# DIRECT DETERMINATION OF THE LIFETIME OF EXCITED COMPOUND NUCLEI BY ANGULAR DISTRIBUTION MEASUREMENTS OF FISSION FRAGMENTS EMITTED FROM SINGLE CRYSTALS

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The lifetime for decay of excited $Np^{238}$ compound nuclei has been determined by measuring the emission distribution of fission fragments from a uranium oxide single crystal bombarded with 10-MeV protons. The proton energy, and with this the compound nuclei excitation energies, were selected to give second-chance fission $Np^{238}$ compound nuclei with lifetimes in the range detectable by this technique.

The angular distribution upon emergence from single crystals of positively charged particles scattered or emitted from crystalline lattice positions shows a regular pattern of minima for emission parallel to low-index atomic axes or planes. This phenomenon has been observed for positrons, protons, $\alpha$ particles, and fission fragments, and the general characteristics of the observed effects are described by the detailed theory of Lindhard.[1]

For emission originating from nuclei displaced from the equilibrium lattice sites, the angular distribution can be used to determine the displacement. The distance that can be measured in this way is of the order of $10^{-8}$ to $10^{-9}$ cm. From the angular distribution of charged particles emitted by decay of a compound nucleus formed by nuclear reaction in a lattice position, it is therefore possible from the known recoil velocity to obtain direct determination of the lifetime. Since the recoil velocity can be adjusted from $10^6$ to $10^9$ cm/sec, this time-of-flight technique can in principle be used to measure lifetimes of the order of $10^{-14}$ to $10^{-18}$ sec.

The possibility of utilizing angular distribution measurements in this way for lifetime measurements was suggested several years ago.[2-4] Recently, Brown, Marsden, and Werner[5] have reported on the use of this technique to set a limit on the lifetime of compound nuclei formed by bombardment of $U^{238}$ with 12-MeV protons. The purpose of this paper is to report on an experimental determination of the lifetime of $Np^{238}$ compound nuclei formed by bombardment of $U^{238}$ with 10-MeV protons and to set a more precise upper limit on the "effective lifetime" of compound nuclei formed by 12-MeV bombardment of $U^{238}$.

Changes in the mean decay position affect both the angular width, $\psi_{1/2}$, and the minimum yield, $\chi$, of the angular distribution.[6] The interatomic potential normal to a row of atoms can be approximated by a screened Thomas-Fermi potential which decreases rapidly in the region of the Thomas-Fermi screening radius, $a$, and has an effective cutoff at a distance, $r_c$, which is a few times larger than $a$. The contribution to the minimum yield by fragments emitted from recoiling compound nuclei can be effectively separated into two components, those emitted within a distance $r_c$ of the row and those emitted outside $r_c$. Particles emitted within $r_c$ in a direction parallel to the row are deflected by the row potential. Such particles have a contribution $\chi_2$ to the minimum yield,

$$\chi_2 = C\frac{\rho^2}{r_0^2} + C\int_0^{r_c} \frac{x^2}{r_0^2} e^{-x/v_\perp \tau} \frac{dx}{v_\perp \tau}. \quad (1)$$

The first term arises from displacements due to lattice thermal vibrations with mean square amplitude $\rho^2$. The second term accounts for a displacement $x$ due to recoil of an exponentially decaying compound nucleus having recoil velocity $v_\perp$ perpendicular to the row and mean lifetime $\tau$. The area associated with each row in a plane perpendicular to the row is $\pi r_0^2 = 1/Nd$, where $N$ is the atomic density in the crystal and $d$ is the atomic spacing along the row. This expression is derived from the Lindhard theory[1] based on a continuum approximation to the row potential which underestimates the minimum yield so the constant $C$ has been included as suggested by Barrett.[7] $C$ can be assumed to be constant only for small values of the displacement. Barrett has found for 400-keV to 5-MeV protons in aluminum and tungsten crystals over a wide temper-

ature range that $C$ was always in the range 2 to 3.[7] Particles emitted outside of $r_c$ can move freely in the row direction and will therefore contribute to the minimum yield by their fractional probability which is $\chi_3 = e^{-r_c/v_\perp \tau}$. In addition, as discussed by Lindhard,[1] it is necessary to include a contribution $\chi_\perp$ to the minimum yield arising from random scattering effects in the crystal or at the crystal surface. The observed minimum yield is $\chi = \chi_1 + \chi_2 + \chi_3$, or

$$\chi - \chi_1' = \frac{2C v_\perp^2 \tau^2}{r_0^2}\left[1 - \left(1 + \frac{r_c}{v_\perp \tau}\right)e^{-r_c/v_\perp\tau} - \frac{1}{2}\left(\frac{r_c}{v_\perp\tau}\right)^2 e^{-r_c/v_\perp\tau}\right] + e^{-r_c/v_\perp\tau}. \quad (2)$$

The correction term $\chi_1'$ contains the thermal vibration effects on Eq. (1) as well as random scattering effects. The first term on the right corresponds to particles emitted within $r_c$ and the second to particles emitted outside $r_c$. For minimum yield changes of more than a few percent, the second term will dominate.

In the present experiment, uranium-dioxide crystals were bombarded with protons in the energy range from 9.0 to 12.0 MeV at the Bell Laboratories–Rutgers University tandem Van de Graaff. Integrated currents of ~30 nA h to ~150 nA h were used. The crystals were cleaved from larger crystals with the ⟨111⟩ axis normal to the crystal surface. A 1-mm diameter proton beam was incident on the target, and the fission fragments were detected with polycarbonate and cellulose acetate films,[8] etched in 6$N$ NaOH under conditions which made the fission-fragment tracks visible, but not tracks due to scattered protons or other light particles resulting from decay or nuclear reactions in the target. The crystals were oriented relative to the incident proton beam such that a ⟨110⟩ axis was 5° from the incident-beam direction, two other ⟨110⟩ axes were 60° from the beam, and a ⟨111⟩ axis (normal to the crystal surface) was 40° from the beam direction. The angular distributions were determined by counting the number of fission-fragment tracks as a function of position on the plastic film in an optical scanning microscope. Each axial direction was scanned in two orthogonal directions chosen such that the scan was not along low-index planar directions.

In these measurements, the observed fission fragments arose from decay of both the primary $Np^{239}$ and the secondary $Np^{238}$ compound nuclei. For this case,

$$\chi - \chi_1' = \left[\frac{1 + (\Gamma_n/\Gamma_f)_2}{1 + (\Gamma_n/\Gamma_f)_1 + (\Gamma_n/\Gamma_f)_2}\right]\chi(v_\perp\tau_1) + \left[\frac{(\Gamma_n/\Gamma_f)_1}{1 + (\Gamma_n/\Gamma_f)_1 + (\Gamma_n/\Gamma_f)_2}\right]\chi(v_\perp\tau_2). \quad (3)$$

The functions $\chi(v_\perp\tau)$ refer to the right-hand side of Eq. (2) and effects of neutron emission on the mean square recoil velocity have been neglected. $\tau_1$ and $\tau_2$ represent the total lifetime for decay of the primary and secondary compound nuclei, respectively. The expressions in brackets are the fraction of the total number of fissions coming from the primary and secondary compound nuclei, respectively, expressed in terms of the branching ratios, $(\Gamma_n/\Gamma_f)$.

From the fission and neutron level-width analysis of Huizenga and Vandenbosch,[9] very short mean decay times ($<10^{-17}$ sec) are expected for proton energies of 12 MeV and higher (up to ~15 MeV) since both the primary $Np^{239}$ and the secondary $Np^{238}$ compound nuclei have high excitaion energies (relative to the estimated fission-threshold energy of ~6.0 MeV). Fast decay is also expected for proton energies of 9.0 MeV or lower since all of the fission fragments come from highly excited $Np^{239}$ compound nuclei. Figure 1 shows the result of a scan of a ⟨111⟩ axial direction for a 12-MeV bombardment. Similar results with somewhat poorer statistical accuracy were obtained for the 9-MeV bombardments.

If it is assumed, as done by Brown, Marsden, and Werner,[5] that the observed minimum yield arises only from the "lifetime terms," i.e., if the scattering term $\chi_1'$ in Eq. (3) is neglected, the above measurement can be used to set an upper limit on the lifetime. The minimum yield of 0.16 from Fig. 1 gives an "effective lifetime"[10] of $\tau_{\text{eff}} \leq 1.3\times10^{-16}$ sec when Eq. (2) is used for $C = 2.5$ and $r_c = 4a \approx 0.4$ Å. This limit is higher than the limit of $<2\times10^{-17}$ sec reported by Brown, Marsden, and Werner.[5] The difference is principally due to the smaller value of the cutoff distance $r_c = 0.1$ Å which they used.

A real lifetime determination requires that $\chi$ and $\chi_1'$ in Eq. (3) be determined under as nearly as possible the same conditions. In the present work, this was done by choosing the proton energy such that both $\tau_1$ and $\tau_2$ are so short that determination of the minimum yield gives $\chi = \chi_1'$. At other proton energies where $\tau_2$ is not negligi-

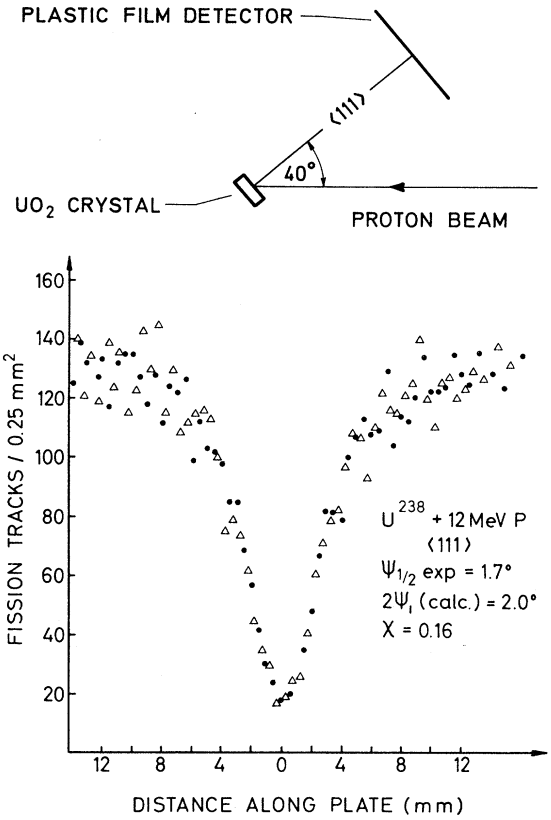


FIG. 1. Fission-track density on a 15-$\mu$m-thick Macrofoil film scanned across a position corresponding to fragment emission parallel to a ⟨111⟩ axis. The triangles and closed circles correspond to scans in two orthonormal directions on the film. A 1-mm-diam proton beam was incident on the uranium dioxide crystal in the geometrical arrangement indicated at the top of the figure. The $\psi_1$ value indicated was calculated from the expression given by Lindhard (Ref. 1) assuming an average fission-fragment proton number of 46.5 and an average energy of 60 MeV.

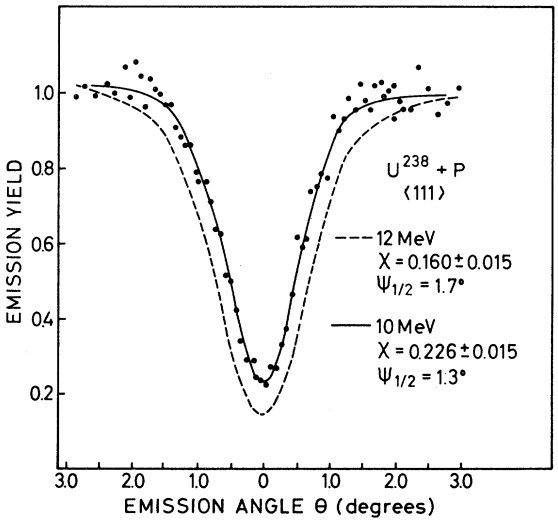


FIG. 2. Measured angular distributions, normalized at large emission angles, of fission fragments emitted parallel to a ⟨111⟩ axis for bombardment of uranium oxide crystals with 10-MeV protons (solid line and closed circles) and with 12- and 9-MeV protons (dashed line). The values indicated for the minimum yield for the 12-MeV curve are the averages, weighted by their respective statistical uncertainty, of three runs at 12 MeV and one at 9 MeV. For the 10-MeV case, the minimum yield is the result of two runs at 10 MeV.

ble, this value for $\chi_1'$ is used to determine $\tau_2$. As noted previously, only very fast fission occurs at proton energies above 12 MeV and below 9 MeV. It is therefore assumed that under such conditions, contributions from the lifetime terms in Eq. (3) are negligible compared to the scattering term $\chi_1'$. At proton energies around 10 MeV, however, the excitation energy of the second-chance fission compound nucleus ($Np^{238}$) is so low that a significant contribution from the last term of Eq. (3) is expected.

A series of experiments were carried out in which 10-MeV bombardments were followed and, in some cases, preceded by 12- or 9-MeV bombardments. Care was taken to ensure that the particle beam was incident on the same spot in each case and that all conditions of beam alignment and crystal orientation were kept constant. Figure 2 shows the results of a scan for a 10-MeV proton bombardment compared with those of a 12-MeV proton bombardment normalized at large emission angles. Three separate 12- and one 9-MeV bombardment were carried out and agreed within statistical accuracy. Two separate 10-MeV bombardments were also in good agreement. The minimum yield values indicated on Fig. 2 are the averages of the various determinations weighted by their respective statistical uncertainties. After correction for scattering effects the minimum yield value gives

$$\left[\frac{(\Gamma_n/\Gamma_f)_1}{1+(\Gamma_n/\Gamma_f)_1+(\Gamma_n/\Gamma_f)_2}\right]\chi(v_\perp\tau_2) = 0.066 \pm 0.021.$$

Estimates[9] of $(\Gamma_n/\Gamma_f)_1 \simeq 1.0$ and $(\Gamma_n/\Gamma_f)_2 = 0.8$ for $Np^{239}$ and $Np^{238}$ compound nuclei at excitation energies of 15.3 and 7.3 MeV, respectively, yield $\chi(v_\perp\tau_2) = 0.183 \pm 0.069$. For $C = 2.5$ and $r_c = 0.4$ Å, this gives a total lifetime $\tau_2 = (1.4 \pm 0.6) \times 10^{-16}$ sec for decay of the $Np^{238}$ compound nuclei and a partial fission lifetime of $(\tau_f)_2 = 2.5 \pm 1.2) \times 10^{-16}$ sec. This result is in good agreement with a partial fission lifetime of $2 \times 10^{-16}$

sec calculated for this case from the expressions given by Huizenga and Vandenbosch[9] using an estimated fission threshold energy of 6.0 MeV. This is surprising since the validity of the calculation is somewhat questionable at low excitation energies.

The mean recoil distance, $v_{\perp}\tau$, from the 10-MeV bombardments, calculated from the measured lifetime, is $0.20 \pm 0.06$ Å. This can be compared with the root mean square thermal vibrational amplitude of 0.07 Å for uranium atoms in $UO_2$.[11] From Andersen's[6] analysis of the dependence on thermal vibrational amplitude of the width of emitted proton angular distributions from tungsten crystals, the width at 0.20-Å mean emission displacement should be 0.5 times the width at 0.07 Å. This is close to the ratio of $0.4 \pm 0.2$ for the change in the width of the angular distribution of fragments from decay of $Np^{238}$ compound nuclei at 10 and 12 MeV after correction of the curves of Fig. 2 for the contribution from $Np^{239}$ using the branching ratio estimates given above. It is difficult to estimate the influence of scattering effects on the width of the angular distribution, but it has been suggested that for axial emission, the width is less sensitive to such effects than is the minimum yield.[6]

Scans of $\langle 110 \rangle$ axial directions in $UO_2$ crystals show even stronger evidence of the importance of scattering effects since they give consistently more narrow and more shallow dips than the $\langle 111 \rangle$ axial scans. This is due to increased scattering by oxygen atomic rows along the $\langle 110 \rangle$ direction[1] and severely limits the usefulness of that direction in lifetime measurements. A more complete discussion of the scattering effects, the detailed analysis, and the associated measurements will be reported elsewhere.[13]

We would like to acknowledge the assistance of P. Ambrosius-Olesen in scanning the plastic detector films and wish to thank J. Lindhard, J. U. Andersen, and J. A. Davies for discussions and guidance in analyzing the observed angular distributions.

Note added in proof.—Similar work has been recently reported by M. Maruyama et al., Phys. Letters 29B, 414 (1969), and K. Komaki and F. Fujimoto, Phys. Letters 29A, 544 (1969), utilizing a somewhat different analytical technique.

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[10]The "effective lifetime" is actually a complicated function of the branching ratios and lifetimes of the members of the decay chain.

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