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K-Shell Internal Ionization and Excitation in β Decay of ^{35}S

by

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Abstract

K-shell internal ionization and excitation (K-IIE) during the β decay of ^{35}S has been investigated by measuring 2.622-keV Cl $K\alpha$ X rays. A flat-crystal X-ray spectrometer combined with a position-sensitive proportional counter (PSPC) has been developed in the present measurement; this is specially designed for high-resolution observations of low-energy photons from radioisotopes. In order to eliminate spurious Cl X rays which are originated from a small amount of chlorine compounds contained in the ^{35}S source as impurities, Cl ions are removed from the original ^{35}S solution by the purification technique with micro-resin column.

As first step to develop the spectrometer, theoretical and experimental analyses have been performed to examine the position resolution of a proportional counter with a 7- μm diameter carbon fiber as resistive anode, which is used to encode one-dimensional positions of incident photons according to the charge-division method. These analyses have clearly shown that the carbon fiber is applicable to the position sensing of the order of 100~200 μm as long as the filling gas is pressurized at near 10 atm. With this analysis, it is concluded that the energy resolution of the spectrometer is much better than semi-conductor detectors in the low-Z region. A PSPC of this type, i.e., with carbon fiber as resistive anode, has been employed in the flat-crystal spectrometer.

With the aid of the spectrometer and the purified ^{35}S source, we have for the first time succeeded in obtaining a high resolution spectra of Cl K X rays emitted in the K-IIE process during the β decay of ^{35}S . Two interesting conclusions have been obtained by careful analyses of these spectra. First, the observed X rays consist of several lines, indicating that the ionization in β decay is much more furious than that in other nuclear decay processes. The strong intensities of satellite lines have made us conclude that multiple ionizations to produce K plus L holes are dominant rather than single K hole creation. Second, the K-hole creation probability presently

determined for ^{35}S is $(2.8 \pm 0.5) \times 10^{-3}$ per β decay, which agrees well with the previous result by Robinson and Holland within the standard deviation. The presently measured value is considerably lower than the self-consistent field (SCF) calculation based on the one-step theory. This fact strongly imply that the SCF calculation is to be modified for low-Z nuclides such as ^{35}S in spite of the good agreement in high-Z region.

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Contents

1. General Introduction	1
1.1. K-shell internal ionization and excitation accompanying β decay	2
1.2. Motivation of the present investigation	3
1.3. Development of the high-resolution X-ray spectrometer	4
1.4. Measurements and main results	4
2. Resolution of Position-Sensitive Proportional Counter with a Resistive Anode Wire of Carbon Fiber	8
2.1. Introduction	9
2.2. Counter system	10
2.2.1. Position-sensitive proportional counter	10
2.2.2. Electronics	11
2.3. Theoretical treatment for position resolution	11
2.3.1. Fluctuations	11
2.3.2. Expression for position resolution	13
2.3.3. Equivalent charge of thermal noise, Δq_x	16
2.4. Measurements	17
2.4.1. Electronic noise, R_{nos}	17
2.4.2. Limit of R_{nos}	19
2.4.3. Total resolution, R	20
2.5. Discussions	20
2.6. Concluding remarks	23
3. Observation of Cl K X rays from ^{35}S Source with a High-Resolution Crystal Spectrometer	36
3.1. Introduction	37
3.2. Apparatus	38
3.3. Source preparation	39
3.4. Performance of crystal spectrometer	41
3.4.1. Detection of low energy photons	41
3.4.2. Rejection of background electrons	42
3.4.3. End effect of the PSPC	42

3.4.4. Discussions.....	43
3.5. High-resolution measurements of low-energy photons.....	44
3.5.1. X rays from ^{35}S sources.....	44
3.5.2. X rays from ^{109}Cd sources.....	46
3.5.3. Discussions for the composite spectrum of Cl $K\alpha$ X-ray group.....	47
4. Determination of the transition probability for K-Shell internal ionization and excitation in β Decay of ^{35}S	59
4.1. Data analysis.....	60
4.1.1. Definitions of X_K and P_K	60
4.1.2. Ratio N_x/N_β	60
4.1.3. Overall detection efficiency D_x	62
4.1.4. Fluorescence yield ω_K	64
4.1.5. Determination of X_K and P_K	65
4.2. Comparison with SCF calculations.....	65
4.3. Concluding remarks.....	67

Chapter 1

General Introduction

1.1. K-shell internal ionization and excitation accompanying β decay

Since every nuclear transition takes place in the whole system of nucleus plus atomic electrons, the atomic electron cloud is perturbed in most of nuclear decays. For example, some vacancies in inner shells are produced through internal conversion (IC) or electron capture (EC). These nuclear decays are regarded as first-order phenomena caused by mixing terms between atomic and nuclear variables in electromagnetic or weak interactions. The perturbation of the electron cloud during nuclear decays generally results in additional ionizations and excitations of inner shells, but with very small probabilities, e.g., typically 5×10^{-4} per β -decay for the K-shell ionization of ^{90}Y ($Z=39$). This process, called K-shell internal ionization and excitation (K-IIE), is one of the various second-order phenomena during nuclear decays, such as internal bremsstrahlung, internal Compton effect and internal pair creation. K-IIE is one of the most interesting subjects among those phenomena, because it is much concerned with the interaction of the nucleus and the atomic electrons. Therefore, the study of K-IIE can provide valuable information about the atomic correlation effect.

Since K-IIE accompanying β decay is signified by the emission of K X rays of daughter elements, the phenomenon can be observed 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 , i.e., the ratio of the total K-hole creation rate to the total β activity of the source. There are not many β emitters suitable for the measurement of P_K . Most β -active nuclides decay to excited states of the daughter and K X rays are emitted after K internal conversion of the excited states. Because of the small probability P_K , the K X rays caused by the K-IIE are usually buried in identical and more intense K X rays caused by K-IC. Boehm and Wu first studied K-IIE during β decays of ^{147}Pm by observing K X rays of the daughter element ^{147}Sm [1]. Since their work, the P_K values for 23 nuclides have been measured with various methods of X-ray detection. In Table 1.1 are given the experimental P_K and two self-consistent field calculations; experimental data in this table are selected from previous measurements with sufficiently high-resolution X-ray spectrometry, such as by Ge and Si detectors or curved-crystal spectrometers and

those obtained with rather pure resolution detectors, such as proportional and NaI(Tl) counters, are removed.

1.2. Motivation of the present investigation

As seen in Table 1.1, there are few P_K values for β -radioactive nuclides with the atomic number Z less than ~ 20 . One of difficulties to measure 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 . Although P_K increases 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 K X-ray emission; ω_K decreases more rapidly with decreasing Z . For the precise P_K measurement of low- Z nuclides, there is the other difficulty in separating X rays of daughter atoms from adjacent X rays of parent atoms, which are emitted by the collision of β particles with parent atoms in the β source. The energy resolution of high-performance semiconductor detectors is not enough to resolve those adjacent X rays in the low- Z region. Critical P_K measurements with an improved nuclear instrumentation are clearly desired for low- Z β -radioactive nuclides.

The P_K measurement for ^{35}S ($Z=16$) was already performed by Robinson and Howland [2] in 1954. 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 for the amount of Cl ions as impurity in ^{35}S sources; first, Cl ions may be mixed into any stage of source preparation procedures because they exist everywhere in the experimental environment and second, the amount of the Cl ions in the original solution gradually increases as a result of the decay of ^{35}S . It is necessary to remove the Cl ions from the original solution of ^{35}S before the source preparation. There is no remark on the removal of Cl ions in the work by Robinson and Howland. The purpose of the present investigation is to obtain new experimental informations on the K-IIE process accompanying β decay of low- Z nuclides. For this purpose, the measurement by Robinson and Howland has been re-performed improving greatly the resolution of X-ray detection and the quality of ^{35}S sources.

1.3. Development of the high-resolution X-ray spectrometer

For the high-resolution observation of X rays from radioactive sources, we have fabricated a flat-crystal X-ray spectrometer combined with a position-sensitive proportional counter (PSPC). In chapter 2, a theoretical analysis concerning the position resolution of a proportional counter with the 7- μm diameter carbon fiber as a resistive anode is described, which is used to encode one-dimensional positions of incident photons according to the charge-division method. The analysis shows that the carbon fiber is applicable to the position sensing of the order of 100~200 μm as long as the filling gas is pressurized up to near 10 atm.

In sec. 3.1~3.4 of chapter 3, the performance of the X-rays spectrometer is described in detail. The spectrometer has been designed taking into account the theoretical analysis concerning a position resolution of PSPC. It is available for observing low energy photons (2~5 keV) emitted from radioactive sources with the energy resolution of 10~50 eV. The absorption of such low energy photons in air has been diminished by flowing hydrogen gas in a chamber which the spectrometer is mounted inside. Large background counts caused by electrons from the source have been greatly decreased by a small magnet settled in front of the source. The experimental procedure to obtain the high-resolution photon spectra is also given in this chapter.

1.4. Measurements and main results

In sec. 3.5 of chapter 3 are described high-resolution measurements of X rays from ^{35}S and ^{109}Cd with the X-ray crystal spectrometer. With an aid of the much better energy resolution of the X-ray spectrometer, we found that there are several components in the Cl K α X rays emitted in the K-IIE process during β decay. With the present spectrometer, we have for the first time succeeded in high-resolution observation of photons of very weak intensity in radiations emitted from the β -active nuclide ^{35}S , i.e., the 2.62-keV Cl K X rays which are emitted as a result of filling up K-holes produced by the K-shell internal ionization and excitation during

the β decay of ^{35}S .

In chapter 4 are explained the analysis of experimental data used to deduce the P_k probability for ^{35}S . The comparison of our result of P_k with theoretical calculations and some discussions on the high-resolution spectrum of Cl $K\alpha$ X-ray group and some suggestions for further work are also given.

Table 1.1. Measured P_K and available theoretical predictions.

Nuclide	$Z^a)$	$E_0^b)$ (keV)	Experimental P_K ($\times 10^4$)			Theoretical P_K ($\times 10^4$)	
					Ref.	CNTM ^{c)}	LS ^{d)}
³⁶ Cl	18	714	22.1	± 3.8	[3]	30	46.09*
⁴⁵ Ca	21	261	24.3	± 3.9	[4]	21.5	28.57*
⁶⁰ Co	28	320	14.2	± 1.4	[5]	12.3	14.7*
⁶³ Ni	29	67	4.6	± 0.4	[6]	11.5	5.54*
⁶⁴ Cu	30	571	11.9	± 0.8	[7]	10.8	14.28*
			11	± 2	[8]		
⁸⁹ Sr	39	1463	8.6	± 0.7	[4]	6.54	8.97*
⁹⁰ Sr	39	546	5.4	± 1	[9]	6.54	7.3*
			6.5	± 0.8	[4]		
⁹⁰ Y	40	2273	7	± 1	[9]	6.24	8.89*
			7.4	± 1.5	[4]		
⁹⁵ Nb	42	160	3.4	± 0.4	[10]	5.66	2.88*
⁹⁹ Tc	44	292	3.89	± 0.16	[11]	5.18	3.88*
			3.9	± 0.3	[4]		
¹¹⁴ In	50	1989	5.4	± 0.4	[12]	4.09	5.42*
¹⁴¹ Ce	59	444	1.79	± 0.11	[13]	3.11	1.81**
¹⁴³ Pr	60	933	2.88	± 0.2	[14]	3.03	2.9*
			2.92	± 0.16	[6]		
¹⁴⁷ Pm	62	224	0.81	± 0.09	[4]	2.88	0.78*
			0.98	± 0.08	[15]		
¹⁵¹ Sm	63	76	0.036	± 0.005	[15]	2.82	0.02*
			0.023	± 0.003	[16]		
¹⁶⁹ Er	69	340	1	± 0.2	[17]	2.47	0.81*
¹⁸⁵ W	75	430	1	± 0.3	[15]	2.25	0.78*
			1	± 0.2	[4]		
²⁰³ Hg	81	214	0.11	± 0.035	[18]	2.1	0.13*
			0.15	± 0.045	[19]		
²⁰⁴ Tl	82	766	1	± 0.1	[20]	2.08	1.05*
			1.12	± 0.11	[6]		
²¹⁰ Bi	84	1160	1.23	± 0.1	[17]	2.04	1.38*

^aZ : atomic number of daughter element;

^b E_0 : maximum beta-ray energy;

^cCNTM : SCF overlap theory by Carlson et al. [21];

^dLS : SCF one-step theory by Law and Suzuki [22] and (*) and (**) indicate ODFS and LDA approximations, respectively as explained in Sec. 4.2.

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Chapter 2

Resolution of PSPC with a Resistive Anode Wire of Carbon Fiber

The contents of this chapter are based on the following paper:

“Resolution of Position-Sensitive Proportional Counter With a Resistive Anode Wire of Carbon Fiber”

D. Ohsawa, S. Masaoka, R. Katano and Y. Isozumi.

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2.1. Introduction

The position-sensitive proportional counter (PSPC) is devised to deliver signals with pulse heights proportional to the distance from one end of entrance window to the position where incident radiations arrive on the window. Various techniques of position sensing for one-dimensional PSPC have so far been developed: (A), charge-division method with resistive anode [1-6]; (B), rise-time method with resistive anode [7-9]; (C), charge-division method with resistive cathode strips [10]; (D), delay-line method with cathode strips [3,11-15]; (E), charge-division method with graded-density cathode [16-18]; (F), charge-division method with cathode printed backgammon pattern on [19,20]; (G), grouped-cathode-wire method [21,22].

Since there is no need to use particular devices for the cathode, PSPC based on the method (A) is much simpler than that based on other methods. One of difficulties in the method (A) is the thermal noise coming from the resistive anode. A simple way to decrease the thermal noise is to employ a wire of higher resistance; the root mean square of the thermal noise is inversely proportional to the square root of anode resistance. There are some materials applicable as the resistive anode wire: carbon-coated quartz, alloy metal and carbon fiber. The electric resistance of commercially available quartz wires is high enough, e.g., 1~50 k Ω /cm. However, the life span of such anodes is shortened through the exfoliation of carbon film on the quartz wire, which is easily caused by electric discharges inside PSPC. Alloy metal wires are stronger than quartz wires. A shortage of alloy metal wires is their comparatively low resistance, e.g., 0.17 k Ω /cm for Karma (70%Ni, 20%Cr, 7%Al+Fe) 10 μ m in diameter and 0.12 k Ω /cm for Nichrome (80%Ni, 20%Cr) 10 μ m in diameter. The resistance of carbon fibers is as high as that of the carbon-coated quartz wires, e.g., 4 k Ω /cm for the 7- μ m diameter wire, while they are more fragile to be handled than the quartz or metal alloy wires. The performance of carbon fiber as resistive anode was extensively examined in the previous works [23-26]. In our previous works [27,28], a PSPC with the carbon fiber as resistive anode was applied to the flat-crystal X-ray spectrometer designed for detecting low-energy photons emitted from radioactive sources. We have recently developed a very simple PSPC of gas-seal type with the 7- μ m diameter carbon

fiber as resistive anode for the X-ray emission spectroscopy [29-31].

A main purpose of this chapter is to make clear a practical limit of position resolution for PSPC with the resistive anode of 7- μm diameter carbon fiber. The PSPC of gas-sealed type and the electronics for operating it are described in Sec. 2.2. In Sec. 2.3 is given a theoretical treatment for estimating the contribution of various fluctuations to the position resolution, *i.e.*, the thermal noise from the resistive anode, other noises from the electronics components and spatial fluctuations in the operation of PSPC. Results of some measurements for those fluctuations and that for the resolution itself are given in Sec. 2.4; a limit of the resolution caused by the electronic noises is 63 μm in length and the best resolution for the present PSPC is 130 μm . Some critical discussions on the present measurements are given in Secs. 2.5 and 2.6.

2.2. Counter system

2.2.1. Position-sensitive proportional counter

In Fig. 2.1 is shown a schematic drawing of the present position-sensitive proportional counter, which is designed for sealing the counter gas with pressures up to 10 atm. The resistive anode wire (AN) is a carbon fiber 7 μm in diameter, which is manufactured by Toray Co. Ltd.; the length of the anode is 100 mm and its total resistance is 42 k Ω . The entrance window (WF) is made of a 200- μm thick aluminum sheet or a 1.5-mm thick beryllium plate. The area of this window is 100 $\times$ 10 mm². The material of the counter frame (CF) is 25-mm thick stainless steel; the shape of the counter is rectangular parallelepiped with the sensitive gas layer 20 mm in thickness. The support (SW) for the entrance window is made of stainless steel 7 mm in thickness. The guide for the anode wire is a hermetic seal with a stainless steel tube 0.8 mm in inner diameter (HS), which is used for the electrical insulation between the anode and the counter frame. Aluminum rings of 0.3 mm in thickness (AR) are used as sealing materials between HS and CF. The sealing material between WF and CF is an aluminum wire of 0.8 mm in diameter. The counter gas is sealed by valves for high pressure uses; the maximum gas pressure is 3 atm for the 200- μm thick aluminum window and 10 atm for the 1.2-mm thick beryllium window.

2.2.2. Electronics

A block diagram of the present position-sensing system by the charge-division method is given by Fig. 2.2. The charge Q_1 received by the preamplifier PA1 (Canberra 2003T) is

$$Q_1 = Q \frac{X_0}{L}, \quad (1)$$

while the charge Q_2 received by the preamplifier PA2 (Canberra 2003T) is

$$Q_2 = Q \left(1 - \frac{X_0}{L}\right), \quad (2)$$

where L is the length of the resistive anode wire and Q is the total electric charge generated by an incident photon which arrives at the position X_0 on the entrance window of PSPC. Outputs from PA1 and PA2 are processed with spectroscopy amplifiers, AMP1 and AMP2 (Ortec 472A), respectively; the output voltage of AMP1, V_1 , is proportional to the position X_0 , while that of AMP2, V_2 , is proportional to $L - X_0$. The sum inverter, SUM (Tennelec TC253), delivers signals proportional to the total charge $Q(=Q_1+Q_2)$, which corresponds to energies deposited in the proportional counter by incident radiations. Outputs of AMP1 and SUM are fed into an analog divider, DIV (Ortec 464), to compute the ratio of $V_1/(V_1+V_2)$. The output from DIV is thus normalized by the energy of the incident radiation and linearly related to the position X_0 . The two-dimensional spectra of position and energy are obtained by piling position and sum-energy signals from DIV in a data acquisition system, PHA (Fast Comtec MPA/PC).

2.3. Theoretical treatment for position resolution

2.3.1. Fluctuations

The total charge Q induced on the anode wire distributes in the direction parallel to the wire because of some physical processes

inherent in the counter operation of PSPC, e.g., the finite range of electrons created by photo-ionization and following Auger transition, the diffusion of electron cloud and the spatial spread of electron avalanches, as discussed in Sec. 2.5. Therefore, the position encoded by the charge-division method is not the incident position X_0 of photons on the entrance window, i.e., what we want to know, but the center of gravity of the charge distribution on the anode wire, X^* , which is given by

$$X^* = \frac{\int_0^L q(x) x dx}{Q}, \quad (3)$$

where $q(x)$ is the charge distribution as a function of the position x on the anode wire and total charge Q is given by

$$Q = \int_0^L q(x) dx. \quad (4)$$

The deviation δx ($\equiv X^* - X_0$) causes the fluctuations δQ_1 and δQ_2 in the charges Q_1 and Q_2 , respectively, which are estimated as

$$\delta Q_1 = \int_0^L q(x) \frac{x}{L} dx - Q \frac{X_0}{L} = Q \frac{\delta x}{L}, \quad (5)$$

$$\delta Q_2 = \int_0^L q(x) \left(1 - \frac{x}{L}\right) dx - Q \left(1 - \frac{X_0}{L}\right) = -Q \frac{\delta x}{L}. \quad (6)$$

The number of electrons passing through the resistive wire statistically changes because of their thermal motions in the resistor. This change also causes the fluctuation in Q_1 and Q_2 as shown in Fig. 2.3 (a) and (b), which are output signals of the preamplifiers PA1 and PA2, respectively; the figure (c) is the sum of the outputs (a) and (b). The input of charge-sensitive preamplifier can be approximated to be at the earth potential, as long as its open-loop gain in the feed-back part is sufficiently high. The preamplifier PA1 in Fig. 2.2 observes the fluctuation from one end of the anode wire, of which the other end is equivalently at the

earth potential. The preamplifier PA2 observes the same fluctuation, but from the contrary side of the anode wire. Therefore, the thermal fluctuation in Q_1 is precisely reversed to that in Q_2 . It is seen in Fig. 2.3 that the sign of the noise (a) is contrary to that of (b) as expected; the amplitude of the sum (c) is much less than those of the noises (a) and (b), since these thermal noises are compensated each other in the sum (c).

Including the contribution from the thermal noise, fluctuations in Q_1 and Q_2 are expressed as

$$\delta Q_1 = Q \frac{\delta x}{L} + \delta q_r, \quad (7)$$

$$\delta Q_2 = -Q \frac{\delta x}{L} - \delta q_r, \quad (8)$$

where $Q \delta x/L$ is the fluctuation caused by the spatial distribution of charges induced on the anode and δq_r is that by the thermal motion of electrons in the resistive anode wire. It is noted that the fluctuation δQ_1 is precisely inverse to δQ_2 .

In addition to the fluctuations δQ_1 and δQ_2 , other fluctuations caused by the noises of electronic components are superimposed on the output from the present position-sensing system given by Fig. 2.2, i.e., charge fluctuations δq_{a1} and δq_{a2} at the input stages of PA1+AMP1 and PA2+AMP2, voltage fluctuation δv_s from SUM and voltage fluctuation δv_d from DIV. All the fluctuations δq_{a1} , δq_{a2} , δv_s and δv_d can be treated to be independent each other because they come out from different sources.

2.3.2. Expression for position resolution

The output voltage from DIV is given by

$$V_d = \left(\frac{V_1}{V_s} \right) V_{const}, \quad (9)$$

$$V_s = V_1 + V_2 = Q H_p G_a, \quad (10)$$

$$V_1 = Q_1 H_p G_a, \quad (11)$$

$$V_2 = Q_2 H_p G_a, \quad (12)$$

where V_d is the output from DIV, V_{const} is 10 V for the divider presently employed (Ortec 464), V_s is the output from SUM, H_p is the charge gain of PA1 and PA2, which is approximately 0.45 V/pC for Canberra 2003T, and G_a is the voltage gain of AMP1 and AMP2. The fluctuation in V_d is deduced as

$$\begin{aligned} \delta V_d &= \delta v_d + V_{const} \delta \left(\frac{V_1}{V_s} \right) \\ &= \delta v_d + V_{const} \left(\frac{\delta x}{L} + \frac{\delta q_r}{Q} + \frac{\delta q_{a1}}{Q} \right) - V_{const} \left(\frac{Q_1}{Q} \right) \left(\frac{\delta q_{a1}}{Q} + \frac{\delta q_{a2}}{Q} + \frac{\delta v_s}{V_s} \right). \end{aligned} \quad (13)$$

Since δx , δq_r , δq_{a1} , δq_{a2} , δv_s and δv_d are independent each other following the Gaussian distribution, the statistical average of a product of any pair of the variables is zero. Then, the statistical average of $(\delta V_d)^2$ is expressed as

$$\begin{aligned} (\Delta V_d)^2 &= (V_{const})^2 \left\{ \left(\frac{\Delta x}{L} \right)^2 + \left(\frac{\Delta q_r}{Q} \right)^2 + \left[\left(\frac{Q_1}{Q} \right)^2 + \left(\frac{Q_2}{Q} \right)^2 \right] \left(\frac{\Delta q_a}{Q} \right)^2 \right. \\ &\quad \left. + \left(\frac{Q_1}{Q} \right)^2 \left(\frac{\Delta v_s}{V_s} \right)^2 \right\} + (\Delta v_d)^2, \end{aligned} \quad (14)$$

where Δs is the root mean square (RMS) of a statistical variable s , which is defined by

$$\Delta s \equiv \text{Av} \left[(\delta s)^2 \right], \quad (15)$$

where $\text{Av}[\delta x_1 \cdot \delta x_2]$ expresses the average of a product of variables δx_1 and δx_2 .

In the derivation of Eq. (14), we assume that RMS noises from PA1+AMP1 are equal to that from PA2+AMP2, i.e., $\Delta q_{a1} = \Delta q_{a2} [\equiv \Delta q_a]$.

The position resolution of PSPC, defined as the full width at half maxima (FWHM), is given by $2.355 \Delta X/L$, where ΔX is RMS of uncertainties in determining the incident position X_0 by the present position-sensing system; the constant $2.355 \left(= 2 \sqrt{2 \ln 2} \right)$ is necessary to transform the RMS values to the FWHM values. Using a relations of

$$\frac{\Delta X}{L} \equiv \frac{\Delta V_d}{V_{const}}, \quad (16)$$

the position resolution R is easily deduced as

$$R \left[\equiv 2.355 \frac{\Delta X}{L} \right] = 2.355 \frac{\Delta V_d}{V_{const}} = \sqrt{(R_{spa})^2 + (R_{nos})^2}, \quad (17)$$

In the above equation, R_{spa} comes from the spatial spread of the charge inherent from the counter operation of PSPC, which depends on the filling gas and the anode voltage, and R_{nos} comes from the thermal noise of the anode wire and additional noises of electronics components, which depends on the resistance of the anode wire and electronics employed, respectively. The explicit expression of R_{spa} is

$$R_{spa} = 2.355 \frac{\Delta x}{L} \quad (18)$$

and that of R_{nos} is

$$R_{nos} = \sqrt{(R_r)^2 + (R_a)^2 + (R_s)^2 + (R_d)^2}, \quad (19)$$

where the partial resolutions R_r , R_a , R_d and R_s are given by

$$R_r = 2.355 \frac{\Delta q_r}{Q}, \quad (20)$$

$$R_a = 2.355 \sqrt{\left(\frac{X_0}{L} \right)^2 + \left(1 - \frac{X_0}{L} \right)^2} \frac{\Delta q_a}{Q}, \quad (21)$$

$$R_s = 2.355 \frac{X_0}{L} \frac{\Delta v_s}{V_s}, \quad (22)$$

$$R_d = 2.355 \frac{\Delta v_d}{V_{const}}. \quad (23)$$

2.3.3. Equivalent charge of thermal noise, Δq_r

Output signals caused by the thermal noise from resistors are shown in Fig. 2.4, which were observed by connecting different resistances in series between preamplifiers PA1 and PA2. It is seen that the amplitude of noises becomes larger as the resistance is smaller. The thermal noise is quantitatively expressed in the form of the equivalent noise charge Q_{enc} , in which the noise intensity of a system consisting of a charge-sensitive preamplifier and a spectroscopy amplifier is converted to the electric charge at the input stage of the preamplifier. By developing the previous theoretical treatments [32,33], Q_{enc} is deduced as

$$Q_{enc}^2 = U_{ps}(n) \left[\left(\frac{kT}{2\sigma} + \frac{qI_g}{4} \right) \tau + \frac{1}{2n-1} \frac{kT}{2} \frac{R_{eq} C^2}{\tau} \right] + 4 U_f(n) A_f C^2, \quad (24)$$

$$U_{ps}(n) = 2 (2n)! \left(\frac{e}{2n} \right)^{2n}, \quad (25)$$

$$U_f(n) = \frac{(n!)^2}{2n} \left(\frac{e}{n} \right)^{2n}, \quad (26)$$

for the RC pulse shaping circuit of one-stage differential plus n-stage integrals which delivers unipolar signals. For the circuit of two-stage differentials plus n-stage integrals to produce bipolar signals, Q_{enc} is deduced as

$$Q_{enc}^2 = B_{ps}(n) \left[\left(\frac{kT}{2\sigma} + \frac{qI_g}{4} \right) \tau + \frac{3}{2n-1} \frac{kT}{2} \frac{R_{eq} C^2}{\tau} \right] + 4 B_f(n) A_f C^2, \quad (27)$$

$$B_{ps}(n) = \frac{(2n)! e^{2(n+1-\sqrt{n+1})}}{2^{2n} (n+1-\sqrt{n+1})^{2n}}, \quad (28)$$

$$B_f(n) = \frac{[(n+1)!]^2 e^{2(n+1-\sqrt{n+1})}}{2(n-1)n(n+1)(n+1-\sqrt{n+1})^{2n}}. \quad (29)$$

In the above expressions, k is Boltzmann constant (8.617×10^{-5} eV/K), T is absolute temperature (K), τ is the time constant of the RC pulse shaping in the spectroscopy amplifier, σ is the resistance of the parallel resistor (Ω), q is the charge of electron (1.602×10^{-7} pC), I_g is the sum of leak currents from PSPC and the field-effect transistor (FET) at the input stage of the preamplifier, R_{eq} is the equivalent noise resistance to express the thermal noise in the FET and A_f is the $1/f$ noise constant for the FET.

According to Eqs. (24) and (27), the equivalent charge of the thermal noise, Δq_r , is given by

$$\Delta q_r = C_n \sqrt{\frac{k T \tau}{2 \sigma}}, \quad (30)$$

where C_n is $U_{ps}(n)$ for unipolar signals and $B_{ps}(n)$ for bipolar signals. The noise charge Δq_r is inversely proportional to $\sqrt{\sigma}$ and decreases as the number of stages of the integral circuit, n , increases.

2.4. Measurements

2.4.1. Electronic noise, R_{nos}

In Fig. 2.5 is given the resolution R_{nos} measured as a function of the charge Q , which were fed to PA1 and PA2 through two resistors with a resistance $\sigma/2$, as seen in Fig. 2.6: (a), $\sigma=1.12$ k Ω ; (b), $\sigma=10.4$ k Ω ; (c), $\sigma=103$ k Ω ; (d), $\sigma=1008$ k Ω ; (e), $\sigma=9390$ k Ω . In the measurement, the output from SUM, V_s , is fixed to 4 V by adjusting the gain G_a of the amplifiers AMP1 and AMP2. Then, Q is given by $4/(H_p G_a)$, H_p is

the charge gain of the preamplifier. Solid curves in Fig. 2.5 were obtained by fitting measured R_{nos} with a model function of

$$R_{nos}(Q) = 2.355 \sqrt{\left(\frac{\alpha}{Q}\right)^2 + \beta^2}, \quad (31)$$

$$\alpha = \sqrt{\frac{\alpha_1^2}{\sigma} + \alpha_2^2}. \quad (32)$$

which are equivalent to Eqs. (19)~(23); the details of χ^2 fit is given in Ref. 34. The agreement between experimental data and estimated curves is satisfactorily good, as seen in Fig. 2.5. Results for free parameters α and β in Eq. (31) are listed in Table 2.1. Using the numerical values in the table, α_1 and α_2 in Eq. (32) were determined by the same χ^2 fit. Final results for α_1 , α_2 and β are

$$\alpha_1 \left[\equiv C_n \sqrt{\frac{k T \tau}{2}} \right] = (16600 \pm 850) \sqrt{k \Omega} \text{ electrons}, \quad (33)$$

$$\alpha_2 \left[\equiv \Delta q_a \right] = (390 \pm 60) \text{ electrons}, \quad (34)$$

$$\beta \left[\equiv \sqrt{\left(\frac{\Delta v_s}{4}\right)^2 + \left(\frac{\Delta v_d}{10}\right)^2} \right] = (2.65 \pm 0.15) \times 10^{-4}, \quad (35)$$

Assuming $T=300$ K and the circuit of two-stage differentials and two-stage integrals in the spectroscopy amplifier, *i.e.*, $C_n=B_{ps}(2)$, the width of output pulses from the amplifier, τ_a , is estimated as

$$\tau_a = \frac{2}{k T} \left[\frac{\alpha_1}{B_{ps}(2)} \right]^2 = (1.7 \pm 0.1) \mu\text{sec}. \quad (36)$$

This value corresponds to the period when the bipolar signal goes up to the maximum, $1.7/2=0.85 \mu\text{sec}$, which is consistent to the time constant of the amplifier presently used (Ortec 472A), *i.e.*, $1.0 \mu\text{sec}$. The equivalent noise of the preamplifier (Canberra 2003T) is nominally 2~3 keV (FWHM, Si), which is equivalent to

$$\Delta q_a = 230 \sim 350 \text{ electrons} \quad (37)$$

This nominal value is also consistent with the value presently determined, i.e., Eq. (34).

2.4.2. Limit of R_{nos}

According to Eqs. (33) and (34), partial resolutions R_r and R_p for the present system are respectively given by

$$R_r = \frac{39000 q}{\sqrt{\sigma_0} Q} \quad (38)$$

$$R_a = \sqrt{\left(\frac{X_0}{L}\right)^2 + \left(1 - \frac{X_0}{L}\right)^2} \frac{920 q}{Q} \quad (39)$$

As Q is much larger, R_r and R_a go to zero and then R_{nos} approaches to a value given by

$$R_{nos}(Q \rightarrow \infty) = \sqrt{R_s^2 + R_d^2} = 2.355 \beta$$

$$(6.26 \pm 0.35) \times 10^{-4} \text{ for } \frac{X_0}{L} = 0.5 \text{ and } V_s = 4 V \quad (40)$$

Thus, a limit in position resolution is 6.26×10^{-4} or $62.6 \mu\text{m}$ in length for the present PSPC system, which results from the noises from SUM and DIV.

In Fig. 2.7, R_{nos} , R_r , R_a and estimated according to Eqs (19), (38), (39) and (40), respectively, as well as experimental R_{nos} are plotted as a function of the input charge Q . The position resolutions measured for 6.4 keV photons are also shown in the figure, which are discussed in Sec. 2.4.3. The agreement between estimated and experimental R_{nos} is rather excellent, indicating the self-consistency of the present treatment for the noise fluctuations. The partial resolutions R_r and R_a linearly decreases as Q increases. The noises from PA1+AMP1 and PA2+AMP2 are one tenth of the thermal noise from the carbon fibre, i.e., $R_a \approx R_r/10$. This means that, for the PSPC system with the $42 \text{ k}\Omega$ resistive anode, it is not necessary to employ the

preamplifier of noise performance as low as presenly used, *i.e.*, 2~3 keV. In the region of $Q < 10^6$ electrons, a main source of the resolution R_{nos} is the thermal noise. When $Q > 10^6$ electrons, the contribution of the thermal noise relatively decreases and R_{nos} approaches to the limit as Q increases. In the region of $Q > 5 \times 10^7$ electrons, R_{nos} is approximately equal to the limit.

2.4.3. Total resolution, R

The position resolution of the present PSPC was measured with the 6.4-keV Fe $K\alpha$ X-ray beam collimated in the width of 25 μm , which was generated by an apparatus arranged for the present measurement. The X rays emitted from an ordinary X-ray tube were guided in a brass pipe of 2 cm in inner diameter and 160 cm in length. A slit made of molybdenum was mounted at the end of the brass pipe; the aperture of slit is 25 $\mu\text{m} \times 10$ mm. To diminish the absorption of the X rays by the air layer, the inside of guide pipe was kept in vacuum by a rotary pump; 40- μm thick kapton films were used as vacuum seal. In the measurement, pressurized gas mixtures, *i.e.*, Ar+5%CH₄ and Ar+5%CH₄ with a pressure of 1 to 10 atm were examined as filled gas; the resolution was better with a higher pressure.

In Fig. 2.7 are shown the position resolutions obtained with the highest pressure, *i.e.*, 10 atm, which were measured as a function of the output charge Q from the PSPC: (a), with Ar+5%CH₄; (b), with Ar+10%CH₄. Differences between measured resolutions and the noise contribution R_{nos} are caused by the spatial fluctuation of Q , *i.e.*, R_{spa} given by Eq. (18). As seen in Fig. 2.7, the difference becomes smaller with decreasing Q and both resolutions measured for Ar+5%CH₄ and Ar+10%CH₄ approach to an asymptotic line corresponding to noises from resistive anode preamplifier, *i.e.*, $\sqrt{(R_r)^2 + (R_a)^2}$. The minimum value of the resolution is 130 μm for Ar+10%CH₄ and 160 μm for Ar+5%CH₄ near $Q = 1.5 \times 10^7$ electrons. When Q is larger than this value, the resolution apparently becomes worse.

2.5. Discussions

The spatial fluctuation of electric charges induced on the anode of PSPC, which makes worse the position resolution of PSPC,

results from some physical processes in the counter operation of PSPC. The first process is the collision of high-energy electrons produced through the interaction of incident photons with the counter gas. For example, the 6.4-keV incident photons mainly produce 3.2-keV electrons in argon gas through the photo-effect, leaving a K-shell hole in argon. Extra electrons with energies of about 2.7 keV are subsequently emitted through the Auger transition of the K-hole state. These electrons generate many electrons with lower energies along their trajectories by ionizing continuously the surrounding gas atoms and molecules. The average length of their trajectories is not negligible compared with the position resolution of PSPC; for example, the range of 3-keV electrons is 200~300 μm in argon gas with a pressure of 1 atm [35,36]. A electron cloud is finally formed after all the electrons are thermalized in the counter gas. Thus, the incident position of the photons is made obscure by the formation of electron cloud, strongly depending on the range of the high-energy electrons in the counter gas. Generalized semi-empirical equations, which are available to estimate the range of the high-energy electrons created in the gas-filled proportional counters, were given by Tabata et al [37]. In a rough approximation, the range of electrons is inversely proportional to the density of filling gas [18].

The second process responsible for the spatial fluctuation is the diffusion of the electron cloud to the anode wire. Being attracted by the electric field, each electron in the cloud drifts to the anode wire. Since the electron cloud is more extended in the travel to anode, the incident position of photons is more obscure. The spread of the electron cloud in density perpendicular to the electric field is expressed by a radial Gaussian distribution [35], of which variance σ_x is

$$\sigma_x = \sqrt{2 \frac{D}{\mu} \frac{L_r}{E}} \quad (41)$$

where D is the diffusion coefficient, μ is the electron mobility L_r is the drift distance and E is the electric field strength. With the approximate forms of D and μ by Rice-Evans [39], it is shown that σ_x is inversely proportional to the square root of the counter gas pressure P and proportional to the square root of the drift distance L_r :

$$\sigma_x \propto \sqrt{\frac{L_r}{P}} \quad (42)$$

The contribution of the lateral diffusion to the position resolution was examined rather in detail in the previous works [40,41]. According to the simple treatment [18], the spread due to the lateral diffusion is estimated to the order of 40 μm for the mean drift length in the present PSPC, *i.e.*, 10 mm; the filling gas is assumed to Ar+10%CH₄.

The third process is the electron avalanches on the anode wire. The operation mode of gas-filled counter depends on the gas gain, *i.e.*, the average of avalanche sizes. The spatial spread of each avalanche is larger with increasing the gas gain. At the large gas gain, which corresponds to the upper side of the proportional counter mode or the transition region from proportional to the self-quenching streamer mode [42,43], the fluctuation in the spatial spread of each avalanche is steeply increased. This effect apparently results in the deterioration of the position resolution.

The effect of these three processes depends on the photon energy to be analyzed, the counter geometry, the filling gas and the applied anode voltage. For usual conditions of PSPC system, the effect of the first process, *i.e.*, the collision of high-energy electrons, is much larger than that of other processes [44]. Some previous works clearly showed that the spatial fluctuation due to the first process can be greatly diminished by increasing the pressure of the filling gas [14,44-46]. For example, the range of 3-keV electrons in argon is decreased from 200~300 μm to 20~30 μm by increasing the gas pressure from 1 atm to 10 atm.

As seen in Fig. 2.7, there is a minimum in both curves (a) and (b) of position resolution measured as a function of the output charge Q ; the resolution becomes worse when the gas gain is larger than the value at this minimum, *i.e.*, $Q=1.5 \times 10^7$. This effect comes from the third process discussed in the preceding section, *i.e.*, the fluctuation of electron avalanches inherent in the counter operation of the high gas gain; the gas gain at $Q=1.5 \times 10^7$ electrons is estimated to 5.9×10^4 , assuming that the W value of the argon gas mixtures, *i.e.*, the average of energy required to produce a pair of electron and positive ion, is 25 eV. Since the anode wire of the present PSPC is

very fine, i.e., 7 μm in diameter, the energy linearity for the incident energy below 10 keV is kept only in the region of gas gains below 10^3 , which corresponds to the anode voltage of 2200 V. Above this gain, the counter does not operate in the proportional region, but in the transitional region. The gas gain 5.9×10^4 , with which we obtained the best position resolution, is in the rather instable region of the counter operation; the photo-peak of the 6.4 keV Fe $K\alpha$ X rays in the energy spectrum is split into two peaks, probably because of the appearance of the self-quenching operation mode. In order to understand the relation between the position resolution and the operation modes, systematical measurements with various mixtures of argon and methane in the pressure range of 1~10 atm are now in progress. The best data for the position resolution is given by Fig. 2.8.

2.6. Concluding remarks

A useful method to improve the position resolution is to suppress the range of photo-electrons by increasing the pressure of filling gas, as discussed before. The increase of gas pressure results in the increase of anode voltage to keep the same gas gain. Because of the fine carbon fiber, the anode voltage for the present PSPC is comparatively low even at high pressures of filling gas; for example, the anode voltage to obtain the gas gain of 6×10^4 for Ar+10%CH₄ with a pressure of 10 atm is 3000 V for the present carbon fiber 7- μm in diameter, while that is more than 5000 V for a 30- μm diameter tungsten wire usually employed in gas-filled counters.

For X-ray measurements with complicated experimental arrangements, it is desirable to use the PSPC in which the counter gas can be sealed; the gas circuit is not necessary for PSPC of that type, but inevitable for PSPC of gas-flow type to supply the constant flow of counter gas. Outer gases coming from the particular cathode plates usually prevent us from making PSPC of gas-seal type based on the backgammon method or the delay-line method; with these methods, it is necessary to mount a specially printed board as a cathode. Thus, the present method with the resistive anode is most suitable for fabricating PSPC of gas-seal type because of its simplest structure.

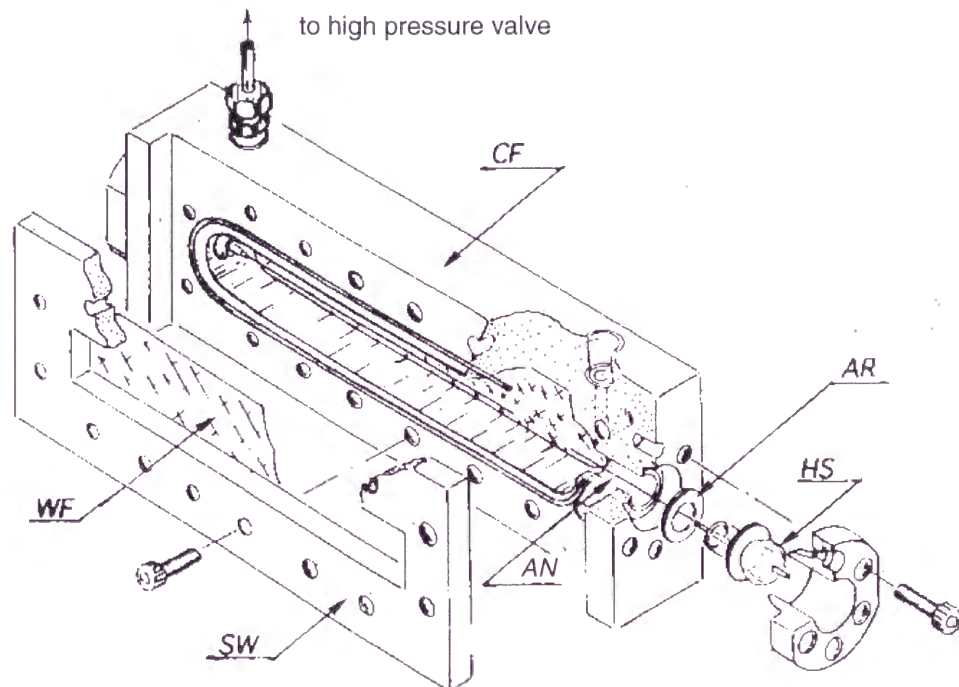


Fig. 2.1. The present position-sensitive proportional counter designed for sealing the counter gas with pressures up to 10 atm: **AN**, resistive anode wire (carbon fiber of 7 μm in diameter); **WF**, entrance window (200- μm thick aluminum sheet or 1-mm thick beryllium plate); **CF**, counter frame (stainless steel 25 mm in thickness); **SW**, support for the entrance window (stainless steel 7 mm in thickness); **HS**, hermetic seal; **AR**, aluminum ring of 0.3 mm in thickness.

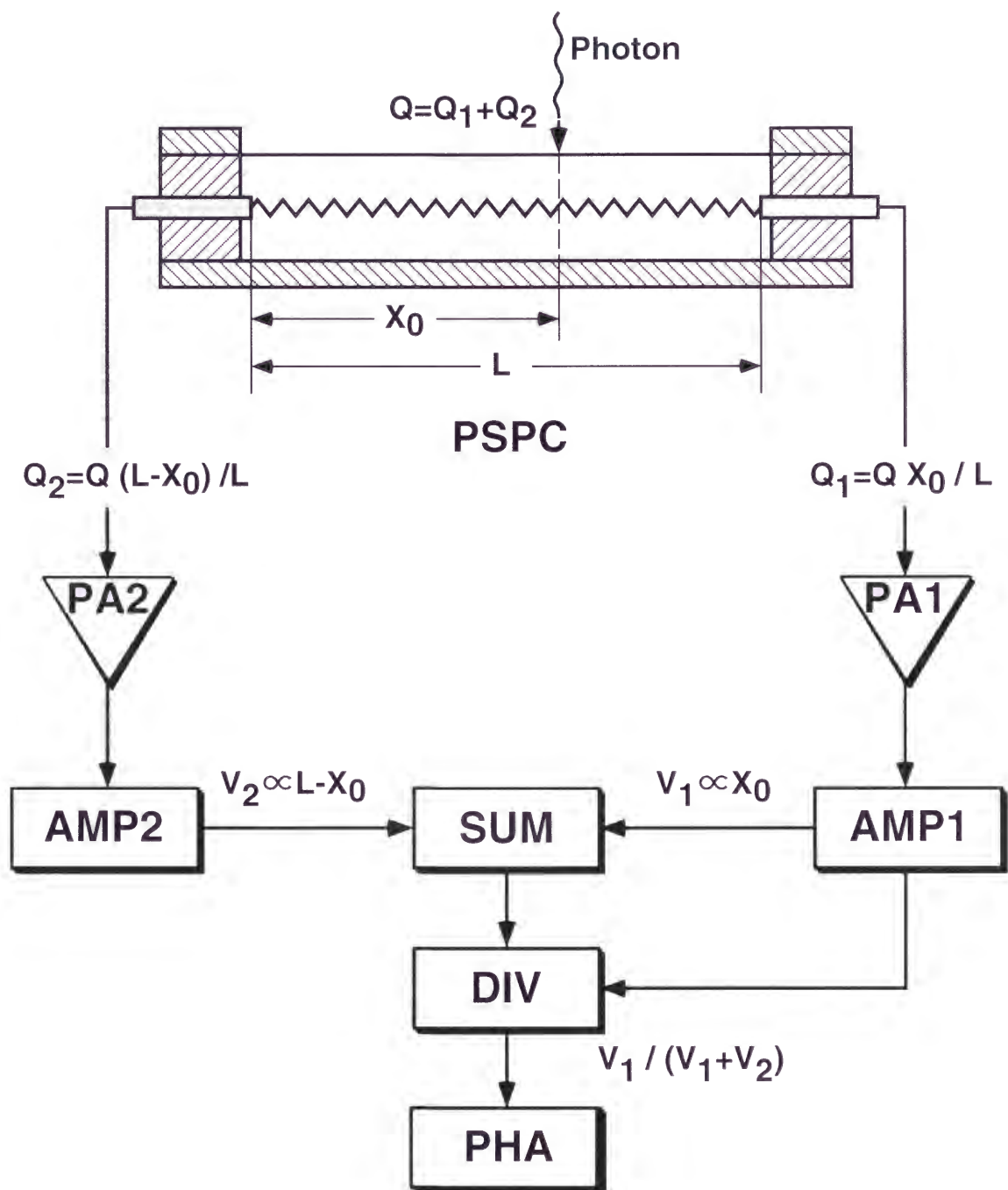


Fig.2.2 Block diagram of the position sensing system by the charge-division method: **PSPC**, position-sensitive proportional counter with resistive anode; **PA1** and **PA2**, charge-sensitive preamplifiers (Canberra 2003T); **AMP1** and **AMP2**, spectroscopy amplifiers (Ortec 472A); **SUM**, sum inverter (Tennelec TC253); **DIV**, analog divider (Ortec 464); **MCA**, multi-channel analyzer (Fast Comtec MPA/PC).

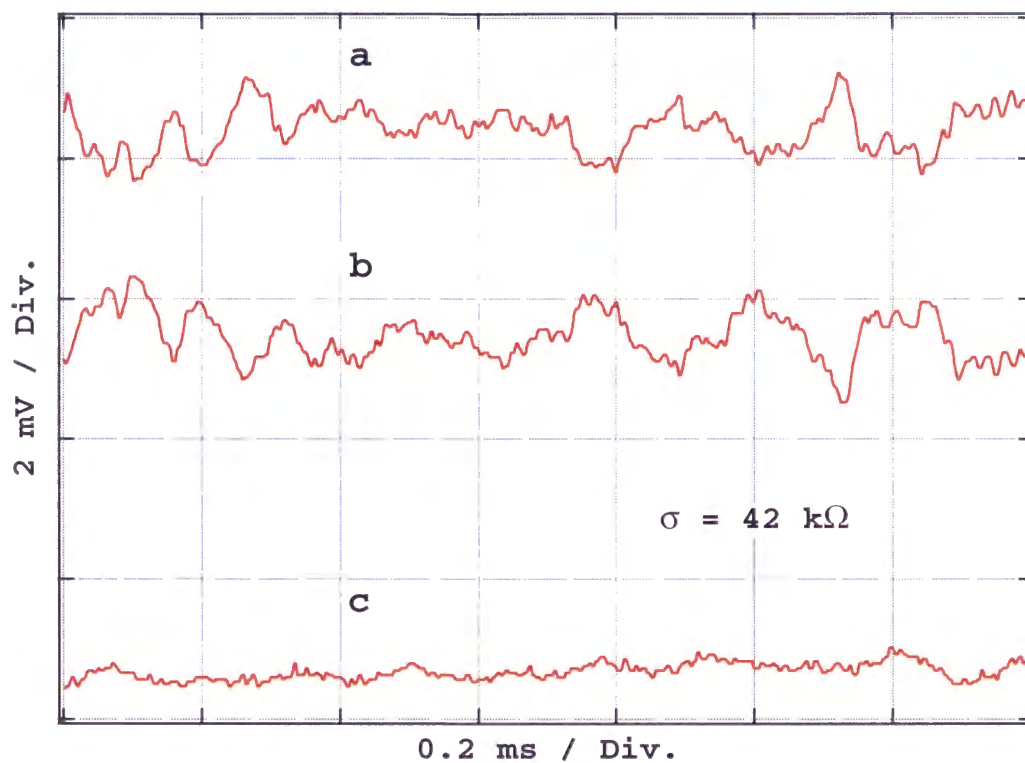


Fig. 2.3. Outputs from the preamplifiers PA1 and PA2 of the present PSPC system with the resistive anode of carbon fiber 7 μm in diameter (42 $\text{k}\Omega$); (a), output from PA1; (b), output from PA2; (c) sum of outputs from PA1 and PA2.

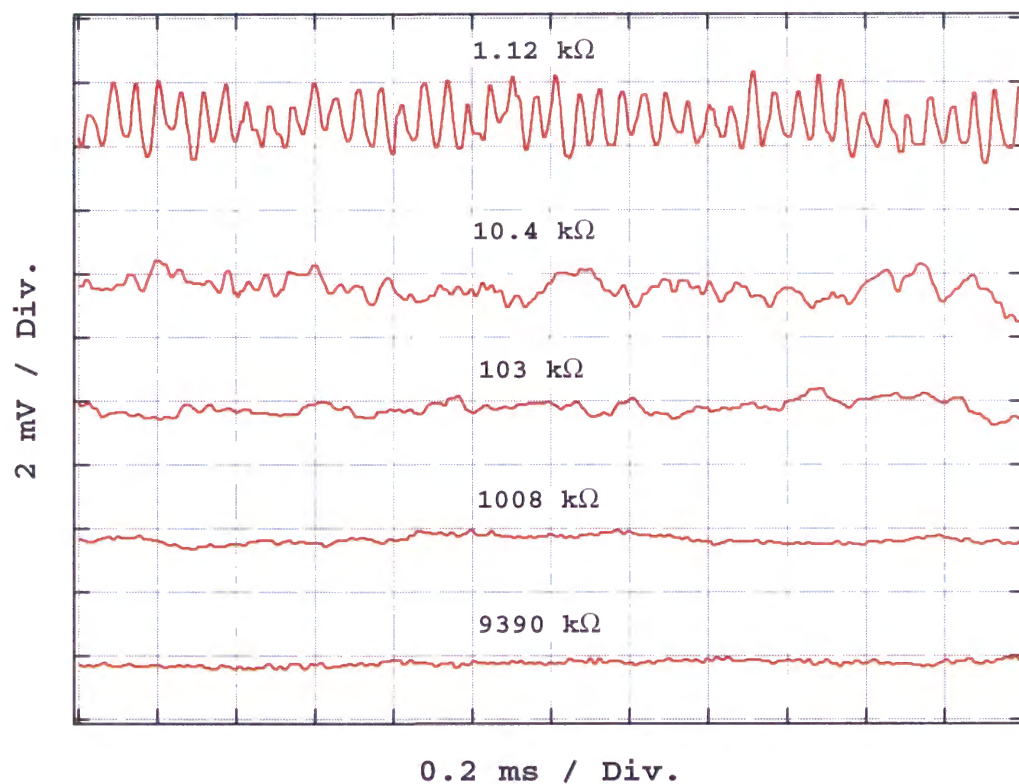


Fig. 2.4. Signals of thermal noises, which were observed with various resistance values connected in series between the preamplifiers PA1 and PA2, *i.e.*, 1.12, 10.4, 103, 1008 and 9390 kΩ.

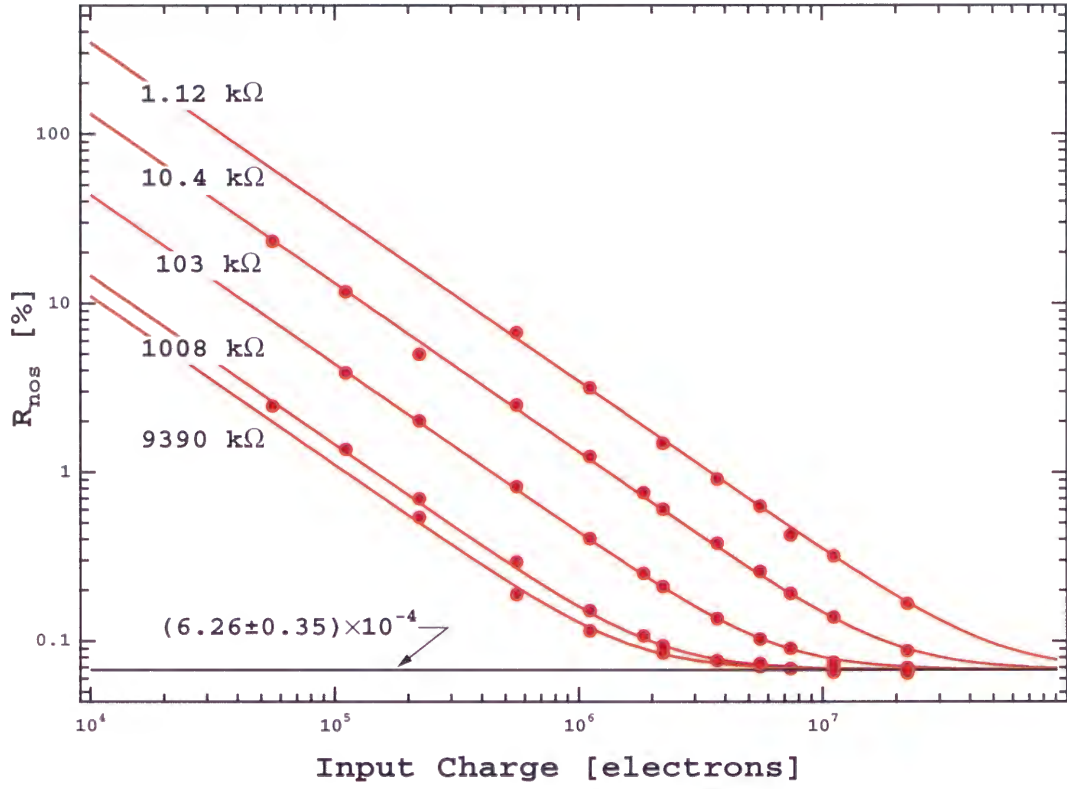


Fig. 2.5. The resolution R_{nos} measured as a function of the input charge Q fed to the system through the arrangement shown by Fig. 2.6; the resistance values connected in series between the preamplifiers PA1 and PA2 are 1.12, 10.4, 103, 1008 and 9390 $k\Omega$.

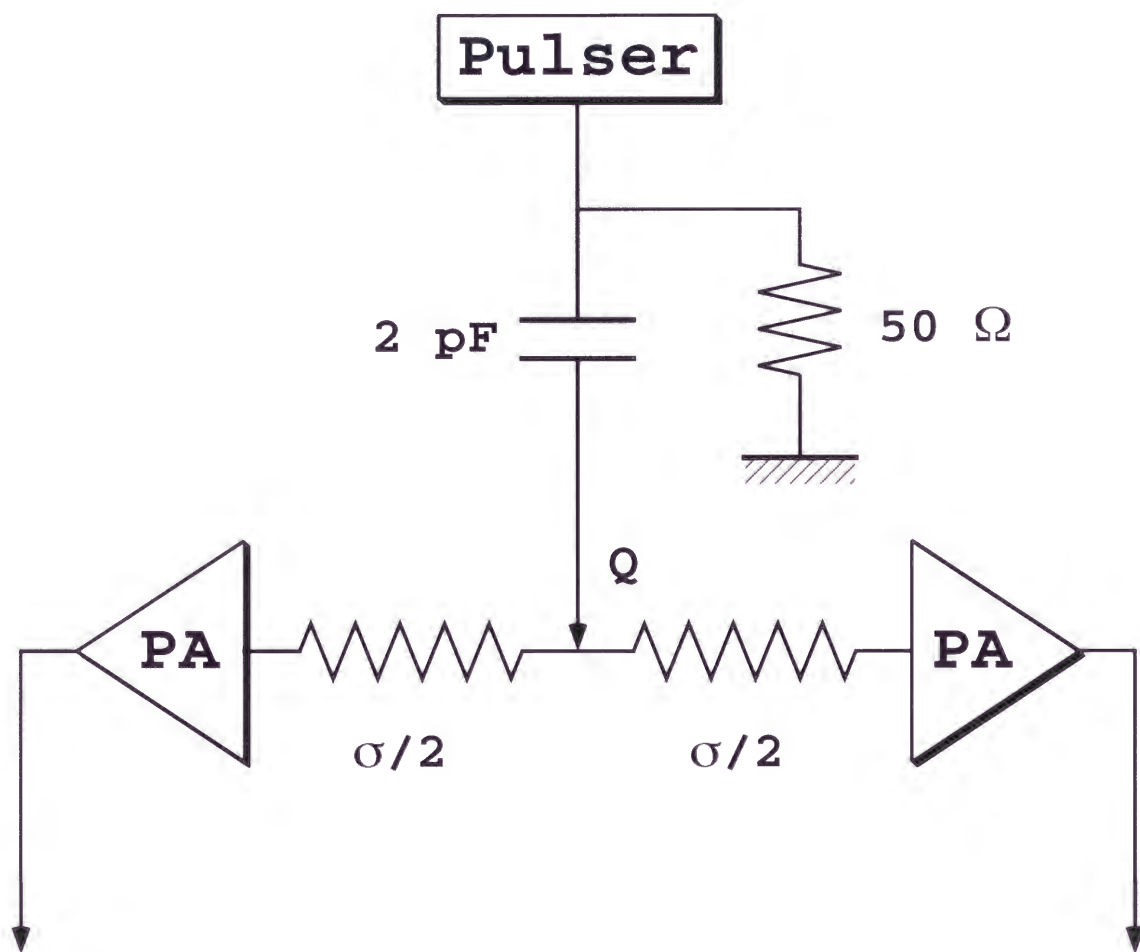


Fig. 2.6. Arrangement for measuring the dependence of the resolution R_{nos} on the input charge Q and the resistance σ .

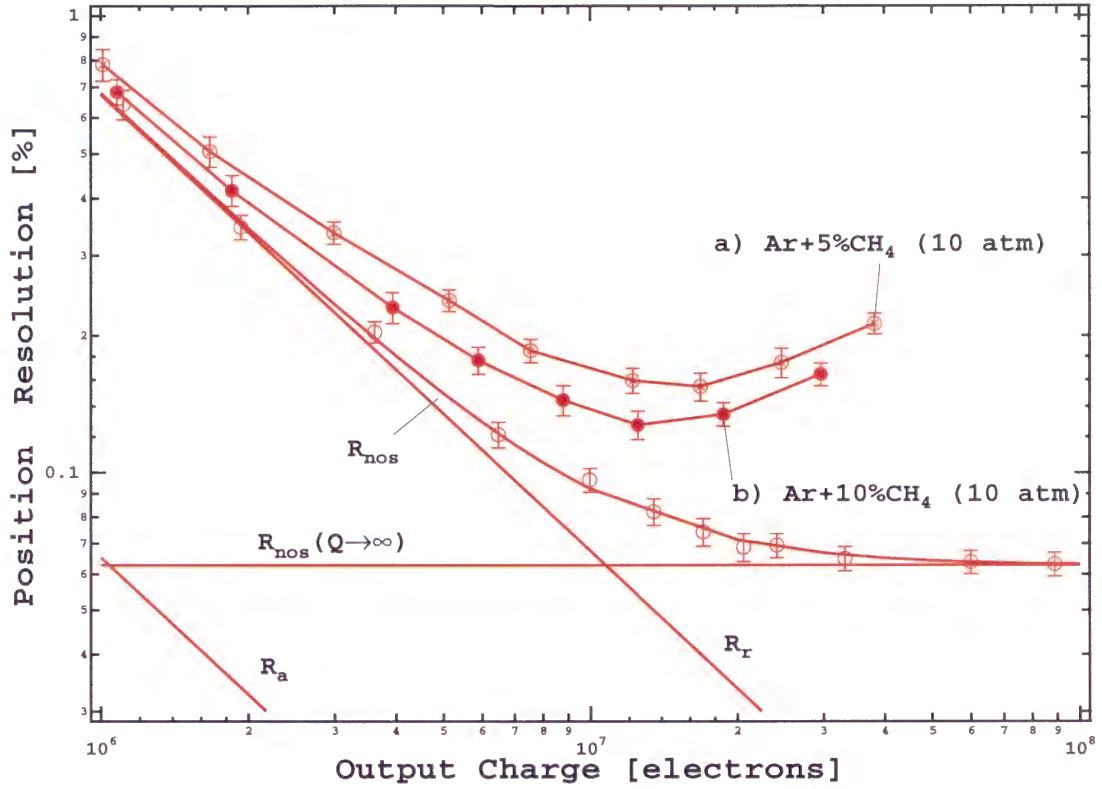


Fig. 2.7. Theoretical and experimental results of position resolutions as a function of the output charge Q from the PSPC system: $\sigma=42 \text{ k}\Omega$, $X_0/L=0.5$ and $V_s=4 \text{ V}$. The resolution R_{nos} coming from various noises are theoretically separated into partial resolutions: R_r , the thermal noise of the carbon fiber; R_a , noises of the preamplifier and spectroscopy amplifier system; $R_{nos}(Q \rightarrow \infty)$, noises of the sum inverter and analog divider. Experimental R_{nos} denoted by open circles were obtained by the same arrangement as Fig. 2.6 with a resistor of $\sigma=42 \text{ k}\Omega$. The position resolution measured for 6.4-keV X rays are given by closed circles: **a**, Ar+5%CH₄ (10 atm); **b**, Ar+10%CH₄ (10 atm).

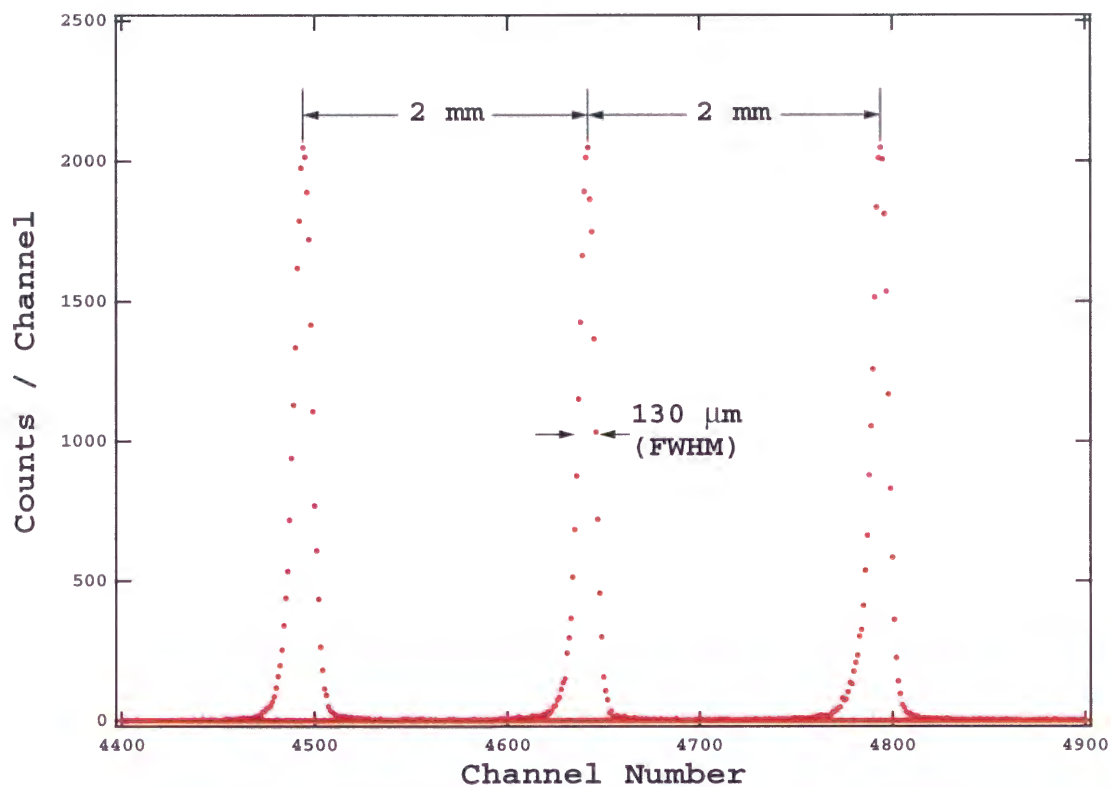


Fig. 2.8. The best position spectrum obtained by the present PSPC system: the resistance of anode, 42 k Ω ; filling gas, Ar+10%CH₄ (10 atm); photon energy, 6.4 keV.

Table 2.1. Fitted values of free aparameters α and β .

σ_0 (k Ω)	α (eletrons)	β ($\times 10^{-4}$)
1.12	14900 $\pm$ 900	2.2 $\pm$ 0.7
10.4	5650 $\pm$ 400	2.70 $\pm$ 0.15
103	1860 $\pm$ 150	2.69 $\pm$ 0.08
1008	620 $\pm$ 30	2.77 $\pm$ 0.05
9390	460 $\pm$ 50	2.95 $\pm$ 0.07

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Chapter 3

Observation of Cl K X rays from ^{35}S Source with a High-Resolution Crystal Spectrometer

The contents of this chapter are based on the following two papers:

“A simple crystal spectrometer for low-energy photons emitted from radioactive sources, Part II”

D. Ohsawa, R. Katano, Y. Ito and Y. Isozumi.

Nucl. Instrum. Methods A 390 (1997) 183.

“K-Shell Internal Ionization and Excitation in β Decay of ^{35}S ”

D. Ohsawa, R. Katano and Y. Isozumi.

Phys. Rev. C in press

3.1. Introduction

A crystal spectrometer of wavelength dispersive type can provide an energy resolution better than 10 eV for low energy photons. The spectrometer is usually operated by the step-scanning method, in which the diffraction angle of crystal is changed step by step and the photon counter is rotated by twice the angle of crystal in each step. Spectrometers of this type are less suited to analyze photons from radioactive sources because of their very low detection efficiencies. Position sensitive detectors (PSD) are often used to obtain photon diffraction spectra without the step-scanning procedure. The detection efficiency of such a spectrometer combined with a PSD is considerably better than that of the ordinary spectrometer. In the previous work [1], we fabricated a flat-crystal spectrometer combined with a position-sensitive proportional counter (PSPC), which is available for analyzing photons emitted from radioactive sources in the energy region of 5~15 keV.

A new flat crystal spectrometer has recently been constructed specially for analyzing photons with energies less than 5 keV. A main purpose of the new spectrometer is to search for faint low energy X rays buried in the large background caused by other radiations from radioactive sources, *e.g.*, higher energy photons, β particles and internal conversion electrons. In this chapter, we describe the performance of the new spectrometer in detail as well as contrivances to eliminate some problems in detecting such low energy photons, *e.g.*, the absorption of photons in air and the high background counts caused by other radiations from the source. An application of the present spectrometer to the observation of an interesting phenomenon, *i.e.*, the K-shell internal ionization and excitation during the β decay of ^{35}S , is also described. In the previous measurement by proportional-counter pulse-height analysis, the photon spectra obtained consist of a superposition of Cl and S K X rays because of the poor energy resolution of the proportional counter. Using the present crystal spectrometer, we have for the first time succeeded to observe the Cl K X rays completely resolved from the S K X rays. The transition probability P_K for the β decay of ^{35}S can be determined with the measured intensities of Cl K X rays and β rays and the K-shell fluorescence yield of chlorine. Details of the determination of P_K are described in the next chapter.

3.2. Apparatus

The geometrical arrangement of counter, crystal and source assemblies in the present spectrometer is schematically shown in Fig. 3.1. Most parts of the spectrometer are made of low-Z materials such as Lucite and aluminium in order to suppress undesired secondary radiations created by the collision of primary radiations from the source with spectrometer materials. The position-sensitive proportional counter (PSPC) is fixed on an aluminium plate, which can slide on an aluminium guide rail (G1) to roughly adjust the distance between crystal (CY) and PSPC.

Two crystals are used for photon diffraction according to the energy range to be analyzed. One is a flat LiF(220) with interplanar spacing $2d$ of 2.848 Å and a crystal size of $50 \times 20 \times 3.5 \text{ mm}^3$, which is adequate for the energy range of 4~12 keV. The other is a flat Ge(111) with the spacing of 6.533 Å and a size of $50 \times 20 \times 5 \text{ mm}^3$, which is available for the range of 2~4 keV. The crystal is installed on the x-axis stage (XS) at the center of turntable to make the center of the crystal surface coincide with the rotational axis of turntable. The crystal and PSPC assemblies are set on the goniometers RS1 and RS2, respectively, and independently rotated around the center of the crystal.

The source assembly consists of three components: radioactive source (S), dump magnet (M) and wedge-shaped collimator with rectangular apertures. This collimator is made of an wedge-shaped lead block attached to a 1-mm thick Lucite plate. The lead block is used to absorb high energy photons while the Lucite plate is necessary to absorb low energy secondaries created by the collision of high energy photons with the lead plate. The aperture of the source side of the collimator is $2 \times 12 \text{ mm}^2$ while that of the crystal side is $18 \times 12 \text{ mm}^2$. The backing material of the source is a carbon strip of 1-mm width and 20-mm length. The radioactive area of the strip is limited to a rectangle of $1 \text{ mm} \times 3 \text{ mm}$. The source strip is attached to a holder (SH) made of 2-mm thick Lucite plate. The source assembly can slide on an aluminium guide rail (G2) to adjust the distance between source and crystal. The rail is fixed on the z-axis stage (ZS) for fine adjustment of z direction of the source assembly.

The PSPC is rotated around the center of the crystal in the

θ -2 θ mode, where the rotation angle of the PSPC is precisely equal to twice that of crystal itself. The range of the source-crystal and crystal-counter distances are from 50 to 75 mm and from 45 to 75 mm, respectively. The detection efficiency of the crystal spectrometer for photons emitted from the source is inherently very low. To obtain better efficiency, both source-crystal and crystal-counter distances can be set to be 50 and 45 mm, respectively, which is the shortest arrangement for the present spectrometer. The whole spectrometer is put into a vacuum-proof desiccator box made of Lucite. To improve the signal-to-noise ratio of the PSPC outputs by eliminating surrounding hums, the desiccator box is mounted inside a small shielding room made of 3-mm thick copper plates.

The anode wire of the PSPC is a carbon fiber of 7- μ m diameter with a resistivity of 4 k Ω /cm (Toray Co., Ltd.). The performance of this fiber as a resistive anode was examined in detail in our previous work [2] and by others [3-5]. The entrance window is a polypropylene film 6.0 μ m in thickness, while the exit window is a polypropylene film 100 μ m in thickness. In order to make the cathode surface electrically conductive, an aluminium layer of about 300-Å thickness is deposited on the polypropylene films by vacuum evaporation. The length of both windows is 60 mm and the width of them is 15 mm. The sensitive thickness of PSPC is 10 mm; its frame is made of a stainless steel plate with the same thickness. A gas mixture of Ar+30%CH₄ is flowed through the PSPC under a little higher pressure than atmosphere (1.10 atm). The incident position of photons on the PSPC is deduced by the ordinary charge-division method, in which the position is estimated as a ratio of the height of output from one end of the resistive anode to the sum of those from both ends of the anode [6-9].

3.3. Source preparation

Three different sources of ⁵⁷Co, ¹⁰⁹Cd and ³⁵S were used: ⁵⁷Co was for the examination of the performance of the present spectrometer while ¹⁰⁹Cd and ³⁵S were for the determination of the K-hole creation probability in the β decay of ³⁵S, P_K.

The original ⁵⁷Co solution with the activity of 37 MBq (Amersham International Plc., England) was carefully purified by the anion exchange resin technique with HCl elution. Then, the chemical form

of the purified ^{57}Co was changed from chloride to nitride. A ^{57}Co source on a 20- μm thick aluminum foil was made by the molecular plating method [10-12]. With some devices for the molecular plating, the radioactive area was confined to a rectangle of 1.0 x 8.0 mm^2 . The source activity was measured to be 19 MBq.

The chemical form of carrier-free ^{35}S purchased from ARC (American Radiolabeled Chemicals, Inc., USA) is Na_2SO_4 in aqueous solution; its specific activity is about 3.7 GBq/ml. Chlorine ions in the original solution was removed by the micro-resin column technique [13,14]; the resin used is Dowex 1- Cl^- and the column size is 0.8-mm diameter x 5-mm length. Successive treatments for the purification of ^{35}S are as follows; (1) 5 ml of 2 N 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 0.01 N HNO_3 to remove away chlorine ions from the resin and (4) a few drops of 2 N HNO_3 to take up the β activity. More than 95 % of the activity was contained in 5 drops (~150 μl) at the last step. No visible residue was seen after adding 10 μl of 0.005 N 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 x 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 polyethylene (CH_2CH_2) 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-run X-ray measurements (45 days at the longest).

The chemical form of ^{109}Cd purchased from NEN (New England Nuclear, USA) is CdCl_2 in 0.1 N HCl (1 ml); its specific activity is about 37 GBq/mg. The original solution with the activity of 37 MBq was purified with a Dowex 50W- H^+ microcolumn (0.8-mm diameter x 5-mm length); cadmium ions was eluted with 1 N HCl (1 μl). More than 80 % of the activity was taken up in 10 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 ^{35}S source.

3.4. Performance of crystal spectrometer

3.4.1. Detection of low-energy photons

The total trajectory of photons from source to counter is 140 mm in the present spectrometer. A serious problem in detecting low energy electrons comes from the absorption of photons in air. A curve (a) in Fig. 3.2 is the relative absorption estimated for the 140-mm path of air as a function of photon energy; the relative absorption is defined by a ratio of the number of photons absorbed in the path to that of incident photons. As seen in the curve (a), the absorption in the air is 7.46% for 10-keV photons and gradually increases with decreasing photon energies.

It was practically difficult to keep a vacuum better than 0.1 Torr inside the desiccator box of the present spectrometer mainly because of the out-gas from Lucite materials. Electric discharges take place in such poor vacuum when the anode voltage is higher than 1300 V. To avoid this discharge, which always destroys a field-effect transistor at the head stage of preamplifier, hydrogen gas was flowed through the desiccator box at a rate of 5~10 cc/min under a gas pressure of 1 atm. The flow of hydrogen gas can suppress the discharge even at an anode voltage of 1800 V, which is high enough to detect photons with energies lower than 3 keV. Helium is less useful than hydrogen to suppress the electric discharge; the discharge takes place in 1-atm helium at an anode voltage of 1550 V.

The counter gas flowed in the PSPC is a gas mixture of Ar+30%CH₄. The gas pressure in the PSPC is kept to 1.10 atm., while the pressure of hydrogen gas in the desiccator is 1.0 atm. Such a small difference in the gas pressures was necessary to suppress the fluctuation of effective thickness of the PSPC by making the window film distended to the outer side. The fluctuations in the gas pressure of the counter gas were kept below 1 % by a hand-made gas-pressure controller in a series of X-ray measurements.

The relative absorption estimated for the 140-mm path of hydrogen as a function of photon energy is given by a curve (b) in Fig. 3.2. It is noted that the flow of hydrogen is inevitable in the present measurement for photons with energies below 3 keV; the relative absorption for a 3-keV photon is 0.16% in hydrogen, while that is 90.5% in air.

3.4.2. Rejection of background electrons

A large number of electrons are emitted from radioactive sources, e.g., β particles from β -active nuclides, Auger electrons competing with the emission of characteristic X rays, conversion electrons competing with the γ ray emission. Apart of these electrons can arrive at the PSPC after being scattered by the crystal and other surrounding materials, increasing background counts in position spectra of low energy photons. As seen in Fig. 3.1, a magnet is installed in the present spectrometer in order to separate the electrons from photons; the electrons passing through the magnet are deflected by the strong magnetic field (~ 1 kGauss).

The effect of the magnet was examined by measuring photons emitted from the ^{109}Cd source. The position spectra given by Figs. 3.3(a) and (b) were obtained with and without the magnet, respectively. Background counts are decreased more than two orders of magnitude, indicating that the magnet is inevitable to observe low energy photons from radioactive sources. Two broad bumps appearing at both sides of the spectra are caused by the end effect of the PSPC, as explained below.

3.4.3. End effect of the PSPC

As seen in Fig. 3.3(a), there are sharp peaks at both ends of the spectrum for Ag L X rays emitted from ^{109}Cd . Instead of such peaks, there are small and broad bumps in the spectrum for Fe K X rays emitted from ^{57}Co . Peaks or bumps always appear at the ends of position spectra when low energy photons are measured in a large background of radiations with higher energies. We found that the intensity of the peak or bump strongly depends on the energy range of background radiations, as shown in Fig. 3.4. In Fig. 3.4(a) is given the energy spectrum for photons emitted from a ^{35}S source, which was obtained by summing outputs from both ends of the resistive anode. In Fig. 3.4(b) are given position spectra corresponding to the five energy regions A, B, C, D and E, which are indicated in the energy spectrum of Fig. 3.4(a). It is clear that the intensity of the bump in the position spectra becomes larger when the corresponding energy region is decreased.

These spurious peaks or bumps at the ends of position spectra are caused by the distortion of the electric field near the ends of the anode wire of the PSPC. The terminals of the anode inside the PSPC are guide tubes of 0.8 mm diameter and 3 mm length. Since the electric field on these terminals is much weaker than that on the anode wire, the gas gain near the ends of anode wire is smaller than the gain on the central part of anode wire. In the terminal regions, pairs of electron and positive ions produced by the ionization process of incident radiations can more easily recombine with each other. Although most of electrons produced in the terminal regions are eliminated by the recombination effect, a part of them are attracted to the ends of the anode wire. Because of the decreased gas gain and the recombination effect, output charges from the ends of anode wire are smaller than those from the central part of anode wire. Thus, events in the bumps are caused by higher energy radiations incident in the terminal region of the anode; it is noted that these events contribute to the position spectra corresponding to regions much lower than that of the incident energy of radiation.

It is difficult to remove the spurious peaks or bumps completely by improving the structure of the inside of PSPC. A way to remove them is to make a PSPC with a size long enough to divide a position spectrum into end parts to be taken off and the central part to be employed as a normal spectrum.

3.4.4. Discussions

In the present experiment for observing the K-IIE process during β decay, the 2.62-keV Cl K X rays to be measured are completely buried in the large background of high energy radiations. The selective detection of photons with energies less than 3 keV is effective in decreasing more the background in the position spectrum. The mixture of Ar+30%CH₄ employed in the present work is not suitable for such detection of low energy photons; since the K-shell absorption edge of argon is 3.21 keV, the detection efficiency of the counter filled with argon mixtures discontinuously increases at this energy and gradually decreases in the higher energy region. Gas systems with an efficiency uniformly decreasing in the higher energy region are more useful for the selective detection. The organic gases of CH₄, C₂H₆, C₃H₈ and C₄H₁₀ are possible candidates for that purpose because

they have no absorption edge except that of carbon at 285 eV.

Charge-sensitive preamplifiers used in the present work are commercially available, designed specially for solid state detectors with rather large input capacitances. Preamplifiers of this type cannot response correctly to pulse signals from the anode with a resistance as low as that of the present PSPC (24 k Ω), because the open-loop gain in the feed-back part is not large enough, *i.e.*, a factor several thousands. It is necessary to develop a preamplifier optimized for the PSPC of charge-division type, which has an anode of relatively small resistance in the range of 10~100 k Ω .

The resolution of the present spectrometer is considerably better than that of the high-resolution nuclear radiation detector for low energy photons, *i.e.*, Si(Li) detector. For example, the resolution for 3-keV photons is 26.9 eV by the present spectrometer and at best 80 eV for the Si(Li) detector. One of difficulties in employing the crystal spectrometer is the preparation of radioactive sources. As shown in this chapter, a source intensity of 10~150 MBq is necessary to obtain a spectrum with enough statistics in a permissible period, *i.e.*, at least a few weeks; this intensity is three orders magnitude stronger than that for ordinary radiation detectors. In spite of this shortage, we believe that the present compact spectrometer can be a powerful tool for the high-resolution detection of low energy photons from radioactive sources until the advent of superconducting radiation detectors in the near future, which are promised to have an energy resolution much better than that of presently available detectors [15-18].

3.5. High-resolution measurements of low-energy photons

3.5.1. X rays from ^{35}S sources

In Fig. 3.5 are given the spectra obtained with 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 $\mu\text{g}/100$ μl solution). Energies corresponding to 2.622-keV Cl $K\alpha_{1,2}$ X rays and 2.308-keV S $K\alpha_{1,2}$ X rays are indicated by arrows in the figure. In order to observe the Cl and S K X rays in the same measured spectrum,

the diffraction angle at the center of Ge crystal, θ_c , was set to 48.4 degree and the source-crystal and crystal-PSPC distances were set to be 50 mm and 45 mm, respectively, which are the shortest arrangement for the present spectrometer. It is seen that the separation of Cl K X rays from S K X rays is sufficient even with such the shortest arrangement. There is a faint bump at the position of S K $\alpha_{1,2}$ X ray in Fig. 3.5(a), which is caused by the self-absorption of β particle in the carrier-free ^{35}S source. This bump became larger when a more intense ^{35}S source was used in the measurement. It is noted that a small amount of S atoms in the source is enough to absorb almost completely the Cl K X rays, as seen in Fig. 3.5(b)

As seen in Fig. 3.5(a) and (b), the background continuum in position spectra is rather large. Energy spectra of photons observed with the PSPC are given by Fig. 3.6. The spectra of Fig. 3.6(a) and (b) were obtained with and without the ^{35}S source, respectively; the regions A in Fig. 3.6(a) was used to make the position spectra (a) and (b) in Fig. 3.5. The large peak in higher energy side of Fig. 3.6(a) is caused by 9.886-keV Ge K X rays while the small peak in lower side is caused by 1.188-keV Ge L X rays. These Ge X rays are emitted through the photo-electric effect of photons at the Ge(111) crystal; most of photons are 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 can not arrive at the crystal because of the strong magnetic field in front of the source. The background spectrum caused by natural radiations is given by Fig. 3.6(b). Now, it is seen that the background continuum in the position spectra given by Fig. 3.5(a) and (b) mainly comes from the Ge X rays emitted from the crystal.

The best-resolution spectrum of Cl K X rays emitted from the purified ^{35}S source is given by Fig. 3.7(a). The Cl 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 degree and both distances from source to crystal and from crystal to PSPC were set to 75 mm, which are the longest arrangement for the present spectrometer. The source intensity was about 370 MBq and the measuring period was 45 days. The spectrum contains several lines; most of these lines may be buried in the large peak of ordinary Cl K $\alpha_{1,2}$ X rays in the spectra (a) in

Fig. 3.5, which was obtained with an un-purified ^{35}S source. Thus, the procedure to remove Cl ions from the original solution is essentially important in the present experiment.

The spectrum (b) in Fig. 3.7 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 the spectrum (a). This spectrum mainly consists of ordinary Cl $K\alpha_{1,2}$ X rays (2.622 keV) and Cl $K\beta_{1,2}$ X rays (2.816 keV), which are emitted by the collision of β particles with Cl atoms in the vinylidene chloride film. The energy of one of peaks in Fig. 3.7(a), i.e., P2 line, coincides with that the Cl $K\alpha_{1,2}$ X rays in Fig. 3.7(b).

3.5.2. X rays from ^{109}Cd sources

The position spectra of photons emitted from the purified ^{109}Cd source are shown in Fig. 3.8. A main peak at the left side of the spectrum is caused by the 2.984-keV Ag $L\alpha_1$ line, which was used to calibrate the detection efficiency of the PSPC for 2.622-keV Cl $K\alpha_{1,2}$ X ray, as explained later. 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 these of Ag $L\alpha_{1,2}$ and Ag $L\beta_1$, because of the steep increase of the detection efficiency of PSPC at the K edge of argon (3.206 keV); the counter gas of PSPC is the gas mixture of Ar+30%CH₄.

From the shape of $L\alpha_{1,2}$ peak, the half width of this peak is estimated to be 1.12 ± 0.01 mm in the unit of length on the PSPC. According to Eq. (A.6) in Ref. 1, the width of 1.12 ± 0.01 mm corresponds to 26.9 ± 0.2 eV in energy for the present spectrometer. The observed width of 1.12 mm is a little larger than the present source width of 1.0 mm. Partial widths coming from the crystal imperfection of Ge(111) and the natural line width of $L\alpha_{1,2}$ peak are estimated to be 0.24 mm and 0.084 mm, respectively, which are relatively small compared with the source width.

There are two other origins of the decrease of resolution in the present spectrometer. One is the position resolution of PSPC caused by the spatial spread of electron avalanches near the anode wire and the thermal noise of the resistive anode; the resolution of the PSPC with the 7- μm diameter carbon fiber was measured to be 0.25~0.40 mm depending on the electronics and the counter gas employed [19]. The other origin is the present geometrical arrangement, where

both source-crystal and crystal-PSPC distances are not long enough, i.e., 75 mm, compared with the width of the Ge(111) crystal, 20 mm. In this case, the incident position of diffracted photons on the surface of PSPC can not be approximated to a straight line perpendicular to the anode wire; the incident position forms an arc of circle corresponding to the diffraction angle of photons. The spread caused by the shape of this arc is roughly estimated to be 0.6 mm in the present case. Thus, the rather poor result for the observed width is mainly caused by the compact arrangement for source, crystal and PSPC in the present spectrometer.

3.5.3. Discussions for the composite spectrum of Cl $K\alpha$ X-ray group

In the present work, we have obtained a 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. 3.7(a). The present observation of the composite spectrum for $K\alpha$ X-ray group strongly indicates that the mechanism of the K-IIE process during β decay is quite different from the inner-shell ionizations and excitations during other nuclear decays such as electron capture, internal conversion, and by the bombardment of light charged particles. Six lines as in Fig. 3.7(b) were assumed for preliminary analysis of the composite spectra of $K\alpha$ X-ray group. The energies are 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-, L- plus M-shell hole states of elements with $Z=9$ to 18 were performed by Maurer and Watson [20]. The energies for P3, P4, P5 and P6 lines in Fig. 3.7(a) 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 Mholes. Thus, the lines P3, P4, P5 and P6 are probably assigned to satellite Cl $K\alpha$ X rays. It is noted that the probability for the multiple ionization and excitation process are relatively large; P2 line corresponding to a single process, while P3, P4, P5 and P6 lines corresponds to the multiple process. The origin of P1 line has not been revealed in the present work; a two-quantum phenomenon such as

the radiative Auger process [21,22] is one of possible candidates. A further detailed analysis for the X-ray group is now in progress.

As discussed later (Sec. 4.2.), the IEE process is caused by the sudden change of nuclear charge during β decay in the model of electron shake-off and shake-up [14]. 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 is 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. The similar high-resolution measurement for higher- Z nuclides are also desired, if possible.

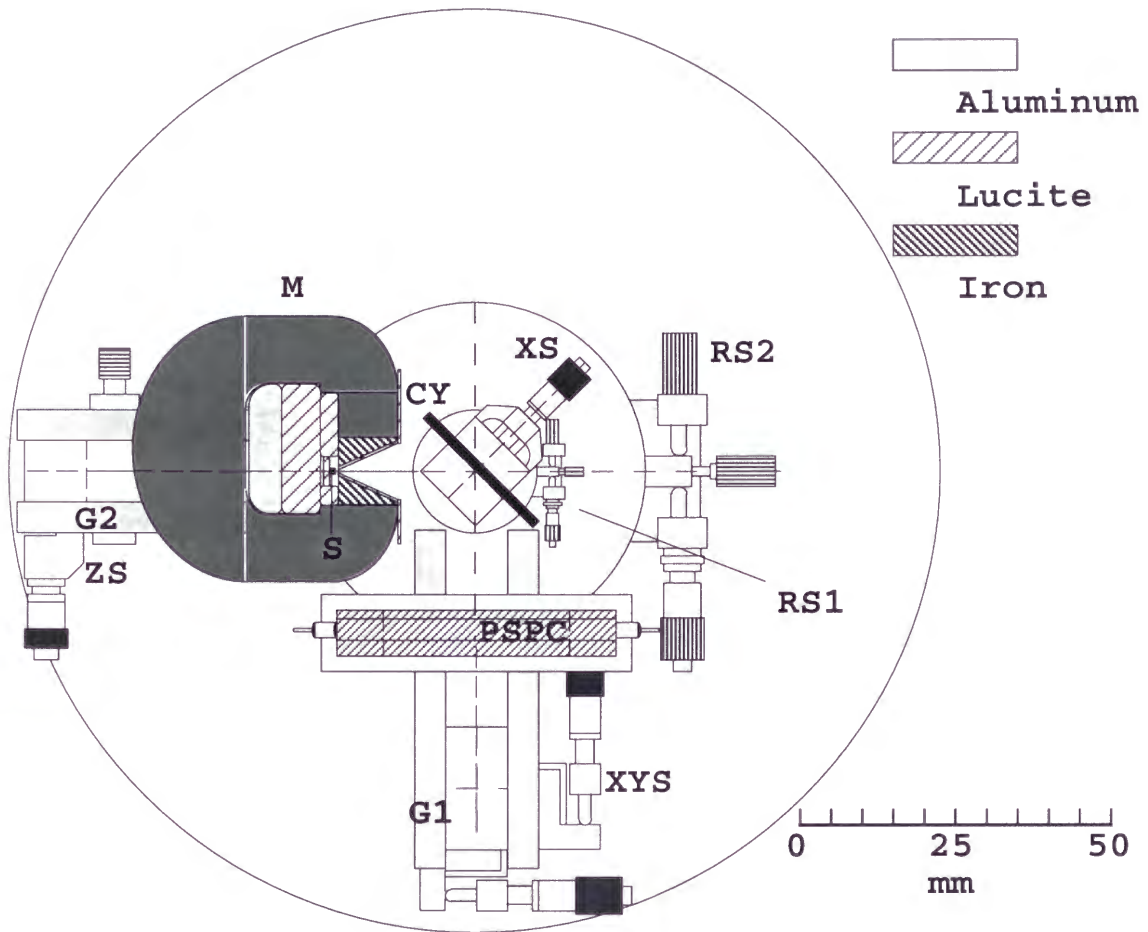


FIG. 3.1. The experimental layout for the X-ray measurement: **PCPC**, position-sensitive proportional counter; **CY**, Ge(111) crystal; **S**, radioactive source; **M**, magnet; **XYS**, xy-axis stage; **XS**, x-axis stage; **ZS**, z-axis stage; **RS1** and **RS2**, rotative stages; **G1** and **G2**, aluminum guide rails.

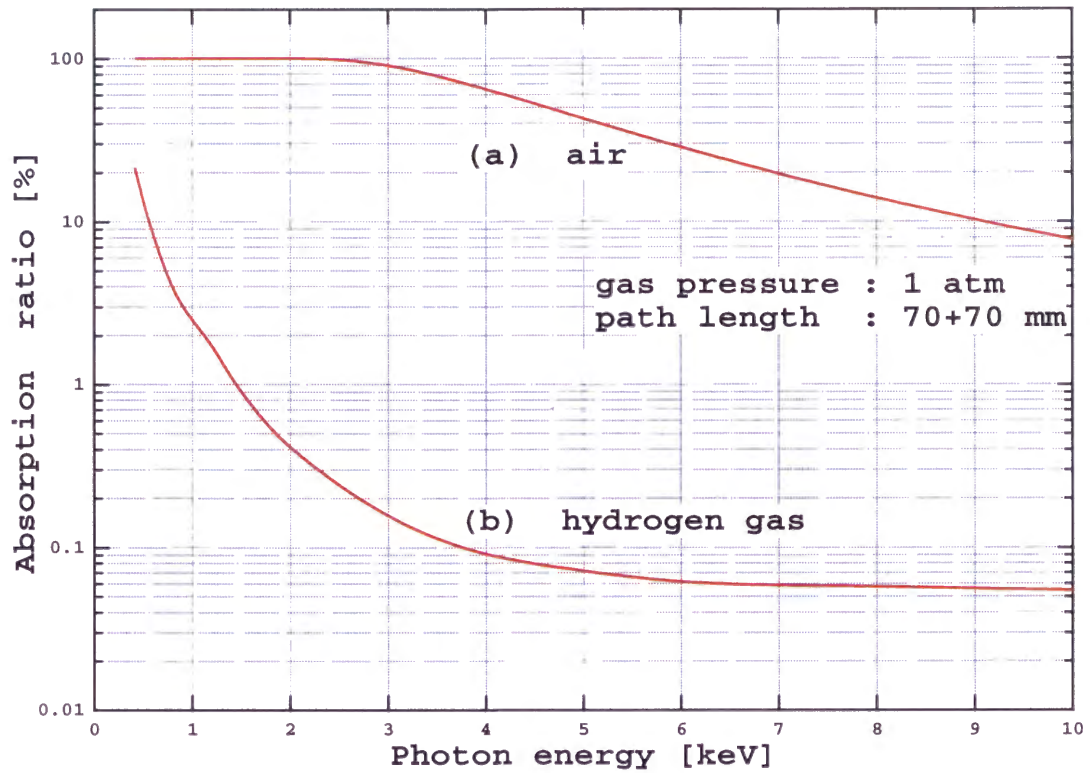


Fig. 3.2. Estimated absorption ratios of photons in the 140 mm path of air and hydrogen gas as a function of photon energy: (a), in air (1 atm); (b), in hydrogen gas (1 atm).

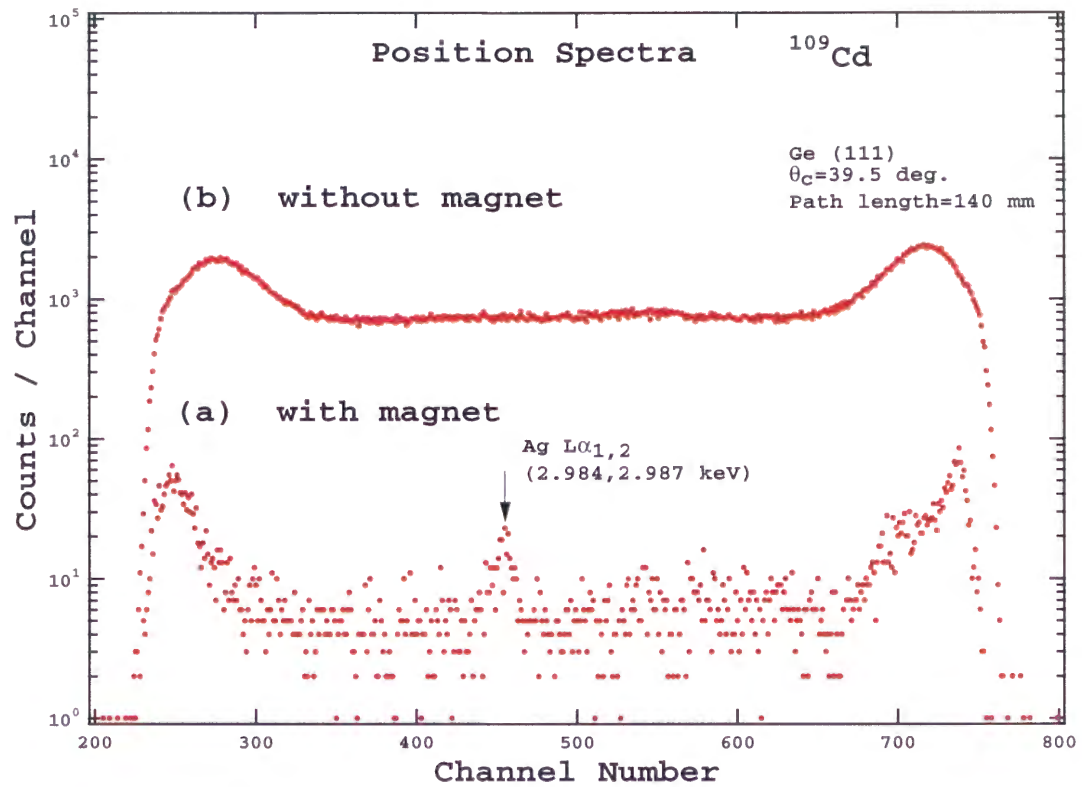


Fig. 3.3. Position spectra of photons emitted from the ^{109}Cd source: (a), without magnet; (b), with magnet. The crystal used is Ge(111) and the diffraction angle at the center of crystal is 39.5° . The source intensity is 32 MBq and the measuring period is about 5 days.

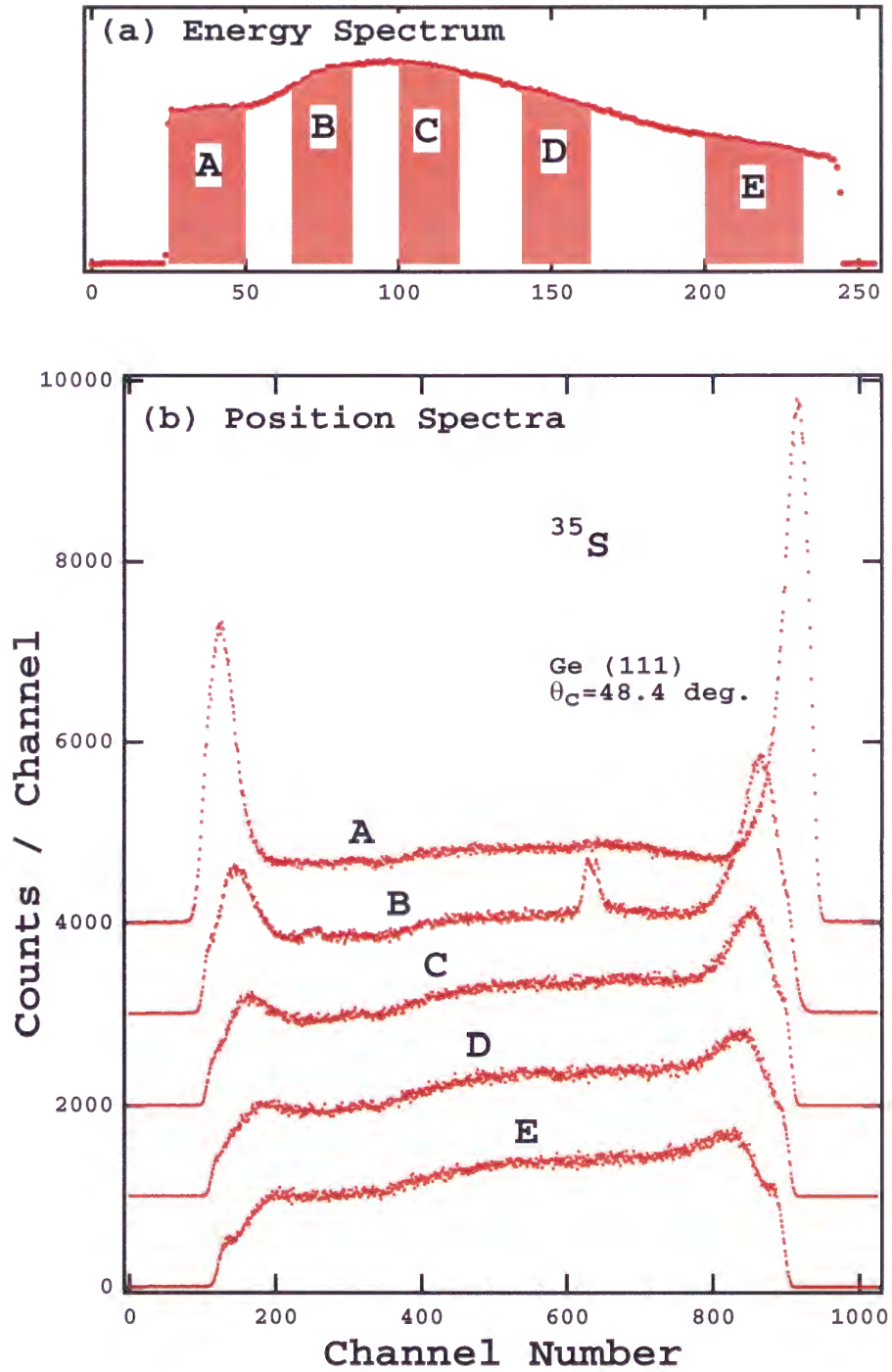


Fig. 3.4. Energy and position spectra obtained for photons emitted from a ^{35}S source: (a), energy spectrum obtained as the sum of outputs from both ends of PSPC; (b) position spectra corresponding to the five regions of A, B, C, D and E in the energy spectra. The crystal used is Ge(111) and the diffraction angle at the center of crystal is 48.4° . The source intensity is 120 MBq and the measuring period is about 14.3 days.

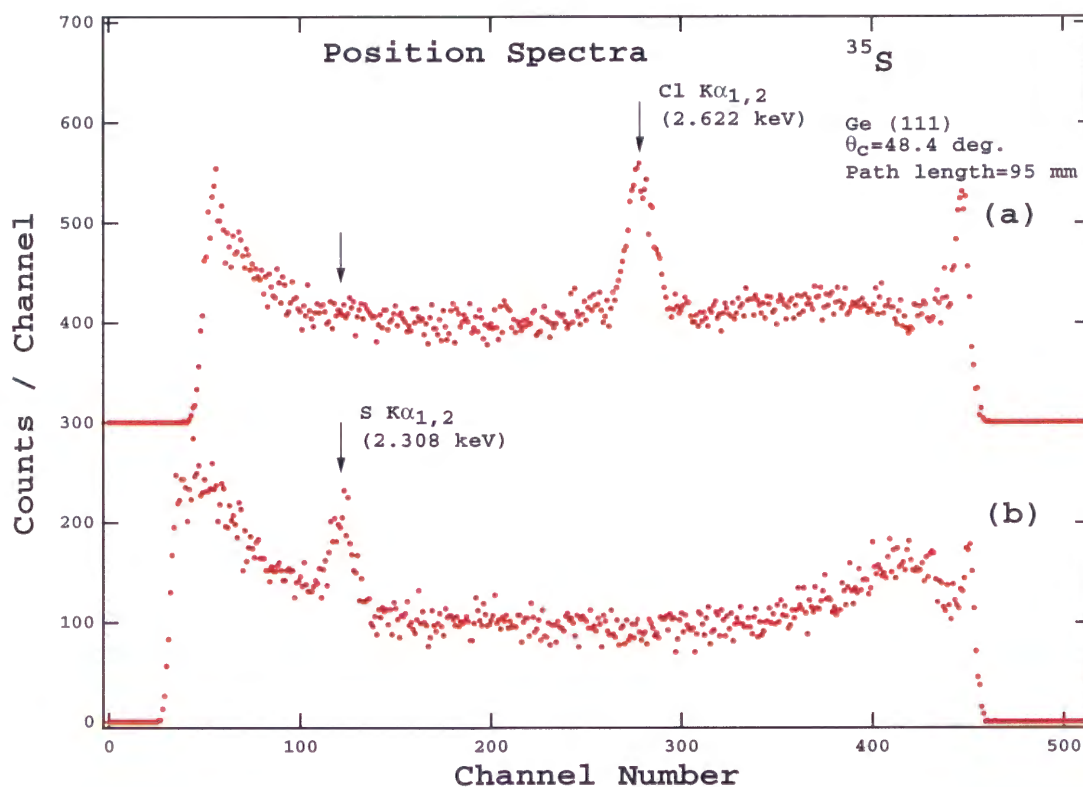


FIG. 3.5. Position spectra of photons emitted from different ^{35}S sources: (a) ^{35}S source prepared from the original solution without the chemical purification; (b) ^{35}S source prepared by containing a small amount of Na_2SO_4 into the original solution.

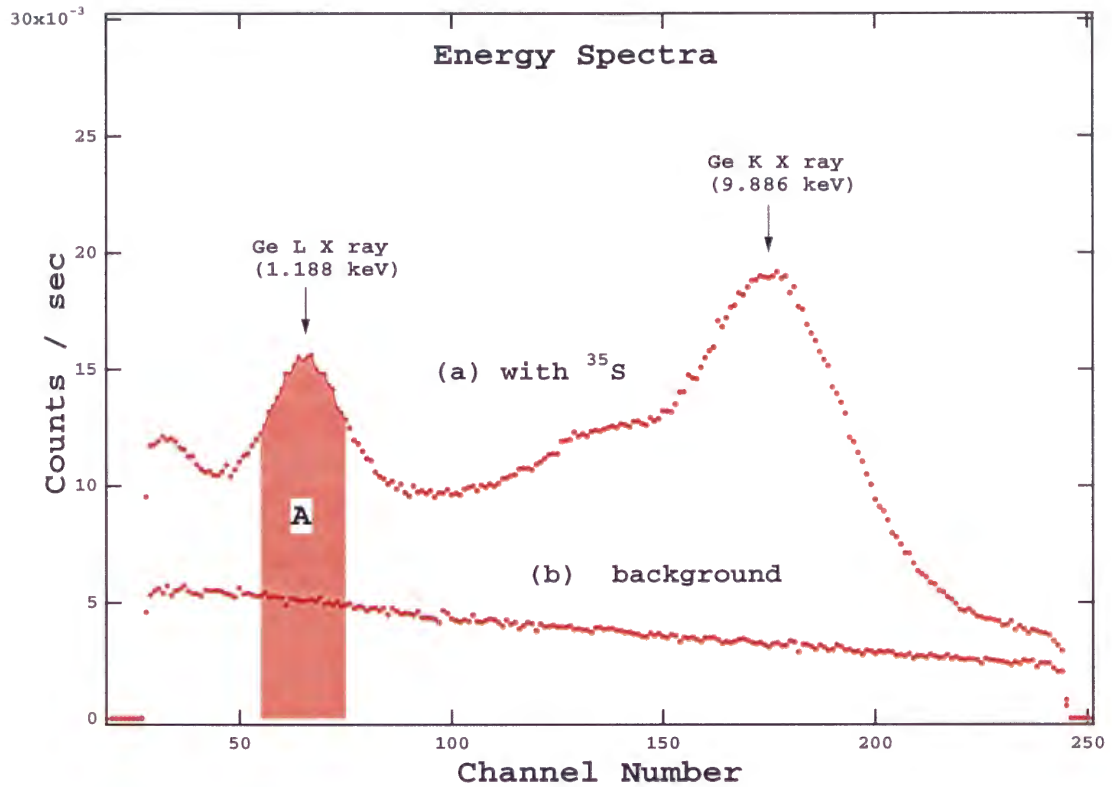


FIG. 3.6. Energy spectra obtained with PSPC: (a), with ^{35}S source; (b), without ^{35}S source. Both spectra were obtained with the same measuring periods (about one week). The linearity in energy scale is not good enough because of the high applied voltage for PSPC (1470 V), which was necessary to obtain the better signal to noise ratio for the position signal of PSPC; the maximum applied voltage to keep the linearity up to 10-keV photon is about 1200 V for the PSPC.

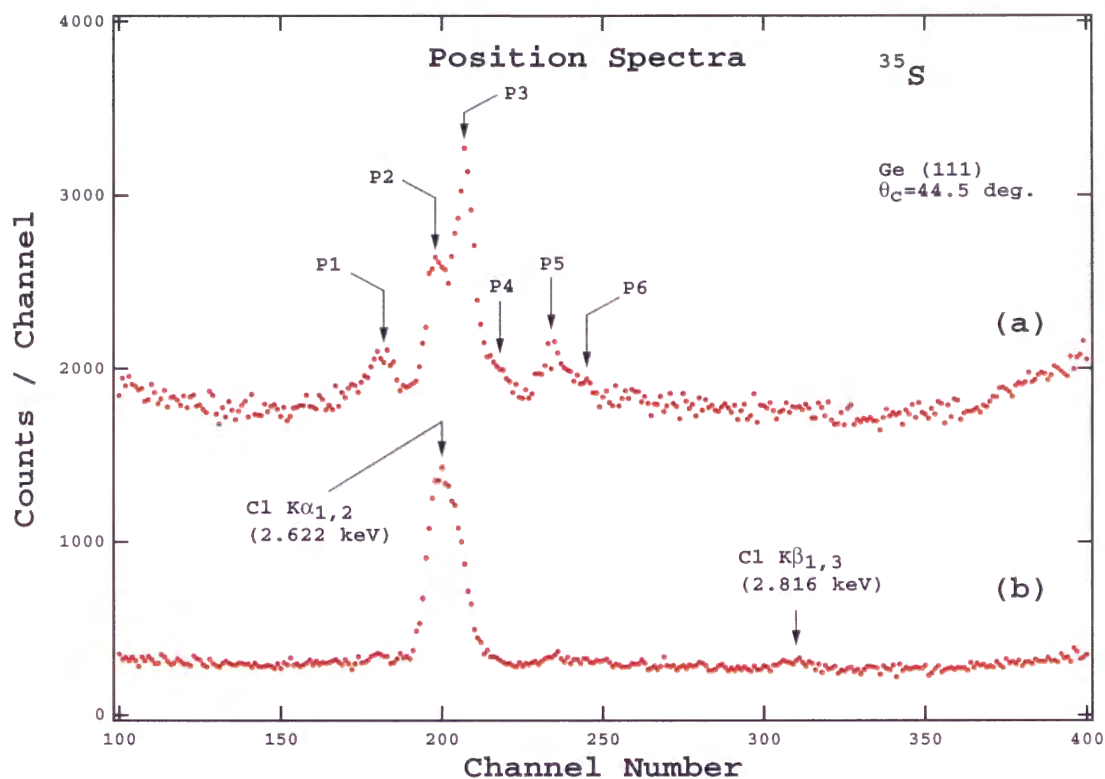


FIG. 3.7. The best-resolution spectra of Cl K X rays emitted from the purified ^{35}S source: (a), covered with a 13.7- μm thick polypropylene film; (b), a 20- μm thick vinylidene chloride film. Arrows P1~P6 in the spectrum (a) indicate the positions of individual lines in the K X-ray group.

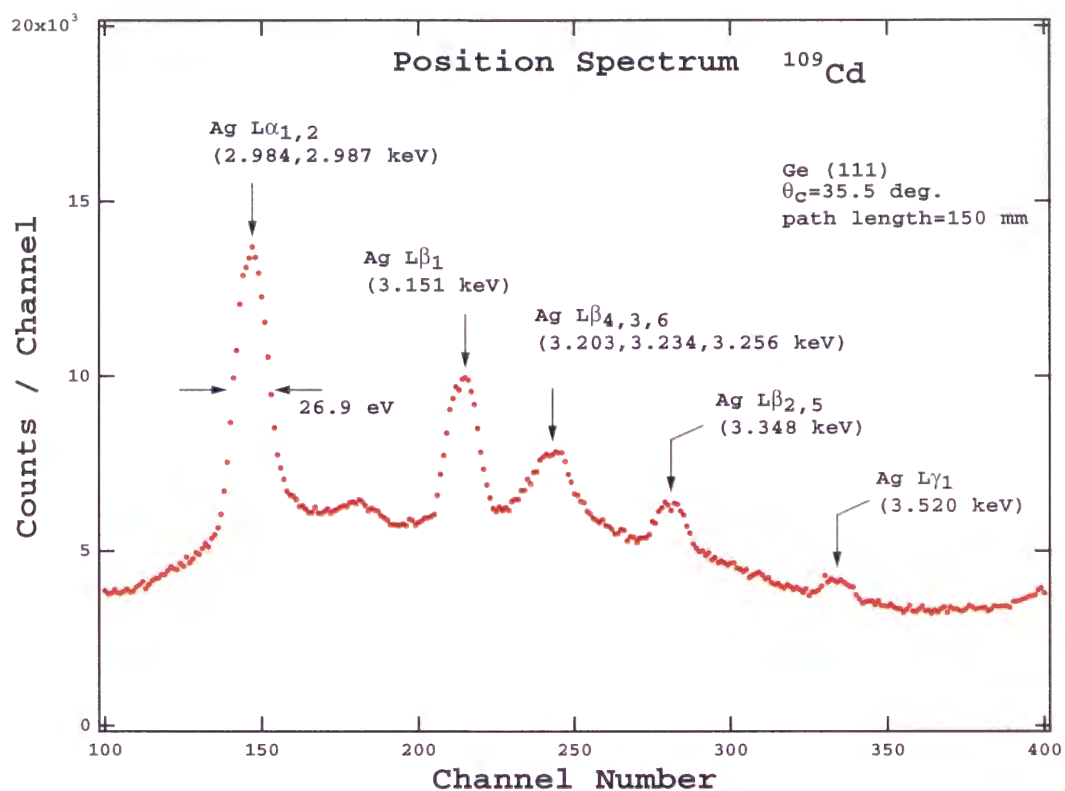


FIG. 3.8. The position spectra of photons emitted from the purified ^{109}Cd source; the Ag $L\alpha_{1,2}$ line in Ag L X-ray group was used to calibrate the overall detection efficiency of PSPC in the X ray spectrometer.

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Chapter 4

Determination of K-Hole Creation Probability per Decay, P_K

The contents of this chapter are based on the following paper:

“K-Shell Internal Ionization and Excitation in β Decay of ^{35}S ”

D. Ohsawa, R. Katano and Y. Isozumi.

Phys. Rev. C in press

4.1. Data analysis

4.1.1. Definitions of X_K and P_K

The X-ray emission probability per decay, X_K , is given by

$$X_K = \frac{N_x}{N_\beta} \cdot \frac{1}{D_x} \quad (1)$$

In the above expression, N_x is the number of 2.622-keV Cl K X rays in the photon spectrum such as Fig. 3.7(a), which was measured with the X-ray crystal spectrometer. The factor N_β is the total number of β particles emitted from the ^{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 the Cl K X ray, which is defined as a ratio of the number of Cl K X rays detected by the PSPC to the number of the total Cl K X rays emitted from the ^{35}S source. Then, the K-hole creation probability per decay, P_K , is defined as

$$P_K = \frac{X_K}{\omega_K}, \quad (2)$$

where ω_K is the K-shell fluorescence yield of chlorine atom.

4.1.2. Ratio N_x/N_β

Two thin ^{35}S sources were prepared in order to determine the ratio N_x/N_β in Eq. (1); the radioactive intensity of the source No. 1 was 58.8 MBq while that of source No. 2 is 93.3 MBq. The geometrical arrangement for the X-ray spectrometer was the same as that for the spectra in Fig. 3.5(a) and (b); θ_c is 48.4 degree and source-crystal and crystal-PSPC distances are shortest, i.e., 50 and 45 mm, respectively. The position spectra of the two ^{35}S source consist of several peaks, as similar to Fig. 3.7(a). The peak area in the position spectra were divided in 6 peaks by the non-linear least square fit [1], in which the shapes of peak and background are assumed to be a Gaussian curve and a straight line, respectively. It is assumed in the present investigation that N_x is given by the sum of each divided peaks. Results of N_x for the sources No. 1 and No. 2 are given in

the forth column of Table 4.1.

The total number of β particles emitted from the ^{35}S source in the period of the X-ray measurement, i.e., N_β in Eq. (1), was determined by the liquid scintillation counter technique. The total number N_β is given by

$$N_\beta = \frac{H_\beta}{D_\beta} \cdot \frac{n_\beta}{a_\beta} \cdot \frac{1}{\eta} \quad (3)$$

In the above equation, H_β is a correction for the decay of ^{35}S in the measuring period:

$$H_\beta = \frac{\exp(\lambda T)}{1 - \exp(-\lambda \Delta T)} \quad (4)$$

where λ is the decay constant of ^{35}S , i.e., $(9.464 \pm 0.008) \times 10^{-8}/\text{sec}$, ΔT is the measuring period of the X-ray measurement, and T is the time interval between the time of the absolute counting of β sources and the start time of the X-ray measurement. The factor D_β in Eq. (3) is the β detection efficiency of the liquid scintillation counter presently used (Beckman LS-6500). The efficiency for this instrument is 0.97 ± 0.01 for ^{14}C ($E_{\text{max}} = 157 \text{ keV}$) and 0.61 ± 0.01 for ^3H ($E_{\text{max}} = 18 \text{ keV}$), respectively, as determined with solutions of these two radioisotopes, which had been previously calibrated by Beckman against identical NBS (National Bureau of Standards) solutions. We independently determined the detection efficiency D_β for β particles from ^{35}S ($E_{\text{max}} = 167.18 \text{ keV}$) using the sulfur-35 reference source prepared by the Du Pont Merck Pharmaceutical Co. The result is 0.960 ± 0.015 , which is consistent with the efficiencies determined by Beckman.

Other factors in Eq. (3), i.e., n_β , a_β and η , are concerned with the measurement of the absolute counting of ^{35}S sources. After the X-ray measurements with the crystal spectrometer, the backing material of β source, i.e., carbon sheet, was immersed in 2 N 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 10 ml scintillation cocktail to measure the β radioactivity. The factor n_β in Eq. (3) is the count rate of the diluted solution obtained by the scintillation counter and a_β is the dilution factor equal to 10^{-6}

in the present measurements. The factor η is the ratio of the activity transferred from the β source to the H_2SO_4 solution, which was determined by the β 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_β and η for each ^{35}S source are listed in Table 4.1.

4.1.3. Overall detection efficiency D_x

The overall detection efficiency of the X-ray spectrometer for the 2.622-keV Cl K X ray, D_x in Eq. (1), was determined by using 2.984-keV Ag L α X ray emitted after the electron capture decay of ^{109}Cd . A thin ^{109}Cd source was prepared for this purpose; its activity is (26.2 ± 1.5) MBq at 19 July 1998. In the present measurement, D_x is given by

$$D_x = \varepsilon \frac{N_{x'}}{N_{x'}^0}, \quad (5)$$

where $N_{x'}$ is the peak counts of the Ag L α X rays in the position spectrum measured with the X-ray spectrometer and $N_{x'}^0$ is the total number of the Ag L α X rays emitted from the ^{109}Cd source in the same period of the X-ray measurement. The source-crystal and crystal-PSPC distances were set to the same as in the measurement for N_x with ^{35}S sources, i.e., 50 and 45 mm, respectively. The ratio of $N_{x'}/N_{x'}^0$ is equal to the overall detection efficiency for the 2.984-keV Ag L α X rays. Therefore, the factor ε in Eq. (5) is necessary to correct the absolute detection efficiency for the energy difference between 2.622-keV Cl K X ray and 2.984-keV Ag L α X ray.

The total number $N_{x'}^0$ in Eq. (5) was determined from

$$N_{x'}^0 = H_{EC} \cdot a_{x'} \cdot R_\gamma \cdot n_{EC}^0, \quad (6)$$

where n_{EC}^0 is the decay rate of the cadmium-109 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 ^{109}Cd source to that from the reference source, $a_{x'}$ is the L α X-ray emission probability per the electron capture decay of ^{109}Cd , which equals to

0.050±0.005 [2]. The factor H_{EC} in Eq. (6) is the correction factor for the decay of ^{109}Cd in the measuring period, of which expression is the same as for the decay of ^{35}S , i.e., Eq. (4); the decay constant λ is $(1.734\pm0.002)\times10^{-8}/\text{sec}$ for the decay of ^{109}Cd .

The factor ε in Eq. (5) is expressed as a product of four correction factors

$$\varepsilon = \varepsilon_1 \cdot \varepsilon_2 \cdot \varepsilon_3 \cdot \varepsilon_4 \quad (7)$$

In the above expression, ε_1 is defined as a ratio of the intrinsic detection efficiency of PSPC for the 2.622-keV photon to that for the 2.984-keV photons. Assuming that the shape of PSPC is rectangular parallelepiped, the approximate intrinsic efficiency can be easily obtained from the simple calculation performed by our previous work [14]. The factor ε_1 was estimated to

$$\varepsilon_1 = 1.33 \pm 0.03 \quad (8)$$

where the error 0.03 comes from an uncertainty in guessing the exact thickness of the gas layer in the PSPC. The factor ε_2 is defined as a ratio of the penetration efficiency of the 2.622-keV photon in the (45+50)-mm pass length of hydrogen gas to that of the 2.984-keV photons. This factor is actually 1.00 according to the present estimation:

$$\varepsilon_2 = 1.00 \pm 0.02 \quad (9)$$

The factor ε_3 is defined as a ratio of the penetration efficiency of the 2.622-keV photon to that of the 2.984-keV photon for the 13.65- μm thick polyethylene (CH_2CH_2) film, with which the ^{35}S and ^{109}Cd sources were covered to avoid the activity contamination. This factor was estimated to be 0.962 using the mass attenuation coefficient for polyethylene:

$$\varepsilon_3 = 0.962 \pm 0.020 \quad (10)$$

The factor ε_4 is defined as a ratio of the intensity of the 2.622-keV photons to that of the 2.984-keV photons diffracted by the Ge(111) crystal. The integrated reflection power for the perfect crystal in

the rotation method is given by

$$P = \frac{\lambda^3}{\sin \theta_c} \propto \frac{1}{E^2}, \quad (11)$$

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 [15]. With the approximation in Eq. (11), the factor ϵ_4 was estimated to be

$$\epsilon_4 = 1.30 \pm 0.03 \quad (12)$$

The determination of D_x with the ^{109}Cd source and that of N_x/N_β with the two ^{35}S sources were alternately performed: the first with ^{109}Cd source, the second with ^{35}S source No. 1, the third with ^{109}Cd source, the fourth with ^{35}S source No. 2 and the fifth with ^{109}Cd source. Results for N_x , N_x^0 and D_x obtained with the ^{109}Cd source are listed in Table 4.2. The values of D_x determined by three runs agrees well each other within the statistical errors in the data of N_x , N_x^0 , indicating that the X-ray spectrometer system worked well constantly in a series of measurements for D_x and N_x/N_β .

4.1.4. Fluorescence yield ω_K

The fluorescence yield ω_K is defined as the probability with which the K-shell vacancy is filled by K X-ray emission processes. As seen in Fig. 3.7(a), the K X rays emitted in the β decay of ^{35}S consist of several components. The ω_K value for each lines is not necessarily the same because the emission mechanism of each line is expected to be different each other as discussed in Sec. IV. There is, however, no way to estimate the ω_K value for each line in the present situation.

Assuming that all the $K\alpha$ lines observed in the present work has the same ω_K value as that for the K X rays emitted from singly ionized atoms, we adopted

$$\omega_K = 0.099 \pm 0.010 \quad (13)$$

as the most reliable value of ω_K for chlorine, which is quoted from the table in Ref. 2. The ω_K values in the table [2] are originally based on both experimental and theoretical informations and up-date values from a polynomial fit to selected experimental data; these values are reasonably applied to singly ionized atoms, since they are not corrected for solid state, chemical or multiple ionization effects. It is not expected that these corrections are not negligibly small for light elements such as chlorine ($Z=17$) in the present case; the ^{35}S source is solid state with a chemical form of Na_2SO_4 on carbon sheet. The rather large error in the adopted value, i.e., $\pm 10\%$, mainly comes from these uncertainties.

4.1.5. Determination of X_K and P_K

In Table 4.3 are given the experimental results for the K X-ray emission probability per decay, X_K , as well as the K-hole creation probability per decay, P_K ; the value in the second column are results by the present work while those in the third column are results by Robinson and Howland [3]. A main part of errors in the present results come from the uncertainties in the factors a_x (the $L\alpha$ X-ray emission probability per electron capture decay of ^{109}Cd) and ω_K ; their uncertainties is 10% for both factors. Statistical errors caused by the X- and β -ray countings in the present work are relatively small compared with these uncertainties.

The errors in the previous results are not equivalent to those in the present results; we have estimated the standard deviations while Robinson and Holland estimated the extreme errors based on the 99:1 probable errors. Using the standard deviation estimated, their results to be compared with the present results 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 . It is now concluded that the present results X_K and P_K agree with the previous values within the standard deviations.

4.2. Comparison with SCF calculations

Theoretical works on K-shell internal ionization and excitation (K-IIE) in these two 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 [4-8], Carlson et al. [9] computed the monopole transition probability of

$$P_K = 2(1 - P_R - P_F), \quad (14)$$

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 ($Z \geq 30$) wave functions. In order to treat the K-IIE process more precisely, Stephan and Crasemann [10], Law and Campbell [11] and Isozumi, Shimizu and Mukoyama [12] developed the one-step approach including the phase-space sharing between three leptons (β , atomic electron and neutrino) and the quantum exchange between two continuum electrons in the final state.

Using SCF functions, Law and Suzuki [13] 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. [9] 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 [14], the agreement between LS calculations 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 4.4; CNTM and LS means the calculation by Carlson et al. [9] and that by Law and Suzuki [13], respectively. LS did not calculate the P_K value for ^{35}S ; their value in Table 4.4 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 4.4, there is an evident difference between the present result and LS value while CNTM value is close by the present result. This strongly implies that the SCF approach by Law and Suzuki is to be modified for the low-Z nuclides ($Z<20$) in spite of the good agreement in high-Z region.

4.3. Concluding remarks

In this work, we have obtained two significant results mainly from the well-resolved spectrum of Cl $K\alpha$ X-ray group, such as Fig. 3.7(a). First, the total probability per β decay of the K-hole creation in the K-IIE process, P_K , for ^{35}S has been determined within a precision enough to be compared with the most reliable calculation by Law and Suzuki [13]. We have found that their theoretical P_K apparently deviates from the value presently determined. It is required that their SCF theory for the K-IIE process is to be re-examined specially for low-Z nuclides. Second, we have obtained a new information on the mechanism of the K-IIE process during β decay. As seen from the intense satellite lines in the spectrum of Cl $K\alpha$ X-ray group, higher shells in addition to 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 per β decay of the K- plus L^n -hole creation in the K-IIE process, where n is the number of L holes. It is urgently desired to estimate theoretically the probability of such multiple ionization and excitation in the K-IIE process during β decay.

Table 4.1. Results for N_x/N_β .

Source	Intensity Measuring		N_x	N_β	η	N_x/N_β
	(MBq)	period (sec)	(counts /10 ³)	(counts /10 ¹³)		(/10 ⁻¹¹)
No. 1	58.8±2.4	487591	2.56±0.21	2.93±0.12	0.679±0.01	8.72±0.81
No. 2	93.3±3.8	865020	6.97±0.40	8.46±0.35	0.548±0.01	8.23±0.58

Table 4.2. Results for D_x .

Run	Measuring	N_x	N_x^0	D_x
	period (sec)	(counts /10 ³)	(counts /10 ¹⁰)	(/10 ⁻⁷)
1	15680	4.06±0.21	2.15±0.24	3.12±0.40
2	24200	5.91±0.23	3.15±0.35	3.10±0.38
3	6213	1.42±0.094	7.71±0.84	3.04±0.41

Table 4.3. Results for X_K and P_K .

	Present work	Rubinson and Holland ^{a)}
$X_K^{b)}$	$(2.8 \pm 0.3) \times 10^{-4}$	$(2.47 \pm 0.6) \times 10^{-4}$
$P_K^{c)}$	$(2.8 \pm 0.5) \times 10^{-3}$	$(2.3 \pm 0.7) \times 10^{-3}$

^aReference [3].

^bdefined as the probability of K X ray emission per β decay.

^cdefined as the probability of K-hole creation per β decay.

Table 4.4. Theoretical P_K values (/ 10^{-3}) by SCF calculations.

Present experiment	Calculation	
	CNTM ^{a)}	LS ^{b)}
2.8 ± 0.5	3.37	$\sim 5.2^{c)}$

^aReference [7].

^bReference [14].

^cObtained by the extrapolation of calculations
in Ref. 13.

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