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A New Method for Measuring Auger Transitions*

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The K fluorescence yields (w_K) of eight elements: zirconium, niobium, molybdenum, rhodium, palladium, silver, cadmium, and tin were measured by a new x-ray method involving the use of a NaI$-$Tl scintillation counter. This counter made it possible to detect the fluorescent x-rays radiated over an effective solid angle of 2π steradians. The values of w_K are Zr—0.645; Nb—0.713; Mo—0.714; Rh—0.779; Pd—0.782; Ag—0.814; Cd—0.819, and Sn—0.840, all with an accuracy of ±3 percent. These results confirm the theoretical predictions of Wentzel that $w_K/(1-w_K)$ is a function of Z^4, and they are in close agreement with the theoretically predicted values of Burhop and Pincherle.

INTRODUCTION

A KNOWLEDGE of the variation of the K fluorescence yield as a function of atomic number (Z) is of importance in studies of internal conversion of γ rays and of K capture.

Broyles, Thomas, and Haynes[1] have recently summarized most of the measurements of the Auger transitions and described the magnetic spectrograph, which they used to determine the K fluorescence yield (w_K) of Ba and Hg. Magnetic spectrographs have been used by several investigators[2–5] for the evaluation of the fluorescence yield of the elements of high atomic number ($Z>46$). These results are more accurate than those obtained with the older x-ray methods of Harms,[6] Compton,[7] Balderson,[8] and others.[9–14] Different observers have found differences of 25 percent in the measured values of w_K with elements of Z between 40 and 50 inclusive.

West, Meyerhof, and Hofstader[15] have shown that the NaI$-$Tl scintillation counter produces a voltage pulse proportional to the energy of the absorbed x-ray. The high density of the NaI crystal makes possible counters which are effectively 100 percent efficient for x-rays and sharply defines the effective region of the counter, thereby permitting accurate determination of the solid angle subtended by the counter. These features have made it possible to develop a method which has permitted more accurate measurement of the K fluorescence yields. The K fluorescence yields of eight of the elements with atomic numbers between 40 and 50 inclusive were determined by the use of the method described below.

EXPERIMENTAL PROCEDURE

Fluorescent radiation is emitted isotropically, and therefore the quantity of this radiation detected by a counter is strongly dependent upon the solid angle subtended by the counter. A counter was designed to detect this radiation over a solid angle of effectively 2π. If the primary radiation is well collimated, then by changing the solid angle subtended by the counter one can distinguish between the primary and fluorescent radiation. A diagram of the experimental equipment is shown in Fig. 1.

The power supply for the tungsten target x-ray tube both regulated the current and, using the Pepinsky[16] circuit for voltage regulation, stabilized the voltage to 1 part in 10 000. This supply has been briefly described by Swartz and Byerly.[17] Voltages of 35–40 kev and currents of 10 ma were used in this experiment.

The general radiation of the x-ray tube was passed through two sets of collimating slits and then diffracted by a calcite crystal. This monochromatic radiation was passed through the monitor slit which served to divide it, allowing part of the x-ray line to reach a Geiger-Müller counter (argon at 70 cm), used as a monitor, and the remaining part to reach the NaI$-$Tl scintillation counter.

A 3-mil foil of each of the elements investigated was placed in the path of the beam, almost in contact with

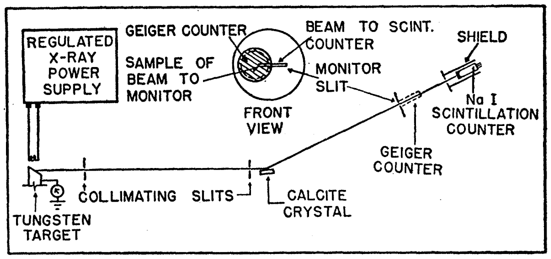

FIG. 1. Experimental arrangement.

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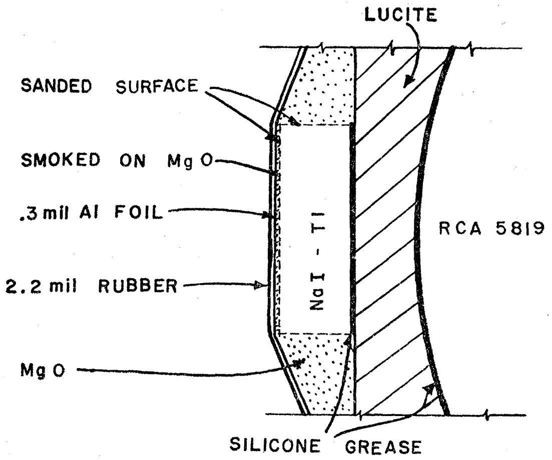

FIG. 2. Detail of NaI—Tl crystal mounting.

the surface of the NaI—Tl crystal, where it subtended a solid angle of effectively 2π steradians. In this position the counter detected both the fluorescent radiation and the portion of the original beam which was not absorbed in the foil. The same foil was then moved 35 cm away from the crystal and placed for convenience at the monitor slit. In this position the solid angle subtended by the counter was negligible. Since the fluorescent radiation is isotropic, less than 0.4 percent of it was detected, whereas the counter still detected the same quantity of the transmitted x-rays, except for the small effects of scattering. The difference between the readings obtained in the above two positions is therefore due to the fluorescent radiation. From the ratio of this fluorescent radiation to the intensity of the incident beam with no absorbers, the fluorescence yield can be obtained.

SCINTILLATION COUNTER MOUNTING

The mounting of the NaI—Tl crystal, which permits the foil to be placed closer than 4 mils to the crystal is shown in Fig. 2. The crystal was packed with MgO. In order to get a very thin layer (0.3 mil) on the front face of the crystal, the MgO was deposited there as smoke from burning Mg ribbon. This layer must be thin so that the x-rays are not appreciably absorbed. The Al foil (0.3 mil) serves as a light and moisture barrier and gives some mechanical protection to the smoked surface. The stretched 2.2-mil latex rubber holds the foil close to the surface of the crystal and gives the mounting more mechanical protection. The total thickness of all materials at the face of the crystal is 2.7 mils, which absorb less than 0.7 percent of the beam at the longest wavelengths (0.7284A) used in this experiment. With this mounting the width at half maximum of the 32-kev line was 28 percent.

RESULTS

A typical pulse-height spectrum obtained with the foil at the crystal is shown in Fig. 3. It is the sum of two basically Gaussian distributions—the spectra of the fluorescent and the transmitted incident radiation. The pulse-height distribution obtained with the foil 35 cm away from the crystal is the spectrum of the transmitted portion of the incident beam.

The two curves were subtracted graphically, and the broken line curve thus obtained represents the fluorescent radiation. This curve has its maximum at approximately the point where the K_α lines would be expected. The curve is not quite symmetrical, being broader on the high-energy side. This is to be expected since the contribution of the K_β line would tend to add more to the high-energy side.

One set of these curves was obtained for each of the eight elements investigated, although only the set for rhodium is shown here. The curves for zirconium and niobium showed almost complete separation of the peaks, while those for cadmium and tin showed relatively less separation. A pulse-height spectrum of the direct beam was also determined when the above curves were obtained.

In Fig. 3 it is seen that the points for higher pulse-height settings taken with the foil at the slit often lie slightly below the same points taken with the foil at the counter. This effect was observed on all of the eight sets of curves. Due to the Gaussian nature of these curves, the fluorescent radiation can be assumed to make no contribution at pulse heights of 36 to 46 volts. This difference can be attributed to scattering. The correction for this effect was made by drawing only one curve in the above region and then drawing the Gaussian curve for the transmitted radiation so that in the 17- to 27-volt region it is symmetric to the single curve drawn at 36 to 46 volts.

A planimeter was used to measure the difference between the curve for the transmitted x-rays drawn as mentioned above and the curve obtained for the combination of transmitted and fluorescent radiation (foil at crystal). The spectrum for the direct beam was plotted and measured and from the ratio of these two areas the ratio of fluorescent to incident radiation was determined. These ratios are given in the first line of Table I.

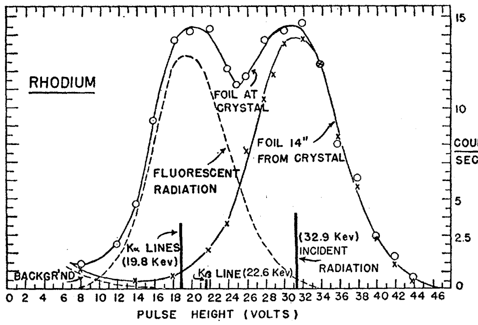

FIG. 3. Pulse height spectra obtained with 3-mil rhodium foil in beam of 32.9-kev x-rays.

TABLE I. Ratio of fluorescent to incident radiation.

Element	Zr	Nb	Mo	Rh	Pd	Ag	Cd	Sn
Graphs	0.0574	0.0633	0.0601	0.0502	0.0732	0.0853	0.1116	0.1410
Integrated counts	0.0613	0.0653	0.0625	0.0531	0.0755	0.0877	0.1121	0.1431
Corrected integrated counts	0.0573	0.0625	0.0604	0.0497	0.0739	0.0858	0.1104	0.1395

The above integrations could also be obtained directly by setting the pulse-height analyzer so as to count all pulses above 8 volts. It is possible to obtain statistics of 1–2 percent by this means in a relatively short time. The ratios obtained are given in the second line of Table I.

It is seen that the ratios for these integrated counts are all approximately 5 percent higher than those obtained with a planimeter. This is to be expected, since with this procedure allowance must still be made for scattering.

At wavelengths longer than that of the K edge, x-rays do not eject K electrons, and consequently no K fluorescent radiation is produced. Using these lower energy x-rays, the difference in the measured transmission of the foil in the two positions (at crystal and 35 cm away) will be due to the scattered x-rays. The ratio of these two readings gives $N_{35}/N_{\text{crystal}}=\exp(-\mu_\gamma t)$, where μ_γ is an "effective scattering coefficient" and t the thickness of the foil. The work of Coade[18] indicates that over the wavelength region between the K edge of the elements investigated and the wavelength of the incident radiation, the scattering coefficient should roughly decrease by a factor of two for all eight elements. If the ratio of transmitted to incident radiations is N_t, then

$$N_t[\exp(\mu_\gamma t/2)-1]=S,$$

where S is the fraction of the incident beam lost by scattering. The value of S was determined for each element, and subtracted from the directly integrated ratios in Table I to give the corrected values shown in line 3.

Since the effect of scattering is of the order of 5 percent of the fluorescent radiation, errors of even 20 and 40 percent in the application of this correction would cause errors of only 1 to 2 percent in the value of the fluorescence yields. Since the corrected values obtained by the two independent methods agree to within an average of 1 percent, it seems reasonable to conclude that the total error due to the effects of scattering is less than 2 percent. Because of their better statistics, the integrated counts are considered more reliable than the data from the pulse-height spectra graphs.

There are, in addition to the above, other sources of error to be considered. As was mentioned earlier, when the foil is 35 cm from the crystal, 0.4 percent of the fluorescent radiation is still detected by the counter.

The rubber, Al foil, and MgO at the face of the NaI−Tl crystal will absorb more of the fluorescent than primary radiation. This is due to the longer wavelengths of the fluorescent radiation and also to the fact that it has a longer average path in the mounting materials. The primary radiation, unlike the fluorescent, is collimated and its path is therefore always perpendicular to the crystal surface. The effect of this greater absorption is also 0.4 to 0.3 percent, and the result of the above two effects is to reduce the experimental value of the measured fluorescence radiation 0.8 percent.

The background counting rate for pulse heights corresponding to photons of 3–12 kev is observed to depend upon the history of the NaI−Tl crystal. This effect was independently observed earlier at this university by Class.[19] Megill and Emigh[20] have also observed the long-term phosphorescence of the NaI−Tl crystal. Exposure to light can increase the background rate in the 6-kev region as much as 5 times, and in the 10-kev region 2 times. This increased rate decays with a half-life on the order of 10 minutes. The exposure of the crystal to an intense beam of 30-kev x-rays can also raise this background level 5–10 percent. This effect, which perhaps can be attributed to phosphorescence, varies very much with different crystals.

With the use of a crystal selected so that this phosphorescence was reduced, this effect still caused the measured fluorescence value to be approximately 0.6 percent too high. This error effectively cancels with the 0.8-percent error mentioned above.

The important sources of error in these experimental ratios are either statistical in origin or due to effects of scattering. These errors are each less than 2 percent.

RELATION OF EXPERIMENTAL RATIOS TO FLUORESCENT YIELD

It can be shown that the number of incident x-rays (dI_x) absorbed photoelectrically by the ejection of a K photoelectron from atoms in a layer dx of the foil is given by

$$dI_x=\tau\frac{(r-1)}{r}N_0e^{-\mu_1 x}dx, \tag{1}$$

where τ is the photoelectric absorption coefficient; μ_1, the total absorption coefficient; r, the absorption jump ratio; and N_0, the number of photons in the direct beam.

Because of the Auger effect only a fraction w_K (the K fluorescence yield) of those atoms which lose a K electron will radiate a K x-ray. These fluorescent K x-rays must, however, pass through the rest of the foil before

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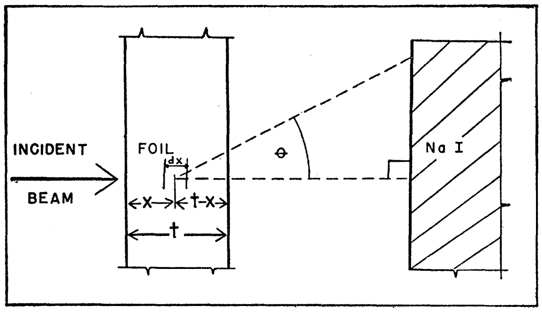

FIG. 4. Diagram showing relationship of t, θ, x, and dx to the foil and NaI−Tl crystal.

they are detected by the crystal and many are therefore reabsorbed. Furthermore, they are radiated isotropically, and the thickness of the foil through which they must pass depends upon the angle θ as seen in Fig. 4. Therefore, the number (dI_j) of K x-rays from the layer dx which are detected by the counter is given by

$$dI_j=\frac{dI_x w_K}{2}\int_0^{\pi/2} e^{-\mu_j(t-x)\sec\theta}\sin\theta d\theta dx, \qquad (2)$$

where t is the thickness of the foil. The difference in the integral between $\theta=90°$ and the actual experimental θ of 89° is less than 0.01 percent.

Allowing for the difference in absorption coefficients of the α_1, α_2, and β lines in the K series, and integrating expression (2) over all dx, the following expression is obtained:

$$I_T=\sum_j C_j\int_0^t dI_j dx=\tau\frac{w_K I_0(r-1)}{2r}\sum_j C_j\int_0^t e^{-\mu_1 x}$$
$$\times\int_0^{\pi/2} e^{-\mu_j(t-x)\sec\theta}\sin\theta d\theta dx=\frac{w_K I_0}{r}(r-1)\sum_j C_j\Gamma_j, \qquad (3)$$

where C_j is the intensity of each line relative to the total intensity of the K series, and Γ_j is defined to be $\frac{1}{2}\tau$ times the double integral.

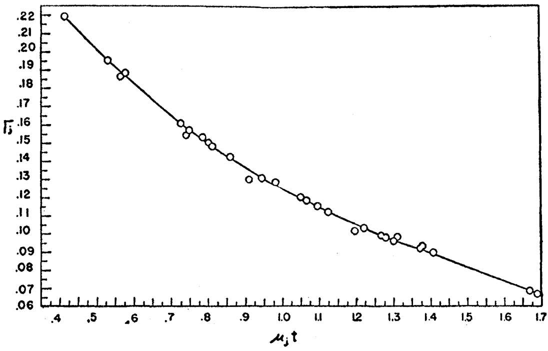

FIG. 5. Plot of

$$\frac{\tau}{2}\int_0^t e^{-\mu_1 x}\int_1^\infty \frac{e^{-\mu_j(t-x)v}}{v^2}dvdx$$

or Γ_j as a function of $\mu_j t$.

If $\sec\theta$ is set equal to v and substituted in the last integral in expression (3), one obtains

$$\int_0^{\pi/2} e^{-\mu_j(t-x)\sec\theta}\sin\theta d\theta=\int_1^\infty \frac{e^{-av}}{v^2}dv, \qquad (4)$$

where $a=\mu_j(t-x)$. This integral is recognized as the Gold[21] integral, which has been tabulated.

Using Gold's values for part of the integral, it was relatively simple to evaluate Γ_j using a small electronic computer (IBM-604). The values of Γ_j obtained are plotted against $\mu_j t$ in Fig. 5. The values of τ and $\mu_1 t$ may vary by more than 100 percent for neighboring points on the curve; yet the value of Γ_j is seen to be primarily a function of $\mu_j t$.

The foil thicknesses—3 mil—were chosen so that $\mu_j t$ was approximately equal to 1, the optimum value for accurate measurement. The same part of the same foil was used for both the fluorescent and absorption measurements eliminating errors due to variations in thickness of the foil and small impurities. Furthermore, it is not necessary to measure t with precision since the fluorescence yield depends upon $\mu_1 t$ and $\mu_j t$, which were obtained directly from the experimental measurements.

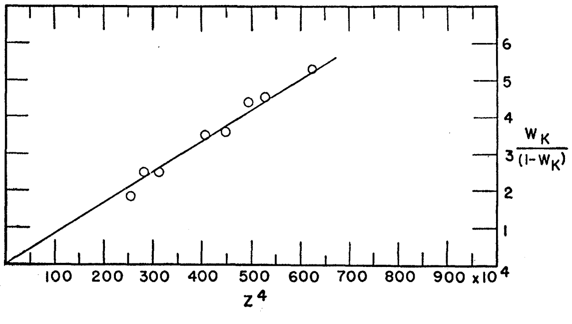

FIG. 6. Plot of Z^4 vs $w_k/(1-w_k)$.

Wavelengths corresponding to those of the $K_{\alpha 1}$, $K_{\alpha 2}$, and K_β lines of the eight elements were isolated from the general radiation of tungsten by the 1.5-meter Bragg single crystal spectrometer. This spectrometer was calibrated and checked by use of the K absorption limits of tin, silver, cadmium, molybdenum, and zirconium.

Before evaluation of the K fluorescence yield it is also necessary to know the absorption jump ratio. The experimental values obtained by different investigators range from 6.7 [22] to 7.8 [23] in the case of silver. Despite this uncertainty in the value of r, the above values would cause only a 1.3-percent difference in the value of $(r-1)/r$. This difference is, however, of the same order of magnitude as the errors probably introduced by the scattering corrections.

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TABLE II. Values of K fluorescence yields.

Element	Zr	Nb	Mo	Rh	Pd	Ag	Cd	Sn
Present results	0.645	0.713	0.714	0.779	0.781	0.813	0.819	0.840
Theoretical[a]	0.67		0.71	0.75	0.78	0.79	0.81	0.83
Magnetic spectrograph[b]						0.83	0.83	
Stephanson[c]	0.69		0.735	0.77		0.81	0.79	0.81
Backhurst[d]			0.785	0.801	0.835	0.838	0.846	0.855
Arends[e]			0.724			0.795		0.825
Berkey[f]			0.79			0.72	0.70	0.66
Balderson[g]			0.97			0.88		
Compton[h]			0.68					

[a] See references 25 and 26.
[b] See reference 2.
[c] See reference 9.
[d] See reference 11.
[e] See reference 12.
[f] See reference 13.
[g] See reference 8.
[h] See reference 7.

FINAL RESULTS

Using the relative intensity of the K lines, from Williams,[24] the weighted contribution of each integral was obtained and summed. With this value, the ratio of fluorescent to incident x-rays, and the value of $(r-1)/r$ which was chosen to be 0.860 for all elements, Eq. (3) was evaluated for w_k. The results are shown in Table II.[2,7–9,11–13,25,26]

The agreement between the scintillation counter results and the theoretical results of Burhop[25] and Pincherle[26] is excellent, in view of the approximations made in the theory. The values for silver and cadmium obtained with the magnetic spectrometer are little over one percent higher than the results from the scintillation counter method. Two samples of silver of different thicknesses were measured as a check on the consistency of the results. The value obtained for the K fluorescence yield of the first sample was 0.817 and that of the second was 0.810.

Wentzel[27] has predicted that $w_K/(1-w_K)$ should vary as the fourth power of Z. The scintillation counter values are seen to give a very close fit to this relationship for the elements studied, as seen in Fig. 6. For $Z>50$, it is quite possible that due to the effects of screening and relativity this relationship is no longer valid, as Broyles, Thomas, and Haynes[1] have mentioned.

With further improvement in scintillation counter techniques, it will be relatively simple to extend this method to the measurement of the L fluorescence yields of elements such as lead and tungsten and to measure the K fluorescence yields of elements such as iron and copper. By making the well-known correction for K escape of iodine,[17] this method also permits rapid and accurate measurement of the K fluorescence yields of elements of high atomic number. Improvement in resolution of the scintillation counter should make it possible to make more accurate corrections for scattering.

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