We are grateful to Dr. William Shive, Dr. James E. Boggs, and Dr. R. Pettit of the Chemistry Department who made available the Raman spectrometer necessary for this work. We are indebted to Dr. W. W. Robertson, Dr. F. W. de Wette, Dr. A. A. Dougal, and Dr. W. C. Duesterhoeft, all of whom we thank for their support. We gratefully acknowledge a significant contribution for the purchase of a laser made by the University Research Institute.

*Work supported by the Joint Services Electronics Program under Contract No. F 44620-71-C-0091.

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L-Subshell Fluorescence Yields for Silver and Tellurium

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(Received 26 July 1972)

We have employed newly developed graphite mosaic crystals to observe the L x-ray spectrum from the decays of ^{109}Cd and ^{125}I. From the measured values of the subshell fluorescence yields, we conclude that either one or both of the Coster-Kronig transitions L_1-L_3M_4 and L_1-L_3M_5 is forbidden in both the silver and tellurium daughters.

Much of the recent progress in the measurement of L-subshell fluorescence yields may be traced to improvements in the resolution of lithium-drifted silicon, Si(Li), detectors.[1] Interest in these measurements stems from the sensitivity of the calculated values of the radiative and, particularly, of the radiationless transition probabilities to the proper choice of atomic wave functions. The Auger yields depend quite critically on the value chosen for the continuum-electron energy. A number of calculations of radiative and Auger yields have appeared recently in the literature which are in good agreement with experiment for high-Z atoms.[2,3] However, the resolution of the Si(Li) detector is not sufficient for study of atoms with $Z \leq 50$.

High-resolution crystal diffraction techniques are also inapplicable for the following reason. To deduce the fluorescence yields from the observed x-ray spectrum, knowledge of the primary vacancy distribution is necessary. This requirement is difficult to fulfill in x-ray or charged-particle excitation experiments since it is generally the photoelectric cross section or the excitation cross section that is itself the object of study. The primary vacancy problem has been solved by using radioactive sources, usually electron-capture β decays or excited nuclear lev-

els which have high internal conversion coefficients, for which the decay scheme is well known. Carrier-free sources (to minimize absorption) of strengths equal to hundreds of millicuries would be required to measure the L-subshell x-ray yields which are of the order of only a few percent for $Z \sim 50$.

We have circumvented both of these difficulties, that of resolution and of efficiency, by exploiting newly developed graphite mosaic crystals.[4] The full width at half-maximum resolution of these crystals in first order for the intense $L\alpha$ transition in silver and tellurium is 65 and 110 eV, respectively. In second order the resolution for the tellurium line is reduced to only 40 eV. Moreover, the efficiency of these crystals is between 1 and 2 orders of magnitude higher than for perfect crystals normally used for this range of x-ray energies. These unique properties should make the graphite mosaic crystals interesting to many physicists. In particular, the crystals are ideally suited to the task of extending the measurements of L-subshell fluorescence yields to the medium-Z region.

A second noteworthy feature in the x-ray spectra of elements with $Z \sim 50$ is the predicted discontinuity in the Coster-Kronig yields. A Coster-Kronig transition is an Auger transition in which a vacancy moves between subshells of a given shell. Thus, for example, a vacancy transfer between L_1 and L_3 with the ejection of an M_5 electron, or, alternatively, the ejection of an L_3 electron during a vacancy transfer between L_1 and M_5, is denoted as L_1-L_3M_5. Such transitions are energetically possible if the separation between the subshells is greater than the binding energy of the M_5 electron. This condition is fulfilled for atoms with $Z < 50$, but fails to be met by atoms with slightly higher atomic number. McGuire[5] has recently analyzed the silver x-ray spectrum recorded by Parratt in 1938.[6] He concludes that the experimental results are in better agreement with calculations if the Coster-Kronig transitions L_1-L_3M_4 and L_1-L_3M_5 are forbidden for silver ($Z = 47$). In addition, McGuire points out that the prediction of the Coster-Kronig yields depends strongly on the procedure used to estimate the continuum-electron energy from the known ionization thresholds.

Our experiment consisted essentially of measuring the intensities of the transition lines in the L-shell x-ray spectrum of the radioactive isotopes ^{125}I and ^{109}Cd. Samples of cyclotron-produced ^{109}Cd and ^{125}I were deposited on copper and silver foils, respectively, and covered with 1-mil beryllium foils. The sources were mounted on a vertical rod which could be rotated to any angle and also translated both horizontally and vertically. The emitted x rays were collimated to approximately 0.25° by a set of nickel slats and impinged on 6 in.^{2} of graphite crystals in the detection chamber. The crystals had a mosaic spread of 0.8°. Diffracted x rays were detected by an all-beryllium proportional counter, 8 in. high with a 2-in.-diam cylindrical detection volume, and a 1-mil beryllium window. Both crystals and counter were mounted on a θ-2θ goniometer mechanism. Source and detection chambers were both parts of a single vacuum system, separated by a $\frac{1}{2}$-in.-thick aluminum wall which served as the collimator holder.

The 2θ angle was advanced in steps of 0.25°, and data were accumulated at each position for intervals ranging from 5 min to 1 h, depending on the source strength. A 4-mCi iodine source was used to obtain the first- and second-order spectra. The second-order spectrum is shown in Fig. 1. An earlier run with a 1-mCi source had yielded essentially identical results for the first-order spectrum. For the cadmium work, a 1-mCi source was used, and the spectrum was observed in first order only. The Bragg condition in second order cannot be met for Cd L x rays and graphite. The results are shown in Fig. 2. The data were analyzed by superposing Gaussian functions, of width determined by the experimental resolution, at the positions predicted by Bragg's law. The measured height of each transition was corrected for transmission

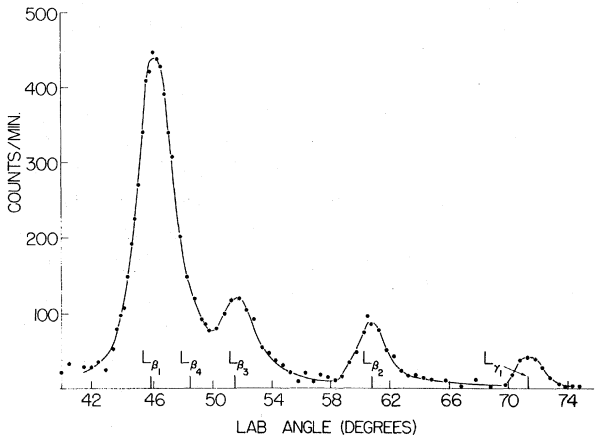

FIG. 1. Portion of the second-order tellurium L x-ray spectrum. The laboratory angle is equal to $2(90 - \theta_B)$, where θ_B is the Bragg angle.

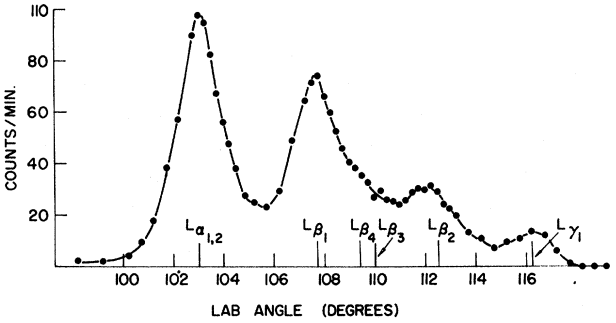

FIG. 2. Silver L x-ray spectrum in first order obtained with a 1 – mCi ^{109}Cd source.

through the beryllium of source and detector, for absorption in the argon of the proportional counter, and for the variation of the integrated reflectivity of the mosaic crystal with Bragg angle and wavelength. Our final results are given in Table I, where the intensity of each transition is expressed relative to the $L\alpha_{1,2}$ composite. The numbers in parentheses for the weakest silver transitions are not measured values. Since these transitions were too weak to be observed, we estimated their intensity relative to the stronger transitions to the same subshell, using the theoretical results of Scofield.[7]

Both ^{109}Cd and ^{125}I decay by 100% electron capture to the first excited state of the daughter nucleus. The L to K capture ratios in iodine[8] and in cadmium[9] have been measured. The multipolarity of the γ transition in the daughter, as well as its internal conversion coefficients, are well known. For iodine we use the values given by Karttunen, Freund, and Fink[10] and Geiger *et al.*,[11] while the values for cadmium are from Diethrich.[12]

TABLE I. Corrected relative intensities of L x-ray transitions observed in the decays of ^{109}Cd and ^{125}I.

Transition	Intensity in silver	Intensity in tellurium
$\eta\ L_2M_1$	(0.02)	0.04
$\beta_1\ L_2M_4$	0.88	0.60
$\gamma_1\ L_2N_4$	0.14	0.14
$l\ L_3M_1$	(0.03)	0.04
$\alpha_2\ L_3M_4$ / $\alpha_1\ L_3M_5$	1.00	1.00
$\beta_2\ L_3N_5$	0.27	0.23
$\beta_4\ L_1M_2$	0.12	0.12
$\beta_3\ L_1M_3$	0.16	0.23
$\gamma_2\ L_1N_2$ / $\gamma_3\ L_1N_3$	(0.05)	0.10

TABLE II. Electron capture probability ratio P_L/P_K and internal conversion coefficients for L and K shells, required for the calculations of the primary vacancy distribution.

	^{109}Cd	^{125}I
P_L/P_K	0.21	0.23
α_T	23	13.65
α_K/α_L	1.18	7.3
$\alpha_{L_1}:\alpha_{L_2}:\alpha_{L_3}$	25:100:98	100:8.9:2.4
n_{KL_1}	0.07	0.05
n_{KL_2}	0.32	0.31
n_{KL_3}	0.57	0.53

The average number of vacancies in the L_i ($i = 1, 2, 3$) subshell created in the filling of a K-shell vacancy is denoted by n_{KL_i}. These numbers have been computed most recently by Venugopala Rao, Chen, and Crasemann[13] and are given in Table II along with the other parameters required for calculating the primary vacancy distribution. The results of the vacancy calculation are presented in the first line of Table III.

Appropriate to the resolution of the work presented here, we may deduce the fluorescence yields ω_i from the following set of formulas:

$$I_{L_3} = (I_L/\bar{\omega}_L)[N_1(f_{13}+f_{12}f_{23})+N_2f_{23}+N_3]\,\omega_3,$$

$$I_{L_2} = (I_L/\bar{\omega}_L)(N_1f_{12}+N_2)\omega_2,$$

$$I_{L_1} = I_LN_1\omega_1/\bar{\omega}_L,$$

where I_L is the total intensity of all L x rays, I_{L_i} is the intensity of L x rays from transitions to the L_i subshell, f_{ij} are the Coster-Kronig transition probabilities, and $\bar{\omega}_L$ is the average L-shell fluorescence yield. Hohmuth, Muller, and Shintlmeister[14] have measured $\bar{\omega}_L$ precisely for the decays investigated in our work and have found the values given in Table III. If their experiment for silver is reanalyzed in terms of the more recent P_L/P_K ratio given in Table II, $\bar{\omega}_L = 0.046$ is obtained.[15] It is clear from the above equations that the individual subshell fluorescence yields are

TABLE III. Primary vacancy distributions and fluorescence yields for the silver and tellurium L x rays.

	Ag	Te
$N_1:N_2:N_3$	0.17:0.32:0.51	0.21:0.28:0.51
$\bar{\omega}_L$	0.046 ± 0.003	0.073 ± 0.007
$\omega_1/\bar{\omega}_L$	0.74 ± 0.07	0.85 ± 0.09
$\omega_2/\bar{\omega}_L$	1.11 ± 0.10	0.97 ± 0.10
$\omega_3/\bar{\omega}_L$	0.76 ± 0.08	0.76 ± 0.08

proportional to $\bar{\omega}_L$, and our results are best given as $\omega_i/\bar{\omega}_L$.

As of this writing, values for the f_{ij} are not available. Based on theory and on experiments at higher Z, the f_{ij} may be restricted to the following ranges: $0.1 < f_{12} < 0.3$, $0.1 < f_{23} < 0.2$, $0.3 < f_{13} < 0.8$. Note that the value of ω_1 is independent of the f_{ij}. We may evaluate minimum and maximum values of ω_2 and ω_3 corresponding to the limits of the f_{ij}, noting that the largest probability, f_{13}, is multiplied by the smallest vacancy number, N_1. Our results are summarized in Table III, where the errors span the minimum and maximum values.

For direct comparison with recent theoretical calculations, we multiply our results by the value of $\bar{\omega}_L$ determined by Hohmuth, Muller, and Shintlmeister to obtain the individual L-subshell fluorescence yields. For silver we find $\omega_1 = 0.034 \pm 0.003$, $\omega_2 = 0.051 \pm 0.005$, $\omega_3 = 0.035 \pm 0.004$. For iodine we deduce $\omega_1 = 0.062 \pm 0.006$, $\omega_2 = 0.071 \pm 0.007$, $\omega_3 = 0.056 \pm 0.006$.

The results for ω_2 are midway between the theoretical values predicted by Chen, Crasemann, and Kostroun[3] and by McGuire.[2] The high values of ω_1, roughly twice those tabulated in Ref. 1, are consistent with the recent re-evaluation of this quantity for silver by McGuire.[5]

For ω_3 we obtain experimental values lower than those calculated by Chen, Crasemann, and Kostroun and by McGuire. This may have the following explanation. In approximately 25% of the cases (for tellurium as well as for silver) the $L\alpha_{1,2}$ transitions occur in the presence of another L-shell hole (double ionization). The existence of this additional hole increases the effective nuclear charge so that $L\alpha_{1,2}$ transitions in the doubly ionized species may overlap $L\beta_1$ transitions in the singly ionized atoms. Similarly, some $L\beta_1$ x rays are shifted into the $L\beta_2$ group. The net effect is that I_{L_3} is reduced, while I_{L_2} is simultaneously increased. Corrections of roughly 15% to our measured intensities, to compensate for the effects of doubly ionized atoms, would bring the experimental values of ω_3 and ω_2 into very good agreement with the theoretical results of Chen, Crasemann, and Kostroun. A cumulative effect of this redistribution of x rays is that the ratio of $L\beta_2$ to $L\alpha_{1,2}$ x rays is increased. In fact, the measured value for this ratio is considerably higher than that predicted by Scofield.[7]

We interpret our results for ω_1 as indicating that the Coster-Kronig rates assumed for L_1-L_3M_4 and L_1-L_3M_5 are too large. The fluorescence yield ω_1 is computed from the equation

$$\omega_1 = \Gamma_R(L_1)/\Gamma(L_1),$$

where $\Gamma_R(L_1)$ is Scofield's radiative width,[7] and $\Gamma(L_1)$ is the total width, including that due to Coster-Kronig transitions. A decrease in the calculated value of $\Gamma(L_1)$ by roughly a factor of 2 is required to explain our results for both silver and tellurium. A decrease of this magnitude would be obtained if either one or both of the Coster-Kronig transitions L_1-L_3M_4 and L_1-L_3M_5 were forbidden for these elements.

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*Work supported in part by a grant from the Research Corporation, and by the National Science Foundation under Grant No. GP-34527.

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