

Nuclear Structure of  $^{120}_{50}\text{Sn}$ 

HIDETSUGU Ikegami

*Institute for Nuclear Study, University of Tokyo, Tanashi-machi, Tokyo, Japan*

(Received July 22, 1960)

The radiations from 5.8-day  $\text{Sb}^{120m}$  have been investigated in detail, using sector-type double-focusing magnetic spectrometers and scintillation counters. The following measurements were performed: coincident gamma-ray spectra, directional correlations of 1.18 Mev  $\gamma$ —1.03 Mev  $\gamma$ , 1.03 Mev  $\gamma$ —0.20 Mev  $\gamma$ , 1.18 Mev  $\gamma$ —0.20 Mev  $\gamma$ , 0.20 Mev  $\gamma$ —0.09 Mev  $\gamma$ , 0.20 Mev  $\text{Ke}^-$ —0.09 Mev  $\gamma$  and 0.20 Mev  $\gamma$ —0.09 Mev  $\text{Ke}^-$ . The 1.18 Mev  $\gamma$ —1.03 Mev  $\gamma$  polarization direction correlation, the conversion electron spectra, and the mean lives of the 2.41- and 2.50-Mev levels have also been investigated. The decay scheme of 5.8-day  $\text{Sb}^{120m}$  proposed by McGinnis, which gives the following assignments to the excited states in  $\text{Sn}^{120}$ , 1.18 Mev ( $2^+$ ), 2.21 Mev ( $4^+$ ), 2.41 Mev ( $6^+$ ), and 2.50 Mev ( $7^-$ ), has been confirmed uniquely. These levels have been interpreted as arising from the neutron configurations  $(2d_{3/2})^2(1h_{11/2})^2$  and  $(2d_{3/2})^3(1h_{11/2})^1$ . The lifetimes

of the 2.41-, 1.18-, and probably 2.21-Mev levels are shorter by a factor 3–4 than those expected for single-proton transitions and may be explained by the neutron transition, assuming a somewhat larger effective charge of neutron in spherical nuclei. The high forbiddenness of the 0.09-Mev  $E1+M2$  transition is ascribable to the simultaneous  $j$ - and  $l$ -forbidden transition between the states involving  $(2d_{3/2})^3(1h_{11/2})^1$  and  $(2d_{3/2})^2(1h_{11/2})^2$  configurations.

No appreciable effects of the nuclear finite size or any nuclear structure dependence on the internal conversion process have been detected even for the highly forbidden 0.09-Mev  $E1+M2$  transition.

In the decay of 16-min  $\text{Sb}^{120}$ , the 1.18-Mev gamma ray has been detected. However, other gamma rays expected from the second level multiplet, which is a characteristic feature of the vibrational model, have not been found.

## 1. INTRODUCTION

THE low-lying states of even-even medium weight nuclei have been explained successfully by Scharff-Goldhaber and Weneser's free vibrational model,<sup>1</sup> Wilets and Jean's shape-unstable model<sup>2</sup> or Davydov and Filippov's asymmetric rotor model.<sup>3</sup> However, these models for medium weight nuclei are not as satisfactory as Bohr and Mottelson's model<sup>4</sup> for heavy nuclei. On the other hand, level structures of most nuclei in the neighborhood of a closed shell have been described very well in terms of the shell model. For Sn isotopes ( $Z=50$ ), however, there are at present few theoretical descriptions and the experimental results available are somewhat ambiguous (see Fig. 1), although the levels of  $\text{Zr}^{90}$  ( $N=50$ ) are well known<sup>5</sup> and have been interpreted quite satisfactorily. Furthermore, Cd isotopes, the neighboring even-even nuclei of Sn, have been interpreted successfully on the basis of the vibrational model.<sup>6</sup> It is, therefore, felt worthwhile to make detailed investigations of Sn isotopes not only for the spin and parity of excited levels and the

level spacing, but also for absolute and relative transition probability, ratio and phase of multipole mixing of gamma transitions and the  $ft$  value in beta decays.

Among Sn isotopes,  $\text{Sn}^{120}$  ( $Z=50$ ,  $N=70$ ) is one of the interesting nuclei because of the highly forbidden electric dipole transition of the 2.50-Mev state which can give rise to an anomalous internal conversion process through the nuclear structure effect first pointed out by Church and Weneser.<sup>7</sup> The experimental data on this process are, however, not sufficiently accurate.

The radioactivity of the long-lived  $\text{Sb}^{120m}$  was investigated by Linder and Perlman by an absorption method.<sup>8</sup> Thereafter, McGinnis investigated the decays of 5.8-day  $\text{Sb}^{120m}$  and 16-min  $\text{Sb}^{120}$  with a scintillation gamma-ray spectrometer.<sup>9</sup> The anisotropies of directional correlations for 1.03 Mev  $\gamma$ —1.18 Mev  $\gamma$ , 0.20 Mev  $\gamma$ —1.03 Mev  $\gamma$ , and 0.09 Mev  $\gamma$ —0.20 Mev  $\gamma$  cascades were measured. The relative intensities of conversion electrons were also studied with a magnetic thin-lens spectrometer. An isomeric state of 11  $\mu\text{sec}$  was assigned to the 2.50-Mev level of  $\text{Sn}^{120}$ . The log  $ft$  value of positron disintegration of 16-min  $\text{Sb}^{120}$  was obtained. However, the order of emission of the 0.09- and 0.20-Mev gamma rays, the spin of the 2.50-Mev level, and the multipole mixing ratio of the 0.09-Mev transition remain somewhat uncertain. Most of his

<sup>1</sup> G. Scharff-Goldhaber and J. Weneser, Phys. Rev. **98**, 212 (1955); J. Raz, Phys. Rev. **114**, 1116 (1959).

<sup>2</sup> L. Wilets and M. Jeans, Phys. Rev. **102**, 788 (1956).

<sup>3</sup> A. S. Davidov and G. F. Filippov, Nuclear Phys. **8**, 237 (1958); **10**, 654 (1959). A. S. Davidov and V. S. Rostovsky, Nuclear Phys. **12**, 58 (1959); T. Tamura and T. Udagawa, Nuclear Phys. **16**, 460 (1960).

<sup>4</sup> A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

<sup>5</sup> R. K. Sheline, Bull. Am. Phys. Soc. **2**, 260 (1957); *Proceedings of the Pittsburgh Conference on Nuclear Structure*, edited by S. Meshkov (University of Pittsburgh and Office of Ordnance Research, U. S. Army, 1957), p. 481. S. Bjørnholm, O. B. Nielson, and R. K. Sheline, Phys. Rev. **115**, 1613 (1959); B. F. Bayman, A. S. Reiner, and R. K. Sheline, Phys. Rev. **115**, 1627 (1959). R. B. Day, A. E. Johnsrud, and D. A. Lind, Bull. Am. Phys. Soc. **1**, 56 (1956).

<sup>6</sup> H. T. Motz, Phys. Rev. **104**, 1353 (1956); B. L. Cohen and R. E. Price, Phys. Rev. **118**, 1582 (1960); T. Tamura and L. G. Komai, Phys. Rev. Letters **3**, 344 (1959).

<sup>7</sup> E. Church and J. Weneser, Phys. Rev. **100**, 943 (1955); **100**, 1241 (1955); **103**, 1035 (1956), Bull. Am. Phys. Soc. **1**, 330 (1956); Phys. Rev. **104**, 1382 (1956). J. Weneser and E. Church, Bull. Am. Phys. Soc. **1**, 181 (1956). T. Green and M. E. Rose, Phys. Rev. **110**, 105 (1958); Bull. Am. Phys. Soc. **2**, 228 (1957). L. S. Kisslinger, Phys. Rev. **114**, 292 (1959). A. S. Reiner, *Proceedings of the 1957 International Conference on Nuclear Structure, Rehovoth, Israel*, edited by J. Lipkin (North-Holland Publishing Company, Amsterdam, 1958); Nuclear Phys. **5**, 544 (1958).

<sup>8</sup> M. Linder and I. Perlman, Phys. Rev. **73**, 1124 (1948).

<sup>9</sup> C. L. McGinnis, Phys. Rev. **98**, 1172 (A) (1955); **109**, 888 (1958).

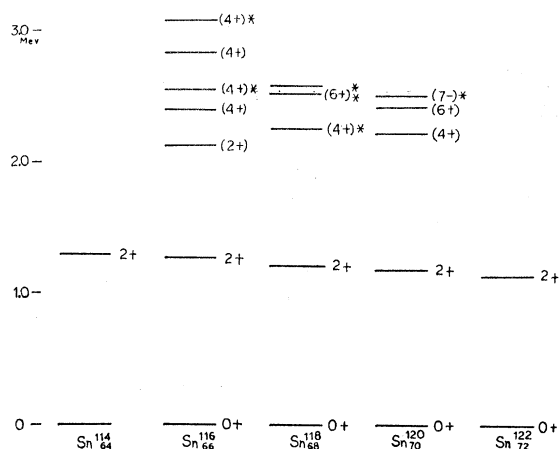


FIG. 1. Comparison of the energy levels of tin isotopes. The asterisks indicate that the level assignments are uncertain.

results are confirmed thoroughly by the present investigation and shown schematically in Fig. 2.

## 2. GENERAL EXPERIMENTAL PROCEDURES

The 5.8-day  $\text{Sb}^{120m}$  was prepared by the irradiation of special spectroscopically pure tin and also 98% isotopically enriched  $\text{Sn}^{120}$  with an internal beam of 12~14 Mev protons in the variable energy 64-inch cyclotron of our Institute.<sup>11</sup> Irradiation with deuterons was not adopted because of the production of undesirable radioactive isotopes. Irradiated tin was dissolved in *aqua regia*, then converted into 8N hydrochloric acid solution and extracted with isopropyl-ether repeatedly. The organic layer was washed with 3N hydrochloric acid and the solvent was evaporated off in the presence of 3N hydrochloric acid containing a small amount of hydro-oxylamine hydrochloride.<sup>12</sup> This residual acid solution was used as the source throughout most of the gamma-ray spectroscopic measurements. In the measurements of the conversion electron, extremely thin sources of the metallic carrier free  $\text{Sb}^{120m}$  were prepared electrochemically by cathodic electrodeposition. As a backing material, a 250- $\mu\text{g}/\text{cm}^2$  thick rubber hydrochloride film was used, on which a 20- $\mu\text{g}/\text{cm}^2$  copper coating was evaporated. The plated sources were about 2  $\mu\text{g}/\text{cm}^2$  in thickness and 3 mm $\times$ 10 mm in area. The metallic Sb sources prepared in this way were almost invisible.

<sup>10</sup> The author wishes to express his gratitude to Mr. K. Itsuki of Osaka Refinery, Mitsubishi Metal Mining Industry, for his kind supply of special pure tin (antimony content less than  $8 \times 10^{-4}\%$ ). The enriched  $\text{Sn}^{120}$  was supplied in the form of  $\text{SnO}_2$  by the Oak Ridge National Laboratory.

<sup>11</sup> S. Kikuchi, I. Nonaka, H. Ikeda, H. Kumagai, Y. Saji, J. Sanada, S. Suwa, A. Isoya, I. Hayashi, K. Matsuda, H. Yamaguchi, T. Mikumo, K. Nishimura, T. Karasawa, S. Kobayashi, K. Kikuchi, S. Ito, A. Suzuki, S. Takeuchi, and H. Ogawa, J. Phys. Soc. (Japan) **15**, 41 (1960).

<sup>12</sup> N. Suzuki and H. Ikegami, Bull. Chem. Soc. Japan (to be published).

The measurements of coincidence and directional correlation of gamma rays were performed with a scintillation coincidence spectrometer with an effective resolving time of  $4 \times 10^{-7}$  sec. Two NaI(Tl) crystals  $1\frac{3}{4}$  in. in diameter and 2 in. long were each coupled to RCA-6342 or DuMont 6292 photomultipliers and shielded laterally by conical Pb shields. The crystals were at a distance of 70 mm from the source. Aluminum or Lucite absorbers were put in front of each crystal to stop all conversion electrons and beta rays. For the directional correlation measurements, the source was always centered so that the single counting rate in the movable detector as a function of position was constant to about one percent. Single counting rates as well as the coincidence rate were recorded.

Conversion electron spectra were investigated by a sector-type double-focusing beta-ray spectrometer with the reference radius  $\rho=18$  cm, (hereafter called as INS-III spectrometer).<sup>13</sup> Electron-gamma directional correlation measurements were accomplished by another small sector-type double-focusing beta-ray spectrometer which was just half the above-mentioned

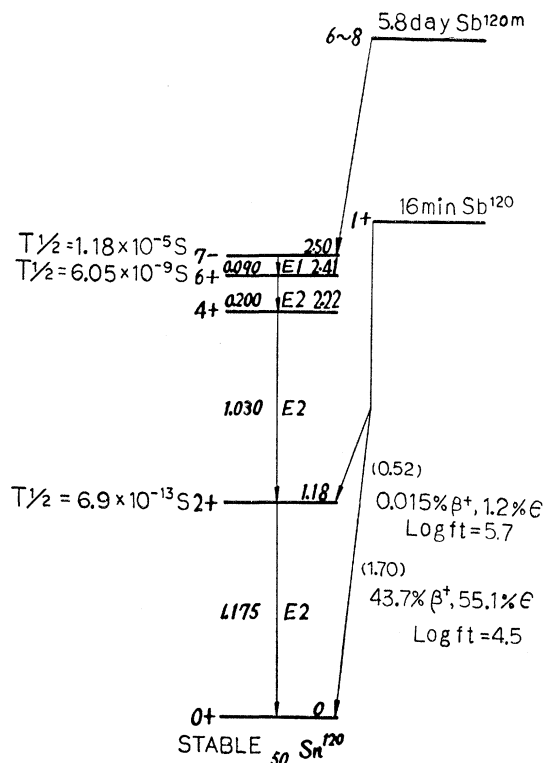


FIG. 2. Level scheme of  $\text{Sn}^{120}$  proposed by McGinnis and confirmed by present data from 5.8-day  $\text{Sb}^{120m}$  and 16-min  $\text{Sb}^{120}$  decays. Some of the values are added and modified by the present measurement.

<sup>13</sup> M. Sakai, H. Ikegami, and T. Yamazaki, Nuclear Instr. (to be published). General properties of a sector-type double-focusing spectrometer are treated by D. L. Judd, Rev. Sci. Instr. **21**, 213 (1950); E. S. Rosenblum, Rev. Sci. Instr. **21**, 586 (1950); H. Ikegami, Rev. Sci. Instr. **29**, 943 (1958).

apparatus (hereafter called as INS-I spectrometer).<sup>14,15</sup> As the detector for low-energy electrons, a GM counter with 10 mm diameter and 500- $\mu\text{g}/\text{cm}^2$  thick mica window was used, whose cutoff energy was less than 20 keV.

A mean life of 2.41-MeV level was measured, using two plastic scintillators<sup>16</sup> 30 mm in diameter and 30 mm long coupled to RCA-6810A photomultipliers and a fast time-to-pulse-height converter.<sup>17</sup> The effective resolving time was estimated as  $6 \times 10^{-10}$  sec for the 1.17- and 1.33-MeV cascade gamma rays from  $\text{Co}^{60}$ .

The 16-min  $\text{Sb}^{120}$  was prepared by the bombardment of 98% enriched  $\text{Sn}^{120}$  with 7–8 MeV protons and the irradiations of natural metallic antimony with fast neutrons from a graphite target bombarded with 24-MeV deuterons.

TABLE I. Summary of all the experimental information on the relative intensities of radiations from  $\text{Sb}^{120m}$ .

| Transition energy (MeV)  | Relative intensities of radiations from 5.8-day $\text{Sb}^{120m}$ |  |         |
|--|--|--|---------|
|  | Gamma-ray (scintillation counter)                                  | Total conversion electron (double focusing spectrometer) |         |
| $1.175 \pm 0.010$  | 1.00   | 1.00   | (E2)    |
| $1.030 \pm 0.010$  | $1.02 \pm 0.05$  | $1.24 \pm 0.10$  | (E2)    |
| $0.200 \pm 0.002$  | $0.99 \pm 0.05$  | $146.4 \pm 10$   | (E2)    |
| $0.090 \pm 0.002$  | $1.04 \pm 0.05$  | $260.6 \pm 20$   | (E1+M2) |
| 0.290  |  | $0.1 \pm 0.1$  | (E3)    |
| $\beta^+ < 0.3\%$ .  |  |  |         |
| 0.290-MeV cross-over branch $< 0.03\%$ .   |  |  |         |
| Coinc. ratio $(K \times \text{ray}) - (0.09 \text{ MeV } \gamma) / (K \times \text{ray}) - (0.20 \text{ MeV } \gamma)$ |  |  |         |
| $= 0.65 \pm 0.07$ at no delay, with 1- $\mu\text{sec}$ resolving time.   |  |  |         |
| Thus, feeding to 2.50-MeV level $> 98\%$ .   |  |  |         |

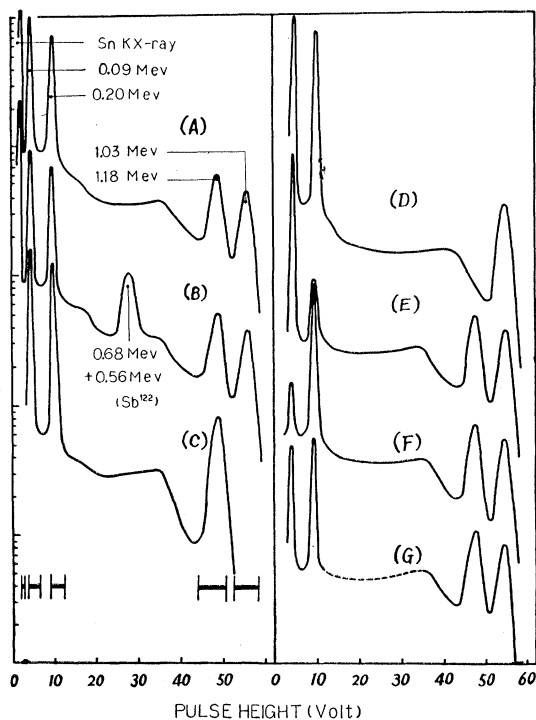


FIG. 3. Spectra of gamma rays from 98% enriched  $\text{Sn}^{120}$  and natural Sn bombarded by 13-MeV protons. (A) and (B) are the single gamma-ray spectra from enriched  $\text{Sn}^{120}$  and natural Sn, respectively. The other curves represent gamma rays in coincidence with (C) 1.18-MeV  $\gamma$ , (D) 1.03-MeV  $\gamma$ , (E) 0.20-MeV  $\gamma$ , (F) 0.09-MeV  $\gamma$ , and (G) Sn K x ray. The horizontal bars represent the positions of the gate window opened in the measurements of coincidence spectra and directional correlations. In (E) and (F), the small peaks at the positions of gating gamma rays are due to the coincidence with Compton-scattered higher energy gamma rays.

### 3. SPECTRA OF GAMMA RAYS AND THEIR COINCIDENCE PROPERTIES

It is known that there are Sn K x rays and four gamma rays whose energies are 0.09, 0.20, 1.03, and 1.18 MeV, respectively, in the decay of 5.8-day  $\text{Sb}^{120m}$ . Figure 3 shows the single gamma-ray spectra and coincidence spectra of each gating radiation measured with a 20-channel pulse-height analyzer and the coincidence circuit described in Sec. 2. The horizontal bars shown in the figure represent the window width opened for respective gating radiations. From these coincidence data, it becomes clear that all four gamma rays are emitted in cascade. The cross-over 0.29-MeV gamma ray was not detected. This fact is consistent with the results derived from the conversion electron spectra as will be shown in Sec. 6. The fact that no pronounced directional correlation resulting from annihilation radiations was observed confirms that no positive electrons were detected by the double-focusing beta-ray spectrometer.

Typical values of intensities of the  $\text{Sb}^{120m}$  radiations are summarized in Table I.

The appearance of 1.18-MeV gamma ray in the decay of 16-min  $\text{Sb}^{120}$  and the observation of 1.16-MeV gamma ray by Coulomb excitation,<sup>18</sup> determine the first excited level of  $\text{Sn}^{120}$  at 1.18 MeV uniquely. Furthermore, we can determine the order of emission of 0.09- and 0.20-MeV gamma rays by the fast delayed coincidence technique which will be described in Sec. 4. Thus it is quite reasonable that the order of radiations is 0.09, 0.20, 1.03, and 1.18 MeV.

From the observed intensity ratio of 0.09- and 0.20-MeV gamma rays coincident with the gating K x ray  $(0.09 \text{ MeV } \gamma - K \times \text{ray}) / (0.20 \text{ MeV } \gamma - K \times \text{ray})$ ,  $0.65 \pm 0.07$ , which is also shown in the lower part of Table I, it is concluded that the feeding to the 2.50-MeV level from the 5.8-day  $\text{Sb}^{120m}$  by electron capture is more than 98%, taking into account the conversion

<sup>14</sup> M. Sakai and H. Ikegami, J. Phys. Soc. (Japan) **13**, 1076 (1958).

<sup>15</sup> H. Ikegami, M. Sakai, and T. Yamazaki, J. Phys. Soc. (Japan) (to be published).

<sup>16</sup> Plastic phosphors are of Polystyrene with 1% *p*-terphenyl, 0.03% POPOP and 0.03% zincstearate.

<sup>17</sup> Y. Ishizaki, N. Yoshimura, E. Takekoshi, and H. Ikegami, J. Phys. Soc. (Japan) (to be published).

<sup>18</sup> P. H. Stelson and F. K. McGowan, Bull. Am. Phys. Soc. **2**, 69 (1957).

coefficients of these gamma rays and the larger decay time of the 2.50-Mev level described in Sec. 4.

#### 4. MEAN LIVES OF THE EXCITED LEVELS OF $\text{Sn}^{120}$

Measurements of nuclear lifetime are valuable from two points of view. First, from the matrix elements of gamma-ray transitions many valuable conclusions can be drawn to check the existing nuclear models and to obtain hints for necessary modifications. Second, if one could investigate the directional correlation, the possibility of perturbation of the nuclear intermediate state by the interaction responsible for the well-known hyperfine structure in atomic spectra could be estimated from the lifetime of the intermediate state.

##### (a) The 1.18- and 2.21-Mev Levels

By Coulomb excitation, the mean life of the first excited level of  $\text{Sn}^{120}$  is determined as  $1.0 \times 10^{-12}$  sec,<sup>18</sup> and this value agrees well with the main features of the systematics of the first excited levels of even-even medium-weight nuclei.<sup>19</sup> From this and the positions of the first and second levels of  $\text{Sn}^{120}$ , it seems to be reasonable to estimate the mean life of the second level as  $2.0 \times 10^{-12}$  sec or less so far as the transition is of  $E2$  type, assuming that the matrix elements of 1.03- and 1.18-Mev gamma transitions are nearly equal. Therefore, we can neglect the perturbation effects of the directional correlations for the 1.18- and 2.21-Mev levels.

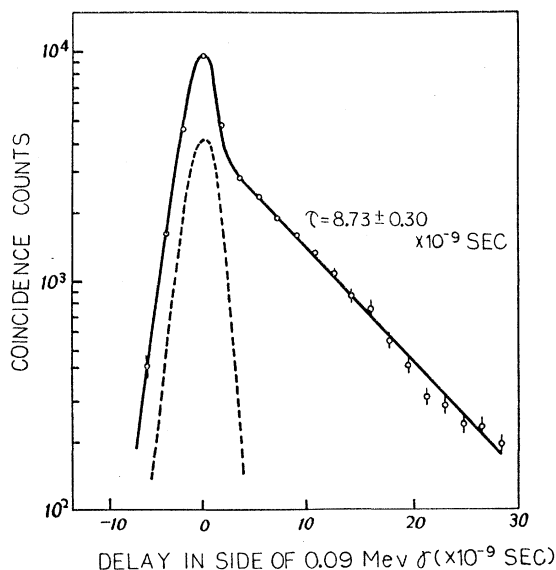


FIG. 4. "Delayed" curve for the 2.41-Mev level of  $\text{Sn}^{120}$ . The solid line is drawn by the least squares method through the experimental points. The "prompt" curve (dashed curve) is obtained by annihilation gamma rays from  $\text{Na}^{22}$ . The prompt coincidence part on the delayed curve is ascribable to the contribution from the Compton-scattered higher energy gamma rays.

<sup>19</sup> G. T. Temmer and N. P. Heydenburg, *Phys. Rev.* **104**, 967 (1956); H. Ikegami, *Genshikaku-Kenkyu* **4**, 104 (1959) in Japanese.

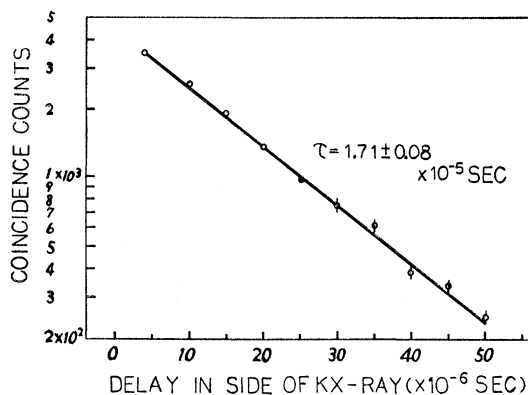


FIG. 5. "Delayed" curve for the 2.50-Mev level of  $\text{Sn}^{120}$ . The solid line represents the least squares fit.

##### (b) The 2.41-Mev Level

Measurement of the fast delayed coincidence between the cascade gamma rays from the 2.50- and 2.41-Mev levels was accomplished using two plastic scintillators<sup>16</sup> coupled to RCA-6810A photomultipliers. The negative pulses produced at the anodes of 6810A cut off the anode current of an E180F pentode and then the pulses were limited and clipped to about  $3 \times 10^{-8}$  sec through RG63/U cable. The time interval between the cascade radiation pulses was measured, using the time-to-pulse-height converter described in Sec. 2 and a 20-channel pulse-height analyzer. Typical data are shown in Fig. 4. The prompt curve obtained by the annihilation gamma rays from  $\text{Na}^{22}$  with the same arrangements of apparatus is also shown for comparison.

The prompt coincidence part on the decay curve is ascribable to the contribution from the higher energy gamma rays. The least squares fitting gives the mean life  $\tau = (8.73 \pm 0.30) \times 10^{-9}$  sec [ $T_{1/2} = (6.05 \pm 0.20) \times 10^{-9}$  sec].<sup>20</sup> Since the 0.20-Mev transition is of pure  $E2$  type and its conversion coefficient  $\alpha$  is 0.133 as shown in Sec. 6, the mean life of the 0.20-Mev gamma transition is  $\tau_\gamma = (9.87 \pm 0.40) \times 10^{-9}$  sec.

##### (c) The 2.50-Mev Level

The lifetime of the 2.50-Mev level is reported as  $T_{1/2} = (1.1 \pm 0.1) \times 10^{-5}$  sec by McGinnis.<sup>9</sup> We have confirmed his result using a coincidence spectrometer with an effective resolving time of  $1 \times 10^{-6}$  sec and an electronic time-delay generator whose pulses can be delayed from  $1 \times 10^{-7}$  sec to  $9 \times 10^{-5}$  sec in discrete steps. Experimental results are shown in Fig. 5 and the least squares fitting yields the mean life  $\tau = (1.71 \pm 0.08) \times 10^{-5}$  sec [ $T_{1/2} = (1.18 \pm 0.05) \times 10^{-5}$  sec]. As the conversion coefficient  $\alpha$  is 0.24 as will be shown in Sec. 6, the mean life of 0.09-Mev gamma transition is  $\tau_\gamma = (2.12 \pm 0.10) \times 10^{-5}$  sec.

<sup>20</sup> H. Ikegami, Y. Ishizaki, and E. Takekoshi (to be published).

TABLE II. Comparison of the experimental transition probabilities with Moszkowski's estimations by the assumption of a single-proton transition.

| Transition             | $\tau_\gamma(\text{exp})(\text{sec})$ | $F = \tau_\gamma(\text{theo})/\tau_\gamma(\text{exp})$ |
|------------------------|---------------------------------------|--|
| 1.18 Mev ( <i>E2</i> ) | $1.00 \times 10^{-12}$                | 4.4  |
| 0.20 Mev ( <i>E2</i> ) | $9.89 \times 10^{-9}$                 | 3.2  |
| 0.09 Mev ( <i>E1</i> ) | $2.12 \times 10^{-8}$                 | $1.7 \times 10^{-8}$                                   |
| 0.09 Mev ( <i>M2</i> ) | $\gtrsim 1.6$                         | $\lesssim 3.5 \times 10^{-5}$                          |

#### (d) Comparison of Experimental Results with the Single-Particle Estimates

With the assignment of multipoles as will be described in Sec. 5 and Sec. 6, the experimental results for the transition probabilities of the 1.18-, 0.20-, and 0.09-Mev gamma rays are compared with the theoretical values assuming a single-proton transition.<sup>21</sup> These are summarized in Table II.

The emission of the 0.09-Mev *E1* gamma ray is expected, because of an admixture of neighboring orbitals in the particle state even if it is highly forbidden, by the effect of simultaneous *j*- and *l*-selection rule between the states of the neutron configuration  $[(2d_{3/2})^3(1h_{11/2})^1]_{7-}$  and  $[(2d_{3/2})^2(1h_{11/2})^2]_{6+}$ . It is, however, interesting to note that a highly forbidden transition such as the 0.09-Mev gamma-ray emission has not yet been found in even-even medium weight nuclei. The same argument would also hold for the 0.09-Mev *M2* transition and therefore its small probability is consistent with the observation.

The favored factors  $F = \tau_\gamma(\text{theo})/\tau_\gamma(\text{exp})$  of 1.18- and 0.20-Mev transitions are of the same order of magnitude. Hence, the characteristic quantities of the states such as magnetic moment and quadrupole moment, etc., may be expected to be almost the same for the 1.18- and 2.41-Mev levels.

#### 5. GAMMA-GAMMA DIRECTIONAL CORRELATIONS

$\text{Sb}^{120m}$  sources in the form of  $\text{SbCl}_3$  in HCl solution described in Sec. 2 were used for most of the correlation measurements in order not to attenuate the directional correlation. The solution was filled in a 5-mm diameter and 5-mm long Lucite case whose wall thickness was 2.0 mm.

The channel widths for the respective gamma rays were chosen to be the same as those in Fig. 3. The number of random coincidences was about one to seven percent of the total number of coincidences in most of the measurements.

Subtracting random coincidences and correcting for the variation of the single counting rates, the data were

<sup>21</sup> In this paper, statistical factors are neglected (i.e.,  $S=1$  in Moszkowski's formula). V. F. Weisskopf, Phys. Rev. **83**, 1073 (1951). J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, New York, 1952), p. 625. S. A. Moszkowski, Phys. Rev. **89**, 474 (1953); *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. XIII. B. Stech, Z. Naturforsch. **7a**, 401 (1952).

fitted by the least squares method to a directional correlation function

$$W(\theta) = \alpha_0 \pm \sigma_0 + (\alpha_2 \pm \sigma_2)P_2(\cos \theta) + (\alpha_4 \pm \sigma_4)P_4(\cos \theta),$$

where  $\sigma_0$ ,  $\sigma_2$ , and  $\sigma_4$  are the probable errors defined by Rose.<sup>22</sup> These would include the contributions from the gamma rays of higher energy, which were absorbed in the scintillators by Compton effect, and had to be corrected as described later. The data were normalized to the form

$$W(\theta) = 1 + (A_2 \pm \Delta_2)P_2(\cos \theta) + (A_4 \pm \Delta_4)P_4(\cos \theta).$$

Then the coefficients  $A_2$  and  $A_4$  were corrected for the finite angular resolution of the detectors in the manner described by Lawson and Frauenfelder,<sup>23</sup> except that the pulse-height analyzer was set to accept only the photopeaks of gamma rays from the collimator. The correction factors were determined for gamma rays of an energy from 0.1 Mev to 1.3 Mev.

The experimental results corrected for accidental coincidences and the variation of single counting rates are presented in Fig. 6 and Fig. 7. The solid curves show the least squares fits to the experimental points.

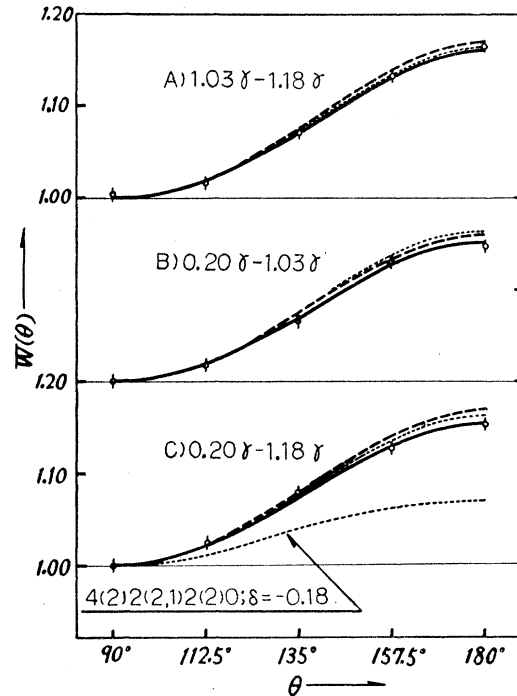


FIG. 6. Experimental directional correlation functions for the gamma rays of  $\text{Sn}^{120}$ . The statistical errors are indicated by vertical bars. The solid lines represent the least squares fit to the experimental points. The broken lines are the correlation functions corrected for the finite solid angle of the detectors. The dotted curves represent the theoretical functions for an  $I+4(2)I+2(2)I$  cascade. In (C), the theoretical function for a  $4(2)2(2,1)2(2)0$  cascade with  $\delta = -0.18$  is also represented for comparison.

<sup>22</sup> M. E. Rose, Phys. Rev. **91**, 610 (1953). See also E. Breitenberger, Proc. Phys. Soc. (London) **A69**, 489 (1956).

<sup>23</sup> J. S. Lawson and H. Frauenfelder, Phys. Rev. **91**, 649 (1953).

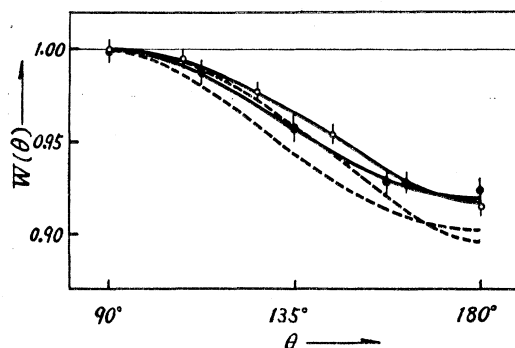


FIG. 7. Experimental directional correlation function of 0.09 Mev  $\gamma$ —0.20 Mev  $\gamma$ . The least squares fitting is shown by the solid line. The open and closed circles indicate the experimental points for the source formed from  $\text{SbCl}_3 \cdot \text{HCl}$  solution and the metallic antimony source, respectively. The broken curves are the experimental directional correlation functions corrected for the finite sizes of source and detectors and also for interfering cascades.

The broken curves are those corrected for the finite angular resolution. The dotted curves are the expected theoretical functions.

#### (a) The 1.03 Mev $\gamma$ —1.18 Mev $\gamma$ Directional Correlation

Correcting for the finite angular resolution, we obtain the coefficients of Legendre's polynomials in the correlation function as

$$A_2 = +0.106 \pm 0.002, \quad A_4 = +0.012 \pm 0.001.$$

Thus, for the 1.03 Mev—1.18 Mev cascade, the spin sequence could be  $4(2)2(2)0$  or  $2(2,1)2(2)0$  with  $\delta = -0.18$ , where  $\delta$  denotes the ratio of the matrix element of the quadrupole transition to that of the dipole transition as defined by Biedenharn and Rose.<sup>24</sup>

#### (b) The 0.20 Mev $\gamma$ —1.03 Mev $\gamma$ Directional Correlation

Since the experimental result was the same as those of 1.03 Mev  $\gamma$ —1.18 Mev  $\gamma$  and 0.20 Mev  $\gamma$ —1.18 Mev  $\gamma$  which will be shown below, it was not necessary to take into account the contribution arising from the coincidence of Compton pulses belonging to the 1.18-Mev gamma ray.

After corrections for the finite angular resolution of detectors, we obtain,

$$A_2 = +0.101 \pm 0.002, \quad A_4 = +0.007 \pm 0.001$$

This also gives the spin sequence  $I+4(2)I+2(2)I$  and therefore it turns out that 0.20- and 1.03-Mev cascade is  $6(2)4(2)2$  or  $4(2,1)4(2)2$  with  $\delta = -0.26$ .

<sup>24</sup> L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. 25, 729 (1953).

#### (c) The 0.20 Mev $\gamma$ —1.18 Mev $\gamma$ Directional Correlation

For the same reason as that given for the 0.20 Mev  $\gamma$ —1.03 Mev  $\gamma$  directional correlation, we need not take into account the contribution from the coincidence of Compton pulses of the 1.03-Mev gamma ray.

Correcting for the finite angular resolution, we obtain

$$A_2 = +0.105 \pm 0.002, \quad A_4 = +0.010 \pm 0.001.$$

This is also the typical form of a pure quadrupole-quadrupole transition of the 4-2-0 type. On the other hand in the case of  $4(2)2(2,1)2(2)0$  sequence with  $\delta = -0.18$ , the expected values are  $A_2 = +0.0482$  and  $A_4 = -0.0057$  as shown in Fig. 6. Hence the spin sequence is found to be  $6(2)4(2)2(2)0$  which is consistent with the measurements on the 1.03 Mev  $\gamma$ —1.18 Mev  $\gamma$  direction polarization correlation and the conversion electrons, as will be described later.

#### (d) The 0.09 Mev $\gamma$ —0.20 Mev $\gamma$ Directional Correlation

Since the mean life of the 2.41-Mev state is  $8.73 \times 10^{-9}$  sec, we have to consider the possibility of a perturbation of the intermediate state by the extranuclear field. Hence, the correlation measurements were performed, using the two kinds of source; dilute solution of  $\text{SbCl}_3$  and electroplated metallic Sb. The total net coincidence counts were  $3 \times 10^5$  for the former source and  $1.5 \times 10^5$  for the latter.

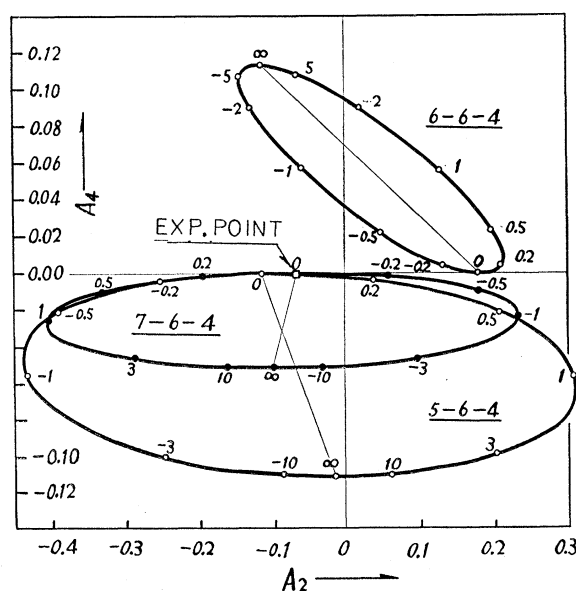


FIG. 8.  $A_2$ — $A_4$  elliptical representations of  $\gamma$ — $\gamma$  directional correlation as a function of the mixing ratio  $\delta$  for the spin sequences 7-6-4, 6-6-4 and 5-6-4. The points on the curves are labelled with the values of  $\delta$ . The open square shows the experimental result for the 0.09 Mev  $\gamma$ —0.20 Mev  $\gamma$  directional correlation.

Through the analysis of the gamma-ray spectra coincident with the 0.09- and 0.20-Mev gamma rays at  $90^\circ$ , the Compton coincidences belonging to the 1.03- and 1.18-Mev gamma rays were estimated ( $\sim 10\%$ ). In order to correct properly for other angles, it was necessary to use the correlation functions described earlier. By subtracting suitably the contribution from the Compton tails of the 1.03- and 1.18-Mev gamma rays and making the corrections for the finite detectors and source dimension,<sup>25,26</sup> we obtain

$$\begin{aligned} A_2 &= -0.0685 \pm 0.0007, & A_4 &= -0.0144 \pm 0.0004, \\ & & & \text{for liquid source;} \\ A_2 &= -0.0707 \pm 0.0012, & A_4 &= +0.0055 \pm 0.0005, \\ & & & \text{for metallic source.} \end{aligned}$$

Since the  $A_2$  values agree with each other within five percent, it seems to be certain that there is no appreciable perturbation effect of the intermediate state by an extranuclear field. Figure 7 represents the experimental results. The weighted mean values from the two sources are

$$A_2 = -0.0692 \pm 0.0020, \quad A_4 = -0.0062 \pm 0.0010.$$

The  $A_2$ — $A_4$  elliptical representations of the directional correlation for the cascades of  $7(2,1)6(2)4$ ,  $6(2,1)6(2)4$  and  $5(2,1)6(2)4$  are shown in Fig. 8. Experimental results fit the spin sequence 7-6-4 with  $\delta = -0.0036$  or 5-6-4 with  $\delta = 0.06$ , eliminating the possibility of 6-6-4. The values of  $\delta$  for these assignments seem to be too small to be decided by measuring the internal conversion coefficient of the 0.09-Mev radiation. The absence of the 0.29-Mev cross-over transition, however, seems to suggest that the former assignment is reasonable.

#### (e) The 1.03 Mev $\gamma$ —1.18 Mev $\gamma$ Direction and Polarization Correlation

A gamma-ray polarimeter of Kyoto Prefectural University was used.<sup>27</sup> Measurements for the 1.03 Mev—1.18 Mev gamma rays of  $\text{Sn}^{120}$  and the 0.89 Mev—1.12 Mev cascade gamma rays of  $\text{Ti}^{46}$  whose spin sequence was well established as  $4+(E2)2+(E2)0+$ , were performed under the same arrangements of the apparatus. The results are represented in Table III.

TABLE III. Polarization-direction correlations for  $\text{Ti}^{46}$  and  $\text{Sn}^{120}$ .

| Nucleus           | Cascade           | $N_{11}/N_1$ ( $\theta=90^\circ$ ) |
|-------------------|-------------------|------------------------------------|
| $\text{Sn}^{120}$ | 1.03 Mev—1.18 Mev | $0.92 \pm 0.03$                    |
| $\text{Ti}^{46}$  | 0.89 Mev—1.12 Mev | $0.92 \pm 0.02$                    |

<sup>25</sup> F. Gimmi, E. Heer, and P. Scherrer, *Helv. Phys. Acta* **29**, 147 (1956).

<sup>26</sup> A. M. Feingold and S. Frankel, *Phys. Rev.* **97**, 1025 (1955).

<sup>27</sup> M. Kawamura, *J. Phys. Soc. (Japan)* **15**, 3 (1960); M. Kawamura, A. Aoki, T. Hayashi, and H. Ikegami, *J. Phys. Soc. (Japan)* (to be published); F. Metzger and M. Deutsch, *Phys. Rev.* **78**, 551 (1950).

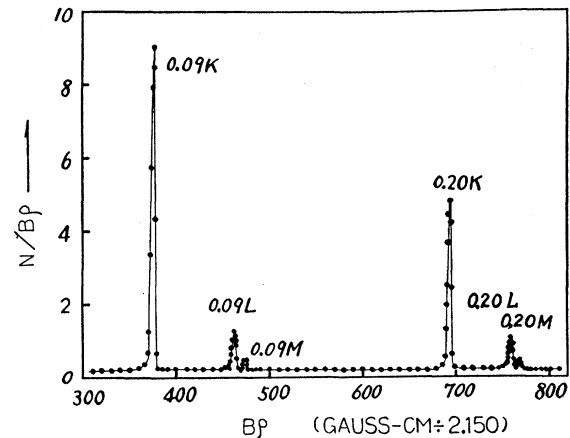


FIG. 9. Conversion electron spectra of 0.09- and 0.20-Mev transitions following 5.8-day  $\text{Sb}^{120m}$  decay. The momentum resolution is 0.5%.

As the expected direction polarization correlation of the  $2+(E2,M1)2+(E2)0+$  sequence with  $\delta = -0.18$  is  $N_{11}/N_1(\theta=90^\circ) = 0.81$ , the spin sequence  $4+(E2)2+(E2)0+$  is determined uniquely and is consistent with the results of the gamma-gamma directional correlation measurements described earlier.

#### 6. SPECTRA OF CONVERSION ELECTRONS

Extremely thin sources of metallic  $\text{Sb}^{120m}$  were prepared electrochemically by the method described in Sec. 2.

The measurements were performed with the INS-III spectrometer using the GM counters whose mica windows were 1.0 and 0.5 mg/cm<sup>2</sup> in thickness. With a  $3\text{-mm} \times 20\text{-mm}$  source and a 3-mm wide detector slit a momentum resolution of 0.5% was obtained, which corresponded to a transmission of 0.2%.

The spectra of the conversion electrons of the lower and the higher energy radiations are represented in Fig. 9 and Fig. 10, respectively.

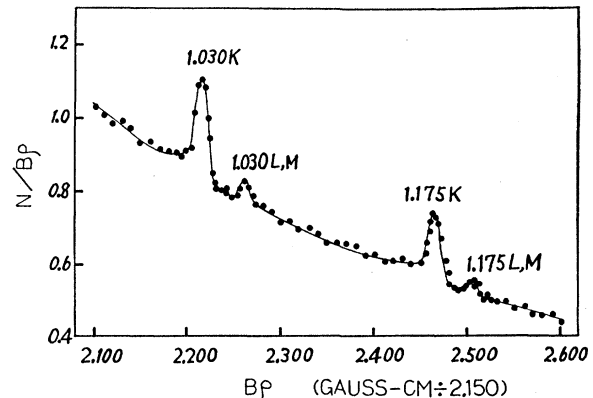


FIG. 10. Electron spectra of the 1.03- and 1.18-Mev transitions. The data are corrected for the decay of  $\text{Sb}^{120m}$ .

TABLE IV. Multipole assignments from the measurements of conversion electrons. The number in parentheses indicates the power of 10 by which the preceding figure should be multiplied. The theoretical internal conversion coefficients are from Rose. The experimental internal conversion coefficients are derived from the relative intensities of the conversion electrons assuming  $\alpha_2^K = 8.85 \times 10^{-4}$  for the 1.18-Mev transition. Some of these assignments are also confirmed by the measurements of  $\text{Ke}^- - \gamma$  and  $\gamma - \text{Ke}^-$  directional correlations (see, Fig. 13).

| $E_\gamma$<br>(Mev) | Expt. $\alpha$       | Theoretical internal conversion coefficient |             |             |             | Assignment |
|---------------------|----------------------|---|-------------|-------------|-------------|------------|
|                     |                      | $E1$  | $E2$        | $M1$        | $M2$        |            |
| 0.09K               | $2.06 \pm 0.10 (-1)$ | $2.10 (-1)$                                 | $1.62 (0)$  | $6.70 (-1)$ | $7.2 (0)$   | $E1$       |
| L                   | $2.5 \pm 0.15 (-2)$  | $2.65 (-2)$                                 | $5.5 (-1)$  | $8.0 (-2)$  | $1.4 (0)$   |            |
| M                   | $5.3 \pm 0.3 (-3)$   | $5.2 (-3)$                                  | $1.3 (-1)$  | $1.7 (-2)$  | $2.9 (-1)$  |            |
| 0.20K               | $1.08 \pm 0.05 (-1)$ | $2.25 (-2)$                                 | $1.11 (-1)$ | $7.10 (-2)$ | $4.15 (-1)$ | $E2$       |
| L                   | $2.1 \pm 0.1 (-2)$   | $2.7 (-3)$                                  | $2.0 (-2)$  | $8.8 (-3)$  | $6.5 (-2)$  |            |
| M                   | $4.1 \pm 0.3 (-3)$   | $5.2 (-4)$                                  | $4.5 (-3)$  | $1.7 (-3)$  | $1.3 (-2)$  |            |
| 1.03K               | $9.6 \pm 1.0 (-4)$   | $4.50 (-4)$                                 | $1.02 (-3)$ | $1.28 (-3)$ | $3.03 (-3)$ | $E2$       |
| L+M                 | $1.6 \pm 0.2 (-4)$   | $6.1 (-5)$                                  | $1.5 (-4)$  | $1.8 (-4)$  | $4.5 (-4)$  |            |
| 1.18K               | ...                  | $3.52 (-4)$                                 | $7.80 (-4)$ | $9.50 (-4)$ | $2.18 (-3)$ | $E2$       |
| L+M                 | $1.3 \pm 0.2 (-4)$   | $4.8 (-5)$                                  | $1.1 (-4)$  | $1.3 (-4)$  | $3.2 (-4)$  |            |

The conversion electrons belonging to the 0.29-Mev cross-over transition were not observed and therefore the branching ratio of this transition was estimated as less than 0.03%. This is consistent with the results from the gamma-ray spectra.

The experimental values for the internal conversion coefficients were derived from the relative intensities of the conversion electrons assuming  $\alpha_2^K = 8.85 \times 10^{-4}$  for the 1.18-Mev transition which was chosen as a reference line. Actually, there is evidence that other electron capture processes might feed the 2.41-Mev state. However, these feedings are estimated to be only two percent or less and therefore cannot affect the conversion coefficient by more than a few percent (see, Table I).

The experimental values of conversion coefficients of the gamma rays from the decay of 5.8-day  $\text{Sb}^{120m}$  are compared with the theoretical values of  $\alpha^{K,L,M}$  obtained by Rose<sup>28</sup> under the assumption of finite nuclear size, in Table IV. In the case of  $Z=50$  and  $0.3 < k < 3.0$ , where  $k$  is the energy of the conversion electron in unit of  $mc^2$ , the internal K-conversion coefficients calculated by assuming either a finite nuclear size<sup>28,29</sup> or a point nucleus<sup>30</sup> do not differ from each other by more than 1.5% even for the magnetic dipole transitions in which the effects of finite nuclear size are most important.

These assignments are all in good agreement with those by gamma-gamma directional correlation meas-

urements. For the 0.09-Mev transition, one might expect the nuclear structure effect of internal conversion process because of its highly forbidden property.<sup>7</sup> A striking anomalous effect, recently reported in the trans-lead odd- $A$  nuclei,<sup>31</sup> was, however, not found in our case.

#### 7. DIRECTIONAL CORRELATIONS INVOLVING K-CONVERSION ELECTRON

As described in Sec. 5(d), it was difficult to decide whether the spin and parity assignment of the 2.50-Mev level is  $7^-$  or  $5^-$  by the internal conversion coefficient of the 0.09-Mev transition. Since the directional correlation of the conversion electron depends on the spin of the nuclear levels, and on the parity change and multipole order mixing of the converted transition, and since in general the dependence of the correlation on the multipole order mixing is different from that of the gamma-gamma directional correlation, the 0.09 Mev  $\text{Ke}^-$ —0.20 Mev  $\gamma$  directional correlation is suitable to decide the spin and parity of 2.50-Mev level.

The preparation of the source is the crucial point in the directional correlation experiments involving low-energy conversion electron or beta particles. In order to prevent electron scattering, extremely thin backing and thin source should be used. Furthermore, the active material should be embedded in a metallic lattice. The latter condition is imperative in this case where the nuclear transitions occur through an electron capture or internal conversion. The electronic shell will be excited by these processes, and the interaction between the nuclear magnetic moment and the excited electronic shell will give rise to a time-dependent perturbation. In a metallic surrounding, however, the recovery time is much shorter ( $< 10^{-12}$  sec) than the mean lifetime of the nuclear intermediate state and consequently there will be no attenuation due to the electron capture or the succeeding electron conversion.

A  $2\text{-}\mu\text{g}/\text{cm}^2$  metallic  $\text{Sb}^{120m}$  source on an extremely

<sup>28</sup> *Internal Conversion Coefficients*, edited by M. E. Rose (North-Holland Publishing Company, Amsterdam, 1958). The  $M$ -shell coefficients refer to a point nucleus and unscreened electrons and should be used with empirical determinations of screening factors. Therefore we have taken the half values of Rose's  $M$ -shell coefficient. M. E. Rose (private communication to Y. Yoshizawa); C. L. McGinnis (see reference 9).

<sup>29</sup> L. Sliv and I. Band, Leningrad Physico-Technical Institute Report 1956 [translation: Report 57 ICCK1, Physics Department, University of Illinois (unpublished)].

<sup>30</sup>  $K$  and  $L$  coefficients for a point nucleus including the effects of screening are given by M. E. Rose, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Appendix IV. See also, M. E. Rose, G. H. Goertzel, B. I. Spinrad, J. Harr, and P. Strong, Phys. Rev. 83, 79 (1951).

<sup>31</sup> F. Asaro, F. S. Stephens, J. M. Hollander, and I. Perlman, Phys. Rev. 117, 492 (1960).



thin backing was prepared by the method described in Sec. 2. The distortion of the line shapes was not recognized even at energy of about 20 keV and the  $KLL$ ,  $KLX$ , and  $KXY$  peaks of the  $K$ -Auger electron group were resolved perfectly. The normal of the source face was oriented at an angle of  $45^\circ$  against the direction of electron observation in the manner described by Gimmi *et al.*<sup>25</sup> The source chamber was made of 2-mm thick Lucite wall. The INS-I spectrometer was set so as to obtain the resolution and the transmission of 2% and 0.3%, respectively.<sup>13,14</sup> The instrumental assembly is shown in Fig. 11.

The electronic devices were almost the same as those used in the measurements of the gamma-gamma directional correlations. Since the attenuation of the directional correlation due to the extranuclear field does not depend on the type of radiation,<sup>32</sup> three directional correlations, 0.09 MeV  $\gamma$ —0.20 MeV  $\text{Ke}^-$ , 0.09 MeV  $\text{Ke}^-$ —0.20 MeV  $\gamma$ , and 0.09 MeV  $\gamma$ —0.20 MeV  $\gamma$ , were measured using the same source, in order to eliminate possible attenuation effects. The experimental results corrected for accidental coincidences ( $<10\%$ ) and the variation of single counting rates are represented in Fig. 12.

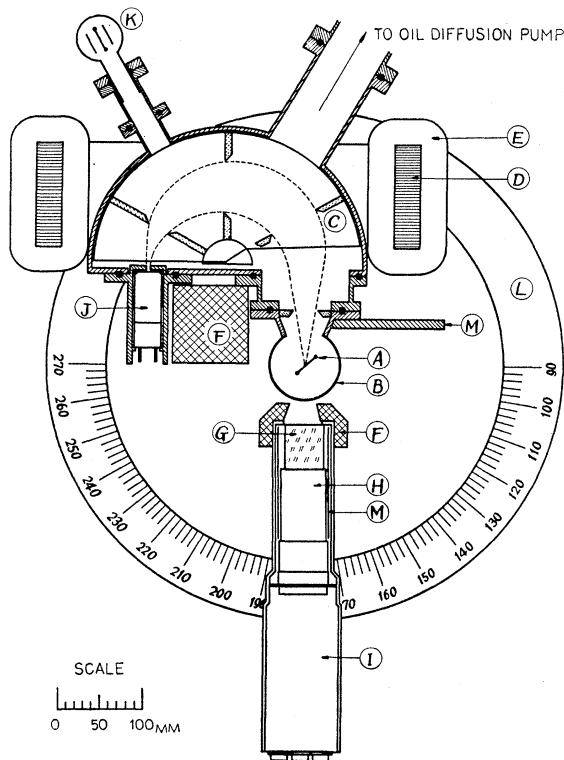


FIG. 11. Instrumental arrangements for the measurements of 0.09-MeV  $\text{Ke}^-$ —0.20-MeV  $\gamma$  and 0.09-MeV  $\gamma$ —0.20-MeV  $\text{Ke}^-$  directional correlations. (A) Source holder; (B) source chamber; (C) aluminum baffle; (D) magnet yoke; (E) coil; (F) lead-shield; (G) NaI(Tl) scintillator; (H) RCA 6342 photomultiplier; (I) pre-amplifier; (J) G-M counter; (K) vacuum gauge; (L) goniometer; (M) magnetic shield.

<sup>32</sup> H. Frauenfelder, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. XIX; R. M. Steffen, in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1955), Vol. 4, p. 293; A. Abragam and R. V. Pound, *Phys. Rev.* **92**, 943 (1953). See also references 24 and 25.

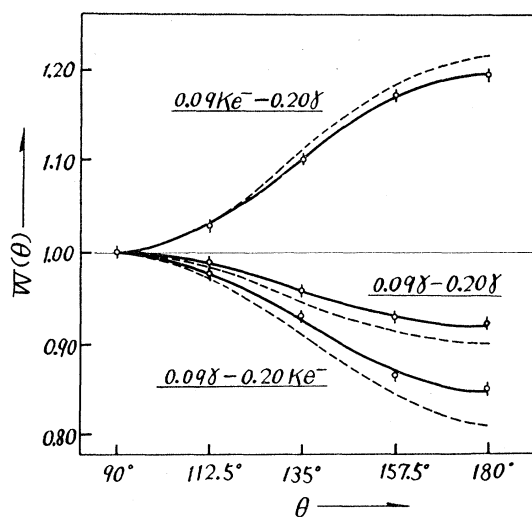


FIG. 12. Experimental directional correlation functions for the 0.09-MeV  $\text{Ke}^-$ —0.20 MeV  $\gamma$ , 0.09 MeV  $\gamma$ —0.20 MeV  $\text{Ke}^-$ . The 0.09 MeV  $\gamma$ —0.20-MeV  $\gamma$  directional correlation is also shown for comparison. The solid lines are the least squares fits drawn through the experimental points. The broken lines represent the experimental directional correlation functions after making corrections as described in Fig. 7.

MeV  $\gamma$ , were measured using the same source, in order to eliminate possible attenuation effects. The experimental results corrected for accidental coincidences ( $<10\%$ ) and the variation of single counting rates are represented in Fig. 12.

#### (a) The 0.09 MeV $\gamma$ —0.20 MeV $\text{Ke}^-$ Directional Correlation

The total net coincidence counts were about  $1 \times 10^5$ . By subtracting the contribution from Compton tails of the 1.03- and 1.18-MeV gamma rays in the same way as described in Sec. 5(d) and correcting for the finite sizes of the source<sup>25</sup> and the electron and gamma-ray spectrometers,<sup>26,33</sup> we obtain

$$A_2(\gamma - \text{Ke}^-) = -0.130 \pm 0.002,$$

$$A_4(\gamma - \text{Ke}^-) = -0.010 \pm 0.001.$$

On the other hand, the point nucleus theory of Biedenharn and Rose predicts<sup>24</sup>

$$A_2(\gamma - \text{Ke}^-) = -0.1282 \pm 0.0015,$$

$$A_4(\gamma - \text{Ke}^-) = 0.007 \pm 0.001, \quad \text{for } 0.20 \text{ MeV } E2;$$

and

$$A_2(\gamma - \text{Ke}^-) = -0.0869 \pm 0.0015,$$

$$A_4(\gamma - \text{Ke}^-) = -0.002 \pm 0.001, \quad \text{for } 0.20 \text{ MeV } M2.$$

The value of  $A_2$  for the 0.20-MeV  $E2$  transition is in good agreement with the experimental value. There-

<sup>33</sup> The correction for the finite angular resolution of the electron spectrometer was performed by a method given by Feingold and Frankel (see references 26). No correction for source scattering has, however, been applied.

fore, it turns out that the assumption of the point nucleus is now applicable to the measurement of the directional correlation involving  $K$ -conversion electrons without serious discrepancies in the case of  $Z=50$ .

(b) The 0.09 Mev  $\text{Ke}^-$ —0.20 Mev  $\gamma$   
Directional Correlation

To investigate the scattering effect of low-energy electrons in the source, the correlations were measured with two very thin sources of 2 and 0.5  $\mu\text{g}/\text{cm}^2$  thickness; and the same results were obtained within experimental errors. The attenuation effect due to the finite thickness of the source backing as reported by the Zürich group<sup>25</sup> was not found, probably owing to the rather good momentum resolution of our beta-ray spectrometer. The total net coincidence counts were about  $1 \times 10^5$ .

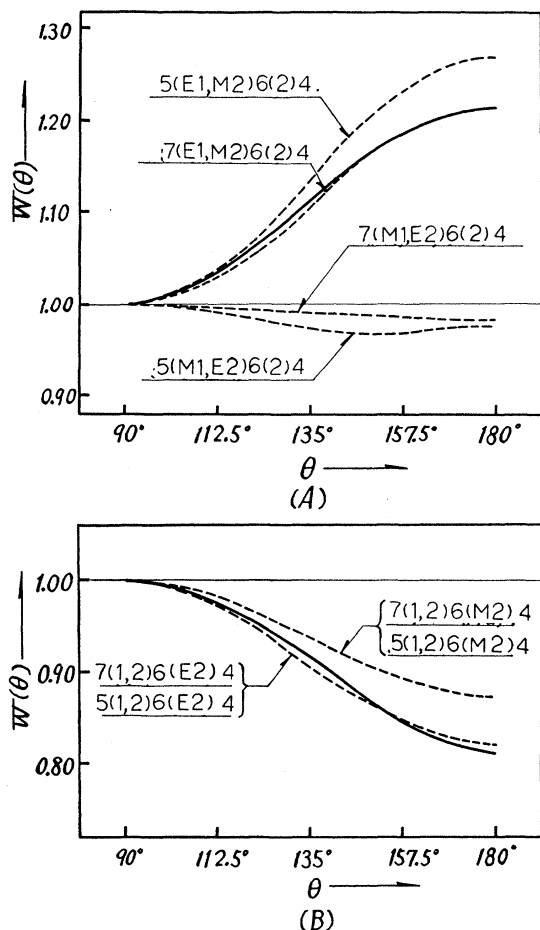


FIG. 13. Comparison of the experimental and theoretical directional correlation functions for the 0.09 Mev—0.20 Mev cascade radiations in  $\text{Sn}^{120}$ . The solid lines represent the corrected experimental directional correlation functions. The broken curves are theoretical functions for the cascades 7-6-4 and 5-6-4. (A) 0.09 Mev  $\text{Ke}^-$ —0.20 Mev  $\gamma$  directional correlation; (B) 0.09 Mev  $\gamma$ —0.20 Mev  $\text{Ke}^-$  directional correlation.

After making all the corrections for the finite sizes of source and detectors, we get

$$A_2(\text{Ke}^- - \gamma) = \pm 0.134 \pm 0.001,$$

$$A_4(\text{Ke}^- - \gamma) = -0.006 \pm 0.001.$$

As was shown earlier, the 0.20-, 1.03-, and 1.18-Mev cascade has the spin sequence  $6(E2)4(E2)2(E2)0$  and therefore the 0.09 Mev  $\text{Ke}^-$ —(1.03 Mev + 1.18 Mev)  $\gamma$  directional correlation function is expected to be the same as that of 0.09 Mev  $\text{Ke}^-$ —0.20 Mev  $\gamma$ . In view of this fact, it is not necessary to subtract the contribution arising from the coincidences with Compton pulses belonging to the 1.03- and 1.18-Mev gamma rays. The expected directional correlation function from the theory for the  $E1+M2$  mixed transition is

$$A_2(\text{Ke}^- - \gamma) = (b_2^e A_2^e + 2\delta_K b A_2 + \delta_K^2 b_2^m A_2^m) / (1 + \delta_K^2),$$

$$A_4(\text{Ke}^- - \gamma) = \delta_K^2 b_4^m A_4^m / (1 + \delta_K^2).$$

The coefficients  $A_2^e$ ,  $A_2$ ,  $A_2^m$ ,  $A_4^m$ ,  $b_2^e$ ,  $b_2^m$ , and  $b_4^m$  are the coefficients tabulated by Biedenharn and Rose.<sup>24,24</sup>  $\delta_K^2$  is the ratio of the ejection rate of  $M2$   $K$ -conversion electrons to that of  $E1$   $K$ -conversion electrons, i.e.,  $\delta_K = (\beta_2^K / \alpha_1^K)^{1/2} \cdot \delta$  where  $\beta_2^K$  and  $\alpha_1^K$  are the  $M2$  and  $E1$   $K$ -internal conversion coefficients, respectively.  $b$  is the particle parameter for a mixed  $E1+M2$  transition defined in the same way as that for a  $E_{L+1}+M_L$  mixture,<sup>24</sup> and its numerical values for a point nucleus are tabulated in the Appendix.

With the  $\delta$  value expected from the experimental results on the 0.09 Mev  $\gamma$ —0.20 Mev  $\gamma$  directional correlation, the coefficients  $A_2$  and  $A_4$  for the spin sequence  $7(E1, M2)6(2)4$  and  $5(E1, M2)6(2)4$  are calculated as

$$A_2(\text{Ke}^- - \gamma) = +0.135 \pm 0.001,$$

$$A_4(\text{Ke}^- - \gamma) = -0.006 \pm 0.001 \quad \text{for } 7(E1, M2)6(2)4;$$

$$A_2(\text{Ke}^- - \gamma) = +0.165 \pm 0.001,$$

$$A_4(\text{Ke}^- - \gamma) = -0.001 \pm 0.001 \quad \text{for } 5(E1, M2)6(2)4.$$

Figure 13 represents the experimental and the theoretical directional correlations of  $\text{Ke}^- - \gamma$  and  $\gamma - \text{Ke}^-$  for the 0.09- and 0.20-Mev transitions. The solid lines represent the pure experimental directional correlation functions corrected as described earlier. The broken curves are the theoretical functions for the cascades 7-6-4 and 5-6-4. From the figure, it is found that the sequence  $5-(E1, M2)6+(2)4+$  is ruled out.

#### 8. POSSIBILITY OF THE EXISTENCE OF A MULTIPLET IN THE SECOND EXCITED STATE

It is very interesting to see whether there will be a splitting of the second excited state owing to a surface vibration in even- $A$  Sn isotopes as found in Cd isotopes.<sup>6</sup>

<sup>24</sup> The notations used in the present paper differ somewhat from those used in reference 24, but in most cases the differences are obvious.

By measuring the decay of the 16-min  $\text{Sb}^{120}$ , we have tried to investigate whether there is such a degeneracy of the second level. The sources were prepared as described in Sec. 2.

The decay of 16-min  $\text{Sb}^{120}$  was traced for about 1–2 hours using a  $1\frac{3}{4}$ -in. diameter and 2-in. long NaI(Tl) crystal optically coupled to a DuMont 6292 photomultiplier and a summing gamma-ray spectrometer with a 3-in. diameter and 3-in. long NaI(Tl) crystal coupled to a DuMont 6363 photomultiplier. The data obtained were, however, somewhat uncertain because of the gain instability of the photomultipliers, due to the variation of the counting rate<sup>35</sup> and the background ascribable to the existence of 5.8-day  $\text{Sb}^{120m}$ .

From the intensity ratio of the 1.18 Mev and annihilation gamma rays, we obtain the branching ratio of the decay to the 1.18-Mev state and the decay to the ground state, which agrees well with McGinnis's result.<sup>9</sup> From the comparison of the summing spectrum of  $\text{Sn}^{120}$  with that of  $\text{Co}^{60}$ , the transition probability to a possible level of about 2.21 Mev if it exists, is smaller than that to the 1.18-Mev level by a factor of about 20 or more and therefore, the  $\log ft$  value is more than 6.5. On the other hand, for some medium weight nuclei whose level sequences are  $0+$ ,  $2+$ ,  $2+$ , the  $\log ft$  values of transitions from  $1+$  to the second excited level are 6.6–5.3.<sup>36</sup> Consequently, there appears to be no degeneracy; however more accurate measurements such as those about the external and the internal conversion electron spectra are necessary, for a definite conclusion to be obtained.

## 9. SUMMARY AND DISCUSSION

Measurements of the spectra of coincidence gamma-rays, conversion electrons, and  $\gamma$ – $\gamma$ ,  $\text{Ke}^-$ – $\gamma$ , and  $\gamma$ – $\text{Ke}^-$  directional correlations and lifetimes of the 2.41- and 2.50-Mev levels of 5.8-day  $\text{Sb}^{120m}$  were performed. The level sequence previously proposed by McGinnis has been confirmed. All the gamma rays except the 0.09-Mev gamma ray, which is a mixture of  $E1$  and  $M2$  with  $\delta = -0.0036$ , are found to be pure electric quadrupole transitions. According to these results, we can expect the low-lying states of  $\text{Sn}^{120}$  to be  $0+$ ,  $2+$ ,  $4+$ ,  $6+$ , and  $7-$  which can be represented by the neutron configurations  $(2d_{3/2})^4$ ,  $(2d_{3/2})^2(1h_{11/2})^2$ , and  $(2d_{3/2})^3(1h_{11/2})^1$ .<sup>37</sup>

The fact that the favored factors of the transitions of the 1.18-, 2.41-, and probably 2.21-Mev states are between 3 and 4.5 seems to prove that these transitions are the neutron jumps between the states characterized by the  $(2d_{3/2})^4$  and  $(2d_{3/2})^2(1h_{11/2})^2$  configurations, as-

suming that the effective charge of the neutron in nuclei is of the same order of magnitude as that of the proton.<sup>38</sup> Therefore if the 2.50-Mev state is also a neutron excitation level, the 0.09-Mev  $E1$  transition whose favored factor is about  $5 \times 10^{-8}$  seems to be a simultaneous  $j$ - and  $l$ -forbidden transition<sup>39</sup> resulting from the change of the configuration from  $(2d_{3/2})^3(1h_{11/2})^1$  with a small amount of impurity to  $(2d_{3/2})^2(1h_{11/2})^2$ . The forbidden 0.09-Mev  $M2$  transition and the absence of a cross-over  $E3$  transition, which is only  $j$  forbidden, can both be accounted for in this manner. However, no example of  $j$ - simultaneous and  $l$ -forbidden electric dipole de-excitation has, so far been found in even-even nuclei. For such a highly forbidden transition one might expect the nuclear structure effect in the internal conversion process. However, the experimental results for the 0.09-Mev  $\text{Ke}^-$  transition are found to agree within experimental errors with the theoretical prediction assuming a point nucleus.

The feeding to the 2.21-Mev level of  $\text{Sn}^{120}$  from the 16-min  $\text{Sb}^{120}$  ( $1+$ ) is smaller than that to the 1.18-Mev level by a factor of about 20 or more and there seems to be no degeneracy of the second excited state of  $\text{Sn}^{120}$ . However, more accurate measurements are required for a more definite conclusion.

## ACKNOWLEDGMENTS

The author wishes to express his gratitude to Dr. M. Sakai, Dr. Y. Ishizaki, Mr. T. Yamazaki of I.N.S. and Dr. M. Kawamura of Kyoto Prefectural University for their valuable discussions and assistance; and to Dr. K. Saito of I.N.S. and Dr. N. Suzuki of Tohoku University for their cooperation in chemical separations of many radioisotopes. He is indebted to the cyclotron crew of the Institute for the operation of the machine throughout the experiment. He is also grateful to Professor M. Sonoda of Kyusyu University for his continuous encouragement and discussions.

## APPENDIX. PARTICLE PARAMETER FOR A $E1+M2$ MIXED DIRECTIONAL CORRELATION

In general, the matrix element for the  $M2$  gamma transition is very small compared to that of the  $E1$  gamma transition. However, under certain circumstances, such as  $j$ - and  $l$ - or  $K$ -forbidden transition,<sup>4</sup> the  $E1$  matrix element may be small, while  $M2$  transition may not be prohibited. As a consequence, the effect of the contribution of the  $M2$  matrix element on the directional correlation cannot be ignored.

<sup>38</sup> A. de-Shalit, Phys. Rev. **113**, 547 (1959). The author is indebted to Dr. H. Horie of Tokyo Institute of Technology for his kind communications on this problem.

<sup>39</sup> H. Horie (private communication); S. A. Moszkowski, see reference 21; M. Goldhaber and A. W. Sunyar, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), Chap. XVI (II).

<sup>35</sup> R. D. Conner, Nuclear Instr. **6**, 337 (1960); W. E. Mott and R. B. Sutton, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin-Göttingen-Heidelberg, 1958), Vol. 45.

<sup>36</sup> M. Sakai (private communication).

<sup>37</sup> S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Math.-fys. Medd. **29**, No. 16 (1955).

TABLE V. Mixed conversion particle parameter  $b$  for  $E1+M2$  mixture.

| $Z \backslash k$ | 0.3     | 0.5     | 1.0     | 1.8     | 3.0     | 5.0     |
|------------------|---------|---------|---------|---------|---------|---------|
| 30               | -0.2069 | -0.3293 | -0.5550 | -0.7463 | -0.8649 | -0.9336 |
| 40               | -0.2032 | -0.3237 | -0.5410 | -0.7268 | -0.8431 | -0.9159 |
| 54               | -0.1980 | -0.3146 | -0.5195 | -0.6913 | -0.8043 | -0.8796 |
| 64               | -0.1939 | -0.3089 | -0.5042 | -0.6651 | -0.7739 | -0.8476 |
| 72               | -0.1903 | -0.3045 | -0.4919 | -0.6436 | -0.7467 | -0.8177 |
| 78               | -0.1872 | -0.3015 | -0.4832 | -0.6273 | -0.7254 | -0.7927 |
| 83               |         | -0.2992 | -0.4762 | -0.6139 | -0.7073 | -0.7715 |
| 88               |         | -0.2968 | -0.4695 | -0.6004 | -0.6889 | -0.7489 |
| 92               |         | -0.2949 | -0.4644 | -0.5898 | -0.6739 | -0.7303 |
| 96               |         | -0.2929 | -0.4595 | -0.5793 | -0.6589 | -0.7112 |

Calculations of  $b$  for the  $E1+M2$  mixed transition were performed by the method outlined by Biedenharn and Rose.<sup>24,40,41</sup> As a check of the results, we also calculated the  $b_2^e$  for  $E1$ ,  $b_2^m$  for  $M2$ , and some  $b$  values for a  $E2+M1$  mixture. The results obtained agree well with those of Biedenharn and Rose within 0.1%. Numerical values of  $b$  for the  $E1+M2$  mixed transition are given in Table V.

<sup>40</sup> J. Matsumoto and H. Ikegami (to be published).

<sup>41</sup> The factor  $-i^{2l(\omega)}$  has been omitted from Eq. (82) of reference 24. Recently, the misprint was also corrected by Church *et al.* [E. Church, M. Rose and J. Weneser, Phys. Rev. **109**, 1299 (1958)].

### $L/K$ -Capture Ratio and $E_L/E_K$ for $\text{Ar}^{37\ddagger,*}$

A. G. SANTOS-OCAMPO AND D. C. CONWAY

Department of Chemistry, Purdue University, Lafayette, Indiana

(Received June 27, 1960)

The  $L/K$  capture ratio of  $\text{Ar}^{37}$  has been determined to be  $0.103 \pm 0.003$  in a high-pressure, multiwire proportional counter. This value is in excellent agreement with the theoretical value of 0.100. The average energy of the  $L$  peak was found to be  $273 \pm 6$  ev. When a new value of  $265 \pm 5$  ev is obtained for the critical  $L$  absorption energy of Cl by interpolation and the effect of the Auger process is considered, it is concluded that the energy to produce an ion pair in Ar at 0.2 kev is  $0.98 \pm 0.04$  times the value at 2.8 kev.

#### INTRODUCTION

IF the correlations between the positions of the electrons are neglected, the theoretical  $L/K$  capture ratio for  $\text{Ar}^{37}$  is 0.082.<sup>1</sup> The theoretical ratio is increased to 0.100 when the Pauli correlations are introduced, although the ratio would probably be somewhat larger if all the correlations were considered.<sup>2</sup>

In the relatively low  $Z$  region the  $L/K$ -capture ratio,  $R$ , can be determined in a proportional counter by use of the relation

$$R = R'(1 - P\omega_K) - P\omega_K k, \quad (1)$$

in which  $R'$  is the observed ratio of the two peak areas in the pulse-height spectrum;  $P$ , the probability of a  $K_\alpha$  x ray escaping the counter;  $\omega_K$ , the  $K$ -fluorescence yield; and  $k$ , the fraction of  $K_\alpha$  x rays in the  $K$  series. It is best to perform the experiment with  $P$  equal to zero because of the uncertainties in the values of  $\omega_K$  and  $k$ .<sup>3</sup> Pontecorvo *et al.*<sup>4</sup> obtained an  $L/K$  capture ratio of 0.087 for  $\text{Ar}^{37}$  in a Xe-filled proportional counter

in which  $P$  was 0.13. A value of  $0.092 \pm 0.010$  or  $-0.005$  was obtained by Langevin and Radvanyi<sup>5</sup> in a Xe filled counter in which  $P$  was 0.026. Their error was estimated from the uncertainty introduced in the extrapolation of the  $L$  peak to zero energy. Kiser and Johnston<sup>6</sup> obtained  $0.102 \pm 0.008$ . However,  $P$  varied from 1–0.4, and a longer extrapolation of the  $L$  peak was required. Values of 0.108 ( $\pm 0.016$ ) for the  $\omega_K$ <sup>7</sup> and 1.0 for the  $k$  of Cl were used in Eq. (1). If the new value<sup>8</sup> for  $\omega_K$  of  $0.093 \pm 0.003$  is used, the capture ratio is increased to  $0.116 \pm 0.011$  (standard deviation). In the present investigation a more accurate value for the  $L/K$ -capture ratio has been determined which can be compared to the theoretical value which has been corrected for the Pauli correlations only.

Recently the low-energy region of the tritium spectrum was examined in a proportional counter using  $P$ -10 gas (9 Ar/CH<sub>4</sub>) and found to deviate from theory.<sup>9</sup> It was noted that the deviation could have been caused by an increase in the energy to produce an ion pair,  $w$ , of 4–6% in the energy interval 1.2–0.25 kev. Most of the electron beam investigations have shown  $w$  to be

<sup>†</sup> Supported in part by the U. S. Atomic Energy Commission.

\* Taken in part from the doctoral dissertation of Augusto G. Santos-Ocampo.

<sup>1</sup> H. Brysk and M. E. Rose, Revs. Modern Phys. **30**, 1169 (1959).

<sup>2</sup> S. Odier and R. Daudel, J. phys. radium **17**, 60 (1956).

<sup>3</sup> B. L. Robinson and R. W. Fink, Revs. Modern Phys. **32**, 117 (1960).

<sup>4</sup> B. Pontecorvo, D. H. W. Kirkwood, and G. C. Hanna, Phys. Rev. **75**, 982, 985 (1949).

<sup>5</sup> M. Langevin and P. Radvanyi, Compt. rend. **241**, 33 (1955).

<sup>6</sup> R. W. Kiser and W. H. Johnston, J. Am. Chem. Soc. **81**, 1810 (1959).

<sup>7</sup> M. Haas, Ann. Physik **16**, 473 (1933).

<sup>8</sup> F. Bertrand, G. Charpak, and F. Suzor, J. phys. radium **20**, 956 (1959).

<sup>9</sup> D. C. Conway and W. H. Johnston, Phys. Rev. **116**, 1544 (1959).