Two-color multiphoton ionization of atoms

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We present a theoretical study on the two-color multiphoton ionization of atoms using a nonperturbative method of solving the Schrödinger equation in momentum space. We generalize the problem for absorption of an arbitrary number of ionization photons and especially focus on the absorption of additional above-threshold high-frequency photons. A systematic calculation on the photoelectron energy spectra for the hydrogen atom is performed. [S1050-2947(97)00704-X]

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Multiphoton ionization of atoms by an intense laser field exhibits a variety of unanticipated nonlinear phenomena. One of the most interesting effects in the above-threshold ionization (ATI) is the significant suppression of the lowest-order peaks in the photoelectron energy spectra with increased laser intensity. The peak-suppression effect is due to the threshold shift during the ionization process. Another striking feature in ATI is that the relative sizes of the consecutive peaks in the spectra become inverted. This "peak-switching" effect is caused by the multiple transitions of the electron through the continua during the redistribution of the final continuum states. The experiment of the simultaneous application of radiation of two different frequencies on atoms by Muller, Van Linden van den Henvel, and Van der Wiel [1] has provided a useful way to unambiguously distinguish between peak-switching and peak-suppression phenomena. In the experiment of Muller, Van Linden van den Henvel, and Van der Wiel, one laser of high frequency promotes the electron from a bound state to the continuum. The second laser of a low frequency has a large enough intensity to induce multiple transitions in the continuum and is responsible for the observed peaks in the photoelectron spectra. Thus, in the two-frequency experiment the ionization process can be separated from the continuum-continuum transition, furnishing a unique way to investigate the nonlinear effects in the photoelectron spectra of ATI. Theoretical studies on the two-color multiphoton ionization of atoms have ranged from the application of the Floquet method [2–4], the S-matrix formalism [5,6], the essential state approach [7,8], and the variational procedure [9] to the direct numerical solution of the time-dependent Schrödinger equation [10–13]. Recently, we used a nonperturbative method of solving the Schrödinger equation in momentum space to investigate the two-color multiphoton ionization of atoms [14]. We generalized the problem for the absorption of an arbitrary number of ionization photons and also included the threshold shifts due both to high-frequency and low-frequency fields. We found that the photoelectron spectrum peaks occur at energies $\epsilon_s = (N_{0H} + s)\hbar\omega_H + n_L\hbar\omega_L - I_i$ where s is the above-threshold high-frequency photon number and I_i is the ionization energy of the initial state. However, the previous work considered only the simple case where no additional above-threshold high-frequency photons are absorbed, i.e., $s=0$ only. In the present work, we shall extend our analysis to the general case of $s>0$ and perform a systematic calculation for

the photoelectron energy spectra of the hydrogen atom. We find that quite different results are obtained when different above-threshold high-frequency photons are absorbed. The mechanism for this different behavior will be discussed.

The method used in this paper has been presented in detail in previous works [14,15]; we give the essential results here. In our problem, the atom with the initial energy E_i is irradiated by two lasers: one a weak laser of relatively high frequency w_H and the other an intense laser of low frequency w_L. For the monochromatic linearly polarized fields

$$\vec{A}_0(t) = \vec{A}_{0j}\cos w_j t, \quad j = H, L \tag{1}$$

the ionization amplitude can be obtained as

$$a_{\vec{k}}(t) = a_{\vec{k}}(0) - \frac{i}{\hbar} b_{\vec{k}}(0) \sum_{n_H, n_L} J_{n_H}(\xi_H) J_{n_L}(\xi_L)$$
$$\times \int_0^t dt' \exp\left[\frac{i}{\hbar}\left(\frac{p^2}{2m} - E_i - n_H\hbar w_H\right.\right.$$
$$\left.\left. - n_L\hbar w_L - Q\right)t'\right], \tag{2}$$

where n_H photons of the high-frequency field are absorbed, causing the ionization of the atom, and n_L photons of the low-frequency field are then absorbed or emitted (for n_L positive or negative), causing the redistribution of the final states. $b_{\vec{k}}(0)$ is a slowly varying function extracted from the wave function and the coupling parameter $\xi_j = e/mcw_j(\vec{k} \cdot \vec{A}_{0j})$, $j=H,L$. The quantity Q is given as

$$Q = Q_H + Q_L, \tag{3}$$

where

$$Q_H = \frac{1}{b_{\vec{k}}(0)} \int d\vec{k}' \left\{ \left[\frac{\hbar^2 k'^2}{2m} - E_i - \frac{e\hbar(\vec{k}' \cdot \vec{A}_{0H})}{mc}\right] a_{\vec{k}'}(0) \right.$$
$$\left. + b_{\vec{k}'}(0) \right\} \langle \vec{k}|V|\vec{k}'\rangle, \tag{4}$$

$$Q_L = \frac{1}{b_{\vec{k}}(0)} \int d\vec{k}' \left(-\frac{e\hbar\vec{k}' \cdot \vec{A}_{0L}}{mc}\right) a_{\vec{k}'}(0)\langle \vec{k}|V|\vec{k}'\rangle. \tag{5}$$
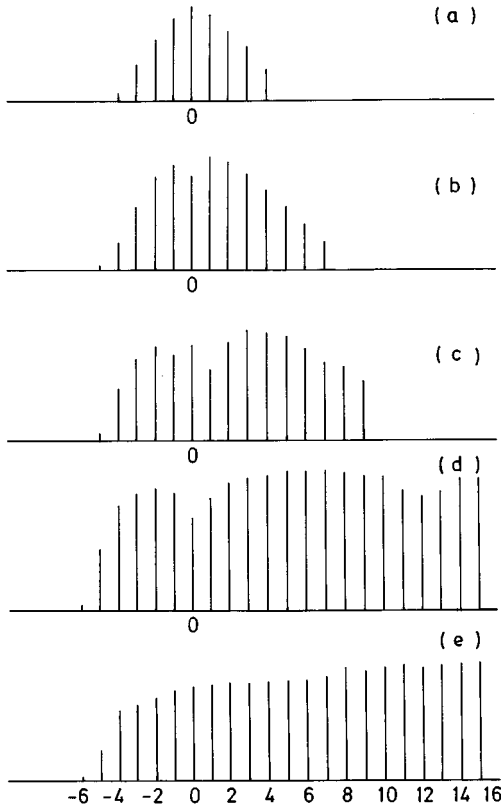
FIG. 1. Probability density $|a_k(t\rightarrow\infty)|^2$, normalized to the value of $n_L=0$, vs the number of exchanged low-frequency photons for $N_{0H}=2(w_H=0.4$ a.u.) and $s=0$. The low-frequency $w_L=0.04$ a.u. The laser field strengths $E_H=0.01$ a.u. and E_L are (a) 0.001 a.u., (b) 0.005 a.u., (c) 0.01 a.u., (d) 0.05 a.u., and (e) 0.1 a.u.

From Eq. (2), the photoelectron spectra peaks occur at energies

$$E_k=\frac{p^2}{2m}=n_H\hbar w_H+n_L\hbar w_L-I_i+Q, \qquad (6)$$

where $I_i=-E_i$ is the ionization energy for the initial state ϕ_i. Equation (6) shows that the energies are shifted by an amount Q, which is an important effect responsible for the peak switching in the photoelectron spectra.

We now present the numerical calculation of the two-color multiphoton ionization for the ground state of the hydrogen atom. Equation (6) can be rewritten as $E_k=(N_{0H}+s)\hbar w_H+n_L\hbar w_L-I_i+Q$, where $n_H=N_{0H}+s$, N_{0H} is the minimum number of the high-frequency photons required for ionization, and s is the number of above-threshold high-frequency photons. We have calculated the probability function $|a_k(t\rightarrow\infty)|^2$ for different high-frequency photon numbers and low-frequency field strengths. A geometry where the two laser fields are in the same direction is assumed. Figures 1–3 show the results of the photoelectron spectra for $N_{0H}=2(w_H=0.4$ a.u.), with $s=0$, 1, and 2, respectively. It can be seen from Fig. 1 ($s=0$) that the spectra for the absorption and emission processes are not symmetric even at low laser field E_L. In general, the sidebands corresponding to absorption are stronger than those of emission. The reason is that for the emission process, there is a cutoff value n_c for emission such that $n_L\hbar w_L+n_H\hbar w_H-I_i>0$. In
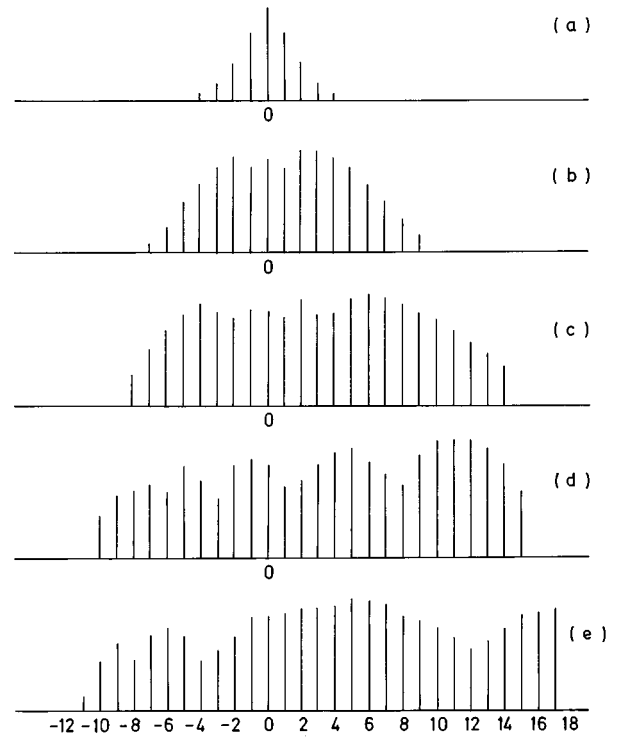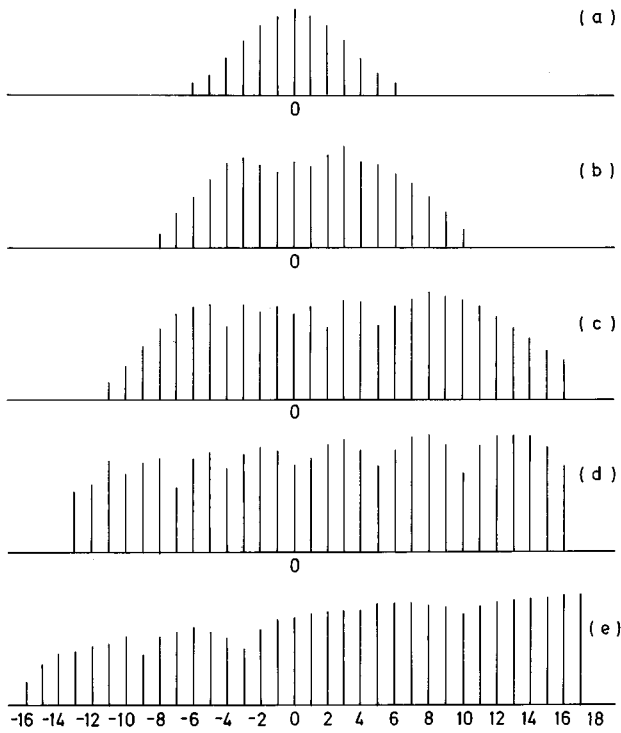

FIG. 2. Same as in Fig. 1, but for $s=1$.

this case ($w_H=0.4$ a.u. and $w_L=0.04$ a.u.), $n_c=-7$. The emission process can only occur from -1 to n_c, while the absorption channels are open from 1 to ∞. Therefore, the probability becomes vanishingly small as n_L approaches n_c and thus the absorption process will have a larger probability than the emission process. Wang, Haus, and Rzażewski [8] have calculated the photoelectron spectra using the essential state method and recently Taïeb, Véniard, and Maquet [13] also studied the spectra using direct numerical solution of the time-dependent Schrödinger equation. In their results, the sidebands of the absorption and emission spectra are found to be symmetric at low field E_L. This is because they consider the simple case that only one high-frequency photon is absorbed to promote the electron directly to the continuum, i.e., the value of w_H is large, thus the cutoff value n_c is also large and its effect is no longer important. In the present work, we have generalized the problem not only for absorption of more than one high-frequency photons ($N_{0H}>1$), but also for absorption of additional above-threshold high-frequency photons ($s\geqslant1$). Our results clearly exhibit peak switching at high field E_L, that is, the relative sizes of the consecutive peaks in the spectra become inverted. It is found that for the case $s=0$ (Fig. 1), the peak switching occurs at many different n_L for the absorption spectra, but only a few instances of peak switching occur for the emission spectra. However, we find quite different results for the cases of $s=1$ and 2 as shown in Figs. 2 and 3, respectively. Here the peak switching occurs at many n_L not only for the absorption process, but also for the emission process. This different behavior between $s=0$ and 1,2 can be seen more clearly in the plot of the probability function versus the low-frequency field strength E_L with different photon multiplicities n_L. Figures 4 and 5 show the results for $N_{0H}=2$, with $s=0$, 1, and 2, for the absorption and emission processes, respectively. In

FIG. 3. Same as in Fig. 2, but for $s=2$.

FIG. 5. Same as in Fig. 4, but for the emission process.

general, the curves increase linearly with the laser field for low-field intensities. As the field intensity increases, the higher-order n_L curves increase more rapidly and eventually overcome the lower-order ones. Thus the peak switching will
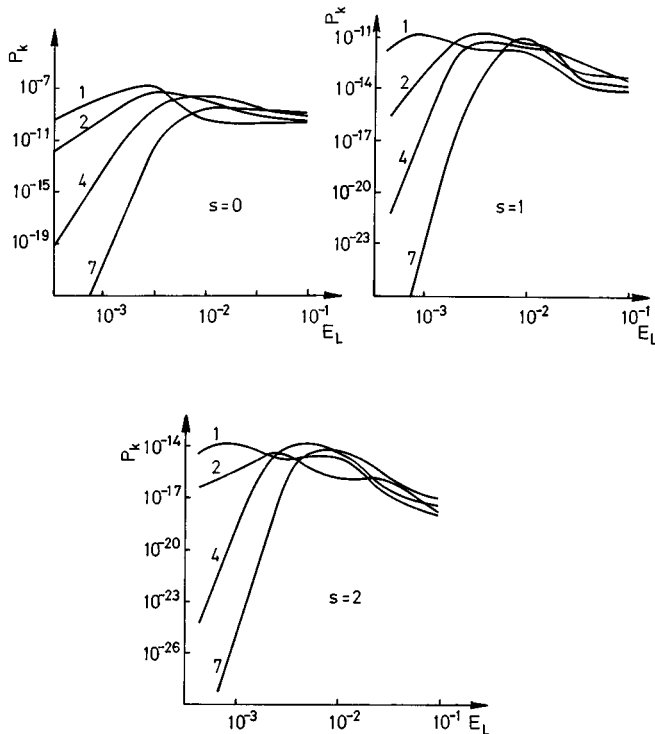

FIG. 4. Probability density as a function of the low-frequency field strength E_L with the exchange of different photon numbers n_L for the absorption process for $N_{0H}=2$, with $s=0$, 1, and 2. The high-frequency field strength $E_H=0.01$ a.u., with $w_H=0.4$ a.u. and $w_L=0.04$ a.u.
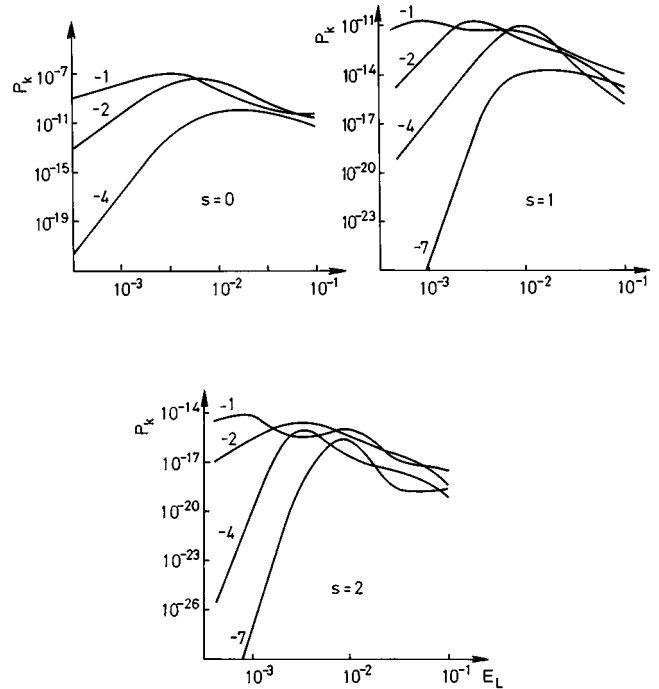
occur at some critical field for higher-order multiplicities n_L. For the absorption process (Fig. 4), the curves are similar for the cases $s=0$, 1, and 2. It can be seen that all the curves reverse when the field E_L increases to higher intensities. The situation is quite different for the emission process (Fig. 5), where for the case $s=0$ the peak switching only occurs at a few instances between $n_L=-1$ and -2 and never occurs for $n_L=-4$. However, for the cases $s=1$ and 2, the peak switchings occur at many different n_L and the curves start oscillating in the high-field region. It is interesting to note that the oscillation becomes more rapid as s gets larger. We have also studied the case of $N_{0H}=3(w_H=0.25)$, with $s=0$, 1, and 2. The above difference in behavior between $s=0$ and 1,2 is again observed. The reason for this is due to the energy shift Q, which is an important effect responsible for the peak switching as we shall discuss in the following.

Figure 6 shows the result of the energy shift Q as a function of the field strength E_L for $N_{0H}=2$, with $s=0$, 1, and 2. For the absorption process [Fig. 6(a)], Q is small for weak-field strength and its effect is negligible, so the lower-order spectra are dominant and no peak switching occurs. As the field E_L increases to higher values, the magnitude of Q increases quite fast for all the cases $s=0$, 1, and 2 and its effect becomes important. The peak switching will then occur successively for each higher-order curve at some critical field and the higher-order n_L curves are eventually all reversed in the high-field region as indicated in Fig. 4. For the emission process, it can be seen from Fig. 6(b) that for the case $s=0$, Q increases very slowly for the lower-order curve ($n_L=-1$) and becomes almost constant for higher-order curves ($n_L=-4$ and -7). Therefore, peak switching occurs only a few times for lower-order multiplicities and never occurs for higher-order ones for the case $s=0$, as we have also pointed out before in Fig. 5. However, when additional above-threshold photons are absorbed ($s=1,2$), Q increases very
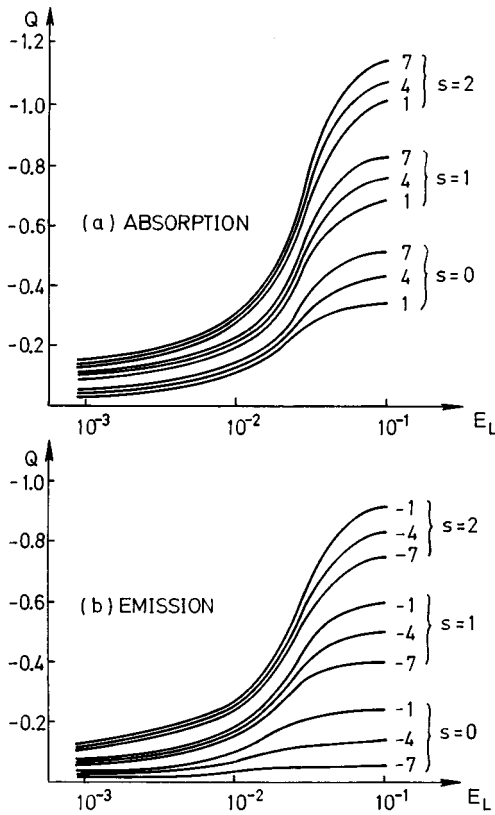
FIG. 6. Energy shift Q as a function of the low-frequency field strength E_L (a.u.) with the exchange of different photon numbers n_L for $N_{0H}=2$, with $s=0$, 1, and 2, for the (a) absorption process and (b) emission process. The high-frequency field strength $E_H=0.01$ a.u., with $w_H=0.4$ a.u. and $w_L=0.04$ a.u.

fast. Therefore, the peak switching will occur at many n_L for both cases. It is also interesting to note that Q increases faster for the case $s=2$ than for $s=1$, thus the oscillation of the probability curve in the high-field region becomes more

rapid for $s=2$ than for $s=1$, as we have also indicated before in Fig. 5. Therefore, our result shows that the energy shift Q is closely related to the peak switching in the photoelectron energy spectra and the correspondence between them seems to be quite consistent. As given in Eq. (3), Q contains two parts: Q_H is the energy shift due to the high-frequency field during the ionization process and Q_L is that due to the low-frequency field during the redistribution of the continuum states. We have studied the competition between Q_H and Q_L. We find that Q_H is larger than Q_L in the low-field E_L region and Q_L becomes much larger than Q_H in the high-field E_L region. This is because the ionization of the atom dominates the process for the lower field E_L, while the re-population of the final states becomes dominant for the higher field E_L. It is also found that the contribution of Q_H will increase as more above-threshold high-frequency photons are absorbed.

In conclusion, we have presented a systematic calculation on the two-color multiphoton ionization of hydrogen atom and extended the problem for absorption of additional above-threshold high-frequency photons. Our results show that quite different behaviors are obtained between the cases $s=0$ and 1,2. We find that for emission spectra the peak switching occurs only at a few instances between low multiplicities and never occurs for higher-order multiplicities for the case $N_{0H}=2$, with $s=0$. This is because the energy shift Q is small for low multiplicities and become almost constant for higher orders. However, the situation becomes quite different for the cases $s=1$ and 2, where Q increases very fast as the field strength E_L increases, especially for the higher-order multiplicities. Therefore, the peak switching will then occur successively for each high-order curve and the curves start oscillating in the high-field regions. It is also found that the oscillation is more rapid for larger values of s.

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