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Laser-induced modifications of selection rules in the Auger effect

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The theory of the Auger effect is extended to include the modifications due to the presence of a strong laser field. The laser makes possible the simultaneous ejection of electrons into many channels, each characterized by the number of exchanged photons. The laser also introduces a new physical axis, which reduces the geometrical symmetry of the process. An estimate is given for the limit of weak fields.

In recent years ionization of atoms and molecules by laser light has received increasing attention both theoretically and experimentally. This interest stems from various facts both fundamental and applied. One of them is that bound-free elementary transitions involve both discrete and continuous states and in principle may be used as a physical mechanism on which to base spectroscopic analysis. Along this line of thought it has been pointed out[1] that laser-assisted Compton scattering by atoms can serve as a technique for measuring the distribution of electronic momenta more precisely as compared to the field-free case.

Among the bound-free processes, the Auger effect in the presence of a laser does not seem to have received any attention although it can be considered one of the most interesting because in its dynamics a crucial role is played by the electron correlation. As is well known[2] an atom ionized in a inner shell undergoes an Auger transition when it reorganizes its electron configuration to attain a lower energy by filling the vacancy with an outer electron. The excess of energy then is imparted to a second electron (Auger electron) which is ejected, leaving the atom doubly ionized.

Here we present an outline of the theory of the Auger effect in the presence of external coherent electromagnetic radiation. It will be shown that the presence of the field introduces a relaxation of the usual selection rules which, in turn, induces important modifications in the angular distribution of the ejected electron that can be easily measured. The atom is approximated by two active electrons moving in the potential of all other electrons assumed to participate only as spectators. To be specific, for the sake of example, we consider an alkali-metal atom with a vacancy in the $1s$ state and we will take the valence electron as one of the two active electrons. Though the laser modifies both bound and free wave functions of the active electrons, for the latter the modifications are expected to be far more significant.

The starting point is the Hamiltonian of the atom interacting with a classical electromagnetic field in the dipole approximation taken in the $\mathcal{E}\cdot\mathbf{r}$ gauge:

$$\mathcal{H}(t)=H_0(t)+e\mathcal{E}(t)\cdot\mathbf{r}_1+e\mathcal{E}(t)\cdot\mathbf{r}_2+e^2/r_{12}\,, \qquad (1)$$

where $\mathcal{E}(t)=\mathcal{E}_0\sin\omega t$ is the laser electric field and ω its frequency; e^2/r_{12} is the Coulomb interaction between the two active electrons and is responsible for the atomic rearrangement, while in H_0 we gather all the kinetic and interaction terms not explicitly included in (1).

The exact form of the S matrix of the process is

$$S_{fi}=-i/\hbar\langle U_f(\mathbf{r}_1,\mathbf{r}_2,t)\,|\,e^2/r_{12}\,|\,\Psi_i(\mathbf{r}_1,\mathbf{r}_2 t)\rangle\,, \qquad (2)$$

the brackets $\langle\cdots\rangle$ indicating integration over time and over $\mathbf{r}_1$ and $\mathbf{r}_2$. In (2) Ψ_i is a solution of the full Schrödinger equation

$$\left[i\hbar\frac{\partial}{\partial t}-\mathcal{H}(t)\right]\Psi_i(\mathbf{r}_1,\mathbf{r}_2,t)=0 \qquad (3)$$

and $U_f(\mathbf{r}_1,\mathbf{r}_2,t)$ the solution of

$$\left[i\hbar\frac{\partial}{\partial t}-[\mathcal{H}(t)-e^2/r_{12}]\right]U_f(\mathbf{r}_1,\mathbf{r}_2,t)=0\,. \qquad (4)$$

As no exact forms of Ψ_i and U_f are known, approximations must be resorted to.

Several methods have been devised in the literature to deal with the laser modifications of the wave functions of the bound electrons[3] but in what follows we will assume that the field is weak enough to leave the atomic states undressed. This assumption is actually not strictly necessary and is made here only for the sake of simplicity since it does not change the essence of the reported effect. Bearing this in mind, if we call $u_0(\mathbf{r})=R_0(r)Y_{00}(\Omega)$ the valence wave function with energy E_0 and $u_a(\mathbf{r})$ the second atomic wave function with energy E_a, the initial state can be written as

$$\Psi_i(\mathbf{r}_1,\mathbf{r}_2,t)=\frac{1}{\sqrt{2}}[u_a(\mathbf{r}_1)u_0(\mathbf{r}_2)+su_a(\mathbf{r}_2)u_0(\mathbf{r}_1)]\times\exp[-i/\hbar(E_a+E_0)t]\,. \tag{5}$$

According to the parity of the initial configuration, s is ± 1.

Usually, in the Auger effect, the outgoing electron has kinetic energy of the order of keV and in this case its wave function is expected to be little affected by the residual ion, while in the stage of ejection it strongly interacts with the laser field. It will be shown later that the modifications in the angular distributions of the Auger electrons affect this interaction significantly. Then, the free-electron wave function is taken in the form of a nonrelativistic Volkov state:

$$\chi_{\mathbf{q}}(\mathbf{r},t)=\exp\left\{i\left[\mathbf{q}+\frac{e\mathcal{E}_0}{\hbar\omega}\cos\omega t\right]\cdot\mathbf{r}-\frac{i}{2m\hbar}\left[\left[\hbar^2q^2+\frac{e^2\mathcal{E}_0^2}{2\omega^2}\right]t+\frac{2e\hbar}{\omega^2}\mathbf{q}\cdot\mathcal{E}_0\sin\omega t+\frac{e^2\mathcal{E}_0^2}{4\omega^3}\sin 2\omega t\right]\right\}. \tag{6}$$

If the laser field is absent, the Volkov state goes over the time-dependent plane wave.

The final wave function will be

$$U_f(\mathbf{r}_1,\mathbf{r}_2,t)=\frac{1}{\sqrt{2}}[u_g(\mathbf{r}_1)\chi_{\mathbf{q}}(\mathbf{r}_2,t)+su_g(\mathbf{r}_2)\chi_{\mathbf{q}}(\mathbf{r}_1,t)]\exp\left[-\frac{i}{\hbar}E_gt\right] \tag{7}$$

with the bare ground-state wave function: $u_g(\mathbf{r})=R_g(r)Y_{00}(\Omega)$ and E_g its energy. The instantaneous energy of the free electron in the field is still buried in $\chi_{\mathbf{q}}(\mathbf{r},t)$. Using the expressions

$$e^{iz\cos\phi}=\sum_k i^kJ_k(z)e^{ik\phi}\,,\qquad e^{iz\sin\phi}=\sum_k J_k(z)e^{ik\phi}\,, \tag{8}$$

and the integral representation of the Bessel function $J_k(z)$,

$$J_k(z)=\frac{1}{2\pi}\int_{-\pi}^{\pi}\exp(-ik\theta+iz\sin\theta)d\theta\,, \tag{9}$$

the ionization probability per unit of solid angle is found as

$$\frac{dP}{d\Omega_{\mathbf{q}}}=\frac{1}{(2\pi)^4}\frac{m}{\hbar^3}\sum_{N=-N_{\min}}^{\infty}|I(N,\mathbf{q}_N)|^2\mathbf{q}_N\,, \tag{10}$$

where

$$\frac{\hbar^2q_N^2}{2m}=E_a+E_0-E_g-\frac{e^2\mathcal{E}_0^2}{4m\omega^2}+N\hbar\omega \tag{11}$$

is the kinetic energy of the outgoing electron after having changed N laser photons, and obeying the condition $\hbar^2q_N^2/2m\geq 0$ from which $N_{\min}$ can be obtained. And

$$I(N,\mathbf{q}_N)=\int_{-\pi}^{\pi}d\theta\tau(\theta,N,\mathbf{q}_N)\,, \tag{12}$$

$$\tau(\theta,N,\mathbf{q}_N)=X(N,\theta,\mathbf{q}_N)T(\mathbf{q}_N,\theta)\,, \tag{13}$$

$$X(N,\theta,\mathbf{q}_N)=\exp\left[-iN\theta-i\frac{e}{m\omega^2}\mathbf{q}_N\cdot\mathcal{E}_0\cos\theta-i\frac{e^2\mathcal{E}_0^2}{8m\hbar\omega^3}\sin 2\theta\right], \tag{14}$$

$$T(\mathbf{q}_N,\theta)=T_1(\mathbf{q}_N,\theta)+sT_2(\mathbf{q}_N,\theta), \tag{15}$$

$$T_1(\mathbf{q}_N,\theta)=\left(u_g(\mathbf{r}_1)\Lambda_{\mathbf{q}_N}(\theta,\mathbf{r}_2)\left|\frac{e^2}{r_{12}}\right|u_a(\mathbf{r}_1)u_0(\mathbf{r}_2)\right), \tag{16}$$

$$T_2(\mathbf{q}_N,\theta)=\left(u_g(\mathbf{r}_1)\Lambda_{\mathbf{q}_N}(\theta,\mathbf{r}_2)\left|\frac{e^2}{r_{12}}\right|u_a(\mathbf{r}_2)u_0(\mathbf{r}_1)\right). \tag{17}$$

Here the round brackets $(\cdots)$ indicate integrations over $\mathbf{r}_1$ and $\mathbf{r}_2$, $\Lambda_{\mathbf{q}_N}(\theta,\mathbf{r}_2)$ stands for the spatial part of a plane wave modulated by the presence of the laser:

$$\Lambda_{\mathbf{q}_N}(\theta,\mathbf{r})=\exp\left[i\mathbf{q}_N\cdot\mathbf{r}+\frac{ie}{\hbar\omega}\mathcal{E}_0\cdot\mathbf{r}\sin\theta\right]. \tag{18}$$

It is mainly the presence of this modulation in the plane wave which introduces relevant modifications on the selection rules usually arising when dealing with the field-free matrix elements which are the counterparts of $T_1(\mathbf{q}_N,\theta)$ and $T_2(\mathbf{q}_N,\theta)$. In fact the procedure to carry out the integrals implicit in (16) and (17) consists in expanding the plane wave in eigenfunctions of the angular momentum:

$$\Lambda_{\mathbf{q}_N}(\theta,\mathbf{r}_2)=(4\pi)^2\sum_{l=0}^{\infty}\sum_{m=-l}^{l}\sum_{l'=0}^{\infty}\sum_{m'=-l'}^{l'}i^{l+l'}Y_{l'm'}^{*}(\mathcal{E}_0)Y_{l'm'}(\Omega_2)Y_{lm}^{*}(\mathbf{q})Y_{lm}(\Omega_2)j_l(q_Nr_2)j_{l'}\left[\frac{e\mathcal{E}_0r_2}{\hbar\omega}\sin\theta\right] \tag{19}$$

and the relative distance between the electrons:

$$\frac{1}{r_{12}}=\begin{cases}\dfrac{4\pi}{r_1}\displaystyle\sum_{\lambda\mu}\frac{1}{2\lambda+1}\left(\frac{r_2}{r_1}\right)^{\lambda}Y^*_{\lambda\mu}(\Omega_1)Y_{\lambda\mu}(\Omega_2), & r_2<r_1,\\[2ex] \dfrac{4\pi}{r_2}\displaystyle\sum_{\lambda\mu}\frac{1}{2\lambda+1}\left(\frac{r_1}{r_2}\right)^{\lambda}Y^*_{\lambda\mu}(\Omega_1)Y_{\lambda\mu}(\Omega_2), & r_1<r_2.\end{cases} \tag{20}$$

Inserting (19) and (20) in (16) and (17), integrals of the general form $\int d\Omega Y^*_{l,m}(\Omega)Y^*_{l',m'}(\Omega)Y_{\lambda\mu}(\Omega)Y_{LM}(\Omega)$ result, which are evaluated with the known techniques of composition of spherical harmonics and application of the triangle conditions.[4] In the absence of the laser, $l'=0$ and $m'=0$ and the usual field-free matrix elements are retrieved. The presence of $Y_{l',m'}(\mathcal{E}_0)$ introduces into the problem a preferential physical axis, let us say the z axis, which strongly modifies the selection rules in (16) and (17) which usually force several angular integrals to vanish. As an example, we give an explicit form for $T_1(\mathbf{q}_N,\theta)$ when $u_a=R_a(r)Y_{10}(\Omega)$, and $u_0=R_0(r)Y_{00}(\Omega)$:

$$T_1(\mathbf{q}_N,\theta)=\frac{4\pi e^2}{\sqrt{3}}\sum_{l=0}^{\infty}(-i)^{2l+1}\begin{pmatrix} l & l+1 & 1\\ 0 & 0 & 0\end{pmatrix}^2[\mathcal{F}(\theta,l,l+1)(2l+3)\sqrt{2l+1}Y_{l0}(\mathbf{q})+\mathcal{F}(\theta,l+1,l)\sqrt{2l+3}(2l+1)Y_{l+1,0}(\mathbf{q})] \tag{21}$$

with the radial part of the integrals given by

$$\mathcal{F}(\theta,n,n')=\int_0^\infty dr_2r_2^2j_n(q_Nr_2)j_{n'}\left(\frac{e\mathcal{E}_0r_2}{\hbar\omega}\sin\theta\right)R_0(r_2)\left(\int_0^{r_2}dr_1\frac{r_1^3}{r_2^2}R_g(r_1)R_a(r_1)+r_2\int_{r_2}^\infty dr_1R_g(r_1)R_a(r_1)\right) \tag{22}$$

and the quantities $\left(\begin{smallmatrix} a & b & c\\ d & e & f\end{smallmatrix}\right)$ representing the usual $3j$ symbols. As a counterpart, we write the corresponding matrix element in the absence of the laser:

$$T_i^{ff}=-i\frac{4\pi e^2}{3}\mathcal{F}(0,1,0)Y_{10}(\mathbf{q}) \tag{23}$$

with

$$\hbar^2q^2/2m=E_a+E_0-E_g\,.$$

This procedure applies to T_2 as well and gives similar results. It is evident, comparing (21) and (23), that the laser interacts strongly with the electron while it is leaving the atom and introduces severe modifications into the angular distribution even if the field intensity is not strong enough to alter the atom wave functions.

Already in the weak-field limit of Eq. (10) we can appreciate the modifications induced by the laser on the angular distribution of the Auger effect. This task is accomplished by developing $X(N,\theta,\mathbf{q}_N)$ and the Bessel functions entering formula (19) in powers of $\mathcal{E}_0/\omega^2$ and retaining only first-order terms. In this case, the integral over θ in (12) is performed analytically and yields three Kronecker δ's which force N to assume the values -1, 0, and 1. Therefore no more than one photon can be exchanged with the radiation field and only three terms survive in the summation of Eq. (10). In this limit the ionization probability for unit of solid angle becomes

$$\frac{dP}{d\Omega_\mathbf{q}}=\frac{dP}{d\Omega_\mathbf{q}}\bigg|^{ff}(1+\lambda\cos^2\Theta)+\delta\,, \tag{24}$$

where

$$\frac{dP}{d\Omega_\mathbf{q}}\bigg|^{ff}=\frac{me^4q}{3\pi\hbar^3}(f_1+3sf_2)^2\cos^2\Theta \tag{25}$$

is the ionization probability for unit of solid angle in absence of laser and Θ the angle between $\mathbf{q}$ and $\mathcal{E}_0$;

$$\lambda=\frac{\mathcal{E}_0^2}{\omega^4}\frac{e^2}{2m^2}q^2\,, \tag{26}$$

$$\delta=\frac{\mathcal{E}_0^2}{\omega^2}q\frac{m\pi}{(2\pi\hbar)^3}A^2\,, \tag{27}$$

$$f_1=\int_0^\infty dr_2r_2^2j_1(qr_2)R_0(r_2)\left[\int_0^{r_2}dr_1\frac{r_1^3}{r_2^2}R_g(r_1)R_a(r_1)+r_2\int_{r_2}^\infty dr_1R_g(r_1)R_a(r_1)\right]\,, \tag{28}$$

$$f_2=\int_0^\infty dr_2r_2^2j_1(qr_2)R_a(r_2)\left[\int_0^{r_2}dr_1\frac{r_1^2}{r_2}R_g(r_1)R_0(r_1)+\int_{r_2}^\infty dr_1r_1R_g(r_1)R_0(r_1)\right]\,, \tag{29}$$

$$A=\frac{2\sqrt{\pi}e^3}{3\sqrt{3}\hbar}\left[f_{32}-f_{30}+3s(f_{42}-f_{40})-3(f_{32}+3sf_{42})\cos^2\Theta\right]\,, \tag{30}$$

$$f_{3i}=\int_0^\infty dr_2r_2^3j_i(qr_2)R_0(r_2)\left[\int_0^{r_2}dr_1\frac{r_1^3}{r_2^2}R_g(r_1)R_a(r_1)+r_2\int_{r_2}^\infty dr_1R_g(r_1)R_a(r_1)\right]\quad(i=0,2)\,, \tag{31}$$

$$f_{4i}=\int_0^\infty dr_2r_2^3j_i(qr_2)R_a(r_2)\left[\int_0^{r_2}dr_1\frac{r_1^2}{r_2}R_g(r_1)R_0(r_1)+\int_{r_2}^\infty dr_{1,}r_1R_g(r_1)R_0(r_1)\right]\quad(i=0,2)\,. \tag{32}$$

Expression (24) amounts to the perturbative expansion of the general formula (10), with the laser intensity present in λ and δ. It will turn out that $\delta \ll \lambda$ so that the modifications are driven essentially by the value of λ. In Eq. (24) λ is smaller than 1, but it gives an idea of the possible trend. Taking for instance $\lambda = 0.1$ we find that this value is obtained with quite realistic values of the other parameters of the process, namely, $\hbar^2 q^2/2m = 870$ eV, $\hbar\omega = 0.1$ eV, $\mathcal{E}_0 = 4\times 10^3$ V/cm, and is legitimate to expect that larger values of the field intensity may be able to give larger modifications in the nonperturbative case. Numerical calculations of the above integrals require the choice of suitable radial wave functions for the three active states entering the formulas. To check their relative importance we have evaluated them using hydrogen wave functions. The results of the numerical work are

$$\left.\frac{dP}{d\Omega_{\mathbf{q}}}\right|^{ff} = (0.7\times 10^{14})\cos^2\Theta \sec^{-1} ,$$

$$\delta = (0.2\times 10^5)(0.2 + 1.38\cos^2\Theta) \sec^{-1} .$$

The last term in (24) is then negligible with respect to the others, albeit it is the only one surviving in the overall probability when $\Theta = 90°$.

In conclusion, in the Auger effect, the presence of a laser field manifests itself through different mechanisms. Two of them are considered as the most significant. First, the ejection into the continuum can take place simultaneously through many channels, each specified by the number of photons emitted or absorbed during ionization [see Eq. (10)]. It will modify the electron final-energy distribution, which is now broader and more discrete. Second, the presence of a laser field manifests itself through a reduction of the geometrical symmetries of the physical process, which in turn brings about interference of many orbital angular momenta, able to produce sizable effects in the angular distributions.

In the same spirit as the laser-assisted Compton scattering, one may conceive of measuring a given Auger transition with and without the presence of a spectator laser field. Then the differences in the measurements would give information on the interaction of the laser field with correlated electrons, and on the modifications of the Auger transition. In turn, hopefully, these can be used to give more precise information on the role of electron correlations in many-electron atoms.

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