$\Omega' \ll \omega$. This distinction is necessary for accuracy to second order in ω^{-1}, and is what distinguishes our derivation from the more familiar Block-Siegert-type derivations, which are valid only to first order in ω^{-1}.

[3]The author is indebted to Professor P. Stehle (private communication) for pointing out the necessity of this step.

[4]This frequency shift is identical to the one found by F. Bloch and A. Siegert [Phys. Rev. 57, 522 (1940)], and agrees with the result of Chang and Stehle [Ref. 1, Eq. (87)] when the latter is expanded to order s^2.

[5]Reference 1, Eq. (77).

[6]By including the effect of a wave function renormalization, however, C. S. Chang and P. Stehle (private communication) find a result in agreement with ours.

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Double-Ionization Effects on Radiative Rates, Auger Rates, and Fluorescence Yields*

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The contributions of double ionization to radiative rates, Auger rates, and fluorescence yields for the filling of atomic K-shell vacancies are calculated by use of the classical Gryzinski methods. The double-ionization correction to the ratio of ionization cross sections for α particles and deuterons incident on $_{22}$Ti is $\sim 6\%$. Recent experimental $_{22}$Ti results of Lewis, Natowiz, and Watson differ from the Born prediction by as much as 7% between 10 and 80 MeV with errors of 2% or less.

Recently, Lewis, Natowiz, and Watson (LNW) have performed a series of precise experiments[1] to test the z dependence of x-ray emission induced by alpha particles ($z=2$) and by deuterons ($z=1$). These experiments, which have been done between 10 and 80 MeV on elements of atomic charge Z from 17 to 29, indicate a behavior on z and Z which deviates from the Born approximation for single ionization. In this paper we shall show that the effect of double ionization on the radiation rate, the Auger rate, and the fluorescence yield are non-negligible, using classical Gryzinski methods[2] to calculate their contributions.

We begin by labeling all of the electrons in our target atom by $n\uparrow$ or $n\downarrow$, depending on their spin. Let us consider the ionization of a K-shell electron with spin up, denoted by $K\uparrow$. The remaining hole may be filled either by a radiative transition from a higher level, or by a nonradiative Auger transition. The radiative rate is given by

$$R^0 = \sum_n R_{n\uparrow}^{K\uparrow} \; . \tag{1}$$

Since we shall neglect spin-dependent forces, this sum extends only over dipole-connected states. Similarly, the Auger rate may be expressed as

$$A^0 = \sum_{\substack{n,m \\ n\neq m}} A_{n\uparrow,m\uparrow}^{K\uparrow} + \sum_{n,m} A_{n\uparrow,m\downarrow}^{K\uparrow} \, , \tag{2}$$

where the first of the lower indices corresponds to the electron which drops into the $K\uparrow$ state, and the second to the electron which is ejected. The fluorescence yield is given by

$$F^0 = R^0/(R^0 + A^0). \tag{3}$$

We now consider double ionization, where the second electron is ejected from a Jth shell. First, consider double ionization of $K\uparrow$ and $J\uparrow$. The new radiative and Auger rates corresponding to the filling of $K\uparrow$ are

$$R^{K\uparrow,J\uparrow} = \sum_{\substack{n \\ n\neq J}} R_{n\uparrow}^{K\uparrow} \, , \tag{4}$$

$$A^{K\uparrow,J\uparrow} = \sum_{\substack{n,m \\ n\neq m \\ n\neq J \\ m\neq J}} A_{n\uparrow,m\uparrow}^{K\uparrow} + \sum_{\substack{n,m \\ n\neq J}} A_{n\uparrow,m\downarrow}^{K\uparrow} \; . \tag{5}$$

Similarly, for double ionization of $K\uparrow$ and $J\downarrow$ we have

$$R^{K\uparrow,J\downarrow} = R^0 \, , \tag{6}$$

$$A^{K\uparrow,J\downarrow} = \sum_{\substack{n,m \\ n\neq m}} A_{n\uparrow,m\uparrow}^{K\uparrow} + \sum_{\substack{n,m \\ m\neq J}} A_{n\uparrow,m\downarrow}^{K\uparrow} \; . \tag{7}$$

By specifying that the J shell has $2n_J$ electrons we may write

$$R^0 = n_J R_{J\uparrow}^{K\uparrow} \, , \tag{8}$$

$$A^0 = n_J(n_{J-1}) A^{K\uparrow}_{n\uparrow,J\uparrow} + n_J^2 A^{K\uparrow}_{n\uparrow,J\downarrow} \ . \tag{9}$$

We now denote the probability for knocking out one $K\uparrow$ electron by $P_{K\uparrow}$, and the probability for knocking out both a K and any other electron A by $P_{K\uparrow,A}$. Assuming that single-particle transition probabilities are not changed because of the additional vacancy, the total radiation rate, including both single and double ionization, may be written as

$$R = n_J R^{K\uparrow}_{J\uparrow}\left(1 - \frac{P_{K\uparrow,J\uparrow}}{P_{K\uparrow}} - \frac{P_{K\uparrow,J\downarrow}}{P_{K\uparrow}}\right) + (n_J - 1) R^{K\uparrow}_{J\uparrow}\left(\frac{P_{K\uparrow,J\uparrow}}{P_{K\uparrow}}\right) + n_J R^{K\uparrow}_{J\downarrow}\left(\frac{P_{K\uparrow,J\downarrow}}{P_{K\uparrow}}\right)$$
$$= R^0\left(1 - \frac{1}{n_J}\frac{P_{K\uparrow,J\uparrow}}{P_{K\uparrow}}\right) \ . \tag{10}$$

Similarly, the new Auger rate is given by

$$A = [n_J(n_J-1)A_{K\uparrow,J\uparrow} + n_J^2 A_{K\uparrow,J\downarrow}]\left(1 - \frac{P_{K\uparrow,J\uparrow}}{P_{K\uparrow}} - \frac{P_{K\uparrow,J\downarrow}}{P_{K\uparrow}}\right) + [(n_J-1)(n_J-2)A_{K\uparrow,J\uparrow} + n_J(n_J-1)A_{K\uparrow,J\downarrow}]\left(\frac{P_{K\uparrow,J\uparrow}}{P_{K\uparrow}}\right)$$
$$+ [n_J(n_J-1)A_{K\uparrow,J\uparrow} + n_J(n_J-1)A_{K\uparrow,J\downarrow}]\left(\frac{P_{K\uparrow,J\downarrow}}{P_{K\uparrow}}\right)$$
$$= A^0 - [2(n_J-1)A_{K\uparrow,J\uparrow} + n_J A_{K\uparrow,J\downarrow}]\left(\frac{P_{K\uparrow,J\uparrow}}{P_{K\uparrow}}\right) - [n_J A_{K\uparrow,J\downarrow}]\left(\frac{P_{K\uparrow,J\uparrow}}{P_{K\uparrow}}\right) \ . \tag{11}$$

Assuming that

$$P_{K\uparrow J\uparrow} = P_{K\uparrow J\downarrow} = P_K\,\xi/2 \ , \quad A_{K\uparrow J\uparrow} = A_{K\uparrow J\downarrow} \ , \tag{12}$$

it may be shown, after a little algebra, that the new fluorescence yield may be expressed as

$$F = F^0[1 + (\xi/2n_J)(1 - F^0) + O(\xi^2)] \ , \tag{13}$$

where we have assumed that ξ, the relative probability of double ionization to single ionization, is small.

In principle, contributions from all available J shells should be included in applications of the above results. In subsequent calculations, however, we shall include only the contributions from the L-shell ($n = 2$, $l = 1$) states since estimates of the contributions from higher shells indicate that their contributions are small.

The contributions of double ionization to R, A, and F may be found by evaluating ξ, which is equal to the ratio of the total double-ionization cross section Q^{ii} to the total single-ionization cross section Q^i. These cross sections may be easily evaluated by using the simple classical equations of Gryzinski. The Gryzinski single-ionization cross section, which has the same z^2 behavior as the Born approximation, is given by

$$Q^i = (\sigma_0/U_i^2)\,G_i(v_q/v_i) \ , \tag{14}$$

where

$$\sigma_0 = \pi e^4 z^2 \tag{15}$$

and $G_i(v_q/v_i)$ is a moderately complicated function of the ratio of the velocity v_q of the bombarding particle to the orbit velocity v_i of the atomic electron which is ionized. The term U_i represents the binding energy of this atomic electron.

In the case of double ionization, there are four types of amplitudes which contribute. The first two represent the case where the two ionized electrons are both ionized by the bombarding particle (one amplitude for each sequence of ionization). The contribution from these terms goes as z^4. The other amplitudes arise when the bombarding electron ionizes one electron which in turn ionizes a second electron on its way out. These amplitudes go as z^2. From Gryzinski's classical expressions one may find the cross section for each process, but one cannot obtain interference terms which go as z^3, for example.

In the region where v_q and v_i are roughly the same order of magnitude, the largest of the above amplitudes is that amplitude corresponding to the bombarding particle first ionizing a K-shell and then an L-shell electron. The expression given by Gryzinski corresponds to

$$Q^{ii} = \frac{2n_K\,2n_L}{4\pi\bar{r}^2}\left(\frac{\sigma_0}{U_i^2}\right)\left(\frac{\sigma_0}{U_{ii}^2}\right) \times G_i\left(\frac{v_q}{v_i}\right) G_Q\left(\frac{U_i}{U_{ii}}, \frac{v_q}{v_i}\right) \ , \tag{16}$$

where $2n_{K(L)}$ is the total number of electrons in the $K(L)$ shell, $\bar{r}$ is the mean distance between electrons, and U_{ii} represents the binding energy of the second atomic electron to be ionized. Taking $U_i/U_{ii} \simeq U_K/U_L = 4$, then we find from Gryzinski that G_Q is a slowly varying function whose value near $v_q \simeq v_i$ is ~ 0.3. The other double-ionization cross sections are at least a factor of 2, and typically a factor of 5 smaller.

In order to evaluate ξ given by

$$\xi = \frac{Q_{ii}}{Q_i} = \frac{2n_K\,2n_L}{4\pi\bar{r}^2}\,\frac{\sigma_0}{U_{ii}^2}\,G_Q\left(\frac{U_i}{U_{ii}}, \frac{v_q}{v_i}\right) \ . \tag{17}$$

We take $\bar{r} \simeq a_0/Z$ and $U_{ii} = \frac{1}{4}(Z^2e^2/2a_0)$, where a_0 is the Bohr radius, take $G_Q \simeq 0.3$, and find that

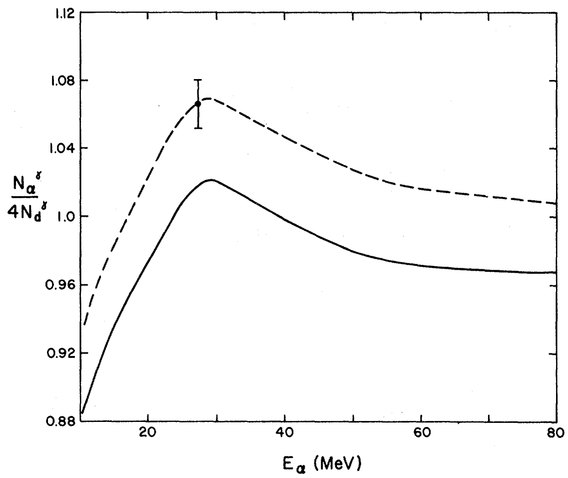

FIG. 1. $N_\alpha^\gamma/4N_d^\gamma$ vs bombarding α particle energy where N_α^γ (N_d^γ) is the number of K x rays per incident α particle (deuteron). Dashed curve represents experimental results of Lewis, Natowitz, and Watson for titanium. Equation (19) is shown by solid line. Born approximation for ratio of single-ionization cross sections is unity.

$$\xi \simeq 60(z^2/Z^2) \ . \tag{18}$$

This result must, of course, be modified for small Z, i.e., $Z \le 9$.

We may now estimate the correction due to double ionization of the result of LNW, who counted all N^γ photons corresponding to transitions into the K shell, using both alpha particles and deuterons as bombarding particles. Using (13) and (18) to estimate these corrections we have

$$\frac{\sigma_\alpha}{4\sigma_d} = \frac{N_\alpha{}^\gamma}{4N_d{}^\gamma}\frac{F_d}{F_\alpha} \simeq \frac{N_\alpha{}^\gamma}{4N_d{}^\gamma}\left[1 - \frac{3(\xi/z^2)}{2(3)}(1-F^0)\right]$$
$$= \frac{N_\alpha{}^\gamma}{4N_\alpha{}^\gamma}\left[1 - \frac{30}{Z^2}(1-F^0)\right] \ . \tag{19}$$

The ratio σ_α/σ_d is the ratio of total single-ionization cross sections.

A typical uncorrected result of LNW is shown for titanium ($Z = 22$) by the dashed line in the Fig. 1, and the results of our corrections are given by the solid line. At high energies this correction will damp out since the G_Q in Eq. (17) goes to zero (as do contributions from the other amplitudes which we have ignored). LNW have performed similar measurements for $17 \le Z \le 29$ with error of 2% or less. They find that the deviation from unity decreases with increasing Z; the maximum experimental deviation from Born approximation decreases from 25% at $Z = 17$ to 4% at $Z = 29$. Using the recent values of Walters and Bhalla[3] for F^0, we find that our correction also decreased from a contribution of 10% at $Z = 17$ to about 2% for $Z = 29$.

We conclude that, while classical calculations of the effects of double ionization on radiation rates, Auger rates, and fluorescence yields are probably correct only to within an order of magnitude, they strongly suggest the need to include double ionization when an accuracy of several percent or less is required over all.

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