Determination of Very Low Transition Energy of ^{235m}U

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With a specially designed 12-stage secondary electron multiplier, a conventional technique for detection of soft electrons below 100 eV has been developed. Applying the retarding method with this multiplier, the transition energy of a nuclear isomer ^{235m}U was determined as to be 30 ± 3 eV. Although there are some restrictions concerning electron sources to be measured, application of the secondary electron multiplier seems to be a useful technique for measurement of soft electrons.

I. INTRODUCTION

The determination of electron energy of less than a few hundred electron volts is usually quite difficult because of the following reasons : (a) Detection of soft electrons requires a special type of detector, for example, a windowless proportional gas counter which is inconvenient for measurement of an electron spectrum, (b) analysis of the observed data is complicated for various kinds of background owing to photoelectrons, secondary electrons or some others.

There are some reports[1~4] as to measurements of the kinetic energy of very soft electrons. Simpson[1] has constructed a low energy electron spectrometer consisting of an electron source, an electron monochromator, a gas cell and an electron energy analyzer. This spectrometer was designed for work in the 5–20 eV primary energy range. Freedman *et al.*[2] have measured the transition energy of ^{235m}U by the use of both magnetic spectrometry employing preacceleration and by electrostatic spectrometry, and succeeded in observing the electron spectrum below a few tens of electron volts. A recently developed type of windowless electron multiplier has been found to be useful for the detection of low energy charged particles. With a Bendix M-306 electron multiplier, Leffel[3] has measured electrons with energies of 100 to 1000 eV. Evans[4] has independently investigated the characteristics of a Bendix continuous-channel electron multiplier and found that, for suitable design of the channel, the efficiency of the channel for the detection of electrons over the energy range 250 eV to 10 keV was greater than 50%.

A conventional method to measure an electron energy in the range below 100 eV has recently been developed with a specially designed secondary electron multiplier, which was manufactured by Mitsubishi Electric Company and modified in some details. Using this multiplier, the very low transition energy of a 26-min isomeric state of ^{235}U has been measured. The uranium sources were prepared from a very thin ^{239}Pu source by the so-called *recoil method,* and by applying a

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retarding method, the integral spectrum of the internal conversion electrons from the isomer was observed. From the analysis of the observed curve, it was found that the resolution of the secondary electron multiplier was fairly good and very soft electrons of energies down to 1 eV could be observed distinguishably. The transition energy determined from the electron spectrum was 30 ± 3 eV and some discussions on the results obtained are also given.

II. EXPERIMENTAL

1. Principle of the Measurement

The experimental method used in the present work is based on the retarding method which is generally a useful technique for determination of electron energy of less than a few hundred electron volts. An electron with a certain kinetic energy can be retarded and finally stopped if the electric field where the electron passes through is equivalent to the initial kinetic energy of the electron concerned. However, there is a difficulty in detection of the electron which has almost been stopped. To deal successfully with this difficulty, preacceleration of the electron in the electric field of several kilovolts or more may be effective when the preacceleration voltage can be supplied with enough accuracy. If the electron can be detected without preacceleration, it may be possible to determine the electron energy with much more accuracy.

By the use of a 12-stage secondary electron multiplier, Model ND 9112 of Mitsubishi Electric Company, originally designed at Nier's Laboratory and modified in some details, it was found that extremely soft electrons could be effectively measured without preacceleration. The essential construction of the multiplier is quite similar to that of a linear type photomultiplier. The Be-Cu dynodes of the multiplier were specially treated by the oxidization process, in which the dynodes were heated in a 10-20 μHg oxgen atmosphere at 500°C for a few minutes. By this treatment, copper oxide decomposes and the dynode surface is covered only by beryllium oxide which has high secondary electron emission efficiency and is stable even in air at room temperature. The overall gain and the dark current both measured in a pressure of 1.0×10^{-5} mmHg with an applied voltage of -3.6 kV were 1.1×10^5 and 1.2×10^{-18} A, respectively. The geometrical arrangement of the dynodes in the multiplier is shown schematically in Fig. 1.

As a soft electron emitter, ^{235m}U was directly deposited on the first dynode, as will be described in the following section. Between the first and second dynodes, a 100-mesh gold-plated grid screen (81% open area) was placed, 2.5 mm from the first one, so as to control the soft internal conversion electrons from the isomer. Since the effective surface of the first dynode has a curvature, the screen was curved as to have the same curvature as that of the first dynode. The edge of the screen was coated with Araldite to keep it in shape as well as to avoid the corona discharge. Then various retarding potentials between the first dynode and the screen were supplied in the range of 0-70 V. These voltages were measured by a conventional transistorized potentiometer and calibrated taking into account the contact potentials of the conducting wires. The voltages between the screen and the second dynode, and other neighboring dynodes were kept constant
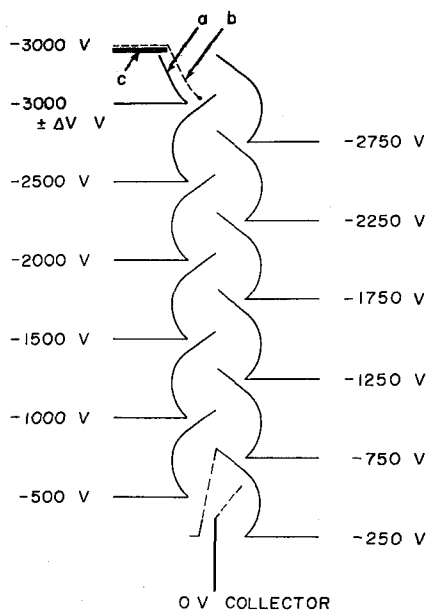
Fig. 1. Geometrical arrangement of the dynodes in the secondary electron multiplier. (a) First dynode (source dynode), (b) 100-mesh gold-plated screen and (c) Lucite.

(250 V) to maintain a constant gain of the multiplier during measurements. Once the internal conversion electrons get through the screen, they are attracted toward the second dynode and eject some secondary electrons from the second dynode, and so on. The outputs from the multiplier for various retarding voltages were counted through a preamplifier and a low-noise linear amplifier. If the retarding voltage becomes to be equal to the initial kinetic energy of the electrons to be measured, they should be completely stopped by the screen. Thus, from the end point of the spectrum, the maximum kinetic energy of the electrons can be determined. The background of the measuring system for each retarding voltage was also measured. It was found that the background of the system without any source depended on the retarding voltage between the first dynode and the screen. Therefore, the observed data of the conversion electrons for each retarding voltage were subtracted by the corresponding values of the background.

Since the half-life of the isomer is only about 26 min[5], measurements must be started as soon as possible after the preparation of the uranium samples. A powerful ion pump of 2400 liters/min (NEC Model 911-1404) connected with a subsidiary rotary pump of 250 liters/min was used and the inner pressure of the multiplier could be pumped out down to 1×10^{-6} mmHg from 1 atm in less than 10 min.

2. Source Preparation

A nuclear isomer ^{235m}U, the first excited level of ^{235}U, has an extremely large internal conversion coefficient being about 10^{20}, and no gamma-ray emission is accompanied by this transition. The internal conversion electrons from ^{235m}U are

expected to be less than a few tens of electron volts[2,6].

As a preliminary step of the work, the recoil method has been investigated for preparation of uranium sources. When [239]Pu disintegrates by emitting an alpha particle with an energy of about 5 MeV, the daughter uranium produced in the excited level may be released from the plutonium source with the recoil energy of about 90 keV. The product in the second or higher excited levels is promptly followed by partly converted gamma-ray transition and the alpha decay of [239]Pu results in production of [235m]U directly or indirectly. To collect the recoiled uranium atoms, a very thin plutonium source of about 2 μg/cm² thickness was prepared electrolytically on a pure copper plate of 3-cm diameter. This source was supplied from Centre d'Études Nucléaires in Saclay. The recoiled uranium atoms from [239]Pu are usually ions, probably +6 or higher. This advantageous ionic state leads us to the successful way of collection of [235m]U on a metallic plate facing to the plutonium source with applying a negative potential to the plate. This is the so-called recoil method.

To obtain the best collecting yield of this recoil method, various conditions which may control the collecting yield of the [235m]U atoms have been examined. Changing the collection period, the distance and the applied voltage between the plutonium source and collector plate in air, the yields of the isomer were measured. The results are shown in Figs. 2, 3 and 4. Taking into consideration these results, the uranium sources were prepared by collecting the isomer for 3 h on a collector plate, placed 6 mm under the plutonium source and applied by −600 V negative potential. Under the condition described above, the [235m]U sources showed the activity of about 5000 counts cm⁻² min⁻¹ at best measured by a low-background 2π gas-flow counter. To secure the maximum intensity of the source, the isomer was collected directly on the first dynode of the multiplier.

Four kinds of uranium samples were prepared: (a) collected on a Cu-Be dynode in air, (b) on a platinum dynode in a pure argon atmosphere (99.998%), (c) on a gold-plated dynode in a pure argon atmosphere and (d) on a gold-plated
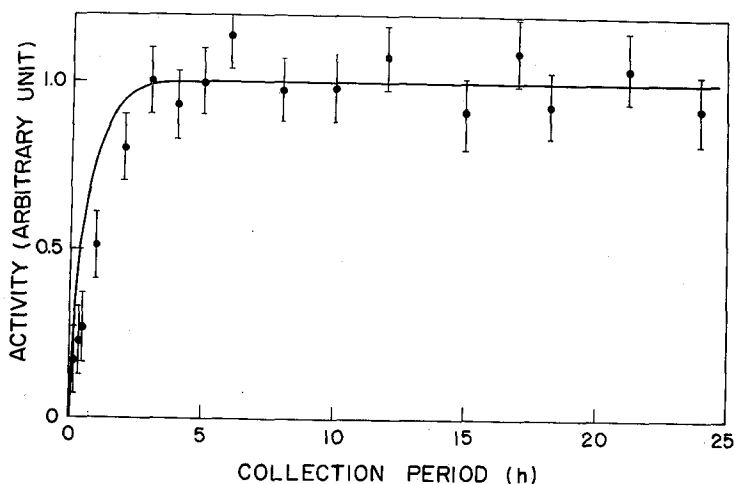

Fig. 2. Dependency of the collecting yield of [235m]U on the collection period. The solid curve indicates the theoretical yield.
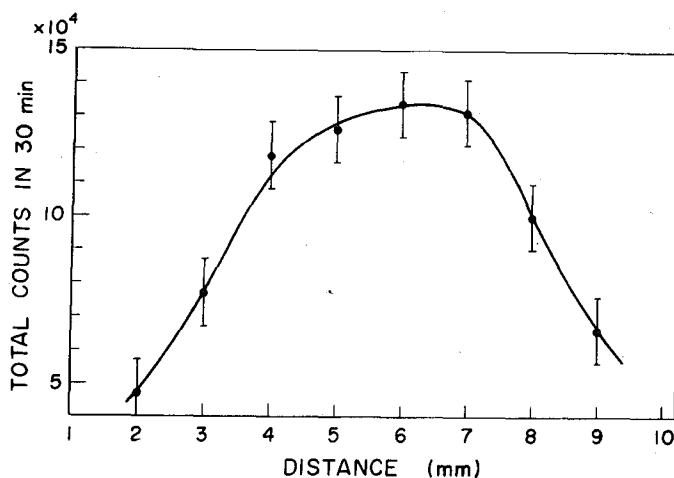
Fig. 3. Dependency of the collecting yield of ^{235m}U on the distance between the collector plate and the ^{239}Pu source. Applied voltage on the collector plate is -600 V and a collection period is 3 h.
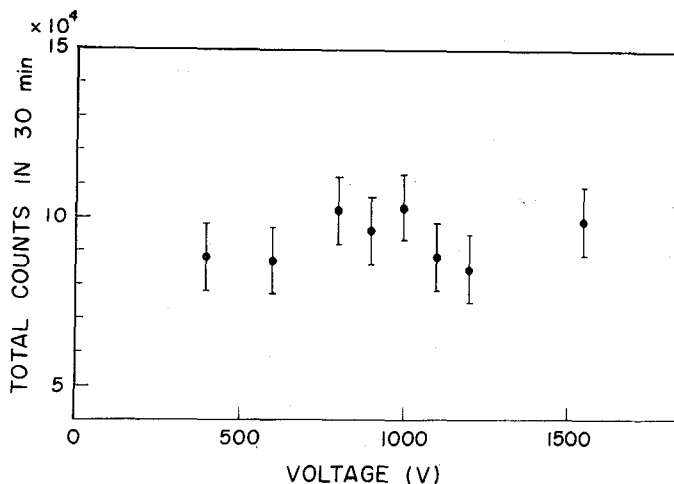

Fig. 4. Dependency of the collecting yield of ^{235m}U on the applied voltage between the collector plate and the ^{239}Pu source. The distance between them is 5 mm and a collection period is 3 h.

dynode in air. To examine the purity of thus prepared samples, the measurements of the source intensities were extended through 5 mean lives. Furthermore, the collected uranium sources were covered by thin collodion films of about 6 μg/cm^2 thickness and counted by the gas–flow counter. It was found that the collodion film cuts down the activity of the uranium source completely. This fact means that the activity consisted of only the soft internal conversion electrons from the isomer. Thus the uranium sources obtained were proved to be perfectly free from any radioactive contaminant. The alpha emission from ^{235}U in the ground state contained in the sources was estimated to be negligible.

III. RESULTS AND DISCUSSION

In Fig. 5 is shown the observed electron spectrum obtained for the uranium samples collected on a gold-plated dynode in a pure argon atmosphere. The results are summed from thirty runs of measurements.
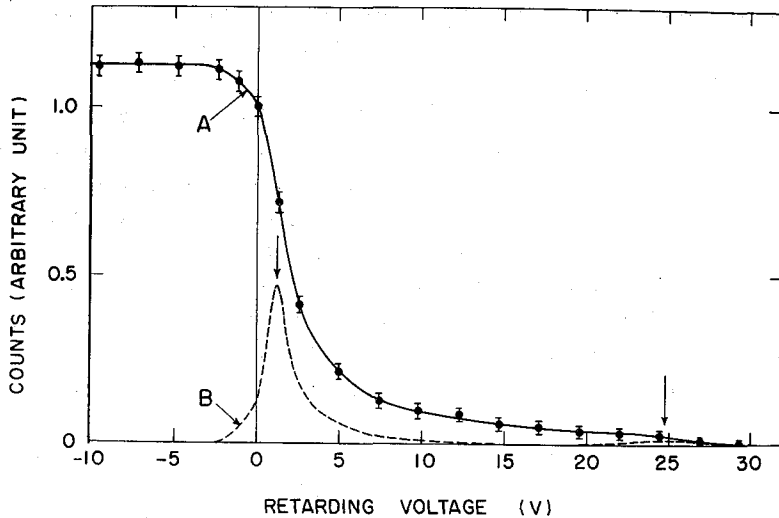

Fig. 5. Solid curve (A) is the observed data (background subtracted) for retarding and accelerating voltages, corrected for decay and normalized to the value for zero potential difference between the first dynode and the grid screen. Dashed curve (B) is the differential spectrum of (A) in arbitrary unit. Two peaks can be seen at 1.2 V and at about 25 V.

Since the transition energy of the isomer is only a few tens of electron volts, only P- or Q-shell electrons can be emitted by the internal conversion process. From this fact, it is expected that there are two electron groups corresponding to P and Q shells. The observed integral spectrum, shown by a solid curve in Fig. 5, indicates two structures which may be considered to be due to the P- and Q-shell conversion electrons. The electron spectrum for attracting voltages becomes saturated rather rapidly. The differential spectrum of the observed curve is also shown by a dashed curve in Fig. 5, where an evident peak at 1.2 eV and a some-what obscure one at about 25 eV are observed. These peaks are rather broad and distorted. This can be understood by some characteristics of secondary electron emission from the dynodes of the multiplier as well as by slight distortion of the electric field between the source dynode and grid screen due to the existence of other dynodes. In the present case the background due to secondary electron emission from the gold-plated grid screen can be neglected.

To obtain the transition energy of the isomer, there are some factors to be considered. First of all, the binding energy of the emitted shell electrons must be added to the measured electron energy. Only available data are those of $P_{II,III}$- and $P_{IV,V}$-shell electrons in uranium which have been known to be 30 and 0 eV[7], respectively. The $P_{II,III}$-shell electrons in uranium are supposed to contribute

predominantly to the observed peak at 1.2 eV. As a possible approximation, therefore, neglecting the effect of binding of uranium atom on the first dynode surface, we can conclude the transition energy be about 31 eV. On the other hand, the $P_{IV,V}$- and Q-shell electrons in a free uranium atom may be assumed to be nearly free. However, in the case of uranium atom bound on a metal surface, the work function of the metal probably gives the threshold energy for emission of an electron from the surface. The work function of Au is 4.6 eV. The transition energy determined from the small peak at 25 eV may be reasonably assumed to be about 30 eV. The second source of error may be caused by different chemical forms of the isomer. When the recoiled uranium ions fly toward the collector plate in air, they probably lose the charge and form uranium oxide. To observe this effect, if any, two kinds of uranium samples were compared, i. e. collected on a gold-plated dynode in air and in a pure argon atmosphere. But the results did not show any evident difference, indicating that the effect of chemical form of uranium on the conversion electron spectrum was not detectable. Thirdly, the effect of different work functions of the base materials was checked by comparing the uranium samples collected on the different dynodes (Cu-Be, Au and Pt). But the results were also negative and dependency of the conversion electron spectrum on the different work functions of the base materials could not be observed. Taking into account all possible sources of errors, the transition energy of ^{235m}U was finally determined as to be 30 ± 3 eV.

With the device described above, it was found that an electron energy below about 100 eV could be determined with an accuracy of 10% or less. An inconvenient feature of the present device is that an electron source should be mounted inside of the multiplier and an electron beam from outer electron gun or others can not be measured. However, a secondary electron multiplier is apparently a useful instrument for detection or measurement of low energy electrons and more developed technique with it can be expected.

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