

## A Double Ionization Chamber for the Differential Method

Hiromasa MAZAKI\*, Eisuke YOSHIOKA\*\*, and  
Shunji KAKIUCHI\*

Received November 5, 1974

A double ionization chamber for measurements of a minute difference in two radioactive sources were constructed. The characteristics of each chamber were studied for various experimental conditions. By applying the differential method with this chamber, relative changes in radioactive decay rate of a nucleus,  $\Delta\lambda/\lambda$ , which is usually of the order of  $10^{-3}\sim 10^{-4}$  can be determined. Some discussions on the differential method are also given.

### I. INTRODUCTION

One of the most convenient methods of measuring a minute difference in intensities between two radioactive sources is the so-called differential method first introduced by Rutherford.<sup>1)</sup> The attractive features of using this method are that one can minimize systematic errors introduced in measurements and it is possible to use a detecting device with an extremely large gain which would not be practical in a single measurement. Furthermore, determinations of the exact amount of the background are not required in the differential method. By these reasons, this method have been used by many workers<sup>2~11)</sup> to investigate minute differences in radioactive decay constants under different chemical or other external conditions.

With an intention of measuring a minute difference in relatively strong gamma-ray sources (10~30 mCi) by means of the differential method, a double ionization chamber was constructed. The chamber consists of two essentially identical ionization chambers which are connected to collect ions of opposite sign. A similar chamber was constructed by Bainbridge *et al.*,<sup>6)</sup> by which they measured changes in the decay constant of  $^{99m}\text{Tc}$  in different chemical states.

In order to achieve the best available conditions in operating the double ionization chamber applied for the differential method, characteristics of each chamber under various conditions were studied. In the present paper, we wish to report details of our experimental work with the chamber, as well as some practical procedures of the differential method.

### II. CHAMBER DESIGN

The construction of the double ionization chamber is shown in Fig. 1. It is constructed by the welded brass and consists of two essentially identical chambers which are

\* 間崎啓匡, 垣内俊二: Laboratory of Nuclear Radiation, Institute for Chemical Research, Kyoto University, Kyoto.

\*\* 吉岡英介: On leave from Hitachi Shipbuilding & Engineering Co., Ltd.

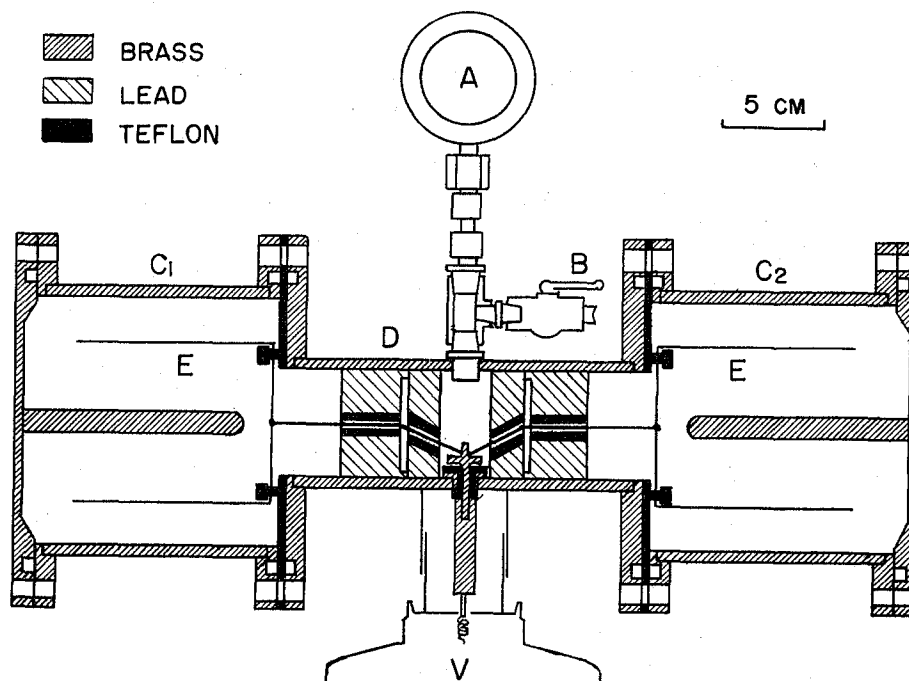


Fig. 1. Double ionization chamber constructed by the welded brass; A) Pressure guage, B) Gas cock to fill and to evacuate the gas, C<sub>1</sub>, C<sub>2</sub>) Two essentially identical ionization chambers, D) Brass pipe connecting two chambers, E) Ion collecting electrode, V) Vibrating reed electrometer.

connected through a brass pipe containing lead blocks in it for gamma-ray shieldings. Not shown in Fig. 1 are lead shields around the central brass pipe. By these shieldings, the leakage of radiations from one source to the other-side chamber can be neglected. The chambers are electrically insulated by teflon plates from the central grounded assembly. Electric potentials ( $< 400$  V) with opposite sign are independently supplied by dry batteries to both chambers. The use of cylindrical collectors (7.5 cm in diameter) is based upon the reasons that we want to avoid different collecting efficiencies for positive ions and electrons (or negative ions) and to prevent gas multiplication due to a large electric field located near the collector. Since the collectors of both chambers are connected at the center, the total output current produced by ions of opposite sign can be made quite small or ideally zero.

The chamber was designed for operation at a pressure of 20 atm at maximum. But, for a long-term operation, the pressure was chosen to be 13 atm and below. No leakage was found even after tests over a month.

Because of the extremely high impedance of the collector, the whole chamber including the head of the vibrating reed electrometer was covered with an iron net for electromagnetic shieldings. Changes in the temperature around the chamber was kept less than  $\pm 1^\circ\text{C}$ . The whole system was set on a goniometer, on which two chambers can be exchanged without moving the radioactive sources. The schematic diagram of the system is shown in Fig. 2.

## Double Ionization Chamber

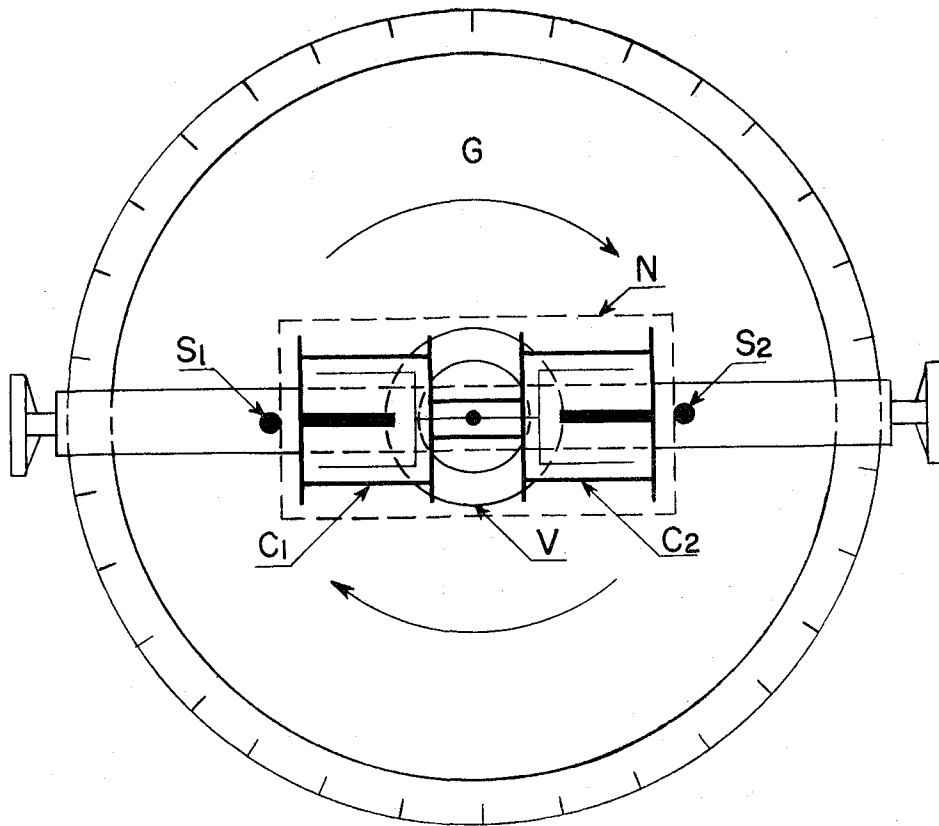


Fig. 2. Schematic diagram of the double ionization chamber set on a goniometer, on which the chamber can be exchanged with respect the sources;  $C_1, C_2$ ) Two identical chambers,  $S_1, S_2$ ) Sources, G) Goniometer, N) Iron net for electromagnetic shieldings, V) Vibrating reed electrometer.

### III. CHARACTERISTICS OF THE CHAMBER

In order to find the optimum condition in operating the double ionization chamber, the detection efficiency of each chamber was studied under various experimental conditions.

#### III. 1. Pressure Dependence

The detection efficiency of the single chamber was measured for various gas pressures up to 13 atm. The chamber was filled with the 90% argon - 10% methane mixture. A  $^{57}\text{Co}$  gamma-ray source (122 keV) was set 3 mm from the brass window (5-mm thick) of the chamber and the output current was measured by the vibrating reed electrometer with the input impedance of  $10^{10}$  ohm. The typical result is shown in Fig. 3. The figure indicates that the detection efficiency naturally increases as the inner gas pressure increases. This means that for better detection efficiency, it is desired to use a pressure as high as possible, provided the chamber stands the pressure.

#### III. 2. Dependence on the Collecting Voltage

Relative outputs for the  $^{57}\text{Co}$  source were measured by changing the collecting voltage

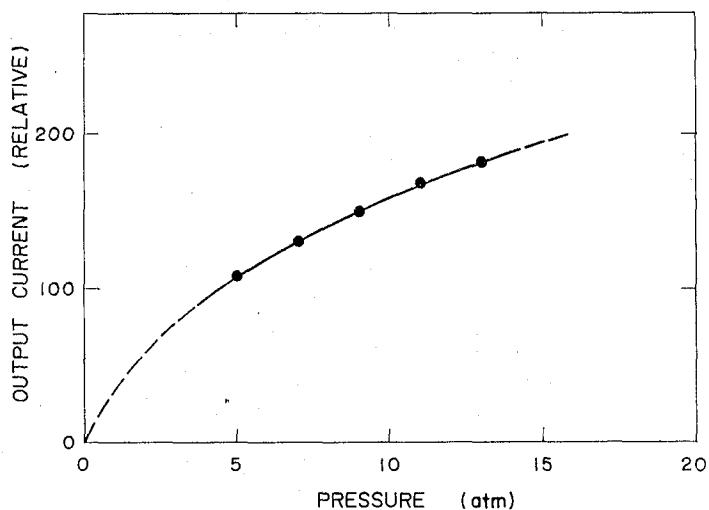


Fig. 3. Pressure dependence of the detection efficiency of the single chamber filled with the 90% argon-10% methane mixture.

in the range of 48~379 V. The gas pressure was chosen as 5, 7, 9, 11, and 13 atm. The results are shown in Fig. 4. At a relatively low pressure, the output current does not depend greatly on the collecting voltage. However, at a high pressure, say 13 atm, the output current gradually increases as the voltage increases.

This phenomena may be interpreted by the electron attachment to positive ions forming neutral molecules. The attachment probability depends in general on the nature of the gas, on the energy of electrons and on the pressure. In the present case, however, the pressure is the only factor which gives rise to the effect on the probability. When the

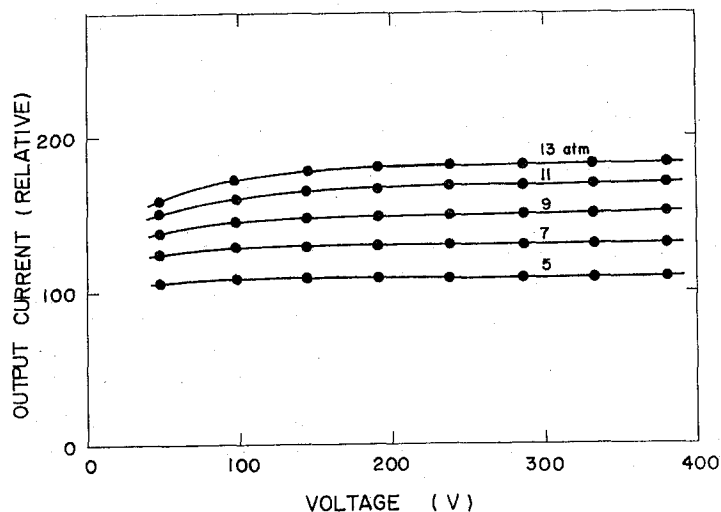


Fig. 4. Dependence of the detection efficiency on the collecting voltage at various gas pressures.

pressure is low enough, the attachment probability can be neglected, but at a high pressure, the recombination of electrons with positive ions plays some role in decreasing the output current. For a given pressure, the attachment probability is decreased with the increased collecting voltage, resulting in increase of the output current.

### III. 3. Dependence on Incident Gamma-Ray Energy

Three kinds of gamma-ray sources were prepared to observe energy dependence of the detection efficiency. These are  $^{57}\text{Co}$  (1.11 mCi, 122 keV),  $^{137}\text{Cs}$  (0.671 mCi, 662 keV) and  $^{60}\text{Co}$  (5.72 mCi, 1.17, and 1.33 MeV). Their absolute intensities were determined by the usual method. Each source was placed at the same position, 30 cm from the window surface of the chamber, and relative output current was measured.

As expected, the relative output current of the  $^{60}\text{Co}$  source is much larger than that of the  $^{57}\text{Co}$ , by a factor of about 10 (see Fig. 5). This is partly due to less attenuation of gamma rays from  $^{60}\text{Co}$  in the window. It seems, therefore, to be more practical to compare efficiencies corrected for attenuation of gamma rays in the 5-mm brass window, of which results are also shown in Fig. 5. It should be noted that a simple correction for attenuation gives only the first approximation, because a build-up factor in the window is not taken into account.

### III. 4. Effect of Window Thickness

A part of incident gamma rays is absorbed or scattered during passage through the window. The fraction of incident gamma rays reached at the inner wall of the chamber depends on energy.

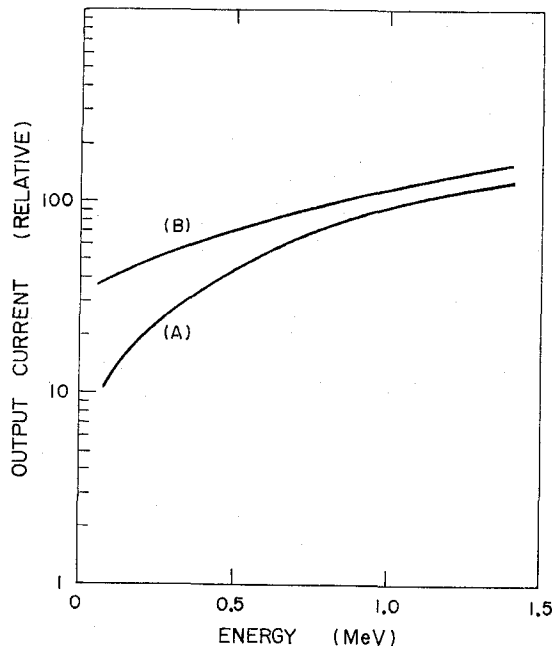


Fig. 5. Dependence of the detection efficiency on the incident gamma-ray energy; A) Observed output current versus gamma-ray energy, B) Corrected for attenuation in the 5-mm thick window of the chamber.

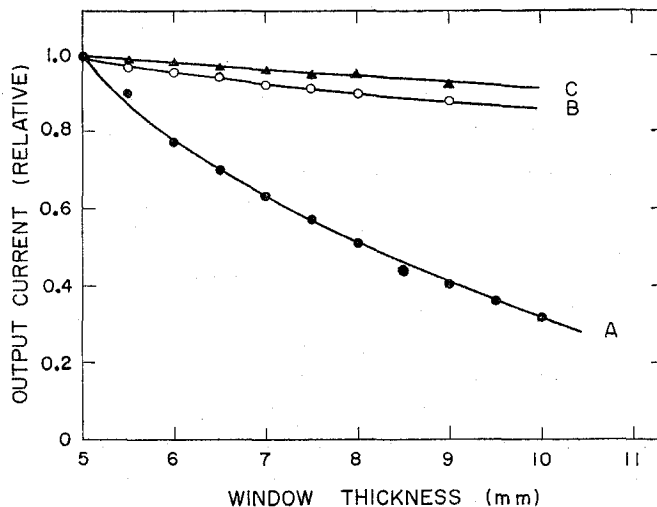


Fig. 6. Dependence of the detection efficiency on the window thickness; A)  $^{57}\text{Co}$  (122 keV), B)  $^{137}\text{Cs}$  (662 keV), C)  $^{60}\text{Co}$  (1.17 and 1.33 MeV).

To find the energy dependence of the fraction passed through the window, three kinds of radioactive sources ( $^{57}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{60}\text{Co}$ ) placed at 30 cm from the window surface were again used without collimators. In Fig. 6 are shown the observed output current as a function of window thickness in the range of 5~10 cm. The results indicate that the thickness of window should be chosen as the minimum which can stand the given inner pressure.

### III. 5. Linearity for Source Intensity

Linearity of the detection efficiency as a function of source intensity is of importance in any measurement of radioactivity. For the use of digital detectors which count numbers of radioactive rays, for example a GM counter, a dead time correction is required to obtain real counts. But, a dead time depends, strictly speaking, on energy of radioactive rays as well as counting rates. By this reason, it is difficult to perform precise measurements when the counting rate becomes comparable with the resolving time of the detection system (usually  $10^4 \sim 10^5$  cps).

Taking into consideration the difficulty in dead time correction, for a large counting rate over  $10^4$  cps, it seems to be more reliable to use analogue detectors which, in principle, do not have a dead time, giving the output as current or voltage instead of counts.

With a  $^{99\text{m}}\text{Tc}$  source (140 keV), of which initial intensity is about 40 mCi, the linearity of the present ionization chamber was investigated. Since  $^{99\text{m}}\text{Tc}$  decays with the half-life of 6.04 h, the output current from the chamber was recorded as a function of source intensity. Observed results are shown in Fig. 7, where the data were corrected for decay. The results indicate the chamber holds its linearity in the range of about 20 mCi and below (corresponding to  $<10^7$  cps in the present experimental condition), but the linearity breaks down in the range over 20 mCi, showing an appreciable effect of recombination and the decrease of the electric field in the sensitive volume of the chamber. Although the break down of the linearity depends on the gas pressure and the design of the chamber, one can

## Double Ionization Chamber

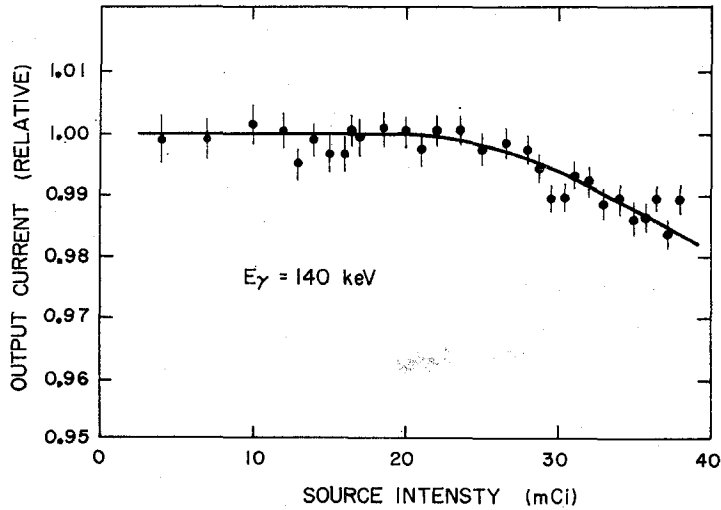


Fig. 7. Linearity of the detection efficiency as a function of source intensity. The gamma-ray source is  $^{99m}\text{Tc}$  (140 keV) with initial intensity of about 40 mCi.

conclude that even an analogue detector has an applicable range for an intensive radioactive source.

### III. 6. Detection Efficiency for Various Gases

The detection efficiency of the chamber depends on the gases which consist of elements with different atomic numbers and ionization potentials.

By the use of the  $^{57}\text{Co}$  source, comparisons of the output currents from the chamber filled with various gases were made. The pressure was chosen as 13 atm in each measurement. The results are listed in Table I. From the results, aged pure argon was used for the further experiment.

## VI. APPLICATION OF THE DIFFERENTIAL METHOD

As mentioned in Sec. I, the most exciting application of the differential method is to measure a small change in decay probability of a radioisotope under different external conditions. Supposing we have two  $^{99m}\text{Tc}$  sources, 1 and 2, under different conditions,

Table I. Detection Efficiency for the 122 keV Gamma Rays with Various Gases

| Gas                                     | Output current<br>(Relative) |
|-----------------------------------------|------------------------------|
| Argon (aged, 99.998%)                   | 100                          |
| 90% argon-10% methane mixture           | 89                           |
| 92.3% argon-7.7% $\text{CO}_2$ mixture  | 81                           |
| 85.2% argon-14.8% $\text{CO}_2$ mixture | 72                           |
| 78.7% argon-21.3% $\text{CO}_2$ mixture | 64                           |
| 72.6% argon-27.4% $\text{CO}_2$ mixture | 59                           |
| 90% helium-10% methane mixture          | 22                           |

where their decay probabilities are  $\lambda$  and  $\lambda + \Delta\lambda$ , respectively. According to the experimental works by Bainbridge *et al.*<sup>6)</sup> and by Mazaki *et al.*,<sup>11)</sup> relative alterations in the decay probability,  $\Delta\lambda/\lambda$ , is of the order of  $10^{-3} \sim 10^{-4}$ . It is practically very difficult to find such a small difference by a single measurement of decay rate.

By the use of the double ionization chamber, the output currents due to sources 1 and 2, measured by chambers  $C_1$  and  $C_2$ , are given as

$$J_{I-1}(t) = G_1 S_{I-1} \exp(-\lambda t), \quad (1)$$

$$J_{I-2}(t) = G_2 S_{I-2} \exp(-\lambda t - \Delta\lambda t), \quad (2)$$

where  $S_{I-1}$  and  $S_{I-2}$  are initial intensities of sources 1 and 2,  $G_1$  and  $G_2$  are the gains of  $C_1$  and  $C_2$ , a suffix I means the first measurement. Since the collecting potentials of  $C_1$  and  $C_2$  are opposite, the output from the vibrating reed electrometer at time  $t$  can be given as

$$E_I(t) = G[G_1 S_{I-1} \exp(-\lambda t) - G_2 S_{I-2} \exp(-\lambda t - \Delta\lambda t)], \quad (3)$$

where  $G$  is the gain of the electrometer.

In the second measurement, denoted by a suffix II, two chambers are interchanged with respect to the sources, *i.e.*,

$$J_{II-1}(t) = G_2 S_{II-1} \exp(-\lambda t), \quad (4)$$

$$J_{II-2}(t) = G_1 S_{II-2} \exp(-\lambda t - \Delta\lambda t), \quad (5)$$

where  $S_{II-1}$  and  $S_{II-2}$  denote the initial activities of the sources 1 and 2 in the second run. As for the first measurement, the output from the vibrating reed electrometer at time  $t$  is given as

$$E_{II}(t) = G[G_1 S_{II-2} \exp(-\lambda t - \Delta\lambda t) - G_2 S_{II-1} \exp(-\lambda t)]. \quad (6)$$

Of particular importance in the differential method is the average difference output which is formed between Eqs. (3) and (6) as

$$E(t) = \frac{1}{2}[E_I(t) - E_{II}(t)]. \quad (7)$$

Taking into account that  $\Delta\lambda \ll 1$ , Eq. (7) multiplied by  $\exp(\lambda t)$  can be represented as

$$\begin{aligned} E(t) \exp(\lambda t) = & \frac{1}{2} G [G_1 (S_{I-1} - S_{II-2}) - G_2 (S_{I-2} - S_{II-1}) \\ & + (G_2 S_{I-2} + G_1 S_{II-2}) \Delta\lambda t]. \end{aligned} \quad (8)$$

Thus, the first and second measurements supply a set of data and the whole procedure is repeated. Equation (8) has a linear form  $y = a + bt$ , being convenient for analysis. This means that for  $\Delta\lambda = 0$ ,  $E(t) \exp(\lambda t)$  holds a constant value, but for  $\Delta\lambda \neq 0$ , it changes with a slope of  $\frac{1}{2} G (G_2 S_{I-2} + G_1 S_{II-2}) \Delta\lambda$  as  $t$  increases. From the experimental value of the slope, one can determine  $\Delta\lambda/\lambda$ .



## V. DISCUSSION

As one of the favorable features of the differential method, we discuss about the case in which either of  $G_1$  or  $G_2$ , the gain of the chamber  $C_1$  or  $C_2$ , changes in a small quantity. Suppose that the activities of source 1 at time  $t$  in the first and second measurements are nearly equal and are denoted by  $S_1$ , and similarly those of source 2 are denoted by  $S_2$ , *i.e.*,

$$S_{I-1} \exp(-\lambda t) \simeq S_{II-1} \exp(-\lambda t) \simeq S_1, \quad (9)$$

$$S_{I-2} \exp(-\lambda t - \Delta\lambda t) \simeq S_{II-2} \exp(-\lambda t - \Delta\lambda t) \simeq S_2. \quad (10)$$

Then, Eqs. (3) and (6) can be written as

$$E_I(t) = G(G_1 G_1 - G_2 S_2), \quad (11)$$

$$E_{II}(t) = G(G_1 S_2 - G_2 S_1). \quad (12)$$

From Eq. (7), the average difference output is

$$E(t) = \frac{1}{2} G(S_1 - S_2)(G_1 + G_2). \quad (13)$$

If either of  $G_1$  or  $G_2$  changes by  $\delta G$ , the resulting small change in  $E(t)$  becomes

$$E(t) = \frac{1}{2} G(S_1 - S_2) \delta G. \quad (14)$$

While if the difference outputs are not averaged, fractional change in  $E_I(t)$  or  $E_{II}(t)$  is given as

$$E_I(t) = G S_1 \delta G \text{ or } -G S_2 \delta G, \quad (15)$$

$$E_{II}(t) = G S_2 \delta G \text{ or } -G S_1 \delta G. \quad (16)$$

Taking into account that  $S_1$  or  $S_2 \gg |S_1 - S_2|$ , it is evident that systematic errors are reduced considerably by averaging the difference outputs of the first and second measurements.

Moreover, the actual observed values of  $E_I(t)$  and  $E_{II}(t)$  involve the dark current of the chamber due to cosmic rays and other radioactive contaminants. But if, before starting measurements, the output current of the vibrating reed electrometer is accurately settled to zero, the dark currents in the first and second measurements are cancelled out by means of Eq. (7). Therefore, in this method, it is not required to determine exact amount of the dark current.

As described above, systematic errors introduced in the measurement of  $\Delta\lambda$  can be greatly reduced, and thus statistical fluctuation in decay phenomena becomes predominant source of errors. In order to find out  $\Delta\lambda/\lambda$ , of which value is of the order of  $10^{-3} \sim 10^{-4}$ , the number of decay phenomena observed in a certain time interval should be at least  $10^8$  counts or more from the statistical requirement. As discussed in Sec. III,5, it is by no means appropriate to use a counting detector. The double ionization chamber is considered to be the most suitable detector for such a strong radioactive source. From this standpoint, the effect of the superconducting state on  $\lambda$  of  $^{99m}\text{Tc}$  is being pursued with the present double ionization chamber. Details of the experiment will appear elsewhere.

### ACKNOWLEDGMENT

The authors wish to express their thanks to Professor S. Shimizu for his stimulating discussions. This work was supported in part by a Scientific Research Grant from the Ministry of Education of Japan.

### REFERENCES

- (1) E. Rutherford, *Sitzber. Akad. Wiss. Wien. Math. natur. Kl.*, **120**, 303 (1911).
- (2) R. Bouchez, P. Daudel, R. Daudel, and R. Muxart, *Compt. rend.*, **227**, 525 (1948); *J. Phys. Radium*, **10**, 201 (1949); P. Benoist, R. Bouchez, P. Daudel, R. Daudel, and A. Rogozinski, *Phys. Rev.*, **76**, 1000 (1949).
- (3) E. Segrè and C. E. Wiegand, *Phys. Rev.*, **75**, 39 (1949); Erratum, *Phys. Rev.*, **81**, 284 (1951).
- (4) R. F. Leininger, E. Segrè, and C. Wiegand, *Phys. Rev.*, **76**, 897 (1949); Erratum, *Phys. Rev.*, **81**, 280 (1951).
- (5) K. T. Bainbridge, M. Goldhaber, and E. Wilson, *Phys. Rev.*, **84**, 1260 (1951).
- (6) K. T. Bainbridge, M. Goldhaber, and E. Wilson, *Phys. Rev.*, **90**, 430 (1953).
- (7) J. J. Kraushaar, E. D. Wilson, and K. T. Bainbridge, *Phys. Rev.*, **90**, 610 (1953).
- (8) S. Shimizu and H. Mazaki, *Phys. Let.*, **17**, 275 (1965).
- (9) H. Mazaki and S. Shimizu, *Phys. Rev.*, **148**, 1161 (1966).
- (10) M. Nishi and S. Shimizu, *Phys. Rev.*, **B 5**, 3218 (1972).
- (11) H. Mazaki, T. Nagatomo and S. Shimizu, *Phys. Rev.*, **C 5**, 1718 (1972).