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One-photon decay of two-hole states in atoms

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The techniques of many-body theory are used to make calculations with Hartree-Fock wave functions of the probability of one-photon decay of two-hole states in the first nonvanishing approximation in the interaction of vacancies. Specific results are obtained for two $1s$ vacancies in neon, two $2p$ and $2s$ vacancies in doubly ionized argon, and two $4d$ vacancies in xenon. The results agree extremely well with recently obtained experimental data on neon and argon. An experimental study is proposed of one-photon decay in a process in which a two-hole state is formed as the result of an Auger process following the removal of an electron from an inner shell by a photon or fast electron.

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1. There have recently appeared both experimental[1–5] and theoretical[6,7] investigations of the mechanisms of decay of highly excited atomic states produced by the removal of two electrons from an inner shell—two-hole states. These phenomena are interesting because their study can give additional information about the interaction between electrons in atoms; in the case of simultaneous decay of two vacancies this is the only information available.

Two-hole states can decay either owing to the ordinary Auger effect, in which the two vacancies are destroyed independently with the emission of two or more photons, or by a radiative process in which one vacancy decays via the Auger effect, and the other in a radiative transition.

There is, however, also a different and extremely interesting possibility for the decay of a two-hole state, in which both vacancies are simultaneously filled by electrons from the outer shells, and the energy released is carried away by a single electron or photon. In the case of emission of an electron the process has received the name of the three-electron Auger effect[1]; the other case, with a photon emitted, is called one-photon decay of a two-hole state.[2–6] Both processes are possible only because of the existence of an interaction between electrons (or holes) in an atom, and consequently are essentially many-electron phenomena. Therefore in theoretical studies on the decay of two-hole states the interaction between vacancies must be taken into account from the beginning.

In the present paper we make an investigation of the one-photon decay of two-hole states in atoms by means of the techniques of the quantum theory of many-bodies.[8] The probabilities of one-photon decay are calculated for two-hole states of several atoms: $(1s)^{-2}$ in Ne, $(2s)^{-2}$ and $(2p)^{-2}$ in Ar^{++}, and $(4d)^{-2}$ in Xe. A preliminary note on the theoretical interpretation and results for this process has been published earlier.[6]

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2. In the case of radiative decay the initial and final states each have two vacancies, i_1, i_2 and j_1, j_2, respectively. The energy difference is carried away by a photon with polarization e and wave vector $\varkappa$. Since the atomic shells containing the vacancies in the initial and final states are very far apart, perturbation theory in terms of the interaction between electrons can be used to calculate the probability.

The probability is given by the expression

$$d\Gamma((e)^{-2} \to \gamma) = 2\pi |M_{fq}|^2 \delta(E_f - E_q + \omega) \frac{\omega^2}{(2\pi c)^3} d\Omega\, d\omega, \qquad (1)$$

where c is the speed of light and M is the amplitude for the process. Atomic units with $\hbar = m = c = 1$ are used throughout.

To determine the probability $\Gamma((e)^{-2} \to \gamma)$ and to identify the transitions corresponding to the decay of the two-hole state, one must know how to calculate with sufficient accuracy not only the amplitude M_{fq} of the process, but also the energy ω of the emitted photon.

In the one-particle Hartree–Fock approximation we can set ω equal to the difference $E_{j_1} + E_{j_2} - E_{i_1} - E_{i_2}$, where E_i and E_j are the Hartree–Fock energies of the hole states i and j. However, the single-ionization potential is not equal to the Hartree–Fock energy E_i, but differs from it by the energy of the rearrangement that occurs in the atomic shell in the ionization process. When two electrons are removed from an atom the double-ionization energy differs from its Hartree–Fock value because of

1) corrections owing to the energy of rearrangement of the atomic shells

$$\Delta E_1 \approx E_{i_1}^{\exp} + E_{i_2}^{\exp} - E_{i_1}^{\mathrm{HF}} - E_{i_2}^{\mathrm{HF}} \qquad (2)$$

($E_i^{\exp}$ is the experimental value of the energy for removing the electron i);

2) corrections caused by the direct interaction of the holes i_1 and i_2:

$$\Delta E_2 = \langle i_1 i_2 | V | i_1 i_2 - i_2 i_1 \rangle \qquad (3)$$

(V is the potential of the Coulomb interaction between electrons). The order of magnitude of the quantity ΔE_2 can be estimated as follows:

$$\Delta E_2 \sim 1/\bar{r} \sim n/Z$$

($\bar{r}$ is the average distance between shells, n is the principal quantum number i_1 (or i_2), and Z is the effective charge acting on the electrons of these shells),

3) corrections associated with virtual excitation of other hole states. For inner shells it can be expressed[9] in terms of the static monopole polarizabilities $\alpha_i^{(0)}$ of the atomic shells outside i:

$$\Delta E_3 = \sum_{\substack{i \le F \\ p > F}} \left| \left\langle p \left| \frac{1}{r} \right| i \right\rangle \right|^2 \frac{1}{E_p - E_i} = \sum_{i > i_1, i_2} \frac{1}{2} \alpha_i^{(0)}(0), \qquad (4)$$

where $i \le F$ indicates summation over occupied states, and $p > F$ that over empty states.

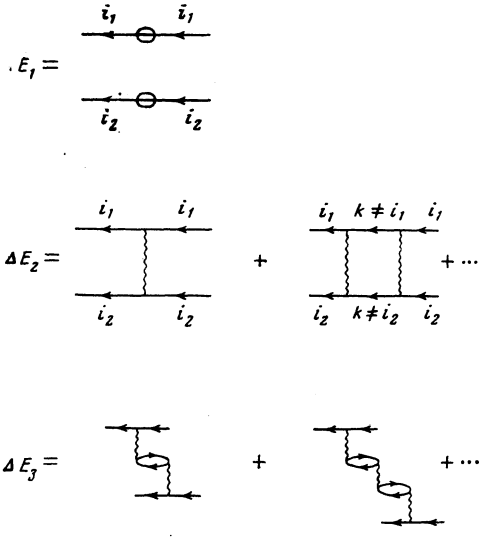

FIG. 1.

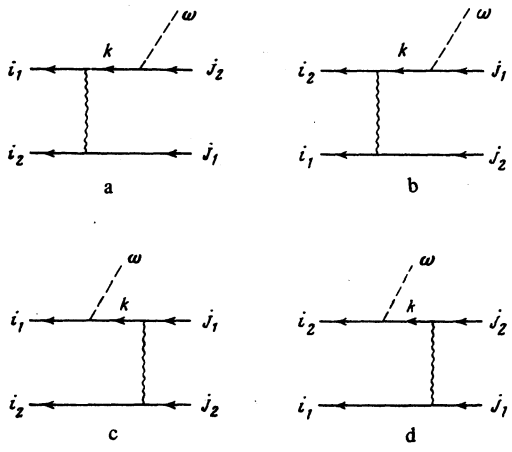

FIG. 2.

In the language of Feynman diagrams the corrections to the double-ionization potential of an atom are determined by the matrix elements represented in Fig. 1, where a circle denotes the real component of the proper-energy part of the one-particle Green's function, a wavy line denotes the Coulomb interaction, and lines with arrows to right or left respectively denote particle or hole states. The values of the various corrections will be derived further on.

In first order in the interelectronic interaction the amplitude M_{fq} for one-photon decay of a two-hole state is determined by the diagrams shown in Fig. 2. Besides the diagrams shown in Fig. 2 we must consider the corresponding exchange diagrams. Using the rules of graphic correspondence,[8] we get the expression for the amplitude

$$M = \sum_{i=1}^{4} M_i.$$

The expressions M_1-M_4 correspond to the diagrams Fig. 2 a–d and can be written

$$M_1=\sum_k \frac{\langle i_1 i_2|U|kj_1\rangle\langle k|(\hat{de})|j_2\rangle}{E_{i_1}+E_{i_2}-E_k-E_{j_1}}, \tag{5}$$

$$M_2=\sum_k \frac{\langle i_2 i_1|U|kj_2\rangle\langle k|(\hat{de})|j_1\rangle}{E_{i_1}+E_{i_2}-E_k-E_{j_2}}, \tag{6}$$

$$M_3=\sum_k \frac{\langle ki_2|U|j_1 j_2\rangle\langle i_1|(\hat{de})|k\rangle}{E_k+E_{i_2}-E_{j_1}-E_{j_2}}, \tag{7}$$

$$M_4=\sum_k \frac{\langle ki_1|U|j_2 j_1\rangle\langle i_2|(\hat{de})|k\rangle}{E_k+E_{i_1}-E_{j_1}-E_{j_2}}. \tag{8}$$

In these equations the summation over k is taken over both hole states and particle states, since the emission of the photon can occur either before or after the interaction between the vacancies; d is the dipole moment operator for the electron; and

$$\langle i_1 i_2|U|j_1 j_2\rangle=\langle i_1 i_2|V|j_1 j_2\rangle-\langle i_1 i_2|V|j_2 j_1\rangle$$

is the difference of the direct and exchange Coulomb matrix elements.

Let us consider some properties of the amplitude for the process. In diagrams c and d the energy denominator is equal to zero for a certain value

$$E_k=E_{j1}+E_{j2}-E_{in} \quad (n=1, 2) \tag{9}$$

of the energy of the virtual state of the electron. In this case the virtual state can become a real state and the one-photon decay process can occur in two stages: first the Auger decay of one vacancy, and then the recombination of the other one. If we neglect the real part of the amplitude M_{j_q} the contribution of this sort of mechanism to the total width $\Gamma((e)^{-2}\to\gamma)$ can be expressed in terms of the partial width of the corresponding Auger transition $i_1\to j_1 j_2 k$ and the partial cross section $\sigma^{i_1 i_2 k}$ for the photoionization:

$$\Delta\Gamma_1=\alpha^2\pi^{-2}\omega^2\sigma_\Gamma^{i_1 i_2 k}\Gamma_A^{i_1 k}. \tag{10}$$

There is an interesting situation when the condition

$$E_{j_n}+E_k\approx E_{i_1}+E_{i_2} \quad (n=1, 2), \tag{11}$$

is satisfied; here E_k is the energy of some internal vacancy of the atom in question. In this case the main contribution comes from diagrams a and b in Fig. 2, since they can have a small energy denominator. Since the states $(i_1)^{-1}(i_2)^{-1}$ and $(j_n)^{-1}(k)^{-1}$ have nearly equal energies, a mixing of these configurations arises if for accidental reasons the matrix element $\langle j_n k|V|i_1 i_2\rangle$ is not small. In the limit of strong configuration mixing, a unique complex is formed which decays just as fast, actually, as a one-hole state. In this case the one-photon decay of the two vacancies is a process of resonance type, in the sense that its probability is $1/\varepsilon^2$, where $\varepsilon=E_k+E_{j_n}-E_{i_1}-E_{i_2}$ appears as the resonance defect. If ε is very small, it is necessary to take into account the width of the "k"th level. The process can be regarded as occurring in two stages: transition of the vacancies i_1, i_2 into j_n, k, and subsequent radiative decay of the "k"th hole. When the width of the hole level is taken into account, the contribution of this mechanism to the total probability of one-photon decay of the two-hole state is given by the following expression:

$$\Delta\Gamma_2=\frac{|\langle i_1 i_2|U|kj_n\rangle|^2}{(E_{i_1}+E_{i_2}-E_k-E_{j_n})^2+\Gamma_k^2/4}W_k, \tag{12}$$

where W_k is the radiation width of the "k"th vacancy.

We now obtain the expression for the total probability $\Gamma((e)^{-2}\to\gamma)$ by summing over the polarizations and integrating over the angles of emission of the photon, and also averaging and summing over the orbital and spin magnetic quantum numbers. We find from Eq. (1)

$$\Gamma((e)^{-2}\to\gamma)=\tfrac{2}{3}\alpha^3\omega^3|M|^2, \tag{13}$$

where α is the fine-structure constant, and the square of the amplitude is given by the expression

$$|M|^2=\frac{1}{(2l_{i_1}+1)(2l_{i_2}+1)}\Big[\sum_{i=1}^{4}|M_i|^2+2\sum_{i<j}\mathrm{Re}\,M_{ij}\Big]. \tag{14}$$

The diagonal terms $|M_i|^2$ are of the form

$$\begin{aligned}|M_1|^2=\sum_{l_i l_2 l_k}\frac{1}{2l_k+1}\Big\{&\frac{\delta_{l_1 l_2}}{2l_1+1}[|R_{1a}(l_k, l_1)|^2+|R_{1b}(l_k, l_1)|^2]\\&-(-1)^{l_1+l_2}\Big[\begin{Bmatrix} l_1 & l_{i_1} & l_k\\ l_2 & l_{i_2} & l_{j_1}\end{Bmatrix}R_{1a}(l_k, l_1)R_{1b}^*(l_k, l_2)\\&+\begin{Bmatrix} l_1 & l_{i_2} & l_k\\ l_2 & l_{i_1} & l_{j_1}\end{Bmatrix}R_{1b}(l_k, l_1)R_{1a}^*(l_k, l_2)\Big],\end{aligned} \tag{15}$$

$$\begin{aligned}|M_3|^2=\sum_{l_i l_2 l_k}\frac{1}{2l_k+1}\Big\{&\frac{\delta_{l_1 l_2}}{2l_1+1}[|R_{3a}(l_k, l_1)|^2+|R_{3b}(l_k, l_1)|^2]\\&-(-1)^{l_{i_1}+l_{i_2}}\Big[\begin{Bmatrix} l_1 & l_{j_1} & l_k\\ l_2 & l_{j_2} & l_{i_2}\end{Bmatrix}R_{3a}(l_k, l_1)R_{3b}^*(l_k, l_2)\\&+\begin{Bmatrix} l_1 & l_{j_2} & l_k\\ l_2 & l_{j_1} & l_{i_1}\end{Bmatrix}R_{3b}(l_k, l_1)R_{3a}^*(l_k, l_2)\Big]\Big\};\end{aligned} \tag{16}$$

the expressions for $|M_2|^2$ and $|M_4|^2$ are found from Eqs. (15) and (16) by the exchanges $i_1\rightleftarrows i_2$, $j_1\rightleftarrows j_2$, and $R_{ia}\rightleftarrows R_{ib}$ $(i=1, 2, 3, 4)$. We do not present here the expressions for the nondiagonal matrix elements, which are much more cumbersome.

The quantities R_{ia} and R_{ib} in Eqs. (15) and (16) are determined from the direct and exchange components of the expressions (5)–(8), where we have only to replace the matrix elements of the Coulomb and dipole operators by their reduced matrix elements $\langle(i_1 i_2\|V_1\|j_1 j_2\rangle$ and $\langle i\|d\|k\rangle$ defined in the usual way.[9, 10]

3. The calculations have been carried out for two $1s$ vacancies in Ne and for two $2s$ and $2p$ holes in Ar^{++}. This choice of the first two atoms is due to the fact that experimental work has recently been reported on the one-photon decay of the two-hole states $(1s)^{-2}$ in Ne[11] and $(2p)^{-2}$ in Ar^{++}.[2] We also studied the decay of the rearrangement of the shells and for direct and virtual interaction between the vacancies, according to Eqs. (2)–(4). As an example we give the values of ΔE_i $(i=1, 2, 3)$ for the transition $(2p)^{-2}\to(3s)^{-1}(3p)^{-1}$ in Ar^{++}:

$$\Delta E_1=10.9 \text{ eV}, \quad \Delta E_2=60 \text{ eV}, \quad \Delta E_3=9.3 \text{ eV}.$$

It can be seen that the corrections amount to about 16

TABLE I.

Atom	Transitions	Energy ω, eV	$\Gamma((e)^{-2}\to\gamma)$ 10^{-6} eV	$\Delta\Gamma_1$, 10^{-6} eV	$\frac{\Gamma((e)^{-2}\to\gamma)}{2\Gamma_A}$ 10^{-4}	$\frac{\Gamma((e)^{-2}\to\gamma)}{2W}$
Ne	$(1s)^{-2}\to(2s)^{-1}(2p)^{-1}$	1810	13,14	0,86	7,7	0,002
Ar^{++}	$(2s)^{-2}\to(3s)^{-1}(2p)^{-1}$	370	40,63	0*	–	0,47
Ar^{++}	$(2s)^{-2}\to(3s)^{-1}(3p)^{-1}$	646	4,76	4,28	7,9	0,01
Ar^{++}	$(2p)^{-2}\to(3s)^{-1}(3p)^{-1}$	500	0,30	0,017	1,8	0,006
Xe	$(4d)^{-2}\to(5s)^{-1}(5p)^{-1}$	122	0,43	0,0010	4,3	0,025

*$\Delta\Gamma_1$ is equal to zero, since the Coster-Kronig transition $(2s)^{-1}\to(3s)^{-1}(3p)^{-1}$ is energy-forbidden.

percent of the total energy.

The energies of the transitions are given in Table I. The summation over intermediate states included integration over the continuous spectrum and was done numerically. The infinite upper limit was replaced with a finite one. Its value was determined by the behavior of the product of the matrix elements of the Coulomb and dipole operators. Where the energy denominator vanishes in diagrams c and d the numerical integration was accomplished with procedures suggested by Bloch.[13] In the sums over particle states the part from discrete levels is rather large. Ordinarily the contributions from the two or three lowest states were included, since the contribution of the discrete levels falls off rapidly with increasing quantum number. All of the necessary wave functions and matrix elements were computed with special programs.[10] The computing was done with a BÉSM-6 computer.

4. The results obtained for the widths of one-photon decays of two-hole states are shown in the table. In the cases considered no state $(4d)^{-2}$ in Xe, in which the energy of the two $4d$ vacancies is rather close to that of the deeper shell $4p$ and the energy denominator becomes small in diagrams a and b of Fig. 2. Owing to this the one-photon decay of the $(4d)^{-2}$ states may be a process of resonance type.

The calculations were made with the Hartree–Fock functions for the hole and electron states. The wave functions for the hole states were found by solving a system of coupled Hartree–Fock equations, except in the case of the decay of two equivalent s-holes, where it turned out to be impossible to determine the wave function simultaneously with those of the other hole states, since the electrons of this subshell are lacking from the atom core. In this case the wave function of the empty s-shell was determined as an excited function in the frozen field of the other hole states. The wave functions of the electronic states were determined in the field of a core in which the two electrons i_1 and i_2 are absent.

For the decay of two $2p$ or $4d$ vacancies we used Hartree–Fock functions averaged over the terms of the given configuration.[12] In the case of the decay of an s subshell the other shells are closed and there is no need to average over terms. The calculation of the energy of the photon emitted included the corrections for particular decay mechanism was distinguished as making a contribution much larger than the others. Therefore it is necessary to consider all of the various mechanisms represented by the diagrams of Fig. 2. The nondiagonal terms M_{ij} make a very important contribution to $|M|^2$, and neglecting them would lead to a large error. This means that the influence on each other of the various decay channels is rather large, and the probability of one-photon decay cannot be regarded as a simple sum of contributions from the various decay mechanisms, so that we could write

$$\Gamma((e)^{-2}\to\gamma)=\sum_{i=1}^{4}\Gamma^{(i)}((e)^{-2}\to\gamma).$$

It is worth while to compare our values of the widths of one-photon decays of two vacancies with twice the widths of one-hole states—the radiative width W and the Auger width Γ_A—which appear when the two vacancies decay independently. Accordingly we calculated these quantities by the formulas

$$W=\frac{1}{6(2l_i+1)}\alpha^3\omega^3|\langle i\|d\|j\rangle|^2, \tag{17}$$

$$\Gamma_A=\frac{4\pi}{2l_i+1}\sum_{l_1l_2l_k}\left\{\frac{\delta_{l_1l_2}}{2l_1+1}\left[|\langle ik\|V_{l_1}\|j_1j_2\rangle|^2+|\langle ik\|V_{l_2}\|j_2j_1\rangle|^2\right]-\begin{Bmatrix} l_i & l_1 & l_{j_1}\\ l_k & l_2 & l_{j_2}\end{Bmatrix}\langle ik\|V_{l_1}\|j_1j_2\rangle\langle ik\|V_{l_2}\|j_2j_1\rangle\right\}, \tag{18}$$

in which the energy is measured in Rydbergs. The values so found for the ratios $\Gamma((e)^{-2}\to\gamma)/2\Gamma_A$ and $\Gamma((e)^{-2}\to\gamma)/2W$ are shown in the table. We note that there is a rather large spread (from thousandths to tenths) in the ratios $\Gamma((e)^{-2}\to\gamma)/2W$, depending on the atom and the initial vacancies i_1 and i_2.

It can be seen from the table that Eq. (10) gives not more than 10 percent of the total width, except in the case of the transition $(2s)^{-2}\to(3s)^{-1}(3p)^{-1}$ in Ar^{++}. Consequently, one-photon decay of two vacancies does not go via two successive steps, Auger effect and recombination, but is a single process going through virtual states. In the study of the decay of two $4d$ vacancies in Xe it is found that the contribution of the intermediate $5p$ hole state is of the same order as that of the $4p$ state, despite the fact that $2E_{4d}-E_{4p}+E_{5p}$. The reason is that the matrix element $\langle 5s\|d\|5p\rangle$ is much larger than $\langle 5s\|d\|4p\rangle$ (by a factor 8), while the Coulomb matrix-elements $\langle 4d4d\|V_1\|4p5p\rangle$ and $\langle 4d4d\|V_1\|5p5p\rangle$ are roughly equal. This means that the one-photon decay of the $(4d)^{-2}$ state in Xe is of nonresonance nature. However, in the production of two vacancies in an inner subshell by the collision of two heavy particles there is a possibility that the energy levels will be shifted for particular values of the collision parameters[2)] so as to make $2E_{4d}=E_{4p}+E_{5p}$. In such a case one should expect a sharp increase of the probability of one-photon decay of the $(4d)^{-2}$ two-hole state.

To make possible a direct comparison with experiment,[2] the calculations for Ar^{++} were carried out for a configuration in which two further electrons were removed from the outer $3p$ subshell. We studied the decays of the $(2s)^{-2}$ and $(2p)^{-2}$ states. It can be seen from the table that the width of the one-photon decay $(2s)^{-2}\to(2p)^{-1}(3s)^{-1}$ is very large in comparison with the widths of the decays $(2s)^{-2}\to(3s)^{-1}(3p)^{-1}$ and $(2p)^{-2}\to(3s)^{-1}(3p)^{-1}$. In the case of the decay of two $2p$ vacancies the calculated value of the ratio $\Gamma((e)^{-2}\to\gamma)/2\Gamma_A$ is $1.8\cdot10^{-6}$,

whereas the experimental value lies in the range $(1.7-4.3)\cdot 10^{-6}$. The spread of the experimental data is explained by the uncertainty about the charge of the ions produced in collisions.[2] In the neon atom, our value for the width of the one-photon decay of the two-hole state $(1s)^{-2}$ is $13.14\cdot 10^{-6}$ eV. It agrees well with the experimental value from[11], which is $14\cdot 10^{-6}$ eV, and differs widely from the value found in[7], $32\cdot 10^{-6}$ eV. This can be explained by the fact that the many-electron nature of the one-photon decay process was not taken into account in[7].

On the basis of these data we can conclude that the mechanism that we have assumed satisfactorily describes the phenomenon of one-photon decay of two-hole states.

In the current experiments[1-5] two-hole states are formed as the result of collisions between two heavy particles. There can be electrons knocked out from outer shells, as in the case of Ar^{++}, and this makes comparison with theoretical calculations somewhat difficult. We propose an experiment in which the two-hole state is formed not in the collision of heavy particles, but as a consequence of the production of a deep vacancy by photoionization of inelastic scattering of fast electrons. The one-hole state so produced decays by an Auger effect, forming two vacancies in a different subshell. The process is quite feasible for observation, since the cross section of photoionization of the 1s shell in Ar, for example, is 10^{-19} cm^2, the Auger width of the 1s vacancy is 0.6 eV, and the partial width of the Auger process is about 90 percent.

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