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Angular Correlation between K and L X Rays in Pb^\dagger

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(Received 12 March 1970)

Angular correlations between K and L x rays in Pb were measured. $NaI(Tl)$ and $Ge(Li)$ detectors were used for the detection of K and L x rays, respectively. The angular correlation functions for the $K-L\alpha$, $K-L\beta$, and $K-Ll$ cascades were found to be $W(\theta) = 1 + (4.13 \pm 0.36) \times 10^{-2} P_2(\cos\theta)$, $W(\theta) = 1 + (1.22 \pm 0.43) \times 10^{-2} P_2(\cos\theta)$, and $W(\theta) = 1 + (23.32 \pm 2.06) \times 10^{-2} P_2(\cos\theta)$, respectively. The results for the $K-L\alpha$ cascade demonstrate the contribution of magnetic quadrupole radiation to the x-ray transitions. When this contribution is taken into account, relatively good agreement with theory is obtained in all cases. Better agreement would be obtained for slightly higher (than theoretically calculated) M_2 contributions to the L x-ray transitions.

I. INTRODUCTION

An atom with a missing electron in one of its inner shells is in a highly excited state from which it deexcites to its ground state through different intermediate states by the emission of x rays and Auger electrons. A partial atomic decay scheme which includes the transitions of interest in the present work is shown in Fig. 1.¹

According to the general theory of angular correlation of successive radiations,² the x rays emitted successively in the process of atomic deexcitation should be directionally correlated.^{3,4} The angular correlation function $W(\theta)$ which is defined as the probability W for two successive x rays to be emitted each into one unit of solid angle in directions at an angle of θ from one another is of the form²

$$W(\theta) = 1 + A_{22}P_2(\cos\theta) + \dots + A_{k \max k \max}P_{k \max}(\cos\theta), \quad (1)$$

where $P_k(\cos\theta)$ are the Legendre polynomials of order k and the coefficients A_{kk} are functions of the atomic angular momenta in the states between which the two successive transitions occur, as well as of the multipolarities of the radiations emitted in the two transitions. For two consecutive x-ray transitions occurring when the excited atom, initially in a state with angular momentum l_i deexcites to a state with angular momentum l_f through an intermediate state with angular momentum I , the coefficients A_{kk} can be expressed as products of two factors, $A_k(1)$ and $A_k(2)$, each of which depends on only one of the transitions involved in the following way:

$$A_k(1) = \frac{F_k(L_1 L_1' I) + 2\delta_1 F_k(L_1 L_1' I) + \delta_1^2 F_k(L_1 L_1' I)}{(1 + \delta_1^2)},$$

$$A_k(2) = \frac{F_k(L_2 L_2' I) + 2\delta_2 F_k(L_2 L_2' I) + \delta_2^2 F_k(L_2 L_2' I)}{1 + \delta_2^2}.$$

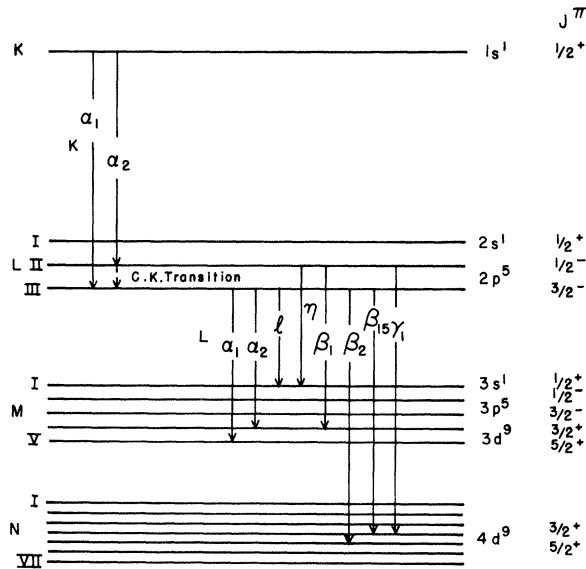


FIG. 1. Partial atomic decay scheme which includes the transitions of interest in the present work.

L_1 , L'_1 and L_2 , L'_2 are the multipolarities of the radiations emitted in the first and the second x-ray transitions, respectively; δ_1 and δ_2 are the mixing ratios of the radiations emitted in the first and second transition, respectively,⁵ and F_k are functions which can be found tabulated in literature⁶ for all cases of interest.

It is assumed in the present discussion that the angular correlation between the x rays is not perturbed by interactions with external fields of the atom in the intermediate state of the x-ray cascade. The magnitude of such possibly perturbing interactions with the nucleus and with the magnetic field due to an incomplete outer atomic shell was discussed by Moellering and Jensen³ and by Beste,⁷ respectively, and found to be negligible in comparison to the natural width of the intermediate atomic level.

Experimentally, the angular correlation between successive x rays has been investigated so far in only a few cases.⁷⁻¹⁰ With the exception of one measurement⁹ the detectors employed, NaI(Tl) crystals^{7,10} and gaseous proportional counters,⁸ did not possess the energy resolution necessary in order to distinguish between the different x-ray transitions involved. The measurements could therefore determine only angular correlation functions averaged over several unresolved cascades. (See, however, Ref. 19.)

Beste⁷ measured the angular correlation between K and L x-rays in lanthanum and found an average anisotropy

$$A = [W(180^\circ) - W(90^\circ)]/W(90^\circ)$$

of $(8 \pm 6)\%$. Konstantinov and Sazonova⁸ measured the angular correlation between K and L x rays in terbium, reporting an average anisotropy A of 16%. No errors are reported in this measurement nor is it clear on what grounds the authors justify their statement that the theoretically expected anisotropy is 16%.

In the present work the angular correlation function was measured for several K - L x-ray cascades in Pb. The L x-rays were detected with a cooled Ge(Li) solid-state spectrometer which enabled the resolution of the L x ray spectrum into the Ll and the $L\alpha$, $L\beta$, and $L\gamma$ groups and thus enabled the measurement of the angular correlations between the K x rays and each of these groups of L x rays, separately.

II. EXPERIMENTAL SET UP

A. X-Ray Source

The Pb x rays were obtained from a source of radioactive 30-year ^{207}Bi which decays to ^{207}Pb by electron capture,¹¹ predominantly by K electron capture, initiating K - L x-ray cascades in ^{207}Pb . A source of ^{207}Bi of 2-mm diameter and of about 40- μCi intensity was prepared by depositing and drying a droplet of ^{207}Bi in HNO_3 solution, on a 1-mil-thick Mylar sheet. The source was placed in the center of an angular correlation table where it was viewed by the K and L x-ray detectors which were capable of being moved independently on the circumference of the table.

B. X-Ray Detectors

A cooled Ge(Li) x-ray spectrometer of cylindrical shape with a diameter of 8 mm and a sensitive depth of 5 mm was used for the detection of the L x rays. The detector, which for operational reasons has to be kept under vacuum and at liquid-nitrogen temperature, was enclosed in a stainless-steel container which admitted the x rays through a 0.25-mm-thick beryllium window. Throughout the measurements the L x-ray detector was kept fixed with the front face of the Ge(Li) crystal at a distance of 29 mm from the ^{207}Bi source. The energy resolution of the Ge(Li) spectrometer was 440 eV [full width at half-maximum (FWHM)] at an energy of 14.4 keV.

The K x rays were detected with a 37-mm-diameter 31-mm-thick NaI(Tl) crystal optically coupled to a RCA 6342 photomultiplier. This detector moved on the circumference of the angular correlation table with the front face of the NaI(Tl) crystal at a distance of 45 mm from the source.

In order to minimize x-ray scattering from one

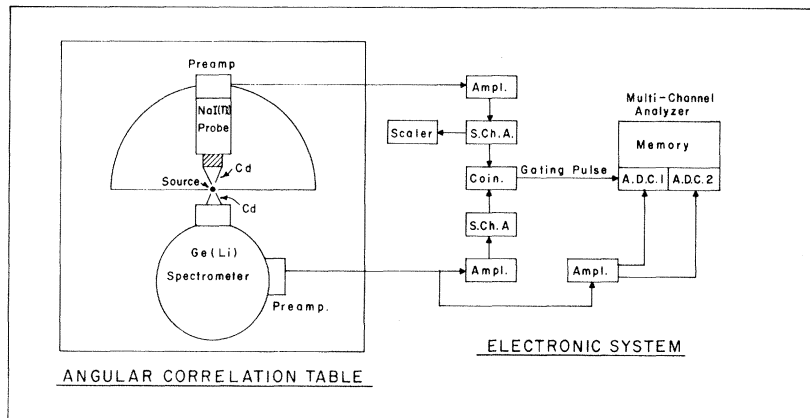


FIG. 2. Schematic diagram of the experimental setup.

detector into another, both detectors were shielded from the sides by cadmium (420 mg/cm^2) and viewed the source through conical cadmium collimators (see Fig. 2).

C. Electronic System

The pulses from the Ge(Li) crystal were passed through a charge-sensitive low-noise preamplifier with a cooled field-effect transistor into an amplifier adjusted for optimal time resolution, and from it the pulses went into a single-channel analyzer, the window of which was set to allow passage of the Pb L x-ray spectrum. Parallel to that, the output of the low-noise preamplifier was also routed to an additional amplifier with shaping time constants set for optimal energy resolution. This amplifier provided two outputs which were conveyed to two analog-to-digital converters (ADC) and were recorded upon analysis into different halves of a 1024 channel memory. The output of one ADC was recorded only when it occurred in coincidence with the detection of a K x ray in the NaI(Tl) crystal. The pulses from the NaI(Tl) detector were similarly passed through a preamplifier, an amplifier, and a single-channel analyzer, the window of which was set to pass the Pb K x-ray spectrum. The outputs of the two single-channel analyzers were introduced into a coincidence system with a resolving time 2τ of 42 nsec. The output of this coincidence system served as an opening gate for the ADC which, as previously mentioned, was set to analyze the L x rays detected in the Ge(Li) crystal only when their detection was accompanied by the detection of a K x ray in the NaI(Tl) crystal. The output of the single-channel analyzer on the K x-ray side was also connected to a scaler, which recorded the number of K x rays detected in the movable NaI(Tl) crystal at each angle of measurement. This number served as

an indication of the degree in which the position of the source differed from strict center and according to it the results of the measurements taken at the different angles were corrected before comparison.

A schematic diagram of the electronic system appears in Fig. 2. Typical L and K x-ray spectra of ^{207}Pb recorded with the Ge(Li) and NaI(Tl) crystals, respectively, are shown in Figs. 3 and 4, and spectra of L x rays detected with the Ge(Li) crystal in coincidence with K x rays detected with the NaI(Tl) crystal are shown in Fig. 5.

III. DATA ACQUISITION AND ANALYSIS

The spectrum of L x rays detected with the Ge(Li) crystal occurring in coincidence with K x rays detected with the NaI(Tl) crystal was measured for different positions of the NaI(Tl) crystal, such that the angles between the directions of the radiations striking the two crystals ranged from 90° to 270° in steps of 30° . The measurements at each angle were of short duration (30 min) and the angles were selected at random in order to minimize the influence of possible instabilities in the electronic system. In addition, in every measurement the spectrum of L x rays detected with the Ge(Li) crystal, without the coincidence requirement, was also simultaneously recorded, as was the number of output pulses from the single-channel analyzer set to pass the K x rays detected with the NaI(Tl) crystal. These data provided monitoring of the stability of every part of the system. Every run consisted of eight measurements, two at 180° and one at each of the other angles.

The data accumulated during a run were treated in the following way: First, the data recorded at each pair of angles symmetric about 180° were added together as were the numbers of the single-channel-analyzer output pulses for the respective

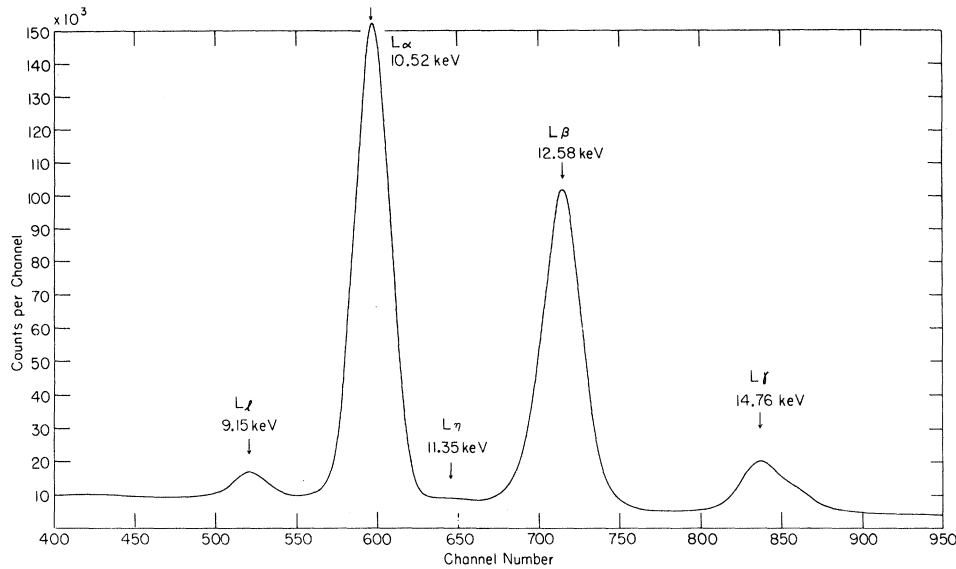


FIG. 3. Spectrum of Pb L x rays measured with the Ge(Li) crystal.

measurements. The number of counts recorded in each pair of measurements in the Ll , $L\alpha$, and $L\beta$ peaks were then calculated (in Fig. 5 the parts of the peaks used in this calculation are marked) and the results, for each of the four pairs of angles were normalized according to the number of the single-channel-analyzer output pulses for the respective pair. Typical normalization factors were 1.000 ± 0.002 ; in only one case was

a normalization factor of 1.004 encountered. Prior to the normalization, background was removed from the Ll peak (9.15 keV).

It was assumed that the background per channel under the Ll peak should be equal to the number of counts accumulated per channel in the region of the spectrum near 7.8 keV, namely, immediately below the Ll peak. It was found out, however, that the single-channel analyzer – the window of which was set to pass the L x-ray spectrum detected with the Ge(Li) crystal – did not pass with full and equal efficiency the parts of the spectrum near the lower and upper thresholds of the window. Since both the Ll peak and the region of the spectrum in the vicinity of 7.8 keV are near the lower threshold of the single-channel-analyzer window, they were passed by the single-channel analyzer with different efficiencies. These efficiencies were determined experimentally, by comparing an L x-ray spectrum accumulated in coincidence with the output pulses of the single-channel analyzer to an L x-ray spectrum accumulated for an equal time without the above coincidence requirement, and the correction was applied accordingly. The contribution of the background to the number of counts in the central parts of the $L\alpha$ and $L\beta$ peaks which were analyzed is very small and was ignored. The spectrum of L x rays detected with the stationary Ge(Li) crystal without the coincidence requirement was essentially identical in all the measurements. Finally, the corrected data accumulated in all the runs, for each pair of angles and each L x-ray peak, were added together and from them the contribution of random coincidences as well as of coincidences due to the

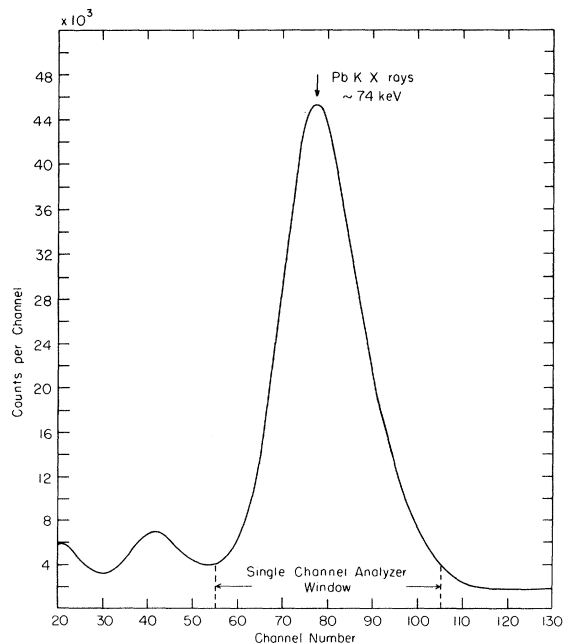


FIG. 4. Spectrum of Pb K x rays measured with the NaI(Tl) crystal.

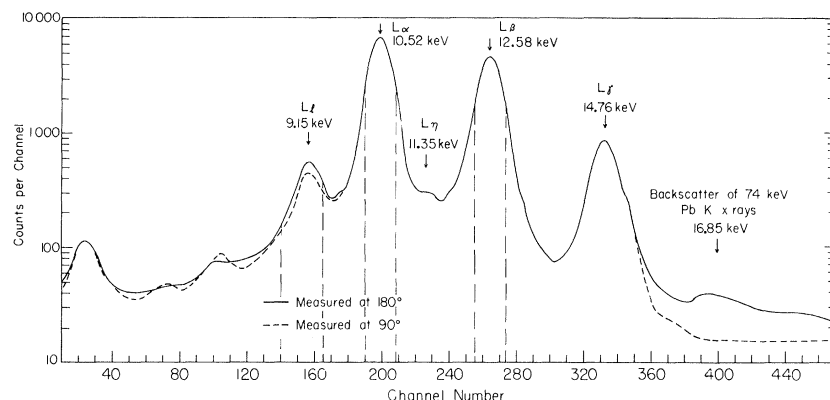


FIG. 5. Spectra of Pb L x rays occurring in coincidence with K x rays, as measured when the angle between the directions of the radiations striking the two detectors were 90° and 180° . Broken lines indicate the parts of the $L\alpha$, $L\beta$, and $L\gamma$ peaks used in the calculations of the angular correlation functions. The contribution to the L x-ray spectrum, measured at 180° of scattered radiation, is evident at the energy of 16.8 keV.

detection of a ^{207}Bi γ ray in the $\text{NaI}(\text{Tl})$ crystal accompanying the detection of an L x ray in the $\text{Ge}(\text{Li})$ crystal, were removed. These contributions were found experimentally to amount together to 6.8, 4.9, and 4.9% of the net number of counts in the $L\alpha$, $L\beta$, and $L\gamma$ peaks, respectively, on the average over the angles, these contributions not being angle dependent.

The contribution to the measured L x-ray spectrum of erroneous coincidences due to scattering of radiation from one detector into another was also investigated. Such a contribution occurs when a $\text{Pb } K$ x ray (~ 74 keV) is Compton scattered by an electron in the $\text{Ge}(\text{Li})$ detector which cannot resolve it from an unscattered $^{207}\text{Pb } K$ x ray. When the detectors are at a 180° angle to one another, the energy imparted to the electron in the $\text{Ge}(\text{Li})$ crystal is ~ 16.6 keV. The contribution of this effect in this case is clearly visible in Fig. 5. However, this contribution is concentrated in the part of the spectrum around 16.6 keV, and it does not affect the other parts of the L x-ray spectrum. The cadmium shielding around the detectors prevented this effect from occurring in the measurements performed at angles other than 180° . This fact was ascertained by measurements performed at all angles and in which an iron absorber of 69 mg/cm^2 was placed between the ^{207}Bi source and the $\text{Ge}(\text{Li})$ detector. This absorber prevented most of the L x rays, originating in the source, from reaching the $\text{Ge}(\text{Li})$ crystal while passing most of the K x rays. In this way the effects of scattering were evaluated and, being found very small, i. e., of the order of 0.1 to 0.2% of the total number of counts in the peaks, they were neglected.

Additional possibilities of obtaining K - L x-ray coincidences which could complicate the data analysis exist in the following cases: (i) when the decay by K or L electron capture of the parent nucleus leads to an excited state of the daughter

nucleus and when, furthermore, this state is de-excited by internal conversion (followed, of course, by emission of daughter's characteristic x rays) within a time interval shorter than or comparable to the resolving time of the coincidence system; and (ii) when the decay of the parent nucleus leads to excited states of the daughter nucleus which decay to the ground state via short-lived (as compared to the resolving time of the coincidence system) intermediate states, and where, furthermore, at least two transitions in each cascade proceed partially by internal conversion.

The number of coincidences of this kind in the present measurement is negligibly small due in part to the fact that in 88% of the cases the decay of ^{207}Bi leads to a long-lived (0.8 sec)¹¹ excited state of ^{207}Pb , and in part to the fact that in all the cascading transitions in ^{207}Pb the internal conversion coefficients are relatively small ($\alpha_K < 0.1$).

The number of such K - L x-ray coincidences was estimated to contribute not more than 1% of the total number of K - L x-ray coincidences accumulated in the present work.

Since, in addition, the rate of occurrence of such coincidences is independent of the angle of measurement, no correction to the data in this respect was made. The possibilities of obtaining such coincidences were mentioned, nevertheless, since they might be extremely important in other cases where radioactive sources are used as x-ray sources in angular correlation measurements.

IV. RESULTS

A. K - $L\alpha$ and K - $L\beta$ Cascades

The final results for these cascades were fitted by a weighted least-square procedure to expressions of the form $N(\theta) = N_0 + N_2 P_2(\cos\theta)$, where $N(\theta)$ is the number of counts accumulated at the angle θ , all corrections included. The fitted curves are shown in Fig. 6. The values of N_2/N_0

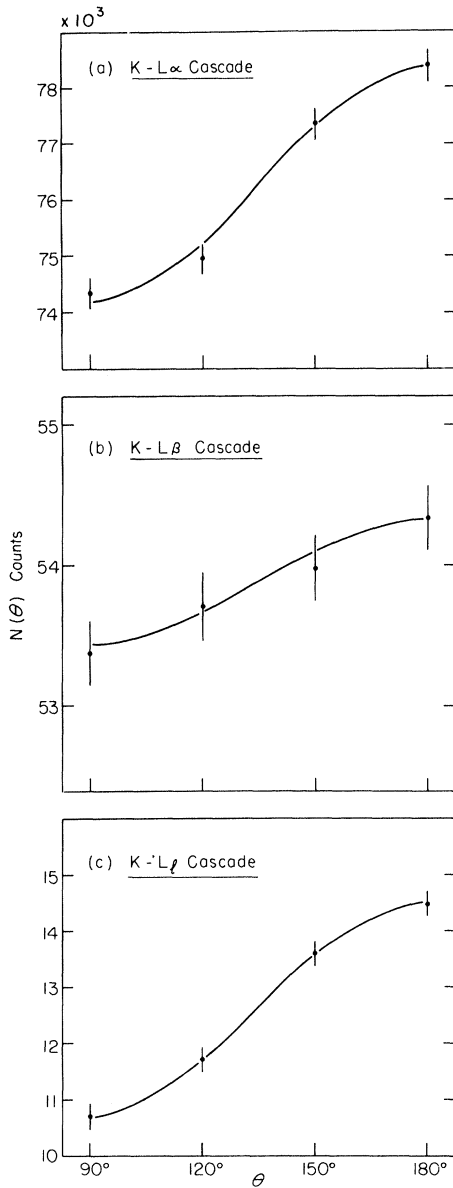


FIG. 6. Results of angular correlation measurements of *K-Lα*, *K-Lβ*, and *K-Ll* cascades in Pb. (a) *K-Lα* cascade: Smooth line represents the function $N(\theta) = 75562 + 2808 P_2(\cos\theta)$. (b) *K-Lβ* cascade: Smooth line represents the function $N(\theta) = 53735 + 589 P_2(\cos\theta)$. (c) *K-Ll* cascade: The smooth line represents the function $N(\theta) = 11999 + 2519 P_2(\cos\theta)$.

for the two cascades were found to be

$$N_2/N_0 = (3.72 \pm 0.32) \times 10^{-2} \quad \text{for the } K-L\alpha \text{ cascade;}$$

$$N_2/N_0 = (1.10 \pm 0.38) \times 10^{-2} \quad \text{for the } K-L\beta \text{ cascade.}$$

These results have to be corrected for the effect of the finite solid angles subtended by the two detectors before they can be compared to the theo-

retically predicted A_{22} values [Eq. (1)] for the respective cascades.

The correction for the finite solid angle of the Ge(Li) detector is negligible due to its small size, while the correction for the solid angle of the NaI(Tl) detector, in the geometry in which it was employed in the present measurements, was evaluated from literature¹² to amount to a multiplication factor of 1.11. After effecting these corrections the measured angular correlation functions can be written as

$$W(\theta) = 1 + (4.13 \pm 0.36) \times 10^{-2} P_2(\cos\theta)$$

for the *K-Lα* cascade;

$$W(\theta) = 1 + (1.22 \pm 0.42) \times 10^{-2} P_2(\cos\theta)$$

for the *K-Lβ* cascade.

B. *K-Ll* Cascade

In addition to the previously described runs of short-duration measurements in which data were accumulated for all the three cascades simultaneously, several runs of long-duration (~450 min) measurements were performed at all angles of measurement, in order to acquire additional data for the lower-intensity *K-Ll* cascade. The data accumulated for this cascade at different angles were compared and normalized according to the data accumulated in the same measurements for the *K-Lα* and *K-Lβ* cascades and the already known angular correlation functions for these two cascades. The final results were fitted by a weighted least-square procedure to the expression $N(\theta) = N_0 + N_2 P_2(\cos\theta)$ (see Fig. 7). The value of N_2/N_0 was found to be

$$N_2/N_0 = (20.99 \pm 1.85) \times 10^{-2}.$$

After effecting the correction due to the finite solid angle of the NaI(Tl) detector the measured angular correlation function of the *K-Ll* cascade can be written as

$$W(\theta) = 1 + (23.32 \pm 2.06) \times 10^{-2} P_2(\cos\theta).$$

V. DISCUSSION

In the present measurements the different *K* x rays were not resolved from one another and neither were all the *L* x rays. Therefore, each of the measured cascades is in fact a mixture of several *K-L* cascades. In order to calculate the theoretically predicted angular correlation functions which are to be compared to the measured ones, the relative contributions of the different *K-L* cascades to the measured mixtures have to be evaluated. According to the atomic decay scheme (see Fig. 1) all of the *L* x rays with the exception of the *Lγ* group can occur in coincidence

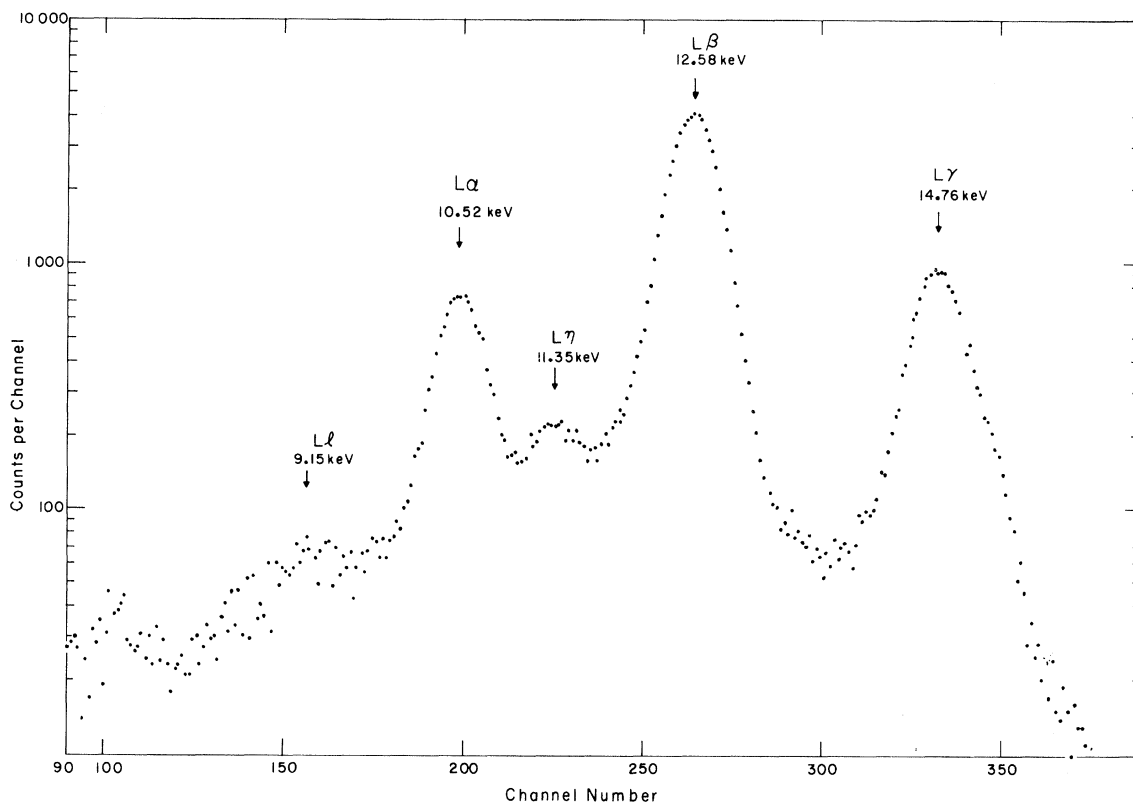


FIG. 7. Spectrum of Pb x rays occurring in coincidence with $K\alpha_2$ x rays. Both K and L x rays were detected with Ge(Li) crystals.

with both $K\alpha_1$ and $K\alpha_2$ x rays; the $L\gamma$ x-ray group can occur in coincidence with $K\alpha_2$ x rays only. An experiment was therefore performed in which the L x-ray spectrum detected with the Ge(Li) crystal was measured in coincidence with $K\alpha_2$ x rays only. In this measurement the K x rays were detected with a second Ge(Li) detector slightly larger (10 mm in diameter) than the one used for the detection of the L x rays. The spectrum of L x rays in coincidence with $K\alpha_2$ x rays is shown in Fig. 7. From this spectrum, the relative intensities of $L\alpha$ and $L\beta$ x rays in coincidence with $K\alpha_2$ x rays relative to the intensity of $L\gamma$ x rays were calculated. These relative intensities – as well as the intensity of the $L\gamma$ x rays in the L x-ray spectra, measured in coincidence with unresolved K x rays detected with the NaI(Tl) crystal – enabled an evaluation to be made of the contribution of L x rays coincident with $K\alpha_2$ x rays to the $L\alpha$ and $L\beta$ peaks in the spectra from which the angular correlation functions were calculated. These contributions were found to amount to $(8 \pm 2)\%$ of the $L\alpha$ peak and to $(75 \pm 5)\%$ of the $L\beta$ peak. The contribution to the $L\gamma$ peak is assumed to be equal to the contribution to the $L\alpha$ peak since both are due to the same Coster-Kronig transition¹³ between the

L_{II} and L_{III} atomic levels.

In the light of these results, the measured angular correlation functions can be compared to the theoretically predicted ones.

A. K - $L\gamma$ Cascade

The data accumulated for this cascade is a mixture of data due to $K\alpha_2$ - $L\gamma$ and $K\alpha_1$ - $L\gamma$ cascades in the ratio of $(8 \pm 2)\%$ to $(92 \pm 2)\%$, respectively. The angular correlation function for the $K\alpha_2$ - $L\gamma$ cascade should be isotropic since the L_{II} atomic level has total atomic angular momentum $\frac{1}{2}$. The theoretically predicted angular correlation function for the $K\alpha_1$ - $L\gamma$ cascade is

$$W(\theta) = 1 + 0.294P_2(\cos\theta).$$

The value of 0.294 for the A_{22} coefficient was arrived at assuming $\delta_1 = -6.25 \times 10^{-2}$ for the $K\alpha_1$ transition and $\delta_2 = +0.795 \times 10^{-2}$ for the $L\gamma$ transition.¹⁴ The signs of δ used here are those of Ref. 2. The angular correlation function predicted for the K - $L\gamma$ cascade as studied here is therefore

$$W(\theta) = 1 + (27.0 \pm 0.6) \times 10^{-2} P_2(\cos\theta).$$

The measured A_{22} coefficient in the angular correlation function of this cascade had the slightly

lower value of $(23.3 \pm 2.1) \times 10^{-2}$. It is interesting to note here that if the M_2 contributions to both the $K\alpha_1$ and the Ll transitions are ignored, the predicted angular correlation function for the $K-Ll$ cascade is

$$W(\theta) = 1 + (23.0 \pm 0.5) \times 10^{-2} P_2(\cos\theta),$$

in very good agreement with the measurements. This agreement is erroneous, however, since the contribution of M_2 radiation to the $K\alpha_1$ transition certainly cannot be ignored as is clear from the results for the $K-L\alpha$ cascade (see following paragraph). An alternative way of reaching better agreement between theory and experiment is to assume that the M_2 contribution to the Ll transition is slightly higher than calculated. An M_2/E_1 mixing ratio of +2%, for the Ll transition instead of the calculated +0.795% would be enough to reach agreement between theory and experiment, within limits of experimental error.

B. $K-L\alpha$ Cascade

The remaining part of the data for the $K-L\alpha$ cascade after the removal of the $(8 \pm 2)\%$ which are due to $K\alpha_2-L\alpha$ coincidences is still a mixture of data due to two different cascades: (i) the $K\alpha_1-L\alpha_1$ cascade due to the consecutive atomic transitions between initial, intermediate, and final atomic states with total atomic angular momenta of $\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$, respectively; for this cascade the theoretically predicted A_{22} coefficient in the angular correlation function is 6.76×10^{-2} . This value of A_{22} was arrived at assuming a value of $\delta = 1.0075 \times 10^{-2}$, for the $L\alpha_1$ transition.¹⁴ (ii) the $K\alpha_1-L\alpha_2$ cascade due to the consecutive atomic transitions between initial, intermediate, and final states with total atomic angular momenta of $\frac{1}{2}$, $\frac{3}{2}$, and $\frac{3}{2}$ respectively; for this cascade the theoretically predicted A_{22} coefficient in the angular correlation function is -24.16×10^{-2} .¹⁵ The relative contributions of these two cascades to the measured $K-L\alpha$ cascade should be in the same ratio as the transition probabilities for radiative deexcitation of the ^{207}Pb atom with a missing electron in the L_{III} shell by $L\alpha_1$ and $L\alpha_2$ x rays, respectively. This ratio, which from simple considerations¹⁶ should have a value of 9, was recently calculated by Scofield¹⁷ to be 8.822. From all these considerations the theoretically predicted angular correlation function for the measured $K-L\alpha$ cascade can be written as

$$W(\theta)(\text{predicted}) = 1 + (3.32 \pm 0.07) \times 10^{-2} P_2(\cos\theta),$$

slightly less anisotropic than the measured angular correlation function of

$$W(\theta)(\text{measured}) = 1 + (4.13 \pm 0.36) \times 10^{-2} P_2(\cos\theta).$$

As in the case of the $K-Ll$ cascade, better agreement with theory would be obtained, assuming for the $L\alpha_1$ transition a slightly higher M_2/E_1 mixing ratio (+2%) than that calculated (1.0075%). In order to check the measured angular correlation function for this cascade, some additional measurements were performed in which the spectrum of L x rays detected with the Ge(Li) crystal was measured in coincidence with $K\alpha_1$ x rays detected with the second Ge(Li) crystal. These measurements were performed only for angles of 90° and 180° and from them a value of $(4.57 \pm 1.00) \times 10^{-2}$ was obtained for the A_{22} coefficient, taking into account the factor 0.92 ± 0.02 for fraction $L\alpha$ here. This is in good agreement with the previously determined value.

It might be worthwhile to remark that in very similar measurements⁹ on the angular correlation function of the same cascade in Tl the value found for the A_{22} coefficient was $(4.0 \pm 1.3) \times 10^{-2}$, in good agreement with the value found in the present measurements in Pb. It should be pointed out that the relatively good agreement between the measured and predicted angular correlation functions for this cascade clearly demonstrates the sensitivity of the x-ray angular correlation function to very slight admixtures of magnetic quadrupole radiation in the predominantly electric dipole x-ray transitions. If these relatively minor contributions would have been neglected, the theoretically predicted A_{22} coefficient in the $K-L\alpha$ angular correlation function measured in this work would have been $(2.25 \pm 0.03) \times 10^{-2}$, in clear disagreement with the measured A_{22} coefficient of $(4.13 \pm 0.36) \times 10^{-2}$.

C. $K-L\beta$ Cascade

The remaining part of the data for this cascade after the removal of the $(75 \pm 5)\%$ which is due to $K\alpha_2-L\beta$ coincidences is a mixture of two cascades, $K\alpha_1-L\beta_2$ and $K\alpha_1-L\beta_{15}$.¹⁸ These two cascades occur between initial, intermediate, and final atomic levels in which the values of the total angular momenta are equal to those for the previously discussed $K\alpha_1-L\alpha_1$ and $K\alpha_1-L\alpha_2$ cascades, respectively. The contributions of the $K\alpha_1-L\beta_2$ and $K\alpha_1-L\beta_{15}$ cascades to the measured $K-L\beta$ cascade are in the same ratio as the transition probabilities for radiative deexcitation of a Pb atom with a missing electron in the L_{III} shell by $L\beta_2$ and $L\beta_{15}$ x rays. This ratio was calculated by Scofield¹⁷ to have the value of 8.935. Taking into account all these factors but neglecting the contribution of magnetic quadrupole radiation to the $K\alpha_1$ and $L\beta_2$ transitions,¹⁹ the expected angular correlation function for the measured $K-L\beta$ cascade would be

$$W(\theta) = 1 + (0.62 \pm 0.13) \times 10^{-2} P_2(\cos\theta),$$

slightly less anisotropic than actually measured. When, however, the M_2 admixtures in the $K\alpha_1$ transition ($\delta = -6.25 \times 10^{-2}$) and the $L\beta_2$ transition for which a value of ($\delta = +1.225 \times 10^{-2}$) was calculated²⁰ are taken into account, the expected angular correlation function becomes

$$W(\theta) = 1 + (0.95 \pm 0.19) \times 10^{-2} P_2(\cos\theta),$$

in good agreement with the experimental results. The A_{22} coefficient in the angular correlation function for this cascade in Tl was measured to be $9(2 \pm 1) \times 10^{-2}$.

VI. CONCLUSION

In the present work, the angular correlation function for the $K-L_1$, $K-L\alpha$, and $K-L\beta$ cascades in Pb were measured. The results, in particular those for the $K-L\alpha$ cascade, demonstrate the necessity of taking into account the small admixtures of magnetic quadrupole radiation to the x-ray transitions. When this is done, relatively good agreement with theory is obtained in all cases. Better agreement would be obtained in all

cases with slightly higher M_2 admixtures in the L x-ray transitions than was theoretically calculated. The experimental results obtained here are similar to previous results⁹ for the same cascades in Tl (see also Ref. 21).

ACKNOWLEDGMENTS

I wish to thank the members of the Arthur A. Noyes Nuclear Chemistry Center at MIT for their hospitality and help during the performance of this work. I wish to thank in particular Professor Charles D. Coryell for his encouragement and useful conversations. Thanks are due also Robert Stewart of the University of Massachusetts, Boston, for his skillful construction of the angular correlation table used in the experiments, Eddie Nellum for his help with parts of the electronic system, as well as the other members of the Department of Physics at the University of Massachusetts, Boston, for useful conversations. I wish to thank also Dr. James H. Scofield of Lawrence Radiation Laboratory, Livermore, California, for making available the results of his calculations on the contributions of magnetic quadrupole radiation to the x-ray transitions.

[†]Present paper is a detailed presentation of a work of which partial results have been published in *Phys. Rev. Letters* **24**, 127 (1970).

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