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LETTER TO THE EDITOR

Coulomb-threshold oscillations and suppression of multiphoton ionization rates

Jerzy Z Kamiński[†]
Physikalisches Institut der Universität Bonn, Nussallee 12, D-5300 Bonn, Federal Republic of Germany

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Abstract. We apply a recently developed quasi-classical expansion for multiphoton processes in strong far-infrared laser fields in order to show that the Coulomb threshold significantly suppresses ionization rates. We also observe a new oscillatory behaviour of differential ionization rate probabilities which is due to the non-analytic behaviour of the Coulomb scattering wavefunction.

Interaction of bound systems, like atoms, molecules or excitons, with intense laser fields is the subject of active experimental and theoretical investigations both in atomic (Agostini and Petite 1988) and solid state physics (Ridley 1982, Combescot and Combescot 1988, Joffre *et al* 1989, Combescot 1990, Kalafati and Kokin 1989). The development of methods that can incorporate a large class of important physical aspects of this interaction and also, very importantly, that can still be tractable numerically, is one of the major problems for theoretical studies. Due to the complexity of many-electron systems, even those without external fields are very difficult to treat, so most of the theoretical methods developed use a one-electron approximation; this approximation is also going to be adopted in the present work. However, even in this case, one has to develop further approximations in order to treat non-perturbatively the action of strong laser fields on matter.

In the case of solid state physics strong modifications of quantum processes by an intense far-infrared laser field have been detected (Ganichev *et al* 1983, 1986). This is due to the fact that the effective coupling of radiation to matter can qualitatively be characterized by a dimensionless parameter $\eta = (I/I_0)(\omega/\omega_0)^{-3}$, where the intensity I and the laser photon energy ω are in units of W cm^{-2} and eV respectively, $I_0 = 3.5 \times 10^{16}$ W cm^{-2} and $\omega_0 = 27.2$ eV (see, e.g., Ganichev *et al* 1986, Kamiński 1990). This means that the softer the laser photons are, the smaller the intensities of radiation necessary to observe the non-perturbative effects that appear for η of the order of one.

In our previous publications (Kamiński 1988, 1990) we have developed a general scheme for non-perturbative studies of multiphoton processes in intense far-infrared laser fields. This method consists in systematic and univocal expansion of transition amplitudes for multiphoton processes with respect to $1/n$, where n is the number of

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[†] On leave of absence from Institute of Theoretical Physics, Warsaw University, Hoża 69, 00-681 Warszawa, Poland.

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laser photons absorbed or emitted by atoms, electrons or other charged carriers. The leading term of this approximation well describes n-photon processes provided that the dimensionless parameter $1/n$ is much smaller than 1. The domain of validity of this expansion scheme has also been discussed.

The present work reports a study of the process of absorption of photons by hydrogen-like quantum systems in solids (like, for instance, centres of the crystal lattice or excitons) subjected to an intense far-infrared laser field in the case when transitions take place from bound states to the scattering ones. We shall focus our attention on hydrogenic systems because in this case the radiationless problem can be solved exactly, which is crucial for the approximation method we are going to apply. Moreover, in order to simplify as much as possible our further considerations, we adopt the effective mass approximation and use units in which the effective mass as well as the strength of the Coulomb potential are equal to one; i.e. we use what can be called the 'atomic units'. However, by the proper scaling of parameters one can easily transform these units into let us say 'excitonic units'; the choice of units is not crucial for the analysis of multiphoton ionization of hydrogenic systems we are going to report in this work.

Keldysh (1965) was the first to study multiphoton ionization of bound systems in the presence of strong radiation. In his well known paper he proposed a non-perturbative approach to this problem, which in principle consists in substitution of the exact wavefunction of ejected electrons by the Volkov wavefunction describing the motion of free electrons in the presence of radiation, i.e. by the wavefunction which neglects the action of the static potential. Certainly, such a substitution is justified for ejected electrons that possess sufficiently large kinetic energy. However, this condition is not satisfied in multiphoton ionization by far-infrared laser fields. An improvement of the Keldysh approach, which accounts for the action of a static potential on ejected electrons, has been presented in Kamiński (1988). The physical interpretation of our approach is that the freed (from a binding static potential) electron follows the changes of the slowly oscillating far-infrared radiation, i.e. its time-dependent effective momentum $\boldsymbol{p}(t)$ equals $\boldsymbol{p}_{\mathrm{av}} - e\boldsymbol{A}(t)$, where $\boldsymbol{p}_{\mathrm{av}}$ is an average momentum and $\boldsymbol{A}(t)$ is the oscillating vector potential of the laser field. For weak fields such a modification causes marginal effects; however, for strong fields we can expect large changes. These expectations are based on the fact that the time-dependent momentum, in a specific geometric configuration, can, from time to time, approach or be equal to zero. On the other hand, it is well known that the Coulomb potential, due to its long-range character, significantly changes radiationless scattering as well as weak-field ionization processes at the threshold (Fano and Rau 1986), i.e. for momenta approaching zero. This is due to the fact that the Coulomb scattering wavefunction is non-analytic at the threshold. Further, by the 'Coulomb threshold' we understand there to be non-analytic behaviour of the scattering wavefunction and other physical quantities for particles moving in a static potential with a Coulomb tail and with energies very close to zero.

The aim of our study is to show that the Coulomb threshold suppresses ionization rates. To this end we have performed a numerical analysis of n-photon ionization probability rates applying the recently developed quasi-classical expansion (Kamiński 1990). The leading term of this expansion for multiphoton ionization processes has already been derived in Kamiński (1988) (see, formulae (40) and (30) with $\psi^{(+)}$ replaced by $\psi^{(-)}$). For the Coulomb potential the radiationless scattering wavefunction is known exactly (see, e.g., the formula (112.7) of Landau and Lifshitz (1958)). This leads us, after applying the effective mass approximation and performing the standard space integration (see, e.g., appendix 7.1 of McDowell and Coleman (1970)) to the following expression for the n-photon ionization probability w_n of a hydrogenic system from its ground state:

$$w_n(\theta,\varphi) = N p_n |A_n(\theta,\varphi)|^2 \tag{1}$$

where N is a constant irrelevant to our further considerations. In the above equation $A_n(\theta,\varphi)$ is a function of the spherical angles that determines the direction of motion of ejected electrons, i.e., the final momentum $\boldsymbol{p}_n$ is equal to $p_n(\boldsymbol{e}_x \sin\theta\cos\varphi + \boldsymbol{e}_y \sin\theta\sin\varphi + \boldsymbol{e}_z\cos\theta)$. This function is given by the following one-dimensional integral:

$$\begin{aligned} A_n(\theta,\varphi) = -8\pi \int_0^{2\pi} \mathrm{d}\phi \exp\Big(\mathrm{i}n\phi - \frac{\mathrm{i}e}{\omega}\int^{\phi} \mathrm{d}\phi'\, \boldsymbol{p}_n\cdot \boldsymbol{A}(\phi') + \ln\Gamma(1-\mathrm{i}\gamma) + \tfrac{1}{2}\pi\gamma\Big) \\ \times \big[-(\mathrm{i}\gamma - 1)e\boldsymbol{A}(\phi)\cdot\boldsymbol{p}_n \xi^{\mathrm{i}\gamma-2}\zeta^{-\mathrm{i}\gamma} + \mathrm{i}\gamma e^2 A^2(\phi)\xi^{\mathrm{i}\gamma-1}\zeta^{-\mathrm{i}\gamma-1} \\ + \tfrac{1}{2}e^2A^2(\phi)(\mathrm{i}\gamma-1)\xi^{\mathrm{i}\gamma-2}\zeta^{-\mathrm{i}\gamma} - \tfrac{1}{2}\mathrm{i}e^2A^2(\phi)\gamma(1-\mathrm{i}|\boldsymbol{p}|)\xi^{\mathrm{i}\gamma-1}\zeta^{-\mathrm{i}\gamma-1}\big] \end{aligned} \tag{2}$$

in which Γ is the Euler gamma function, $\boldsymbol{p} = \boldsymbol{p}_n - e\boldsymbol{A}(\phi)$, $\gamma = 1/|\boldsymbol{p}|$, $\xi = 1 + p_n^2$ and $\zeta = 1 - p_n^2 + 2e\boldsymbol{A}(\phi)\cdot\boldsymbol{p}_n - 2\mathrm{i}|\boldsymbol{p}|$; let me note at this point that due to the ponderomotive potential (see, e.g., the formula (30.9) of Landau and Lifshitz (1960)) $p_n^2 = -1 - \frac{1}{2}\alpha_0^2\omega^2 + 2n\omega$ for a circularly polarized laser field of the form $e\boldsymbol{A}(\phi) = -(\alpha_0\omega/\sqrt{2})(\boldsymbol{e}_x\sin\phi + \boldsymbol{e}_y\cos\phi)$. This is the integral we calculate numerically.

The ionization probability rates are presented in figures 1–3 as functions of $\alpha_0 = (I/I_0)^{1/2}(\omega/\omega_0)^{-2}$. We see that ionization probability rates initially rise monotonically and afterwards start to oscillate. These oscillations can be attributed exactly to the Coulomb threshold. Indeed, as follows from figure 1, for a given n there is the one value of α_0, which we call the critical value α_n^{cr}, around which we observe 'infinitely' rapid oscillations with 'infinitely' small amplitudes. The critical value α_n^{cr} corresponds to the case in which the effective time-dependent momentum $\boldsymbol{p}(\phi) = \boldsymbol{p}_n - e\boldsymbol{A}(\phi)$ is equal to zero. For the circularly polarized laser field this can only happen for $\theta = \pi/2$. However, these rapid oscillations (although not 'infinitely' rapid) also appear for θ in the vicinity of $\pi/2$, as shown in figure 2, because in this case $\boldsymbol{p}(\phi)$ can, for some values of ϕ, be very close to zero. If θ sufficiently differs from $\pi/2$, we do not observe the Coulomb-threshold oscillations. These rapid oscillations can hardly be detected experimentally, because the electromagnetic field generated by lasers does not have a constant intensity. This means that ionization probability rates have to be averaged over stochastic fluctuations of laser-field parameters. Such an average will certainly smooth out rapid oscillations, but slow oscillations may remain. Moreover, it is clear that the averaged ionization probability rate $\langle w_n\rangle$ exhibits the deep minimum for $\langle\alpha_0\rangle = \alpha_n^{\mathrm{cr}}$, sandwiched by the two large maxima. Hence, we see that the Coulomb threshold suppresses ionization probability rates, i.e. leading, to some extent, to the stabilization (or localization) of bound systems. A more detailed numerical analysis will be presented elsewhere, where different polarizations of the radiation will also be considered.

Figure 3 shows the dependence on α_0 of the total ionization probability rate w_{T},

$$w_{\mathrm{T}}(\theta,\varphi) = \sum_n w_n(\theta,\varphi) \tag{3}$$

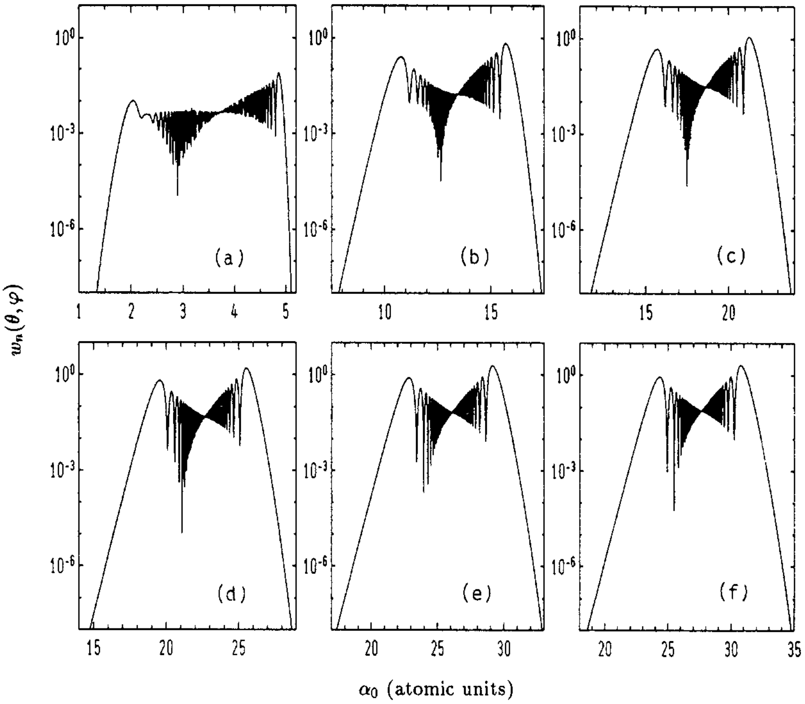

Figure 1. Differential ionization rate probabilities $w_n(\theta,\varphi)$ for $\theta = \frac{1}{2}\pi$, $\varphi = 0$ and for a circularly polarized laser field of frequency $\omega = 0.024$ (in atomic units) as functions of α_0 (in atomic units): (*a*) $n = 21$, (*b*) $n = 23$, (*c*) $n = 25$, (*d*) $n = 27$, (*e*) $n = 29$, (*f*) $n = 30$.

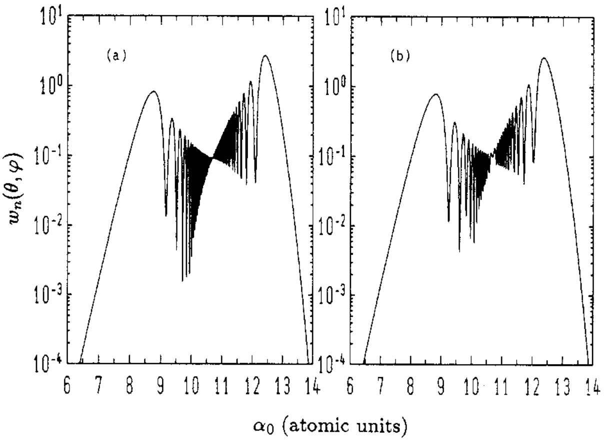

Figure 2. Differential ionization rate probabilities $w_n(\theta,\varphi)$ for $n = 13$, $\varphi = 0$ and for a circularly polarized laser field of frequency $\omega = 0.049$ (in atomic units) as functions of α_0 (in atomic units): (*a*) $\theta = \frac{1}{2}\pi$, (*b*) $\theta = 0.95 \times \frac{1}{2}\pi$.

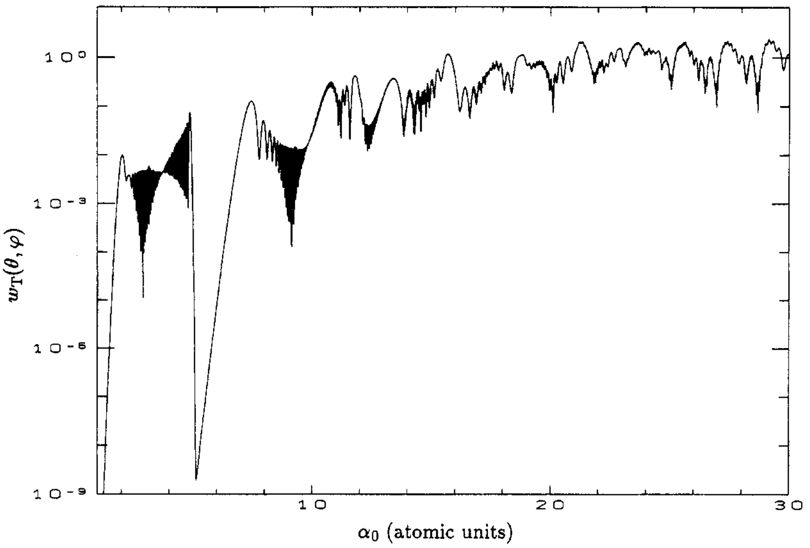

Figure 3. The total differential ionization rate probability $w_T(\theta, \varphi)$ for $\theta = \frac{1}{2}\pi$, $\varphi = 0$ and for a circularly polarized laser field of frequency $\omega = 0.024$ (in atomic units) as a function of α_0 (in atomic units).

where the summation runs over the opened channels, i.e. over those n for which $p_n^2 > 0$. We see that for small α_0 the ionization rate w_T rises initially, and has a deep minimum for $\alpha_0 \approx 5$; however, for large α_0 we observe the saturation of the ionization rate. This finding is, to some extent, in agreement with a recently reported exact numerical analysis of one-dimensional model atoms (Su *et al* 1990).

To summarize, we have shown that the Coulomb threshold causes the suppression and the stabilization of ionization rates. Moreover, the non-analytic character of this threshold leads to the so-called Coulomb-threshold oscillations of w_n as functions of α_0. The existence of these oscillations can be attributed, by means of quantum defect theory (Seaton 1983) to the Rydberg states of bound systems. It also appears that the total ionization rate w_T exhibits 'erratic' behaviour as the function of α_0. This behaviour, however, is due to the superposition of regular Coulomb-threshold oscillations and cannot be prescribed to any sort of chaos. Let me note in closing that a similar 'erratic' behaviour of the ionization probability has been observed in the numerical model simulation (Scharf *et al* 1989). It is, however, questionable whether such behaviour can be observed experimentally because, as has already been mentioned, the laser-field parameters fluctuate and an average over them has to be performed in the end.

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