

Ionization of inner atomic shells during resonance excitation of the valence electron

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The Auger effect from inner shells, which arises from resonance excitation of the valence electron of an atom by a weak electromagnetic wave, is discussed. The mechanism leading to an Auger transition is based on the residual Coulomb interaction between the valence electron and the core electrons. On the assumption that the wave field is switched on adiabatically, the probability of the Auger effect of the inner electrons of the atom is determined.

1. INTRODUCTION

The excitation of an electron shell arising in an atom can be removed in various ways, including through a nonradiative transition (Auger effect from inner shells). Various types of Auger processes arising from relaxation transitions in the electron shells of atoms have been closely studied both theoretically and experimentally (see, for example, the survey in Ref. 1).

In the presence of an external electromagnetic wave, Auger transitions acquire several characteristic features. In the last few years, experimental² and theoretical³⁻⁵ studies have dealt with the effects of multiphoton excitation and ionization from the inner shells of many-electron atoms in the field of a strong electromagnetic wave. In this field, a large group of electrons of the outer shells participate in collective motion, and excite the core electrons through the Coulomb interaction.

Also of interest is a situation in which in the initial state of an atom located in the field of a weak electromagnetic wave, no vacancy is created in advance in its electron shell, and the quantum energy and wave intensity are insufficient for direct ionization from inner shells. In the case of resonance excitation of the valence electron by an external wave, relaxation to the ground state may be accompanied by spontaneous emission or by transfer of the wave energy to an electron of the inner shell. This transfer is due to the residual Coulomb interaction of the valence electron and an inner electron. Multiple repetition of this process may also lead to ionization of inner electrons.

In Ref. 6, an expression was derived for the polarization potential, which acts on inner electrons in the course of resonance excitation of the valence electron. This potential was found by the psi-function formalism of a two-level system, and the probability of multiphoton ionization from the inner shells was estimated to be vanishingly small.

As will be shown here, the probability of the Auger effect from inner shells is much smaller than the probability of spontaneous relaxation between the resonance levels of the valence electron. Therefore, in the time necessary for ionization from an inner shell, the atom can be excited repeatedly by the field of the external wave to an upper state, then undergo relaxation to a lower one. In view of this fact, inclusion of dissipative processes, which assumes the use of the density matrix method for the calculations, is essential. When dissipative interactions are neglected, the states of the valence

electron in the field of a resonance wave are satisfactorily described by the well known functions of the Rabi problem.⁷

2. STATEMENT OF THE PROBLEM

We consider an alkali metal atom in the field of a quasisynchronous electromagnetic wave with a given law governing the switching-on of the electric field strength, $\mathbf{E}(t)$. The interaction of the valence electron with the wave will be described by the operator ($\hbar = c = 1$)

$$V(\mathbf{r}, t) = -eE(t)(\mathbf{e}\mathbf{r})\cos\omega t, \quad (1)$$

where \mathbf{r} is the radius vector of the electron and \mathbf{e} is the unit vector of polarization of the wave (below, a wave linearly polarized along the z axis will be considered: $\mathbf{e} = \mathbf{e}_z$).

We assume that when $t \rightarrow -\infty$, the field is switched off continuously, and when $t \rightarrow +\infty$, the field strength amplitude reaches a steady-state value E_0 .

We consider the case of single-photon resonance, when for the valence electron the field frequency ω is close to the transition frequency: $\omega_{21} + \Delta$ and $|\Delta| \ll \omega_{21}, \omega$.

In addition, we assume that the frequency detuning Δ is smaller than the field width $\Gamma_f \sim d_{21}E_0$, where d_{21} is the dipole matrix element of the transition in a two-level system.

The most appropriate method of describing the effect considered in this work is the approach using the density matrix formalism. In this approach, the Coulomb interaction of the valence electron with a core electron is described by the operator

$$\begin{aligned} V_c(\mathbf{r}_1, t) &= e \int \rho(\mathbf{r}_2, t) \frac{e}{|\mathbf{r}_1 - \mathbf{r}_2|} dV_2 \\ &= \int \rho_{21}(t) \psi_1^{(0)*}(\mathbf{r}_2) \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_2^{(0)}(\mathbf{r}_2) dV_2, \end{aligned} \quad (2)$$

where $\rho_{21}(t)$ is the nondiagonal element of the density matrix, calculated on the basis of the eigenfunctions $\psi_1^{(0)}$ and $\psi_2^{(0)}$ of the two-level system, and \mathbf{r}_1 and \mathbf{r}_2 are the radius vectors of the Auger electron and valence electron, respectively. We note that only the off-diagonal element of the density matrix, essential for the effect discussed, has been left standing in Eq. (2).

The equations describing the evolution of the density matrix contain the resonance interaction between the atom and the wave, together with the operator corresponding to dissipative processes.⁸

The interaction (2) will be taken into account in accordance with perturbation theory. This approach implies the satisfaction of several criteria which must be met by the main parameters of the problem. Thus, the amplitude of the field strength E_0 of the wave must be large enough that the field width Γ_f can be on the order of the spontaneous width Γ_s . As will be shown, the condition $\Gamma_f \sim \Gamma_s$ is optimal in terms of the probability of the Auger effect from the inner shell. On the other hand, the field E_0 must not be too high, so that the ionization probability of the valence is less than the probability of the Auger effect.

In addition, inclusion of the interaction (2) in accordance with perturbation theory assumes that the mixing amplitude of the resonance states in the external field is substantially greater than the mixing amplitude due to the residual Coulomb interaction with the Auger electron. Fulfillment of this condition imposes a certain upper bound on the allowed magnitude of the detuning Δ . A criterion for the detuning Δ can be obtained, for example, from a comparative estimate of the diagonal corrections of the second approximation to the eigenfunctions of the system. The mixing amplitude of the resonance states by the external field is determined by the parameter $\alpha \approx (\Gamma_f/\Delta)^2$. A quantity having a similar meaning and arising from the residual Coulomb interaction of the valence electron and Auger electron is given by the parameter

$$\alpha_c = \int \frac{\left| \left\langle 1p \left| \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \right| 2i \right\rangle \right|^2}{[\varepsilon_p - (E_i^{(0)} + \omega)]^2} \frac{d\mathbf{p}}{(2\pi)^3}, \quad (3)$$

where $\varepsilon_p = p^2/2m_e$ is the energy of the Auger electron in the continuous spectrum; $E_i^{(0)} = -I_0$ is the energy of this electron in the initial bound state (I_0 is the energy of ionization from an inner shell).

The matrix element in Eq. (3) is given by the expression

$$\begin{aligned} & \left\langle 1p \left| \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \right| 2i \right\rangle \\ &= \iint e^{-i\mathbf{p}\mathbf{r}_1} \psi_1^{(0)*}(\mathbf{r}_2) \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_2^{(0)}(\mathbf{r}_2) \psi_i^{(0)}(\mathbf{r}_1) dV_1 dV_2, \end{aligned} \quad (4)$$

where the function $\psi_i^{(0)}(\mathbf{r}_1)$ describes the initial state of the Auger electron.

The calculation of the integral in Eq. (4) is appreciably simplified if one takes into consideration the comparative sizes of the outer shell and inner shells of the atom. The characteristic distances along r_1 and r_2 in Eq. (4) are determined by the radii of the corresponding shells: for r_2 , it is the radius of the valence shell of the atom, and for r_1 , the radius of the inner shell. As a rough estimate, one can assume that the radius of the ionizable shell is $r_s \sim a_0 Z_s \text{ Ry}/I_0$, where $a_0 = \hbar^2/m_e e^2$ is the first Bohr radius of the hydrogen atom; Z_s is the effective charge of the core for the shell from which the Auger transition takes place; and $\text{Re} = m_e e^4/2\hbar^2 = 13.6 \text{ eV}$.

It follows from the above estimate that for the electrons of inner shells $r_1 \ll r_2$, and in the dipole approximation the matrix element in Eq. (4) has the form

$$\begin{aligned} \left\langle 1p \left| \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \right| 2i \right\rangle &\approx - \int e^{-i\mathbf{p}\mathbf{r}_1} e\mathbf{r}_1 \psi_i^{(0)}(\mathbf{r}_1) dV_1 \\ &\times \int \psi_1^{(0)*}(\mathbf{r}_2) \nabla \left(\frac{e}{r_2} \right) \psi_2^{(0)}(\mathbf{r}_2) dV_2. \end{aligned} \quad (5)$$

Calculation using Ehrenfest's theorem and the conditions $I_0 \gg \omega$ of the integrals in Eq. (5) leads to the following estimate of the amplitude:

$$\alpha_c \approx \left(\frac{m_e \omega_{21}^2 a_0^2 \text{ Ry}}{I_0^2} \right)^2 \sim \left(\frac{I \text{ Ry}}{I_0^2} \right)^2,$$

where I is the binding energy of the valence electron.

From the estimates obtained, we have the following condition for the magnitude of the ratio of the field width to the frequency detuning

$$\Gamma_f/\Delta > I \text{ Ry}/I_0^2. \quad (6)$$

In a real situation, it is not difficult to satisfy the inequality (6).

3. PROBABILITY OF THE AUGER EFFECT FROM INNER SHELLS OF THE ATOM

We consider the initial problem of the Auger effect from an inner shell in second-order perturbation theory in the residual Coulomb interaction between the valence electron and Auger electron. The results thus obtained make it possible, on the one hand, to show the general structure of the expression for the transition amplitude, and on the other hand, to obtain a relative estimate in comparison with the amplitude of direct photoionization to second order in the interaction with the external field $\mathbf{E}(t)$. Incidentally, we note that in writing the final expression for the probability of the Auger effect from an inner shell of the atom, we shall use the basic results of Ref. 9 for direct multiphoton ionization of an atom in the homogeneous field of an external wave. We recall that there, the multiphoton ionization effect is chiefly determined by the perturbation of the photoelectron wave function in the continuum. In the present work, a similar approach is used, whereby the continuum functions of the Auger electron appear in the intermediate states of a composite matrix element.

For the time evolution of the density matrix elements, we confine ourselves to the simplest case, in which the transverse T and longitudinal τ relaxation terms in the two-level system are equal: $T = \tau$. When the condition $\Delta < \Gamma_f$ is satisfied, and the field is switched on adiabatically, the nondiagonal element of the density matrix is given by the expression⁸

$$\rho_{21}(t) = \frac{1}{2} \frac{r_0 T}{1 + (r_0 T)^2} e^{-i\omega t + \lambda t}, \quad (7)$$

where $r_0 = d_{21} E_0$, and the parameter λ is included in the law $\exp(\lambda t)$ of adiabatic switching-on of the wave field ($\lambda \rightarrow +0$). The solution (7) was obtained in the approximation $T/\tau_s \ll 1$, where $\tau_x = 1/\lambda$ is the characteristic time at which the field is switched on.

The amplitude of the Auger transition in second-order perturbation theory in the residual Coulomb interaction of a core electron with the valence electron is

$$a_{fi}^{(2)}(t) = \frac{1}{4} \left[\frac{r_0 T}{1 + (r_0 T)^2} \right]^2 \frac{\exp[i(\varepsilon_p - E_i^{(0)} - 2\omega - i\lambda)t]}{\varepsilon_p - E_i^{(0)} - 2\omega - i\lambda} \times \int \frac{\langle p1 | \frac{e^2}{r_{12}} | p12 \rangle \langle p1 | \frac{e^2}{r_{12}} | 2i \rangle}{\varepsilon_{p_1} - E_i^{(0)} - \omega - i\lambda} d\mathbf{p}_1 / (2\pi)^3. \quad (8)$$

As follows from Eq. (8), the condition $r_0 T \sim \Gamma_f / \Gamma_s \sim 1$ is optimal from the standpoint of the magnitude of the probability of the Auger effect. In the limiting case of a very weak external field, when we have $r_0 T \ll 1$, the probability is vanishingly small. In the opposite limiting case $r_0 T \gg 1$ (this condition can be interpreted as the absence of relaxation in the two-level system: $T = \tau \rightarrow \infty$), the probability of the Auger effect in our formulation vanishes. This result can be explained as follows. In the framework of the psi-function formalism, a two-level system in a resonance field is generally described by the superposition of two quasienergy functions Ψ_+ and Ψ_- , which become the wave functions of the corresponding unperturbed states of the system when the field is switched on.⁷ The functions Ψ_+ and Ψ_- form a complete system of wave functions, and in the compound matrix element of the transition probability amplitude, summation over virtual states that include both functions is necessary. It is easy to ascertain that interference of the individual components of the sum changes the probability amplitude to zero in the approximation in which the Rabi problem is solved. A result different from zero is obtained when small corrections $\pm \Omega$ ($\Omega = 1/2 [\Delta^2 + r_0^2]^{1/2}$ being the Rabi frequency) to the quasienergies of intermediate states are taken into account in the energy denominators. As a result, the transition probability amplitude in second-order perturbation theory, calculated with the aid of psi-functions, is found to be proportional not to $(r_0/\Omega)^2$, but to the substantially smaller factor $r_0^2/\Omega(I_0 - \omega)$. It goes without saying that this situation also remains true in perturbation theory of higher orders, so that the small quantity Ω is included in the amplitude denominators only once together with the large energy factors $I_0 - \omega, I_0 - 2\omega, \dots$.

The matrix elements in the composite matrix element of the expression (8) are given by the equations

$$\langle p1 | \frac{e^2}{r_{12}} | 2i \rangle = \iint e^{-i\mathbf{p}_1 \cdot \mathbf{r}_1} \psi_1^{(0)*}(\mathbf{r}_2) \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_2^{(0)}(\mathbf{r}_2) \psi_i^{(0)}(\mathbf{r}_1) dV_1 dV_2, \quad (9)$$

$$\langle p1 | \frac{e^2}{r_{12}} | p12 \rangle = \iint e^{-i\mathbf{p}_1 \cdot \mathbf{r}_1} \psi_1^{(0)*}(\mathbf{r}_2) \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_2^{(0)}(\mathbf{r}_2) e^{i\mathbf{p}_1 \cdot \mathbf{r}_1} dV_1 dV_2.$$

To understand the essence of further approximation, we perform in the matrix element $\langle p1 | e^2/r_{12} | p12 \rangle$ the integration with respect to the relative radius vector $\mathbf{r}_1 - \mathbf{r}_2$. We thus obtain

$$\langle p1 | \frac{e^2}{r_{12}} | p12 \rangle = - \frac{4\pi e^2}{|\mathbf{p} - \mathbf{p}_1|^2} \int e^{-i(\mathbf{p} - \mathbf{p}_1) \cdot \mathbf{r}} \psi_1^{(0)*}(\mathbf{r}) \psi_2^{(0)}(\mathbf{r}) dV. \quad (10)$$

To get an estimate, we confine ourselves to the case of the Auger effect at the ionization threshold, when $p \approx 0$. A combined analysis of the expression (10) and of the energy denominator in the amplitude (8) indicates that the main contribution to the magnitude of the integral over $d\mathbf{p}_1$ in Eq. (8) is due to the range of values $p_1 \lesssim 1/a_0$. In this range of values of intermediate momentum and in view of the fact that $I \ll I_0$, the quantity ε_{p_1} can be neglected in the propagator (8). This makes it possible, in estimating the amplitude, to use the convolution theorem in the 8 composite matrix element. In this approximation, the integration with respect to $d\mathbf{p}_1$ in Eq. (8) leads to the following expression for the composite matrix element:

$$\int \psi_1^{(0)*}(\mathbf{r}_2) \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_2^{(0)}(\mathbf{r}_2) \psi_i^{(0)*}(\mathbf{r}_2') \frac{e^2}{|\mathbf{r}_1 - \mathbf{r}_2'|} \psi_2^{(0)}(\mathbf{r}_2') \psi_i^{(0)}(\mathbf{r}_1) dV_1 dV_2 dV_2'.$$

Since the psi-function of the Auger electron in the initial $\psi_i^{(0)}(\mathbf{r}_1)$ is present here in the integral, further calculation permits the use of the dipole approximation in the Coulomb interaction of the electrons. Finally, we obtain the following expression for the transition amplitude:

$$a_{fi}^{(2)}(t) = \frac{\exp[i(\varepsilon_p - E_i^{(0)} - 2\omega - i\lambda)t]}{\varepsilon_p - E_i^{(0)} - 2\omega - i\lambda} \frac{1}{I_0 - \omega} \int (\mathbf{e} \cdot \mathbf{E})^2 \psi_i^{(0)}(\mathbf{r}) dV, \quad (11)$$

where the quantity

$$\mathbf{E} = \frac{1}{2} \frac{r_0 T}{1 + (r_0 T)^2} \left\langle 1 \left| \nabla \left(\frac{e}{r} \right) \right| 2 \right\rangle = \frac{m_e \omega_2}{2e} \frac{r_0 T}{1 + (r_0 T)^2} \langle 1 | z | 2 \rangle | \mathbf{e}_z \rangle \quad (12)$$

has the meaning of amplitude of the effective field which the valence electron exerts on the Auger electron [in the derivation of Eq. (12), the nature of the polarization of the external wave is taken into account, and the approximate equality $\omega_{21} \approx \omega$ is used].

The amplitude of direct photoionization from an inner shell of the atom in second-order perturbation theory in the field $\mathbf{E}(t)$ is given by the expression

$$\tilde{a}_{fi}^{(2)}(t) = \frac{\exp[i(\varepsilon_p - E_i^{(0)} - 2\omega - i\lambda)t]}{\varepsilon_p - E_i^{(0)} - 2\omega - i\lambda} \frac{1}{4\omega} \int e^{-i\mathbf{p} \cdot \mathbf{r}} (\mathbf{e} \cdot \mathbf{E}_0)^2 \psi_i^{(0)}(\mathbf{r}) dV. \quad (13)$$

For photoionization at the threshold ($p \approx 0$), we obtain the following estimate from Eqs. (11) and (13):

$$a_{fi}^{(2)}(t) / \tilde{a}_{fi}^{(2)}(t) \sim \frac{\omega}{I_0 - \omega} \left(\frac{E}{E_0} \right)^2 \sim \frac{\omega}{I_0 - \omega} \left(\frac{I}{\Gamma_s} \right)^2 \quad (\text{for } \Gamma_f \approx \Gamma_s). \quad (14)$$

As is evident from the relation (14), the resonance excitation of the valence electron by the field of a weak wave leads to an appreciable strengthening of the effect of multiphoton ionization from the inner shell.

Using the concept of effective field in higher-order perturbation theory, we obtain an expression for the transition probability amplitude that agrees with the analogous amplitude of the Keldysh problem, expanded as a series in the interaction of the electron with the external wave. We then obtain the following expression (in ordinary units) for estimating the probability of the Auger effect per unit times:⁹

$$w \approx A \omega (I_0/\hbar\omega)^{3/2} \exp [2\langle x \rangle I_0/\hbar\omega + 1 - I_0/\hbar\omega] \cdot (1/2\tilde{\gamma})^{2(I_0/\hbar\omega + 1)}, \quad (15)$$

where A is a numerical factor of order unity; $\langle x \rangle$ signifies an integer-valued part of x ; and the parameter

$$\tilde{\gamma} = \frac{2(2m_e I_0)^{1/2}}{m_e \omega z_{12} r_0 T / [1 + (r_0 T)^2]} \quad (16)$$

is analogous to the adiabaticity parameter γ of Ref. 9.

For the subsequent numerical estimates, along with the expression (15), we shall give an expression for the probability of resonance ionization $w^{(r)}$ of an atom from the ground state through an intermediate level. In the case of single-photon resonance, when $n_s = 1$ and $e^2 E_0^2 \alpha / 4\hbar\omega \ll 1$ hold (α being a coefficient determining the Stark shift of the resonance level in the external field), we have¹⁰

$$w^{(r)} \approx \frac{1}{4} \frac{\Gamma_f^2}{\Delta^2 + \Gamma_f^2/4} w, \quad (17)$$

where w is the probability of direct ionization from the resonance level, described by a formula analogous to (15), with the corresponding values of the adiabaticity parameter γ and binding energy I of the valence electron in the resonance state.

4. DISCUSSION OF RESULTS; CONCLUSION

As an example, we consider the potassium atom ($5P_{1/2} \rightarrow 4S_{1/2}$ transition, $\hbar\omega = 3.07$ eV), for which the matrix elements of the corresponding dipole transition are known.¹¹ The intrinsic width of the $5P_{1/2}$ level is $\Gamma_s \approx 1.1 \times 10^{-7}$ eV. The field width $\Gamma_f = (1/2)eE_0 z_{12}$ for the resonance transition turns out to be $\Gamma_f \approx 0.65 \times 10^{-8} E_0$ eV and becomes of order of the intrinsic width at low field strength, $E_0 \approx 17$ V/cm.

One more fact, which sets an additional lower bound on the allowable values of E_0 , should be taken into consideration. The observation conditions should ensure that both the Doppler and collisional broadenings of the atomic levels be small compared to the field width: $\Gamma_D, \Gamma_C < \Gamma_f$. To decrease the effects associated with the Doppler width, the experiment should be performed with sufficiently well collimated atomic beams, the direction of the wave being transverse to the beam. At small beam divergence angles $\theta = d/2l$ (d being the channel diameter, and l , the channel length), the transverse temperature of the atoms T_{\perp} is related to the longitudinal temperature $T_{\parallel} \approx T$ as follows: $T_{\perp} \approx T\theta^2$. For $T = 10^{-3}$ K, $l = 1$ m, and $d = 1$ cm, the Doppler width is

$\Gamma_D \approx 5 \times 10^{-8}$ eV, and thus, the field strength $E_0 \approx 17$ V/cm is sufficient to satisfy the inequality $\Gamma_f > \Gamma_D$.

Having used Eq. (16), we obtain an estimate of the value of the parameter γ , which determines the ionization from the $3P$ shell of the potassium atom, and the number of wave quanta $K_0 = \langle I_0/\hbar\omega + 1 \rangle$ necessary for ionization from this shell. Near exact resonance (for $\Delta < \Gamma_f$), for the values used in this work ($I_0 \approx 17$ eV, $\hbar\omega = 3.07$ eV, $z_{12} \approx 6^{1/2} a_0$), the parameter $\tilde{\gamma}$ in the optimal regime $\Gamma_f \approx \Gamma_s$ is $\tilde{\gamma} \approx 16.6$. The number of quanta is $K_0 = 6$.

Substituting the numerical values given above into Eq. (15), we find for the probability of the Auger effect from the $3P$ shell

$$w^{(3P)} \approx 20 \text{ sec}^{-1}.$$

Similarly, for the probability of ionization of the valence electron by a wave field of strength $E_0 \approx 17$ V/cm, we obtain with the aid of the expression (17)

$$w^{(4S)} \approx 20 \text{ sec}^{-1}.$$

A comparison of the two probabilities indicates that in a weak field with strength $E_0 \approx 17$ V/cm, the condition $w^{(3P)} \approx w^{(4S)}$ applies.

The effect of ionization from an inner shell can be observed by recording the UV photons ($\hbar\omega \approx 13$ eV). We also note that the ratio of the probabilities of the Auger effect and ionization of the valence electron is determined by the factor

$$w^{(3P)}/w^{(4S)} \sim [\gamma^K/\tilde{\gamma}^{K_0}]^2$$

(K being the number of wave quanta required for ionization from the $4S$ level), which depends very strongly on the magnitude of the field strength E_0 . The probability of the Auger effect has a maximum at field strength $E_0 \approx 17$ V/cm ($\Gamma_f \approx \Gamma_s$), and the probability of ionization of the valence electron increases continuously as a function of E_0 ($\propto E_0^{2K}$).

It follows from the estimates obtained above that the exit velocity of K^+ ions acted upon by a laser wave of strength $E_0 \approx 17$ V/cm reaches a value of 10^{11} ion/cm³ sec (at a beam density of 10^{10} cm⁻³).

We neglected the Coulomb interaction of the Auger electron in the continuous spectrum with a hole in an inner shell of the ion core. As was shown in Ref. 12, inclusion of this interaction leads to an appreciable increase in ionization probability per unit time.

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