agreement with the result we obtain using a Ge(Li) detector.

From an average of our three measurements we obtain a mixing ratio for the 161-keV transition of $\delta=0.64\pm0.05$.

Figure 7 shows the theoretical $K/(L+M)$ ratio of the 161-keV transition, A $(\gamma\text{-}e_K)$, and $A_4(\gamma\text{-}e_K)$ as a function of λ, where the mixing ratio used in these calculations is $\delta=0.64$. $A_2(\gamma\text{-}e_K)$ yields two values for λ. These values are $\lambda=41\pm10$ and $\lambda=163\pm5$. $A_4(\gamma\text{-}e_K)$ could, in principle, choose between these values of λ but in this case, because of the large uncertainty in A_4, we get agreement with both values although it favors the larger value, the most probable values of λ obtained from A_4 being 85 and 163.

Experimentally, we find the $K/(L+M+\cdots)$ ratio of the 161-keV transition to be

$$K/(L+M+\cdots)=4.0\pm0.3.$$

The large error quoted for this measurement is due to estimates of the systematic error in the background subtraction, not statistics. Comparing this value to the theoretical $K/(L+M)$ ratios, we find $\lambda=50_{-22}^{+19}$ and $\lambda=205_{-19}^{+20}$. Although this measurement seems to confirm the lower value of λ, it does not give as clear cut a choice as one might wish. In comparing the ratio of K-shell conversion to all higher subshells we might reasonably expect this ratio to be a few percent lower than the $K/(L+M)$ ratio. As can be seen from Fig. 7, a small increase in this ratio, $K/(L+M+\cdots)$, would reduce the lower λ and raise upper value of λ making a choice of $\lambda=41\pm10$ even more reasonable.

A more compelling argument can be made in terms of the K-conversion coefficient. The K-conversion coefficient corresponding to $\lambda=163$ is $\alpha_K=0.095$. Although agreement among the published values of the K-conversion coefficient for this transition is not good, it seens very unlikely that it could be this low.

Thus, we find the best estimate of the mixing ratio of the 161-keV transition to be $\delta=0.64\pm0.05$. The positive mixing ratios obtained from the $K/(L+M)$ ratio and from the $\gamma\text{-}e_K$ directional correlation are $\delta=1.10_{-0.22}^{+0.38}$ and $\delta=1.02_{-0.11}^{+0.35}$, respectively. The discrepancy between the mixing ratios can be resolved by assuming that dynamic nuclear structure effects exist in 161-keV transition and that the value of the nuclear structure parameter (taking into account the uncertainty in the mixing ratio) is 40 ± 15.

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Search for Double K-Shell Ionization in the Decay of Xe^{131m}†

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The deexcitation of the $h_{11/2}$ isomeric 164-keV level in Xe^{131m} has been reinvestigated for the possible simultaneous emission of two K-conversion electrons. The double-decay process was searched for by observing the x-ray coincidence spectrum with Ge(Li)-NaI(Tl) and Ge(Li)-Si(Li) coincidence spectrometers. Although the characteristic x ray of xenon was actually observed in the coincidence spectrum, it was found that this was not primarily due to the simultaneous emission of two K electrons. Our experiment places an upper limit on the possible occurrence of double K-electron emission per single-photon decay: $W_{e_Ke_K}/W_\gamma<1.4\times10^{-3}$. From the observed photon spectrum in coincidence with K x rays, an upper limit of 2×10^{-2} is obtained for the probability of $e_K\text{-}\gamma$ transitions per single-photon decay.

I. INTRODUCTION

Electromagnetic transitions between two nuclear energy states can, in principle, take place via a second-order process in competition with the more probable single-quantum emission. In such a second-order decay the transition energy is shared between two photons, a photon and a conversion electron, or two conversion electrons. In particular, for a transition from a state of energy E_1 and angular momentum $\mathbf{j}_1$ to a state of energy E_2 and angular momentum $\mathbf{j}_2$, two radiations of multipolarities and energies $\mathbf{L}_1$, $\hbar\omega_1$ and $\mathbf{L}_2$, $\hbar\omega_2$ can be emitted with the restriction that $\mathbf{L}_1+\mathbf{L}_2=\mathbf{j}_1-\mathbf{j}_2$ and $\hbar\omega_1+\hbar\omega_2=E_1-E_2$. The three modes for this decay are illustrated by diagrams A, B, and C in Fig. 1. The second-order process is generally several orders of magnitude less probable than the corresponding one-quantum emission, and observation is made even more difficult because of the continuous energy spectrum for the radiation that results from the sharing of the transition energy. However, in transitions where the first-order process is forbidden or severely hindered, the second-order decay may be observable. If such is the case, nuclear-structure information can be obtained from a careful study of the second-order process.

The theory of the double-decay process for atomic transitions was worked out by Goeppert-Mayer,[1] and

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† Work supported in part by the Research Corporation of America and the Alumni Development Fund, University of Oklahoma.

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TABLE I. Summary of available experimental results on nuclear double-quantum emission.

Nucleus	Energy (MeV)	Multi-polarity	Results[a]	Ref.
C^{12}	4.43	$E2$	$E1E1/E2 \leq 1.7 \times 10^{-4}$	a
			$E1E1/E2 \leq 0.5 \times 10^{-4}$	b
O^{16}	6.05	$E0$	$E1E1/E2 \leq 1.1 \times 10^{-4}$	b
	6.13	$E3$	$\gamma\gamma/\gamma \leq 5 \times 10^{-4}$	c
Ca^{40}	3.35	$E0$	$\gamma\gamma/\text{pair} \leq 2.1 \times 10^{-3}$	d
			$\gamma\gamma/\text{pair} \leq 4 \times 10^{-4}$	e
K^{41}	1.29	$M2$	$\gamma\gamma/\gamma \leq 6 \times 10^{-5}$	f
Rb^{85}	0.514	$M2$	$\gamma\gamma/\gamma \leq 1.2 \times 10^{-5}$	f
Zr^{90}	1.76	$E0$	$\gamma\gamma/E0 \leq 6 \times 10^{-4}$	g
			$\gamma\gamma/\text{pair}+e \leq 1.8 \times 10^{-4}$	e
			$\gamma\gamma/\text{pair}+e \leq 8 \times 10^{-5}$	h
Ag^{109}	0.087	$E3$	$\gamma\gamma/\gamma \leq 1.9 \times 10^{-5}$, $e_K e_K/\gamma = (8_{-1.7}^{+0.6}) \times 10^{-3}$, $\gamma e_K/\gamma \leq 1.5 \times 10^{-3}$	i
In^{113}	0.393	$M4$	$e_K e_K/e_K \leq 2 \times 10^{-5}$	j
In^{114}	0.192	$E4$	$\gamma\gamma/\gamma \leq 3 \times 10^{-5}$	k
			$e_K e_K/e_K < 2.2 \times 10^{-4}$, $\gamma e_K/e_K < 4.5 \times 10^{-4}$	l
			$e_K e_K/e_K \leq 1.3 \times 10^{-4}$	m
Xe^{131}	0.164	$M4$	$X_K X_K/e_K+\gamma = 4.5 \times 10^{-4}$	n
			$\gamma\gamma/\gamma \leq 2.2 \times 10^{-5}$, $e_K e_K/\gamma = (3.6 \pm 0.7) \times 10^{-3}$, $e_K\gamma/\gamma < 3 \times 10^{-2}$	o
			$X_K X_K/e_K+\gamma \leq 2 \times 10^{-5}$, $e_K\gamma/\gamma < 2 \times 10^{-2}$	this work
Ba^{137}	0.662	$M4$	$\gamma\gamma/\gamma = (6.4 \pm 3.1) \times 10^{-6}$	p

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[n] Reference 10.

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[q] For clarity, the symbol W has been dropped for transition probabilities (e.g., $W_{\gamma\gamma}$ becomes $\gamma\gamma$). The notation in each case is as used by the authors in presenting their results.

this effect has been observed recently in the emission spectrum of a deuterium-neon plasma.[2] For nuclei, theoretical work has been done by Eichler and Jacob,[3] Eichler,[4] and most recently by Grechukhin.[5,6,7] A rather detailed discussion of the nuclear-structure effects on the second-order transition probability is given in Ref. 7, which includes estimates of these effects calculated on the basis of several nuclear models. A similar discussion for the particular case of In^{114m} is given by Church and Gerholm.[8]

Efforts to detect second-order effects in nuclear

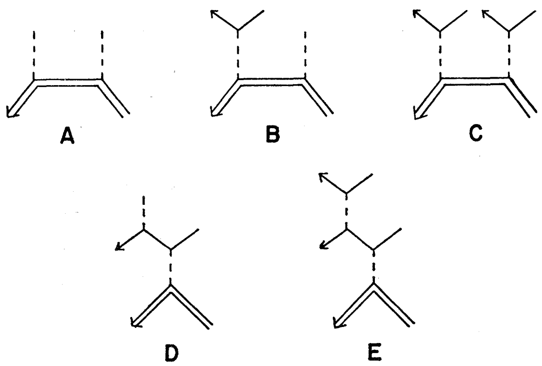

FIG. 1. Possible second-order processes leading to two photons, a photon and electron, or two electrons. (A) Double-photon emission. (B) Conversion-electron–photon emission. (C) Double conversion-electron emission. (D) Internal Compton effect. (E) Conversion of the internal Compton effect.

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transitions have been made repeatedly. A summary of experimental results is given in Table I. Except for the case of the 662-keV $M4$ transition in Ba^{137}, no positive evidence of the observation of two-photon decay [Fig. 1(A)] has been reported. However, observation of the simultaneous emission of two K x rays has been reported for the 87-keV $E3$ transition in Ag^{109m} [9] and the 164-keV $M4$ transition in Xe^{131m}.[10,11]

We have reinvestigated the decay of Xe^{131m}, looking particularly for the simultaneous emission of two K x rays. In the earlier of the two previous measurements on Xe^{131m}, Alväger and Ryde[10] found a value for the ratio of the emission of two K x rays per decay of 4.5×10^{-4}, from which a value relative to the emission of a single photon can be deduced to be $W_{e_Ke_K}/W_\gamma=2.4\times10^{-2}$. After careful consideration of possible sources of interference, they concluded that this represented the observation of a real double-decay process [Fig. 1(C)]. In a more recent experiment, Knauf, Sommer, and Klewe-Nebenius[11] have found a value of $W_{e_Ke_K}/W_\gamma=(3.6\pm0.7)\times10^{-3}$, and they attribute this either to a real double-decay process or to internal conversion of the internal Compton effect [Fig. 1(E)]. Although there is an order-of-magnitude difference between these two results, they both indicate the observation of double K conversion. Knauf *et al.*[11] have also measured an upper limit for double-photon decay relative to single-photon decay of $W_{\gamma\gamma}/W_\gamma\leq2.2\times10^{-5}$. An upper limit for double conversion calculated[11] from this result yields a value of $W_{e_Ke_K}/W_\gamma\leq4.5\times10^{-4}$, which would seem to rule out the possibility of a double-decay process. Listengarten[12] has suggested that the origin of the two coincident x rays is the conversion of the internal Compton effect (ICE). He estimates theoretically that the probability for this effect is

$$(W_{e_Ke_K})_{\mathrm{ICE}}/W_\gamma=\frac{4\alpha}{3\pi}\,\alpha_K(k_0,L)\,k_0\int_{k_1}^{k_2}\frac{dk}{k}\,\tfrac{1}{2}\alpha_K(k,L), \qquad (1)$$

where $\alpha=1/137$, $k_0=E_1-E_2$, $k_1=E_{e_K}$, $k_2=k_0-E_K$, E_K is the electron binding energy, and $\alpha_K(k,L)$ is the K conversion coefficient for a transition of multipolarity L. The energies are in units of the electron rest mass. For Xe^{131m}, Eq. (1) yields a value of 1.6×10^{-2}, several times larger than the measured value given in Ref. 11.

A further motivation for the reinvestigation of the decay of Xe^{131m} stems from the fact that in the two cases where positive evidence for double conversion-electron emission was found (Xe^{131}, Ag^{109}) the measurements involved the observation of coincident x rays, while experiments searching for the same effect by observing the electrons directly (In^{113m}, In^{114m}) produced only upper limits that were much lower than the x-ray results (see Table I). This would indicate that some interference may be present in the x-ray data that does not affect a direct measurement of conversion electrons. Furthermore, the latter results are in disagreement with estimates of the conversion of the ICE. For In^{113m}, Eq. (1) predicts a value three times larger than the reported experimental upper limit.[13] It seems likely that the difficulty here is due to interference from bremsstrahlung radiation produced by conversion electrons from the first-order decay. If an energy-selecting window is placed on the x-ray peak, it will always include part of the continuous bremsstrahlung spectrum, and, if used as a coincidence gate, will always produce counts in the x-ray peak of the coincidence spectrum. It should be noted here, however, that corrections for this effect were considered in both previous experiments on Xe^{131m}.

In the present experiment, the photon spectrum following the decay of Xe^{131m} has been observed in coincidence with xenon K x rays. Advantage has been taken of the superior resolution of semiconductor detectors to carefully study the effects of the continuum spectrum on the x-ray–x-ray coincidence rate. Section II of this paper contains a description of the experimental procedure and a presentation of the results. Section III contains a discussion of these results.

II. EXPERIMENTAL PROCEDURE AND RESULTS

Two experimental configurations were used in the measurements. In both cases a Ge(Li) detector was used to obtain a high-resolution photon spectrum following the decay of Xe^{131m} in coincidence with K x rays of xenon. In the first experiment to be discussed, a NaI(Tl) crystal served as the detector in the gate channel. It was replaced by a Si(Li) detector for the second experiment in order to take advantage of the superior energy resolution of this detector to reduce the fraction of the continuum photon spectrum included in the energy-selecting window.

Accidental coincidences were subtracted for equal time intervals from the data during the course of each experiment. Errors quoted are based on counting statistics and uncertainties in source intensity, detector-efficiency measurements, and background extrapolations.

A. Source Preparation

Radioactive Xe^{131} was obtained as a daughter product from a solution of 50-mCi I^{131}. The iodine solution was kept frozen in an evacuated container maintained at dry-ice temperature for 14 days. The xenon gas was then collected by condensing it in an ampule kept at liquid-nitrogen temperature. The sample was transferred to the isotope separator at Argonne National Laboratory, where Xe^{131} was collected on a 3.5-mg/cm^2 aluminum backing. The source diameter was approximately 3 mm. Two sources were made with initial

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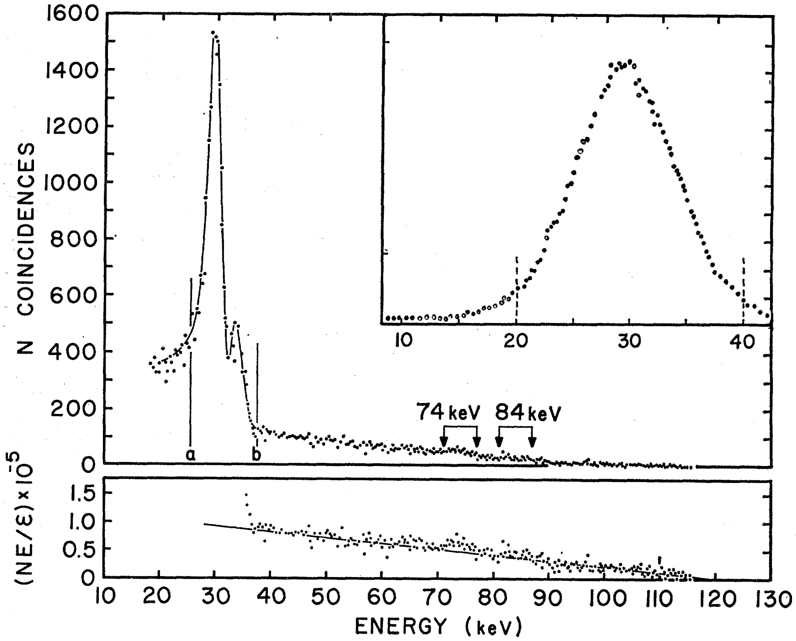

FIG. 2. X-ray coincidence spectrum for Ge(Li) detector gated with a NaI(Tl) detector. The insert to the right contains the NaI(Tl) gate; the lower curve contains the continuous spectrum divided point by point by the Ge(Li) efficiency and multiplied by the energy.

strengths of 10^5 and 10^4 dis/sec, respectively. The I^{131} contamination was measured to be $<0.02\%$ of the Xe^{131} activity in each case.

B. Ge(Li)-NaI(Tl) Coincidence Measurement

The source was mounted between a 4-cm^3 Ge(Li) detector and a 5.1-cm-diam, 0.8-mm-thick NaI(Tl) scintillation detector. A copper-lined lead shield with a hole slightly larger than the source diameter was placed between the two detectors in order to reduce the backscatter from one detector to the other. A Teflon absorber of 56 mg/cm^2 was placed between the source and the scintillation detector to stop the conversion electrons which could penetrate the 0.12-mm beryllium entrance window of the NaI(Tl) crystal. The germanium crystal was encased in a 137 mg/cm^2 aluminum can; no additional absorber was placed between it and the source.

Spectra from the Ge(Li) detector were recorded with a multichannel analyzer gated by the output of a standard crossover-timing coincidence circuit. The coincidence circuit had a time resolution adjustable between 50 and 200 nsec, and the resolving time was always set large enough to ensure 100% coincidence efficiency. The gate signal was obtained by the requirement that a suitable portion of the spectrum from the NaI(Tl) detector selected by a single-channel analyzer be in coincidence with the Ge(Li) spectrum.

The coincidence spectrum, observed with a 20-keV window in the NaI(Tl) channel that accepted 95% of the x-ray peak (20–40 keV) is shown in Fig. 2. The x-ray peak from the gate channel is shown in the insert of Fig. 2. The x-ray peak is expected to contain events both from the simultaneous emission of two x rays and from those events in which an x ray entered the Ge(Li) detector while a continuum photon with an energy between 20 and 40 keV is simultaneously detected by the NaI(Tl) detector. This continuum photon may either originate directly from a nuclear transition or be external bremsstrahlung. The coincidence rate between continuum photons and x rays is observed to be substantial. The spectrum of the photon continuum which results when the coincidence rate above 40 keV is multiplied at each energy by the energy E and corrected for detector efficiency ϵ is a straight line as is shown in the lower portion of Fig. 2. This result is in good agreement with the observation of Ref. 11 and is expected from a thick-target bremsstrahlung spectrum.

The coincidence rate in the x-ray peak of the Ge(Li) spectrum (interval *ab* in Fig. 2) has been evaluated. After subtracting the continuum background obtained by extrapolating the photon spectrum through the interval *ab*, there remains a coincidence rate, denoted by $n_{cx}=n_{xx}+n_{xp}$, which is the sum of the x-ray–x-ray coincidence rate and the x-ray–continuum-photon coincidence rate. These may be expressed as

$$n_{xx}=2N_0\epsilon_1(x)\epsilon_2(x)f_xW_{xx},$$

$$n_{xp}=N_0\epsilon_1(x)\epsilon_2(x)f_pW_{xp},$$

where N_0 is the source strength, $\epsilon_1(x)$ and $\epsilon_2(x)$ are the respective average efficiencies of the Ge(Li) and NaI(Tl) detectors for the x-ray region, f_x and f_p are the respective fractions of the x-ray peak and continuum spectrum accepted in the window of the single-channel analyzer, and W_{xx} and W_{xp} are the respective probabilities per disintegration for emission of two simultaneous x rays and the emission of an x-ray–

continuum-photon pair. W_{xx} and W_{xp} include all mechanisms for producing these effects; ϵ_1 and ϵ_2 include geometric as well as intrinsic efficiencies.

In order to study the effect of the continuum background on the coincidence rate in the interval *ab*, this rate was observed as a function of window width in the gate channel and compared with the singles rate in the gate channel. The results of this comparison, normalized for a window width of 30 keV, are plotted against window width in Fig. 3. All energy intervals are centered at 29.5 keV. The straight line represents the ratio f_p expected from the observed continuum. If the coincidences in the interval *ab* were all true x-ray–x-ray events they should follow the pattern of the gate singles as the window width is varied. As seen in Fig. 2, this is clearly not the case. In fact, the measured coincidence rate follows the ratio f_p quite closely, which indicates the observed coincidences are predominately due to x-ray–continuum events. It is also clear from Fig. 3 that the ratio of accepted x ray to the continuum background is optimal for window widths from 10 to 20 keV. Measurements have therefore been made at window widths of 20, 14, 10, and 5 keV.

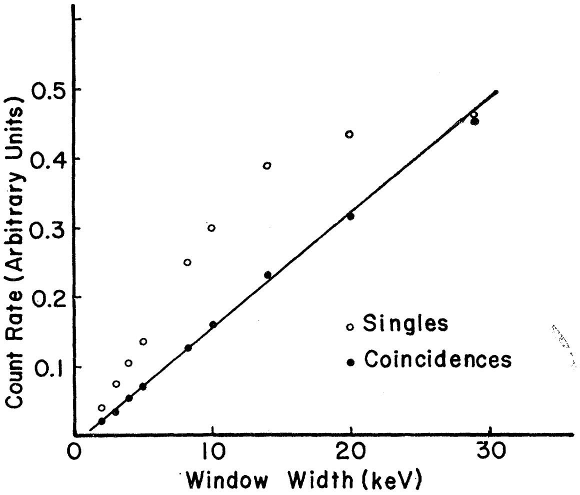

FIG. 3. Comparison of singles rates in the gate channel with the coincidence rate in the interval *ab* (Fig. 2) as a function of window width. The straight line is a plot of the fraction of the continuum spectrum, f_p, accepted in the single-channel-analyzer window.

The probability for double x-ray emission (W_{xx}) can be obtained from the measured coincidence rate (n_{cx}) in the interval *ab* by subtracting the contribution from x-ray–continuum events. This contribution was determined experimentally by first placing the window above the x-ray peak and observing the x-ray–continuum coincidence rate with no x rays in the window. This rate is given by

$$n_{xp}'=N_0\epsilon_1(x)\epsilon_2'(p)f_p'W_{xp},$$

where $\epsilon_2'(p)$ is the efficiency for continuum photons in the new energy interval and f_p' is the fraction of the total continuum spectrum included in the new window. From the result of this measurement, we have

$$n_{xp}=[\epsilon_2(x)f_p/\epsilon_2'(p)f_p']n_{xp}',$$

where ϵ_2 and ϵ_2' are measured quantities determined from single rates. The ratio f_p/f_p' of the fractions of the photon spectrum accepted in the window is obtained from the observed coincidence spectrum corrected for detector efficiency. The region underlying the x-ray peak was obtained by extrapolation (see the lower portion of Fig. 2). The probability for double x-ray emission is then given by

$$W_{xx}=[n_{cx}/2N_0\epsilon_1(x)\epsilon_2(x)f_x]-[n_{xp}/2N_0\epsilon_1(x)\epsilon_2(x)f_x]$$
$$=R_1-R_2,$$

where R_1 and R_2 represent the respective bracketed ratios. The results of this calculation for different window widths are as follows:

Window (keV)	R_1	R_2	W_{xx}
20	$(54.0\pm0.8)\times10^{-5}$	$(52.3\pm2.0)\times10^{-5}$	$(1.7\pm2.2)\times10^{-5}$
14	$(41.8\pm0.8)\times10^{-5}$	$(41.5\pm1.7)\times10^{-5}$	$(0.3\pm1.9)\times10^{-5}$
10	$(38.4\pm0.8)\times10^{-5}$	$(36.8\pm1.5)\times10^{-5}$	$(1.6\pm1.7)\times10^{-5}$
5	$(37.5\pm1.0)\times10^{-5}$	$(39.4\pm1.6)\times10^{-5}$	$(-1.9\pm1.9)\times10^{-5}$

C. Ge(Li)-Si(Li) Coincidence Measurement

The experiment described above clearly implies that the coincidences in the x-ray peak of the Ge(Li) coincidence spectrum are predominately due to x-ray–continuum events. As a consequence of this, a second measurement was undertaken in which the contribution to the photon continuum by external bremsstrahlung was reduced, with an accompanying improvement in the peak-to-continuum ratio f_x/f_p. To achieve this improvement, an experimental arrangement similar to that described above was used, except that the source was placed between two beryllium plates to stop the electrons in low-*Z* material and the NaI(Tl) detector in the gate channel was replaced with a Si(Li) detector. This arrangement results in a 30% reduction of the continuum photon spectrum as measured in the energy range 47–130 keV, and the better resolution of the

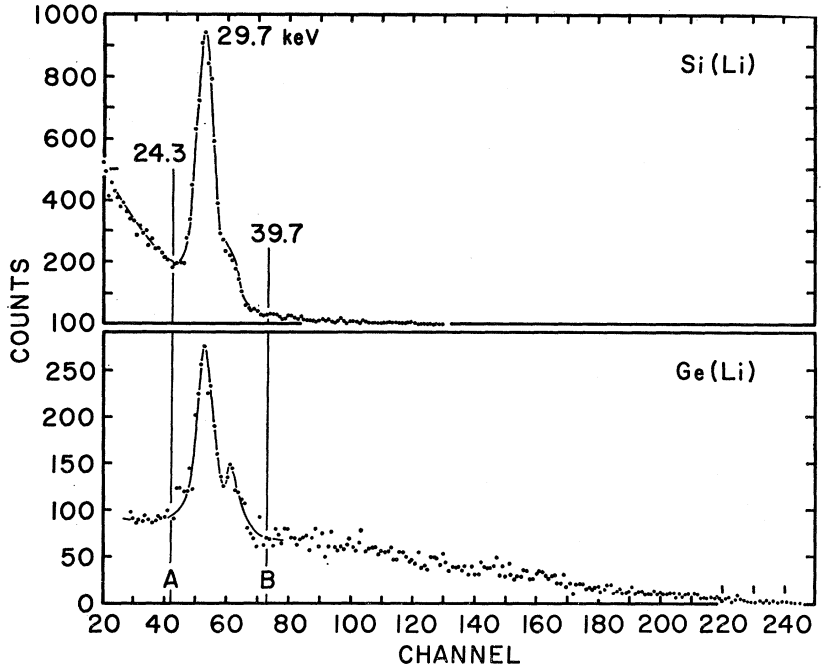

FIG. 4. The upper curve is the coincidence spectrum from the Ge(Li) detector; the lower curve depicts the x-ray gate from the Si(Li) detector.

Si(Li) detector makes it possible to cover completely the x-ray peak with a 16-keV window. The coincidence spectrum from the Ge(Li) detector along with the x-ray gate from the Si(Li) detector is shown in Fig. 4.

As before, the count rate in the x-ray peak of the Ge(Li) spectrum after subtraction of the continuum background is denoted by $n_{cx}=n_{xx}+n_{xp}$, where

$$n_{xx}=2N_0\epsilon_1(x)\epsilon_3(x)f_xW_{xx},$$

$$n_{xp}=N_0\epsilon_1(x)\epsilon_3(x)f_pW_{xp},$$

and where $\epsilon_3(x)$ is the efficiency in the energy interval AB (see Fig. 4) of the Si(Li) detector. To determine W_{xx} it is again necessary to measure n_{xp}. A somewhat different approach was used to accomplish this measurement than in the previous experiment, which takes advantage of the superior resolution of the Si(Li) detector. In this case the two channels were reversed, and a coincidence spectrum from the Si(Li) detector was recorded. Because of its very low efficiency, the Ge(Li) detector was replaced in this measurement by a NaI(Tl) detector. Once the coincidence spectrum is obtained, the continuum spectrum may be extrapolated beneath the x-ray peak. If the contribution of the continuum to the count rate in the interval AB of the Si(Li) spectrum is denoted by n_{xp}', then

$$n_{xp}'=N_0\epsilon_2(x)\epsilon_3(x)f_pW_{xp}.$$

From this we obtain

$$n_{xp}=n_{xp}'[\epsilon_1(x)/\epsilon_2(x)].$$

It is assumed in writing this relation that all of the x-ray peak was included in the NaI(Tl) gate, and care was taken to ensure this was the case. We thus obtain for the probability per disintegration of double x-ray emission,

$$\begin{aligned} W_{xx}&=[n_{cx}/2N_0\epsilon_1(x)\epsilon_3(x)]-[n_{xp}'/2N_0\epsilon_2(x)\epsilon_3(x)]\\ &=(15.0\pm1.5)\times10^{-5}-(16.8\pm2.0)\times10^{-5}\\ &=(-1.8\pm2.5)\times10^{-5}. \end{aligned}$$

III. DISCUSSION OF RESULTS

From the results of the above experiments an upper limit may be placed on the occurrence of two coincident K x rays per disintegration of Xe^{131m}. The weighted average of the measurements is $W_{xx}=(0.14\pm1.8)\times10^{-5}$ and from this $W_{xx}\leq2\times10^{-5}$. For comparison with previous work, an upper limit for the emission of two K electrons relative to the emission of a single γ ray may be calculated from the observed double x-ray emission probability W_{xx}:

$$W_{e_Ke_K}/W_\gamma=(1+\alpha)\omega_K^{-2}W_{xx}\leq1.4\times10^{-3}.$$

Here the values $\omega_K=0.87$ [14] and $\alpha=53.3$ for the fluorescent yield and total conversion coefficient have been used, where α is calculated from $\alpha_K=32.1\pm0.4$ as measured in Ref. 11, $K/L=1.83$, and $K/M=8.8$.[15] A value of $\alpha_K=31.7\pm0.9$ was obtained from data in the present experiment, in good agreement with Ref. 11.

The upper limit for $W_{e_Ke_K}/W_\gamma$ obtained in this work

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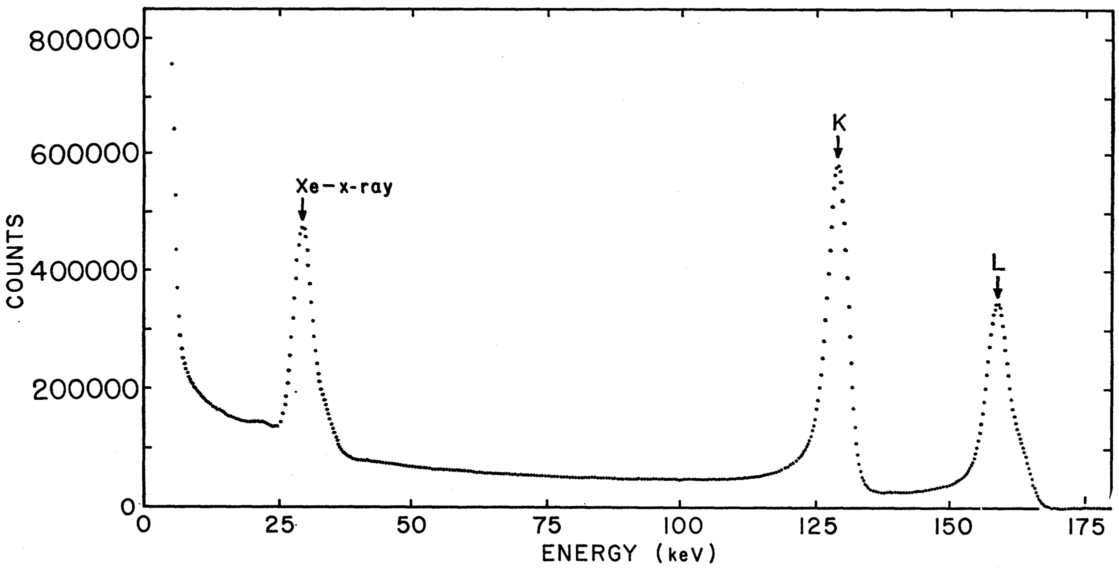

FIG. 5. Singles electron spectrum taken with the Si(Li) detector.

is consistent with the expectation $W_{e_Ke_K}/W_\gamma \leq 4.5\times10^{-4}$ calculated from the upper limit for double γ-ray emission reported in Ref. 11. This result also indicates that the theoretical estimate for the contribution from conversion of the internal Compton effect [Eq. (1)] is too large by at least a factor of 10.

An upper limit may also be obtained for a two-step nuclear transition in which one of the radiations is a conversion electron [Figs. 1(B) and 1(D)]. If in Fig. 4 all the observed coincidence counts in the energy interval 47–130 keV are assumed due to this process, the limit is

$$W_{e_K\gamma}/W_\gamma \leq 2\times10^{-2}.$$

Here use was made of the theoretical expressions given in Eq. (28) of Ref. 4 (with $\tau=\tau'=0$) to estimate the total continuum spectrum by extrapolation from this energy interval. This limit is 30% smaller than that obtained in Ref. 11 (3×10^{-2}) and probably reflects the reduction of the external bremsstrahlung interference achieved by encasing the source in beryllium. It seems apparent from this result that at least a large fraction of the events in the continuum coincidence spectrum must be attributed to interference from external bremsstrahlung.

No evidence for feeding of the 80-keV, $\frac{1}{2}^+$ first excited state is found in either the γ-ray or conversion-electron spectra. A sample electron spectrum taken with a Si(Li) detector is shown in Fig. 5. There is clearly no evidence for additional conversion lines in this spectrum, and an upper limit for the intensity of a transition to the 80-keV level relative to the K line of the isomeric transition is 2×10^{-4}. The Ge(Li) coincidence spectrum in Fig. 2 shows a slight indication of a line at 74 keV (see especially the spectrum in the lower portion of this figure). This has been identified as the lead fluorescent x ray from the antiscatter shield between the detectors. Assuming the 80.16-keV transition has multipolarity $M1$, we find the probability for a transition to the 80.16-keV level from the isomeric state to be $<2\times10^{-5}$ per decay.

Note added in proof. R. Vukanović, L. Samuelsson, M. Migahed, L. Westerberg, and L. O. Edvardson [Phys. Letters **29B**, 576 (1969)] have reported the observation of double internal conversion in the decay of In^{114}: $W_{ek}W_{ek}/W_{ek} = (1.7\pm0.3)\times10^{-5}$, consistent with the upper limits of previous experiments listed in Table I.

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