Nonlinear Light Scattering Accompanying Multiphoton Ionization

J. H. Eberly and Q. Su
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

J. Javanainen
Department of Physics and Astronomy, University of Connecticut, Storrs, Connecticut 06268
(Received 27 July 1988; revised manuscript received 11 October 1988)

We confirm in computer experiments the presence of unexpected plateau and cutoff features in high-order harmonic generation accompanying multiphoton ionization. We show the connection between these features and above-threshold ionization.

PACS numbers: 42.50.Hz, 32.80.Rm

The quantum-mechanical theory of nonlinear susceptibilities is well known[1] and is based on perturbation theory. However, it has been shown that light intensities above about 10^{12} W/cm^2 would be sufficient to call into question the validity of both perturbation theory and all conventional expressions for polarizability and susceptibility.[2,3] These are the same high intensities associated with above-threshold ionization (ATI).[4]

Now two series of experiments[5,6] have actually observed nonlinear optical processes in atomic gases with high-power lasers operating above this value of intensity. Several unexpected phenomena have been reported. These include the following: (1) a "plateau" of high-order harmonics (up to the 33rd harmonic has been reported) in which successive harmonics are approximately equal in strength or falling off slowly; (2) a relatively abrupt "cutoff" above a certain harmonic order; and (3) a continuous background of light scattered at all wavelengths between the high-order harmonics.

A theoretical interpretation of these phenomena has not appeared, and in view of the real uncertainties inherent in such difficult experiments it may be questioned as to what extent the reported results are fundamental atomic effects. For example, the gas pressures used were not extremely low, and one may ask whether many atom or propagation or plasma processes played a role.[7] In this Letter we report computer experiments that contribute theoretical evidence that the effects observed are indeed fundamental single-atom effects, and our results suggest that they can be expected in essentially all atomic vapors.

In addition, we suggest, on the basis of a nonperturbative analysis of the atomic wave function, that at least two of the unexpected features found in the light scattering spectra are intimately related to features of the corresponding ATI photoelectron spectra, which have been extensively studied in different laboratory experiments.[4] Our computer experiments allow the first comparison of corresponding ATI and high-order harmonic spectra *taken with the same laser pulse,* and we believe that they corroborate our suggestion.

Now we describe our computer light scattering experiments. As mentioned above, it is doubtful whether any susceptibility of finite order can be qualitatively reliable, let alone numerically accurate, in the range of laser intensities (10^{13}–10^{16} W/cm^2) of the recent laboratory light scattering experiments.[5,6] Thus, we do not attempt to compute a susceptibility, but compute instead a more fundamental quantity, the ionizing atom's time-dependent dipole moment. This is easily obtained directly from the atomic wave functions: $D(t) = \langle \Psi(t) | ex | \Psi(t) \rangle$. Unfortunately, exact wave functions $|\Psi(t)\rangle$ do not exist for any atom, even for atomic hydrogen, in the presence of a strong laser field $\epsilon_0 \sin\omega t$. However, by solving Schrödinger's partial differential equation (in atomic units),

$$[-\tfrac{1}{2}\partial^2/\partial x^2 + V(x) - x\epsilon_0(t)\sin\omega t]\Psi(x,t) = i\,\partial\Psi(x,t)/\partial t\,, \quad (1)$$

we can compute *ab initio* wave functions numerically with good accuracy.[8] We include a density-of-time points not only sufficient for convergence of the wave function calculation but also sufficient for subsequent Fourier resolution of the highest harmonic frequency to be analyzed. Furthermore, since the recent light scattering experiments[5,6] showed similar results for a variety of atoms, we suppose that, for the moment at least, we are not compelled to introduce the many details that distinguish real atoms from each other and so choose a simple "representative" $V(x)$ for the binding potential.[8] In Fig. 1 we show the lowest bound levels of our model atom.

The coherently scattered light power at a given frequency ω is proportional to the square of $D(\omega)$, the Fourier transform of the dipole expectation value $D(t)$. Thus we can take $|D(\omega)|^2$ to represent the coherent photon spectra. We report results for the photon spectra obtained in ten-photon ionization experiments. We have chosen two square laser pulses of 16.25 cycles duration and field strengths of 0.04 and 0.05 a.u. (intensity between 10^{13} and 10^{14} W/cm^2).

In Fig. 2 we show the computed photon spectra in

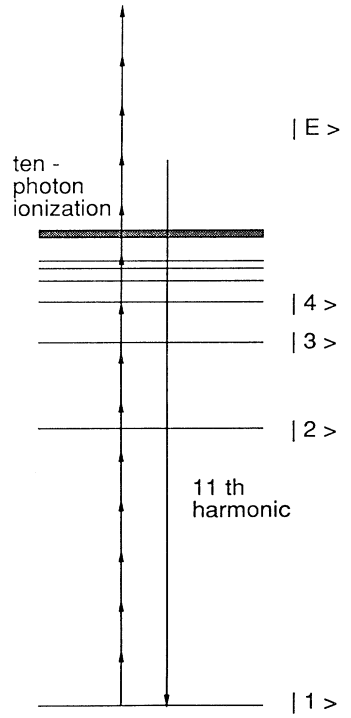

FIG. 1. Upward arrows indicate laser-photon absorption, and the downward arrow indicates eleventh harmonic generation, superposed on a diagram showing only the lowest-energy levels of our model atom, approximately to scale.

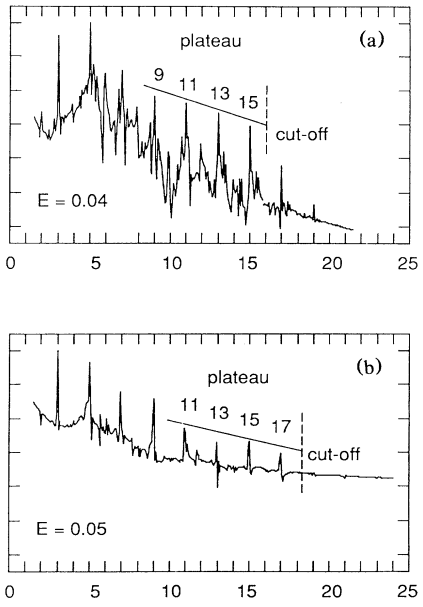

FIG. 2. Light scattering spectra computed for a square 16.25-cycle laser pulse. The horizontal axis unit is laser-photon energy, and several peaks are labeled by their harmonic number. The peak electric field strength (in atomic units) is (a) 0.04 and (b) 0.05. The locations of "plateaus" and cutoffs are indicated. The vertical axes are logarithmic and the curves in (a) and (b) are independently normalized. The third harmonic peak is 7 times stronger in (b).

these cases. *Both spectra contain the unexpected features of the recent data from laboratory experiments*[5,6] mentioned above: (1) a "plateau" of several approximately equal-strength high-order harmonic peaks, (2) a relatively abrupt high-harmonic cutoff after the plateau, (3) a low-level continuous background of light frequencies between the harmonics.

Since our computer results follow from the purely one-electron theory of Eq. (1), they cannot possibly be attributed to collective or propagation or plasma processes. Thus the appearance of all three unexplained features of the experimental data[5,6] provides independent evidence that the laboratory experiments may have observed new fundamental single-atom phenomena. Note that the extreme simplicity of our "representative" atomic model is an advantage rather than a deficiency in drawing conclusions of this kind.

Another advantage (not exploited up to now) of numerical experiments with *ab initio* wave functions is that the same electron and its wave function can be analyzed in several ways. In Fig. 3 we show the ATI photoelectron spectra that would be obtained *from the same laser pulses* as produced the photon spectra in Fig. 2. This is the first report of a comparison of ATI spectra and photon spectra under identical conditions.

Several of the photoelectron peaks in Figs. 3(a) and 3(b) are labeled according to the number of photons needed to create them. Note that in Fig. 3(a) the ten-photon peak has already disappeared and, between Figs. 3(a) and 3(b) the eleven-photon peak also shifts below threshold. These shifts agree with the ac Stark shift or so-called ponderomotive threshold shift: $\Delta E = e^2\epsilon_0^2/4m\omega^2$, as expected.

More interesting is the envelope of peak heights. We observe that the photon number at which the ATI peak envelope cuts off is nearly or exactly the same as the harmonic number of the cutoff of the corresponding photon spectrum. Furthermore, the photon numbers of the harmonic peaks in the "plateau" regions correspond well with the photon numbers of the major ATI peaks.[9] Both ATI and harmonic spectra have continuous backgrounds. In other words, our data show that the main features of the harmonic spectra that are new to nonlinear optics all appear to be counterparts of features of ATI spectra that have been recorded[4] in the last five years.

Now let us see how to explain this situation without the benefit of computer analysis. We consider the atomic-state vector, expanded in the bare ($\epsilon_0 = 0$) basis: $|\Psi(t)\rangle = \sum_m a_m(t)\exp(-iE_m t)|m\rangle$, where of course the sum is interpreted as an integral when E_m is in the con-

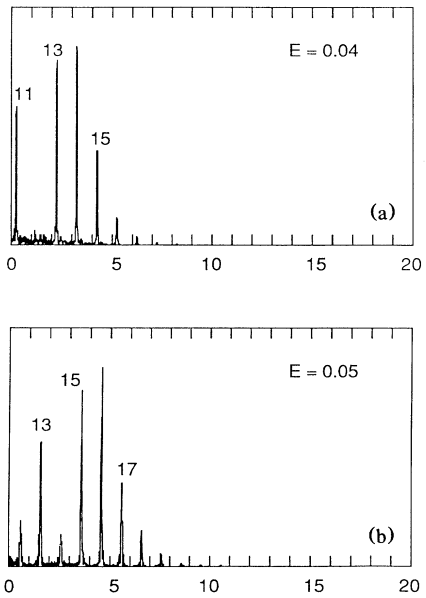

FIG. 3. Computer photoelectron (ATI) spectra obtained from the same laser pulses used in Fig. 2. The horizontal axis is electron kinetic energy in units of photon energy. Various peaks are labeled according to the number of photons needed to produce them by excitation from the ground state. The vertical axes are linear, and the fifteen photon peak is 4 times stronger in (b).

tinuum. The atomic dipole moment is given by

$$D(t)=\sum_{k,m} x_{km}a_k^* a_m \exp[i(E_k-E_m)t]\,.$$

We can assume that the ground-state amplitude remains much larger than any continuum-state amplitude. Thus, at least for the continuum contributions, the dipole moment is well approximated by

$$D(t)\approx\sum_m x_{0m}a_m^* a_0 \exp(iE_m t)+\text{c.c.}$$

This clearly shows that, to a good approximation, the amplitude of a high qth harmonic is proportional to $a_{q\omega}^* a_0$ (where $E_m=q\omega$, and the zero energy is at the ground state: $E_0=0$), and the scattered power at the qth harmonic is proportional to $|a_0|^2|a_{q\omega}|^2$.

Note that, in addition to the energy phases $\exp[-iE_m t]$, the amplitudes a_m could themselves vary rapidly enough to obfuscate the origin of the Fourier peaks. This occurs if the electron populations $|a_m|^2$ vary on an optical-frequency scale, implying an ionization rate of the atom more rapid than ω. The qth harmonic peak would be affected if the ionization rate were on the order of $q\omega$. At the intensities under consideration here, our atom ionizes at a rate less than ω and thus much less than 13ω or 19ω, so we can safely ignore the possibility of high frequencies in the a_m's to a good approximation.

Using the same state vector $|\Psi(t)\rangle$, we now compute the ATI probability density $P(E_m)=|\langle E_m|\Psi\rangle|^2$ for the corresponding photoelectron peak $E_m=q\omega$, the peak associated with the absorption of q photons from the ground state. This probability is trivially $P(q\omega)=|a_{q\omega}|^2$.

This derivation is fully nonperturbative and does not employ the rotating-wave approximation. There is no assumption that $|a_{q\omega}|^2$ is proportional to the qth power of the intensity, or that $a_0\approx 1$. The result shows that precisely the same amplitude $a_{q\omega}$ is responsible for both a high-order harmonic peak and the corresponding ATI peak. This means that at least the qualitative characteristics of the ATI peak envelope should be reproduced in the high-order harmonics.[3] That is, ATI peaks of comparable strength should produce harmonic peaks of comparable strength, and the locations of cutoffs should be approximately the same in ATI as in harmonic spectra. Also, there should be no qth harmonic if there is no q-photon ATI peak but not *vice versa*. Since $a_{q\omega}$ is nonzero for all q, there are ATI peaks for all integers q, but the dipole matrix element x_{0m} obeys dipole selection rules, so $D(t)$ and the photon spectra will have strong contributions only from odd-valued harmonics. We believe that an inspection of the spectra presented in Figs. 2 and 3 supports these conclusions.[10]

Of course, more data are needed to test these conclusions more widely, and establish their limits. Almost certainly they will not hold well for low-order harmonics, which can be generated by bound-bound as well as bound-free transitions. However, we have already found confirming results in another, quite different, regime of ATI and harmonic generation, in connection with two-photon ionization, and also using longer laser pulses and pulses that are smoothly turned on and off. These findings will be presented elsewhere.

In summary, we have presented the results of multiphoton ionization computer experiments with a model atom. We believe our atom is realistic enough to allow these following conclusions: (1) the recent observations of slow harmonic decrease,[5,6] and more particularly the cutoff plateau behavior[6] will be found to be true of any atomic species, not only noble gases; and (2) high-order harmonic generation is necessarily associated with ATI production in a specific way. In addition, our observation of plateau and cutoff features provides independent evidence that (despite the nonnegligible gas pressures employed) the real experiments were observing fundamental single atom, not many atom or plasma effects in light scattering.

We thank S. L. Chin, L. A. Lompre, and G. Mainfray for discussions, and the Allied-Signal Corporation and the John Von Neumann Center for support of computing resources. Since completing this work, we have learned that a near replication of the harmonic peak structure in xenon of Ref. 6 has been obtained from a computer ex-

periment by K. Kulander and B. W. Shore (private communication).

---

[1]Y. R. Shen, *Principles of Nonlinear Optics* (Wiley, New York, 1984).

[2]S. E. Harris, Phys. Rev. Lett. **31**, 341 (1973); Y. Gontier, M. Poirier, and M. Trahin, J. Phys. B **13**, 1381 (1980); M. Aymar and M. Crance, *ibid.* **14**, 3585 (1981); L. Pan, K. T. Taylor, and C. W. Clark, Phys. Rev. Lett. **61**, 2673 (1988).

[3]A formalism for nonperturbative light-scattering theory has been given by B. W. Shore and P. L. Knight, J. Phys. B **20**, 413 (1987).

[4]Recent reports are given, for example, by U. Johann, T. S. Luk, H. Egger, and C. K. Rhodes, Phys. Rev. A **34**, 1084 (1986); G. Petite, P. Agostini, and F. Yergeau, J. Opt. Soc. Am. B **4**, 765 (1987); P. H. Bucksbaum, M. Bashkansky, R. R. Freeman, T. J. McIlrath, and L. F. DiMauro, Phys. Rev. Lett. **56**, 2590 (1986).

[5]C. K. Rhodes, Phys. Scr. **T17**, 193 (1987); A. McPherson, G. Gibson, H. Jara, U. Johann, I. A. McIntyre, K. Boyer, and C. K. Rhodes, J. Opt. Soc. Am. B **4**, 595 (1987).

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

[7]Systematic studies of pressure dependence have so far been made only for low harmonics: A. L'Huillier, L. A. Lompre, M. Ferray, X. F. Li, G. Mainfray, and C. Manus, Europhys. Lett. **5**, 601 (1988); R. Rosman, G. Gibson, K. Boyer, H. Jara, T. S. Luk, I. A. McIntyre, A. McPherson, J. C. Solem, and C. K. Rhodes, J. Opt. Soc. Am. B **5**, 1237 (1988).

[8]We have discussed the details of our numerical methods, and the nature of the representative atom for which we compute $\Psi(x,t)$, in J. Javanainen, Q. Su, and J. H. Eberly, Phys. Rev. A **38**, 3430 (1988). The bound states contain a Rydberg sequence, the positive-energy states are Coulombic, and parity is a good quantum number. The ground state is at -0.67 a.u., and a few other low-lying levels are shown in Fig. 1.

[9]In each of our spectra in Fig. 2, the lowest peak in the plateau region corresponds to an electron peak near to but below the threshold (and thus not shown in Fig. 3). The high-lying Rydberg states are not resolved and behave exactly like part of the electron continuum during our short pulses.

[10]Phase matching has been ignored in our treatment because we are treating single-atom scattering. In addition, phase matching usually takes effect through a power-law factor $(f_{\mathrm{PM}})^q$. This appears to be true at the intensities of interest as well as all lower intensities, according to preliminary calculations at Livermore and Saclay [A. L'Huillier, K. Kulander, and B. W. Shore (private communication)]. In this case, phase matching will produce exactly the same effect, namely, a drop in harmonic intensity by the factor $(f_{\mathrm{PM}})^2$, between the eleventh and thirteenth harmonics as between the 99th and 101st. That is, it cannot be responsible for any relatively abrupt "cutoff" at a particular harmonic number.