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The photoelectron energy spectra of the above-threshold ionization of atomic hydrogen

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Abstract. The above-threshold photoelectron energy spectra of the hydrogen atom in an intense field are calculated by solving the Schrödinger equation in momentum space, which facilitates the extraction of the rapidly varying part of the wavefunction. It is found that the peak switching and the suppression of low-energy peaks in the above-threshold ionization is clearly manifested in this theory. We found that the apparent threshold shifts of the ATI peaks are strongly dependent on the final continuum state. The frequency dependence of the energy spectrum is also investigated.

1. Introduction

One of the most interesting effects investigated in the multiphoton ionization of atoms by an intense laser field is the above-threshold ionization (ATI), where the energy spectrum of the ejected electron consists of a series of peaks centred at energies $\varepsilon_s = (N_0+s)\hbar\omega - I_i$, where s is the above-threshold photon number and I_i is the ionization energy of the initial state (Agostini *et al* 1979, Kruit *et al* 1981, Petite *et al* 1984, Burnett *et al* 1993). Qualitatively speaking, the peaks are due to the absorption of more than the minimum number (N_0) of photons required to ionize the atom. There are many other non-linear effects that manifest themselves in the experimental results of ATI, such as the failure of I^N dependence of the rate of N-photon ionization, the peak-switching of the ATI photoelectron spectra, and the significant suppression of the lowest-order peak with increasing laser intensity. The ATI phenomenon was also found to be strongly dependent on the laser frequency. For the same laser intensity, but higher frequency, the number of observed ATI peaks is small (Fabre *et al* 1982). Recently, a new series of higher order above-threshold ionization peaks in the photoelectron energy spectra was also observed (Rottke *et al* 1993). These striking features in ATI have invoked a great amount of theoretical interest. A perturbative method was first used (Gontier and Trahin 1980, Aymar and Crance 1981, Gao and Starace 1989). However, the observed relative decrease of the lower-order ATI peaks indicates that the lowest-order perturbation treatment is not adequate and should be replaced either by a high-order perturbation calculation or by some non-perturbative methods. Calculations of the latter type were performed recently. They have ranged from the application of Floquet theory (Chu and Cooper 1985, Potvliege and Shakeshaft 1990, Dörr *et al* 1992, Madajczyk *et al* 1992), which takes advantage of the periodicity of the Hamiltonian; the essential-state approach (Bialynicka-Birula 1984, Edwards *et al* 1984, Deng and Eberly 1984, 1985, Yao *et al* 1989, Piraux *et al* 1991), which points to the importance of saturation in continuum–continuum transitions; to the direct numerical solution of the time-dependent Schrödinger

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equation (Kulander 1987a, b, Javanainen *et al* 1988, Collins and Merts 1989, Krause *et al* 1992, Cerjan and Kosloff 1993).

Studying the photoelectron spectra from atoms irradiated by an intense laser field is a useful means of obtaining information about the multiphoton processes taking place. Various characteristics of photoelectron spectrum have been studied and are reasonably understood. Some previous works (Muller *et al* 1983, Mittlemann 1984, Freeman and Bucksbaum 1986) point to the importance of the ponderomotive energy which is the cycle-average kinetic energy due to the electron oscillating in the laser field. Typical values for free electron ponderomotive shifts in a 10^{13} W cm^{-2} laser are around 1.06 eV, compared with the shift of only 0.008 eV for the ground state in hydrogen (Freeman and Bucksbaum 1991). Therefore, the Rydberg and continuum states shift upward, relative to the lower bound states, and produce an increase in ionization threshold approximated by the ponderomotive shift. However, some of the experimental works (Lompre *et al* 1985) claim an absence of the ponderomotive effect. Javanainen and Eberly (1988) have done a numerical study of intense laser photoionization and found that the deviation of the threshold shift from the ponderomotive energy is quite large, up to 20% for a low-frequency field and even larger for the case of a high-frequency field. Milonni and Ackerhalt (1989) pointed out that the confusion that has surrounded the ponderomotive shift is connected with an incorrect treatment of the A^2 term in the Hamiltonian. In their results, they have shown that the threshold shift is due to the $A \cdot p$ interaction, not the ponderomotive energy. Recently, Mulser *et al* (1993) generalized the concept of the ponderomotive potential of a free electron to the atoms in an intense field, and found that the so-called ponderomotive term in the Hamiltonian corresponds to no physical reality. Pan *et al* (1986, 1988, 1989) analysed the effect of a laser field on the ionization potential of atoms and showed that the ponderomotive-potential theory does not describe the apparent threshold shift in the ATI photoelectron energy spectrum. They also calculated the level shift based on the perturbative theory. It was found that the threshold intensity decreases rapidly with the order of perturbation and the result may not correctly describe the situation at high laser intensity. Thus, they conclude that the mechanism that causes the apparent threshold shift is still an open question. The purpose of this paper is to present a systematic calculation of the above-threshold photoelectron energy spectra, and hopefully provide a deeper understanding of this problem.

Recently a new and efficient method of solving the time-dependent Schrödinger equation for a system undergoing multiphoton processes has been introduced (Shakeshaft and Dörr 1988, Shakeshaft and Han 1988). The novel feature of this method is that the Schrödinger equation is solved in the momentum space, which facilitates the extraction of the rapidly varying part of the wavefunction. A preliminary calculation for a one-dimensional system has been performed, and the dynamics of ATI was also briefly investigated (Shakeshaft and Han 1988, Han 1990). In this paper, we shall extend this method to give a detailed calculation for the above-threshold photoelectron spectra of the hydrogen atom, and also discuss the dependence of the spectra on the intensity and the frequency of the laser field.

2. Theory

We consider an electron initially, at $t = 0$, bound by the atomic potential V in the state $|\Phi_i(t)\rangle = |\phi_i\rangle \exp(-iE_i t/\hbar)$, with E_i the initial energy. The time-dependent Schrödinger equation is

$$i\hbar \frac{d}{dt}|\Psi(t)\rangle = \left(\frac{p^2}{2m} + V + H_I(t)\right)|\Psi(t)\rangle \tag{1}$$

where $H_1(t) = -(e/mc)\boldsymbol{A}\cdot\boldsymbol{p}$ is the interaction with the applied electromagnetic field.

For the case with $V = 0$, which is just a free electron moving in the radiation field, the solution to equation (1) was originally derived by Volkov (1935) and can be written in the following form

$$|\chi_k(t)\rangle = \exp\{-\mathrm{i}[E_\mathrm{i}t/\hbar + \theta_k(t)]\}|k\rangle \tag{2}$$

where $|k\rangle$ is the eigenvector of $\boldsymbol{p}$ with momentum eigenvalue $\hbar k$ normalized as

$$\langle r|k\rangle = (2\pi)^{-3/2}\mathrm{e}^{\mathrm{i}k\cdot r} \tag{3}$$

and the real phase $\theta_k(t)$ is given by

$$\theta_k(t) = \frac{1}{\hbar}\int_0^t \mathrm{d}t' \left(\frac{\hbar^2k^2}{2m} - E_\mathrm{i} + H_1(t')\right). \tag{4}$$

For a monochromatic, linearly polarized field $\boldsymbol{A}(t) = \boldsymbol{A}_0\cos\omega t$, we have

$$\theta_k(t) = \frac{1}{\hbar}\left(\frac{\hbar^2k^2}{2m} - E_\mathrm{i}\right)t - \frac{1}{\hbar}\frac{e\boldsymbol{A}_0\cdot\boldsymbol{p}}{mc\omega}\sin\omega t. \tag{5}$$

We shall use the wavefunction $|\chi_k(t)\rangle$ as our basis. The solution for equation (1) can be written as

$$|\Psi(t)\rangle = \int \mathrm{d}\boldsymbol{k}\, a_k(t)|\chi_k(t)\rangle. \tag{6}$$

Therefore, $a_k(t)$ represents, for $t \to \infty$, the amplitude for finding the electron which has escaped from the atomic potential V and is moving freely through the field.

Substituting the expansion of equation (6) into equation (1), an inhomogeneous integro-differential equation can be obtained for the coefficient $a_k(t)$

$$\mathrm{i}\hbar\frac{\partial a_k}{\partial t} = \exp[\mathrm{i}\theta_k(t)]b_k(t) \tag{7}$$

where

$$b_k(t) = \int \mathrm{d}k' \exp[-\mathrm{i}\theta_{k'}(t)]a_{k'}(t)\langle k|V|k'\rangle. \tag{8}$$

Using equation (5), we can formally integrate equation (7) over t,

$$a_k(t) = a_k(0) - \frac{\mathrm{i}}{\hbar}\sum_n J_n(\xi)\int_0^t \mathrm{d}t' \exp\left[\frac{\mathrm{i}}{\hbar}\left(\frac{p^2}{2m} - E_\mathrm{i} - n\hbar\omega\right)t'\right]b_k(t') \tag{9}$$

where $a_k(0)$ can be determined from the initial boundary condition, and we have used the Fourier–Bessel expansion

$$\exp(-\mathrm{i}\xi\sin\omega t) = \sum_n J_n(\xi)\exp(-\mathrm{i}nwt) \tag{10}$$

with

$$\xi = \frac{ep \cdot A_0}{\hbar mc\omega}$$

It has been shown that (Shakeshaft and Dörr 1988, Shakeshaft and Han 1988), because of the phase factor $\exp(i\theta_k(t))$ on the right-hand side of equation (7), the function $a_k(t)$ varies rapidly with both k and t. On the other hand, $b_k(t)$ varies relatively slowly with k and t. Consequently, we can interpolate $b_k(t)$. It has been investigated (Han 1990) that the function $b_k(t)$ has significant effect on the ionization process. By assuming $b_k(t)$ is constant in t, it can be proved that equation (9) reduces to the Keldysh–Faisal–Reiss approximation (Keldysh 1964, Faisal 1973, Reiss 1980, 1990). Thus the KFR approach, which is generally regarded as providing the best model for explainant the ATI, is only a special limiting case of our approach by ignoring the time dependence of the function $b_k(t)$. In this limiting situation, it has also been shown that the threshold shift is due to the ponderomotive energy which is commonly believed responsible for the suppression of the low-energy ATI peaks at high field intensity. This is of course not correct. A realistic treatment of the problem must include the slowly time-varying part in $b_k(t)$. Explicit and implicit methods have been proposed for solving this problem (Shakeshaft and Dörr 1988). In the explicit method, we extrapolate $b_k(t)$ using the Taylor series expansion

$$b_k(t) = b_k(0) + \dot{b}_k(0)t + \cdots. \tag{11}$$

The time derivative of $b_k(t)$ can be obtained from equation (8)

$$\dot{b}_k(t) = \int \mathrm{d}k' \left[-\frac{\mathrm{i}}{\hbar} \dot{F}_{k'}(t) \exp\left(-\frac{\mathrm{i}}{\hbar} F_{k'}(t) \right) a_{k'}(t) + \exp\left(-\frac{\mathrm{i}}{\hbar} F_{k'}(t) \right) \dot{a}_{k'}(t) \right] \langle k|V|k' \rangle$$

where

$$F_k(t) = \left(\frac{\hbar^2 k^2}{2m} - E_\mathrm{i} \right) t - \frac{e\hbar k \cdot A_0}{mc\omega} \sin \omega t.$$

Using equation (7) for $\dot{a}_k(t)$, we obtain

$$\dot{b}_k(t) = -\frac{\mathrm{i}}{\hbar} \int \mathrm{d}k' \left[\dot{F}_{k'}(t) \exp\left(-\frac{\mathrm{i}}{\hbar} F_{k'}(t) \right) a_{k'}(t) + b_{k'}(t) \right] \langle k'|V|k' \rangle. \tag{12}$$

Substituting equation (12) into (11) and to a first-order approximation, we find

$$b_k(t) = b_k(0) \exp(-\mathrm{i}Qt/\hbar) \tag{13}$$

where

$$Q = \frac{1}{b_k(0)} \int \mathrm{d}k' \, [\dot{F}_{k'}(0) a_{k'}(0) + b_{k'}(0)] \langle k|V|k' \rangle. \tag{14}$$

Please note that in the zero-order approximation we keep only the first term in the expansion of equation (11) so that $b_k(t) = b_k(0)$, which is a constant and in this limit our method reduces to the KFR approximation as we pointed out before. Therefore, the result in equation (13) obtained by taking the first-order approximation can be regarded as providing new modifications for the KFR model. The convergence of the expansion

series in equation (11) has also been checked in a preliminary calculation for the previous work (Shakeshaft and Han 1988) where the one-dimensional electron-potential scattering was studied by also including the t^2 term in the Taylor series expansion of the function $b_k(t)$. It was found that inclusion of the t^2 term will not qualitatively change the results obtained by the first-order approximation and the convergence is thus reasonably good. Therefore, we use the first-order approximation in equation (13) so that the computational difficulty involved in the three-dimensional case studied here can be avoided. Substituting equation (13) into (9) and using equation (10), we obtain the ionization amplitude

$$a_k(t) = a_k(0) - \frac{\mathrm{i}}{\hbar}\sum_n J_n(\xi)b_k(0)\int_0^t \mathrm{d}t' \exp\left[\frac{\mathrm{i}}{\hbar}\left(\frac{p^2}{2m} - E_\mathrm{i} - Q - n\hbar\omega\right)t'\right]. \tag{15}$$

This result shows that the ATI peaks occur at electron energies.

$$E_k = \frac{p^2}{2m} = n\hbar\omega - I_\mathrm{i} + Q \tag{16}$$

where $I_\mathrm{i} = -E_\mathrm{i}$ is the ionization energy for the state ϕ_i. Therefore, the ATI peaks are shifted by an amount Q, which is responsible for the peak switching and suppression of the low-energy peaks in ATI.

Table 1. Positions of the ATI peaks as a function of field strength E_0 for $\omega = 0.04$ au. s is the above-threshold photon number. Q is the energy shift. P is the ponderomotive energy.

E_0(au)	s	E_k (eV)	Q (eV)	P (eV)
0.001	0	0.48	−0.06	−0.004
	1	1.41	−0.22	−0.004
	2	2.34	−0.38	−0.004
	3	3.42	−0.39	−0.004
0.005	0	0.21	−0.33	−0.106
	1	1.05	−0.58	−0.106
	2	2.02	−0.70	−0.106
	3	3.10	−0.71	−0.106
0.01	1	0.85	−0.78	−0.42
	2	1.76	−0.96	−0.42
	3	2.81	−1.02	−0.42
	4	3.86	−1.03	−0.42
0.015	1	0.52	−1.11	−0.95
	2	1.51	−1.21	−0.95
	3	2.48	−1.33	−0.95
	4	3.51	−1.37	−0.95

3. Results and discussion

In this section we present the numerical calculation of the above-threshold ionization for the ground state of the hydrogen atom. We have calculated the ionization probability, $|a_k(t \to \infty)|^2$, as a function of photoelectron energies for different laser field strengths.

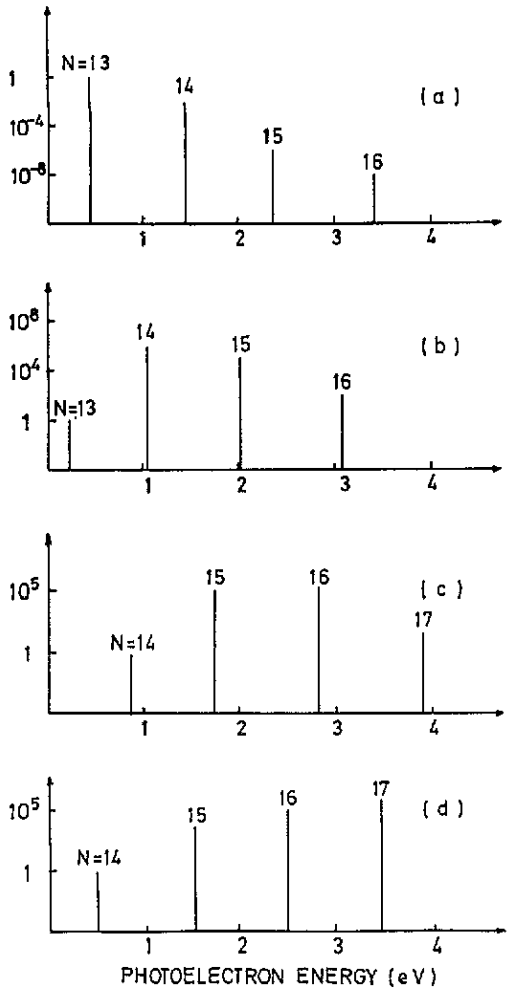

Figure 1. Relative heights of ATI peaks, normalized to the first peak, for $\omega = 0.04$ au. The laser field strength E_0 are (a) 0.001 au, (b) 0.005 au, (c) 0.01 au, (d) 0.015 au.

The expression for $|a_k(t \to \infty)|^2$ can be easily obtained from equation (15) by letting $t \to \infty$. The electronic wave vector k is calculated using equation (16). Since Q is also a k-dependent function, no analytic expression for k can be obtained and a numerical solution of equation (16) for k is carried out. Figure 1 shows the result for those photoelectron energies corresponding to the ATI peaks with $\omega = 0.04$. It is clearly seen that as we increase the magnitude of the field, the peaks shift to lower energies. In addition, the amplitude of the lowest peak diminishes and practically disappears at higher fields. The maximum peak is switched from the first continuum to the second, and then from the second to the third, and so on. Hence, the bell-shaped structure of the photoelectron spectrum, which is a striking feature of ATI is formed; and the highly-lying continuum states successively become most populated. From figure 1, it can also be seen that the centres of peaks are found at positions corresponding to the condition given in equation (16). It is interesting to note that the shifts Q, which come from the $A \cdot p$ interaction, are different from different final states. This result is different from some previous calculations where the shifts are due to the ponderomotive energy, which is the same for all states. As we have pointed out before, the mechanism that causes the apparent threshold shift is still an open question. In our

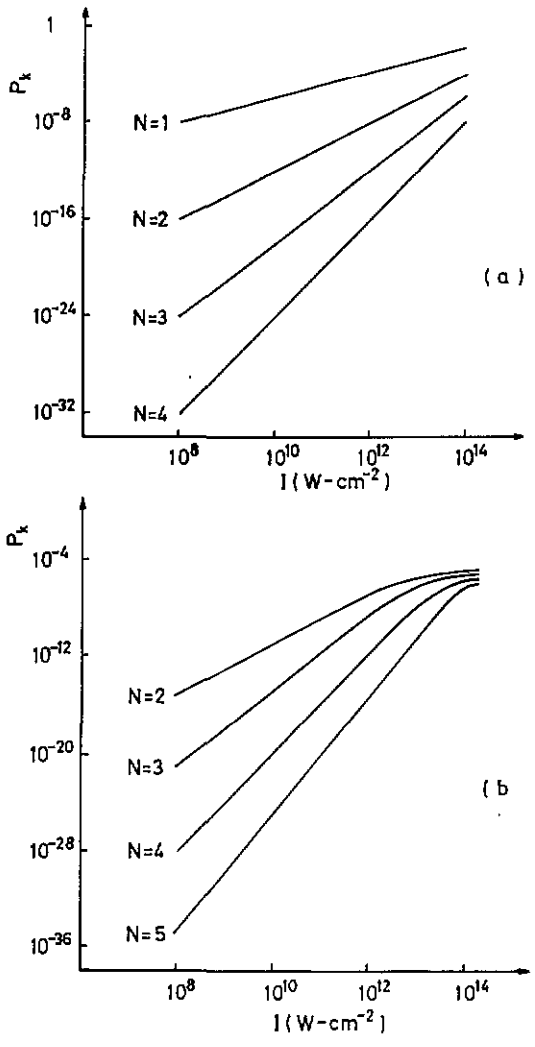

Figure 2. Probability density $|a_k(t \to \infty)|^2$ as a function of field intensity I for frequency (a) $\omega = 0.55$ au and (b) $\omega = 0.28$ au. N is the total number of photons absorbed.

results, the energy shifts are found to have no relevance to the ponderomotive energy but rather are strongly dependent on the final continuum states. Chu and Cooper (1985) have calculated the field-dependent threshold shift for the ground state of the hydrogen atom using the Floquet method, and obtained a similar result to ours in that the threshold shifts are different for different final states. They only calculated the threshold shift of the lowest peaks for high frequency cases and found that the shift is small, but they predicted that it can be quite large for low frequency cases, i.e. for many-photon ionization. From the study on the frequency-dependence of the ATI peaks as shown below, we have actually found that the energy shift Q is small for high frequencies, but becomes large for low frequencies. Therefore, our results are consistent with theirs. In table 1, we present a more quantitative description of these findings. The photoelectron peak energy E_k and the energy shift Q are calculated with different field strengths for $\omega = 0.04$. The ponderomotive energies are also shown in the table for comparison. For a fixed value of field strength the ponderomotive shift is the same for all peaks. However, our energy shifts Q, which depend strongly on the atomic energy state, have different values for different ATI peaks.

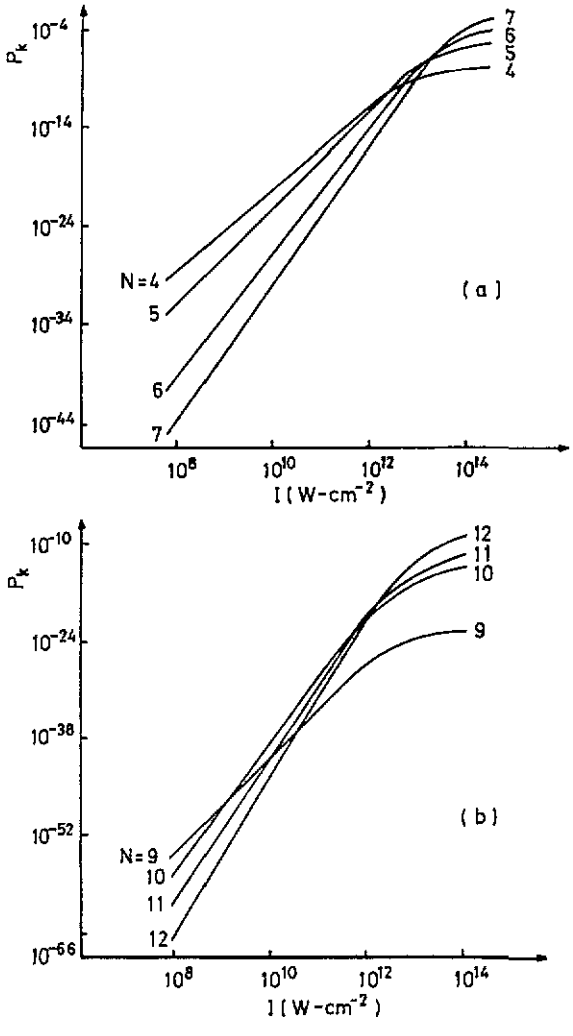

Figure 3. Same as in figure 2, but for frequency (a) $\omega = 0.14$ au and (b) $\omega = 0.06$ au.

We have also studied the frequency dependence of the ATI peaks. The results are shown in figures 2 and 3 for $\omega = 0.55$ au ($N_0 = 1$), 0.28 au ($N_0 = 2$), 0.14 au ($N_0 = 4$) and 0.06 au ($N_0 = 9$). It is found that, at high frequency, the low-energy ATI peaks are dominant and the I^N dependence is recovered. As the frequency is lowered, the high-order peaks begin to increase very rapidly with field intensity, and eventually overcome the lowest peak. As shown in figures 2 and 3, the I^N dependence is clearly observed for the case of $\omega = 0.55$ au ($N_0 = 1$). As ω decreases to 0.28 au ($N_0 = 2$), the curve begins to deviate from the I^N law in the high field region. As the frequency decreases further to $\omega = 0.14$ au ($N_0 = 4$) and $\omega = 0.06$ au ($N_0 = 9$), the higher-order ATI peaks increase very fast with field intensity, and eventually the order of the curves is reversed. We found that the peak inversion occurs at about intensity 0.5×10^{13} W cm^{-2} for $\omega = 0.14$ au, and occurs at a lower intensity 10^{10}–10^{11} W cm^{-2} for $\omega = 0.06$ au. It is interesting to note that the peak inversion is closely related to the energy shift Q. In table 2, we show the values of Q for different frequencies ω and field strengths. It is found that when ω is large, Q is small and thus its effect is negligible. As ω is decreased, Q increases with higher-order peaks and becomes

Table 2. The energy shift Q for different frequencies ω and field strengths E_0. N is the total number of photons absorbed.

ω (au)	E_0 (au)	N	Q (eV)	ω (au)	E_0 (au)	N	Q (eV)
0.55	0.001	1	−0.02	0.14	0.001	4	−0.03
		2	−0.11			5	−0.14
		3	−0.24			6	−0.31
	0.005	1	−0.08		0.005	4	−0.15
		2	−0.23			5	−0.42
		3	−0.46			6	−0.62
	0.01	1	−0.15		0.01	4	−0.42
		2	−0.36			5	−0.85
		3	−0.58			6	−0.98
0.28	0.001	2	−0.02	0.06	0.001	9	−0.06
		3	−0.12			10	−0.18
		4	−0.25			11	−0.32
	0.005	2	−0.11		0.005	9	−0.25
		3	−0.32			10	−0.52
		4	−0.51			11	−0.65
	0.01	2	−0.28		0.01	9	−0.68
		3	−0.53			10	−0.91
		4	−0.79			11	−1.01

comparable to the photon energy $\hbar\omega$. Therefore, the effect due to Q becomes sizable and the peak inversion finally occurs at some critical field. It is also worth noting that the ponderomotive shift in the KFR model used in previous calculations increases only with the field strength but remains the same for peaks of all orders. However, our energy shift Q increases not only with the field strength but also with the order of the peaks (it can be seen from table 2 that the tendency of magnitude of Q to increase with the order of peaks is quite fast). Therefore, these two effects of increasing Q are combined together to give some enhancement for the interesting results obtained in figure 3.

4. Conclusion

Based on the method of solving the Schrödinger equation in momentum space, the photoelectron energy spectrum of the above-threshold ionization of hydrogen atom is calculated for different field strengths and frequencies. The peak switching and the suppression of the low-energy peaks are clearly manifest in our results. It is found that the threshold shift of the ATI peaks, which comes from the $\boldsymbol{A} \cdot \boldsymbol{p}$ interaction, is strongly dependent on the atomic energy state. We found that the shift is small for the lower energy peaks, but becomes quite large for high-order peaks. We have also studied the frequency dependence of the ATI peaks. For high frequency cases, the threshold shift is small, and the I^N law is observed. For low frequency cases, the shift, which increases very fast with both the field intensity and the order of the peaks, becomes comparable with the photon energy and peak inversion will eventually occur at some critical field.

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