**Title**
Direct Proof of Electron-Capture-Delayed-Fission Process

**Journal**
Physical review letters, 63(23)

**Authors**
Hall, H.L.
Gregorich, K.E.
Henderson, R.A.
et al.

**Publication Date**
2017-12-04

LBL-27425 C1
Preprint

**Lawrence Berkeley Laboratory**
UNIVERSITY OF CALIFORNIA

Submitted to Physical Review Letters

# First Direct Proof of EC-Delayed Fission

H.L. Hall, K.E. Gregorich, R.A. Henderson, C.M. Gannett,
R.B. Chadwick, J.D. Leyba, K.R. Czerwinski, B. Kadkhodayan,
S.A. Kreek, D.M. Lee, M.J. Nurmia, and D.C. Hoffman

June 1989

NUCLEAR SCIENCE DIVISION

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

LBL-27425

# First Direct Proof of EC-Delayed Fission

H. L. Hall, K. E. Gregorich, R. A. Henderson, C. M. Gannett,
R. B. Chadwick, J. D. Leyba, K. R. Czerwinski, B. Kadkhodayan,
S. A. Kreek, D. M. Lee, M. J. Nurmia, and D. C. Hoffman

*Nuclear Science Division*
*Lawrence Berkeley Laboratory*
*University of California*
*1 Cyclotron Road*
*Berkeley, CA 94720*

## Abstract

This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

# First Direct Proof of EC-Delayed Fission

H. L. Hall, K. E. Gregorich, R. A. Henderson, C. M. Gannett,
R. B. Chadwick, J. D. Leyba, K. R. Czerwinski, B. Kadkhodayan,
S. A. Kreek, D. M. Lee, M. J. Nurmia, and D. C. Hoffman

*Nuclear Science Division*
*Lawrence Berkeley Laboratory*
*University of California*
*1 Cyclotron Road*
*Berkeley, CA 94720*

## Abstract

We have studied the electron-capture-delayed fission ($\varepsilon$DF) of $^{234}$Am. The half-life of $^{234}$Am was determined to be $2.32 \pm 0.08$ minutes and the measured ratio of delayed fissions to plutonium K x-rays was $(2.2 \pm 0.6) \times 10^{-4}$. The observed fissions were unequivocally assigned to an EC-delayed fission process in americium by coincidence measurements between the K-capture x-rays and the subsequent fission.

This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

Delayed fission is a nuclear decay process in which a decaying nucleus populates excited states in its daughter nucleus, which then fission. These states can be above the fission barrier(s) of the daughter (yielding prompt fission), within the second well of the potential energy surface (a fission shape isomer), or within the first well of the potential energy surface (an electromagnetic isomer), as illustrated in Figure 1. This decay mode is believed to influence the production of heavy elements from multiple neutron capture processes [1,2,3,4] followed by $\beta$ decay, such as the astrophysical r-process and nuclear weapons tests [5,6]. Delayed-fission processes may also provide a sensitive probe of fission barriers in the heavy element region [7].

Experimental observations of fission tracks from EC-delayed fission ($\varepsilon$DF) were first reported [8,9] in the light americium and neptunium isotopes as early as 1966. In 1969, Berlovich and Novikov [10] noted that the nuclei in question met the conditions required for delayed fission, although the observed fissions were not specifically attributed [11] to delayed-fission processes until 1972. Habs *et al.* [12] observed fissions which they attributed to $\varepsilon$DF in $^{232}$Am in 1978. Gangrskiĭ *et al.* [13] have reported delayed fission in several neutron-deficient transcurium nuclei, and an $\varepsilon$DF branch has been tentatively assigned [14] to $^{242}$Es. Recently, $\varepsilon$DF has been reported [15] in the region of $^{180}$Hg.

However, there have been no measurements of the time-correlation between the EC decay and the subsequent fission of the daughter. All reports of $\varepsilon$DF have assigned the observed fissions to this decay mode by inference, using fission half-life systematics to rule out spontaneous (ground-state) fis-

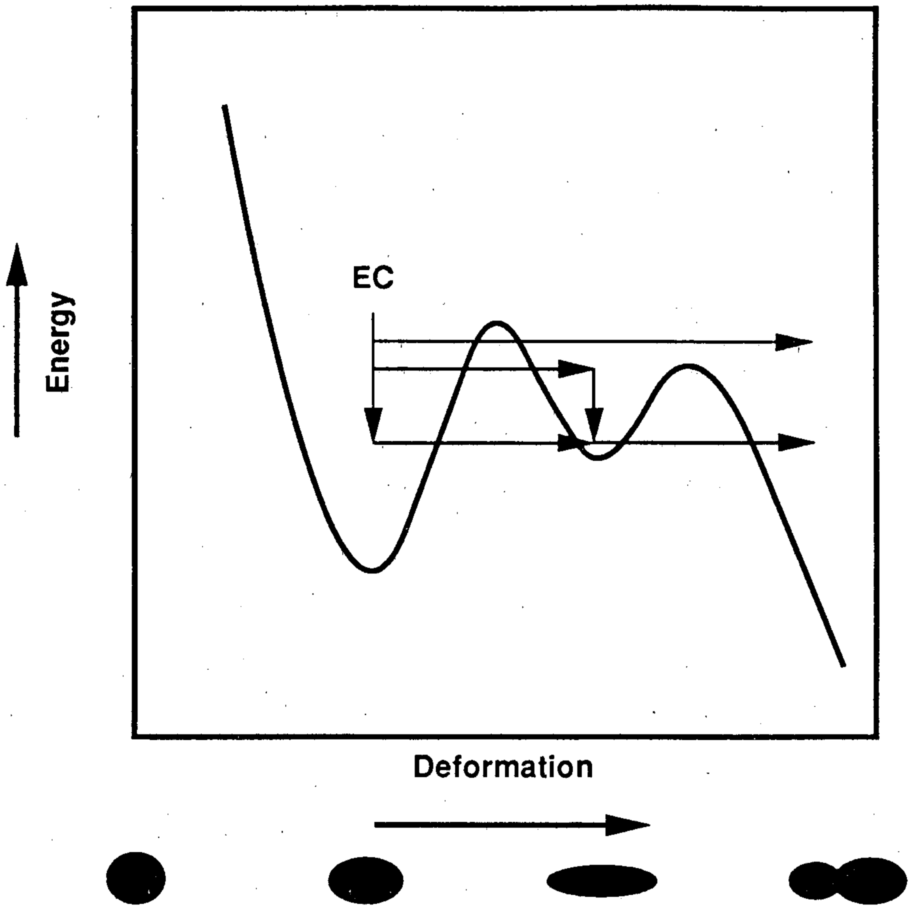

Figure 1: Schematic illustration of the delayed-fission process.

sion of the parent. We sought to study a presumed delayed-fissile nucleus in sufficient detail to measure its fission properties and to verify directly the EC-delayed-fission hypothesis.

$^{234}$Am ($t_{1/2} = 2.6$ min [11,16]) was chosen for this study, and was produced by irradiating multiple $^{237}$Np targets [17] with 70-73 MeV $\alpha$ particles from the LBL 88-Inch Cyclotron. The recoiling reaction products were swept away from the targets with a KCl/He-jet, and transported to various collection sites for subsequent measurements. The use of multiple targets allowed the production of a large number of fission events, permitting us to measure the fission properties of $^{234}$Am and to measure the time correlation between the plutonium x-rays from K-capture in $^{234}$Am and the subsequent fission of the $^{234}$Pu.

From on-line data taken with our rotating-wheel system and from radiochemical data, we measured the fission properties of this decay mode. The half-life of $^{234}$Am was found to be $2.32 \pm 0.08$ minutes. The delayed-fission probabilty was determined to be $(2.2 \pm 0.6) \times 10^{-4}$ from the measured ratio of fissions to plutonium K x-rays, assuming the contribution to the K x-ray intensity from internal conversion is negligible and the K/L capture ratio in $^{234}$Am is very large. Based on 1188 observed coincident fision fragments, the mass division of the $^{234}$Am $\varepsilon$DF mode was found to be highly asymmetric, with a total kinetic energy of $175 \pm 5$ MeV. These measurements will be discussed in more detail in a complete paper [18] to follow this Letter.

The time correlation between EC and fission was measured using aerosols collected directly from the KCl/He-jet transport system without any chemical separation. The aerosols were collected on a tantalum foil and, after a collection interval of four minutes, the foil was placed before a light-tight transmission-mounted 300-mm$^2$ silicon surface barrier (SSB) detector operated in air. The SSB detector and foil were sandwiched between two germanium $\gamma$ detectors.

Since low-energy fission is typically accompanied by about 10 prompt $\gamma$ rays [19,20], a high overall $\gamma$ detection efficiency would cause many of the true K x-ray events to sum with these prompt $\gamma$ rays and be recorded at a too high an energy. This diminishes the efficiency for the K x-ray photopeaks. On the other hand, too low an overall $\gamma$ efficiency would also reduce the efficiency for detecting the x-ray, and hence the x-ray–fission correlations. By measuring the prompt $\gamma$ rays from spontaneous fission of a source of $^{252}$Cf, the distance between the $\gamma$ detectors and the sample was adjusted to bring the summing rejection level to 50% in each detector. As long as the $\gamma$ multiplicity of the $^{234}$Am $\varepsilon$DF is not grossly different than that of $^{252}$Cf, this would maximize the number of detected x-ray–fission correlations. In the final configuration, each detector subtended a solid angle of about 6.7% of $4\pi$, giving a 50% summing rejection level. This resulted in an overall correlation detection efficiency (using both detectors) of 6.7% for each detected fission.

The signal from the SSB detector provided a common start for two electronic time-to-amplitude convertors (TACs). The stop signals for the first and second TACs were provided by the first and second $\gamma$ detectors, respectively. The time window on the TACs was ±500 ns. Calibrations were obtained using the prompt $\gamma$ rays from the fission of $^{252}$Cf and the $\gamma$ rays in

Table 1: Observed and expected x-ray intensities from the correlated x-ray–fission data. Expected x-ray intensities are taken from the **Table of Isotopes**. [21].

| X-ray | E/keV | $I_{\text{theo}}$ | No. Observed[a] | $I_{\text{obs}}$ |
|---|---|---|---|---|
| Pu $K_{\alpha 2}$ | 99.55 | 0.299 | 4 | 0.15 ± 0.08 |
| Pu $K_{\alpha 1}$ | 103.76 | 0.479 | 14 | 0.54 ± 0.18 |
| Pu $K_{\beta 1'}$ | 116.9 | 0.162 | 7 | 0.27 ± 0.11 |
| Pu $K_{\beta 2'}$ | 120.6 | 0.060 | 1 | 0.04 ± 0.04 |

[a] Approximately 6 ± 3 of these events are due to the continuum of prompt $\gamma$ rays from fission.

---

coincidence with the $\alpha$ particles from the decay of $^{249}$Cf. The timing resolution of the TACs was ~25 ns full-width at half-maximum (FWHM), and the energy resolution of the $\gamma$ detectors was ~1.5 keV FWHM for the Pu K x-ray region. Upon detection of a fission event in the SSB detector, the amplitudes of the pulses (if any) in all detectors and the TACs were measured and recorded, along with the time at which the event occurred. The start and stop times of the measurement of each sample were also recorded.

Approximately 1000 samples were collected and measured in this manner in a forty hour irradiation. Figure 2 shows the x-ray and $\gamma$ spectrum of those events in prompt coincidence with the fission signal. Observed and expected x-ray intensities for plutonium are given in Table 1.

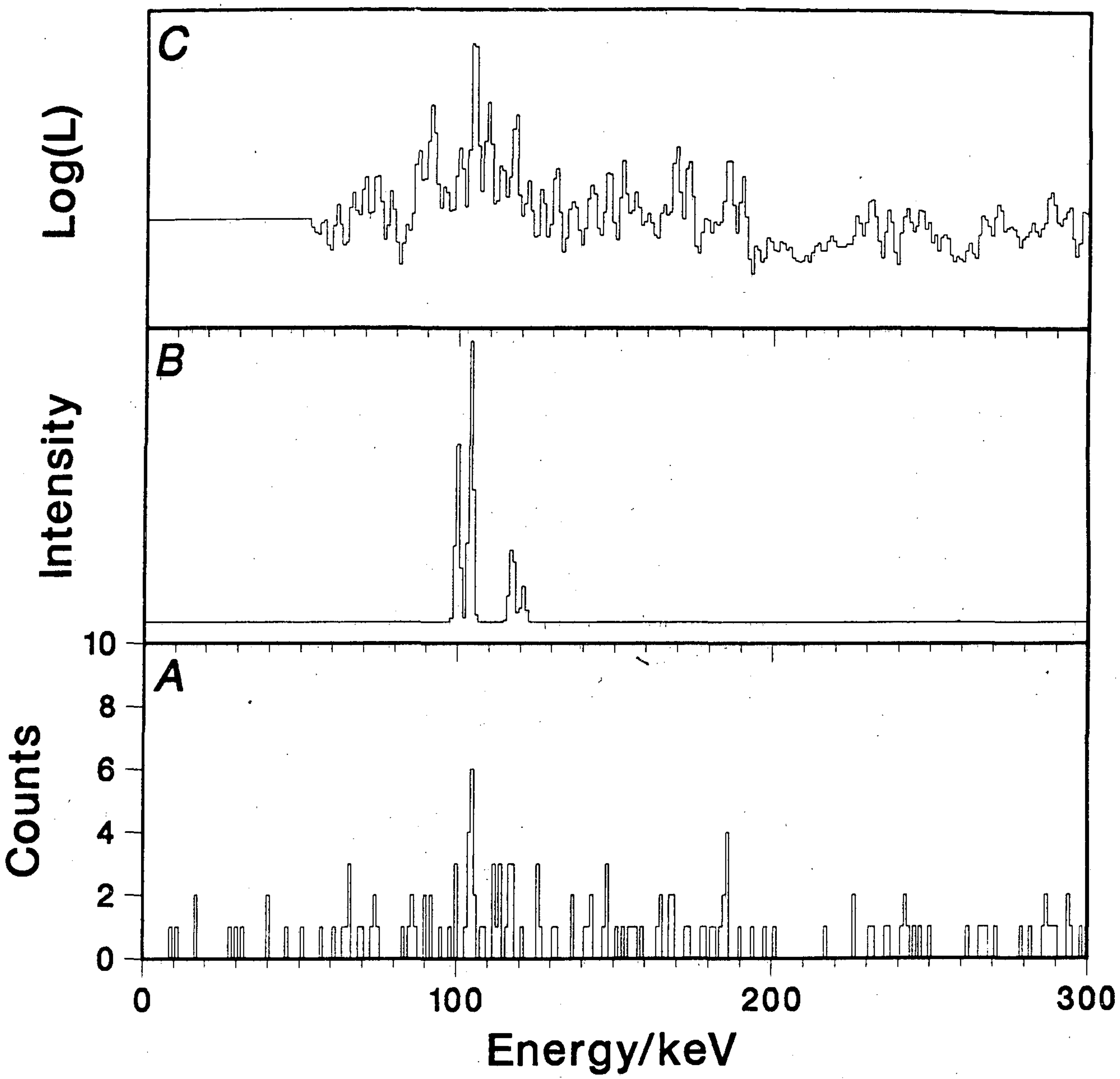

Figure 2: X-ray–fission correlation results. *A*: X-rays and $\gamma$ rays in coincidence with delayed fission from $^{234}$Am. *B*: An idealized plutonium K x-ray spectrum, based on the measured detector resolution and an expected prompt $\gamma$-ray continuum. *C*: The liklihood function for the position of the ideal spectrum (*B*) in the data (*A*). In *C*, the liklihood is plotted as a function of the $K_{\alpha 1}$ position of the ideal spectrum. From the liklihood functions, the $K_{\alpha 1}$ energy was found to be 103.6 ± 0.5 keV and the total number of K x-rays was found to be 19 ± 5.

No evidence was observed for fission delay times longer than the timing resolution of the experiment, about 25 ns. The fact that plutonium x-rays can be seen requires that the lifetime of the fissioning state be longer than the time it takes the orbital electrons to cascade down and fill a K-vacancy. The time required for this is on the order of $10^{-17}$ seconds [22]. We can therefore set limits on the half-life of the fissioning state(s) of $10^{-8}$ ns $< t_{\frac{1}{2}} <$ 25 ns. If the nucleus is heavily damped in the second well (*i.e.* tunnelling proceeds through the inner fission barrier and the nucleus then $\gamma$ decays to the lowest state in the second well before fissioning), these limits are also limits on the lifetime of the shape isomer $^{234f}$Pu. This is consistent with other plutonium fission isomers, which have half-lives on the order of 20-40 ns [21].

The fission-$\gamma$ coincidence data in Figure 2 also show what appear to be photopeaks at about 112, 126, 147, 168, and 185 keV. Although the counting statistics for these photopeaks are poor, prompt $\gamma$ rays from fission fragments do not display such structure [19]. It is possible that these $\gamma$ rays are a result of $\gamma$ deexcitation of levels of $^{234}$Pu in the second well. If this is the case, the correlation of these $\gamma$ rays supports the hypothesis that nuclear motion in the second well is strongly damped. Unfortunately, the lack of $\gamma$-$\gamma$-fission coincidence data precludes constructing a level scheme for $^{234f}$Pu. However, x-ray—$\gamma$—fission coincidence from delayed fission should provide a unique probe for studying the level structure of the shape isomer. Such a measurement might best be done with a high efficiency multiple $\gamma$-detector array such as the proposed **GAMMASPHERE** [23].

Our measurement of x-ray—fission correlations in the decay of $^{234}$Am unequivocally proves that this decay mode is indeed EC-delayed fission. This is the first $\varepsilon$DF process for which direct proof has been obtained. The only other time-correlated measurement of a delayed-fission process is for $\beta$-delayed fission of $^{256m}$Es [24].

## Acknowledgements

This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

The authors wish to thank the staff and crew of the LBL 88-Inch Cyclotron for their assistance.

## References

[1] E. M. Burbidge, G. R. Burbidge, W. A. Fowler, and F. Hoyle, *Rev. Mod. Phys. 29*, 547 (1957).

[2] C.-O. Wene and S. A. E. Johansson, *Phys. Scripta 10A*, 156 (1974).

[3] C.-O. Wene, *Astron. & Astrophys. 44*, 233 (1975).

[4] H. V. Klapdor, T. Oda, J. Metzinger, W. Hillebrandt, and F. K. Thielman, *Z. Physik A 299*, 213 (1981).

[5] R. W. Hoff, *in* **Weak and Electromagnetic Interactions in Nuclei,** H. V. Klapdor, ed., Springer-Verlag (Heidelburg, 1986), 207.

[6] R. W. Hoff, *Inst. Phys. Conf. Ser. No. 88/J. Phys. G:Nucl. Phys. 14 Suppl.*, S343 (1986).

[7] Yu. A. Lazarev, Yu. Ts. Oganessian, and V. I. Kuznetsov, *Joint Institutes for Nuclear Research, Dubna, Report No. JINR–E7–80–719* (1980).

[8] V. I. Kuznetsov, N. K. Skobelev, and G. N. Flerov, *Yad. Fiz. 4*, 279 (1966) [*Sov. J. Nucl. Phys. 4*, 202 (1967)].

[9] V. I. Kuznetsov, N. K. Skobelev, and G. N. Flerov, *Yad. Fiz. 5*, 271 (1967) [*Sov. J. Nucl. Phys. 5*, 191 (1967)].

[10] É. E. Berlovich and Yu. P. Novikov, *Dok. Akad. Nauk SSSR 185*, 1025 (1969) [*Sov. Physics—Doklady 14*, 349 (1969)].

[11] N. K. Skobelev, *Yad. Fiz. 15*, 444 (1972) [*Sov. J. Nucl. Phys. 15*, 249 (1972)].

[12] D. Habs, H. Klewe–Nebenius, V. Metag, B. Neumann, and H. J. Specht, *Z. Physik A 285*, 53 (1978).

[13] Yu. P. Gangrskiĭ, M. B. Miller, L. V. Mikhaĭlov, and I. F. Kharisov, *Yad. Fiz. 31*, 306 (1980) [*Sov. J. Nucl. Phys. 31*, 162 (1980)].

[14] R. Hingman, W. Kuehn, V. Metag, R. Novotny, A. Ruckelshausen, H. Stroeher, F. Hessberger, S. Hofmann, G. Muenzenberger, and W.

Reisdorf, *Gessellschaft für Schwerionenforschung, Darmstadt, Report No. GSI 85-1*, 88 (1985).

[15] Yu. A. Lazarev, Yu. Ts. Oganessian, I. V. Shirokovsky, S. P. Tretyakova, V. K. Utyonkov, and G. V. Buklanov, *Europhys. Lett. 4*, 893 (1987).

[16] L. P. Somerville, A. Ghiorso, M. J. Nurmia, and G. T. Seaborg, *Lawrence Berkeley Laboratory Nuclear Science Division Annual Report, 1976-1977, Report No. LBL-6575*, 39 (1977).

[17] H. L. Hall, M. J. Nurmia, D. C. Hoffman, *Nucl. Inst. Meth. A276*, 649 (1989).

[18] H. L. Hall, K. E. Gregorich, R. A. Henderson, C. M. Gannett, R. B. Chadwick, J. D. Leyba, K. R. Czerwinski, B. Kadkhodayan, S. A. Kreek, D. M. Lee, M. J. Nurmia, D. C. Hoffman, C. E. A. Palmer, and P. A. Baisden, to be submitted to *Phys. Rev. C* (1989).

[19] D. C. Hoffman and M. M. Hoffman, *Ann. Rev. Nucl. Sci. 24*, 151 (1974).

[20] D. C. Hoffman and L. P. Somerville, *in* **Charged Particle Emission from Nuclei Vol. III**, D. N. Poenaru and M. Ivascu, eds., (CRC Press, Boca Raton, Fla., 1988) 1.

[21] C. M. Lederer, V. M. Shirley, E. Browne, J. M. Dairiki, R. E. Doebler, A. A. Shihab-Eldin, L. J. Jardine, J. K. Tuli, and A. B. Buyrn, **Table of Isotopes, 7th Ed.** (John Wiley & Sons, New York, 1978).

[22] J. H. Scofield, *At. Data Nucl. Data Tables 14*, 121 (1974)

[23] "GAMMASPHERE - A Proposal for a National Gamma Ray Facility," M.-A. Delaplanque and R. M. Diamond, eds., *Lawrence Berkeley Laboratory Publication No. PUB-5202* (1988).

[24] H. L. Hall, K. E. Gregorich, R. A. Henderson, D. M. Lee, D. C. Hoffman, M. E. Bunker, M. M. Fowler, P. Lysaght, J. W. Starner, and J. B. Wilhelmy, *Phys. Rev. C 39*, 1866 (1989).

LAWRENCE BERKELEY LABORATORY
TECHNICAL INFORMATION DEPARTMENT
1 CYCLOTRON ROAD
BERKELEY, CALIFORNIA 94720