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Suppression of peak switching in above-threshold ionization spectra

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The essential-state approach is employed to describe above-threshold ionization (ATI) of a single atom. Populations of consecutive peaks in electron spectra are calculated by integrating over space and taking into consideration laser spatial shapes. A significant effect of finite interaction volume on electron energy distributions is found, originating from either the limited acceptance of the detectors or size of aperture. In particular, peak switching of electron spectra, a striking feature of ATI, is suppressed and even destroyed by a large aperture or detector acceptance. These effects must be considered before any comparison can be made between theory and experiment.

I. INTRODUCTION

Above-threshold ionization (ATI), which refers to an atomic process carried out by the absorption of additional photons over the minimum number required for ionization, has recently been an intriguing subject in atomic physics. The interest in this subject is stimulated mainly by the discovery of many novel features in this ionization process, such as the so-called "peak switching" of the electron spectra,[1-4] i.e., the relative sizes of the consecutive peaks in the spectra become inverted when the field strength exceeds a certain value and the largest peak is switched gradually to higher electron energies with further increasing laser intensity.

To account for these novel features in the electron spectra, a variety of theoretical models have been proposed. Peak switching has been attributed to a large ac Stark shift of the ionization potential,[5] to saturation of continuum-continuum transitions,[6-8] and to modulation of the continuum energies by diagonal continuum-continuum couplings[9-13] in strong field, and so on. It has also been pointed out[3,14,15] that several other mechanisms, such as electron angular distributions, space-charge effects, and laser pulse shapes, might contribute appreciably to the relative amplitudes of the successive peaks and must consequently be considered before any comparison is made between theory and experiment. In this paper, we will indicate another factor that remarkably affects the ATI spectra, and especially the peak-switching phenomenon, and demonstrate the importance of the factor by considering a very simple example of a Gaussian laser beam. This factor is the finite-interaction volume experimentally observed due to the limited acceptance of the electron (ion) detector or to the size of the aperture used to attenuate the laser beam.

Briefly stated, ATI experiments are performed typically as follows: an amplified, very intense laser beam is focused into and irradiates the gas target which is filled in a vacuum chamber at low pressure, with outgoing electrons or ions being detected by an electron or ion spectrometer. In many cases, with the help of an aperture, only the center part of the laser beam is selected (see, e.g., Ref. 1) and focused. Furthermore, the detector acceptance is usually finite, which leads to considerable modification of the electron and ion yields. The modification resulting from finite acceptance has already been considered by Sogard[16] in the case of ordinary nonresonant multiphoton ionization (without continuum-continuum transitions), and he has indicated the important effect of both the finite acceptance and the shape of the lens used. In view of these facts, the size of the aperture and the finite acceptance will certainly alter the ionization volume and thus the ion yield and the electron distributions which will be discussed in the present work.

II. STATE PROBABILITIES FOR A SINGLE ATOM

For a given atom exposed to a laser field with local intensity I, the probabilities for the atom to be in different states satisfy the following equations:

$$i\dot{u}_0=D_{01}\int d\omega_1 u_{\omega_1}\ , \tag{1}$$

$$i\dot{u}_{\omega_1}-(\omega_1-n\omega)u_{\omega_1}=D_{10}u_0+D_{12}\int d\omega_2 u_{\omega_2}\ , \tag{2}$$

$$i\dot{u}_{\omega_l}-[\omega_l-(n+l-1)\omega]u_{\omega_l}=D_{l,l-1}\int d\omega_{l-1}u_{\omega_{l-1}}+D_{l,l+1}\int d\omega_{l+1}u_{\omega_{l+1}}\ ,\quad l=2,3,\ldots, \tag{3}$$

whereas in the standard saturation theory of ATI,[6-8] only essential states are concerned including the ground state $|0\rangle$ and different continuum bands with the energies ω_l. The ground state is coupled by absorption of n photons to the first continuum, and the continuum bands are successively coupled to one another with matrix elements

$D_{l,l+1}$ ($l=1,2,\dots$). Note that the detailed continuum structure is ignored as usual in order to focus on the essentials only. Following Deng and Eberly,[8] by introducing a new set of quantities,

$$K_l=\int d\omega_l u_{\omega_l} , \tag{4}$$

and performing some manipulations, we obtain the state probabilities for a specific atom; and for the case of square pulse,

$$P_0=|u_0|^2=\exp(-2\pi D_{10}^2 R_1 t) , \tag{5}$$

$$P_l=\int d\omega_l |u_{\omega_l}|^2=R_1[1-\exp(-2\pi D_{10}^2 R_1 t)]\prod_{q=2}^{l} Z_q R_q^2 ,$$

$$l=1,2,\dots, \tag{6}$$

where Z_l are the intensity-dependent parameters determining whether the continuum-continuum couplings are strong or not, and they are defined as

$$Z_l=\pi^2|D_{l,l+1}|^2 . \tag{7}$$

R_l are a set of continued fractions satisfying the following recursion relation:

$$R_l=\frac{1}{1+Z_l R_{l+1}} . \tag{8}$$

Equations (5) and (6) together with Eqs. (7) and (8) determine the state probabilities for a given atom. If the laser field is uniform in the interaction volume, these equations also describe the photoelectron energy spectrum. In Fig. 1 we plot according to these equations the populations of different continuum bands labeled with l versus laser intensity. In calculating these curves, considering that continuum-continuum matrix elements are generally decreasing functions[17,18] of l, we choose them according to a simple empirical rule:[8]

$$D_{l,l+1}/D_{l-1,l}=\beta . \tag{9}$$

Note that the laser intensity I is defined as Z_1 [see Eq. (7)] and T is the duration of the square pulse. In addition, D_{10}^2 is proportional to I^n.

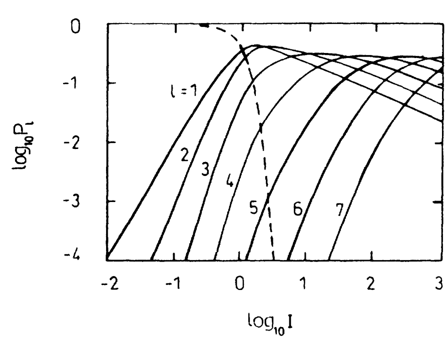

FIG. 1. Log-log plot of state probabilities vs laser intensity I for different continuum states labeled with l, which also relates to amplitudes of consecutive peaks in electron spectra for uniform field. The dashed line is the ground-state probability and I is defined as Z_l [see Eq. (7)]. Parameters are $n=2$, $T=10$, and $\beta=2$.

It is readily observed from these curves that (i) amplitudes of consecutive peaks decrease with index l for a weak field; (ii) as the laser intensity exceeds a certain value, the relative size of the first over the second peak become inverted, with the first peak being suppressed; (iii) with further increasing laser intensity, the largest peak is successively switched from the first peak to a higher one (peak switching). These features are manifest in most ATI experiments and are well reproduced here. Yet the laser spatial shape is ignored here and a uniform field distribution is assumed, which is certainly unjustified in most cases. Note that the curves labeled with l also signify state probabilities for a single atom.

III. POPULATIONS OF CONSECUTIVE ELECTRON PEAKS

Strictly speaking, Eqs. (5) and (6) apply only to a single atom. As a laser beam is usually used in ATI experiments, atoms at different locations will "feel" different local laser intensities and consequently associate with different degrees of ionization. Take, for a simple example, the saturation regime. Atoms in the central part of the laser beam are fully ionized, but those at the edge of the beam are weakly ionized so that the total ion yield monotonously increases with laser intensity, even though the atoms at the focus are depleted. This is the saturation effect of the ion-yield curves observed in most multiphoton ionization (MPI) experiments.

Now we turn to deal with the electron numbers in the continuum. It is worth reminding the reader that the squares of all the dipole couplings in Eqs. (5) and (6) are proportional to the local intensity $I(\mathbf{r})$ acting on the atom, with D_{12}^2 proportional to $I^n(\mathbf{r})$. As a result, these two equations rely on coordinates of the atom and describe the behavior of the local atom. The total number of ions N_i is the integration of $1-P_0$ over the interaction volume, and so are the electron numbers. By the total electron (ion) number we henceforth mean the number of electrons (ions) that reach the detectors.

As the acceptance of the detector is usually limited, N_i corresponds to the ions created in the volume within the range of detector acceptance, i.e.,

$$N_i=\rho_0\int_{V_I}[1-P_0(\mathbf{r})]d\mathbf{r} , \tag{10}$$

where the atomic gas is assumed to be homogeneously filled in a vacuum chamber with density ρ_0, and V_I is the interaction volume "seen" by the detector. Analogously, the electrons within the lth continuum band or the amplitude of the lth peak in the electron spectrum are

$$N_l=\rho_0\int_{V_I}P_l(\mathbf{r})d\mathbf{r} . \tag{11}$$

To see how the detected interaction volume V_I influences the electron distributions, we consider a simplified example of a real experiment. We consider a Gaussian laser beam with the beam profile

$$I(\mathbf{r})=\begin{cases} I_0 e^{-(r/r_0)^2} & \text{if } r<r_l ,\\ 0 & \text{otherwise} ,\end{cases} \tag{12}$$

where the small variation in the interaction volume along the laser direction is neglected, I_0 is the maximum value of Z_l, r_0 is the beam waist, and r is the radius from the central axis of the laser beam. Note that the limitation of the beam in Eq. (12) originates from either the size of the aperture that attenuates the laser beam or finite detector acceptance. The electron number N_l is then

$$N_l=N_0\int_0^{r_l} P_l(r)2\pi r\, dr , \tag{11$'$}$$

with N_0 being the number of neutral atoms initially in the cylindrical volume with waist r_0 and r_l being the radius of the cylindrical interaction volume "observed" by the detectors. A similar treatment has already been performed by Sogard,[16] who discussed the usual nonresonant multiphoton ionization. Here we will deal with a more general MPI process with continuum-continuum transitions involved, that is, ATI.

Displayed in Fig. 2 is the laser intensity dependence of populations of different continua. Note that $r_l=2r_0$ is chosen so that only the central part of the laser beam is detected. Other parameters are the same as those in Fig. 1. The dashed line is the ion-yield curve. It is evident that peak switching manifests itself in these curves, but the relative amplitudes are less sensitive to the field strength than in Fig. 1, where all atoms are exposed to the same laser intensity I_0. In other words, peak switching is not so pronounced as in the case of a homogeneous field. In Fig. 3, we calculate these electron-yield curves for $r_l=10r_0$ and for the same parameters as in Fig. 2. Again the dashed line is the ion-yield curve. Peak switching is now completely destroyed in the laser intensity range of interest, and the populations of consecutive peaks in the electron spectrum decrease with index l as usual. In addition, the saturation effect due to the depletion of neutral atoms at the focus and to the expansion of the ionization region is evident in the ion-yield curve, which is consistent with experimental results.[2,3]

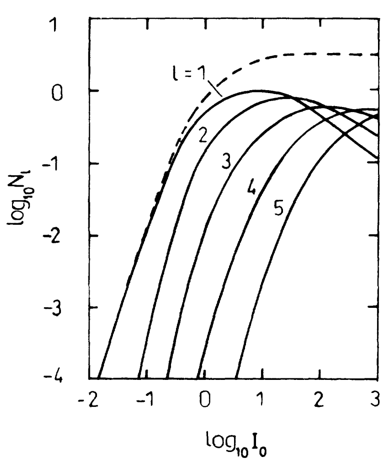

FIG. 2. Log-log plot of amplitudes of different electron peaks vs laser intensity. The beam profile is given by Eq. (12) with $r_l=2r_0$.

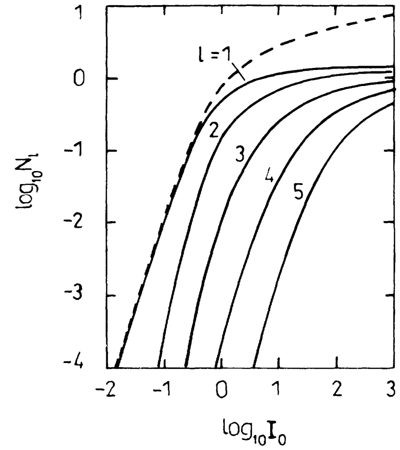

FIG. 3. The same as Fig. 2, except for $r_l=10r_0$, showing the destruction of peak switching in ATI spectra by finite-interaction volume.

IV. DISCUSSIONS AND SUMMARY

Suppression and even destroying of peak switching in electron spectra is not difficult to understand. As for a given atom, whether probabilities for consecutive continuum states are inverted or not (we will refer to the inversion as "probability inversion" but reserve the term "peak switching" to the same phenomenon for numerous atoms) depends on the local intensity it experiences (for details see Fig. 1); atoms at different locations feel different field strengths and therefore are of different probability distributions in the continuum. For instance, atoms at the center part of the laser beam are exposed to the strongest field and the most pronounced probability inversion happens to these atoms. On the other hand, atoms at the edge of the beam are exposed to the weak-field part, and they most likely absorb n photons and stay at the first continuum rather than absorb a larger number of photons and reach the higher-lying continua. The resulting electron distribution detected is determined by the average of the probability distribution over separate atoms. If one detects only the central part of the whole interaction volume in which most atoms experience a strong field (the case of Fig. 2), the inverted probability distribution among continuum bands is predominant, and the whole electron spectrum exhibits peak switching. If, however, the electron detector accepts electrons ejected from a larger part of the interacting atoms, most atoms detected experience the weak part of the laser beam, though the atoms at the focal center still exhibit probability inversion. In this case, atoms undertaking lowest-order ionization contribute most to the electron distribution and therefore wash out the strong-field effects. Particularly, peak switching of electron spectra is less pronounced or even destroyed, which highlights the importance of finite detector acceptance in calculating the detected electron spectra.

The finite-interaction volume is not necessarily attributed to the limited acceptance of the detectors. In many cases, an aperture is used in many experiments to select the center part of the laser beam. In this situation, the size of the aperture also remarkably affects the electron distribution. Roughly speaking, the aperture cuts off the boundary part of a laser beam and restricts the interaction volume to the center of the beam. The population in the l continuum may still be approximately expressed as Eq. (11′), and the results are unchanged. For example, peak switching will be suppressed or even broken down for a large aperture. In contrast, a small aperture must be used in order to observe more effectively the phenomenon of peak switching in ATI spectra.

In the foregoing analysis, we have taken a simple example of the beam profile with the form of Eq. (12). The results will be improved if a more realistic laser spatial shape is considered. Moreover, diagonal couplings have been ignored and only resonant continuum-continuum couplings have been taken into account. This implies that here we deal with the case of a not very intense light field, in which resonant continuum-continuum couplings dominate. Delicate calculations can be made and the present work may be extended to higher field intensity if diagonal continuum-continuum couplings are involved. Nevertheless, the qualitative results will be unchanged, since the qualitative intensity dependence of populations for different peaks remains as in Fig. 1, in spite of the inclusion of the diagonal continuum-continuum couplings.[9-13] We will discuss these problems elsewhere.

In summary, a very simple model approach is applied to investigate the laser intensity dependence of the amplitudes of successive peaks in ATI spectra, taking account of the laser spatial shape. It is found that the electron distribution is sensitive to the "observed" interaction volume. For large detector acceptance or aperture size, peak switching of ATI spectra, a striking feature of ATI, is suppressed and even destroyed. As a consequence, a relatively small aperture should be used in planning an ATI experiment to observe the characteristic feature, and the detector acceptance and the aperture size must be taken into account before any comparison can be made between theory and experiment.

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