Title
AN INVESTIGATION OF THEINFLUENCE OF ANGULAR MOMENTUM ON FISSION PROBABILITY

Authors
Gilmore, John
Thompson, Stanley G.
Perlman, I.

Publication Date
1962-06-08

UCRL-9304 Rev.

University of California

Ernest O. Lawrence Radiation Laboratory

Berkeley, California

For Journal-Phys. Rev.

UCRL-9304 Rev.

UNIVERSITY OF CALIFORNIA
Lawrence Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

AN INVESTIGATION OF THE INFLUENCE OF ANGULAR MOMENTUM ON FISSION PROBABILITY

John Gilmore, Stanley G. Thompson, and I. Perlman

June 8, 1962

AN INVESTIGATION OF THE INFLUENCE OF ANGULAR MOMENTUM ON FISSION PROBABILITY

John Gilmore, Stanley G. Thompson, and I. Perlman

Lawrence Radiation Laboratory
Berkeley, California

June 8, 1962

ABSTRACT

A nuclear emulsion technique has been used to determine total fission cross sections in the following heavy-ion bombardments: (C^{12} + Tm^{169}; O^{16} + Ho^{165}), (O^{16} + Tm^{169}; Ne^{20} + Ho^{165}), (C^{12} + Re^{185}; O^{16} + Ta^{181}), (O^{16} + Re^{185}; Ne^{20} + Ta^{181}). Each pair of bombardments resulted in the same compound nucleus, and excitation energies could be made equal in the two cases by adjustment of bombarding energies.

The ratio of the fission cross section to a calculated compound-nucleus-formation cross section, σ_f/σ_c, was taken as a measure of fission probability in each bombardment. Larger fission probabilities were observed to occur for the systems having greater angular momenta.

AN INVESTIGATION OF THE INFLUENCE OF ANGULAR MOMENTUM ON FISSION PROBABILITY

John Gilmore, Stanley G. Thompson, and I. Perlman

Lawrence Radiation Laboratory
Berkeley, California

June 8, 1962

Studies of heavy-ion-induced nuclear reactions have included a number of investigations of the fission process. One conclusion from these investigations is that fission represents a significant fraction of the total reaction cross section, even for relatively light nuclei. In the bombardment of rhenium with N^{14} ions, Druin, Polikanov, and Flerov report that fission accounts for about 30% of the reaction cross section at a bombarding energy of 100 Mev.[1] This proportion increases to more than 50% when Au^{197} and Bi^{209} are bombarded with heavy ions.

One factor contributing to such high probabilities for fission is that the compound nuclei are neutron-deficient, with relatively large values of the fissionability parameter Z^2/A. High neutron binding energies in these compound nuclei also favor the competition of fission over neutron emission.

Compound nuclei formed in heavy-ion bombardment are further characterized by formation with as much as 100 $\hbar$ of angular momentum. The possibility that angular momentum may affect fissionability has been discussed by G. M. Pik-Pichak.[2] Using the liquid drop model to evaluate the saddle-point deformation and rotational energies, Pik-Pichak showed that the barrier against fission decreases with increasing angular momentum. Calculations of this type have also been performed by Hiskes.[3] Another approach to the evaluation of an angular momentum effect has been taken by Halpern[4] and by Huizenga and Vandenbosch.[5] These authors point out that if an appreciable part of the excitation energy is taken up in rotational motion, the level width for neutron emission becomes small relative to that for fission.

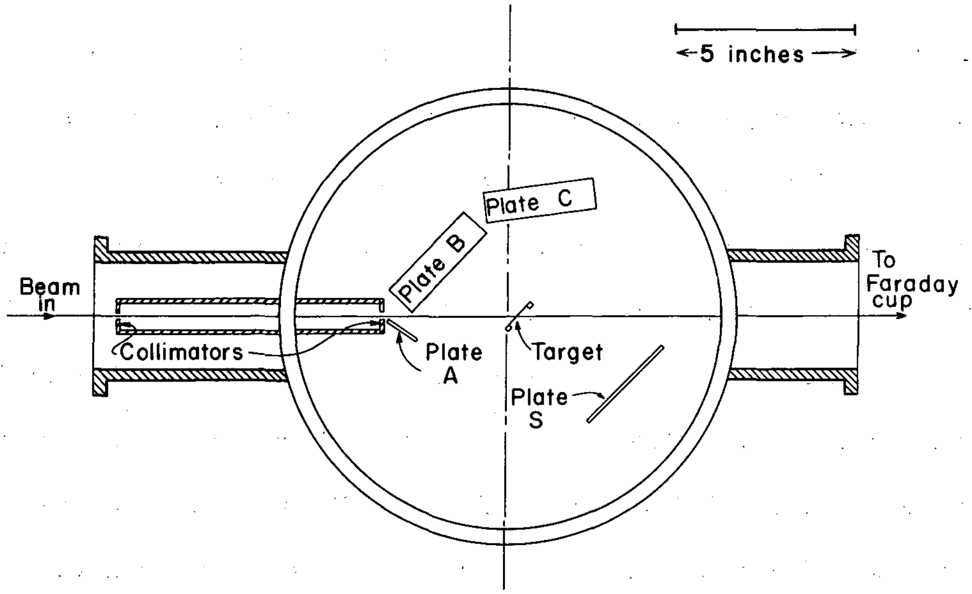
The object of this work was to determine the effect of angular momentum on the probability for fission in heavy-ion bombardment. Pairs of isotopes were bombarded with different heavy ions to give the same compound nucleus. These target isotopes were in the region of Z from 67 to 75, where heavy-ion fission probabilities increase rapidly with bombarding energy. For a given excitation energy of the compound nucleus the angular momentum brought in by the heavier ion was in general greater because of its larger mass and radius. Angular momenta and fission probabilities in the two bombardments were then correlated to reveal the direction and magnitude of any angular momentum effect. The quantity taken as a measure of fission probability was the ratio of an experimentally determined fission cross section to a calculated compound-nucleus formation cross section. For adequate sensitivity, a region of the periodic system was selected in which the fission cross sections would be well below the geometric cross section and yet not vanishingly small.

EXPERIMENTAL PROCEDURE

Cross sections for fission were determined by integration of fission-fragment angular distributions. The fission chamber (Fig. 1) contained nuclear emulsions which intercepted fission fragments recoiling from the target. The chamber had been designed by Goldberg and Reynolds for experiments in heavy-ion elastic scattering.[6] In our work, emulsion holders were rearranged so that emulsions recorded fission fragments leaving the target at angles of from 60 to 178 deg with respect to the beam direction. To improve recognition of fission fragment tracks, emulsions were mounted so that fragments entered the plate at angles of less than 30 deg to the surface.

The heavy-ion beam was momentum-analyzed and collimated by two 1/8-in. collimators fixed 7.5 in. apart. Aluminum foils were used to degrade the initial ion energy of 10.4 MeV/nucleon. Required foil thicknesses were determined from Hubbard's range-energy tabulations.[7] At the conclusion of each bombardment, the beam intensity was reduced and an emulsion placed between the second collimator and target to intercept a few hundred heavy ions. From a determination of the range spectrum and reference to the range-energy relationships for heavy ions by Heckman et al.,[8] the energy and homogeneity of the degraded beam were established. The width of the energy distribution at half-maximum was found typically to be 2%.

Emulsions were scanned under 1000 × magnification at 5-mm intervals along the plate center line. At each point sufficient area was scanned to detect at least 300 fission-fragment tracks.



MU-19346

Fig. 1. Fission chamber.

Correction curves giving the intercepted solid angle and angle of recoil for each area scanned were determined from measurements of the chamber. The error in recoil angle arising from error in measurements was estimated as ±0.5 deg. Finite collimator size and scattering in the target were calculated to result in a half-width in angular resolution of approx. 3 deg. The correction curves were checked by applying them to observations made with an α-particle source of known intensity placed at the target position. Within limits of expected standard deviation, the angular distribution was isotropic and the calculated and measured rates of α-particle emission from the source agreed to within 1%.

After passing through the target, the beam entered a Faraday cup where the charge was collected for integration by a 100%-feedback electrometer. A quadrupole magnet was placed around the mouth of the Faraday cup to prevent escape of electrons from the cup. Values of integrated beam current given by the electrometer and inferred from a determination of Rutherford scattering cross section agreed to within 4%.

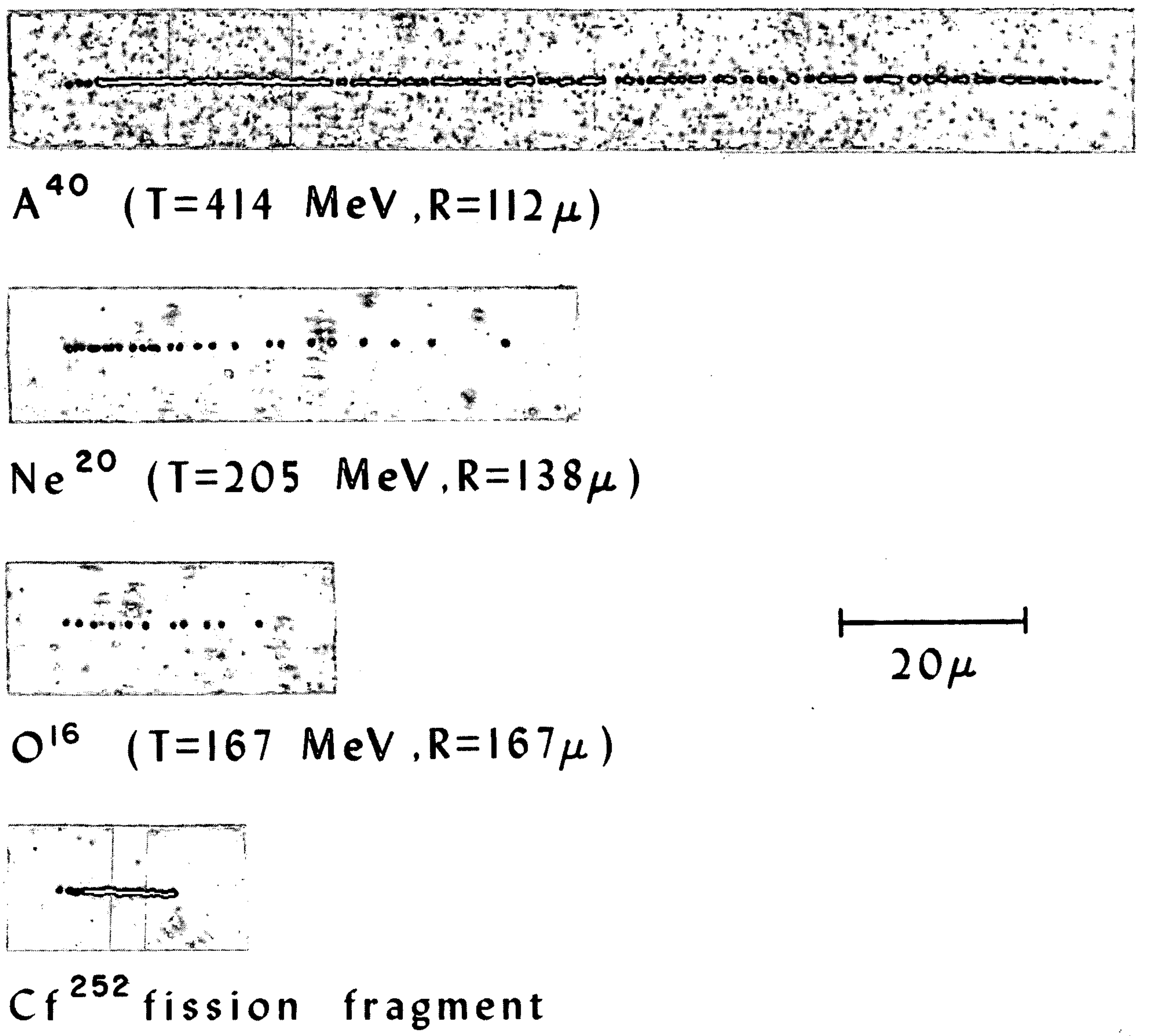
Targets of Ho^{165}, Tm^{169}, Ta^{181}, Re^{185}, and Re^{187} were prepared in approx 0.6 mg/cm^2 thickness on 1.2 mg/cm^2 nickel backings. Targets of the separated isotopes Re^{185} (96.0% Re^{185}, 4.0% Re^{187}) and Re^{187} (98.6% Re^{187}, 1.2% Re^{185}) were electroplated, following the procedure of Levi and Esperson.[9]

Emulsions used in the chamber were 50μ thick, coated on 1 × 3-in. glass slides. Preliminary experiments with the fission chamber indicated a need for an emulsion-developer combination which would permit fission-fragment tracks to be discriminated from tracks of short-range heavy ions. Adequate discrimination was finally achieved through use of Ilford K minus 2 emulsions processed with a modification of a developer given by Stevens.[10] Development of 50-μ-thick emulsions was carried out for 40 min at 19 ± 19 C. in a developer of the composition

sodium phosphate (tribasic)	25 g,
sodium hydrogen phosphate (dibasic)	25 g,
sodium sulfite (anhydrous)	40 g,
potassium bromide	4 g,
sodium bisulfite	0.05 g,
p-aminophenol hydrochloride	0.3 g,
water -- to make	1 liter.

A series of K minus 2 emulsions was exposed to several different heavy-ion beams and to a Cf^{252} source of fission fragments (Fig. 2). Ends of the tracks are at the left edge of Fig. 2. For A^{40}, the entire range of the ion is recorded and the maximum in rate of energy loss, dT/dR, is seen as a continuous region a few microns from the end of the track. For the Ne^{20} ion only a fraction of the total range is recorded, and decreased grain spacing near the end of the track again reflects the increasing rate of energy loss. The O^{16} ion is recorded only as a few grains in the region of maximum dT/dR, and in plates exposed to N^{14} and C^{12} ions only random grains were developed. Fission-fragment tracks from Cf^{252}, on the other hand, were continuous except at the very end, where the track often broke down into one or two grains as dT/dR approached zero.

In early stages of this work, scanning was complicated by the presence of a surface blackening of the emulsion which often obscured the fission-fragment tracks lying immediately beneath the surface. When a pair of charged plates located between the target and emulsion caused a displacement of the blackened region, it became apparent that electrons, probably arising as δ rays from passage of the ion beam through the target, where responsible for



ZN-2563

Fig. 2. Heavy-ion tracks in Ilford K minus 2 emulsion.

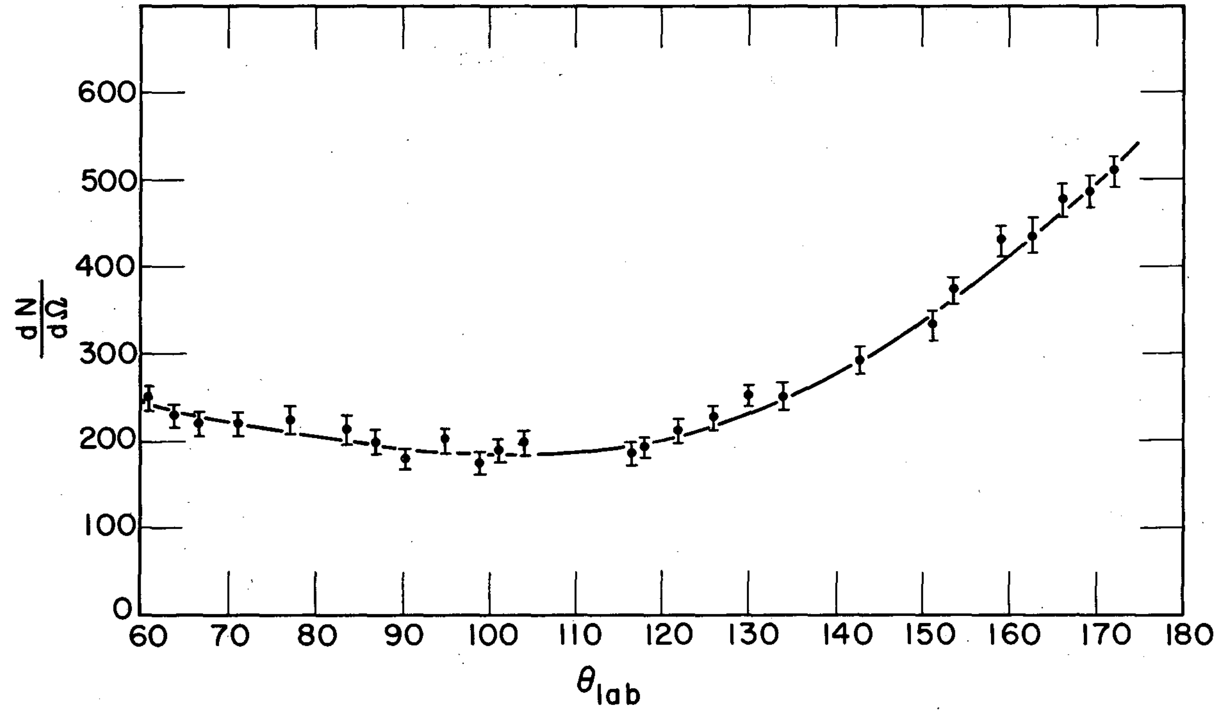
the blackening. A permanent magnet constructed by Goldberg and Reynolds[11] was placed near the target in later bombardments to deflect electrons away from the emulsions.

ANALYSIS OF DATA

An example of the angular distributions from which fission cross sections were derived is shown in Fig. 3. This distribution represents data from plates B and C (Fig. 1) for the bombardment of Re^{185} with 79.2-MeV C^{12} ions. A transformation of the data of Fig. 3 to the center-of-mass (c.m.) system is shown in Fig. 4. The transformation was made with reference to a most probable mass and kinetic energy, using the tables of Marion, Arnette, and Owens.[12] Terrell's[13] correlation of kinetic energy release with $Z^2/A^{1/3}$ of the fissioning nucleus has been found to be valid for heavy-ion fission of Au^{197},[14] and Bi^{209},[15] and was used to determine the most probable kinetic energy. Corresponding to the results of radiochemical studies of fragment mass distributions in fission of Au^{197} by C^{12} and N^{14} ions, [16,17] a division of the fissioning nucleus into equal fragments was assumed to be most probable. Calculations of differential fission cross sections are not, however, particularly sensitive to these choices of most probable fragment mass or kinetic energy. A 20% change in the value of either quantity would be reflected in a change of only 3% in the fission cross section.

Each value of $dN/d\Omega$ (Fig. 4) was multiplied by $d\Omega/d\theta = 2\pi \sin\theta$ and the resulting distribution integrated over θ from $\pi/2$ to π to give the number of fission fragments emitted in the backward hemisphere. This number was taken as one-half the total fragments emitted, following the assumption that the angular distribution is symmetric about 90 deg c.m. Such symmetry is observed in heavy-ion fission angular distributions for which data have been recorded at small angles.[18]

In Tables I, II, and III and Figs. 5, 6 and 7 total fission cross sections are shown as a function of the bombarding energy of the ion. Indicated uncertainties pertain only to counting statistics.



MU-21021

Fig. 3. Angular distribution of fission fragments from the bombardment of Re^{185} with 79.2-MeV C^{12} ions (lab system).

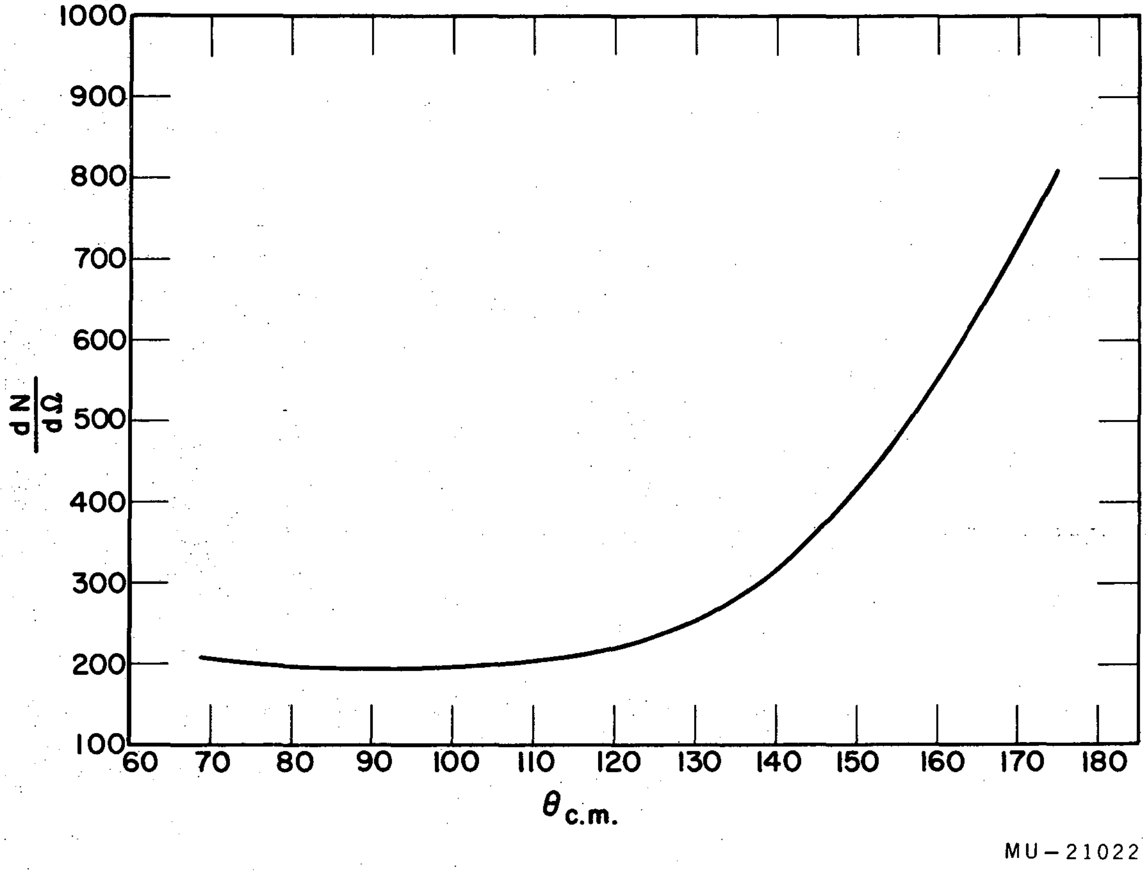


Fig. 4. Angular distribution of fission fragments from the bombardment of Re^{185} with 79.2-MeV C^{12} ions (c.m. system).

Table I. Results of bombardment with C^{12} ions

Target	Energy of bombarding ion (MeV)	Fission cross section and standard deviation (barns)	
Tm^{169}	89	0.074	0.015
	104	0.386	0.052
	124	1.28	0.09
Re^{185}	79	0.089	0.006
	89	0.270	0.020
	106	0.652	0.023
	124	1.03	0.03
Re^{187}	99	0.339	0.013
	124	0.826	0.027

Table II. Fission cross sections and standard deviations for bombardments with O^{16}

Target	Energy of bombarding ion (MeV)	Fission cross section and standard deviation (barns)	
Ho^{165}	94	0.0052	0.0015
	109	0.079	0.005
	132	0.243	0.013
	167	0.440	0.036
Tm^{169}	94	0.019	0.003
	104	0.079	0.012
	116	0.227	0.020
	148	0.573	0.048
	167	0.769	0.038
Ta^{181}	88	0.044	0.007
	89	0.071	0.005
	94	0.173	0.014
	98	0.346	0.026
	109	0.761	0.027
	122	1.18	0.06
	136	1.45	0.05
	151	1.69	0.08
	167	1.89	0.08
Re^{185}	94	0.303	0.018
	104	0.625	0.026
	116	1.05	0.05
	148	1.54	0.05
	167	1.80	0.06
Re^{187}	167	1.74	0.05

Table III. Fission cross sections and standard deviations for bombardments with Ne^{20}

Target	Energy of bombarding ion (MeV)	Fission cross section and standard deviation (barns)	
Ta^{181}	99	0.059	0.009
	116	0.736	0.063
	118	0.780	0.066
	138	1.57	0.09
	164	2.05	0.09
	178	2.19	0.14
	211	2.47	0.10
Ho^{165}	116	0.094	0.013
	118	0.121	0.015
	138	0.351	0.041
	139	0.348	0.037
	168	0.552	0.045
	178	0.626	0.048
	198	0.789	0.077
	204	0.791	0.065

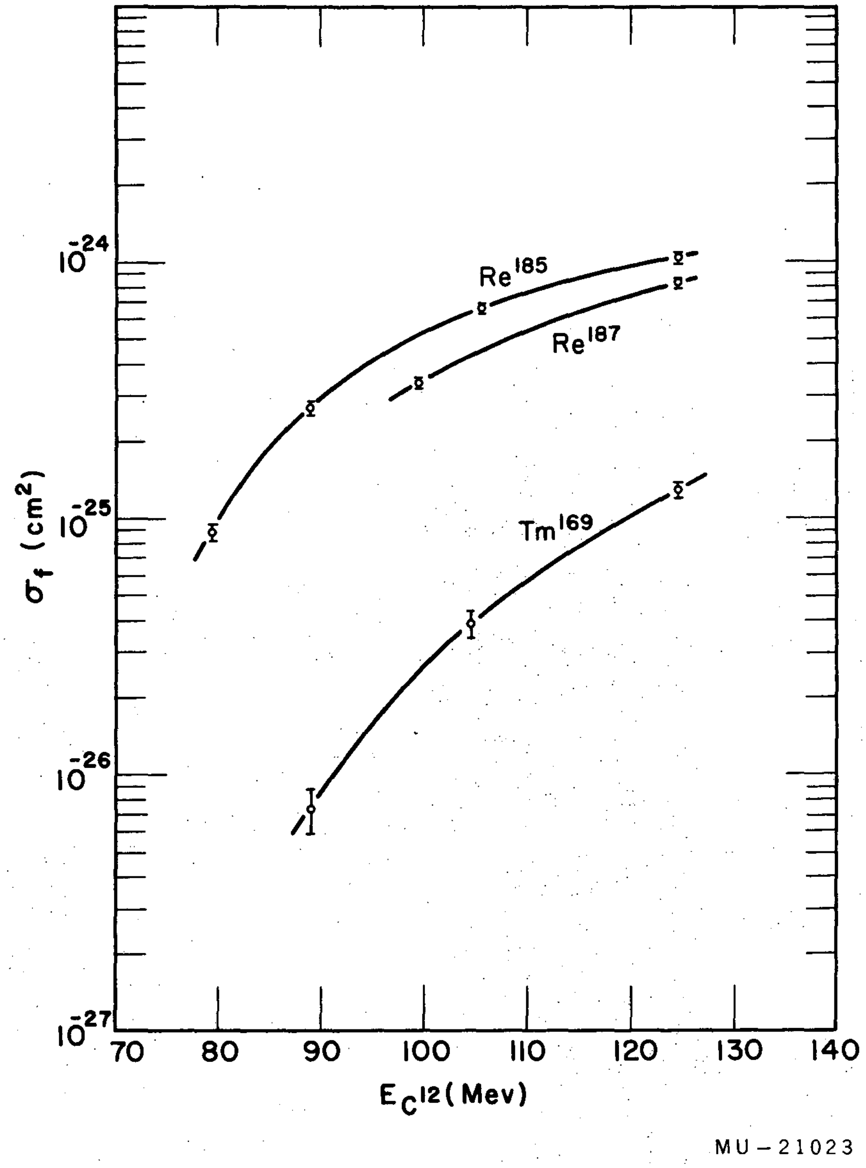
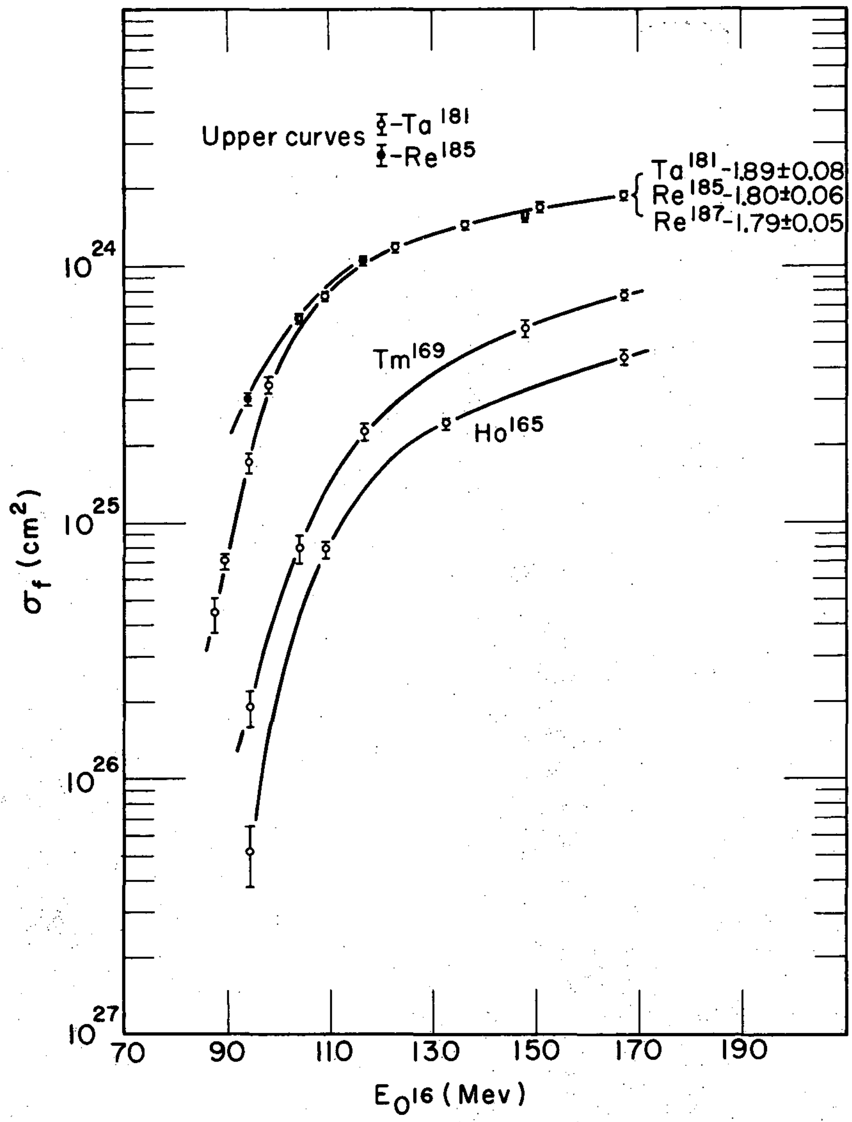
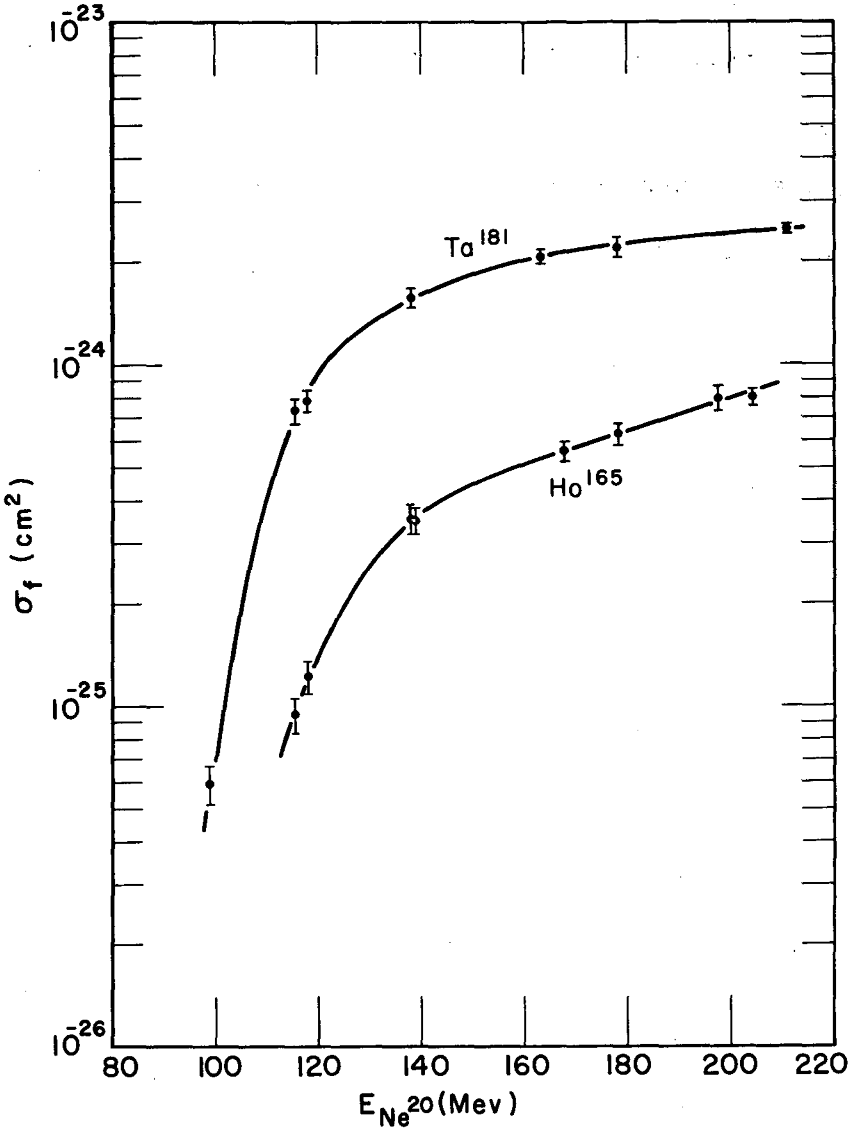


Fig. 5. Fission cross sections for bombardment with C^{12} ions.



MU-21024

Fig. 6. Fission cross sections for bombardment with O^{16} ions.



MU-21025

Fig. 7. Fission cross sections for bombardment with Ne^{20} ions.

Cross sections for compound-nucleus formation, σ_c , were taken from the calculations by Thomas.[19] The model used for these calculations was based on a square-well nuclear potential with a radius parameter of 1.5×10^{-13} cm.

Classical values of maximum orbital angular momentum for spherical nuclei were given by the expression

$$\ell_{max}(\hbar) = [2\mu(R_1+R_2)^2 (E_{cm}-B)]^{1/2} , \qquad (1)$$

where μ = reduced mass of system,

$R = 1.5 \times 10^{-13} A^{1/3}$ cm,

E = c.m. energy of system,

B = Coulomb barrier.

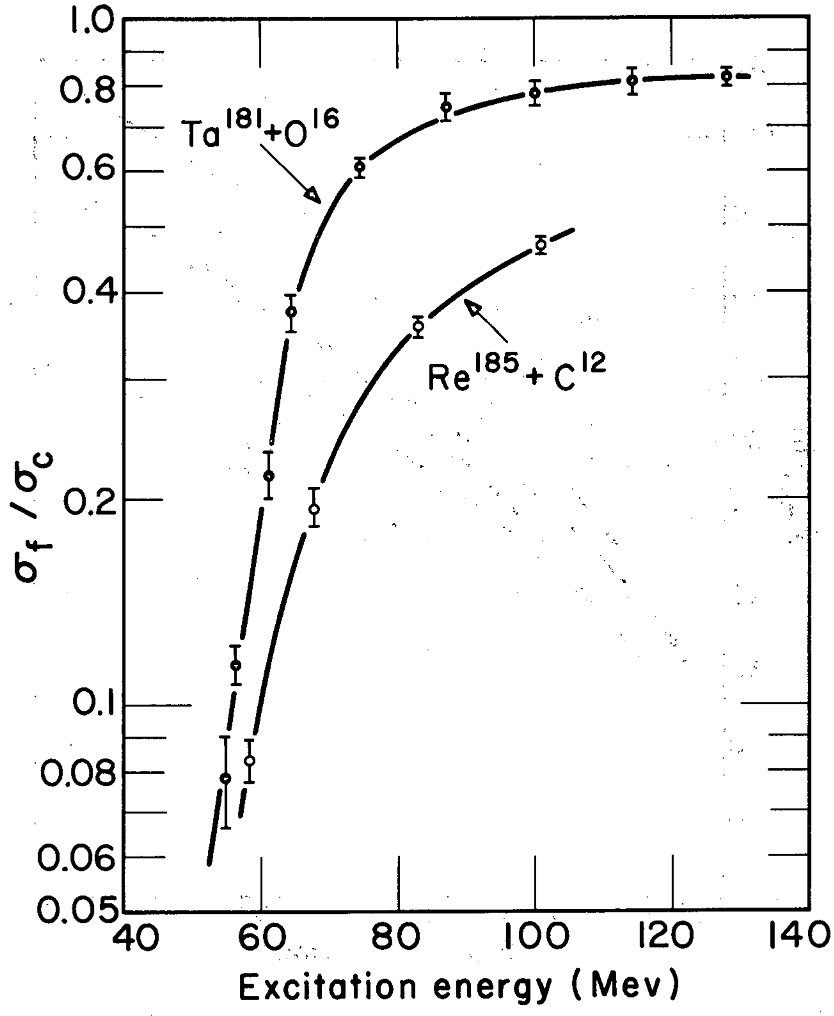
Strong ground-state deformations of target isotopes in this work may affect both ℓ_{max} and σ_c . An estimate of this effect was made by including the quadrupole potential and substituting the semimajor axis of an ellipsoid for the target radius in Eq. (1). Details of this estimate, as well as a discussion of the effect of a mutual polarization of bombarding ion and target on the Coulomb barrier, are given in reference 20.

Fission probabilities, σ_f/σ_c , and angular momenta were calculated and compared for the following pairs of target—bombarding particle systems:

$Re^{185} + O^{16}$, $Ta^{181} + Ne^{20}$, $Tm^{169} + O^{16}$, $Ho^{165} + Ne^{20}$.

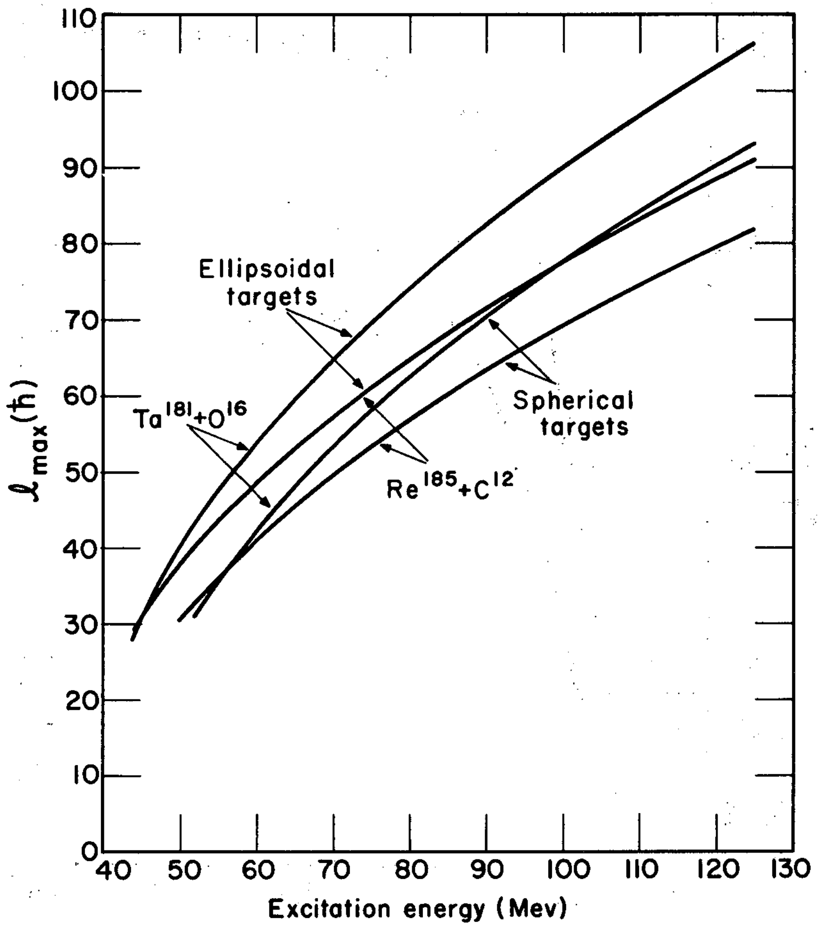
$Re^{185} + C^{12}$, $Ta^{181} + O^{16}$, $Tm^{169} + C^{12}$, $Ho^{165} + O^{18}$.

An example of the results is given in Figs. 8 and 9, showing ℓ_{max} and σ_f/σ_c , respectively, for $Re^{185} + C^{12}$ and $Ta^{181} + O^{16}$.



MU-21044

Fig. 8. Probability for fission in the bombardments $Ta^{181} + O^{16}$ and $Re^{185} + C^{12}$ (square-well σ_c).



MU-21036

Fig. 9. Maximum angular momentum (classical) as a function of excitation energy in the compound nucleus for $Ta^{181} + O^{16}$ and $Re^{185} + C^{12}$.

DISCUSSION

Comparison of Figs. 8 and 9 shows that fission probabilities are higher for the target—bombarding particle system having greater angular momentum. Similar correlations were found in the other pairs of bombardments. These findings support the conclusions of Pik-Pichak[2] and Halpern,[4] but uncertainties in both σ_c and ℓ limit our results to only qualitative significance. Studies of fission-fragment angular distribution[21] and correlations by Sikkeland et al.[22] indicate that fission of the light targets studied here occurs only following compound-nucleus formation. But these authors' work also indicates that a significant fraction of the reaction cross section for heavy ions is due to processes in which only a fraction of the bombarding-particle nucleons amalgamate with the target. For the bombardment of U^{238} with 124-MeV C^{12} ions, 25 ± 5% of the calculated compound-nucleus cross section[19] was estimated to go into these non-compound-nucleus interactions. Further indication of particle breakup reactions is found in Britt and Quinton's measurements[23] of angular distributions and energy spectra of charged particles emitted in reactions of heavy ions with Au^{197} and Bi^{209}. Angular distributions are peaked in the forward direction, and energies of most forward-emitted α particles are consistent with heavy-ion breakup rather than evaporation from compound nuclei.

These results suggest that the calculated σ_c used in deriving fission probability should be regarded as an upper limit to the true compound-nucleus-formation cross section. Further, since non-compound-nucleus reactions probably involve those heavy ions with the largest impact parameters, our classical calculation of angular momentum should also represent an upper limit. The investigations by Sikkeland et al. do indicate, however, that the proportion of non-compound-nucleus interaction is relatively independent of bombarding particles. This result encourages us to assume that the reductions in calculated ℓ and σ_c due to non-compound-nucleus reactions are similar in each of the systems, leading to a given compound nucleus, and to conclude that a relatively larger fission probability is found for higher angular momentum.

Continuing studies of fission-fragment angular correlation and charged-particle emission will help to define the extent of non-compound-nucleus interaction and its effect on angular momentum deposit; more complete knowledge of σ_c and the ℓ distribution may then warrant reinterpretation of our data to establish more precise relationships between angular momentum and fissionability.

ACKNOWLEDGMENTS

We are pleased to acknowledge useful discussion of this work with Dr. John M. Alexander, Dr. Torbjørn Sikkeland, Dr. Wladyslaw J. Swiatecki, Dr. Victor E. Viola, and Mr. Frank Plasil.

FOOTNOTE AND REFERENCES

[*] Work done under the auspices of the U.S. Atomic Energy Commission.

1. V. A. Druin, S. M. Polikanov, and G. N. Flerov, Soviet Physics—JETP 5, 1059 (1957).
2. G. A. Pik-Pichak, Soviet Physics—JETP 7, 238 (1958).
3. John R. Hiskes, The Liquid-Drop Model of Fission: Equilibrium Configurations and Energetics of Uniform Rotating Charged Drops (Ph. D. Thesis), Lawrence Radiation Laboratory Report UCRL-9275, June 1960 (unpublished).
4. I. Halpern, Ann. Rev. Nuclear Sci. 9, 245 (1959).
5. J. R. Huizenga and R. Vandenbosch, Nuclear Fission, to be published in Nuclear Reactions, Vol. 2 (North-Holland Publishing Co., Amsterdam, Netherlands).
6. E. Goldberg and H. L Reynolds, Phys. Rev. 112, 1981 (1958).
7. Edward L. Hubbard, Range—Energy Relation for Heavy Ions in Metals, Lawrence Radiation Laboratory Report UCRL-9053, Jan. 1960 (unpublished).
8. H. H. Heckman, B. L. Perkins, W. G. Simon, F. M. Smith, and W. H. Barkas, Phys. Rev. 117, 544 (1960).
9. R. Levi and G. A. Espersen, Phys. Rev. 78, 231 (1960).
10. G. W. W. Stevens, in Fundamental Mechanisms of Photographic Sensitivity (Symposium) (Butterworth's Scientific Publications, London, 1951), p. 310.
11. E. Goldberg, H. L. Reynolds, and D. D. Kerlee, Phys. Rev. 119, 2009 (1960).
12. J. B. Marion, T. I. Arnette, and H. C. Owens, Oak Ridge National Laboratory Report ORNL-2574 (1959).

13. J. Terrell, as presented by R. B. Leachman, Second United Nations Conference on the Peaceful Uses of Atomic Energy, Geneva (United Nations, Geneva, 1958), Vol. 15, 231.

14. G. E. Gordon, A. E. Larsh, T. Sikkeland, and G. T. Seaborg, Phys. Rev. 120, 1341 (1960).

15. H. C. Britt and A. R. Quinton, Phys. Rev. 120, 1768 (1960).

16. H. Marshall Blann, Fission of Gold with 112-MeV C^{12} Ions: a Yield-Mass and Charge- Distribution Study (Ph. D. Thesis), Lawrence Radiation Laboratory UCRL-9190, May 1960 (unpublished).

17. N. I. Tarantin, Iu. B. Gerlit, L. I. Guseva, B. F. Miasoedov, K. V. Filippova, and G. N. Flerov, Soviet Physics—JETP 7, 220 (1958).

18. Victor E. Viola, Angular Distributions from Heavy-Ion-Induced Fission (Ph. D. Thesis) Lawrence Radiation Laboratory Report UCRL-9619,March 1961 (unpublished).

19. T. D. Thomas, Phys. Rev. 116, 703 (1959).

20. John Gilmore, The Effect of Angular Momentum on Fission Probability (Ph. D. Thesis) Lawrence Radiation Laboratory Report UCRL-9304, July 1960 (unpublished).

21. T. Sikkeland, A. E. Larsh, and G. E. Gordon, Phys. Rev. 123, 2112 (1961).

22. T. Sikkeland, E. L. Haines, and V. E. Viola, Phys. Rev. 125, 1350 (1962).

23. H. C. Britt and A. R. Quinton, Phys. Rev. 124, 877 (1961).