Two-color above-threshold ionization of atoms

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A nonperturbative method is used to study the two-color multiphoton ionization of atoms by solving the Schrödinger equation in momentum space, which facilitates the extraction of the rapidly varying part of the wave function. Our method generalizes the problem for cases where an arbitrary number of ionization photons are absorbed, and also takes into account the threshold shifts due to both the high- and low-frequency fields. The photoelectron energy spectra of the hydrogen atoms are calculated and the dependence on the laser field strengths and frequencies is discussed. [S1050-2947(96)05205-3]

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I. INTRODUCTION

The simultaneous application of radiation of two different frequencies has led to interesting new phenomena in multiphoton ionization of atoms, and attracted many experimental and theoretical investigations [1–10]. In the experimental study of the above-threshold ionization (ATI) of atoms by Muller *et al.* [1], one laser emitted light with frequency in the ultraviolet region, while another emitted light in the infrared region. The ultraviolet light was responsible for the ionization of atoms, while the more intense laser in the infrared region was responsible for the repopulation between the continua and the observed multiple peaks in the photoelectron spectra. Therefore the two-color experiments provide an opportunity for separating the ionization process from the continuum-continuum transition, and pose challenges for the theoretical studies. Another interesting feature of this process is that the sidebands due to the emission of infrared photons are stronger than the absorption of the photons, resulting in an asymmetric photoelectron spectra. Leone *et al.* [4] have studied the two-frequency ionization of hydrogen atoms using a *S*-matrix formalism, and calculated the angular distribution and the photoelectron spectra. Rzażewski, Wang, and Haus [5] have applied the essential state model to the study of two-color above-threshold ionization, and the photoelectron energy spectra were also derived. Dörr and Shakeshaft [3] have investigated the problem using the Floquet method. Both the resonance-enhanced multiphoton ionization and nonresonance ionization were calculated. Recently, Zhou and Rosenberg [6] have developed a variational procedure for two-color ionization and tested this method numerically for a one-dimensional problem. In all these previous works, the high-frequency field is considered to be either weak enough to be treated perturbatively or to have a large enough frequency that only one high-frequency photon is absorbed to promote the electron directly to the continuum, thus avoiding the complications due to the threshold shift.

Recently, an efficient method of solving the time-dependent Schrödinger equation for a system undergoing multiphoton processes was introduced [11–13]. An important feature of this method is that the Schrödinger equation is solved in momentum space, which facilitates the extraction of the rapidly varying part of the wave function. In the present paper, we shall apply this nonperturbative method to investigate the two-color multiphoton ionization of atoms and generalize the problem for absorption of an arbitrary number of ionization photons. We shall also take into account the threshold shifts due to both the high-frequency and low-frequency fields. A detailed calculation of the above-threshold photoelectron spectra of the hydrogen atom will be performed and the dependence on the laser field strengths and frequencies will also be discussed.

II. THEORY

We consider the ionization of an atom by two lasers, one a weak laser of relatively high-frequency w_H, and the other an intense laser of low-frequency w_L. The interaction between the electron and the two radiation fields is

$$H_I(t) = H_I^H(t) + H_I^L(t), \tag{1}$$

with

$$H_I^j(t) = -\frac{e}{mc}\vec{A}_j(t)\cdot\vec{p} + \frac{e^2}{2mc^2}A_j^2(t), \quad j=H,L, \tag{2}$$

where $A_H(t)$ and $A_L(t)$ are the vector potentials for the high-frequency and low-frequency fields, respectively. The $A^2(t)$ term can be removed by a trivial contact transformation [14].

The method used in this paper has been presented in detail elsewhere [13]. We briefly review the essential features here. The time-dependent wave function $\Psi(t)$ is expanded in terms of the Volkov functions [15] $\chi_{\vec{k}}(t)$ which are solutions for problems where the atomic potential V vanishes and the electron moves freely in the radiation fields,

$$|\Psi(t)\rangle = \int d\vec{k}\, a_{\vec{k}}(t)|\chi_{\vec{k}}(t)\rangle. \tag{3}$$

For the monochromatic linearly polarized fields,

$$\vec{A}_j(t) = \vec{A}_{oj}\cos w_j t, \quad j=H,L, \tag{4}$$

the Volkov function has the form

$$|\chi_{\vec{k}}(t)\rangle = \exp\{-i[E_i t/\hbar + \theta_{\vec{k}}(t)]\}|\vec{k}\rangle, \tag{5}$$

where E_i is the initial energy, $|\vec{k}\rangle$ the eigenvector of $\vec{p}$, and the phase factor $\theta_{\vec{k}}(t)$ is given by

$$\theta_{\vec{k}}(t) = \frac{1}{\hbar}\left(\frac{\hbar^2 k^2}{2m} - E_i\right)t - \frac{e}{\hbar m c w_H}\vec{p}\cdot\vec{A}_{oH}\sin w_H t$$

$$- \frac{e}{\hbar m c w_L}\vec{p}\cdot\vec{A}_{oL}\sin w_L t. \tag{6}$$

Substituting $|\Psi(t)\rangle$ of Eq. (3) into the time-dependent Schrödinger equation leads to a set of first-order inhomogeneous integrodifferential equations for the coefficient $a_{\vec{k}}(t)$. A slowly varying function $b_{\vec{k}}(t)$ is extracted and we can extrapolate it using a Taylor-series expansion. Following the procedures described in Ref. [13], it is easy to obtain the ionization amplitude

$$a_{\vec{k}}(t) = a_{\vec{k}}(0) - \frac{i}{\hbar}\sum_{n_H}\sum_{n_L} J_{n_H}(\xi_H)J_{n_L}(\xi_L)b_{\vec{k}}(0)$$

$$\times \int_0^t dt' \exp\left[\frac{i}{\hbar}\left(\frac{p^2}{2m} - E_i - n_H\hbar w_H\right.\right.$$

$$\left.\left. - n_L\hbar w_L - Q\right)t'\right], \tag{7}$$

where $a_{\vec{k}}(0)$ can be determined from the initial boundary condition and the coupling parameter $\xi_j = e/mcw_j(\vec{k}\cdot\vec{A}_{0j})$, $j = H, L$. The quantity Q is given as

$$Q = Q_H + Q_L, \tag{8}$$

$$Q_H = \frac{1}{b_{\vec{k}}(0)}\int d\vec{k}'[\dot{F}_{\vec{k}'}^H(0)a_{\vec{k}'}(0) + b_{\vec{k}'}(0)]\langle\vec{k}|V|\vec{k}'\rangle, \tag{9}$$

$$Q_L = \frac{1}{b_{\vec{k}}(0)}\int d\vec{k}' \dot{F}_{\vec{k}'}^L(0)a_{\vec{k}'}(0)\langle\vec{k}|V|\vec{k}'\rangle, \tag{10}$$

with

$$F_{\vec{k}}^H(t) = \left(\frac{\hbar^2 k^2}{2m} - E_i\right)t - \frac{e\hbar(\vec{k}\cdot\vec{A}_{oH})}{mcw_H}\sin w_H t, \tag{11}$$

$$F_{\vec{k}}^L(t) = -\frac{e\hbar(\vec{k}\cdot\vec{A}_{oL})}{mcw_L}\sin w_L t. \tag{12}$$

Equation (7) shows that the photoelectron spectra peaks occur at energies

$$E_k = \frac{p^2}{2m} = n_H\hbar w_H + n_L\hbar w_L - I_i + Q, \tag{13}$$

where $I_i = -E_i$ is the ionization energy for the initial state ϕ_i. It can be seen that Eq. (13) describes the process in which n_H photons of the high-frequency field are absorbed causing ionization, and then n_L photons of the low-frequency field are absorbed or emitted (for n_L positive or negative), causing the redistribution of the continuum states. As we have pointed out before, all the previous calculations are limited to the case of $n_H = 1$. Our result has generalized the problem for absorption of an arbitrary number of high-
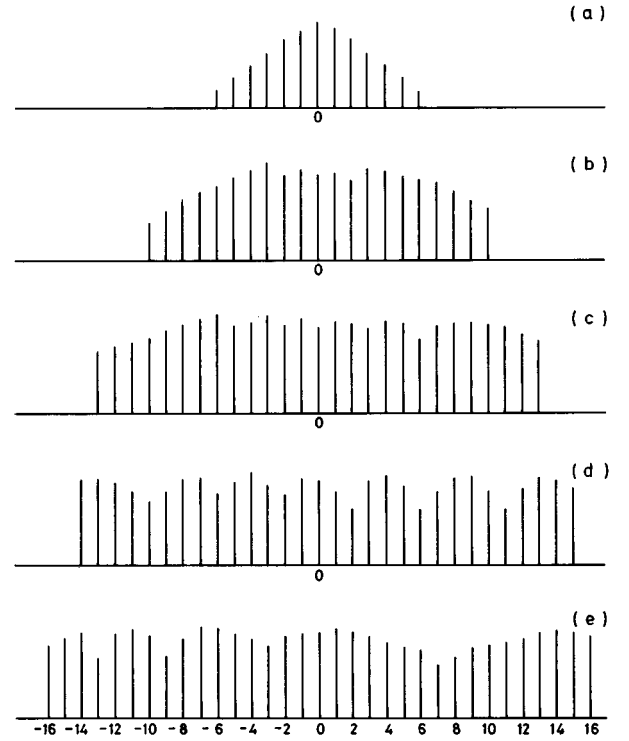

FIG. 1. Probability density $|a_k(t\to\infty)|^2$, normalized to the value of $n_L=0$, vs the number of exchanged low-frequency photons for $w_H=1.8$ a.u. and $w_L=0.04$ a.u. The laser field strengths $E_H=0.001$ a.u. and E_L are (a) 0.001 a.u., (b) 0.005 a.u., (c) 0.01 a.u., (d) 0.05 a.u., (e) 0.1 a.u.

frequency photons. More importantly, Eq. (13) shows that the energies are shifted by an amount Q, which comes from the $\vec{A}\cdot\vec{p}$ interaction and is an important effect responsible for the peak switching in the photoelectron spectra. As given in Eq. (8), Q contains two parts, where 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.

III. RESULTS AND DISCUSSION

In this section we present the numerical calculation of the two-color multiphoton ionization for the ground state of the hydrogen atom. We have calculated the probability function, $|a_k(t\to\infty)|^2$, for different values of the high-frequency photon energy and low-frequency field strengths. We assume a geometry where the two laser fields are in the same direction. The expression for $|a_k(t\to\infty)|^2$ can be easily obtained from Eq. (7) by letting $t\to\infty$. The electronic wave vector k is calculated using Eq. (13). Since Q is also a k-dependent function, no analytic expression for k can be obtained and a numerical solution of Eq. (13) for k is carried out. We first study the case where only one high-frequency photon is absorbed for ionization, i.e., $n_H=1$. Figure 1 shows the result of the photoelectron spectra for $w_H=1.8$ a.u. and $w_L=0.04$ a.u. The values are normalized to that of $n_L=0$. The results for a lower high-frequency value $w_H=1.0$ a.u. is shown in Fig. 2. It can be seen from Fig. 1 that at a small value of the low-frequency field strength ($E_L=0.001$ a.u.), the sidebands of the absorption and emission spectra are symmetric. As the
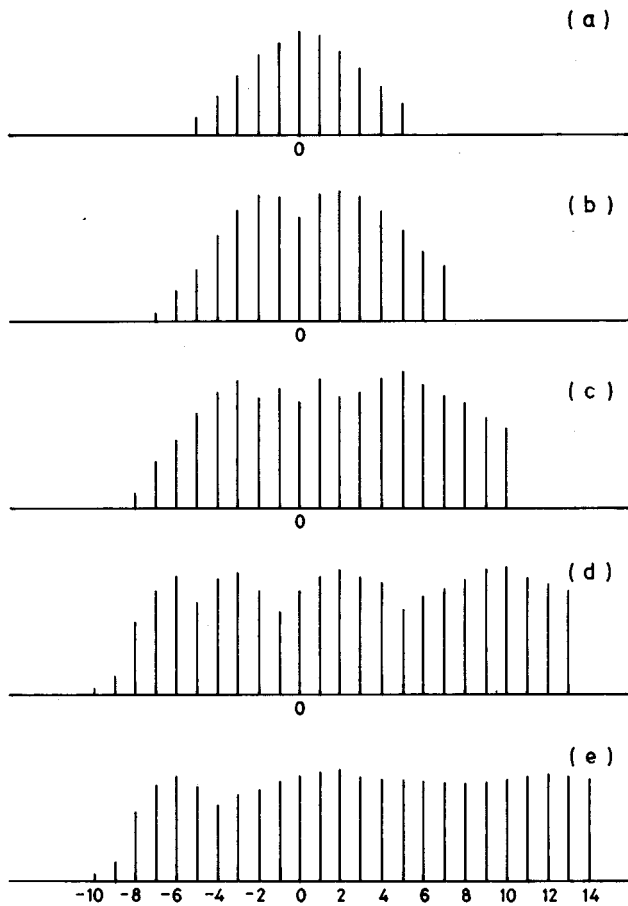
FIG. 2. Same as in Fig. 1, but for $w_H=1.0$ a.u.

FIG. 3. Same as in Fig. 1, but for $n_H=2$ ($w_H=0.4$ a.u.) and $E_H=0.01$ a.u.

field E_L increases, the photoelectron spectra become asymmetric. In general, the sidebands corresponding to the emission are stronger than those of absorption, at least for the small values of n_L. Our results also exhibit peak switching, that is, the lowest-order peak is no longer the strongest one. It is found that at a lower field strength $E_L=0.005$ a.u., the peak switching occurs only between $n_L=-3$ and $+3$, and that the probability function then decreases for larger $|n_L|$. When E_L increases ($E_L>0.01$ a.u.), the peak switching occurs at many different n_L and the probability function exhibits oscillation. In the oscillatory region the maximum and minimum generally occur at different n_L for the emission and absorption sidebands. Leone *et al.* [4] have calculated the photoelectron spectra using a S-matrix formalism and obtained similar results as above. Therefore our results are consistent with their's in this case. However we obtain a different result for the case of a lower value $w_H=1.0$ a.u. as shown in Fig. 2, in which it is clearly seen that the absorption sidebands are always stronger than the emission ones. This is due to the reason that for the emission process the coupling parameter ξ_L is small and, in addition, it must be remembered that there is a cutoff value n_C for the emission such that $n_L\hbar w_L+n_H\hbar w_H-I_i>0$. In our case ($w_H=1.0$ a.u., $w_L=0.04$ a.u.), $n_C=-12$. 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 ones. We
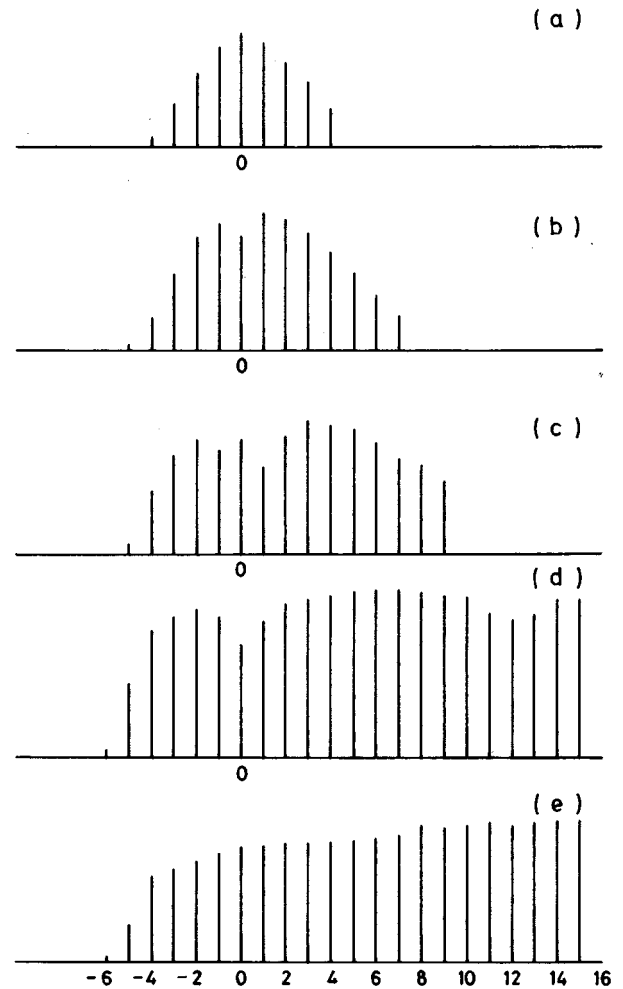
have also extended the calculation to cases where absorption of more than one high-frequency photons is necessary for ionization. Figures 3 and 4 show the results for $n_H=2$ ($w_H=0.4$ a.u.) and $n_H=3$ ($w_H=0.25$ a.u.), respectively. In these cases, the photoelectron spectra are no longer symmetric even for the small field E_L. In general, the absorption probabilities are larger than those of the emission ones. It is also interesting to note that for the emission spectra in Fig. 3, only a few instances of peak switching occur and no peak switching occurs at all in Fig. 4, even in the high E_L fields. 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 later. For the absorption spectra the behavior is similar to that of $n_H=1$, however, the oscillation is much slower. In order to present a more qualitative description of these findings, we have plotted the probability function versus the low-frequency field strengths E_L for different photon multiplicities $\pm n_L$ as shown in Figs. 5, 6, and 7 for $n_H=1$, 2, and 3, respectively. Figure 5 clearly shows that for low intensities, the curves increase linearly with the laser field E_L. As the intensity increases, the higher-order n_L curves increase rapidly and eventually overcome the lower-order ones. Thus, the peak-switching will occur successively at some critical fields for higher-order multiplicities n_L. In the
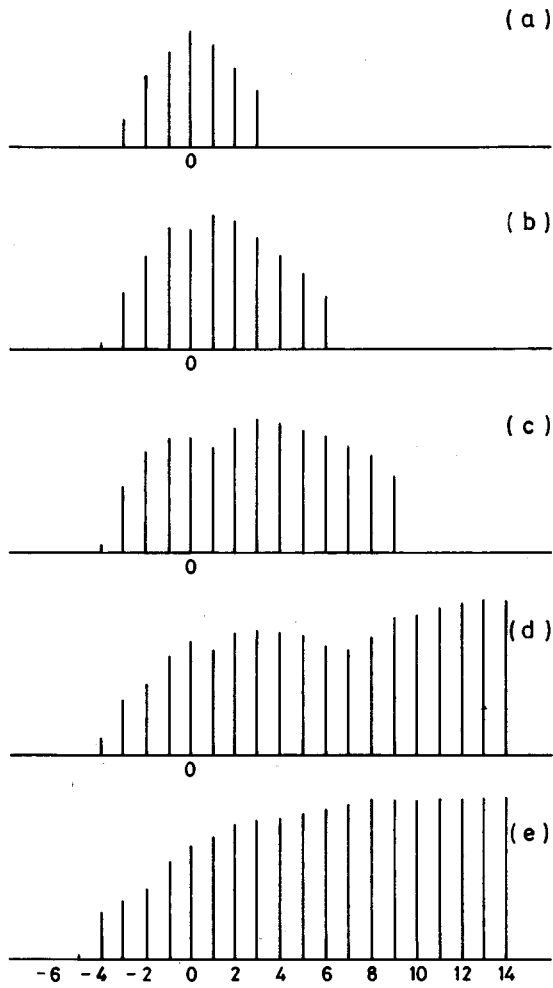
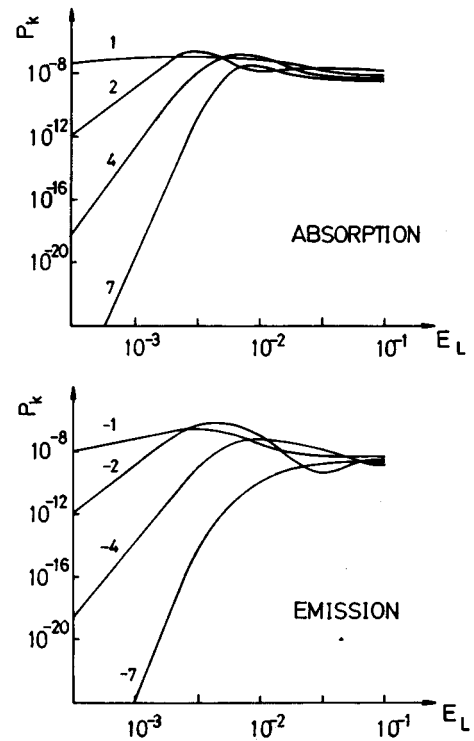FIG. 4. Same as in Fig. 3, but for $n_H=3$ ($w_H=0.25$ a.u.).

FIG. 5. Probability density as a function of the low-frequency field strength E_L (a.u.) for the absorption and emission of the different photon number n_L. The high-frequency field strength $E_H=0.001$ a.u. and $w_H=1.0$ a.u., $w_L=0.04$ a.u.
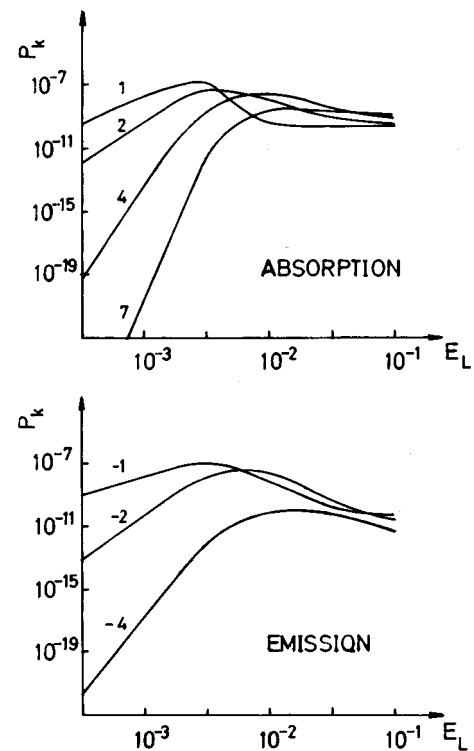
high-field region, both emission and absorption spectra start oscillating and approach saturation because an increasing number of channels open for a high-field strength, which eventually have comparable probabilities. However the situation becomes quite different for the cases of $n_H=2$ and $n_H=3$. As shown in Fig. 6 ($n_H=2$), the peak switching occurs only between the curves $n_L=-1$ and $n_L=-2$ for the emission process and never occurs for $n_L=-4$. This behavior becomes more prominent for $n_H=3$, as shown in Fig. 7, where the different n_L emission curves never cross each other, and therefore no peak switching occurs at all. The reason is due to the effect of the energy shift Q as we just pointed out and shall discuss shortly. For the absorption process, the curves are similar to that in Fig. 5. However, it is interesting to note that, all the curves reverse and decrease when the field E_L increases to higher intensities as shown in Figs. 6 and 7.

As we have pointed out before, all the previous calculations include only the ponderomotive shift due to the low-frequency field and neglect the effect due to the high-frequency field. In our calculation, the energy shifts due to both the high-frequency and low-frequency fields are taken into account. In the following, we shall discuss the relationship of the energy shift with the peak switching just discussed in Figs. 5, 6, and 7. Figure 8 shows the results for the energy shift Q as a function of the field strength E_L for the

FIG. 6. Same as in Fig. 5, but for $n_H=2$ ($w_H=0.4$ a.u.) and $E_H=0.01$ a.u.
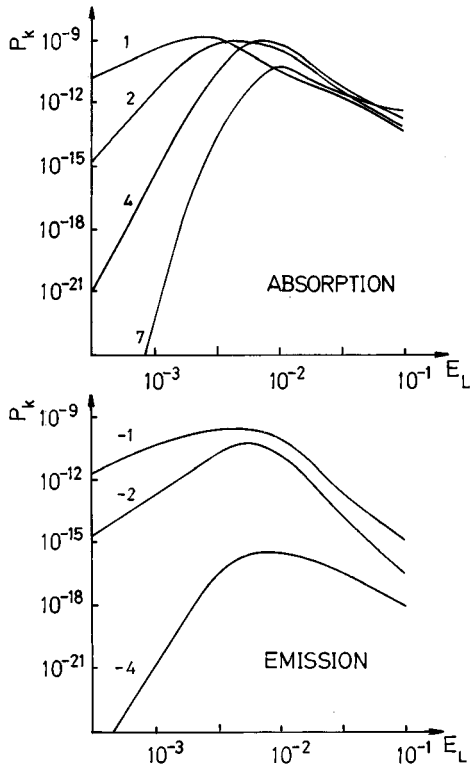
FIG. 7. Same as in Fig. 6, but for $n_H = 3$ ($w_H = 0.25$ a.u.).

case of $n_H = 1$. For the small field strength, Q is small and its effect is negligible, so the lower-order spectra are dominant and no peak switching occurs. But for higher values of E_L ($E_L = 0.01$ a.u.~0.05 a.u.), the magnitude of Q increases quickly and its effect becomes sizable. The peak switching will then occur successively for each higher-order curve at some critical field in this region as shown in Fig. 5. It is worth noting that our energy shift Q, which comes from the $\vec{A} \cdot \vec{p}$ interaction, has different values for different final states. This differs from previous calculations where the
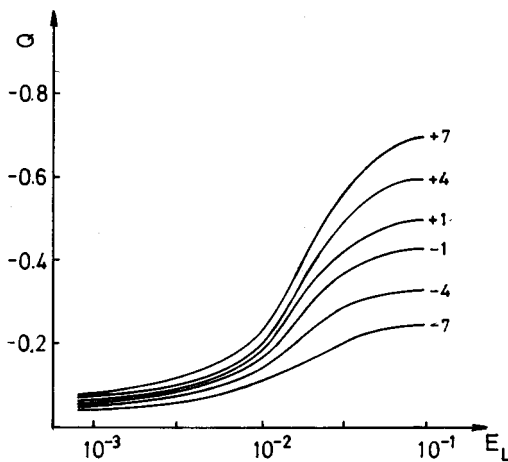

FIG. 8. The energy shift Q as a function of the low-frequency field strength E_L (a.u.) for the different numbers of exchanged photons n_L. The high-frequency field strength $E_H = 0.001$ a.u. and $w_H = 1.0$ a.u., $w_L = 0.04$ a.u.
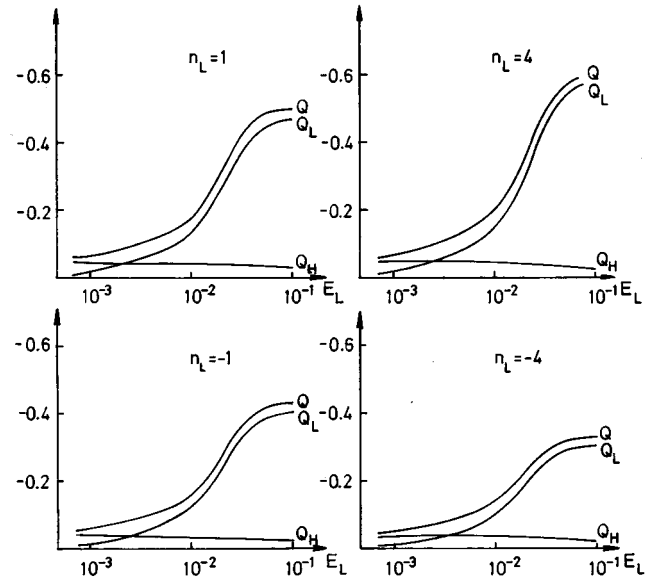

FIG. 9. Same as in Fig. 8, but also includes the energy shifts Q_H and Q_L.

shifts are due to the ponderomotive energy, which increases only with the field strength but remains the same for all states. However, our energy shift Q increases not only with the field strength, but also with the final continuum states (as seen from Fig. 8, at a fixed E_L the tendency of the magnitude of Q to increase with the order of curves is quite fast). Therefore these two effects of increasing Q are combined together to enhance the interesting results obtained in Fig. 5. We have also studied the competition between Q_H and Q_L due to the high-frequency field and the low-frequency field, respectively. Figure 9 shows the results for different multiplicities n_L. It is found that for lower E_L, Q_H is larger than Q_L, while Q_L becomes dominant for the higher field E_L. This is in accordance with our expectations, since 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. For the lower field E_L, it is the ionization of the atom that dominates the process and thus Q_H is larger than Q_L. As the field E_L increases, the repopulation of the final states becomes dominant and then Q_L becomes much larger than Q_H. We have also studied the cases for $n_H = 2$ and $n_H = 3$ with the results shown in Figs. 10 and 11, respectively. In these cases, the energy shifts Q for the absorption processes ($n_L = 1, 4$) increase very rapidly with the field E_L, especially for the higher-order multiplicities so that the higher-order n_L spectra become more important and the curves are eventually all reversed in the high-field region as indicated in Figs. 6 and 7. The situation becomes quite different for the emission processes ($n_L = -1, -4$), in which Q increases slowly for the case $n_H = 2$ (Fig. 10), which leads to only a few peak-switching occurrences as we have also pointed out before in Fig. 6. This behavior becomes even more clear for the case $n_H = 3$ as shown in Fig. 11. It can be seen that the energy shift Q increases much more slowly for the emission processes, especially for higher-order curves such as $n_L = -4$, where Q is almost a constant. Here, the peak switching never occurs and the curves do not cross each other at all as shown
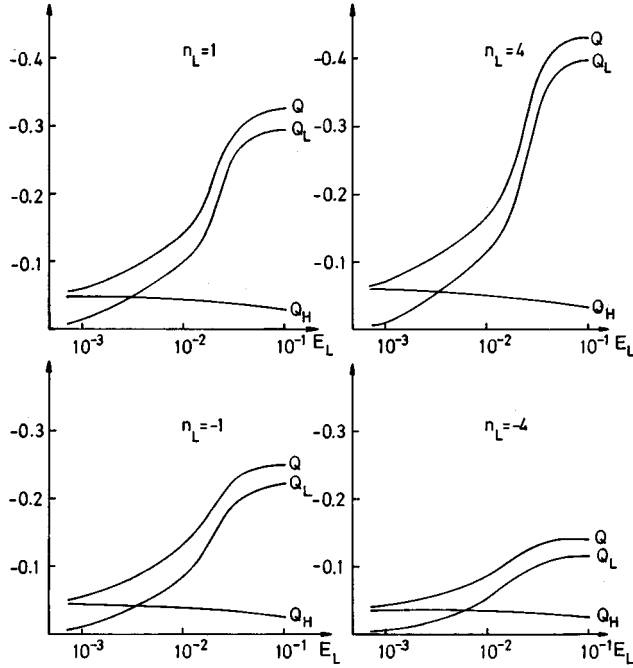
FIG. 10. Same as in Fig. 9, but for $n_H = 2$ ($w_H = 0.4$ a.u.) and $E_H = 0.01$ a.u.

in Fig. 7. Therefore our result shows that the energy shift Q is closely related to the peak switching in the photoelectron spectra, and the correspondence between them seems to be quite consistent.

IV. CONCLUSION

We have studied the two-color above-threshold ionization of atoms by using a nonperturbative method of solving the Schrödinger equation in momentum space. Our method generalizes the problem for cases where an arbitrary number of ionization photons are absorbed, and also takes into account the threshold shifts due to both the high-frequency and the low-frequency fields. We have calculated the photoelectron energy spectra for hydrogen atoms with different field strengths and frequencies, and found that an important effect responsible for the peak switching in the photoelectron spectra is due to the energy shift Q. We found that for the case of $n_H = 1$, the energy shift is small for low laser intensity and the lower-order spectra dominate so that no peaking switch-
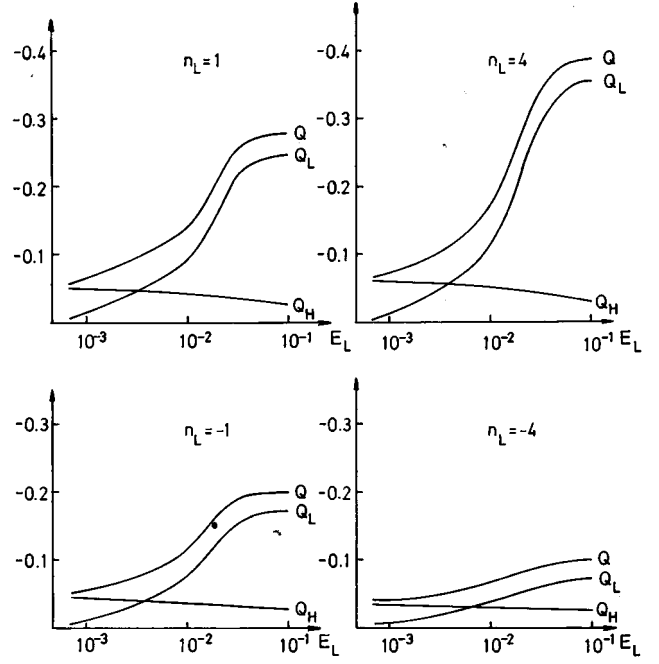

FIG. 11. Same as in Fig. 10, but for $n_H = 3$ ($w_H = 0.25$ a.u.).

ing occurs. As the field strength E_L increases, Q increases quite rapidly, and peak switching will eventually occur at some critical field. We have also investigated the competition between the energy shifts due to the high- and low-frequency fields and found that at the low-field E_L, it is the ionization of the atom that dominates the process so that Q_H is larger than Q_L. When the field E_L increases to higher values, the redistribution of the continuum states then dominates, and Q_L becomes much larger than Q_H. We have also extended the investigation for the cases of $n_H = 2$ and 3. It is found that for the absorption process, Q increases very rapidly with the laser field E_L, and the higher-order multiplicity curves will all be reversed in the high-field region. However the situation differs greatly for the emission process, in which Q increases much more slowly and becomes almost constant for the higher-order curves so that no peak switching occurs at all.

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