β Spectrum of I^{129} and Its Decay Scheme*

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The decay of I^{129} has been studied with several instruments including a magnetic lens spectrometer, a scintillation spectrometer utilizing a technique in which the activity is introduced into a NaI(Tl) crystal, and a proportional counter. A soft beta radiation whose end-point value is measured to be 150±5 kev and a gamma radiation of 38 kev are observed. The shape of the beta spectrum is studied and the K-conversion coefficient and the K/L conversion ratio for the 38-kev gamma radiation are determined. The beta and gamma radiations are shown to be in coincidence and an upper limit is set for a possible beta branch to the ground state of Xe^{129}. Spin assignments are made which agree with shell model theories of the nucleus and with data available on the decay of the 8-day isomer of Xe^{129}.

INTRODUCTION

RADIOACTIVE I^{129} is a long-lived fission product which emits soft β^- and γ radiations. Lifetime measurements have been made by several experimenters[1] who reported values of 1.72 and 3×10^7 yr, which they obtained by determining the disintegration rates of weighed amounts of I^{129}. Absorption[1] and proportional counter[2] techniques have been used to measure the maximum energy of the β spectrum, and values close to 0.12 Mev have been given. The soft γ radiation has been reported to be 0.039 Mev.[2]

The β decay of I^{129} leads to Xe^{129}. Information about the first two excited states of this isotope has been obtained through study of the decay of the 8-day isomer of Xe^{129}. At first a 196-kev gamma radiation was observed associated with this isomer which was identified as a magnetic 4-pole transition. In order to build a decay scheme consistent with the measured ground-state spin of $\frac{1}{2}$ and shell model theories, Goldhaber and Sunyar[3] postulated a low-lying intermediate state. Following shell model theories, the second excited state was assumed to be an $h_{11/2}$ level and the first excited state a $d_{3/2}$ level. Thus, the 196-kev $M4$ transition could be the $h_{11/2}\rightarrow d_{3/2}$ transition and a $d_{3/2}\rightarrow s_{1/2}$ transition would follow to the ground state. Recently, a second transition of 40 kev in the decay of ${}_{54}Xe^{129m}$ was confirmed by observing its L-conversion line in a magnetic spectrometer.[4] Presumably, the 40-kev line observed in ${}_{54}Xe^{129m}$ and the 39-kev line in I^{129} are the same $d_{3/2}\rightarrow s_{1/2}$ transition in ${}_{54}Xe^{129}$.

The spin of the ground state of I^{129} has been measured and given to be 7/2 (the shell model predicts $g_{7/2}$). If the β transition observed in I^{129} leads to the first excited state in Xe^{129}, the spin and parity change involved in the β transition ($g_{7/2}\rightarrow d_{3/2}$) is $\Delta I=2$, (no), which belongs to the second forbidden parity unfavored group. A beta branch from the $g_{7/2}$ ground state of I^{129} to the $s_{1/2}$ ground state of Xe^{129} ($\Delta I=3$, no) is not completely ruled out on the basis of theoretical lifetime estimations. Considerations based upon the known ft value of Be^{10} would indicate a branching ratio of 10^{-3} for the ground-state transition. It appeared to us, therefore, that a determination of the β branching ratio in I^{129} would be of interest, and, further, that a theoretical interpretation of the forbidden shape of the beta spectrum would add to the knowledge of the second forbidden parity unfavored group, of which, to date, ony four other cases[5] (Cl^{36}, Tc^{99}, Cs^{137}, Cs^{135}) are known to exist and have been investigated. Photon studies were made to complete our knowledge concerning the decay scheme of I^{129}.

CHARACTERISTIC X-RAYS AND GAMMA RADIATION

A preliminary investigation of the soft photon radiations of I^{129} with a NaI(Tl) scintillation spectrometer revealed the presence of photons with 30-kev energy. By the use of Cd, In, Sn, and Sb critical absorbers, it was established that these photons were

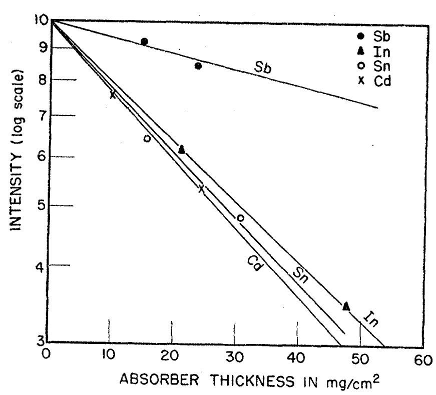

FIG. 1. Critical absorption of x-rays and γ rays of I^{129} in Sb, Sn, In, and Cd. The break between Sb and the rest indicates the presence of Xe x-rays.

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[1] Katcoff, Schaeffer, and Hastings, Phys. Rev. 82, 688 (1951); Parker, Creek, Herbert, Lantz, and Martin, Oak Ridge National Laboratory Report ORNL-286, September, 1949 (unpublished).

[2] C. J. Borkowski and A. R. Brosi, Oak Ridge National Laboratory Report ORNL-607, 1950 (unpublished).

[3] M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).

[4] S. Thulin and I. Bergstrom, Phys. Rev. 85, 1055 (1952).

[5] C. S. Wu, Physica 18, 989 (1952).

Xe K x-rays. The experimentally obtained characteristic absorption curves are shown in Fig. 1.

The soft radiations were then carefully examined with a krypton-filled proportional counter. A pronounced Xe K_α line (with a slight bump due to the K_β line) was revealed, but, in addition, the existence of a weak line at 38 kev was shown (Fig. 2). This agrees with the findings of Borkowski and Brosi.[2]

If it is assumed that the observed Xe K x-rays are due to the internal conversion of the 38-kev gamma radiation, the K-conversion coefficient for this gamma ray can be estimated by comparing the sum of the areas under the K_α, K_β and escape peaks with that of the 38-kev line after corrections for window absorption, counter efficiency, and fluorescence yield have been applied. The K-conversion coefficient thus obtained is 22 ± 4.

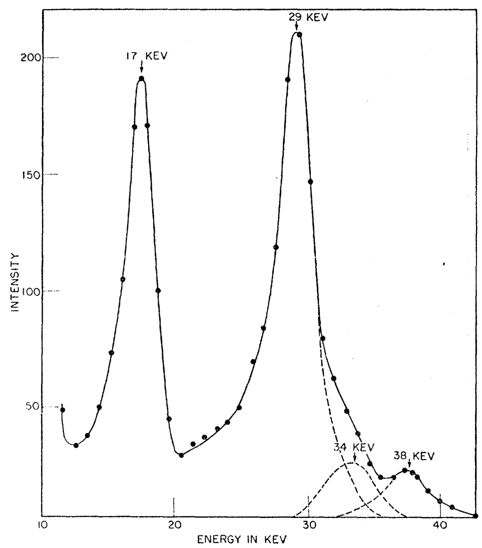

FIG. 2. Proportional counter observation of the radiations of I^{129}. The 29-kev and 17-kev peaks are due to the xenon K_α line and an escape peak, while the broadening of the 29-kev peak at 34 kev may be due to the xenon K_β line. A definite indication of a 38-kev gamma ray is shown.

Table I lists theoretical K-conversion coefficients for various multipoles, computed with the help of the calculations of Spinrad and Keller,[6] for radiations with energies approaching the K-shell binding energies. The use of these calculations in the case of I^{129} is justified by the nearness of the K-shell binding energy Xe (34.6 kev) to the energy of the 38-kev radiation.

The observed value of 22 ± 4 for α_K is in agreement with either $M1$ or $E2$ radiation. Evidence based on the K- to L-conversion ratio of the 38-kev radiation which will be presented below indicates $M1$ to be the preferred assignment. This is in agreement with a $d_{3/2}\rightarrow s_{1/2}$ transition.

[6] B. I. Spinrad and L. B. Keller, Phys. Rev. 84, 1056 (1951).

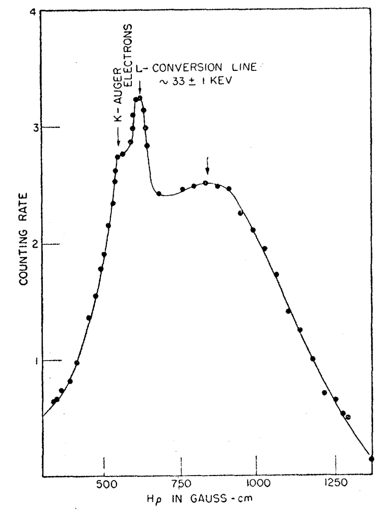

FIG. 3. The momentum distribution of the I^{129} β spectrum.

THE L-CONVERSION LINE

The β spectrum and electron conversion lines were studied with the Columbia University magnetic lens spectrometer. The investigation of a low-energy β spectrum of a highly forbidden transition is a difficult task. Because of its extremely long half-life ($t_{1/2}=3\times10^7$ yr) the specific activity of the I^{129} sample was very low. The source was prepared by depositing 200 μg/cm^2 of KI onto a Formvar film whose thickness was 10 μg/cm^2. Even though the purity of the I^{129} was 85 percent, the source was equivalent to only $\sim$0.05 μC/cm^2, so that an extended source 1 cm in radius was necessarily used. The resolution was on this account broadened to 6–8 percent. The counter used had a Formvar window of $\sim$25 μg/cm^2.

Figure 3 shows the momentum distribution of the spectrum. The line appearing at $H\rho=620$ is the L-conversion line of the 38-kev gamma radiation. This same L-conversion line was also observed by Thulin and Bergstrom[4] in the Xe^{129m} e^- spectrum. The energy of the gamma ray calculated from the conversion line is $E_\gamma=E_{L^-}+W_L=33+4.6=37.6\pm1$ kev. The little peak on the left side of the L-conversion line is due to K-Auger electrons.

A comparison of the area under the L-conversion line with the area under the beta spectrum enables us to determine α_L, the L-conversion coefficient. This combined with the above measured value of α_K gives us a value for the K/L conversion ratio, which is about 10:1. Whereas $\alpha_K=22\pm4$ is in agreement with either

TABLE I. Theoretical K-conversion coefficients.

Electric Multipole	α_K	Magnetic Multipole	α_K
$E1$	8	$M1$	19
$E2$	23	$M2$	300
$E3$	40		

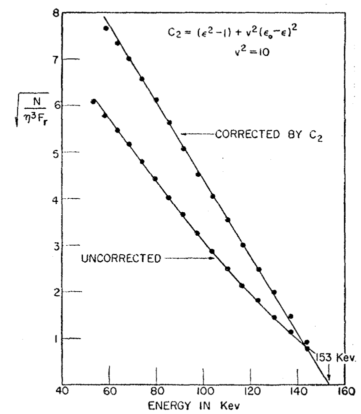

FIG. 4. The Kurie plot of the I^{129} β spectrum. N is the counting rate per momentum interval. The value 10 for v^2 in the correction factor C_2 agrees with the value of $v^2=9.5$, which fits the Cs^{135} beta spectrum which also results from a $g_{7/2}\rightarrow d_{3/2}$ transition.

$M1$ or $E2$, the K/L ratio indicates the 38-kev gamma ray to be an $M1$ transition (for $E2$ the K/L ratio is[7] around $\sim$0.2). Therefore, the preferred assignment for the 38-kev transition is $d_{3/2}-s_{1/2}$.

The Beta Spectrum

The continuous beta spectrum yields an end point higher than those previously reported. It is 150±5 kev. The Kurie plot is not linear but shows a slight curvature convex toward the energy axis (Fig. 4). The distortion due to the source thickness and broad resolution used was checked by using a S^{35} source prepared by mixing the S^{35} with KI and covering a circular area of 1-cm radius with 200 μg/cm^2 of $S^{35}-$KI mix. It was found that distortion above 50 kev was negligible (Fig. 5).

It is important to ascertain experimentally whether a beta branch to the ground state of Xe^{129} is negligibly small. A verification of this would be to show that each beta particle emitted in the decay of I^{129} is followed by a 38-kev gamma radiation. The technique[8] developed at the Brookhaven National Laboratory of growing NaI(Tl) crystals with a trace of the radioactive substance mixed in it enables one to answer this question. Because of the nearly 100 percent efficiency in detecting the 38-kev line in the NaI(Tl) crystal, the pulse height detected should be proportional to the sum of the beta-particle energy and the gamma energy, because each beta particle emitted to the excited state of Xe^{129} is followed by a 38-kev emission. The minimum pulse height in the beta distribution, due to those β particles emitted with practically zero energy, should be equivalent to 38 kev. This implies that the pulse distribution should give practically zero intensity at the low-energy region and then suddenly increase to a high counting rate at 38 kev. Figure 6 shows this remarkable discontinuity as anticipated. From the counting rate below the 38-kev region, one could put an upper limit to the β branch to the ground state of Xe^{129} of less than 1 percent. This is in agreement with the theoretical estimation based on the available ft value of Be^{10} which is also a $\Delta I=3$, (no) transition. The ft value of Be^{10} after being corrected for its average value of $\bar{D}_2$ is around 10^{12} sec. Now the average value[9] of $\bar{D}_2$ for the ground-to-ground transition in I^{129} is 1.5×10^{-4}. If one assumes that the corrected ft value of all transitions with $\Delta I=3$, (no) should have about the same order of magnitude, then the uncorrected f_0t for this transition will be $\sim10^{16}$ sec. It is a factor of 10^3 larger than the observed $f_0t\sim10^{13}$ sec. This implies that the branching ratio must be of the order of $10^{-2}-10^{-3}$.

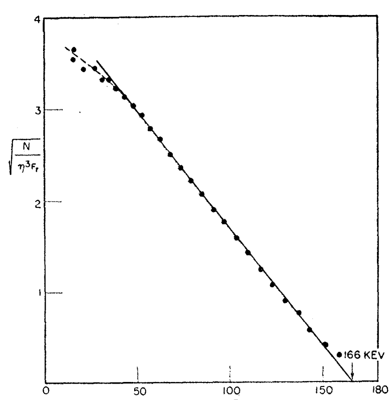

FIG. 5. The Kurie plot of the S^{35} β spectrum. This spectrum was used to check distortion due to source thickness and broad resolution. A source was prepared by mixing S^{35} with enough KI ($\sim$100 μg/cm^2) to simulate conditions existing for the I^{129} source. The spectrum shows negligible distortion above 50 kev.

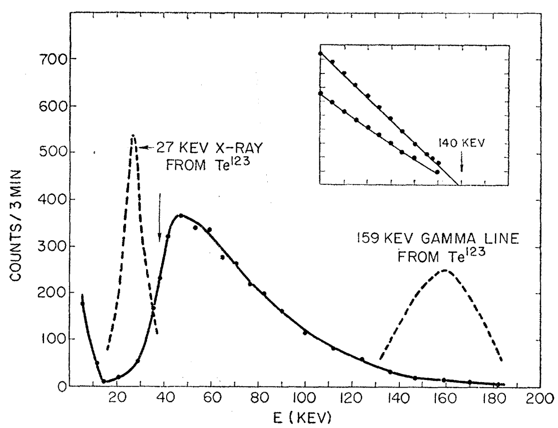

FIG. 6. Energy distribution of the I^{129} β spectrum as seen with an internal source scintillation spectrometer. The insert shows Kurie plots of the spectrum, the upper plot of the two being corrected by $C_2=(\epsilon^2-1)+v^2(\epsilon_0-\epsilon)^2$, where $v^2=10$.

[7] M. Goldhaber, Physica 18, 1091 (1952).

[8] Scharff-Goldhaber, der Mateosian, Goldhaber, Johnson, and McKeown, Phys. Rev. 83, 480 (1951); E. der Mateosian and A. Smith, Phys. Rev. 88, 1186 (1952).

[9] J. Davidson, Phys. Rev. 82, 48 (1951).

INTERPRETATION OF THE β SPECTRUM

Recent analysis of the superallowed transitions have shown that the contributions from the Gamow-Teller (G-T) and Fermi interactions in the β-decay process are comparable. This means that the beta-decay interaction is not a single interaction but rather a linear combination of approximately equal parts of G-T and Fermi interactions.

In the past, one has been able to fit the parity unfavored second forbidden β spectra by tensor or polar vector interaction alone. The reason behind this possibility is the very close energy dependences between the correction factors associated with the various matrix elements. For instance, if the transition energy$\ll$ Coulomb barrier energy, then the correction factors for the nuclear matrix elements of R_{ij}, $R_{ij}T_{ij}$ are related by $C_{Rij}\approx -2C_{Rij^*Tij}=4C_{Tij}$. Likewise, the correction factors of $R_{ij}{}^*A_{ij}$ and $T_{ij}{}^*A_{ij}$ are very close in shape,

$$C_{Rij^*Aij}\approx -2C_{Tij^*Aij}.$$

Therefore, the correction factor for the (S,T) combination can be written in terms of the tensor or polar vector interaction alone. It explains why it has been found possible to fit the observed $\Delta I=2$ (no) spectra with one of these interactions alone.

The algebraic form of the correction factor for second forbidden interactions with the following assumptions:

(i) transition energy $\ll$Coulomb barrier energy,
(ii) G-T interaction $\approx$Fermi interaction,
(iii) $S_{ijk}\equiv 0$,

can be expressed as

$$C_2=p^2+v^2q^2=(\epsilon^2-1)+v^2(\epsilon_0-\epsilon)^2,$$

where

$$v^2=\left[\frac{2(\alpha Z/2\rho)(R_{ij}+\frac{1}{2}iT_{ij})-A_{ij}}{(\alpha Z/2\rho)(R_{ij}+\frac{1}{2}iT_{ij})-A_{ij}}\right]^2=\left(\frac{2\eta-a}{\eta-a}\right)^2.$$

One outstanding relationship between v^2 and a/η is that in the neighborhood of $a/\eta=1$ the value of v^2 becomes very large. The value of v^2 used in C_2 to fit the I^{129} β spectrum is 10 (Figs. 4 and 6). This is in good agreement with the value of $v^2=9.5$ in C_2 for the Cs^{135} β spectrum which also results from a $g_{7/2}\rightarrow d_{3/2}$ transition.

CHEMICAL PREPARATION

The radioactive I^{129} used in this investigation was made available to us through the kind arrangement of Dr. Parker of Oak Ridge National Laboratory. The I^{129} was a dilute solution of $NaOH+Na_2SO_3$. The total amount of iodine was around 2 mg. The isotopic concentration is approximately 85 percent of I^{129} and 15 percent of I^{127}.

To prepare a NaI source, the iodine was first extracted with CCl_4. To the CCl_4 layer, 10 cc of distilled water containing about 5 mg Zn dust was added. The mixture was shaken and the aqueous layer was separated and filtered through a medium filter stick. The solution was passed through a cation exchange resin (Dowex–50, Na-form) to convert it to NaI. However, NaI is not suitable for use in magnetic spectroscopic investigation as the NaI source will quickly become deliquescent as soon as it is exposed to moist air. For this purpose a portion of the I^{129} was converted to KI by ion exchange and a source of approximately 200 $\mu g/cm^2$ was evaporated onto a thin Formvar film.

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