distribution in argon at 15 Torr for several laser intensities: (a) $3 \times 10^{13} \text{ W/cm}^2$, (b) $2.2 \times 10^{13} \text{ W/cm}^2$, (c) $1.6 \times 10^{13} \text{ W/cm}^2$ (from Li *et al.* [7]).

Our model potential is

$$V(r) = \frac{2\pi}{\kappa m} \delta(r) \frac{\partial}{\partial r} r, \quad (1)$$

where the regularization operator $(\partial/\partial r)r$ acts on the following wave function. It supports one bound state with energy $\kappa^2/2m$ and wave function $\exp(-\kappa r)/r$. The quasi-energy wave function $\Psi_0(r, t)$ of the bound state in the presence of an external laser field with arbitrary polarization has been obtained [5] and can be used to calculate HHE [6].

The intensity of HHE with frequency Ω and polarization ϵ is proportional to the square of the matrix

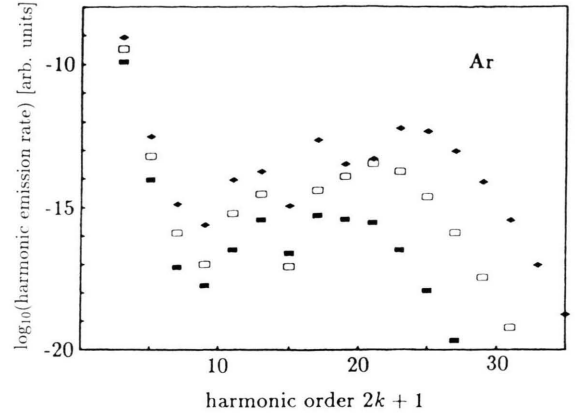


Fig. 2. Logarithm of the harmonic intensities emitted by the model argon atom for the same laser intensities as in Fig. 1.

element

$$M = (2\pi\Omega)^{1/2} \int d^3r dt e^{i\Omega t} \Psi_0(r, t)^* e r \cdot \epsilon \Psi_0(r, t). \quad (2)$$

This integral can be reduced to a one-dimensional quadrature which has to be carried out numerically.

The model atom defined by the potential (1) depends on just one parameter, viz. κ , which is adjusted to the ionization energy of the atom under consideration. Figure 2 displays the harmonic intensities calculated from (2) for one single model argon atom. These results can be confronted with the experimental results obtained in argon at 15 Torr. Data as well as calculations are for a linearly polarized laser field. Due to inversion symmetry only odd harmonics are produced. The calculations reproduce the general features of the data including the rapid initial drop-off of the harmonic intensities followed by a dip which goes over into a ragged plateau. The plateau has a fairly well defined rim, from where the harmonic intensities drop by about one order of magnitude per harmonic. It is remarkable that the data show, for reasons not understood, no emission of the 13th harmonic, while the calculations yield a strong suppression of the 15th harmonic. This is further explored in Figure 3. The parameter η on the abscissa is the ratio of the ponderomotive energy over the energy of one laser photon, which is proportional to the laser intensity. Figure 3 demonstrates that the suppression persists within an intensity range large enough for the effect to show up in experiments which invariably involve an intensity distribution. Figure 4 concentrates on the pronounced structure of a particular harmonic inten-

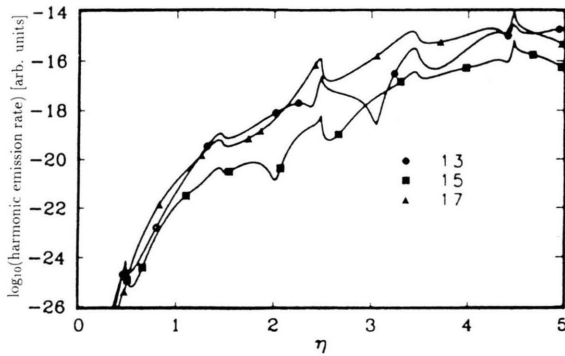


Fig. 3. Logarithm of the 13th, 15th, and 17th harmonic intensities for the model argon atom as a function of the laser intensity.

sity versus laser intensity. The prominent spikes correspond to intensities at which ATI channels close. This comes about as follows: for high laser intensities the bound electron, in order to become free, has to acquire not only the ionization energy $|E_0|$ but also the wiggle energy of a free electron in a laser field, viz. the ponderomotive energy U_p which is proportional to the laser intensity. Hence, whenever with increasing intensity the quantity $(|E_0| + U_p)/(\hbar\omega)$ goes through an integer N , the electron must absorb one more electron in order to be ionized (that is, $N + 1$ in place of N). This channel closing is related to the sharp spikes in HHE. The close relationship can be understood by noticing that (2) essentially calculates a polarization

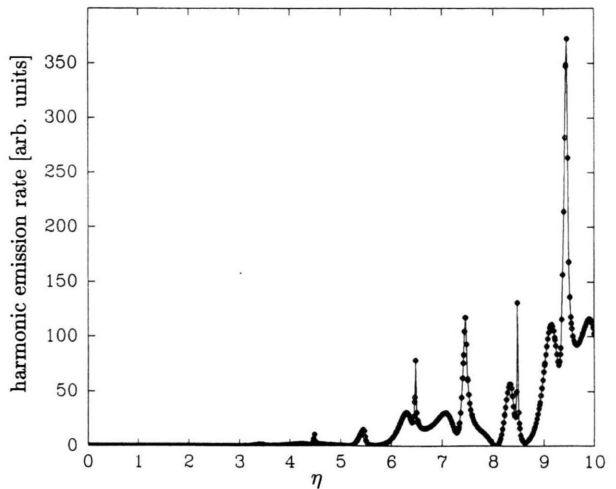


Fig. 4. Intensity of the 17th harmonic for the model argon atom as a function of the laser intensity.

whose real and imaginary parts are related via a dispersion relation.

In summary, the simple delta-function model atom interacting with a classical laser field exhibits a surprisingly rich structure. While quantitative agreement with the real world should not be expected, the qualitative features derived from this model can be expected to show up in real atoms as well.

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