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K-shell internal ionization and excitation in β decay of ^{35}S

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Using a flat-crystal x-ray spectrometer designed for low-energy photons from radioactive sources, the *K*-shell internal ionization and excitation (*K*-IIE) in the β decay of ^{35}S has been investigated by measuring chlorine *K* x rays from carefully purified ^{35}S sources. We have succeeded for the first time in obtaining a high-resolution spectrum of the $K\alpha$ x-ray group emitted after the β decay of ^{35}S, which consists of at least six lines, indicating that multiple ionizations and excitations including *L* and *M* shells are dominant in the *K*-IIE process during the β decay of low-*Z* nuclides. The *K*-hole creation probability for ^{35}S determined is $(2.8 \pm 0.5) \times 10^{-3}$ per decay, which agrees well with the previous result by Rubinson and Howland within the standard deviation. The comparison with available theoretical calculations is also given.
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I. INTRODUCTION

One of the atomic processes accompanying β decay is *K*-shell internal ionization and excitation (*K*-IIE); a *K*-shell electron of the parent nuclide goes to the continuum of the daughter in internal ionization, while it goes to an unoccupied higher-bound state in internal excitation. A main mechanism of the *K*-IIE process is so-called electron shake-off and shake-up caused by the sudden change of the Coulomb charge of the nucleus during β decay [1,2]. Since the *K*-IIE process is signified by the emission of *K* x rays of daughter elements, the phenomenon can be examined by the detection of these *K* x rays. A measurable quantity to be compared with theory is the *K*-hole creation probability per decay, P_K, which is of the order of 10^{-3}–10^{-4} depending on the atomic number *Z*.

In 1953, Boehm and Wu first studied the *K*-IIE process during the β decay of ^{147}Pm by observing *K* x rays of the daughter ^{147}Sm [3]. Since their work, the P_K values for more than 20 nuclides have been determined with various methods of x-ray detection. However, there are few P_K values for nuclides with $Z<20$. One of difficulties in measuring the P_K of such low-*Z* nuclides comes from the fact that the *K* x-ray emission in the *K*-IIE process is less intense with decreasing *Z*. According to a rough theoretical prediction [4,5], P_K increases as $1/Z^2$ with decreasing *Z*. The increase in P_K is compromised by the decrease of the *K*-shell fluorescence yield ω_K, i.e., the probability with which the *K* hole is filled through x-ray emission; ω_K decreases more steeply with decreasing *Z*. The other difficulty in the measurement of low-*Z* nuclides is separation between the x rays of the daughter atom and those of the parent atom, which are emitted by the collision of β particles with parent atoms in the β source. The resolution of high-performance semiconductor detectors is not enough to resolve those adjacent x rays in the low-*Z* region.

The P_K measurement for ^{35}S ($Z=16$) was performed by Rubinson and Howland in 1954 [6]. In their measurement by proportional-counter pulse-height analysis, the photon spectra obtained consist of a superposition of Cl and S *K* x rays due to the poor energy resolution of proportional counter. In the measurement with ^{35}S, we are to be careful of the amount of chlorine ions as impurity in ^{35}S sources. First, chlorine ions may be mixed into solutions at any stage of source preparation, because they exist everywhere in the experimental environment. Second, the amount of chlorine ions in the original solution gradually increases as a result of the decay of ^{35}S. There is no remark on the removal of chlorine ions in the work by Rubinson and Howland.

Critical P_K measurements with improved nuclear instrumentation are clearly desired for low-*Z* nuclides. We report here in detail our recent experiment to obtain a reliable P_K value for ^{35}S with a flat-crystal x-ray spectrometer, for which the energy resolution is much better than semiconductor detectors in the low-*Z* region. Technical improvements in the present P_K measurement are the energy resolution in x-ray detection and the almost complete removal of chlorine ions from ^{35}S sources. Furthermore, with the aid of the excellent resolution of the x-ray spectrometer, we have found that there are several components in the Cl $K\alpha$ x rays emitted in the *K*-IIE process during β decay. In Sec. II, the experimental procedure to obtain the high-resolution photon spectra is described. The analysis of experimental data and the result of the P_K probability for ^{35}S are given in Sec. III. The comparison of this result with theoretical calculations is given in Sec. IV. Discussions on the high-resolution spectrum of the Cl $K\alpha$ x-ray group and suggestions for future work are given in Secs. V and VI.

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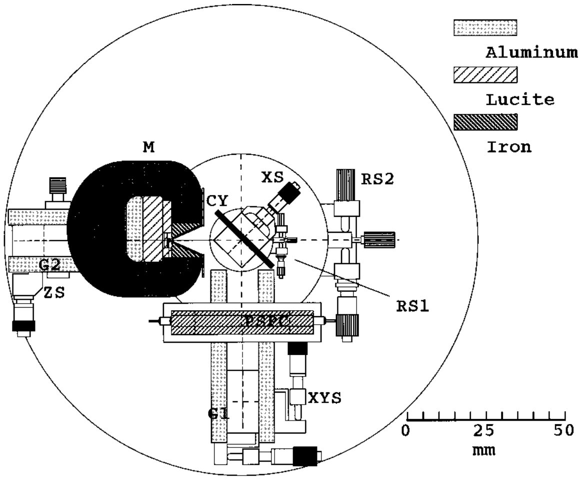

FIG. 1. The experimental layout for the high-resolution x-ray measurement: PSPC, position-sensitive proportional counter; CY, Ge(111) crystal; S, radioactive source; M, magnet; XYS, xy-axis stage; XS, x-axis stage; ZS, z-axis stage; RS1 and RS2, rotative stages; G1 and G2, aluminum guide rails.

II. EXPERIMENTAL PROCEDURE

A. Apparatus

An improved version of the flat-crystal x-ray spectrometer developed in our previous works [7,8] was used for the high-resolution measurements of low-energy photons emitted from radioactive sources ^{35}S and ^{109}Cd. The geometrical arrangement of position-sensitive proportional counter (PSPC), crystal, and source in the spectrometer is schematically shown in Fig. 1. The PSPC has a single anode wire consisting of 7-μm-diam carbon fiber with a resistance of 4 kΩ/cm; the sensitive area is $60\times15\times10\,\mathrm{mm}^3$ and the window is a 6-μm-thick aluminized polypropylene film. The position of incident photons on the PSPC was obtained by the charge-division method, in which the position is deduced as a ratio of the pulse height of an output signal from one end of the resistive anode to the sum of those from both ends of the anode. Position and energy signals from an analog divider (Ortec 464) were stored in the list mode of the data acquisition system (First Comtec MPA/SBB). The performance of the PSPC with a carbon-resistive anode is described in Ref. [9].

The diffraction crystal is Ge(111), of which the size is $50\times20\times5$ mm^3 and the interplanar spacing $2d$ is 6.5327 Å. The PSPC is rotated around the center of crystal in the θ-2θ mode, where the rotation angle of the PSPC is precisely equal to twice that of crystal itself. All electrons from a radioactive source were bent away by the magnetic field ($\sim$1 kG) of a magnet, inside which the source was mounted, as shown in Fig. 1. All metal parts around the source were carefully covered with Lucite to suppress the production of any external radiations. The whole assembly of the present spectrometer was installed in a plastic desiccator, which was then put in an electromagnetic shielding box made of 3-mm-thick copper plates to eliminate electronic noises from surroundings. To obtain the highest x-ray detection efficiency with the present spectrometer, the source-crystal-PSPC arrangement was set to the shortest, i.e., 50 mm for the distance between source and crystal and 45 mm for that of crystal and PSPC. The longest arrangement to achieve the best resolution was 70 mm for both distances.

The counter gas for the PSPC was Ar+30% CH_4. In order to avoid the absorption of low-energy photons in air, hydrogen gas was made to flow through the desiccator at a rate of 5–10 ml/min under a pressure of 1 atm. The pressure in the PSPC was kept a little higher than that of hydrogen gas, i.e., 1.10 atm. Such a small difference between the pressures is necessary to suppress a change in the sensitive thickness of the PSPC by making the counter window (6-mm-thick polypropylene film) distended to the outer side. The fluctuation in the pressure of counter gas was kept below 1% by a handmade pressure controller.

B. Source preparation

Two kinds of radioactive sources, i.e., ^{35}S and ^{109}Cd, have been prepared in the present measurements. The energy of the Ag $L\alpha_{1,2}$ line emitted from the electron-capture decay of ^{109}Cd is 2.984 keV, which is near to that of the Cl $K\alpha_{1,2}$ line, i.e., 2.622 keV. The Ag $L\alpha_{1,2}$ line from a calibrated ^{109}Cd source has been used to determine the absolute intensity of the Cl $K\alpha$ x rays emitted from ^{35}S, as described in Sec. III.

The chemical form of carrier-free ^{35}S purchased from ARC (American Radiolabeled Chemicals) is Na_2SO_4 in aqueous solution; its specific activity is about 3.7 GBq/ml. Chlorine ions in the original solution were removed by the microresin column technique [10,11]; the resin used is Dowex 1-Cl$^-$ and the column size is 0.8 mm diameter $\times$5 mm length. Successive treatments for the purification of ^{35}S were as follows: (1) 5 ml of 2N HNO_3 and then 1 ml of distilled water to change the chemical form of the resin from Dowex 1-Cl$^-$ to Dowex 1-$NO_3{}^-$, (2) 0.1 ml of the original solution to adsorb the β activity on the resin, (3) 5 ml of 0.01N HNO_3 to remove away chlorine ions from the resin, and (4) a few drops of 2N HNO_3 to take up the β activity. More than 95% of the activity was contained in five drops ($\sim$150 μl) at the last step. No visible residue was seen after adding 10 μl of 0.005N NaOH to the drops and drying up them. The addition of NaOH is necessary to avoid the serious contamination in heating the radioactive drops; Na_2SO_4 is much more stable than H_2SO_4 at high temperatures. After the activity was solved in a drop of distilled water with a volume of about 2 μl, the drop was put on a carbon strip of 1 mm width and 20 mm length. The active area on the strip was about 1 mm$\times$3 mm, and the intensity was 370 MBq at the highest. The strip was glued on the 2-mm-thick Lucite plate. The active area was covered with a 13.7-μm-thick polypropylene film; this is helpful to avoid the contamination of the whole part of the x-ray spectrometer inside the vacuum-proof desiccator in the long-running x-ray measurements (45 days at the longest).

The chemical form of ^{109}Cd purchased from NEN (New England Nuclear) is $CdCl_2$ in 0.1N HCl; its specific activity is about 37 GBq/mg. The original solution with the activity of 37 MBq was purified with a Dowex 50W-H$^+$ microresin

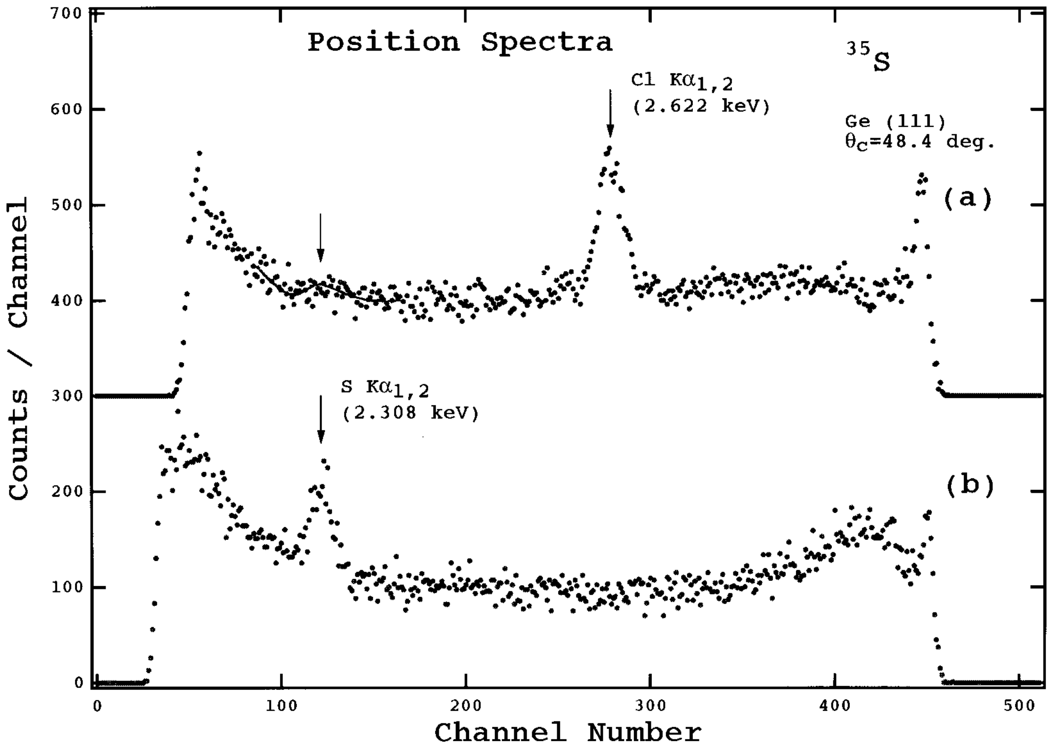

FIG. 2. Position spectra of photons emitted from different ^{35}S sources: (a) A source prepared from the original solution without the chemical purification and (b) a source prepared by adding a small amount of Na_2SO_4 to the original solution.

column (0.8 mm diameter×5 mm length); cadmium ions were eluted with 1N HCl. More than 80% of the activity was taken up in ten drops of the elution (~300 μl). The ^{109}Cd source on the carbon strip was prepared from these drops following the same procedure as that for the ^{35}S source.

C. High-resolution measurements of low-energy photons from ^{35}S and ^{109}Cd sources

In Fig. 2 are given position spectra of low-energy photons emitted from two different ^{35}S sources, which were prepared from the original solution without the chemical purification: (a) from the original solution itself and (b) from the solution admixed by a small amount of Na_2SO_4 (~10 μg/100 μl solution). Energies corresponding to 2.622-keV Cl $K\alpha_{1,2}$ and 2.308-keV S $K\alpha_{1,2}$ x rays are indicated by arrows in the figure. In order to catch both Cl and S *K* x rays in the same spectrum, the diffraction angle θ_c was set to 48.4° with the shortest source-crystal-PSPC arrangement. It is seen that the separation of Cl *K* x rays from S *K* x rays is sufficient even with the shortest arrangement.

There is a faint bump of S $K\ \alpha_{1,2}$ x ray in the position spectrum (a) in Fig. 2, which was caused by the self-absorption of β particles in the carrier-free ^{35}S source. This bump became larger when a more intense ^{35}S source was used in the measurement. No peak of Cl *K* x rays appears in the spectrum (b), indicating that a small amount of sulfur atoms in the source is enough to absorb almost completely those x rays.

Energy spectra observed with the PSPC are given in Fig. 3, of which the blackish region indicates the energy width to construct the position spectra from list-mode acquisition data: (a) with the ^{35}S source and (b) without a source. The large peak in the higher-energy side of the energy spectrum (a) is caused by 9.886-keV Ge *K* x rays, while the small peak in the lower side is caused by 1.188-keV Ge *L* x rays. These Ge *K* and *L* x rays are emitted through the photoelectric effect at the Ge(111) crystal, which is caused by photons coming from the ^{35}S source; they mainly consist of internal bremsstrahlung emitted in the β decay and external bremsstrahlung produced by the collision of β particles with surrounding materials near the source. Note that β particles from the source cannot arrive at the crystal because of the strong magnetic field in front of the source. As shown by the energy spectrum (b) in Fig. 3, the intensity of natural background radiation is relatively small compared with that of Ge x rays. This indicates that the background continuum in position spectra, e.g., (a) and (b) in Fig. 2, is mainly caused by the Ge x rays from the crystal.

The position spectrum of Cl *K* x rays emitted from the purified ^{35}S source is given by Fig. 4(a). The chlorine impurity in the original solution was almost completely removed by the chemical purification in the source preparation. In this measurement, the diffraction angle θ_c was set to 44.5° with the longest source-crystal-PSPC arrangement. The source intensity was about 370 MBq, and the measuring period was 45 days. There appear at least six lines in the spectrum, as indicated by the arrows P1–P6. These lines are hidden by the big peak of Cl $K\alpha_{1,2}$ x rays in spectrum (a) in Fig. 2, which was obtained with an unpurified ^{35}S source. Thus the removal of chlorine ions from the original solution is essen-

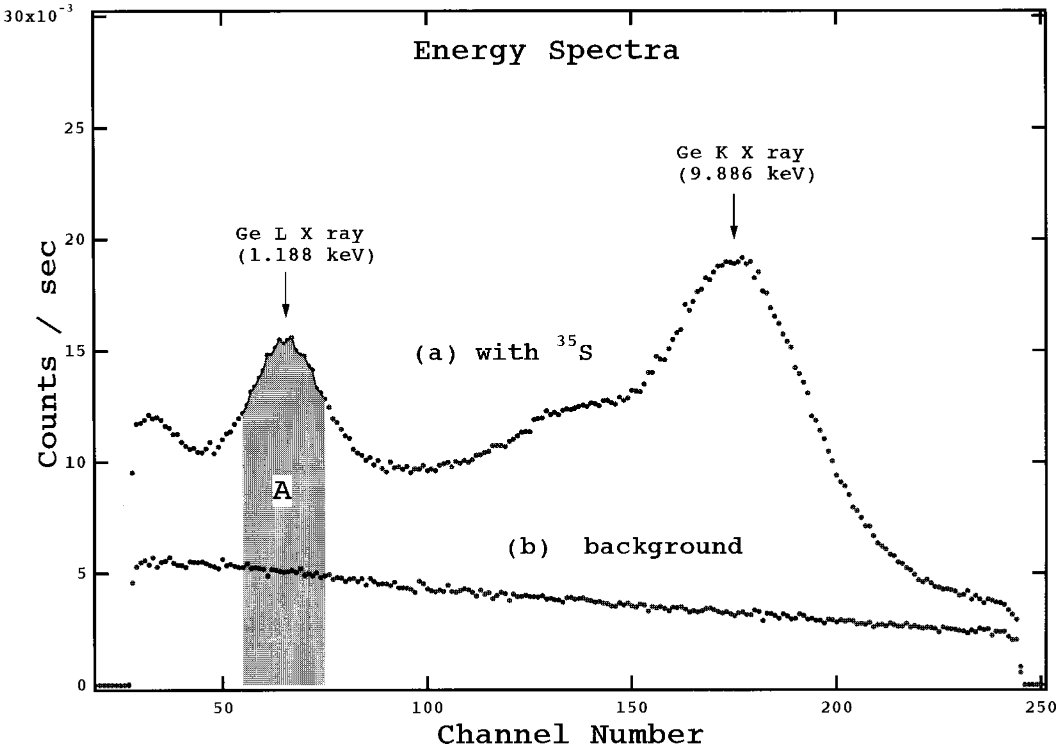

FIG. 3. Energy spectra obtained with the PSPC: (a) with the ^{35}S source and (b) without a source. Both spectra were obtained with the same measuring periods (about 1 week). The linearity in energy scale is not good because of the high voltage applied to the PSPC (1470 V), which was necessary to obtain the better signal-to-noise ratio for the position signal of the PSPC; the maximum voltage to keep the linearity for up to 10-keV photons is about 1200 V.

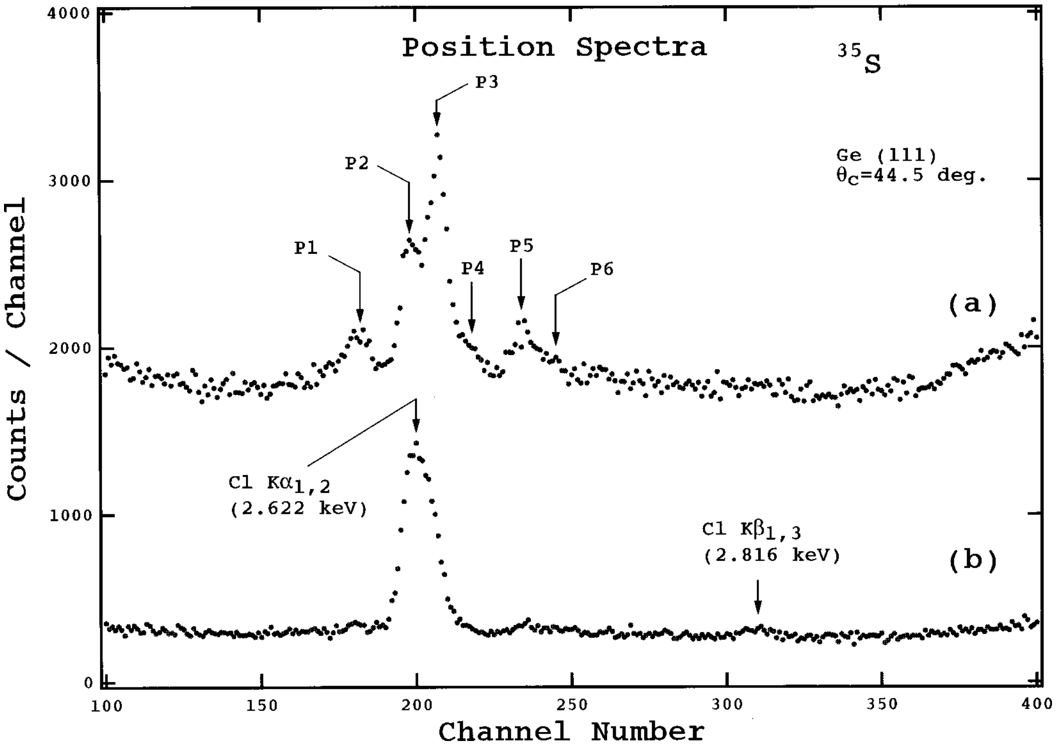

FIG. 4. The best-resolution spectra of Cl K x rays emitted from the purified ^{35}S source: (a) covered with a 13.7-μm-thick polypropylene film and (b) a 20-μm-thick vinylidene chloride film. Arrows P1–P6 in the spectrum (a) indicate the positions of individual lines of the Cl $K\alpha$ x-ray group.

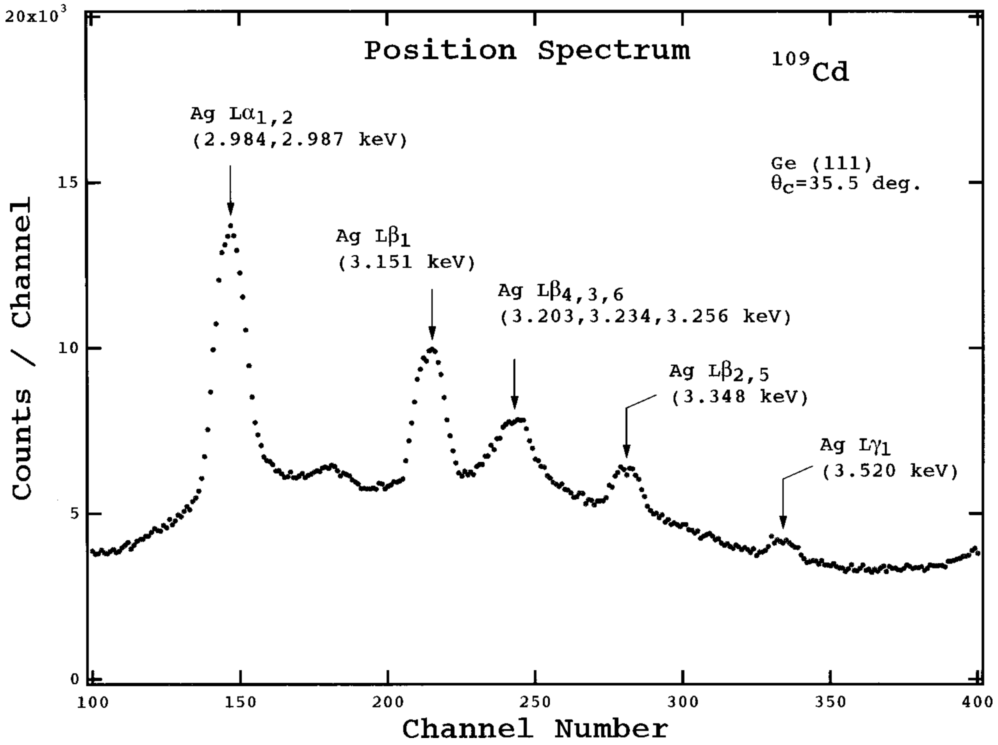

FIG. 5. The position spectra of photons emitted from the purified ^{109}Cd source; the Ag $L\alpha_{1,2}$ line in the Ag L x-ray group was used to calibrate the overall detection efficiency of the PSPC in the flat-crystal x-ray spectrometer.

tially important in the present experiment.

Spectrum (b) in Fig. 4 was obtained by covering the same purified source with a 20.0-μm-thick vinylidene chloride film; the experimental arrangement is precisely the same as that for spectrum (a). Spectrum (b) consists of ordinary Cl $K\alpha_{1,2}$ x rays (2.622 keV) and Cl $K\beta_{1,3}$ x rays (2.816 keV), which are emitted by the collision of β particles with chlorine atoms in the vinylidene chloride film. The energy of one of peaks in the spectrum (a), i.e., P2 line, coincides with that of the Cl $K\alpha_{1,2}$ x rays in spectrum (b).

The position spectra of photons emitted from the purified ^{109}Cd source are shown in Fig. 5. In this measurement, the diffraction angle θ_c was set to 35.5° with the longest source-crystal-PSPC arrangement. A main peak at the left side of the spectrum is caused by the 2.984-keV Ag $L\alpha_{1,2}$ line, which was used to calibrate the detection efficiency of the PSPC for the Cl $K\alpha_{1,2}$ x rays, as explained in Sec. III. The intensities of Ag $L\beta_{3,4,6}$, Ag $L\beta_{2,5}$, and Ag $L\gamma_1$ lines are about one order of magnitude enhanced comparing with those of Ag $L\alpha_{1,2}$ and Ag $L\beta_1$, because of the steep increase of the detection efficiency of the PSPC at the K edge of argon (3.206 keV).

III. DATA ANALYSIS

The K x-ray emission probability per β decay, X_K, is given by

$$X_K=\frac{n_x}{N_\beta}\frac{1}{D_x}. \tag{1}$$

In the above expression, n_x is the number of Cl K x rays in the position spectrum measured with the flat-crystal x-ray spectrometer. The factor N_β is the total number of β particles emitted from a ^{35}S source in the same period of the x-ray measurement. The factor D_x is the overall detection efficiency of the spectrometer for Cl K x rays, which is defined as a ratio of the number of Cl K x rays detected by the PSPC to the total number of Cl K x rays emitted from the ^{35}S source. The ratio n_x/N_β was determined with two different ^{35}S sources, while the efficiency D_x was determined with a ^{109}Cd source. The technical details to obtain n_x/N_β and D_x are summarized in Appendixes A and B, respectively.

The K-hole creation probability per decay, P_K, is

$$P_K=\frac{X_K}{\omega_K}. \tag{2}$$

As seen in Fig. 4, the spectrum of K x rays emitted in the K-IIE process during the β decay is apparently different from that of ordinary K x rays emitted from atoms singly ionized or excited by electron bombardments. The ω_K value, which is to be used in Eq. (2), is not necessarily the same as that for the ordinary K x rays. There is, however, no way to estimate the ω_K value for the K-IIE process in the present situation. In the data analysis for P_K, we have adopted

$$\omega_K=0.099\pm0.010 \tag{3}$$

for chlorine, which is the most reliable value for the chlorine atom singly ionized or excited [12].

In Table I are given the experimental results for X_K and P_K; the present results are in the second column, while those

TABLE I. Results for probabilities X_K and P_K.

	Present work	Rubinson and Howland[a]
X_K[b]	$(2.8\pm 0.3^{d})\times 10^{-4}$	$(2.47\pm 0.35^{e})\times 10^{-4}$
P_K[c]	$(2.8\pm 0.5^{d})\times 10^{-3}$	$(2.3\pm 0.6^{e})\times 10^{-3}$

[a] Reference [6].

[b] K x-ray emission probability per β decay.

[c] K-hole creation probability per β decay.

[d] Standard deviation.

[e] Standard deviation estimated from the 99:1 probable error in Ref. [6].

by Rubinson and Howland [6] are in the third column. A main part of errors in the present results come from ω_K and P_x (the $L\alpha$ x-ray emission probability per electron capture decay of ^{109}Cd, as defined in Appendix B), of which uncertainties are 10% for both factors. Statistical errors caused by x- and β-ray countings in the present work are relatively small compared with these uncertainties.

The errors in the results by Rubinson and Howland are not equivalent to those in the present ones; we have estimated standard deviations, while they estimated extreme errors based on the 99:1 probable errors. Using the standard deviation, their results to be compared with the present ones are given by $(2.47\pm 0.35)\times 10^{-4}$ for X_K and $(2.3\pm 0.6)\times 10^{-3}$ for P_K; the extreme error in their results is 0.6 for X_K and 0.7 for P_K. As seen in Table I, the present X_K and P_K values agree with the previous ones within the standard deviations.

IV. COMPARISON WITH SCF CALCULATIONS

Theoretical works on K-shell internal ionization and excitation (K-IIE) in the past three decades revealed that many-body effects, e.g., electron-electron correlation and screening by other electrons, should explicitly be taken into account for a quantitative comparison of measured P_K data of high reliability. A brief historical sketch of theoretical investigations of the K-IIE process during β decay is given below. Following the approach of electron shake-off and shake-up based on the simple overlap integral [5,13–16], Carlson *et al.* [17] computed the monopole transition probability of

$$P_K = 2(1 - P_R - P_F), \tag{4}$$

where P_R is the probability for a K electron to remain in the K shell, while P_F is the probability for excitations to occupied shells which are forbidden by the Pauli principle, and the factor of 2 accounts for two electrons in the K shell. They used self-consistent-field (SCF) wave functions for bound electrons, i.e., nonrelativistic Hartree-Fock ($Z<30$) and relativistic Hartree-Fock-Slater ($Z\geqslant 30$) wave functions. In order to treat the K-IIE process more precisely, Stephas and Crasemann [18], Law and Campbell [19], and Isozumi, Mukoyama, and Shimizu [20] developed the one-step approach including the phase-space sharing between three leptons (β, atomic electron, and neutrino) and the quantum exchange between β and atomic electrons in the final state.

TABLE II. Theoretical P_K values ($/10^{-3}$) for ^{35}S.

	SCF Calculations	
Present experiment	CNTM[a]	LS[b]
2.8 ± 0.5	3.37	$\sim 5.2^{c}$

[a] Reference [17].

[b] Reference [2].

[c] Obtained by the extrapolation of numerical results in Ref. [2].

Using SCF functions, Law and Suzuki [2] performed systematic calculations according to the one-step approach. The Dirac-Fock-Slater model with optimized potentials for the exchange interaction was employed for the ground state of the parent atom. The configuration in the final state was carefully reconsidered in their SCF calculations. It is implicitly assumed in the SCF calculation by Carlson *et al.* [17] that the final continuum electron moves in the SCF potential defined from the solution of the final ground-state ion; the K-shell occupation in this case is 2. However, the actual final state compels the K-shell occupation to be 1, and one should find the SCF functions for the configuration with a hole in the K shell. They used SCF wave functions for the final state with such a configuration, which were evaluated from three different approximations for the exchange interaction part in the SCF potentials, i.e., the unmodified potential, the modified SCF potential using the local density approximation, and the modified SCF potential by the average Fermi momentum approximation. As already discussed [1], the agreement between calculations by Law and Suzuki and experimental values are excellent except for low-Z nuclides.

Theoretical P_K values for ^{35}S, which were deduced from SCF calculations, are listed in Table II; ''CNTM'' and ''LS'' mean the calculation by Carlson *et al.* [17] and that by Law and Suzuki [2], respectively. Law and Suzuki did not calculate the P_K value for ^{35}S; their value in Table II was obtained by the extrapolation of their results for ^{36}Cl ($Z=17$), ^{45}Ca ($Z=20$), and ^{60}Co ($Z=27$). As seen in Table II, there is an evident difference between the present result and LS value, while the CNTM value is close by the present result. This implies that the SCF approach by Law and Suzuki is to be modified for low-Z nuclides ($Z<20$) in spite of the good agreement for high-Z nuclides.

V. DISCUSSIONS FOR THE COMPOSITE SPECTRUM OF Cl $K\alpha$ x-RAY GROUP

In the present work, we have obtained very interesting spectra of $K\alpha$ x rays emitted in the K-IIE process during β decay of ^{35}S. A typical example of such spectra is given by Fig. 4(a). The present observation of the composite spectrum for the $K\alpha$ x-ray group strongly indicates that the mechanism of the K-IIE process during β decay is quite different from that of the same process during other nuclear decays, e.g., electron capture and internal conversion, and by the bombardment of light charged particles.

The peak area in the position spectra was divided into six peaks by a nonlinear least squares fit [21], in which the shapes of peak and background were assumed to be a Gauss-

TABLE III. Results of measurements with ^{35}S sources.

Source	Intensity (MBq)	Measuring period (sec)	n_x[a] (counts/10^3)	N_β[b] (counts/10^{13})	η[c]	n_x/N_β (/10^{-11})
No. 1	58.8 ± 2.4	487,591	2.56 ± 0.21	2.93 ± 0.12	0.68 ± 0.01	8.72 ± 0.81
No. 2	93.3 ± 3.8	865,020	6.97 ± 0.40	8.46 ± 0.35	0.55 ± 0.01	8.23 ± 0.58

[a] Peak counts of Cl *K* x rays in the position spectrum obtained with the flat-crystal x-ray spectrometer.

[b] The total number of β particles emitted from the ^{35}S source in the same period of the x-ray measurement.

[c] One of the correction factors necessary to deduce the absolute intensity of each ^{35}S source (see Appendix A).

ian curve and a straight line, respectively. A preliminary analysis determined the energies of the six lines to be 2597 eV for P1, 2622 eV for P2, 2636 eV for P3, 2653 eV for P4, 2680 eV for P5, and 2692 eV for P6. The systematic Dirac-Fock calculations for energies of *K* x rays emitted through the decay of the multiple *K*- and *L*–plus–*M*-shell hole states of elements with $Z = 9-18$ were performed by Maurer and Watson [22]. The energies for P3, P4, P5, and P6 lines agree with the Dirac-Fock calculations; P3, P4, P5, and P6 correspond to the states with one, two, three, and four holes in the *L* shell, respectively, in addition to a single *K* hole plus some *M* holes. Thus the lines P3, P4, P5, and P6 are assigned to satellite Cl $K\alpha$ x rays. It is noted that the probability for the multiple ionization and excitation process is relatively large; the P2 line corresponds to a single process, while the P3, P4, P5, and P6 lines correspond to the multiple process. The origin of the P1 line has not been revealed in the present work; a two-quantum phenomenon such as the radiative Auger process [23,24] is one of the possible candidates. A further detailed analysis for the $K\alpha$ x-ray group is now in progress.

The internal ionization and excitation during β decay (IIE) are caused by the sudden change of nuclear charge in the model of electron shake-off and shake-up [1]. Since the change of nuclear charge in β decay is unity, i.e., $Z \rightarrow Z + 1$, the effect of the change to atomic electrons becomes larger for lower-*Z* nuclides. It be expected that the intensities of satellite $K\alpha$ x rays may be relatively increased for lower-*Z* nuclides. We are planning to perform x-ray measurements for simple β-decaying nuclides of ^{32}P, ^{33}P, ^{45}Ca, and ^{63}Ni with the same instrument as used in the present work. Similar high-resolution measurements for higher-*Z* nuclides are also desired, if possible.

VI. CONCLUDING REMARKS

In this work, we have obtained two significant results mainly from the well-resolved spectrum of the Cl $K\alpha$ x-ray group, such as Fig. 4(a). First, the *K*-hole creation probability, P_K for ^{35}S has been determined within a precision enough to be compared with the most reliable calculation by Law and Suzuki [2]. We have found that their theoretical P_K apparently deviates from the value presently determined. It is evident that their SCF theory for the *K*-IIE process should be reexamined for low-*Z* nuclides. Second, we have obtained new information on the mechanism of the *K*-IIE process during β decay. As seen from the intense satellite lines in the spectrum of the Cl $K\alpha$ x-ray group, higher shells in addition to the *K* shell are also ionized or excited in most events of the *K*-IIE process. From the measured spectrum of the $K\alpha$ x-ray group, it is possible to deduce the partial probabilities of the *K*–plus–L^n-hole creation in the *K*-IIE process, where *n* is the number of *L* holes. It would be valuable to have theoretical estimates of the probability of such multiple ionization and excitation in the *K*-IIE process during β decay.

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APPENDIX A: DETERMINATION OF THE RATIO n_x/N_β

Two purified ^{35}S sources were prepared to determine the ratio n_x/N_β in Eq. (1); their intensities were 58.8 and 93.3 MBq. In the measurements to determine the P_K probability, the shortest source-crystal-PSPC arrangement was employed for the most effective x-ray detection. The factor n_x is given by a sum of $K\alpha$ and $K\beta$ peak counts in the position spectrum measured with the spectrometer. Peak counts of six lines in the Cl $K\alpha$ x-ray group were summed up as the $K\alpha$ x-ray counts. The counts of $K\beta$ x rays were actually zero

TABLE IV. Results of measurements with the ^{109}Cd source.

Run	Measuring period (sec)	$n_{x'}$[a] (counts/10^3)	$N_{x'}$[b] (counts/10^{10})	D_x[c] (/10^{-7})
1	15 680	4.06 ± 0.21	2.15 ± 0.24	3.12 ± 0.40
2	24 200	5.91 ± 0.23	3.15 ± 0.35	3.10 ± 0.38
3	6 213	1.42 ± 0.094	7.71 ± 0.84	3.04 ± 0.41

[a] Peak counts of the 2.984-keV Ag $L\alpha_{1,2}$ x rays in the position spectrum obtained with the x-ray crystal spectrometer.

[b] The total number of the 2.984-keV Ag $L\alpha_{1,2}$ x rays emitted from the ^{109}Cd source in the same period of the x-ray measurement.

[c] Overall detection efficiency of the flat-crystal x-ray spectrometer for the Cl *K* x rays, which is defined by Eq. (B1).

because the $K\beta$ line was so weak as to be buried in the background, as seen in Fig. 4(a).

After the x-ray measurement with the flat-crystal spectrometer, the whole carbon sheet of ^{35}S source was immersed in 2N H_2SO_4 solution with a volume of 1 ml, which was then diluted to 100 ml with distilled water. A part of the solution with a volume of 0.1 ml was further diluted to 100 ml, of which 0.1 ml was dissolved in a 10-ml scintillation cocktail to measure the β radioactivity. Then the total number of β particles, N_β, is given by

$$N_\beta = \frac{H_\beta}{D_\beta} \frac{n_\beta}{a_\beta} \frac{1}{\eta}. \tag{A1}$$

In Eq. (A1), H_β is a correction for the decay of ^{35}S in a measuring period:

$$H_\beta = \frac{\exp(\lambda T)}{1-\exp(\lambda \Delta T)}, \tag{A2}$$

where λ is the decay constant of ^{35}S, i.e., $(9.464 \pm 0.008) \times 10^{-8}$/sec, ΔT is the period of the x-ray measurement, and T is the interval between the start time of the x-ray measurement and the time of the absolute β-ray counting of the ^{35}S source. The factor D_β in Eq. (A1) is the β detection efficiency of the liquid scintillation counter presently used (Beckman LS-6500), which was determined to be (0.960 ± 0.015) for ^{35}S with an aid of a ^{35}S reference source prepared by Du Pont Merck Pharmaceutical. The factor n_β is the count rate of the diluted solution obtained by the scintillation counter, and a_β is the dilution factor equal to 10^{-6}. The factor η is a ratio of the activity transferred from the ^{35}S source to the H_2SO_4 solution, which was determined by the β-ray countings of the carbon backing sheet before and after immersing it in the H_2SO_4 solution.

Final results of n_x/N_β as well as measured values of n_x, N_β, and η for the two ^{35}S sources are listed in Table III.

APPENDIX B: DETERMINATION OF THE EFFICIENCY D_x

The detection efficiency, D_x in Eq. (1), is given by

$$D_x = \varepsilon_1 \varepsilon_2 \varepsilon_3 \varepsilon_4 \frac{n_{x'}}{N_{x'}}, \tag{B1}$$

which was determined by using 2.984-keV Ag $L\alpha_{1,2}$ x rays emitted after the electron capture decay of ^{109}Cd; the intensity of a ^{109}Cd source prepared for this purpose was 26.2 MBq. In Eq. (B1), $n_{x'}$ is the peak counts of Ag $L\alpha_{1,2}$ x rays in the position spectrum such as in Fig. 5 and $N_{x'}$ is the total number of Ag $L\alpha_{1,2}$ x rays emitted from the ^{109}Cd source in the same period of the x-ray measurement. Source-crystal and crystal-PSPC distances were exactly the same as for the measurement to determine n_x/N_β with ^{35}S sources, i.e., 50 and 45 mm, respectively.

The ratio of $n_{x'}/N_{x'}$ in Eq. (B1) is the overall detection efficiency for the 2.984-keV Ag $L\alpha_{1,2}$ x rays. The factors ε_1, ε_2, ε_3, and ε_4 are necessary to correct the difference in energies of Cl $K\alpha_{1,2}$ x ray (2.622 keV) and Ag $L\alpha_{1,2}$ x ray (2.984 keV): ε_1, for the intrinsic detection efficiency of the PSPC; ε_2, for the efficiency to penetrate the $(45+50)$-mm path length of hydrogen gas; ε_3, for the efficiency to penetrate the 13.65-μm-thick polyethylene film, with which ^{35}S and ^{109}Cd sources were covered to avoid the activity contamination; ε_4, for diffraction by the Ge(111) crystal. The factor ε_1 was estimated to (1.33 ± 0.03) from the simple calculation performed by our previous work [25]. The factor ε_2 is actually 1.00. The ε_3 value was estimated to (0.962 ± 0.002) from the mass attenuation coefficient of polyethylene.

The integrated reflection power for the perfect crystal in the rotation method is given by

$$P = \frac{\lambda^3}{\sin\theta_c} \propto \frac{1}{E^2}, \tag{B2}$$

where θ_c is the Bragg angle of diffracted photons which goes perpendicular through the center of the PSPC window, and λ and E are the wavelength and energy of incident photon, respectively [26]. With the approximation in Eq. (B2), the factor ε_4 was estimated to be (1.30 ± 0.03).

The total number $N_{x'}$ in Eq. (B1) was determined from

$$N_{x'} = H_{EC} P_{x'} R_\gamma I_{EC}, \tag{B3}$$

where I_{EC} is the absolute intensity calibrated for a ^{109}Cd reference source prepared by Laboratoire de Mesure des Rayonnements Ionisants, R_γ, is the intensity ratio of the 88-keV γ ray emitted from the purified ^{109}Cd source to that from the reference source, $P_{x'}$ is the $L\alpha_{1,2}$ x-ray emission probability per decay, which is equal to 0.050 ± 0.005 [12]. The factor H_{EC} is a correction factor for the decay of ^{109}Cd in the measuring period, of which the expression is the same as for the decay of ^{35}S, i.e., Eq. (A2); the decay constant λ is $(1.734 \pm 0.002) \times 10^{-8}$/sec for ^{109}Cd.

Three experimental runs to determine the efficiency D_x were performed with the same ^{109}Cd source. Results for $n_{x'}$, $N_{x'}$, and D_x are listed in Table IV.

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