

Differential spectrum of the conversion electrons and the excitation energy of $(1/2^+)$ -uranium-235 isomer

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The differential spectrum of the conversion electrons of ^{235m}U has been measured for the first time. The conversion spectrum has been compared with the x-ray electron spectrum of UF_4 for the case of polycrystalline UF_4 matrix impregnated with the isomer. On this basis it is concluded that the electrons of the UF_4 "valence band," $6p_{1/2^-}$, $6p_{3/2^-}$ -uranium electrons—and possibly $2s$ -electrons of fluorine—take part in the conversion process. An isomer excitation energy of 76.8 ± 0.5 eV is determined.

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The decay of the "26-minute" $(1/2^+)$ -isomer ^{235m}U into the ground $(7/2^-)$ -state of ^{235}U is a unique case of the conversion of the nuclear E3-transition into the outer electron shells of the uranium atom.^{1,2} The transition energy, according to recent data, is only 74 ± 4 eV.³ The spectrum of ^{235}U conversion electrons has been investigated many times⁴⁻⁷; however, up to now there has been no success in determining the structure of the spectrum. Nevertheless, it was assumed that conversion occurs into the $(6p_{1/2})^2$ -, $(6p_{3/2})^4$ - and $6d$ -shells of uranium.⁷

We have recently measured the differential spectrum of the ^{235m}U conversion electrons, identified its structure and determined the isomeric transition energy more precisely. Uranium tetrafluoride (UF_4) was chosen as the matrix for the isomer.

Alpha decay of ^{239}Pu ($25 \mu\text{g}/\text{cm}^2$, 100 cm^2) was used to obtain the ^{235m}U . The isomer—the recoil atoms in the alpha decay of ^{239}Pu —were collected in an atmosphere of chemically pure helium (100 mm Hg) by means of an axially symmetrical electrostatic field ($100 \text{ V}/\text{cm}$) on the surface of a polycrystalline UF_4 layer, deposited beforehand on a gold foil. To avoid destruction of the UF_4 layer, the corona discharge current at the layer surface was limited to $\sim 1 \mu\text{A}$. It was expected that drift deep in the UF_4 layer and subsequent isotopic exchange would contribute to the formation of the compound $^{235m}\text{UF}_4$. No build-up of uranium oxides on the surface of the UF_4 layer was detected, even after it had been exposed to the isomer collection conditions, to judge from a comparison of the x-ray electron spectra¹⁾ of UF_4 , which we measured before and after the isomer. The ^{235m}U sources had an area of $1.5 \times 5 \text{ mm}^2$ and exhibited an initial activity of 10^5 decays/sec.

The measurements of the differential spectrum of the ^{235m}U conversion electrons and the UF_4 x-ray electron spectrum were performed on the Hewlett-Packard model HP5950A ESCA spectrometer.⁸ We extended the energy range of the electron-optical portion of the spectrometer ($300\text{--}1500 \text{ eV}$) to $0\text{--}2500 \text{ eV}$ without significantly altering the physical characteristics of this instrument. In particular, the half-width we measured at the half-height of the $\text{Au } 4f_{7/2}$ x-ray electron line amounted to $\sim 0.8 \text{ eV}$. The

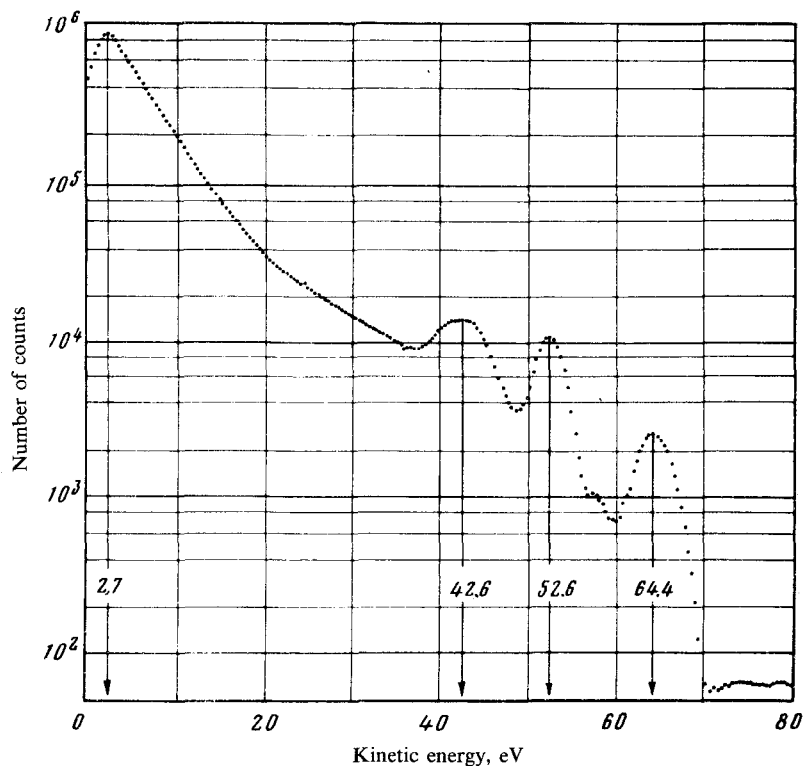


FIG. 1. Conversion electron spectrum of ^{235m}U .

energy scale of the spectrometer was calibrated with an accuracy of ~ 0.5 eV by finding, with the aid of an HP2100A computer, the maxima of the $\text{Cu } 2p_{1/2}$, $\text{C } 1s$ and $\text{Au } 4f_{7/2}$ x-ray electron lines, the binding energies of which are well known.⁹ The origin of the binding energy scale coincided with the position of the Fermi level (E_f) at the "zero-cutoff" (boundary of the conduction band) of the x-ray electron spectrum of palladium. Under the conditions existing for the measurement of the ^{235m}U conversion spectrum the spectrometer resolution, which we determined in terms of the cutoff of the secondary electron peak in the vicinity of the "zero energy" electrons of the ^3T isotope, was no worse than 0.5 eV.

Figure 1 shows the conversion electron spectrum of the ^{235m}U isomer, inserted into a polycrystalline UF_4 matrix. This is the result of summing five separate spectra, measured with different ^{235m}U sources. The spectrum was smoothed three times with respect to a running sequence of five successive points. Instrument background was subtracted out. Three peaks are readily apparent in the conversion electron spectrum of ^{235m}U , the locations of which correspond to electron kinetic energies of 42.6, 52.6 and 64.4 eV. These peaks lie at the beginning of the rise of a very intense and asymmetrical maximum with an energy of about 2.7 eV. The low-energy peak is produced by electron stopping processes in the UF_4 layer and by secondary electron emission. We

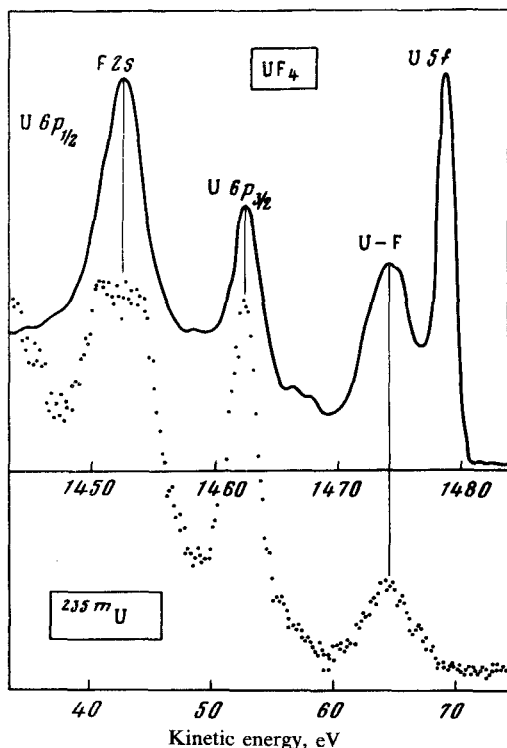


FIG. 2. Comparison of conversion and x-ray electron spectra.

observe exactly this same maximum in the spectrum of low-energy electrons from ^3T , ^{242}Cm , ^{252}Cf , etc., radioactive sources. Its overwhelming intensity in the ^{235m}U conversion spectrum attests to a relatively deep penetration of the isomer into thickness of the UF_4 layer. The three other peaks of the spectrum are conversion peaks.

In order to identify the conversion peaks and determine the ^{235m}U excitation energy, we compared the conversion and x-ray electron spectra obtained by us from the same UF_4 layer under completely identical measurement conditions. Portions of these two spectra, coinciding in terms of the relative position of the maximums, are shown in Fig. 2.

The smooth curve in the upper part of Fig. 2 is the valence-band region of the UF_4 x-ray electron spectrum. The curve, formed by the points in the lower part of the figure, refers to the ^{235m}U spectrum in the vicinity of its conversion lines.

We will use the interpretation of the UF_4 x-ray electron spectrum suggested in Ref. 10. For simplicity we will also use the one-electron notations. The rightmost peak of the x-ray electron spectrum (1478.7 eV) should correspond to the $5f$ -electrons of uranium. Right next to it is the intrinsic "(U-F)-valence band of UF_4 " with a peak at 1474.2 eV. The next peak at 1462.4 eV corresponds to the $6p_{3/2}$ spin component of the orbital doublet ($6p_{3/2} - 6p_{1/2}$) of uranium. The other $6p_{1/2}$ -component of the doublet is located near 1452 eV and nearly coincides in energy with the more intense $2s$ -line of fluorine.

The peaks of the conversion spectrum coincide with the "valence band," $6p_{3/2}$ -peak and (F $2s$ -U $6p_{1/2}$)-double peak of the x-ray electron spectrum of UF_4 . The last peak in the conversion spectrum is also double. From comparison of the spectra it can be concluded that a conversion of the isomeric transition in the ^{235m}U case occurs into the electrons of the "valence band," the $(6p_{3/1})^4$, $(6p_{1/2})^2$ -electrons of uranium, and, apparently, into the $2s$ -electrons of fluorine.

The comparison of the x-ray electron and conversion spectra (see Fig. 2) makes it possible to determine the excitation energy of the ^{235m}U without resorting to the work function value of the sample and the binding energy of the electrons taking part in the conversion process of the isomeric transition. The energy balance for the excitation of any line in conversion and photoeffect processes can be written in the following manner:

$$E^* = T_c + E_{\text{bind}} + e\phi, \quad (1)$$

$$h\nu = T_{\text{ph}} + E_{\text{bind}} + e\phi, \quad (2)$$

where E^* is the excitation energy of the ^{235m}U , $h\nu$ is the energy of the excited x-radiation, E_{bind} is the binding energy of the electrons of the excited shell, $e\phi$ is the work function of the sample material, and T_c and T_{ph} are the kinetic energies of the conversion electrons and photoelectrons, respectively. It follows from Eqs. (1) and (2) that

$$E^* = h\nu + (T_c - T_{\text{ph}}). \quad (3)$$

In our experiments the x-ray electron spectrum of UF_4 was excited by monochromatized characteristic radiation of aluminum ($Al_{K\alpha}$), the energy of which is $h\nu = 1486.6$ eV. The difference $(T_c - T_{\text{ph}})$ measured from the results of a series of five measurements amounted to (1409.8 ± 0.5) eV. Thus, the energy of the uranium-235 isomeric transition $(1/2^+) \rightarrow (7/2^-)$, as determined by us, is 76.8 ± 0.5 eV.

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¹The x-ray electron spectrum is the spectrum of photoelectrons appearing when the sample is irradiated with x-rays.

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