Resonant and virtual processes of above-threshold ionization of negative ions

I. Yu. Kiyan and V. P. Kraĭnov

Institute of General Physics, Academy of Sciences of the USSR, Moscow
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A theoretical investigation is made of the relative role of two alternative above-threshold ionization processes: a direct multiphoton transition from the ground state to the final state in a continuous spectrum and a cascade (multistage) transition when an atomic electron absorbs the minimum number of photons necessary for the ionization and is scattered in the electromagnetic radiation field absorbing a further photon from this field. The task is reduced to determination of the relative contributions made to the compound matrix element by various intermediate states in the continuum. It is shown that in the case of the zero-range potential the cascade process may be significant when the number of photons participating in the ionization process is small in the limit of high radiation frequencies. The cascade process makes a small contribution to the compound matrix element when the nonlinearity of the ionization process is high, i.e., in this case the cross section of the above-threshold multiphoton ionization cannot be factorized.

1. INTRODUCTION

The phenomenon of the above-threshold ionization of atoms and ions is currently attracting much attention. This phenomenon is interesting because an atomic system is dissociated by a monochromatic electromagnetic field in such a way that an electron is transferred to a continuous spectrum and the energy spectrum of the emitted electrons has several maxima separated from one another by $\hbar\omega$, where ω is the electromagnetic field frequency. These maxima are detectable, of course, only if the electromagnetic radiation intensity is sufficiently high. The maxima appear because an electron continues to absorb photons from the external electromagnetic field even in the continuous spectrum provided there is a third body, which in this case is the atomic core (in the absence of a third body the real absorption of photons is forbidden, because then the laws of conservation of energy and momentum cannot be satisfied simultaneously). In fact, above-threshold ionization occurs also in the case of tunnel ionization of an atom in an alternating field: it can be regarded as the absorption of a very large number of photons followed by the emission of electrons with a very wide energy spectrum.[1]

An analysis of the above-threshold ionization on the basis of a quantum-mechanical theory of transient perturbations involves the use of multiphoton matrix elements of higher order than the lowest that causes the electron being ionized to go over to a continuous spectrum. Such multiphoton matrix elements contain summation over the intermediate atomic states in the discrete spectrum and integration over to the intermediate atomic states in the continuous spectrum.

We shall now consider possible resonances of such matrix elements. The process of summation over the states in the discrete spectrum gives rise to a random resonance, which appears only at a certain frequency of an external electromagnetic field. In the case of summation over the states in the continuous spectrum (i.e., integration), there are no resonances of the usual multiphoton ionization. However, in the above-threshold ionization case there are resonances when the energy of an intermediate state in the continuous spectrum is equal to the sum of the energy of the initial state of the atomic system and the energy of an integral number of photons of an external electromagnetic field.

The present paper deals with the contribution made by these resonances to the magnitude of the multiphoton matrix element governing the above-threshold ionization of an atomic system. We shall consider two alternative situations: 1) the above-threshold ionization is governed primarily by the intermediate states in the vicinity of a resonance; 2) the main role is played by nonresonant intermediate states in the continuous spectrum (together with the intermediate states in the discrete spectrum). In the former case the above-threshold ionization can be regarded as a cascade (multistage) process in which real absorption of above-threshold photons takes place, i.e., the corresponding multiphoton matrix element contains simply the product of one-photon dipole matrix elements between the continuous-spectrum states corresponding to the energies at the resonances. In the latter case the absorption of above-threshold photons is virtual. The tunnel ionization in an alternating field also represents vertical absorption of a large number of photons.

We shall now consider the mathematical difference between the two situations mentioned above.[2] We shall begin with a multiphoton matrix element representing the absorption of one above-threshold photon. The matrix element has a real part corresponding to the integral, in the sense of its principal value for a single resonance in the contnuous spectrum. However, it also has an imaginary part associated with bypassing of a simple pole at the point representing this resonance. If the imaginary part predominates, the process of absorption of an above-threshold photon is of cascade nature, but if the real part of the integral predominates, we can speak of virtual absorption of an above-threshold photon.

The real and imaginary parts of a multiphoton matrix element of the above-threshold ionization of the hydrogen atom were calculated in Ref. 3 on a computer. On the whole, we may conclude that the two parts are of the same order of magnitude, although in the case of different final states (differing in respect of the orbital quantum number) we can have a great variety of specific situations when either the real or the imaginary part predominates. However, the results of Ref. 3 apply only to two specific values of the frequency of the external electromagnetic field. It is shown in Ref. 4 that

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in the case of the hydrogen atom if $\hbar\omega \gg E_n$ (E_n is the energy of the nth term of the hydrogen atom) the real and imaginary parts of a two-photon matrix element are of the same order of magnitude, whereas in the case when $\hbar\omega \sim (1.5-3)E_n$ the imaginary part predominates, i.e., the transition is of cascade nature. Therefore, in the case of the Coulomb potential we cannot draw any unambiguous conclusion about the nature of the above-threshold absorption of photons by an electron in a continuous spectrum.

We shall try to draw specific conclusions about the above-ionization process, based on analytic expressions for the above-threshold ionization probabilities. The analytic problem can be solved for the zero-range potential, which as a rough approximation applies to the above-threshold ionization of negative ions.

A review of the published results on the multiphoton ionization considered on the basis of the zero-range potential can be found in the book of Demkov and Ostrovskiĭ[5] (see information also given in Ref. 6) and we shall assume that the reader is familiar with this topic.

The problem considered here resembles the problem of the scattering of an electron by a negative ion in the field of an external electromagnetic wave; this electron can be forced to absorb or emit wave photons, so that the problem is whether the process is real or virtual. If an electron located near an ion absorbs virtually two photons, then the corresponding two-photon matrix element is dominated by the principal value of the integral. However, if the absorption of two photons occurs consecutively, then the two-photon matrix element is dominated by the imaginary part representing bypassing of a simple pole at an energy equal to the sum of the energy of the initial state and the energy of one photon. This question is not considered in an analysis[7] of the forced absorption and emission of photons in a strong electromagnetic field.

2. TWO-PHOTON ABOVE-THRESHOLD IONIZATION IN ZERO-RANGE POTENTIAL

In the case of the zero-radius potential there is a single bound state $|0\rangle$ with a wave function[5]

$$\psi_0 \propto \exp(-\varkappa r)/r. \tag{1}$$

Here, $\varkappa^2 = -2E_0$ and E_0 is the energy of the bound state. This state has a zero orbital quantum number.

In a continuous spectrum only the s waves differ from plane wares; their scattering phase is

$$\delta_s = -\operatorname{arctg}(p/\varkappa), \tag{2}$$

where p is the electron momentum (we are assuming everywhere that $m = \hbar = 1$).

The interaction of an electron with an external electromagnetic field can be represented by $V = \mathbf{r}\cdot\mathscr{E}$ although a gauge transformation of the interaction $V = \mathbf{pA}/c + \mathbf{A}^2/2c^2$ ($\mathscr{E}$ is the intensity of the electromagnetic field in the electromagnetic wave, $\mathbf{A}$ is the vector potential, and $\mathscr{E} = \dot{\mathbf{A}}/c$) gives the same results for multiphoton matrix elements.

A two-photon matrix element of a transition from the ground state $|0\rangle$ to a state $|p\rangle$ in the continuous spectrum can be written in the form

$$V_{p0}^{(2)} = \int \frac{V_{pp'}V_{p'0}\,dp'}{E_{p'} - E_0 - \omega - i\varepsilon}, \qquad \varepsilon \to +0. \tag{3}$$

Here, $V_{pp'}$ and $V_{p'0}$ are one-photon matrix elements and $E_{p'} = p'^2/2$. Since $E_0 + \omega > 0$, the integrand in Eq. (3) has a simple pole at $p'^2 = 2(E_0 + \omega)$ (see the Introduction). Consequently, Eq. (3) can be represented in the form

$$V_{p0}^{(2)} = \pi i \frac{V_{pp'}V_{p'0}}{2[2(E_0+\omega)]^{1/2}}\,\delta(p' - [2(E_0+\omega)]^{1/2}) + \mathrm{P}\int \frac{V_{pp'}V_{p'0}\,dp'}{E_{p'} - E_0 - \omega}. \tag{4}$$

The first term on the right-hand side of Eq. (4) represents a contribution to the two-photon matrix element made by a resonant intermediate state $|p'\rangle$ in the continuous spectrum. The second term, representing the principal value of the integral, represents the contribution of the rest of the continuum. The problem of the role of resonances in the continuous spectrum discussed in the Introduction reduces to which of the terms in Eq. (4) is the dominant one.

Two-photon transitions in the d and s states are generally possible from the ground state of the zero-radius potential with zero orbital momentum. In the case of both channels the initial $|0\rangle \to |p'\rangle$ is the same; the matrix element $V_{p'0}$ of this transition is well known[5]:

$$V_{p'0} \propto \mathscr{E}p'/(p'^2 + \varkappa^2)^2. \tag{5}$$

In the case of transitions from the intermediate p' state to the final s and d states the matrix element $V_{pp'}$ is of the following form (with allowance for the fact that the s wave has a nonzero phase, see above):

$$V_{pp'}(p \to d) \propto \mathscr{E}\left\{\frac{2}{pp'}\frac{d}{dp}\delta(p'-p) - \frac{4}{p^3}\delta(p'-p)\right\}, \tag{6}$$

$$V_{pp'}(p \to s) \propto \frac{\mathscr{E}}{(p^2+\varkappa^2)^{1/2}} \times \left\{\frac{\pi\varkappa}{2}\frac{d}{dp'}\left(\frac{\delta(p-p')}{pp'}\right) + \left[\frac{p'}{(p'^2+p^2+i\varepsilon)^2} + \frac{p'}{(p'^2-p^2-i\varepsilon)^2}\right]\right\}, \qquad \varepsilon \to +0. \tag{7}$$

In view of the condition $p' = p$, the terms in these expressions (containing the delta function and its derivative) contribute only to the principal value of the integral in Eq. (4) [second term on the right-hand side of Eq. (4)], since the conditions $p' = p = [2(E_0 + 2\omega)]^{1/2}$ and $p' = [2(E_0 + \omega)]^{1/2}$ in the first term on the right-hand side of Eq. (4) are obviously incompatible.

Therefore, in the case of circular polarization of an external electromagnetic field (*spd* transition) the whole contribution to the compound matrix element comes not from the resonant intermediate state, but from the final state in the continuous spectrum. In other words, the pole approximation is invalid and the transition is not of cascade nature.

This conclusion can also be understood qualitatively. In the case of a cascade transition the probability of an *spd* transition can be factorized. However, the probability of a one-photon *pd* transition in the continuous spectrum is zero since the p and d waves are not distorted by the zero-range potential and a free electron cannot absorb photons from a monochromatic electromagnetic wave. Therefore, the *spd* transition can only be virtual.

We can see that the delta function does not vanish at the point $p'=p$, which differs from the point $p'=[2(E_0+\omega)]^{1/2}$, where the denominator of the integrand in the second term on the right-hand side of Eq. (4) vanishes. The compound matrix element is then of the form

$$V_{p0}^{(2)}(spd) \propto \mathscr{E}^2(2\omega+E_0)/\omega^4. \tag{8}$$

If the field is linearly polarized, we can expect not only the *spd* transition, but also an *sps* transition. In the latter case it is somewhat more difficult to calculate the compound matrix element, because it contains not only the delta-function term from Eq. (7), but also the following integral:

$$I=\int_0^\infty \frac{p'^3dp'}{[{}^1/_2p'^2-(\omega+E_0)-i\varepsilon'](p'^2+\varkappa^2)^2} \times\left[\frac{p'}{(p'^2-p^2+i\varepsilon)^2}+\frac{p'}{(p'^2-p^2-i\varepsilon)^2}\right]. \tag{9}$$

Here, $\varepsilon\to+0, \varepsilon'\to+0$ are the rules for bypassing poles. We can use the form (3) for the two-photon matrix element.

The integral of Eq. (9) is easily calculated by the theory of residues. Consequently, the two-photon matrix element of the *sps* transition is

$$V_{p0}^{(2)}(sps) \propto \frac{\mathscr{E}^2}{\omega^{1/2}}\left[2^{1/2}i\frac{(E_0+\omega)^{3/2}}{\omega^4}+\varkappa\frac{\omega+E_0}{\omega^4}\right]. \tag{10}$$

The imaginary term in square brackets of Eq. (10) determines the contribution of the resonant intermediate state with an energy $E_p=E_0+\omega$ to the *sps*-transition probability. We can see from Eq. (10) that the role of the resonant states in the continuous spectrum is the one-photon threshold, i.e., at the energy $\omega+E_0\ll|E_0|$ is small. However, if $\omega\gg|E_0|$, the intermediate resonance states make [as follows also from Eq. (10)] a dominant contribution to the probability of the *sps* transition. In fact, in this case we cannot say that the transition becomes of cascade nature, since the *sps*-transition probability becomes comparable with the probability of the *spd* transition.

We can understand Eq. (10) qualitatively as follows. In the case of the *sps* transition we find that, in contrast to the *spd* transition considered above, the matrix element can be factorized, since the *s* wave is distorted by the zero-range potential and the probability of the one-photon *ps* transition in the continuous spectrum differs from zero.

It is interesting to note that the probability of the two-photon above-threshold *sps* transition is zero when $\omega=|E_0|$, i.e., when the first photon corresponds exactly to the limit of the continuous spectrum. However, at this energy the probability of the above-threshold two-photon ionization corresponding to the *spd* transition differs from zero, as demonstrated by Eq. (8).

3. PROCESS OF ABOVE-THRESHOLD IONIZATION IN THE CASE OF A HIGH DEGREE OF NONLINEARITY

The expressions obtained above for the two-photon matrix element can be used also to find the probability of multiphoton above-threshold ionization of a negative ion in the case of one-photon above-threshold absorption (when the field frequency is $\omega\ll|E_0|$). The process of absorption of the minimum (necessary for the ionization) number of photons K is then replaced by the absorption of one photon of frequency $\Omega=K\omega$. We shall now consider the validity of this approximation.

We shall use $V_{ik}=z_{ik}\mathscr{E}$ to denote a dipole matrix element of an ik transition (and assume, to be specific, that the field is linearly polarized). The above-threshold $(K+1)$-photon matrix element is then

$$V_{0p}^{(K+1)}=\sum_{kln\ldots}\frac{V_{0k}V_{kl}V_{ln}\ldots V_{p'p}}{(E_k-E_0-\omega)(E_l-E_0-2\omega)\ldots(E_{p'}-E_0-K\omega)}. \tag{11}$$

It can be rewritten formally in the form

$$V_{0p}^{(K+1)}=\sum_{p'}\frac{V_{0p'}^{(K)}V_{p'p}}{(E_{p'}-E_0-\Omega)}, \tag{12}$$

where V is the matrix element of the K-photon process and $\Omega=K\omega$. The approximation consists in the replacement of $V_{0p'}^{(K)}$ by the one-matrix element $V_{0p'}$ of the field with an effective intensity $\mathscr{E}'$. The question is when this replacement is valid.

According to Eq. (5), if $p'\ll\varkappa$, which follows from the multiphoton nature of the process, we find that

$$V_{0p'}\propto\mathscr{E}'p'/\varkappa^4. \tag{13}$$

It follows from Ref. 6 that the multiphoton matrix element has the same dependence on p' if the final state $|p'\rangle$ is characterized by the orbital quantum number $l=1$. The value of K must then be odd and, moreover, the above-threshold state $|p\rangle$ should have an orbital quantum number $l=0$, so that the selection rules indicate that the $|p'\rangle$ state must have only $l=1$. The channels characterized by an increase in l in the course of absorption of a large number of photons are "suppressed" in the multiphoton ionization probability, but their low statistical weight is due to a high centrifugal barrier.[1] Naturally, this discussion cannot be regarded as a rigorous proof that the replacement in question is justified.

We are thus dealing now with a two-photon transition involving first the absorption of a photon with a high frequency Ω and then with a low frequency ω ($\Omega\gg\omega, \Omega+E_0>0$). The expressions for the corresponding two-photon matrix elements can be obtained by analogy with the considered case of photons of the same frequency ω. The results are then as follows:

$$V_{p0}^{(2)}(spd) \propto \mathscr{E}\mathscr{E}'(\Omega+E_0+\omega)/\Omega^2\omega^3 \tag{14}$$

and

$$V_{p0}^{(2)}(sps) \propto \frac{\mathscr{E}\mathscr{E}'}{\Omega^{1/2}}\left[2^{1/2}i\frac{(\Omega+E_0)^{1/2}}{\omega^2\Omega^2}+\varkappa\frac{(\Omega+E_0-\omega/2)}{\omega^2\Omega^2}\right]. \tag{15}$$

Equations (14) and (15) are simplified by dropping terms of higher order of smallness relative to ω/Ω, so that the above expressions for the $\omega=\Omega$ case cannot be derived from them.

It should be pointed out that, with the exception of the imaginary part of Eq. (15), which corresponds to a cascade transition through a resonant intermediate state, the other terms in Eqs. (14) and (15) are due to the contribution of the final state to the compound matrix element. In this case the multiphoton ionization process can be regarded as con-

sisting of a one-photon transition to the final state followed by the absorption of photons of the same energy from an external electromagnetic field. This justifies qualitatively the above replacement of the multiphoton matrix element with the one-photon element.

It follows from Eqs. (14) and (15) that the probability of a cascade transition in the continuous spectrum through its resonant intermediate state is always negligible compared with the probability of the direct multiphoton process. It therefore follows that the approximation of cascade absorption of above-threshold photons in the continuous spectrum is not justified in the case of the Coulomb or the zero-range potentials.

CONCLUSIONS

We shall conclude by finding the limits of validity of perturbation theory when applied to the problem in hand. We shall find the relationship between the probabilities of the above-threshold and normal ionization processes. The probability of the one-photon ionization by a field $\mathscr{E}'$ of frequency Ω is (see Eq. (5))

$$W^{(1)} \propto \mathscr{E}'^2 (\Omega + E_0)^{1/2} / \Omega^4. \tag{16}$$

If we assume that $\Omega + E_0 \sim \omega$ and divide the two-photon transition probability deduced from Eqs. (14) and (15) by Eq. (16), we obtain the criterion of the validity of perturbation theory in the case of the zero-range potential:

$$W^{(2)}/W^{(1)} \sim \mathscr{E}^2/\omega^3 \ll 1. \tag{17}$$

It was derived earlier in Ref. 6.

This criterion is not identical with the criterion of a multiphoton transition in the Keldysh theory[8]:

$$1/\gamma^2 = \mathscr{E}^2/2\omega^2 E_0 \ll 1. \tag{18}$$

Here, γ is the adiabatic parameter. The reason may be that this theory is incorrect as a result of the absence of the gauge invariance of the transition amplitudes. This problem is discussed in greater detail in Ref. 9.

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[1]L. P. Rapoport, B. A. Zon, and N. L. Manakov, *Theory of Multiphoton Processes in Atoms* [in Russian], Atomizdat, Moscow (1978), p. 140.
[2]N. B. Delone, S. P. Goreslavsky, and V. P. Krainov, J. Phys. B **16**, 2369 (1983).
[3]Y. Gontier, N. K. Rahman, and M. Trahin, Europhys. Lett. **5**, 595 (1988).
[4]S. I. Marko, Thesis for Candidate's Degree [in Russian], Voronezh State University (1985).
[5]Yu. N. Demkov and V. N. Ostrovskiĭ, *The Zero-Range Potential in Atomic Physics* [in Russian], Leningrad State University (1975).
[6]N. L. Manakov and A. G. Faĭnshteĭn, Zh. Eksp. Teor. Fiz. **79**, 751 (1980) [Sov. Phys. JETP **52**, 382 (1980)].
[7]F. V. Bunkin, A. E. Kazakov, and M. V. Fedorov, Usp. Fiz. Nauk **107**, 559 (1972) [Sov. Phys. Usp. **15**, 416 (1973)].
[8]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].
[9]V. P. Kraĭnov, *The Atom in an Extremely Strong Optical Field* (*Lecture*) [in Russian], Engineering-Physics Institute, Moscow (1989).

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