$<E<3\,E_0$, the difference is practically constant and, for the case of *Pb* and $d=0.5\times10^{-13}$ cm, is of the order 0.20×10^{-13} cm. On the other hand the assumption that δE be constant would require

$$R=r_b+d\left[1-\left(\frac{r_b}{d}\right)\left(\frac{\delta E}{2E-E_0}\right)\right]. \quad \text{(A-9)}$$

Our uncertainty regarding these various assumptions and further, the result from optical analyses[19] that the nuclear potential itself appears energy dependent, cause us to refrain from using the above equations to correct the data. Nonetheless we think the analysis does provide a qualitative understanding of the observed angular and energy dependence of R.

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Transition in 26-min U^{235m} of Less Than 23 Electron Volts

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The conversion electron spectrum from the decay of 26-min U^{235m} has been examined by both magnetic spectrometry employing preacceleration and by electrostatic spectrometry. The energy distribution is peaked at 0.5 electron volts, with evidence of structure at 5 ev and at 19 ev, leading to an estimated upper limit of 23 ev for the transition energy. Tests of the performance of the apparatus were made with the conversion spectrum of the low-energy transition in 6-hr Tc^{99m}, redetermined as 2.15 kev; with the "zero energy" peak of Am^{241} and with the spectrum of photoelectrons excited from silver with ultraviolet light.

I. INTRODUCTION

THE existence of a very low-energy isomeric state in U^{235} following the unhindered 5.150-Mev α decay of Pu^{239} ($\frac{1}{2}+$) was recently discovered independently by Huizenga *et al.*[1] and by Asaro and Perlman.[1] The 26.16 ± 0.03 min isomeric decay[2] is thus expected to go by an $E3$ transition from a presumed $\frac{1}{2}+$ state to the (measured) $\frac{7}{2}-$ ground state of U^{235}.

That the very soft radiation was indeed coming from U^{235m} was established[1] both by chemical separation of uranium from Pu^{239} and by observing the decay of electrostatically collected recoils from the Pu^{239} α-decay. The radiation is detectable in a windowless Bradley *PCC*-11 proportional counter, which has, for this activity, a plateau 75 volts long of slope 10% per 100 volts. At the upper end of this plateau the counting rate exceeds 50% of the α counting rate (in 2π geometry) of the parent Pu^{239} sample from which the recoils are collected. This lower limit on the counter efficiency corresponds, as will be shown, to the counting of single electrons of energy less than 19 ev ejected per decay event. Identification of the radiation as very weak electrons was established by the observation that an absorbing film of ~2 μg/cm^2 of Formvar laid in intimate contact with the sample backing ranged out the count completely. For quanta above 1 ev such an absorber is over 90% transparent.

The first actual energy determination was made[3] in an internal sample proportional spectrometer in which it was shown that the pulse-height distribution from U^{235m} was nearly identical with that from single low-energy photoelectrons ejected from the wall of the counter with ultraviolet light. In the work reported here by magnetic and electrostatic spectrometry the radiation is seen to possess the magnetic rigidity and energy characteristic of negative electrons distributed in energy in the range 0–19 ev, peaked sharply at 0.5 volt.

II. SOURCE PREPARATION

Electrostatic collection of charged recoils from the α emission in Pu^{239} was used for preparing essentially weightless sources for this study. An "emanating" film of 8 mg of Pu^{239} evaporated on about 200 cm^2 of 1-mil Al foil folded into a 4-inch diameter cup constituted

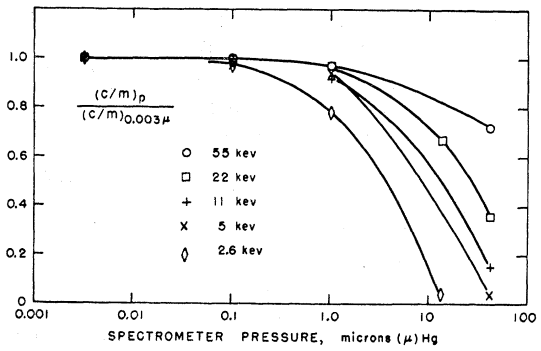

FIG. 1. The effect of spectrometer pressure on electron lines of various energies. The ordinate is the counting rate at any pressure relative to that at 0.003 μ Hg. The path length in the spectrometer is about one meter. The source of conversion lines is Am^{241}.

[1] Huizenga, Rao, and Engelkemeir, Phys. Rev. **107**, 319 (1957); I. Perlman (private communication).
[2] J. R. Huizenga (private communication).
[3] D. W. Engelkemeir (private communication).

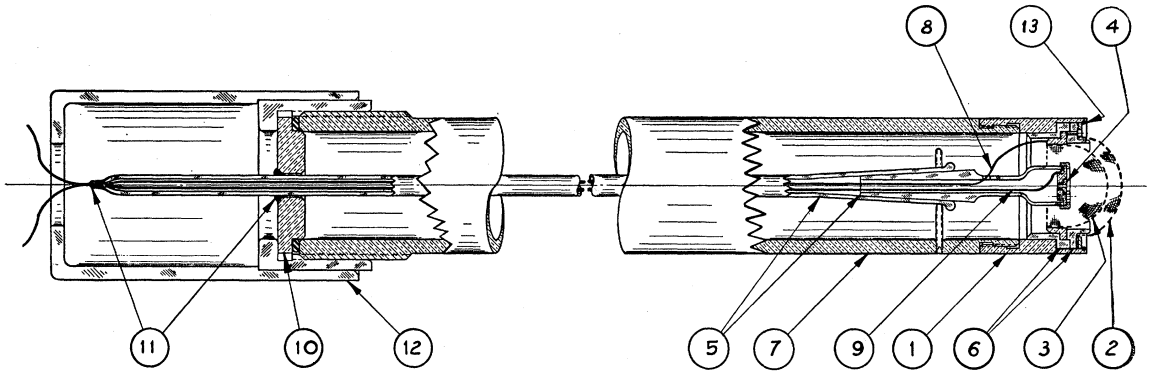

FIG. 2. Source preaccelerator and electrostatic analyzer (1) threaded end cap holding grid assembly; (2) grounded outer grid; (3*) insulated inner grid; (4) $\frac{1}{8}$-inch source disk supported on $\frac{3}{8}$-inch disk held with clamp ring on (5) quartz stem, enclosing high voltage lead to source and grid, supported on three radial screws near end of female joint; (6*) lucite insulators for inner grid; (7) brass tube, 2 ft long; (8*) inner grid contacting wire passing through quartz stem pump-out hole; (9) source-contacting wire; (10) gasket pressure plate; (11) wax vacuum seals; (12) Lucite cap; (13) retainer ring for grid assembly. Items marked with an asterisk were omitted in the first preacceleration experiments. The constant accelerating potential is applied between source and grounded outer grid; the analyzing potential is applied between source and inner grid.

the anode. Recoils were collected in a flowing helium atmosphere onto 3-mm diameter platinum or brass foil cathodes held near the center of the anode cup using 2–4 kilovolts, enough to limit the corona discharge to a few milliamperes. With this system, collection efficiency was erratic; at best $\sim 10^6$ disintegrations/min of U^{235m} activity, as measured on the Bradley counter, could be obtained in one hour. Such a source has about 10^{-7} of one atom layer of U^{235m} if uniformly deposited. The uniformity of the count from separate segments of the source shows this to be roughly the case. No α counts were collected, indicating the absence of aggregate recoil.

III. LOW-ENERGY MAGNETIC SPECTROSCOPY WITH PREACCELERATION

The Argonne double-lens spectrometer was adjusted for a transmission of 1% and a resolution of $<2\%$ for the 3-mm diameter sources. The adjustment of the earth's field degausser was checked by flip coil and by a one-meter long special oscilloscope tube focusing 500 volt electrons. A standard demagnetizing procedure with a fifteen-foot square coil and a variac was used before each run; we have found this necessary to eliminate the polarization of the steel paneled walls, floor and ceiling of the laboratory which is induced when high energies are focused in the spectrometer.

For detector we used an end-window, loop-wire, proportional counter, with propane gas (at a pressure of 8 cm Hg) flowing continuously. A double-layer water-spread Formvar-film counter window about 2–5 $\mu g/cm^2$ thick was supported on a 50% open area Lectromesh grid. The window cutoff was 800 ev. Although very good plateaus are obtained with this counter, especially for low-energy electrons, we ran most of the experiments at or below the plateau knee, at which voltage the sensitivity for minimum ionizing cosmic-ray secondaries is reduced. Thus the normal background of 8 counts/min is cut to about 3 counts/min, while counter performance is still stable.

With liquid nitrogen in the side arm trap, the pressure in the spectrometer was maintained at $(1–2)\times 10^{-6}$ mm Hg in these experiments. Figure 1 presents the effect of gas scattering on electrons of various energies focused through the one-meter spectrometer path as a function of gas pressure. At 2.6 kev a pressure of 10^{-4} mm Hg reduces the transmitted intensity by only 3% and this is near the lowest energy (2.0 kev) examined in the present experiments at fiftyfold lower pressure.

Source mount and preaccelerator are shown in Fig. 2. This figure shows the final design using two concentric grids (described later). For the first experiments, the inner grid, part No. 3, its insulating washers, No. 6, and its connecting lead, No. 8, were omitted. The 3-mm source was centered on a 1-cm metal disk mounted on the insulating quartz stem through which the wire lead was sealed. It was placed in the normal spectrometer source position. Accelerating voltages in the range 1–5 kilovolts between the source and the enclosing grounded 1.3-cm radius, 16-mesh hemispherical copper screen gave no increase in background count with the spectrometer focused to the accelerating energy.[4] An electrostatic voltmeter of 3000-volt range was calibrated with a precision resistive divider and Rubicon type B potentiometer to ± 3 volts at the values most used for acceleration.

Am^{241}

The performance of the system was tested with a volatilized ~ 30 $\mu g/cm^2$ Am^{241} source on Al. With 1.5 kv

[4] This absence of an instrumental "zero"-energy line enabled us to measure energies as low as zero. With thicker counter windows, 87 $\mu g/cm^2$, necessitating accelerating voltages of 10 or more kilovolts, a prominent "null-line" was observed by Schneider, Huber, Humbel, de-Shalit, and Zünti, Helv. Phys. Acta, **25**, 259 (1952), extending up to ~ 1 kev.

acceleration the line shapes of the L_3 line of the 26-kev transition and the L_1 line of the 33-kev transition at 8.75 and 10.79 kev, respectively, are unaffected, but the transmission increases by 20% compared to the no-acceleration case. The shift in line position agreed with the measured acceleration voltage within 15 ev.

With increasing accelerating voltage the "zero"-energy Am^{241} peak intensity increased, from zero at the 800-ev window cutoff to a saturation value at ~5 kev, tracing out the transmission curve of the Formvar window.[5] The electrons in this group may be partially undegraded disintegration Auger electrons and partly degraded higher energy primaries or secondaries generated in source or backing by alpha particles or gamma rays. The integrated intensity of the group is 20–40 times the total of all higher energy conversion lines, which suggests that they are mainly secondary electrons. The line shape showed a sharp low-energy dropoff and a long tailing off to the high-energy side, with a half-width several times that of a homogeneous line. This excessive width turned out to be due to the acceleration action on very low-energy electrons. The low-energy intercept occurred at the accelerating voltage plus 45 volts with the spectrometer calibration similarly taken from the low-energy intercept of the L_3 line of the 60-kev transition.

P^{32}

To obtain a rough idea of the intensity of a "zero-energy" degraded and backscattered peak from a sample of a pure high-energy beta emitter, a P^{32} source was deposited from solution on to a one-mil Al backing. The intensity of the zero-energy peak was only ~10% of the integrated continuum intensity and the zero-energy peak dropped off as rapidly on the high side as did that of Am^{241}. An accelerating potential of two thousand volts was used.

Tc^{99}

The 2-kev $E3$ transition in 6-hr Tc^{99m} was examined with an invisible volatilized Tc_2O_7 source. The spectrum is shown in Fig. 3 with zero and with 2000 volts acceleration illustrating the increase in window transmission for the approximately 1.6-kev conversion electrons. The transition energy is computed to be 2.15 (±0.03) kev, with relative conversion coefficients $M_{1+2+3}:M_{4+5}:N=6:1:0.9$ with, roughly, $M_1/M_{2+3}\approx 1$. Schneider *et al.*,[4] obtained 2.0 kev.

U^{235m}

Several attempts to detect the radiations of U^{235m} with the accelerator with up to 5 kv acceleration gave negative results, with samples which, as counted in the Bradley counter, should have given thousands of counts per minute in the spectrometer. The failure is ascribed to the lack of spherical symmetry of the accelerating system, in which the stronger field at the edges of the source mount pulls very low-energy electrons along a direction normal to the spectrometer axis, and away from the 15°–25° acceptance angle of the spectrometer.

Following a suggestion of D. Hutchison of this laboratory, we modified the accelerator by adding the intermediate hemispherical grid (Fig. 2) of $\frac{1}{8}$ in. smaller radius than the outer grid; the inner grid was held at the source potential. With this shield the low-energy electrons are undeflected in the field-free space until they reach the grid, and then the acceleration is sufficiently radial that they continue their original trajectories. With this grid the zero-energy peak in Am^{241} became symmetrical with the expected 2% spectrometer resolution, at all accelerating voltages, and the peak-value energy was determined to be 10±35 electron volts; the spectrometer was calibrated with the L_1 line of the 33-kev transition in the source. With U^{235m} sources in the device, one peak (Fig. 4), with the spectrometer resolution, was observed. The spectrum was surveyed magnetically in the range 0–4 kev initial energy, and a limit on the relative intensity of a line of energy above 50 ev of 0.003 may be set.

With an accelerating potential of 2165 volts[6] the peak

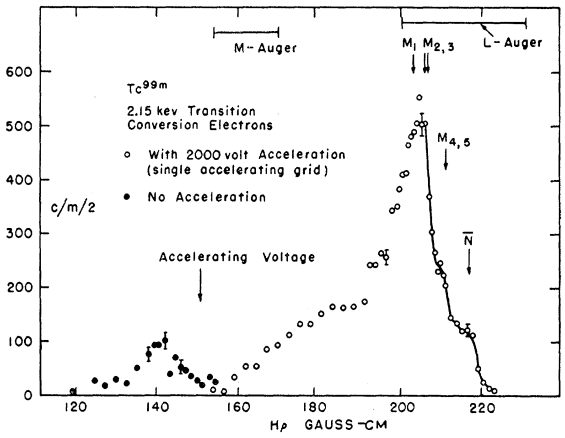

FIG. 3. Conversion electrons from the 2.15-kev transition in 6-hr Tc^{99m}. Arrows indicate the calculated positions of the lines originating from the indicated shells for a transition energy of 2.15 kev and an accelerating potential of 2000 volts at the source. The solid dots show the same lines with no acceleration, indicating the effect of the counter window whose cutoff is ≈800 ev. *L*-Auger electrons (resulting from conversion of the 140-kev transition) will be found in this region of the spectrum between 200 and 230 $H\rho$ (with 2000-volt acceleration) but their intensity is low (see reference 4) compared to 2.15-kev conversion. *M*-Auger electrons will be found in the region 154 to 170 $H\rho$. Note absence of peak at accelerating voltage.

[5] This increase in the peak rate, which follows the expected window transmission curve, together with the slight increase at 9 kev noted above, with 2-kv accelerating voltage, shows the absence, in our setup, of the very large increase in transmission observed by Schneider *et al.*[4] for very small initial energy.

[6] At 2 kv the window transmission is about 70%. At higher accelerating voltages with the double grid system, increases in

energy was determined as 8 ± 10 ev. The peak intensity corresponded to approximately one electron emitted per disintegration as computed from the observed count on the sample in the Bradley counter, and from the spectrometer luminosity corrected for accelerator, window and supporting grid transmission. All points on the peak (half-width=48 volts at 2165 volts) decayed with the 26-min lifetime.

IV. ELECTROSTATIC SPECTROMETRY

As no internal line standard for checking the spectrometer calibration was available in the U^{235m} sources, the accuracy of the energy determination was limited by possible instability of this factor and by error in measuring the accelerating voltage. Also the resolution of possible structures of the order of a few volts was limited by the 2% momentum resolution at 2165 volts.

To secure this more detailed information the accelerating system of Fig. 2, but with both grids made of 200-mesh stainless steel, was employed as an integral electrostatic spectrometer by connecting a separate lead to the inner grid to which, then, relatively small retarding (negative) voltages could be applied. With fixed accelerating voltage (2165 volts) between source and outer grid, electrons originating in the source with sufficient kinetic energy to surmount the potential barrier imposed by the inner grid will be focused in the spectrometer at their initial energy plus 2165 volts. For fixed initial energy the line position will be magnetically fixed and the intensity will drop more or less sharply with increasing retarding voltage, depending on the resolution of the electrostatic spectrometer. However, for electrons which originate at the inner grid, as secondaries from energetic radiation, the acceleration will increase with increasing retarding voltage and the line will shift upwards in the magnetic spectrometer but maintain constant peak intensity. This behavior enables one to distinguish the origin of the electrons. For energy ranges small compared to the resolution width of the magnetic spectrometer at 2165 volts, which is about 50 volts, the focusing current may be left fixed without materially affecting the detection efficiency for the electrons just being transmitted by the electrostatic spectrometer. This resolution is about 1.2% in momentum; the normal spectrometer resolution of 1.8% for a 3-mm source is improved with the electrostatic double-grid accelerator at low energies.

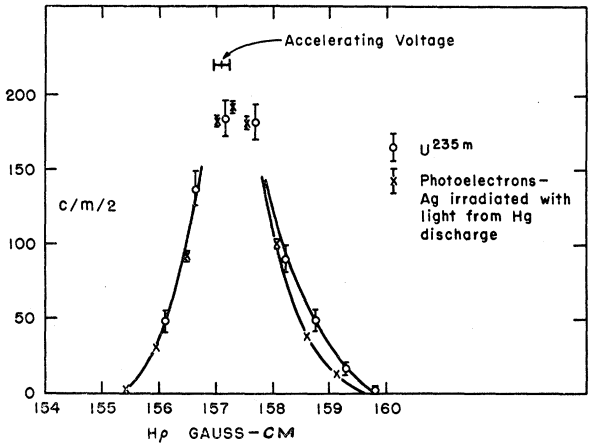

FIG. 4. The magnetic spectrometer analysis of the electrons from the decay of U^{235m} (open circles) with 2165 volts acceleration at the source. The position of the peak of a 2165 (±5) volt line computed from the spectrometer calibration is shown by the horizontal bar labeled "accelerating voltage." The crosses are obtained from the photoelectrons ejected from a silver disk in the source position on irradiation with the light from a mercury discharge. The intensity of the two lines has been normalized at the peak and a small shift ($+0.5$ $H\rho$) has been made in the position of the photoelectron peak to bring the left sides into coincidence for convenience in comparing the shapes. This shift represents an uncertainty in the spectrometer calibration constant of the magnetic spectrometer for the photoelectron source, due to a source position shift. The energy determination is actually derived from the electrostatic analysis. Note that the excess line width of the U^{235m}, less than 10 volts, is consistent with the electrostatic analysis.

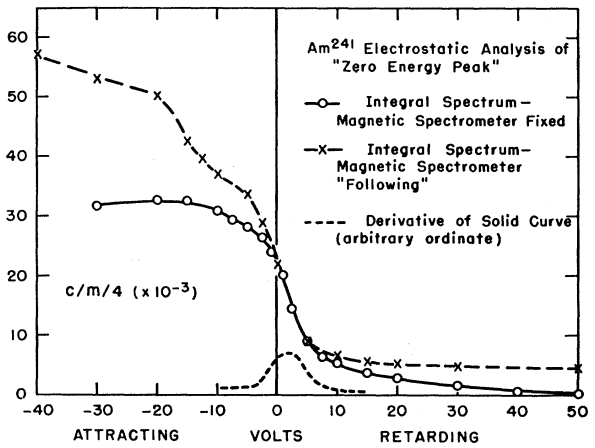

FIG. 5. Electrostatic analysis of the "zero-energy" peak in Am^{241}. The open circles (solid curve) describe the integral spectrum with the magnetic spectrometer current fixed to focus electrons whose energy is the main accelerating voltage. The crosses (dashed curve) describe the integral spectrum with the magnetic spectrometer setting varied to focus electrons whose energy is the main accelerating voltage plus the "retarding" voltage; thus the magnetic spectrometer "follows" the momentum associated with the electrons which originate at the inner grid as secondaries produced by alphas, gammas, and higher energy conversion electrons. For an explanation of the curve shape at attracting voltages, see text. The dotted curve is the derivative of the solid curve showing the energy distribution of the Am^{241} "zero-energy" peak, whose maximum is at two volts.

Am^{241}

Figure 5 exhibits the "zero-energy peak" integral intensity of Am^{241} as a function of retarding voltage. The solid curve is the intensity observed with the magnetic spectrometer focus fixed at 2165 volts and the dashed curve gives the maximum intensity as the

background of several counts/min were observed, which were shown to be due to electrons originating at the inner grid as "corona."

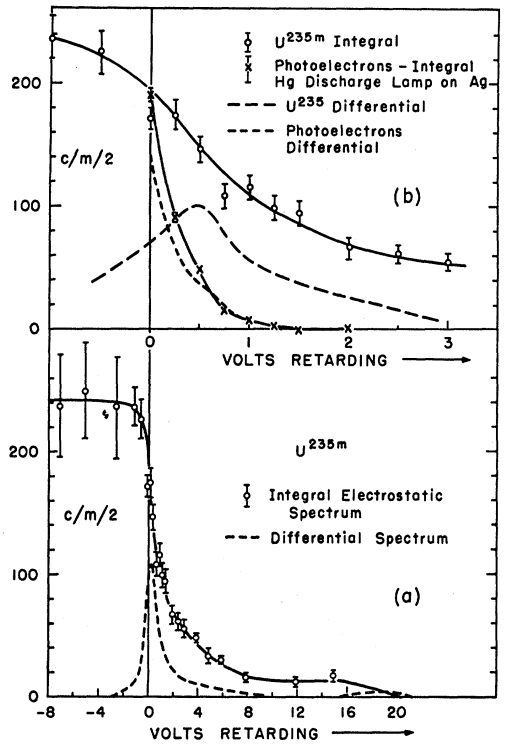

FIG. 6. (*a*) Electrostatic analysis of the electrons emitted in decay of U^{235m}. Dashed curve shows the differential distribution. (*b*) Same data on an expanded energy scale. The crosses and the short-dash curve are integral and differential plots of the electrostatic analysis of photoelectrons ejected from a silver disk in the source position on irradiation with a mercury discharge lamp. The comparison gives some idea of the effect of the instrumental resolution on the U^{235m} spectrum. *Note added in proof.*—An experimental point at 20 volts "retarding" and intensity of 1 ± 2 c/m/2 was inadvertently omitted from the figure.

magnetic spectrometer is shifted in focus to follow the retarding voltage. The constant intensity observed on the shifting line to the right (dashed) indicates the presence of some secondaries generated at the inner grid with zero energy. Differentiation gives the energy distribution of the dotted curve,[7] which is peaked at 2 ev in agreement with the results of magnetic spectroscopy.

The curves to the left of zero give the results with the inner grid voltage positive. The continuation of the rise in intensity with small positive voltage (<1 volt) may be in part due to the finite resolution of the electrostatic spectrometer and partly to the existence of a contact potential between source mount and inner grid, if its polarity retards electrons. For low-energy secondary electrons ejected backwards towards the source from the inner grid, increasing "attracting" voltages should increase the collection efficiency into the accelerating field between the two grids. This effect probably accounts for the increased rate on the "following" curve at −20 to −50 volts.

[7] In the first electrostatic spectrometer measurements on Am^{241} and U^{235m}, coarser 16-mesh screens were used. With these a much broader differential distribution was obtained, peaked at ~25 volts, presumably due to accelerating-field "leak" through the coarser grids. We are indebted to D. W. Connor for pointing out this effect.

U^{235m}

Figure 6 presents the analogous results for U^{235m} in which data from several runs is summed. Here no evidence of a shifting peak due to secondaries appears, and a narrower differential distribution is obtained than for Am^{241}. Decay of the counting rate was observed to follow a 26-min half-life at 0, 4, and 6 volts. The low counting rates made it difficult to verify the decay of the points beyond 6 volts but the straightness of the 6-volt decay curve suggests that the points up to 19 volts are due to U^{235m} decay rather than "corona." In agreement with the magnetic spectroscopy, the distribution maximum occurs at 0.5 ± 0.2 electron volts. There is evidence of structure in the distribution near 5 volts and perhaps at 19 volts which we judge to be intrinsic to U^{235m}, and not due to instrumental effects, by comparison to the photoelectron spectrum (see below). In some runs there was some evidence of a shifting line of low intensity, several counts/min, above about 2–3 volts ascribed to corona between the grids, but not in the "best" runs.

Calibration

To determine whether the observed electron spectrum of U^{235m} was homogeneous in energy and whether any effects not considered could shift the apparent energy, the resolution curve of the electrostatic spectrometer was approximately determined by measuring the spectrum of low-energy photoelectrons. These were generated on a $\frac{1}{8}$-in. silver disk at the source position by light from a quartz mercury arc admitted to the magnetic spectrometer through a quartz window. Consideration of the photoelectric work function of silver and the intensity distribution of the mercury lamp indicates the most probable photoelectron energy to be within $\frac{1}{2}$ volt of zero. A narrower distribution than for U^{235m} was indeed obtained, peaked at zero volts (Fig. 6, dashed curves).

Inspection of Fig. 6 shows that application of a resolution correction based on the photoelectron spectrum will obviously result in a spectrum with peaks located approximately as indicated above. In consideration of experimental uncertainties, there is little point in attempting the correction; we merely conclude that a nearly continuous distribution of U^{235m} decay electrons peaked sharply at $\frac{1}{2}$ volt but extending at least to 19 volts was observed, and that this apparent spread in energy represents, in the main, an intrinsic distribution and not an instrumental irresolution.

V. DISCUSSION

To the measured electron energies must be added appropriate binding energies to obtain transition

energies. These electron binding energies must be those appropriate to orbitals in an uranium atom (or uranium oxide molecule) bound to the metal substrate. Even for the free atom the binding energies in the outermost shells are not well established, but for such a bound atom the minimum binding energy would appear to be the electronic work function of the surface. Though the orbitals of the ejected electrons are not known, it is clear that conversion can only be occurring in the P and Q shells in which binding energies for the free atom range from a few up to about 50 ev. Since only about one electron is ejected per disintegration there must be few Auger electrons accompanying each conversion electron. If we take the upper energy limit of the electrons as 19 ev, we may reasonably assume it to correspond to a conversion electron from the least bound shell and add only about 4 ev for the metal work function to obtain an upper limit of 23 ev for the transition energy.

For the decay of a $\frac{1}{2}+$ state of 23-ev excitation to a $\frac{7}{2}-$ state by $E3$ transition, the theoretical lifetime for photon emission is $\sim 10^{15}$ years, if one uses Weisskopf's formula. The experimental lifetime of 26 minutes corresponds to a conversion coefficient $\alpha_3 = 10^{19}$. For whatever the comparison may be worth, one notes that the theoretical value for the threshold K conversion coefficient for hydrogen (13.6 ev transition energy) for $E3$ transition is[8] $\alpha_3 = 2\times 10^{15}$.

[8] B. I. Spinrad, Phys. Rev. **98**, 1302 (1955).

As stated in the introduction, no photon counts were observed through a 2-μg/cm^2 optically transparent absorber. However, the process of conversion in the outer shells might reasonably be expected to be accompanied by the emission of photons in the optical or ultraviolet range, of the order of one such photon per electron. A source of U^{235m} on a 3-mm brass disk was placed on an Eastman 103 a-F ultraviolet sensitive plate for one hour. No image was observable on developing, despite over 10^7 disintegrations having irradiated the emulsion. As a spot should be visible with about 10^8 photons on this area, the result is credible. Examination of the photon spectrum with a photomultiplier cooled to liquid nitrogen temperature should be undertaken.

For the anomalous rotational band based on the $\frac{1}{2}-$ level ($\frac{3}{2}-$ level at 12.5 kev, $\frac{5}{2}-$ level at 50.8 kev),[9] one calculates a decoupling parameter[10] $a = -0.628$.

It is interesting to consider the experimental possibility of exciting this very low-lying state by Coulomb excitation by using moderate-energy intense beams of ions or electrons.

ACKNOWLEDGMENTS

Mr. Howard Ruhde of the Metallurgy division kindly prepared the evaporated plutonium emanating source.

[9] K. N. Shliagin, Zhur. Eksptl. i Teort. Fiz. **30**, 817 (1956) [translation: Soviet Phys. JETP **3**, 663 (1956)].

[10] A Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 16 (1953).

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Excited States in Mn^{56}†

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An electrostatic accelerator and a broad-range magnetic spectrograph were employed to investigate the excited states of Mn^{56} by means of the $Mn^{55}(d,p)Mn^{56}$ reaction. The proton groups from thin manganese targets were observed at 10, 30, and 60 degrees with an incident deuteron energy of 7.0 Mev and at 20 degrees with 6.6-Mev deuterons. In the region of excitation in Mn^{56} up to 7 Mev, 124 excited states were found. The ground-state Q-value for the reaction is 5.047±0.005 Mev, and the first excited state is at 0.025±0.004 Mev.

FOR some time, this laboratory has been engaged in a general program of nuclear spectroscopy, and recently much of the research has been concerned with the determination of the energy-level schemes of the nuclei between mass numbers 40 and 70. The present paper reports the results of studies on the excited states of Mn^{56} carried out through the $Mn^{55}(d,p)Mn^{56}$ reaction.

Previous investigations of the bound states of Mn^{56} have been carried out through the (d,p) and (n,γ) reactions. The results of these studies have been summarized,[1] and they indicate the existence of sixteen

† This work has been supported in part by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

* Lieutenants, U. S. Navy.

‡ On leave from the National University of Mexico.

[1] *Nuclear Level Schemes*, $A = 40$–$A = 92$, compiled by Way, King, McGinnis, and van Lieshout, Atomic Energy Commission Report TID-5300 (U. S. Government Printing Office, Washington, D. C., 1955).