Metastable States of Ge^{73}†*

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The decay of As^{73} to Ge^{73} has been investigated with scintillation counters. The K capture is followed by a 53.9-kev gamma ray which in turn is followed by a 13.5-kev transition to the ground state. The total conversion coefficient of the 53.9-kev gamma is 4.7 and the lifetime is between 900 microseconds and 10 seconds. The half-life of the 13.5-kev transition is 4.6 microseconds. The available evidence points to the 53.9-kev transition being $M2$ and the 13.5-kev transition $E2$ with a possible small admixture of $M1$. Spin and parity assignments are discussed.

AS part of a general search for nuclear isomers with lifetimes in the millisecond range, it seemed pertinent to examine further the disintegration scheme of As^{73}. Johansson,[1] on evidence obtained from measurements with a β-ray lens spectrometer, suggested the decay scheme shown in Fig. 1. It was of particular interest to investigate the lifetime of the 53.9-kev transition, as half-lives in the millisecond range are not expected to be very common.[2] A knowledge of the lifetimes is also helpful in verifying the spin and parity changes involved. In addition, it was desirable to confirm the existence of the 13.5- and 53.9-kev gamma rays by means of scintillation spectrometer techniques. The order of emission of the gamma rays was also investigated and found to be the reverse of that proposed by Johansson on the basis of shell theory.

SOURCE PREPARATION

As^{73} was obtained from Oak Ridge as carrier-free arsenite in 2.8M HCl. Metallic arsenic was precipitated from a hot concentrated HCl solution to which had been added 0.2-mg arsenic carrier (as AsO_2^-) and a few tenths of a gram of sodium hypophosphite. In order to make sources, portions of the precipitate were transferred to 0.001-inch Al foils, dried, and covered with a film of 1 percent zapon. In addition, a NaI crystal was grown from a melt containing some of the metallic arsenic and 0.5 percent TlI. This crystal retained only a small fraction of the added activity and gave a somewhat poorer resolution than commercial NaI(Tl) crystals.

At the time the final measurements were made, the As^{74} activity was less than 1 percent of As^{73}, and no difficulties were encountered due to the presence of As^{74}. No other radioactive contaminants were found.

EXPERIMENTAL RESULTS

Gamma-Ray Spectrum

The spectrum of the arsenic source was investigated with a scintillation spectrometer using a $\frac{1}{2}\times\frac{1}{2}\times\frac{3}{3/2}$-inch cleaved NaI(Tl) crystal[3] which was sealed against moisture with a 0.001-inch aluminum foil. The associated electronic equipment consisted of DuMont K1186 photomultipliers, regulated high-voltage power supplies,[4] a modified version of the non-overload amplifier,[5] and a gray wedge pulse-height analyzer.[6] The spectra were photographed on Kodak type IV-0 spectroscopic film, then printed with such magnification that the intensity scale corresponds to that of commercial semilog paper. Different points on an isodensity line, traced on such paper, would then have the correct relative intensities.

Figure 2 shows a trace of the pulse-height spectrum obtained with an external As^{73} source. The energy scale was calibrated with silver K x-rays (22.0 kev) from a Cd^{109} source. Since the gray wedge used covers an intensity ratio of only 1:100, different exposures were required in order to examine the desired range. In

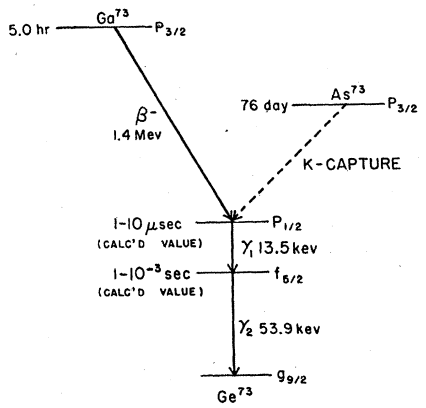

FIG. 1. Johansson's decay scheme for As^{73} and Ga^{73}.

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[1] S. Johansson, Arkiv. Fysik 4, 273 (1952).

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

[3] The NaI(Tl) was supplied by the Harshaw Chemical Company.

[4] W. A. Higinbotham, Rev. Sci. Instr. 22, 429 (1951).

[5] R. L. Chase and W. A. Higinbotham, Rev. Sci. Instr. 23, 34 (1952).

[6] Bernstein, Chase, and Schardt, Rev. Sci. Instr. 24, 437 (1953).

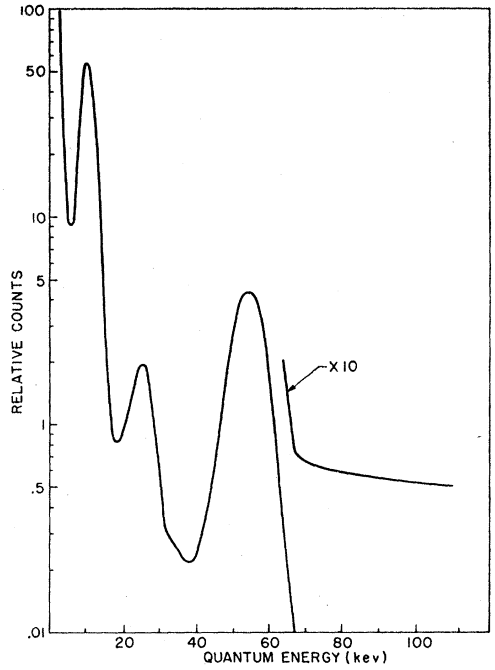

FIG. 2. Pulse-height spectrum of an As^{73} source external to a NaI(Tl) crystal.

addition to the 9.9-kev germanium x-rays and 53.9-kev gamma rays,[7] a line appears at 25 kev which is caused by 53.9-kev quanta ejecting photoelectrons from iodine in the NaI(Tl) scintillator without subsequent capture of the iodine K radiation in the crystal. The 13.5-kev gamma ray[1] is not detectable because of its very high conversion coefficient (see Table II). No line at 81 kev, corresponding to the 75-kev conversion electrons reported by Johansson, was observed. The statistics in the background are such that it would have been possible to see a line 0.2 percent as strong as the 53.9-kev line. Johansson measured the ratio in intensity of the 75-kev conversion electrons to the K conversion electrons associated with the γ_2 transition and found it to be 0.1 percent. Thus, the 81-kev quanta, if present in our source, must have almost as high an internal conversion coefficient as the 53.9-kev line. Similarly, the crossover transition, which would appear at 67.4 kev, is not found.

The pulse height spectrum obtained with the NaI(Tl) crystal containing As^{73} is shown in Fig. 3a. Since this source was much weaker than the external source, the tail of the photomultiplier noise is more noticeable and decreases the peak-to-trough ratio of the 9.9-kev line. The escape peak at 25 kev is absent because the activity is distributed throughout the volume of the crystal. There is still no indication of the 13.5-kev transition reported by Johansson.[1] The gray wedge analyzer requires 200 microseconds to record an event and is insensitive during this period. Consequently, the 13.5-kev transition, if it exists, must follow either the 9.9- or 53.9-kev radiations within this time. Visual observation of individual sweeps on a Tektronix oscilloscope showed small pulses following the 53.9-kev pulses within a few microseconds, but no pulses were observed after the 9.9-kev pulses. Figure 3b shows the spectrum of these delayed events. Special trigger circuits were used to make the gray wedge analyzer sensitive only during the time from 5 to 20 microseconds following a pulse with an amplitude between 40 and 60 kev. Two separate energy measurements gave values of 13.7 and 14.1 kev, respectively, in agreement with Johansson's γ_1 transition. However, this transition definitely follows rather than precedes the 53.9-kev gamma ray. The observation that no 53.9-kev pulses were seen following the 9.9-kev x-rays on the Tektronix scope proves that the 53.9-kev transition must be delayed by more than a few microseconds.

Observations with the internal source permit some qualitative conclusions regarding transitions from As^{73} to the 13.5-kev and ground levels in Ge^{73}. If a long time constant is used in the amplifier, pulses delayed by only 2 or 3 microseconds will add to the height of the original pulse. Pictures taken in this way showed a pronounced asymmetry on the high side of the 53.9-kev line, but left the 9.9-kev line unchanged. This indicates that, at most, few delayed 13.5-kev quanta follow directly after a K capture; in other words, no direct decay was observed from As^{73} to the 13.5-kev level. The relative areas under the 9.9- and 53.9-kev peaks (Fig. 3a) give directly the ratio of K captures to total 53.9-kev transitions. Because the photomultiplier noise background could not be evaluated accurately, only an upper limit of 1.5 can be given for this ratio. This shows that K capture to the ground state is not a very probable mode of decay.

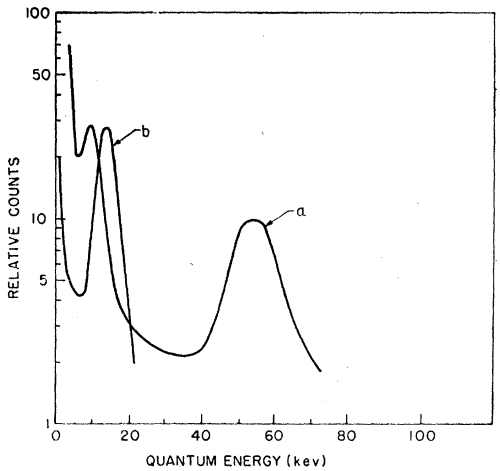

FIG. 3. Pulse-height spectra of an As^{73} source incorporated in a NaI(Tl) crystal. Curve *a* shows the singles spectrum, curve *b* the spectrum of pulses following the 53.9-kev gamma ray.

[7] Because of their greater accuracy, Johansson's energy values have been used throughout the paper.

The relative intensities of the 9.9-, 13.5-, and 53.9-kev photolines can be used to evaluate conversion coefficients. The ratio of the number of K x-rays to that of unconverted 53.9-kev gamma rays was measured with the $\frac{3}{32}$-inch NaI(Tl) crystal and external source. The best value of this ratio was obtained from the ratio of the steps in the integral bias curve, the escape radiation being included with the 53.9-kev photoline. After correction for absorption in the 0.001-inch aluminum foil, the ratio is 6.2.

To look for unconverted 13.5-kev gammas, a proportional counter filled to two atmospheres with a mixture of 90 percent argon and 10 percent methane was used. No evidence for 13.5-kev photons was found, but an upper limit of 1/1400 could be set for the ratio of 13.5-kev quanta to K x-rays. A smaller contribution of 13.5-kev gammas would have been masked by the high energy tail of the x-ray peak. In arriving at the upper limit, account was taken of the relative counting efficiencies (88 and 59 percent) and resolutions (14 and 12 percent full width at half-maximum) at 9.9 and 13.5 kev, respectively, and of absorption in the 1.27 g/cm^2 of beryllium used to absorb the betas from As^{74}.

The experimental intensity ratios listed in Table I were used to calculate the total internal conversion coefficient (α_2) of the 53.9-kev gamma ray, the K/L conversion ratio (K_1/L_1) and a lower limit for the total internal conversion coefficient (α_1) of the 13.5-kev transition. All As^{73} disintegrations were assumed to proceed according to the decay scheme of Fig. 5. The fluorescence yield of the germanium K x-rays was taken as 0.56,[8] the fraction of L capture in As^{73} as 0.11; this latter value was computed according to Rose and Jackson[9] for a decay energy of 320±30 kev.[10] The results of these calculations for α_2, K_1/L_1, and α_1 are given in Table II. As a check on the internal consistency of the data, the ratio $(K_2+L_2+M_2)/(L_1+M_1)$ was evaluated using (L_1+M_1)/Auger, K x-rays/γ_2, and α_2. The calculated ratio is in good agreement with Johansson's experimental value.

TABLE I. Experimental ratios of the intensities of various radiations from As^{73}.

Intensity ratio	Experimental value
$K_2/(L_2+M_2)$	5.6±0.3[a]
L_1/M_1	5.4±0.5[a]
(L_1+M_1)/Auger	0.77[a,b]
(K x-rays)/γ_2	6.2±0.6
(K x-rays)/γ_1	$\geqslant$1400

[a] See reference 1.
[b] For purposes of calculation, a 10 percent experimental error was assigned to this value.

[8] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).
[9] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).
[10] This decay energy was obtained from the threshold of the $Ge^{73}(p,n)As^{73}$ reaction reported by C. C. Trail and C. H. Johnson, Phys. Rev. **91**. 474 (1953).

TABLE II. Conversion coefficients and ratios of conversion electron intensities for the two transitions in Ge^{73}.

Intensity ratio	Values: Calculated from the data of Table I	Values: Measured by Johansson
$\alpha_2=(K_2+L_2+M_2)/\gamma_2$	4.7±0.6	
K_1/L_1	0.63±0.22	<0.5
$\alpha_1=(K_1+L_1+M_1)/\gamma_1$	$\geqslant$1300	
$(K_2+L_2+M_2)/(L_1+M_1)$	1.25	1.2

Measurements of Half-Lives

A gray wedge time-delay analyzer[11] was used to record the data. In this instrument the radiation preceding the delayed transition triggers the blanked, horizontal time sweep of an oscilloscope. The delayed event stops the horizontal sweep and is displayed as a vertical line trace. The face of the oscilloscope is photographed through a gray wedge mounted in such a way that its transparency decreases in the vertical direction. The gray wedge was calibrated as described by Bernstein *et al.*;[6] the time markers from a DuMont 256D oscilloscope were used to calibrate the horizontal sweep. The performance of the analyzer was checked by triggering the horizontal and vertical sweeps by random events. This gave the expected horizontal line.

Since γ_1 is completely internally converted, the internal source was used to measure the half-life of the 13.5-kev level. The discriminators on the time-delay analyzer were set such that any pulse between 40 and 70 kev would start the time sweep. A subsequent pulse between 7 and 70 kev started the display sweep. Figure 4 shows the resulting picture taken on Kodak type IV-O film. The average half-life measured from it and another picture is 4.6±0.5 microseconds. The assigned error is due to the uncertainty in the wedge calibration. Another possible source of error lies in the random after-pulses[12] created in the photomultiplier by the 53.9-kev pulses. This effect was minimized by using a photomultiplier with silver-magnesium dynodes.[13] It was not determined exactly how much of an error this effect could have introduced, but Fig. 3b shows very little evidence of after-pulses.

In an attempt to measure the half-life of the second excited level of Ge^{73}, a krypton-filled proportional counter, channeled on germanium K x-rays, was used to trigger the time sweep of the analyzer. The display sweep was triggered by the photoline of the 53.9-kev gammas as detected with a $\frac{3}{4}$-inch diameter by $\frac{1}{16}$-inch thick NaI(Tl) crystal. Pictures were taken with a Polaroid Land camera; although this results in decreased accuracy for half-life measurements, the data are obtained more rapidly. Time sweeps of 3, 8, 26, 60, 150, and 400 microseconds were used. The source strengths were progressively decreased as the sweep

[11] Schardt, Bernstein, and Chase, Phys. Rev. **90**, 353 (1953).
[12] Mueller, Best, Jackson, and Singletary, Nucleonics **10**, No. 6, 53 (1952).
[13] DuMont K1186.

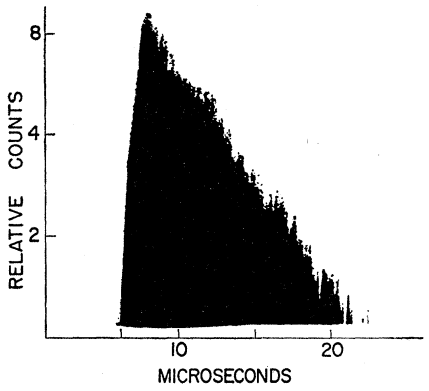

FIG. 4. The decay of the 13.5-kev level as observed on the time-delay analyzer. The measured half-life is 4.6±0.5 microseconds.

time was increased so that, if one half-life occurred during the sweep duration, the number of real coincidences would have been at least ten times the number of chance coincidences. There was no evidence of any decay even when using the 400-microsecond sweep.

Measurements with the 900-microsecond sweep of the time-delay analyzer were made with the internal source in order to take advantage of its greater detection efficiency and improved geometry.[14] The discriminators associated with the time and display sweeps were set to trigger on pulses from 5.5 to 65 kev and 40 to 65 kev, respectively. Again no decay was found, though a lifetime comparable to the sweep time would have been observable on the Polaroid prints. Thus a lower limit of 900 microseconds was set for the half-life of the 53.9-kev transition.

It was possible to set an upper limit on the lifetime of the 53.9-kev transition by making a rapid chemical separation of arsenic from germanium and looking immediately for a decay in the germanium fraction. The procedure[15] used is as follows: An equal volume of 47 percent hypophosphite-free HI was added to a solution of pentavalent As^{73} (containing 0.2-mg As carrier) in $3N$ HCl. The AsI_3 formed was extracted into $CHCl_3$, 2–4 mg of germanium holdback carrier (as GeI_4) were added and the $CHCl_3$ layer was extracted with $1M$ H_2SO_4. Less than 0.4 percent germanium is carried into the H_2SO_4 acid layer, while the arsenic is removed as AsO_2^- with an efficiency >95 percent. The time required for the layers to separate and to drain the $CHCl_3$ layer into a thin-walled glass vessel for counting is ~15 seconds. γ_2 was detected by means of a standard scintillation spectrometer connected to a Brush recorder. In the two experiments performed the germanium fraction had decayed to background within the time of separation. Thus the half-life must be less than 10 seconds, since otherwise the decay of γ_2 would have been discernable on the recorder tape.

While this work was in progress, Campbell[16] measured the half-life of the 53.9-kev transition. His value is 0.33±0.05 second.

DISCUSSION

The available evidence on the decay schemes of As^{73} and Ga^{73} is summarized in Fig. 5. Johansson's spin and parity assignments,[1] which were made partly on the basis of shell theory, have been revised to take into account the order of emission of the two gamma rays. The Ge^{73} ground state is known to be $g_{9/2}$ from the measured spin[17] and from shell theory. The ground states of Ga^{73} and As^{73} are almost certainly $p_{3/2}$ states, in agreement with shell structure and in analogy to the measured spins and moments[17] of Ga^{71} and As^{75}. The logft values for the transitions from Ga^{73} and As^{73} to the second excited state (67.4 kev) of Ge^{73} are 5.9 and 5.4, respectively, indicating allowed transitions. Thus the 67.4-kev level could be $p_{1/2}$, $p_{3/2}$, or $f_{5/2}$; any of these assignments is compatible with the single-particle model. However, the $f_{5/2}$ assignment can be eliminated because the 67.4-kev crossover transition was unobserved ($<2\times10^{-3}$ of unconverted 53.9-kev transitions). According to Blatt and Weisskopf,[18] the theoretical $M2$ gamma half-life for an $f_{5/2} \longrightarrow g_{9/2}$ transition of 67.4 kev is $\sim10^{-3}$ sec, a factor of about 10^5 shorter than the experimental lower limit.

The 53.9-kev transition can be labeled fairly definitely as $M2$. Extrapolation of the K conversion coefficient data of Rose *et al.*[19] to lower energies with the aid of Spinrad's[20] threshold values shows that the experimental value, $\alpha_K=4.0$, is compatible only with a $M2$ or $E2$ assignment. The measured K/L ratio of 6 to 7[1] is in excellent agreement with the value 6.8 obtained from Goldhaber and Sunyar's[2] curve for $M2$, but not with the value 1.7 for $E2$. In the case of the 13.5-kev transition, the evidence points to an $E2$ assignment, with a possible small admixture of $M1$. Transitions of multipole order greater than two can be ruled out on the basis of lifetime considerations. The threshold values for the total K conversion coefficients,[20] ~64 ($E1$), ~44 ($M1$), ~670 ($E2$) can be used to eliminate pure $E1$ and $M1$ type transitions since the experimental lower limit for α_K is 450. The measured K/L ratio, 0.63±0.22 is higher than the value, ~0.15, obtained by extrapolating Goldhaber and Sunyar's[2] curve for $E2$ from $Z^2/E=60$ to $Z^2/E=75$.

[14] With the external source, processes resulting in Auger and internal conversion electrons could not be detected. The calculated accidental rate is low enough with the internal source to permit use of a 10-millisecond sweep time. However, 900 microseconds is the longest sweep for which the analyzer was designed.

[15] We are indebted to Dr. René J. Prestwood, Los Alamos Scientific Laboratory, for suggesting the separation method.

[16] E. C. Campbell (private communication).

[17] Way, Fano, Scott, and Thew, *Nuclear Data*, National Bureau of Standards Circular 499 and Supplements (U. S. Government Printing Office, Washington, D. C., 1950).

[18] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

[19] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

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

However, a recent accurate measurement[21] of the K/L ratio for the 84-kev transition in Yb^{170}, 0.36 ± 0.04 at $Z^2/E=57$, indicates that the $E2$ curve is too low for large values of Z^2/E; considering the experimental accuracy of the original data used to construct the curve, such a displacement is not unreasonable. Even so, the observed K/L ratio for the 13.5-kev transition appears to be somewhat high for pure $E2$, and a small admixture of $M1$ $(K/L\sim7.4)^2$ cannot be excluded.

Since the 67.4-kev level of Ge^{73} is either $p_{1/2}$ or $p_{3/2}$, the 13.5-kev level must be either $5/2+$ or $7/2+$, respectively. Thus the two possible assignments for the gamma cascade are

$$p_{1/2}\xrightarrow{M2}5/2+\xrightarrow{E2}g_{9/2},\quad p_{3/2}\xrightarrow{M2}7/2+\xrightarrow{M1+E2}g_{9/2}.$$

The intermediate level cannot be interpreted on the basis of the single particle model since no low-lying $d_{5/2}$ or $g_{7/2}$ levels can be expected for the forty-first neutron in Ge^{73}. A reasonable assumption would be that three neutrons are in the $g_{9/2}$ shell and that this $(g_{9/2})^3$ configuration has spin $5/2+$ or $7/2+$. The $(g_{9/2})^3$ configuration is also compatible with the fact that the 53.9-kev transition has a half-life about 600 times longer than would be expected for a single particle $M2$ transition. The gamma half-life of the 13.5-kev transition ($\geq6\times10^{-3}$ sec) is to be compared with the theoretical values,[18] 4×10^{-2} sec for $E2$, and 9×10^{-9} sec for $M1$. Thus, at first glance, the lifetime would appear to favor a pure $E2$ assignment for the transition and therefore a $5/2+$ designation for the 13.5-kev level. However, as discussed above, this level most likely has a $(g_{9/2})^3$ configuration and therefore one would expect the transition to the ground state to be considerably slower than a single particle $E2$ transition. A $7/2+$ assignment for the 13.5-kev state and a mixed $M1+E2$ transition to the ground state is thus a definite possibility, although this interpretation would require an exceedingly small matrix element for the $M1$ transition.

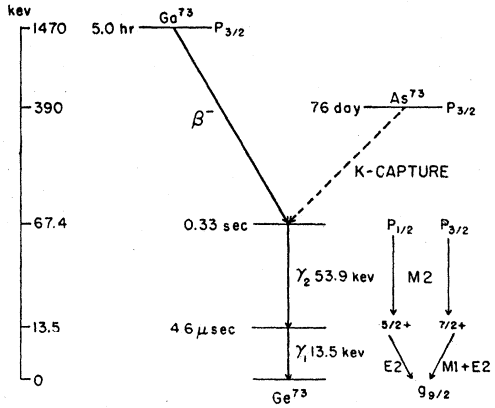

FIG. 5. Revised decay scheme of As^{73} and Ga^{73}.

The fact that the lifetimes of the two gamma transitions are long compared with those expected for $M2$ and $M1$ transitions, respectively, points to a relatively high purity of the postulated $(g_{9/2})^3$ configuration of the intermediate level. This is in accord with recent considerations on mixed configurations[22] which make it plausible that purer configurations occur at the beginning than in the middle of shells.

The authors would like to thank Dr. M. Goldhaber and Dr. J. Mihelich for valuable discussions. A great deal of help with electronic problems was given by Mr. R. L. Chase, who also designed the time-delay analyzer.

Note added in proof.—In a recent paper (Compt. rend. **236**, 1872 (1953)) R. Barloutaud and M. Sartory propose a decay scheme for As^{73} substantially different from that of Fig. 5 and incompatible with the evidence given in the present paper. However, Dr. Barloutaud has kindly informed us that his experimental data are consistent with our decay scheme.

[21] Graham, Wolfson, and Bell, Can. J. Phys. **30**, 459 (1952).

[22] A. De-Shalit and M. Goldhaber (private communication).