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Attempted Coulomb excitation of the spontaneous-fission isomeric state in ^{239}Pu†

C. E. Bemis, Jr., F. Plasil, R. L. Ferguson, E. E. Gross, and A. Zucker
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
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Attempts to Coulomb excite the 8.5 μsec spontaneous-fission isomeric state in ^{239}Pu with 100- and 117-MeV ^{20}Ne ions have led to negative results. A cross section upper limit of 3.2×10^{-34} cm^2, integrated over ^{20}Ne laboratory scattering angles $90° \to 180°$, was determined.

[NUCLEAR REACTIONS ^{239}Pu(^{20}Ne, ^{20}Ne′) spontaneous-fission isomer, $E = 100$ and 117 MeV; measured $T_{1/2}$, coinc, Coulomb excitation yield; deduced upper limit $\sigma(E, \theta)$ for $\theta = 90–180°$. Enriched target.]

Fission induced by the time-dependent electromagnetic field of a passing ion is an intriguing possibility that has been investigated theoretically by several authors. Guth and Wilets[1] and Wilets, Guth, and Tenn[1] have used a classical model which is adiabatic and involves no intrinsic target excitation. Beyer *et al.*[2] have used a quantum mechanical approach which considers excitation through the β-vibrational states and have evaluated cross sections for both the Coulomb-induced fission process and the related case of spontaneous-fission isomer excitation. Classical dynamical and quantum mechanical calculations have been reported by Reisenfeldt and Thomas[3] and extended by Holm and Greiner,[4] the latter considering the important influence of nuclear forces on the Coulomb excitation processes.

The above calculations show that Coulomb induced fission is likely only with very heavy projectiles (e.g., Xe at ~5 MeV/nucleon), and that the cross sections are quite substantial ($d\sigma/d\Omega \sim 1$ mb/sr) when scattering is in the backward direction. The various cross-section estimates differ, however, by orders of magnitude.

The excitation and subsequent observation of spontaneous-fission isomeric states is inherently easier to perform experimentally than the pure Coulomb-induced fission process, because the fission events are delayed relative to the time of excitation by the lifetime of the isomeric state. Observation of delayed fission events in pulsed beam experiments can serve as a very sensitive indicator of the population of fission isomeric states.

We have chosen to investigate the possible Coulomb excitation of spontaneous-fission isomeric states using ^{20}Ne ions accelerated at the Oak Ridge isochronous cyclotron (ORIC), as a prelude to the exciting possibility of observing the direct Coulomb-fission process. The direct process may be observable using newer heavy ion facilities which are capable of accelerating Xe and still heavier ions. The Coulomb excitation of spontaneous-fission isomeric states in the second minimum of the nuclear potential is presumed to be similar to the processes most likely involved in direct Coulomb fission, since both require the nucleus to be transformed to much larger equilibrium deformations. An understanding of the Coulomb-excitation processes responsible for the excitation of spontaneous-fission isomeric states should therefore be of value in planning experiments designed to observe the case of pure Coulomb induced fission.

The 8.5 μsec fission isomeric state in ^{239}Pu, which has previously been produced in the ^{239}Pu(d, pn) and ^{238}Pu(d, p) reactions,[5] the ^{236}U(α, n) and ^{238}U($\alpha, 3n$) reactions,[6] the ^{239}Pu(n', n') and ^{240}Pu($n, 2n$) reactions,[7] the ^{239}Pu(γ, γ') reaction,[8] and the ^{240}Pu(γ, n) reaction,[9] was chosen for our investigations because the isomeric state is sufficiently well characterized and the target material ^{239}Pu could be prepared in sufficiently high isotopic quality to preclude possible interferences from the ground-state spontaneous fission activity of even-even isotopic impurities. From analyses of some of the fission-isomer excitation functions,[10] the excitation energy of the isomeric state was determined to be 2.20 ± 0.20 MeV.

A pulsed-beam system, based on the use of 1-m long parallel deflection plates with a 2-cm spacing, was developed for these experiments. The plates were located in a beam line just following the beam extraction point from the cyclotron and it was found that the application of 6 kV to one of the plates was sufficient to deflect a well-focused 100–120 MeV ^{20}Ne^{+5} beam ~1.5 cm in the horizontal plane at a distance of ~10 m from the plates in the beam transport system. At this point, another plate was used to intercept the deflected beam while the undeflected beam was transmitted to the target station. The separation between the deflected and undeflected beams was limited to only ~1.5 cm because of the ion optical influence of a quadrupole doublet and a 45° bending magnet located between the deflection plates and the interception point. A pulse generator, using an Eimac 4CW-25000A radial beam-power tetrode in the switch-tube mode, produced square-wave high-voltage pulses which were applied to one of the deflection plates. Tests of the system using 20-μsec wide pulses indicated that, after corrections due to the time of flight of the ions from the deflection plates to the target assembly, the decay time of the beam at the target station was ~80 nsec and that the beam was completely removed ~200 nsec after the generation of the deflection pulse.

The 120 $\mu g\,cm^{-2}$ isotopically pure ^{239}Pu target on a 1.1 $mg\,cm^{-2}$ Ni foil backing was prepared in an isotope separator. A recoil catcher foil, also 1.1 $mg\,cm^{-2}$ Ni foil, was placed ~3 cm downstream at an angle of 45° to the beam and was viewed by two large area Si(Au) surface-barrier detectors located at 90° to the beam on either side of the catcher foil. Each detector subtended a solid angle of ~20% of 4π sr. The recoiling ^{239}Pu atoms resulting from elastic and inelastic scattering had to pass through the Ni target backing before stopping in the Ni catcher foil. This arrangement only allowed those ^{239}Pu recoils which resulted from elastic and inelastic scattering of 100-MeV ^{20}Ne ions at laboratory angles greater than ~90° to reach the collector foil. Back-scattered ions were expected to yield the largest spontaneous-fission isomer excitation probabilities. The residual range of the ^{239}Pu recoils in the Ni catcher foil resulted in implantation depths up to ~0.65 mg cm^{-2} in the 1.1 $mg\,cm^{-2}$ Ni catcher. The range of typical fission fragments in Ni is ~5 $mg\,cm^{-2}$ and fission fragments emerging from either side of the collector could, therefore, be recorded in the detectors with $\gtrsim$70% of their initial kinetic energy. The two detectors were operated in a fast coincidence mode and both detector pulses were processed together with the output of a time-to-amplitude converter which indicated the time of the fission event relative to the end of a 20-μsec beam burst. The three parameter correlated data were stored in a buffer memory and recorded on magnetic tape.

Two experiments were conducted using 100 and 117 MeV $^{20}Ne^{+5}$ ions for a combined total of ~10^4 particle μC. Only one delayed fission event was observed in these experiments which corresponded to a production cross section of ~3.2×10^{-34} cm^2 integrated over ^{20}Ne laboratory scattering angles in the 90–180° range dictated by our experimental arrangement.

Several possible Coulomb excitation paths leading to the fission isomeric state have been considered by us in the hope of understanding the very low limit on the excitation cross section. Using the Winther–de Boer semiclassical $E2$ coupled-channels computer program[11] for multiple Coulomb excitation, which has been expanded to include $E1$, $E3$, and $E4$ excitations, we have evaluated the cross sections expected (a) for direct $E2$ excitation of the isomer from the ^{239}Pu ground state band, (b) for multiple $E2$ Coulomb excitation through states at or near ~5.5 MeV,[10] which is the height of the first potential energy barrier, and (c) for virtual $E1$ Coulomb excitation through the giant dipole resonance. For the case of direct $E2$ excitation, we have used an upper limit for $B(E2)\uparrow$ which corresponds to the 8.5 μsec lifetime of the isomeric state. The $B(E2)\uparrow$ value is very small, ~10^{-8} single particle units, and corresponds to an excitation cross section of ~1.5×10^{-36} cm^2, well below our upper limit. Similarly, cross sections of $\leq10^{-36}$ cm^2 are expected if single particle $E2$ transitions by a variety of excitation paths are allowed to populate the isomer through states at or near the height of the first barrier.

Multiple $E1$ excitation through the giant dipole resonance (GDR) structure at ~12–13 MeV however can lead to quite large cross sections comparable to our upper limit of ~3.2×10^{-34} cm^2. Using an energy-weighted sum-rule limit to estimate the $E1$ strength from the ground state to the GDR, we arrive at $B(E1)\uparrow\sim6.5\times10^{-25}\ e^2\,cm^2$ for the $T_<$ resonance. If we allow the transition from the ground-state GDR to the isomeric state to have the same strength, we estimate a cross section of ~10^{-32} cm^2. Our cross section limit would imply that $B(E1;\ \text{GDR}\rightarrow\text{Isomer})/B(E1;\text{Ground}\rightarrow\text{GDR})\lesssim1/100$.

We have recently learned of similar Coulomb excitation experiments by Gangrsky *et al.*[12] using 60 MeV ^{12}C ions and 740 MeV ^{136}Xe ions in attempts[12] to excite the 200 nsec fission isomer in ^{238}U. Cross section upper limits of 10^{-33} cm^2 and 10^{-31} cm^2, respectively, were determined. A cross section upper limit of 3×10^{-28} cm^2 sec^{-1} has also recently been determined by Ngô, Péter, and Tamain[13] for the direct Coulomb fission of ^{238}U using ^{84}Kr ions. Although our limits for ^{20}Ne ions and the fission isomer of ^{239}Pu are somewhat lower than the above, we conclude that further attempts to Coulomb-excite fission isomers will have to be postponed until the use of much heavier ions is a reality.

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