Loss of harmonic generation in intense laser fields

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Harmonic generation and above-threshold ionization are both consequences of the nonlinear response of atoms to intense laser fields. In this paper, we examine what happens to the single-atom harmonic spectrum at high intensities, when multiphoton absorption ceases to be the dominant process of ionization. At these intensities, ionization can occur by the electron wave packet passing directly over the resultant barrier of the atomic potential and the incident electric field. In consequence of this, the harmonics produced are of a lower strength and can be swamped by the production of a large background to the harmonic spectrum. This background is caused by the ionized electron rescattering from the nucleus in the presence of the laser field.

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I. INTRODUCTION

It has long been recognized that correlations should exist between the photoelectron and harmonic spectra that are observable when an atom is exposed to an intense electric field [1]. For example, one would expect the highest-order harmonic generated in a particular pulse to be related to the number of photons that the ionizing electron absorbs when it forms the higher-order above-threshold ionization (ATI) peaks.

The mechanism by which ATI and harmonic spectra are formed is generally well understood at intensities around 10^{14} W/cm^2 or lower [2] (in spite of the fact that any theoretical analysis cannot rely on perturbation theory). It is the aim of this paper to examine the behavior of an ionizing atom when it is exposed to fields of even higher intensity, where direct multiphoton excitation and ionization ceases to be the dominant ionization process. At these high intensities, the atom ionizes as the electron passes over the resultant potential barrier of the atomic nucleus and the incident electric field.

We use a numerical method of obtaining the solutions of a one-dimensional Schrödinger equation with a model potential representing the atomic nucleus, and use it to study the response of an atom to pulses of various intensities. The numerical method is outlined in Sec. II. In Sec. III we present our results and examine the differences between harmonic and photoelectron spectra produced at different intensities, and hence by the alternative mechanisms of direct multiphoton absorption and over-the-barrier ionization. In addition to examining these spectra, we also plot both the ionization probabilities and electron accelerations as a function of time. Comparing these plots for different intensities, we are able to draw conclusions about the physical processes that are occurring in the pulse. We also briefly compare our results with those produced by calculations using classical atoms. In Sec. IV we discuss the relationship between our calculated single-atom spectra and the multiatom spectra that could be obtained experimentally.

II. THE NUMERICAL CALCULATION

Because perturbation theory is not valid in the intensity regime that we are concerned with here, we have employed a numerical method to carry out the calculation. Many numerical models have been used previously to examine multiphoton effects, the models being of widely different complexity [3–5]. We choose to employ a one-dimensional fully-time-dependent method, with the calculation being carried out in the Kramers-Henneberger [6] frame, which has the advantage of being fast to compute for a very wide range of parameters. Details of the calculation have been presented elsewhere [5]. However, we present below a brief summary of the technique for a single-atom model in which the atomic potential is modeled by the frequently used screened potential [4,5,7]

$$V(x)=\frac{-1}{[(1+x^2)]^{1/2}}\ , \tag{2.1}$$

which has a Coulomb tail and hence supports a Rydberg series of bound states. We use a time-dependent electric field given by

$$\mathcal{E}(t)=\mathcal{E}_0\sin(\omega t)\sin^2\left[\frac{\pi t}{T}\right]\ , \tag{2.2}$$

where T is the pulse length and $\mathcal{E}_0$ is the peak electric field. This smooth pulse avoids any "turn-on" transients. The harmonic spectrum $|D(\omega)|^2$ is calculated by taking the Fourier transform of the time-dependent acceleration of the electron calculated at each time step in our calculation [8], such that

$$D(\omega)\propto\int_0^T\ddot{d}(t)e^{-i\omega t}dt\ , \tag{2.3}$$

where $\ddot{d}(t)$ is calculated using Ehrenfest's theorem as

$$\ddot{d}(t)=\left\langle\phi(x,t)\left|\frac{x}{(1+x^2)^{3/2}}-\mathcal{E}(t)\right|\phi(x,t)\right\rangle\ . \tag{2.4}$$

Here $\phi(x,t)$ is the solution of the one-dimensional length-gauge Schrödinger equation

$$i\frac{\partial\phi}{\partial t}(x,t)=-\frac{1}{2}\frac{\partial^2\phi}{\partial x^2}(x,t)+V(x)\phi(x,t)+\mathcal{E}(t)x\phi(x,t)\ . \tag{2.5}$$

We use the length gauge here for ease of calculation and we refer the reader to Ref. [8] for further calculational details. It should be emphasized that this only gives the single-atom response: we will return to this point below.

III. MECHANISMS OF IONIZATION

At lower intensities, the dominant method of ionization is multiphoton excitation to real or virtual bound levels and thence out into the continuum. For long pulses, the photoelectron spectrum consists of well-defined ATI peaks separated by the photon energy. For short pulses, these ATI peaks split up into several subpeaks, many of which can be accounted for in terms of multiphoton resonances between ac Stark-shifted bound states [9,10].

Similarly, when the harmonic spectra produced by these pulses are examined, they consist of peaks, predominantly located at odd multiples of the photon energy, again modified by the occurrence of bound-state resonances [11]. If such resonances do occur, then the particular harmonic that corresponds to the order of the multiphoton resonance can be dramatically enhanced. However, such enhancement can only affect a single harmonic. To increase the magnitude of all the harmonics, one can instead increase the incident laser intensity.

Recent experiments have shown that high-intensity lasers are capable of producing both high-order and relatively high-strength harmonics [12]. However, we explain below that there exists a limit to the increase that one can achieve in the single-atom harmonic intensity simply by increasing the driving-field intensity.

Keldysh pointed out in 1965 that ionization by a process other than straightforward multiphoton absorption was possible [13]. To examine the effect of intensity on the harmonic spectrum, we roughly divide the ionization process into two regimes. In the first regime, applicable at low intensities, ionization occurs by the electron passing through a series of virtual levels in the atom (real levels if a resonance exists) and out into the continuum, where it can continue to absorb photons and form the higher ATI peaks. Harmonics are generated by transitions between these bound levels, or from the continuum to these bound levels [14].

In the second regime, applicable at higher intensities, ionization occurs by the electron passing over the resultant potential of the atomic nucleus and the incident electric field (see Fig. 1). This can only occur when the electric field is greater than some critical field so that the maximum of the resultant potential lies at a lower energy than the binding energy of the atom.

Of course, the division of ionization into these two regimes is arbitrary. The pictures represent two extreme views of the ionization process and, for intermediate intensities, one can view the ionization process as having some contribution from the electron tunneling through the barrier presented by the resultant potential [15], or alternatively ionization can occur when the electron is excited to higher levels and then passes directly over the barrier. However, considering over-the-barrier (OTB) ionization and multiphoton absorption ionization as two distinct processes can explain the relatively abrupt changes in the photoelectron and harmonic spectra that occur as the intensity increases.

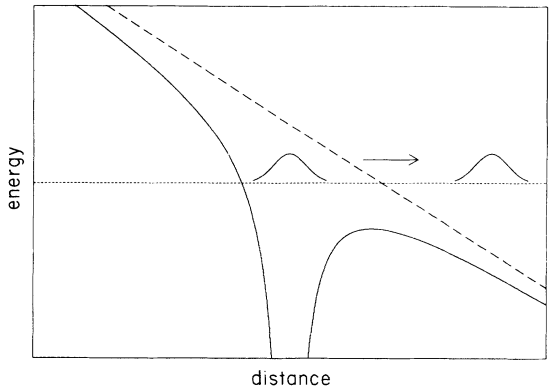

FIG. 1. Schematic diagram of over-the-barrier ionization. The combined effect of the incident electric field (dashed line) and the nuclear potential results in a potential barrier (solid line) that has a peak value on one side that is less than the binding energy of the atom (dotted line). Hence the electron can pass over the barrier and away from the atom.

This picture of over-the-barrier ionization has been used previously with success by Augst *et al.* [16]. By using a simple classical time-independent picture of OTB ionization, they have shown that there is good agreement between the intensities at which OTB ionization is predicted and the experimentally measured intensities.

We follow the method of Augst *et al.* [16] by formulating a simple time-independent classical theory to predict the intensity at which OTB ionization becomes possible. The resultant potential due to our model atom and incident electric field is given by

$$V_r(x)=\frac{-1}{[(1+x^2)]^{1/2}}-\mathcal{E}x\ , \tag{3.1}$$

where the sinusoidal variation of the incident electric field has been neglected. Then the maximum point of the potential is at x_m, where

$$\frac{dV_r}{dx}=\frac{x_m}{(1+x_m^2)^{3/2}}-\mathcal{E}=0\ . \tag{3.2}$$

Therefore we can see that OTB ionization can occur approximately when

$$|E_g|\le\frac{1+2x_m^2}{(1+x_m^2)^{3/2}}\ . \tag{3.3}$$

For our model potential, $|E_g|=0.669\,78$, where E_g is the ground-state energy and hence the critical field is 0.1195 a.u. (5.01×10^{14} W/cm^2). (This is the critical field for our model potential: for atomic hydrogen, the critical

field is 0.0625 a.u.) Using this value of the critical field, we can compare the results of calculations both above and below this critical intensity to see whether the changeover to OTB ionization has a significant effect on the harmonic spectrum.

Figures 2 and 3 show various plots for two intensities: the lower intensity, 2.0×10^{14} W/cm^2, is below the critical intensity and the other, 3.2×10^{15} W/cm^2, is above. All the plots are for an incident pulse of wavelength 300 nm and pulse length of 50 fs.

Figures 2(a) and 3(a) show the effect that increasing the peak intensity has on the harmonic spectrum: clearly there is less harmonic generation at the higher intensity, with the harmonic strength being reduced by an order of magnitude. Examining the photoelectron spectra for these two intensities [Figs. 2(b) and 3(b)], one can see that the ATI peaks cease to be distinguishable at the higher intensity, showing a close correlation between the photoelectron and harmonic spectra.

This decrease in the harmonic strength can be explained in terms of the ionization mechanism. When an atom ionizes by multiphoton absorption, the ionizing electron passes through a series of virtual bound levels of the atoms (some of these levels may be real if any multiphoton resonances occur). This process is *quasiperiodic* and relatively *slow* because the matrix element of the multiphoton transition is small. Also the electron remains close to the nucleus and so it can absorb further photons, or alternatively emit harmonics. In addition, when the electron is promoted into the continuum, it is *still close to the nucleus* and hence capable of emitting high-order harmonics.

In contrast, ionization by the OTB mechanism is a very rapid process once the critical electric field is reached: the electron wave function passes over the barrier in a fraction of a laser cycle. Once over the potential barrier, the electron accelerates down the potential and away from the nucleus. It is the *rapidity* and *aperiodic* nature of this process that prevents harmonic generation. The wave packet *quickly moves away from the core* and there is very little time for harmonics to be generated.

The response of the electron to the incident electric field during the remaining part of the pulse becomes that of a free electron and the electron scatters from the nucleus. Such scattering events modify the harmonic spectrum by increasing the background. This is significant because this background can be large enough to wash out any harmonics produced on the rising edge of the pulse before the critical field is reached. The level of this background increases dramatically with increasing peak intensity. In increasing the peak intensity from 5×10^{14} W/cm^2 to 5×10^{15} W/cm^2, the background increases by orders of magnitude. It is this scattering background that destroys any regular structure in the photoelectron spectrum.

Further evidence for this fast OTB ionization mechanism being responsible for the lack of harmonics is found in plots of the acceleration. Figure 2(c) is a plot of the acceleration of an electron as a function of time in a pulse with a subcritical electric field, with the $\langle\phi|\mathcal{E}(t)|\phi\rangle$ term in Eq. (2.4) not included, so as to illustrate more

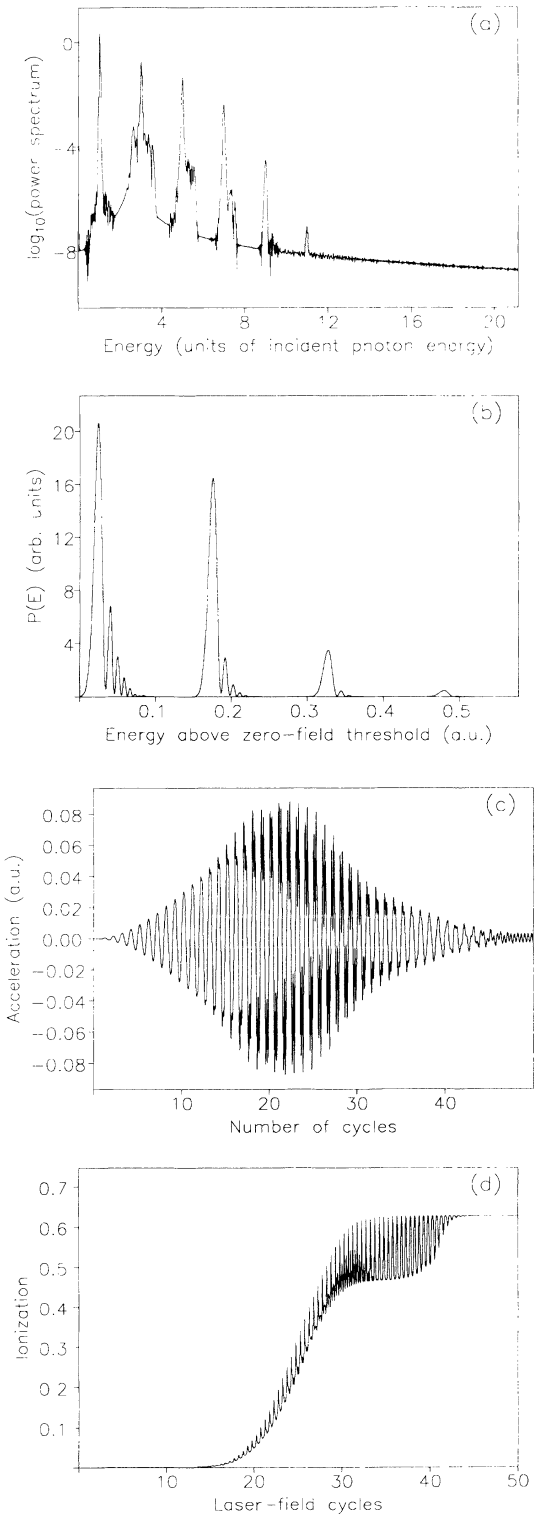

FIG. 2. (a) Plot of the harmonic spectrum: $\log_{10}$ of acceleration $|D(\omega)|^2$ against photon energy in units of the incident photon energy. (b) Photoelectron spectrum: continuum population $P(E)$ as a function of energy E in atomic units. (c) Acceleration as a function of time, with the electric-field term removed. (d) Ionization probability as a function of time. All plots are for a 50 fs pulse of wavelength 300 nm and at an intensity of 2.0×10^{14} W/cm^2.

clearly how the harmonics of the field are produced. One can see that the electron is being accelerated and hence emitting radiation throughout the pulse, reaching a maximum acceleration at the maximum of the pulse.

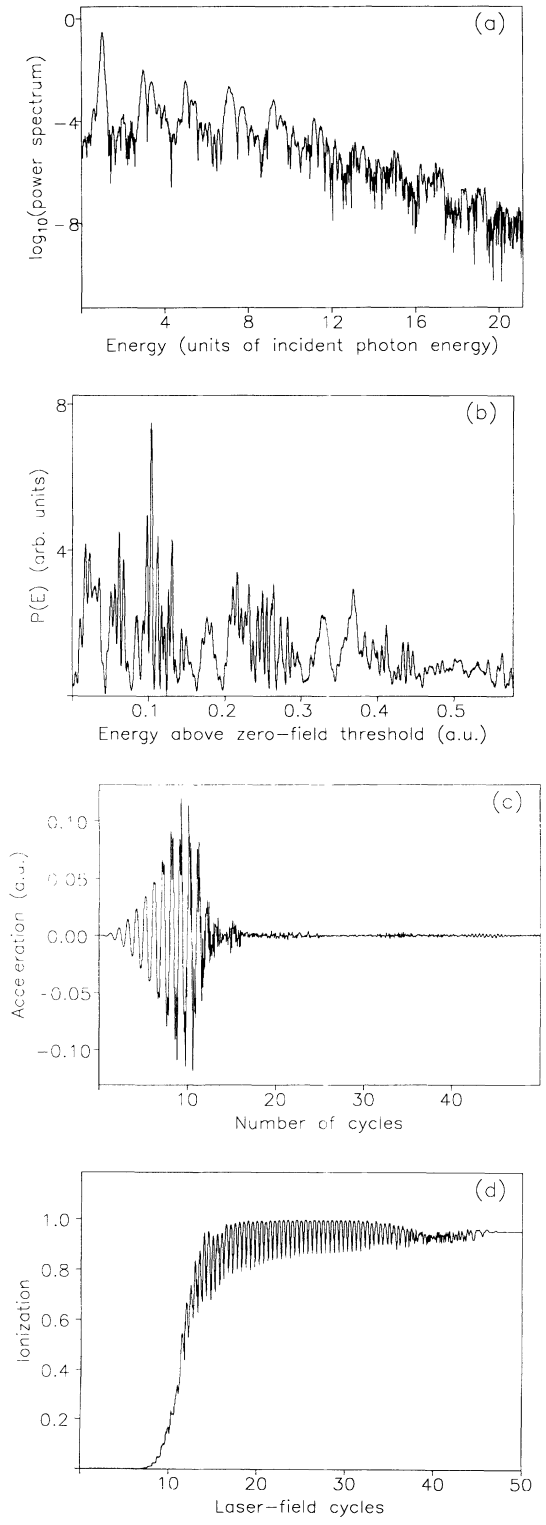

FIG. 3. As in Fig. 2, but for an intensity of 3.2×10^{15} W/cm^2.

By contrast, examining the acceleration of the electron in the high-intensity pulse [Fig. 3(c)], one can see that the acceleration ceases abruptly once the critical field is reached. Once past this critical field, the electron is free and it moves away from the atom. Hence the only acceleration after ionization is due to the electric field at the fundamental frequency and so no harmonics are produced. One can appreciate that it is when the electron is close to the nucleus that harmonics are produced, so that once the critical field is reached, there is little probability of harmonic generation.

One can also examine the ionization probability as a function of time [Figs. 2(d) and 3(d)]. The ionization probability is calculated by taking the sum of the overlaps of the evolved wave function with the field-free bound states of the atoms and subtracting this from unity. At the lower intensity, one can see a steadily increasing ionization probability with a maximum ionization rate at the center of the pulse when the electric field is a maximum. On the other hand, the ionization probability plot of the high-intensity case shows a very rapid increase in ionization once the critical field is reached. The ionization probability leaps up to unity at a time which is comparable to that predicted by our simple theory ($t\approx449$ a.u.).

It is interesting to compare our observations with those obtained by considering classical atoms. Chu and Yin [17] have used the technique developed by Leopold and Percival [18] to examine classical atoms exposed to electric fields greater than the critical field at optical frequencies. Amongst other mechanisms, they identify a direct ionization mechanism that can be considered to be the classical analogue of over-the-barrier ionization. Once initiated, such ionization proceeds within a fraction of a cycle, as is noticed in our quantum-mechanical calculation. Bandarage, Maquet, and Cooper [19] have also identified this direct ionization process. However, these classical results are calculated from the atomic dipole moment, the use of which can introduce a spuriously large background into the harmonic spectrum (Reference [8] discusses this further).

IV. MULTIATOM SPECTRA

In this section we will be discussing the important question as to what relationship our single-atom calculation has to experimentally observable harmonic spectra. The first point to emphasize is that we have, of course, not calculated the full two-time dipole-dipole correlation function. In common with the calculations of many others, in a sense we have only calculated the coherent radiation part of the single-atom spectrum. This is related to the coherent spectrum that could be observed in the forward direction of a multiatom harmonic-generation experiment, as has been discussed by various workers [20]. Eberly and Rzazewski have shown, for example, that the coherent single-atom spectrum plays a dominant part in determining the form of the observed multiatom spectrum in most cases [21].

However, the single-atom spectrum is modified by propagation effects as the emitted radiation passes

through the medium driven by the intense field. Various propagation codes have been developed [22] that allow for the investigation of these propagation effects, but these codes assume that the single-atom spectrum consists of a series of very narrow harmonic peaks. Of course this is far from the case at high-field strengths, as can be seen from Fig. 3(a). In this case, the harmonic peaks are broad and the contrast between the peak heights and the background is significantly reduced. Therefore, it is not straightforward to predict how propagation effects will alter the single-atom high-field harmonic spectrum.

With this in mind we confine ourselves to the conclusion that the individual harmonic strengths are *not increased* by increasing the incident intensity beyond some limiting value. They are, in fact, decreased in height and broadened by the decrease in the time available for harmonic generation and also by the scattering of the ionized electron. This conclusion will hold even if the background is modified by the effects discussed above.

V. CONCLUSION

We have shown that the mechanism of ionization plays a vital role in the generation of a harmonic spectrum. In particular, the change to over-the-barrier ionization at some critical value of incident electric field means that there exists an upper limit to the intensity that one can use to obtain a strong discrete harmonic spectrum.

If the peak pulse intensity never reaches this critical intensity then harmonic generation can be considerable and there is a low background to the peaks. On the other hand, if the peak pulse intensity is above this critical value, then once that critical intensity is reached, complete ionization takes place rapidly. Harmonics can be produced on the rising edge of the pulse, but this production ceases at the critical intensity and for the remaining part of the pulse, the ionized electrons undergo bremsstrahlung scattering events which build up the background of the discrete harmonic spectrum and the peak-background contrast can be drastically reduced. So increasing the intensity of the incident pulse does not necessarily increase the harmonic-production rate, and indeed can even *decrease* the contrast between the harmonic peaks and the background, or wash out the peaks altogether.

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[1] See, for example, J. H. Eberly, Q. Su, and J. Javanainen, Phys. Rev. Lett. **62**, 881 (1989); M. Dörr, R. M. Potvliege, and R. Shakeshaft, J. Opt. Soc. Am. B **7**, 433 (1990).

[2] See, for example, M. Pont, R. M. Potvliege, and R. Shakeshaft, *Proceedings of the 5th International Conference on Multiphoton Processes,* edited by G. Mainfray and P. Agostini (CEA, Paris, 1990), p. 105; K. C. Kulander, K. J. Schafer, and J. L. Krause, *ibid.* p. 119, P. Lambropoulos, *ibid.* p. 133; L. Davidovich, *ibid.* p. 145.

[3] J. Javanainen, J. H. Eberly, and Q. Su, Phys. Rev. A **38**, 3430 (1988).

[4] See, for example, K. C. Kulander, Phys. Rev. A **36**, 2726 (1987); X. Tang, A. Lyras, and P. Lambropoulos, Phys. Rev. Lett. **63**, 3269 (1989); B. Sundaram and L. Armstrong, Jr., Phys. Rév. A **38**, 152 (1988); K. J. LaGattuta, *ibid.* **40**, 683 (1989); L. A. Collins and A. L. Merts, *ibid.* **40**, 647 (1989); M. S. Pinzola, G. J. Bottrell, *ibid.* **40**, 659 (1989); J. K. Liakos and M. Horbatsch, *ibid.* **40**, 685 (1989); P. Krstic and M. H. Mittleman, *ibid.* **40**, 587 (1989).

[5] V. C. Reed and K. Burnett, Phys. Rev. A **42**, 3152 (1990); K. Burnett, P. L. Knight, B. Piraux, and V. C. Reed, Phys. Rev. Lett. **66**, 301 (1991); V. C. Reed and K. Burnett, Phys. Rev. A **43**, 6217 (1991).

[6] H. A. Kramers, *Collected Scientific Papers* (North-Holland, Amsterdam, 1956), p. 272; W. C. Henneberger, Phys. Rev. Lett. **21**, 838 (1968).

[7] X. Tang and S. Basilie (unpublished).

[8] The use of the time-dependent acceleration of the electron instead of the atomic dipole moment to calculate the harmonic spectrum has been explained in a Brief Report by K. Burnett, V. C. Reed, J. Cooper, and P. L. Knight, Phys. Rev. A **45**, 3347 (1992). In this paper it is pointed out that when the dipole moment of an atom is significant, then it is computationally incorrect to multiply simply the spectrum obtained from the dipole moment by the factor ω^4; instead it is necessary to use the acceleration of the ionizing electron. This results in harmonic spectra which have backgrounds that are independent of the size of the computational grid. The distinction between these two methods is particularly important at high-field intensities when the final dipole moment of the atom can be large, where using the dipole moment to calculate the harmonic spectrum can result in a spuriously large background that swamps any structure in the harmonic spectrum.

[9] For experimental results see G. Petite, P. Agostini, and F. Yergeau, J. Opt. Soc. Am. B **4**, 765 (1987); R. R. Freeman, P. H. Bucksbaum, H. Milchberg, S. Darack, D. Schumacher, and M. E. Geusic, Phys. Rev. Lett. **59**, 1092 (1987).

[10] R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **41**, 1609 (1990); R. M. Potvliege and R. Shakeshaft, *ibid.* **40**, 3061 (1989); M. Dörr, Ph.D. thesis, University of Southern California, 1990.

[11] K. C. Kulander, B. W. Shore, J. Opt. Soc. Am. B**7**, 502 (1990); B. W. Shore and K. C. Kulander, J. Mod. Opt. **36**, 857 (1989); W. Becker, S. Long, and J. K. McIver, Phys. Rev. A **41**, 4112 (1990); B. Sundaram and P. W. Milonni, Phys. Rev. A **41**, 6571 (1990); X. F. Li, A. L'Huillier, M. Ferray, L. A. Lompré, and G. Mainfray, Phys. Rev. A **39**, 5751 (1989) and references therein.

[12] A. L'Huillier, L. A. Lompré, G. Mainfray, and C. Manus, *Proceedings of the 5th International Conference on Multiphoton Processes,* edited by G. Mainfray and P. Agostini

(CEA, Paris, 1990), p. 45; M. Ferray, A. L'Huillier, X. F. Li, L. A. Lompré, G. Mainfray, and C. Manus, J. Phys. B **21**, L31 (1988); A. McPherson, G. Gibson, H. Jara, U. Johann, T. S. Luk, I. McIntyre, K. Boyer, and C. K. Rhodes, J. Opt. Soc. Am B **4**, 595 (1987).

[13] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys.—JETP **20**, 1307 (1965)].

[14] B. W. Shore and P. L. Knight, J. Phys. B **20**, 413 (1987).

[15] M. V. Ammosov, M. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys.—JETP **64**, 1191 (1986)]; A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, *ibid.* **50**, 1393 (1966) [**23**, 924 (1966)].

[16] S. Augst, D. Strickland, D. D. Meyerhofer, S. L. Chin, and J. H. Eberly, Phys. Rev. Lett. **63**, 2212 (1989).

[17] Shih-I Chu and R. Y. Yin, J. Opt. Soc. Am. B **4**, 720 (1987).

[18] J. G. Leopold and I. C. Percival, J. Phys. B **12**, 709 (1979).

[19] G. Bandarage, A. Maquet, and J. Cooper, Phys. Rev. A **41**, 1744 (1990).

[20] B. Sundaram and P. W. Milonni, Phys. Rev. A **41**, 6571 (1990); P. L. Knight and P. W. Milonni, Phys. Rep. **66**, 21 (1980); J. H. Eberly (unpublished).

[21] J. H. Eberly and K. Rzazewski (private communication).

[22] A. L'Huillier, X. F. Li, and L. A. Lompré, J. Opt. Soc. Am. B **4**, 527 (1990).