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LETTER TO THE EDITOR

Dynamics of the above-threshold ionization of atoms in an intense field

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Abstract. Based on the method of solving the Schrödinger equation in momentum space, which facilitates the extraction of the rapidly varying part of the wavefunction, it can be shown that the Keldysh-Faisal-Reiss approximation is only a limiting situation of the general approach for the strong-field ionization. We have also studied the suppression of the low-energy peaks in the above-threshold ionization and some interesting points are discussed.

Above-threshold ionization (ATI) is the production of higher-energy electrons that have absorbed additional photons over the minimum number (N_0) required for ionization (Agostini *et al* 1979, Kruit *et al* 1981, Petite *et al* 1984). The experimental results for the energy spectra of ejected electrons consist of a series of equally spaced peaks centred at the 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. Various theoretical models have been proposed to account for the observation. The Keldysh-Faisal-Reiss (KFR) approximation (Keldysh 1965, Faisal 1973, Reiss 1980) has generally been regarded as providing the best model for explaining the ATI. Recently, however, some difficulties and questions have been raised regarding the KFR approximation. Javanainen and Eberly (1989) computed numerically the above-threshold ionization electron spectra and found that the KFR model offers an unreliable representation of the ionization process. Milonni (1988) has shown that the Keldysh approximation is questionable under conditions of strong ionization. Mittleman (1989) also pointed out that the KFR theory has some conceptual difficulties since physically equivalent Hamiltonians give radically different results when treated by this method. One purpose of this letter is to present a simple interpretation of the KFR approximation, which is found to be a special limiting case of the general approach for the multiphoton process.

In ATI, another striking feature is the so called 'peak switching'. Above a certain intensity of the field, the first and possibly even the second peaks become smaller than the subsequent peaks and eventually disappear. Some previous works (Muller *et al* 1983, Mittleman 1984, Chu and Cooper 1985, Freeman *et al* 1986) point to the importance of the ponderomotive energy which is the quiver kinetic energy of an electron in the field. 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 for intense laser photoionization and found that the deviation of the threshold shift from the ponderomotive energy is quite large, up to 20% for the lower-frequency field and even larger for the case of a high-frequency field. Pan *et al*

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(1986, 1988, 1989) analyse the effect of a laser field on the ionization potential of atoms and show that the ponderomotive-potential theory does not describe the apparent threshold shift in the ATI photoelectron energy spectrum. They also calculate the level shift based on the perturbation theory. It is 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. 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. Their approach rests on the assumption that the binding potential is sufficiently 'short ranged' that the eigenstate of $p^2/2m+V(r)$ may be taken to be the eigenstates of $p^2/2m$ and $\boldsymbol{p}$. This assumption seems to be oversimplified in the sense that a correct electron final state should account for the joint influence of the radiation and of the Coulomb field. 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). Some preliminary calculations for a one-dimensional system are performed. The results are interesting in that the multiphoton process shows with many peaks in the transition probability function. In this letter, we extend this theory to investigate the dynamics in ATI. In our results, it can be shown explicitly that the suppression of the low-energy peaks in ATI has no relevance to the ponderomotive energy and the mechanism for the apparent threshold shift is clearly manifested in the theory.

We start from the time-dependent Schrödinger equation

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}|\Psi(t)\rangle=\left(\frac{p^2}{2m}+W+H_{\mathrm{I}}(t)\right)|\Psi(t)\rangle \tag{1}$$

where W is the atomic potential and $H_{\mathrm{I}}(t)$ is the interaction with the applied field

$$H_{\mathrm{I}}(t)=-\frac{e}{mc}\boldsymbol{A}(t)\cdot\boldsymbol{p}+\frac{e^2}{2mc^2}A^2(t). \tag{2}$$

Initially, at $t=0$, the electron is bound by the atomic potential W, in the state i represented by $|\Phi_i(t)\rangle=|\phi_i\rangle\exp(-\mathrm{i}E_it/\hbar)$, with E_i the initial energy. Let $|\boldsymbol{k}\rangle$ denote the eigenvector of $\boldsymbol{p}$ with momentum eigenvalue $\hbar\boldsymbol{k}$ normalized so that

$$\langle\boldsymbol{k}'|\boldsymbol{k}\rangle=\delta(\boldsymbol{k}'-\boldsymbol{k}) \tag{3a}$$

$$\int \mathrm{d}\boldsymbol{k}|\boldsymbol{k}\rangle\langle\boldsymbol{k}|=1. \tag{3b}$$

To solve equation (1) we may write

$$|\Psi(t)\rangle=\exp(-\mathrm{i}E_it/\hbar)\int \mathrm{d}\boldsymbol{k}\exp[-\mathrm{i}\theta_{\boldsymbol{k}}(t)]a_{\boldsymbol{k}}(t)|\boldsymbol{k}\rangle \tag{4}$$

where

$$\theta_{\boldsymbol{k}}(t)=\frac{1}{\hbar}\int_0^t \mathrm{d}t'\left(\frac{\hbar^2k^2}{2m}-E_i+H_{\mathrm{I}}(t')\right). \tag{5}$$

It is clear to see that the state $\exp\{-\mathrm{i}[E_it/\hbar+\theta_{\boldsymbol{k}}(t)]\}|\boldsymbol{k}\rangle$ satisfies the time-dependent Schrödinger equation with $W=0$. Therefore, from equation (4), $a_{\boldsymbol{k}}(t)$ represents, for $t\to\infty$, the amplitude for finding the electron which has escaped from the atomic potential W and is moving freely through the field.

Substituting the expansion of equation (4) into equation (1), we obtain an inhomogeneous equation for $a_k(t)$:

$$i\hbar \frac{\partial a_k}{\partial t} = \exp[i\theta_k(t)] b_k(t) \tag{6}$$

where

$$b_k(t) = \int dk' \exp[-i\theta_{k'}(t)] a_{k'}(t) \langle k | W | k' \rangle. \tag{7}$$

Formally integrating equation (6) over t, we obtain

$$a_k(t) = a_k(0) - \frac{i}{\hbar} \int_0^t dt' \exp[i\theta_k(t')] b_k(t') \tag{8}$$

where $a_k(0)$ can be determined from the initial boundary condition. It has been shown (Shakeshaft and Dörr 1988, Shakeshaft and Han 1988) that because of the presence of the phase factor $\exp[i\theta_k(t)]$ on the right-hand side of equation (6), the function $a_k(t)$ varies rapidly in k when t is large, and it varies rapidly in t when k is large. On the other hand, $b_k(t)$ varies relatively slowly in k and t. Consequently, we can interpolate $b_k(t)$. Let us discuss several interesting points.

(i) If $b_k(t)$ is indeed constant in t, from equation (8)

$$a_k(t) = a_k(0) - \frac{i}{\hbar} b_k(0) \int_0^t dt' \exp[i\theta_k(t')]. \tag{9}$$

For the monochromatic, circularly polarized field

$$A(t) = a[\hat{e}_x \cos(\omega t) + \hat{e}_y \sin(\omega t)] \tag{10}$$

we get

$$\theta_k(t) = \frac{1}{\hbar}\left(\frac{\hbar^2 k^2}{2m} - E_i\right) t + \left(\frac{e^2 a^2}{2mc^2}\right) t - \frac{eap_\perp}{\hbar mc\omega} [\sin(\omega t - \psi) + \sin\psi] \tag{11}$$

where $p_\perp = (p_x^2 + p_y^2)^{1/2}$ and $\psi = \tan^{-1}(p_y/p_x)$.

In this case we obtain

$$a_k(t) = a_k(0) - (i/\hbar) b_k(0) \sum_n \exp(in\psi) J_n(\xi) \exp[(-i/\hbar)\xi \sin\psi]$$
$$\times \int_0^t dt' \exp\left[\frac{i}{\hbar}\left(\frac{p^2}{2m} - E_i + P - n\hbar\omega\right) t'\right] \tag{12}$$

where $P = e^2 a^2 / 2mc^2$ is the ponderomotive term and we have used the Fourier-Bessel expansion

$$\exp[-i\xi \sin(\omega t - \psi)] = \sum_{n=-\infty}^{+\infty} J_n(\xi) \exp[-in(\omega t - \psi)] \tag{13}$$

with

$$\xi = \frac{eap_\perp}{\hbar mc\omega}.$$

The result in equation (12) was originally introduced by Keldysh (1964) and later developed by Faisal (1973) and Reiss (1980). Thus the KFR approximation is only a

special case of our approach by considering $b_k(t)$ as constant in t. From equation (12) it follows that the energy spectra of the photoelectron should have peaks at

$$E = p^2/2m = n\hbar\omega - I_i - P \tag{14}$$

where $I_i = -E_i$ is the ionization potential for the state ϕ_i. Therefore the ponderomotive term P gives a threshold shift which is commonly believed responsible for the suppression of the low-energy ATI peaks at high field intensities. However, it should be remembered that the above result is only a limiting situation that neglects the time-dependence of $b_k(t)$. This is of course not correct. A realistic treatment of the problem must include the slowly time-varying part in $b_k(t)$. We discuss the effect of $b_k(t)$ as follows.

(ii) We can rewrite $b_k(t)$ in equation (7) as

$$b_k(t) = \exp[-(\mathrm{i}/\hbar)Pt]B_k(t) \tag{15}$$

where

$$B_k(t) = \int \mathrm{d}\boldsymbol{k}' \exp[-(\mathrm{i}/\hbar)F_{k'}(t)]a_{k'}(t)\langle \boldsymbol{k}|W|\boldsymbol{k}'\rangle \tag{16}$$

$$F_k(t) = (\hbar^2k^2/2m - E_i)t - \frac{eap_\perp}{mc\omega}[\sin(\omega t - \psi) + \sin\psi]. \tag{17}$$

If we temporarily ignore the time dependence in $B_k(t)$, we obtain from equation (8)

$$\begin{aligned} a_k(t) &= a_k(0) - (\mathrm{i}/\hbar)B_k \int_0^t \mathrm{d}t' \exp[\mathrm{i}\theta_k(t')] \exp[-(\mathrm{i}/\hbar)Pt'] \\ &= a_k(0) - \frac{\mathrm{i}}{\hbar} B_k \sum_n \mathrm{e}^{in\psi} J_n(\xi) \exp[-(\mathrm{i}/\hbar)\xi \sin\psi] \int_0^t \mathrm{d}t' \\ &\quad \times \exp\left[\frac{\mathrm{i}}{\hbar}\left(\frac{p^2}{2m} - E_i - n\hbar\omega\right)t'\right]. \end{aligned} \tag{18}$$

This expression, unlike the KFR result, indicates that the ATI peaks have no ponderomotive shift. This is because the ponderomotive force does not affect the atomic states and every state, bound or continuum, should be shifted by exactly the same amount P, and therefore the transition energies cannot be affected by P. In fact, the A^2 term in the Hamiltonian can be removed by a trivial contact transformation (Kroll and Watson 1973) and so it cannot be the source of relative shift of the levels.

(iii) Of course, the correct treatment must also take the time dependence in $B_k(t)$ of equation (16) into account. This will give rise to an additional level shift for the level i. In so doing, we have to solve the integro-differential equations of equations (6) and (7). Explicit and implicit methods have been proposed (Shakeshaft and Dörr 1988) for solving these equations. In the explicit method, we extrapolate $B_k(t)$ using the Taylor series expansion

$$B_k(t) = B_k(0) + \dot{B}_k(0)t + \ldots. \tag{19}$$

The time derivative of $B_k(t)$ can be obtained from equation (16)

$$\begin{aligned} \dot{B}_k(t) = \int \mathrm{d}\boldsymbol{k}' \Big[&-\frac{\mathrm{i}}{\hbar} \dot{F}_{k'}(t) \exp[(-\mathrm{i}/\hbar)F_{k'}(t)]a_{k'}(t) \\ &+ \exp[(-\mathrm{i}/\hbar)F_{k'}(t)]\dot{a}_{k'}(t) \Big] \langle \boldsymbol{k}|W|\boldsymbol{k}'\rangle \end{aligned} \tag{20}$$

where

$$\dot{F}_k(t) = \hbar^2 k^2/2m - E_i - (eap_\perp/mc)\cos(\omega t - \psi). \tag{21}$$

Using equation (6) for $\dot{a}_k(t)$ and equation (15), equation (20) becomes

$$\dot{B}_k(t) = -\frac{\mathrm{i}}{\hbar}\int \mathrm{d}\boldsymbol{k}'[\dot{F}_{k'}(t)\exp[(-\mathrm{i}/\hbar)F_{k'}(t)]a_{k'}(t) + B_{k'}(t)]\langle \boldsymbol{k}|W|\boldsymbol{k}'\rangle. \tag{22}$$

Substituting equation (22) into equation (19) and to a first-order approximation we find

$$B_k(t) \simeq B_k(0)\exp[(-\mathrm{i}/\hbar)P_i t] \tag{23}$$

where

$$P_i \simeq \frac{1}{B_k(0)}\int \mathrm{d}\boldsymbol{k}'[\dot{F}_{k'}(0)a_{k'}(0) + B_{k'}(0)]\langle \boldsymbol{k}|W|\boldsymbol{k}'\rangle. \tag{24}$$

From equation (15), we have

$$b_k(t) = B_k(0)\exp[(-\mathrm{i}/\hbar)(P + P_i)t]. \tag{25}$$

Substituting equation (25) into equation (8) and using equation (11), we can obtain the ionization amplitude

$$a_k(t) = a_k(0) - \frac{\mathrm{i}}{\hbar}B_k(0)\sum_n \mathrm{e}^{\mathrm{i}n\psi}J_n(\xi)\exp[(-\mathrm{i}/\hbar)\xi\sin\psi]\int_0^t \mathrm{d}t'$$
$$\times\exp\left[\frac{\mathrm{i}}{\hbar}\left(\frac{p^2}{2m} + I_i - P_i - n\hbar\omega\right)t'\right]. \tag{26}$$

This result shows that the ATI peaks occur at electron energies

$$E = p^2/2m = n\hbar\omega - I_i + P_i. \tag{27}$$

Therefore, the ATI peaks are shifted by an amount P_i due to the $\boldsymbol{A}\cdot\boldsymbol{p}$ interaction, but not the ponderomotive shift P. It is interesting to note that P_i may be positive or negative depending on the relative magnitudes of the quantities a_k, $\dot{F}_k$ and B_k. When P_i is negative, from equation (27), the effective ionization potential increases, which corresponds to the suppression of the low-energy peaks in ATI. This is the case for the experiments carried out so far. However, when P_i is positive, the ionization potential will be lowered and the low-energy peaks might not be suppressed even at very high intensities. This situation has not been observed and it might correspond to the case with very high field frequency.

In conclusion, based on the method of solving the time-dependent Schrödinger equation in momentum space, it was shown that the KFR approximation is only a limiting situation of the general approach for multiphoton ionization. It seems that the Keldysh approximation is likely to fail for a strong field for which the time dependence in the slowly varying function $b_k(t)$ may become important for the ionization process. We have also derived a more general expression for describing the shift of the low energy peaks in ATI. Calculations are in progress and will be reported later.

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