Angular dependence of fluorescent x rays*

James H. Scofield
Lawrence Livermore Laboratory, University of California, Livermore, California 94550
(Received 21 June 1976)

The angular correlation between ionizing and fluorescent x rays is calculated including the dependence on the linear polarization of either x ray. The calculation is done relativistically including all the radiation multipoles using single-particle wave functions calculated in the Hartree-Slater model. The results are given for the ionization of a $2p_{3/2}$ electron with the subsequent filling by a $n = 3$ electron for $Z = 30$, 79, and 92.

INTRODUCTION

The radiation emitted by atoms which have been excited in an asymmetric manner will, in general, be asymmetric and polarized. In this paper we treat the example of the x rays emitted in filling an inner-shell vacancy created by x-ray ionization. The x rays emitted by states with angular momentum greater than $\frac{1}{2}$ will be correlated with respect to the direction of the ionizing x rays, and, if the initial x ray is linearly polarized or the final x ray's polarization is measured, there will be a correlation between the polarization direction and emission direction.

The ionization process leads to a particular density matrix of the magnetic substates of the intermediate state. The small contribution from scattering with the nonresonant intermediate states is neglected here. The subsequent decay from the intermediate state is nonisotropic. The density matrix resulting from photoionization has previously been examined by Flügge, Mehlhorn, and Schmidt[1] and by Oh and Pratt.[2] Flügge, Melhorn, and Schmidt use the nonrelativistic cross-section results of McGuire.[3] Oh and Pratt supplement nonrelativistic calculations with relativistic Born-approximation results. The present calculation is done relativistically, including the higher radiation multipoles. As in the other calculations, single-particle wave functions based on the Hartree-Slater theory are used. This same model is used for the treatment of the radiative decay of the single-vacancy state.

THEORY

The calculation is simplified by the technique for the treatment of angular correlations developed principally for nuclear transitions.[4] The density matrix of the intermediate atomic state is calculated in the coordinate system with the incoming photon along the z direction. By using the spherical tensor components of the density matrix, the transformation of the z axis to the outgoing photon's direction is easy. The dependence of the decay process on the density matrix is then calculated in this coordinate system. With linear polarization of the radiation included, the calculation is carried out with the polarization along the x direction; rotations taken into account are those of the initial polarization direction by the angle ϕ_1 into the scattering plane and of the final polarization direction away from it through the angle ψ_2. The expression for the angular correlation is thus a sum of terms, each with a factor dependent on the two separate transitions and the angular transformation.

With the final electron's direction not observed, the angular correlation between the ionizing and fluorescent x rays is proportional to

$$W = 1 + \sum_{k>0,\ \text{even}} \Big(B_k(\gamma_1)A_k(\gamma_2)\,P_k(\cos\theta) + Q_1 B_k^{\perp}(\gamma_1)A_k(\gamma_2)\,P_k^{(2)}(\cos\theta)\cos 2\phi_1 + Q_2 B_k(\gamma_1)A_k^{\perp}(\gamma_2)P_k^{(2)}(\cos\theta)\cos 2\psi_2 + Q_1 Q_2 \frac{(k+2)!}{(k-2)!}\,B_k^{\perp}A_k^{\perp}\,\frac{\mathrm{Re}[D_{22}^k(\phi_1,\theta,\psi_2) + D_{2-2}^k(\phi_1,\theta,\psi_2)]}{2}\Big).$$

Our notation and development follows that given by Steffen.[5] The $B_k(\gamma_1)$ and $A_k(\gamma_2)$ are the zero components of the spherical tensors of the density matrix for the initial ionizing and the final reradiating transitions, respectively. The $B_k^{\perp}(\gamma_1)$ and $A_k^{\perp}(\gamma_2)$ are proportional to the second component. P_k and $P_k^{(2)}$ are the Legendre and associated Legendre polynomials. The amount of polarization of the initial radiation is Q_1, and Q_2 is the response to the polarization of the emitted radiation. These range from 0 for no polarization to 1 for complete polarization. The $D_{ij}^k(\phi, \theta, \psi)$ are the components of the transformation matrix from the initial coordinate system to the final.

Lloyd[6] included the polarization-polarization term and gave the D matrix specialized to the

particular combination that occurs.

We use a single-particle description of the process. The dependence of the final-state wave function in the photoionization process on the total angular momentum due to the coupling between the outgoing electron and the vacancy is not included. This coupling may be important near the threshold for photoionization. The ionization and decay are here treated as occurring in complete subshells.

The B's and A's are given in terms of the reduced matrix elements of the radiation multipole fields between the single-particle states as

$$B_k(\gamma) = R^{-1} \sum_{LL'} F_k(LL' j_0 j_1) \times \langle \epsilon j_0 || A_L^{(\pi)} || j_1 \rangle \langle \epsilon j_0 || A_{L'}^{(\pi')} || j_1 \rangle^*,$$

$$B_k^{\perp}(\gamma) = R^{-1} \sum_{LL'} (-1)^{\Lambda_{\pi'}} f_k(LL') F_k(LL' j_0 j_1) \times \langle \epsilon j_0 || A_L^{(\pi)} || j_1 \rangle \langle \epsilon j_0 || A_{L'}^{(\pi')} || j_1 \rangle^*,$$

where

$$R = \sum_L |\langle \epsilon j_0 || A_L^{(\pi)} || j_1 \rangle|^2,$$

$$f_k(L, L') = - \left(\frac{(k-2)!}{(k+2)!} \right)^{1/2} \begin{pmatrix} L & L' & k \\ 1 & 1 & -2 \end{pmatrix} \Big/ \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix},$$

$$F_k(L, L', j_0, j_1) = (-1)^{j_1 + j_2 - 1} \times [(2k+1)(2L+1)(2L'+1)(2j_1+1)]^{1/2} \times \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix} \begin{Bmatrix} L & L' & k \\ j_1 & j_1 & j_0 \end{Bmatrix}.$$

Here j_0 denotes the angular momentum of the outgoing electron, and j_1 that of the subshell from which it was ionized; $\Lambda_{\pi'} = 0$ (1) for electric L' (magnetic) multipoles. The reduced matrix elements are defined by

$$\langle j_0 | A_{LM}^{(\pi)} | j_1 \rangle = (-1)^{j_0 - m_0} \begin{pmatrix} j_1 & L & j_0 \\ m_1 & M & m_0 \end{pmatrix} \langle j_0 || A_L^{(\pi)} || j_1 \rangle.$$

The expressions for $A_k(\gamma)$ and $A_k^{\perp}(\gamma)$ are obtained by replacing j_0 by j_2, the final-vacancy angular momentum. The extra phase factor which occurs in the treatment of two successive emissions does not enter.

We have previously given expressions for the reduced matrix elements.[7] The expressions as given yield imaginary values for some of the reduced matrix elements. Purely real expressions are obtained by including a factor i^l in the electrons' single-particle wave functions, where l is the orbital angular momentum.

TABLE I. Calculated values of A_2 and $A_2^{\perp}$ for the allowed L-to-M transitions.

Z		Ll	$L\alpha_2$	$L\alpha_1$	Average $L\alpha$
30	A_2	0.499	−0.399	0.101	0.051
	$A_2^{\perp}$	−0.250	0.200	−0.0498	−0.025
79	A_2	0.487	−0.392	0.115	0.063
	$A_2^{\perp}$	−0.252	0.199	−0.0485	−0.023
92	A_2	0.483	−0.390	0.120	0.068
	$A_2^{\perp}$	−0.253	0.199	−0.0479	−0.023

IONIZATION OF THE L_3 SUBSHELL

Here we present results for ionization of a $2p_{3/2}$ electron with subsequent filling of the vacancy by an M-shell electron. Calculations are performed for atomic numbers $Z = 30$, 79, and 92. For the $j = \frac{3}{2}$ intermediate state, only the $k = 0$ and 2 terms enter. Explicitly,

$$P_2(\mu) = \tfrac{1}{2}(3\mu^2 - 1), \quad P_2^{(2)}(\mu) = 3(1 - \mu^2).$$

The $E1$ transitions dominate the radiation and ionization processes for low energies. The value $f_2(1, 1) = \frac{1}{2}$ assures that for polarized radiation the entire dependence is relative to the polarization direction for the $E1$ transitions. Table I lists the values calculated for $A_2(\gamma_2)$ and $A_2^{\perp}(\gamma_2)$ for the M-to-L transitions: $Ll(L_3 - M_1)$, $L\alpha_2(L_3 - M_4)$, and $L\alpha_1(L_3 - M_5)$. The $L\alpha$ lines are not easily resolved experimentally; hence average values of A, weighted by the calculated emission rates are given. The radial matrix elements are calculated in the Hartree-Slater model as used previously.[8]

Angular correlations between the emitted x rays in a K-L-M vacancy cascade depend on the same A_2 factors. Experiments[9, 10] indicate good agreement with theory in the case of the $L\alpha$ component while the measured asymmetry coefficient for the Ll component is smaller than the theoretical value.

The photoionization matrix elements are calcu-

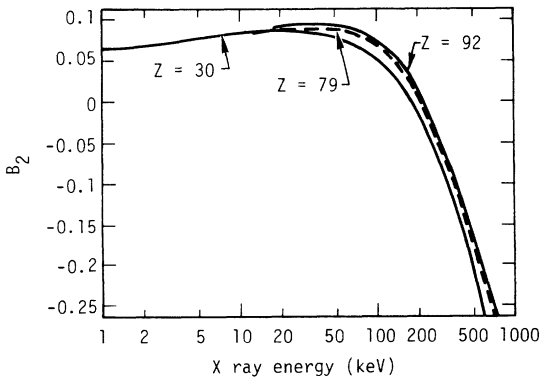

FIG. 1. Calculated values of B_2 as a function of x-ray energy for $Z = 30$, 79, and 92, for ionization of the L_3 subshell.

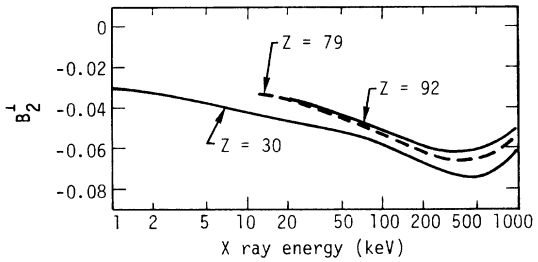

FIG. 2. Calculated values of $B_2^{\perp}$ as a function of x-ray energy.

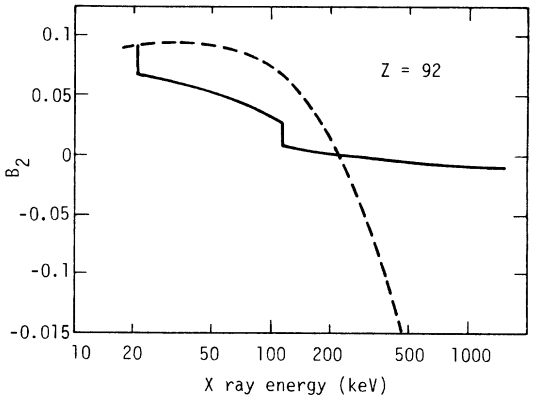

FIG. 3. Calculated value of B_2 for $Z=92$. The dashed curve is for ionization of the L_3 subshell alone; the solid curve includes the transfer of vacancies from the more tightly bound subshells.

lated in the same model as those for the decay, with the necessary modification for the normalization of the continuum wave functions. Figure 1 shows the B_2 value and Fig. 2 the $B_2^{\perp}$ value as a function of energy for $Z=30$, 79, and 92. For nonrelativistic energies, B_2 is an average of the values 0.5, −0.4, and 0.1 for the $E1$ transitions to the s, $d_{3/2}$, and $d_{5/2}$ continuum states. In terms of magnetic subshell populations, N_j of the intermediate state, B_2 can be expressed as $(N_{3/2}-N_{1/2})/(N_{3/2}+N_{1/2})$, with the populations depending only on the absolute value of the magnetic quantum number. Oh and Pratt[2] point out that in the nonrelativistic Born-approximation limit B_2 has the value of 0.1.

For photoionization energies above the limit for ionization of a more tightly bound subshell, radiative, Coster-Kronig, or Auger transitions may transfer the inner vacancy to the L_3 subshell, populating the magnetic substates equally. In principle, the small energy differences between the L x-ray diagram lines and the satellites following Coster-Kronig and Auger transitions could be used to discriminate against the latter. In Fig. 3 we show for $Z=92$ the value of B_2 diluted by the transitions originating from deeper-levels. The photoionization cross sections were calculated; Coster-Kronig coefficients $f_{12}=0.1$, $f_{23}=0.2$, and $f_{13}=0.6$ were interpolated from the values given by Bambynek *et al.*[11] The transfer probabilities from the K shell were taken from Table IV.VI of Ref. 11. For ionization energies above the K edge, the bulk of the vacancies in the L_3 subshell originate in the K shell.

For a particular incident x-ray energy and final x-ray transition, the appropriate values of B_2 and A_2 must be multiplied together to obtain the coefficient of P_2. For uranium we thus have a maximum value of $A_2 B_2 = 0.044$ for the Ll transition and 0.0062 for the $L\alpha$ average in the region below the L_2 edge.

---

*Work performed under the auspices of the U. S. Energy Research and Development Administration under Contract No. W-7405-Eng-48.

[1]S. Flügge, W. Mehlhorn, and V. Schmidt, Phys. Rev. Lett. 29, 7 (1972).

[2]S. D. Oh and R. H. Pratt, Phys. Rev. A 10, 1198 (1974).

[3]E. J. McGuire, Sandia Laboratories Report No. SC-RR-70-721, 1970 (unpublished).

[4]For reviews of the treatment of angular correlations, see L. C. Biedenharn and M. E. Rose, Rev. Mod. Phys. 25, 729 (1953); S. Devons and L. J. B. Goldfarb, *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1957), Vol. 42, p. 362; H. Frauenfelder and R. M. Steffen, *Alpha-, Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland, Amsterdam, 1966), Vol. 2, p. 977; and H. J. Rose and D. M. Brink, Rev. Mod. Phys. 39, 306 (1967).

[5]R. M. Steffen, Los Alamos Scientific Laboratory Report LA-4565-MS, 1971 (unpublished), available from the National Technical Information Service, Springfield, Virginia 22151. See also K. S. Krane and R. M. Steffen, Phys. Rev. C 2, 724 (1970).

[6]S. P. Lloyd, Phys. Rev. 85, 904 (1952).

[7]J. H. Scofield, *Atomic Inner-Shell Processes*, edited by B. Crasemann (Academic, New York, 1975), Vol. 1, p. 265.

[8]J. H. Scofield, Phys. Rev. 179, 9 (1969).

[9]E. S. Macias and M. R. Zalutsky, Phys. Rev. A 9, 2356 (1974) (gives a summary of earlier measurements).

[10]M. R. Zalutsky, E. S. Macias, and A. L. Catz, Phys. Rev. A 11, 75 (1975).

[11]W. Bambynek, B. Crasemann, R. W. Fink, H. U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. V. Rao, Rev. Mod. Phys. 44, 716 (1972).