

are also inappropriate for these nuclei.

Because of the large number of missed levels, resonance-parameter distributions and average values of resonance parameters obtained by multilevel fitting of the low-energy resonances of fissile nuclei can be unreliable. It appears that a more reliable approach for obtaining certain average resonance parameters is by a statistical comparison of experimental and simulated cross sections. Such an approach has been presented recently.<sup>9</sup>

I wish to express appreciation to J. Blons, D. R. Mathews, M. S. Moore, L. W. Nordheim, and G. deSaussure for help and discussion on certain aspects of this and related work. The University Computer Center under Robert Swanson has been most generous in its time and assistance.

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## Perturbation of the $^{235m}\text{U}$ Decay Rate by Implantation in Transition Metals

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(Received 18 August 1972)

When implanted in transition metals, the decay rate of  $^{235m}\text{U}$  is influenced by the free-electron concentration of the host metal. The results can be qualitatively described by means of the "rigid-band" and "screened-potential" models.

The  $^{235}\text{U}$  nucleus has a  $\frac{1}{2}^+$  isomeric state decaying by an  $E3$  transition to the  $\frac{7}{2}^-$  ground state with a decay constant  $\lambda$  of about  $0.026 \text{ min}^{-1}$ . The transition energy has the exceptionally low value of  $73 \pm 5 \text{ eV}$ , and proceeds by internal conversion in the outermost electron subshells, most probably the  $(6p_{1/2})^2$ ,  $(6p_{3/2})^4$ , and  $6d$  shells (or  $P$  II, III, and IV shells, respectively) of uranium.<sup>1</sup> These shells, and consequently the isomeric decay rate, can be perturbed by changes in the valence-electron subshells  $(5f)^3(6d)(7s)^2$ . In a previous publication,<sup>2</sup> we have shown that indeed the decay constant changes with the metal into which the  $^{235m}\text{U}$  nuclei are implanted. We present here more accurate and more numerous results, showing that this effect is due to the interaction between the outer atomic electrons of the  $^{235m}\text{U}$

impurity and the free electrons of the metallic environment and not to some chemical effect as proposed earlier.<sup>2</sup>

The isomeric level is fed by the  $\alpha$  decay from  $^{239}\text{Pu}$ , thus a  $^{235m}\text{U}$  source is obtained by collecting the recoils from a thin  $^{239}\text{Pu}$  deposit. If the collector foil and the Pu layer are both under high vacuum, the  $^{235m}\text{U}$  nuclei keep their recoil energy ( $\leq 90 \text{ keV}$ ), and penetrate into the collector with a range of about 100 to 250 Å, depending on the metal used. In our previous study,<sup>2</sup> the activity of the  $^{235m}\text{U}$  sources was followed with a proportional flow counter, the transfer between the collector position and the counter being done under Ar (1 atm). This procedure has several drawbacks, the main one being the possibility of surface oxidation and gas adsorption by the col-

lector during the transfer and the subsequent counter operation.

The results presented here have been obtained with an improved experimental setup.<sup>3</sup> The Pu deposit and the counter are within the same vacuum vessel, and a rotary motion allows the transfer from the collecting to the counting position without breaking the high vacuum (about  $5 \times 10^{-7}$  Torr). The counter is a Channeltron electron multiplier (Bendix CEM-4039). This setup not only avoids the drawbacks mentioned above, but has the additional advantages of excellent counter stability and increased counting rate due to the lower electron detection threshold of a Channeltron as compared to a proportional counter. Initial intensities ranged from 40 to 200 cm sec, depending on the collector used, with a background of 0.3 to 0.6 cm/sec. As a check of the overall stability and reproducibility, it was found that the external error of a series of consecutive measurements (each collector was measured at least five times) was about equal to the internal error, i.e., the average of the statistical errors of the individual measurements. As the total number of counts registered per mea-

surement reaches values of 1 to  $5 \times 10^5$ , one can easily measure  $\lambda$  with an accuracy of 0.1% to 0.2% (standard errors).

The metallic foils used as collectors were chemically etched, except for Rh, Ir, Pt, and Au, which were mechanically polished. It was found that the surface state of the collector is not very critical,<sup>4</sup> indicating that we are detecting almost exclusively implanted nuclei, and not those lying eventually on the surface. It should be added that the total number of  $^{235m}\text{U}$  recoils collected remains well below the threshold value for any interaction between the implanted U atoms themselves.

The experimental decay constants of  $^{235m}\text{U}$  implanted in transition metals are shown in Figs. 1 and 2. It is expected<sup>5</sup> that  $\lambda$  will be mainly influenced by the free-electron concentration of the host metal; thus the results are displayed on Fig. 1 as a function of the atomic concentration on a doubly logarithmic scale. "Size effects" may also occur when the atomic radii of host and guest atoms differ by more than about 15%. Inspection of the metallic radii<sup>6</sup> shows that, in the cases studied here, this effect should be weak or absent. It also implies that the implanted U impurities will mainly occupy substitutional positions.

For the noble metals (group Ib), the atomic concentration  $n_a$  is, in principle, equal to the free-electron concentration  $n_e$ . However, the actual situation may be slightly different. Information about it can be derived from a comparison between the experimental Hall constants ( $R_H$ )<sub>obs</sub>, and the calculated ones based on the hypothesis of one free electron per atom. In Fig. 1(a) the  $\lambda$ 's obtained for Ag and Au are given for two

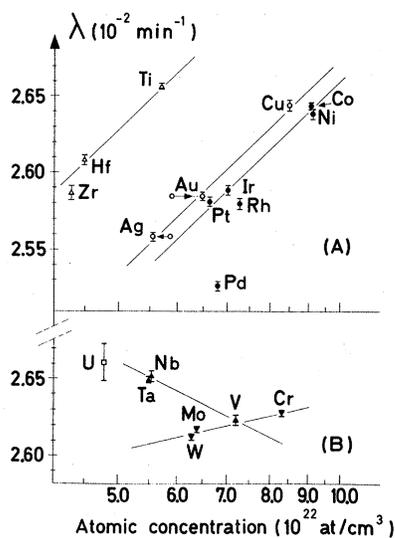


FIG. 1. Decay constant of  $^{235m}\text{U}$  implanted in various transition metals versus the atomic concentration of the host metal. (a) Open circles, group Ib (monovalent elements); closed circles, group VIII (divalent and trivalent elements); open triangles, group IVb (quadrivalent elements); (b) closed triangles, group Vb (pentavalent elements); inverted closed triangles, group VIb (hexavalent elements). For Au and Ag, the data points without error flags correspond to the nominal atomic concentrations (=nominal free-electron concentrations) and the points with error flags, to electronic concentrations deduced from the Hall coefficients (see text).

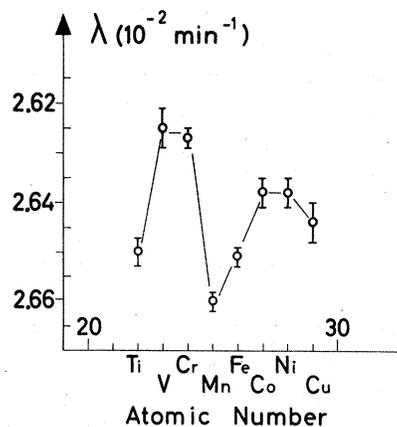


FIG. 2. Decay constant of  $^{235m}\text{U}$  implanted in the transition metals.

atomic concentrations: one corresponding to the nominal atomic concentration, and the other to the concentration deduced from  $(R_H)_{obs}$  for Au, Ag, and Cu and normalized to one free electron per Cu atom. It is seen that with this correction, a straight line can be fitted through the  $\lambda$ 's of Cu, Ag, and Au within the experimental errors.

For bivalent Ni and Pt and trivalent Co and Ir of group VIII, and quadrivalent Ti and Hf of group IVb, it is seen that a straight line with the same slope as for the Ib group can be drawn through the data. The apparent lack of fit of Zr, Rh, and Pd might be due to either one or several of the following effects: (i) a discrepancy with respect to the assumption underlying Fig. 1, i.e.,  $n_a = n_e$ ; (ii) an anomalous distribution of the implanted U nuclei between interstitial and substitutional positions; (iii) improper surface etching.

For the pentavalent Vb (V, Nb, Ta) and hexavalent VIb (Cr, Mo, W) groups, the effect of the atomic concentration on  $\lambda$  is much weaker [see Fig. 1(b)]; for the Vb group, the trend of the effect is even reversed as compared to the Ib group. For comparison,  $\lambda$  for U metal is also shown; the statistical accuracy is low because of the high background activity of the U collector; in addition to this, a systematic error due to surface oxidation should probably be added.

For metals with valencies higher than 1, it is not possible to use  $(R_H)_{obs}$  for obtaining more correct values for  $n_e$ , as discussed above for the noble metals. Blokhin, Satchenko, and Nikiforov<sup>7</sup> have deduced  $n_e$  values for some of the transition metals of the 3d period from the relative intensity of the  $K\beta_5$  x-ray lines, but for the present purpose it is meaningless to use these results for correcting only the 3d element of a given group, and not the 4d and 5d ones.

It is known that the electronic interaction between a metal and an impurity atom depends on the relative number of valence electrons of the host and guest atoms.<sup>5</sup> When the perturbation is small, the "rigid-band" model can be applied; this is the case when the difference in valencies between the host atom and a substitutional impurity is either 0 or 1. When this difference is larger than 1, the free-electron concentration in the immediate vicinity of the impurity will be modified in such a way that the local perturbation is canceled, i.e., the impurity is screened by the electron gas. This screening has a relaxation length (usually called the screening length) which is proportional to  $(n_e)^{-1/6}$  and has dimensions of the order of the atomic radius of the impurity.

The results obtained with  $^{235m}\text{U}$  (mainly hexavalent atom) can be qualitatively explained with these "rigid-band" and "screened-potential" models. When the valency difference is equal to or larger than 2 [Fig. 1(a)], the screening effect dominates; the data for the monovalent Ib group indicate that the isomeric transition probability is proportional to the power  $1/(12.81 \pm 0.94)$  of the free-electron concentration, i.e.,

$$\lambda(^{235m}\text{U}) \propto (\text{screening length})^{-1/2}.$$

When this difference is 1 or 0 [Fig. 1(b)], the valence electrons of U are taken up in the conduction band of the host metal; in this case, the net effect on  $\lambda(^{235m}\text{U})$  is due to the detailed interaction of the conduction band on the valence and conversion electron subshells of  $^{235m}\text{U}$ . The decay of  $^{235m}\text{U}$  nuclei implanted in the transition metals of the first period (Fig. 2) illustrates this point<sup>8</sup>: The variation of  $\lambda$  with atomic number displays a double hump which is probably due to the well-known splitting of the 3d band.<sup>5,9</sup>

Further inspection of Fig. 1(a) indicates that, except for Pd, the results obtained with all the monovalent, divalent, and trivalent metals are grouped together, while those for the quadrivalent metals form another distinct group. This may be due to the fact that U can eventually be quadrivalent, thus implantation in quadrivalent transition metals may lead to a peculiar situation, being intermediate between the extreme "rigid-band" and "screened-potential" models.

It is difficult at present to make a quantitative evaluation of these effects, as the theoretical conversion coefficients of the  $^{235}\text{U}$  isomeric transition are not known.

The author thanks Mme. G. De Corte for excellent computer work, and M. L. Vansteelandt, M. E. Mies, and their colleagues for very reliable technical support.

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<sup>3</sup>A detailed report on the experimental techniques is being prepared for publication. We only mention here the collecting geometry, which has to be close to  $2\pi$  for reaching adequate counting rates. The distance between collector and Pu deposit was 1 mm, the diameter of the Pu deposit was 25 mm, and the diameter of the collectors ranged from 10 to 20 mm. Under these conditions, the distribution of the implanted  $^{235m}\text{U}$  nuclei has a maximum within a few angstroms below the collector

surface and decreases almost linearly up to the range value.

<sup>4</sup>The following checks were performed: (i) changing the etching or cleaning procedure of the collector surface; (ii) changing the threshold energy of the electrons detected from 0 eV up to 15 eV by means of a properly biased control grid located between the collector and the Channeltron input, so as to detect preferentially the less deeply implanted <sup>235m</sup>U. No measurable difference in decay rate was found, while differences of up to 5% were found with unclean collectors, e.g., for Pt with traces of hydrocarbons on the surface:  $\lambda(0 \text{ eV}) = +2.583 \pm 0.006 \times 10^{-2} \text{ min}^{-1}$  and  $\lambda(15 \text{ eV}) = (2.445 \pm 0.029) \times 10^{-2}$

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<sup>8</sup>It may be noticed that the relative variation of  $\lambda(^{235m}\text{U})$  between the two extreme cases studied here, namely, Mn [ $\lambda = (2.6613 \pm 0.0020) \times 10^{-2} \text{ min}^{-1}$ ] and Pd [ $\lambda = (2.5269 \pm 0.0030) \times 10^{-2} \text{ min}^{-1}$ ] amounts to (5.2  $\pm$  0.2)%.

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### <sup>4</sup>He( $\gamma, p \pi^-$ ) Cross Section Around $\Delta(1236)$

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(Received 31 March 1972)

The cross section of the reaction  $^4\text{He} + \gamma \rightarrow p + \pi^- + (ppn)$  has been measured in the region of the first resonance for various values of the recoil momentum. An anomaly is observed for high values of this momentum and a tentative explanation is suggested.

The experiment described in this Letter was performed in order to understand how pion photoproduction on a quasifree nucleon in <sup>4</sup>He is affected by the proximity of the other nucleons, near the  $\Delta(1236)$  resonance.<sup>1</sup>

The central idea of this experiment was to compare the photoproduction cross section

$$\gamma + ^4\text{He} \rightarrow p + \pi^- + (ppn), \quad (1)$$

for a given recoil momentum  $\vec{P}_R$  of the residual ( $ppn$ ) nucleus, to the elementary one  $\gamma + n \rightarrow p + \pi^-$ , because in these reactions we could easily detect the two emitted products.

For low values of  $P_R$  the probability of finding the neutron far enough from the  $A - 1$  other nucleons is large, and the nucleus may be described correctly by the independent-particle model. Thus, we may think that the impulse approximation is valid. In this model we have  $-\vec{P}_R = \vec{P}_0$ , where  $\vec{P}_0$  is the momentum of the target neutron before the interaction. Then the cross section for the Reaction (1) as a function of the invariant mass  $Q$  of the proton-pion pair can be accurately predicted from the cross section for the elementary reaction and the nucleon momentum distribution  $\mathcal{O}(P_0)$ .

As we were interested in looking for a departure from the predictions of this model, we decided to study the photoproduction at high values of  $P_R$ .

Our experimental setup permitted us to determine  $Q$  and  $\vec{P}_R$  by measuring the pion and the proton four-momenta in two magnetic spectrometers when the proton, the pion, and the photon lie in the same plane. The spectrometer for the pion could analyze a maximum momentum of 400 MeV/ $c$  and had a momentum acceptance of 6%. The corresponding figures for the proton spectrometer were 700 MeV/ $c$  and 12%. The particles were detected in each focal plane by a counter telescope consisting of two plastic scintillators. The particle identification was made by means of the energy losses and by the difference in time of flight. The target was 0.65-g/cm<sup>2</sup> liquid helium. The low counting rate of this coincidence experiment requires the use of a high-intensity photon beam with a duty cycle as high as possible: The Saclay linear accelerator facility offered us these conditions.<sup>2</sup> The bremsstrahlung beam intensity was measured by a Wilson-type gas quantummeter. We estimate that the systematic error in absolute value is less than 10%.

The kinematics of (1) are governed, in the lab frame, by the following equations:

$$\vec{\nu} = \vec{P}_p + \vec{P}_\pi + \vec{P}_R, \quad (2)$$

$$\nu^0 + M_{4\text{He}} = P_p^0 + P_\pi^0 + M_{3\text{He}} + T_R + E_R, \quad (3)$$

where  $M_{4\text{He}}$  and  $M_{3\text{He}}$  are the ground-state masses of <sup>4</sup>He and <sup>3</sup>He; ( $\vec{\nu}, \nu^0$ ), ( $\vec{P}_\pi, P_\pi^0$ ), and ( $\vec{P}_p$ ,