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FAST AND SLOW FISSION

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ABSTRACT

Measurements of alpha particle induced fission of actinide nuclei and fission of the composite system ^{170}Yb formed in ^{12}C and ^{20}Ne bombardments both show significantly greater neutron emission prior to fission than is consistant with current statistical models. Implications of these results are discussed in the context of possible extreme models: 1) the enhancement of fission at low excitation energies due to shell effects; 2) the inhibition of fission at high excitations due to a limiting of the fission width and 3) the possibility of significant neutron emission during the descent from saddle to scission. In addition the apparent incompatability between current models of incomplete fusion processes and the analysis of light heavy ion induced fission which ignore incomplete fusion is discussed.

INTRODUCTION

Over the past ten years the properties of actinide fission at excitation energies within a few Mev of the barrier have become rather well defined and fission decay properties are reasonably well understood in terms of fundamental characteristics of the underlying potential energy surface.[1,2] These studies show that deformed nuclear shells[3] have a very important effect on this potential energy surface and that macroscopic symmetries[4] of the nuclear shape in the region of the saddle points have a dramatic effect on the relative fission decay rates. In contrast fission at higher excitation energies or in cases involving large angular momenta is more poorly understood at either a fundamental or empirical level.

In this paper we will try to bring together a variety of experimental results which suggest that:

1. At all angular momenta and excitation energies above ∿20 MeV fission is much slower than expected in most conventional models.

2. Present interpretations of fission data from light heavy ion experiments appear fundamentally inconsistent with current concepts of incomplete fusion.

"SLOW" FISSION

At relatively low excitation energies it has been found[4,5] that collective enhancements of the level densities have an important effect on fission probabilities for actinide nuclei. Figure 1 shows the effect of a triaxial shape at the first peak of the fission barrier on the fission probability for ^{237}Np. The microscopic statistical model used to calculate P_f for the axially symmetric case assumes a stable γ deformation and the fission enhancement comes from the additional rotational levels associated with this shape. This triaxial shape comes as a result of a triaxial shell which lowers the potential energy and an antishell which raises the potential energy surface for the axially symmetric shape.[6] Since the effect of these shells is expected to diminish at higher excitation energies it is expected that the saddle point shape will then undergo a transition to the axially symmetric shape characteristic of the liquid drop potential energy surface and this transition will result in a decrease in fission probability. There are currently no reliable theoretical estimates of the energy region where this transition might occur or even a formulation of how to quantitatively include such a transition in the microscopic statistical models used to calculate fission probability distributions. There are, however, several sources of experimental evidence that suggest that first chance fission probabilities have significantly decreased at excitation energies as low as 20 MeV from values expected in a statistical model calculation with a triaxial first barrier.

For the fissioning system ^{236}U Madland and Nix[7] have performed an unfolding of (n,f) data for a series of uranium isotopes to obtain an estimate of the first chance fission probability (P_f). Their results shown in Fig. 2 suggest that P_f may have dropped by about a factor of two at $E^* \sim 21$ MeV as compared to the plateau value observed in the $E^* = 6$-12 MeV region. For comparison a statistical model fit to the theshold region with a triaxial first barrier predicts a slow increase in P_f in the $E^* = 12$-20 MeV region.

Another method of obtaining information on the high energy behavior of P_f was developed by Cheifetz and Fraenkel.[8] They showed that measurements of energy and angular distributions of the neutrons in coincidence with fission can be used to deduce the average numbers of neutrons before and after fission. This technique utilizes the fact that prefission neutrons are emitted approximately isotropically from the center of mass for the fissioning system whereas the postfission neutrons come from the fully accelerated fragments. Using this technique the following systems have been studied: 12 MeV p + $^{238}\mathrm{U}$[8], 45 MeV α + $^{209}\mathrm{Bi}$, $^{232}\mathrm{Th}$, $^{233,238}\mathrm{U}$ and $^{239}\mathrm{Pu}$[9]; 155 MeV p + $^{209}\mathrm{Bi}$, $^{238}\mathrm{U}$[10]; and $^{170}\mathrm{Yb}$ excited by $^{12}\mathrm{C}$ + $^{158}\mathrm{Gd}$ and $^{20}\mathrm{Ne}$ + $^{150}\mathrm{Nd}$ reactions.[11]

The results from experiments on actinide nuclei are sumarized in Table I. It has been previously concluded[9,10] that these data suggest a decrease in Γ_f/Γ_n at high excitation energies. Analysis of 45 MeV $^4\mathrm{He}$ data indicate in a model independent manner that the average excitation energy of the fissioning nucleus is approximately 10-20 MeV (i.e. 20-30 MeV dissipated on prefission neutrons on the average). This result coupled with a total fission probability of $\sim$1 is incompatable with a simple statistical model calculation. However, at least qualitatively the data appear consistent with a model where Γ_f/Γ_n is strongly enhanced at low energies due to the triaxial shape at the first barrier and this enhancement begins to disappear in the 10-20 MeV excitation energy range.

Thus, for actinide nuclei both the unfolded P_f for $^{236}\mathrm{U}$[7] and the neutron emission measurements[9,10] appear empirically consistent and in terms of current theories of fission seem to suggest that the triaxial shell at the first barrier is washing out in the excitation energy range 10-20 MeV. There are currently no detailed theoretical calculations which can either substantiate or refute this hypothesis. A more radical hypothesis to explain these results would be that the statistical model itself is starting to breakdown in this energy region so that Γ_f does not rise as fast as estimated from fits to low energy data while Γ_n remains approximately statistical. One version of such a model has recently been suggested by Grange and Weidenmüller.[12] A third possibility could be that the transition from saddle to scission is much slower than previously estimated[10] so that neutron emission becomes probable from the fissioning system after it has passed the saddle. Invoking a one body dissipation mechanism does lead to longer saddle to scission times ($\sim10^{-20}$sec)[14] but at $\sim$20-40 MeV excitation energies this should still be shorter than neutron emission times ($\sim10^{-19}$ + few x 10^{-20} sec).

The recent results of Gavron et al[11] on the compound system $^{170}\mathrm{Yb}$ have generated renewed interest in these questions because for this very different system neutron measurements again indicate an anomolously large number of prefission neutrons compared to statistical model calculations for the 194 MeV $^{12}\mathrm{C}$ and 174 MeV $^{20}\mathrm{Ne}$ bombardments. The recent data at various detection angles for neutron spectra in coincidence with fission and evaporation residue products are shown in Figures 3-4. The spectra have been fit using a monte-carlo simulation technique with the constraint that the prefission neutron spectra would have the same temperature as that determined in an evaporation residue experiment. Qualitatively, the similarity of the two sets of data suggest a large number of prefission neutrons. Table II shows that the results from the fits to the fission coincidence data confirm this expectation. Furthermore, it is found that predictions of a statistical model incorporating a rotating liquid drop fission barrier with fermi gas level densities can not reproduce these results and at the same time fit fission cross section data in this mass region.

In discussing the actinide results presented above it seemed natural to first suggest a hypothesis based on the enhancement of Γ_f at low energies due to a triaxial shell that had previously been predicted and experimentally verified. The appearance of a similarly anomalous Γ_f/Γ_n for ^{170}Yb at high angular momentum would seem to suggest a more general phenomenon possibly in connection with the dynamics of fission. However, it could still be that shells and rotational enhancements are important in ^{170}Yb since calculations[13] of the ground state shapes for spins of ∿60-80ħ indicate a triaxial shape for neutron numbers greater than 90 in nearby Erbium isotopes. At the other extreme it could be that a significant fraction of the neutrons are emitted between saddle and scission. At the relevant excitation energies (∿50-150 MeV) neutron emission times become as short as a few x 10^{-21} sec which could be shorter than the saddle to scission time if the one body dissipation hypothesis is correct.[14]

Finally, data is also available for the reaction 155 MeV p + ^{209}Bi.[10] Here again the large number of prefission compound neutrons and the measured spallation cross sections can not be reproduced in a normal statistical model calculation.[10] In this case an internucleon cascade calculation is used to estimate the excitation energy distribution for the "compound" residues that then decay by neutron emission and fission. This case seems intermediate between the actinide and ^{170}Yb cases discussed above. The mean excitation energies are intermediate between these two cases and the angular momenta involved are modest. This is quite a different shell region from either actinide or rare earth nuclei (i.e. ^{210}Po is near a doubly magic spherical shell) and one would not a priori expect specific shell generated enhancement effects to be the same as in actinide nuclei. The evidence for the existence of large numbers of prefission neutrons in three very different regions would seem to suggest a common general mechanism but as discussed above we can not at present identify a single dominant effect that might be important in these different cases.

The present situation can best be summarized as follows: (1) Experiments indicate an anomalous ratio of prefission to postfission neutrons for actinides at excitation energies above ∿20 MeV, for ^{170}Yb at excitation energies of 135 and 170 MeV and for ^{210}Po at an average excitation energy of ∿100 MeV; (2) Theoretical hypothesis involving enhancements of Γ_f at low energies <u>or</u> limiting of Γ_f at high energies <u>or</u> the emission of neutrons between saddle and scission could qualitatively explain these results; (3) Quantitative theoretical models are needed to sort out the relative importance of these very different physical effects and (4) Predictions and conclusions from current statistical model analyses of fission data at moderate excitation energies may be suspect since these models are incapable of qualitatively reproducing the experimental ratios of prefission to postfission neutrons.

For fission of the composite system ^{170}Yb induced by 194 MeV ^{12}C and 174 MeV ^{20}Ne projectiles the calculated maximum angular momenta contributing to fusion are 72 ħ and 79 ħ respectively. These angular momenta are above the values $\ell \sim 65$ ħ for which the rotating liquid drop model (RLD) predicts[15] that the fission barrier equals the neutron binding energy but still below the values ($\ell \sim 85$ ħ) where the fission barrier is expected to vanish and thus, one would expect to observe significant cross sections for compound fission as discussed in the preceeding section. An additional experiment with 239 MeV ^{20}Ne projectiles leads to a critical angular momentum of 99 ħ which is well into the region of vanishing fission barriers. This represents a case where much of the "fusion" cross section is in a region where normal statistical models do not apply since $B_f = 0$. Initial analysis of the data[11] indicated that for this reaction, fission was "fast" relative to the characteristic neutron evaporation time. However, a subsequent, more comprehensive evaluation of these data[16] indicated an error in the analysis. When corrected the resulting spectra in coincidence with fission fragments resemble the spectra in coincidence with evaporation residues indicating that fission is a slow process even at angular momenta at which the barrier is zero. Similar results have also been published by Hilscher et al.[17]

LIGHT HEAVY ION REACTIONS

There is considerable evidence from evaporation residue studies than an entrance channel limit exists for the angular momentum of a fused system formed in light heavy ion reactions. For example the cross section data[18] for the ^{12}C + ^{160}Gd reaction suggest a limiting angular momentum of 43 ± 3 ħ with higher partial waves contributing to an incomplete fusion process where only part of the projectile is captured. This interpretation seems substantiated by γ ray multiplicity experiments[19] for the system ^{16}O + ^{154}Sm which seem to show a saturation of the maximum angular momentum at values of about 50 ħ. These results have been sucessfully interpreted in terms of an entrance channel model[20,21] of incomplete fusion which seems to give a reasonable overall picture of these reactions when they lead to the formation of evaporation-like residue products. However, no attempt has yet been made to reconcile these results with light heavy ion induced fission data and the statistical analysis of these data in terms of a rotating liquid drop model. It should especially be noted that in the mass ∼170 region the rotating liquid drop model predicts that the fission barrier should equal the neutron binding energy at an angular momentum of ∼60 ħ significantly above the cutoff expected from incomplete fusion for the ^{12}C reaction. Current statistical models[11,15,22] used to analyse the above neutron emission data and the available fission cross section data do not include any provisions for entrance channel limitations to the angular momentum of the fused systems and most of the fission reactions in these models come from angular momenta near or above the region where $B_f = B_n$. Clearly, the fission models and the incomplete fusion model are inconsistent in their present form.

At present the data for light heavy ion induced fission reactions in the rare earth region are limited and the statistical models are necessarily of a qualitative nature because of the assumptions of rotating liquid drop fission barriers and fermi gas level density distributions. In general the deficiencies of this model can be masked by treating the ratio of level density parameters, a_f/a_n, and a renormalization constant for the fission barrier as adjustable parameters. In practice this means that data from a single reaction can quite often be fit by a range of parameters and it seems possible that effects due to incomplete fusion might be masked. There do exist, however, a few cases where fission excitation functions exist for several reactions leading to the same composite system. The most extensive data are from Sikkeland and coworkers [23,24] for the systems ^{181}Re formed in ^{12}C, ^{16}O and ^{22}Ne bombardments and ^{186}Os formed in ^{11}B, ^{12}C and ^{16}O bombardments. In addition ^{186}Os has also been studied in an (α,f) experiment.[25] In order to try to look for possible effects due to incomplete fusion we have tried to refit these data with a statistical model[26]. The results are shown in Figs. 5 and 6 as a ratio of the experimental to calculated cross sections versus the critical angular momentum for fusion from a Bass Model. Figure 5 also shows the calculated limiting angular momentum in the entrance channel from the Wilczynski model of incomplete fusion[21]. For reference we show the RLD calculation of the fission barrier[15]. Because of the limitations in both experiment (energy variation via degrader foils) and the calculations (RLD + Fermi gas level density) it is not possible to make a definitive conclusion from these results but we believe that these comparisons do not show any strong evidence for decreased experimental cross sections for $\ell > \ell_w$ especially in the ^{16}O, ^{22}Ne cases. In the ^{12}C case which should be most affected by incomplete fusion the data do not go very far into the region of interest. For ^{12}C the ^{181}Re data show a decrease in $\sigma_{expt}/\sigma_{calc}$ (but always remaining above 1) while the ^{186}Os data show $\sigma_{expt}/\sigma_{calc} \sim$ constant but at a value of .6 - .7. Clearly more extensive data and improved modeling are needed to assess the importance of entrance channel limitations on the fusion-fission process but particularly for ^{16}O and ^{22}Ne bombardments it seems difficult to reconcile the large cross sections (500-1000 mb) at the highest energies with an entrance channel limit to complete fusion.

CONCLUSIONS

In this paper we have tried to draw on both relatively new experimental results and some considerably older data to point out that there exist several areas in which we do not yet understand the fission process and light heavy ion reactions at a relatively fundamental level. First data from neutron emission experiments indicate that fusion-fission processes seem to occur much slower than expected from current statistical models in a variety of systems including ^{170}Yb, ^{210}Po and several actinides at modest excitation energies. The results from experiments in these different regions of mass, energy and angular momentum seem very similar but current most plausible explanations are quite different. For actinides this effect could be created by shell effects on Γ_f and for ^{170}Yb the apparent low values of Γ_f/Γ_n could result from the misidentification of neutrons emitted between saddle and scission as being compound nucleus neutrons. In both cases there are also alternative explanations and a comprehensive understanding will require both more experimental results and more quantitative fission calculations.

An additional problem in trying to understand the angular momentum dependence of fissionlike processes is that there are still uncertainties in the basic character of the light heavy ion reactions that are most useful in creating composite systems with angular momenta in the 50-150 $\hbar$ region. In particular, existing statistical models of heavy ion induced fission reactions do not include (nor seem to require) the concept of entrance channel limits to the angular momentum (i.e., incomplete fusion) of fused systems which seems necessary to explain existing data on evaporation residue production. This apparent contradiction might be explained in models including one or more of the following extremes: (1) fission models may have disguised the incomplete fusion effects by variations in their arbitrary parameters, (2) a fast fission-like process may complete directly with the fast particle emission that feed the incompletely fused evaporation residues (but fission seems abnormally slow, i.e., many precision neutrons), and (3) could a significant fraction of the residue events identified as incomplete fusion be coming from slow alpha particle evaporation from superdeformed shapes[28] and thus compete with compound fission.

Because of the uncertainties and ambiguities in our understanding of fission and light heavy ion reactions it seems doubtful that meaningful estimates of important physical quantities (e.g. fission barrier) can be reliably extracted from measured fission data. However, it does seem promising that more detailed experiments could lead to new insights on macroscopic nuclear properties.

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Table I. Average numbers of neutrons emitted prior to fission (ν_{pre}) and after fission (ν_{post}) for series of reactions involving actinide nuclei. Statistical model calculations assume a single fission barrier and fermi gas level densities. Data and calculations are from Fraenkel et al (Ref. 9) and Cheifetz et al (Ref. 10).

Reaction	$P + {}^{238}U$	$\alpha + {}^{232}Th$	$\alpha + {}^{233}U$	$\alpha + {}^{238}U$	$\alpha + {}^{239}Pu$	$P + {}^{238}U$
E* (MeV)	18	40	39	39	38	a
Expt ν_{pre}	0.62±.25	2.9±.9	3.3±1.5	3.6±1.6	2.7±0.8	5.8±1.0
Expt ν_{post}	3.9 ±.2	4.4±.3	4.2±1.7	4.6±0.7	5.1±0.3	5.1±0.5
Expt $\sigma_{\alpha,4n}/\sigma_f$	——	0.02	0.0002	——	0.0004	0.03 mb[b]
Calculations for $a_f = a_n = A/20 \rightarrow A/8$						
ν_{pre}	.2 →.4	∼2.7	∼1.8	∼2.8	∼2.2	5.8
$\sigma_{\alpha,4n}/\sigma_f$		.05→.7	.2→.7	.1→.5	.5→.8	4.4 mb[b]
Calculations for $a_f = 1.33\ a_n = A/20$						
ν_{pre}	——	.04	.03	.07	.06	——
$\sigma_{\alpha,4n}/\sigma f$	——	0	0	0	0	——

[a] E_{Lab} = 155 MeV

[b] value for evaporation residue cross section.

Table II. Reactions and Results from Experiments of Gavron et al (Ref. 11) involving the composite system ^{170}Yb. ℓcrit is the critical angular momentum associated with fusion as calculated from a Bass Model

Reaction	$^{12}C+^{158}Gd$	$^{20}Ne+^{150}Nd$	$^{20}Ne+^{150}Nd$
E_{Lab} (MeV)	192	176	239
E* (MeV)	169	135	191
ℓcrit (ħ)	72	79	99
Expt ν_{pre}	6±1	5±1	1±1
Expt ν_{post}	3±1	3±1	8±1
Calc. ν_{pre}[a]	3.4	2.2	
Calc. ν_{pre}[b]	2.2	1.0	

[a] $a_f/a_n = 1.0$ $B_f = 0.8$ RLD

[b] $a_f/a_n = 1.04$ $B_f = 0.98$ RLD

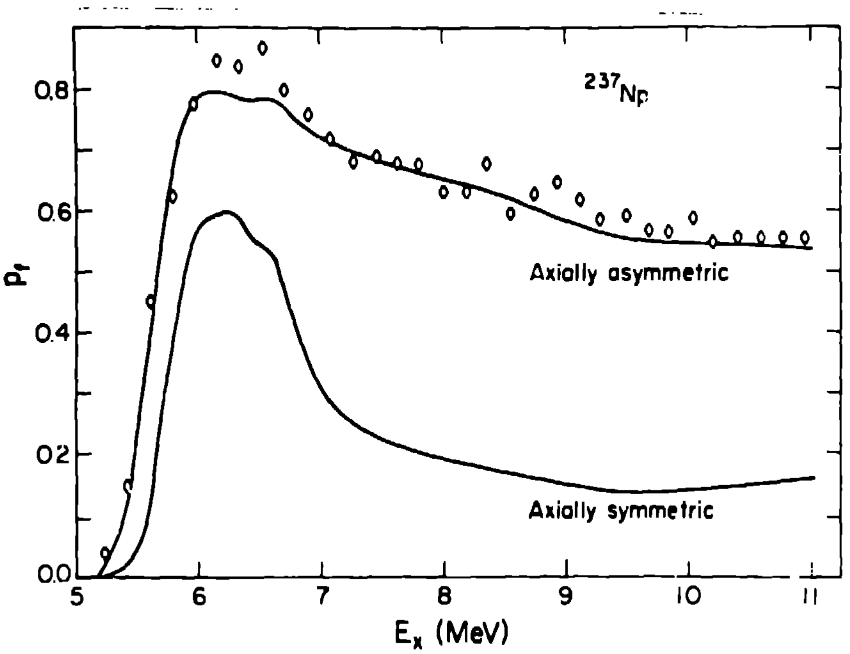

Fig. 1. Fission probability for ^{237}Np compared to calculations using a microscopic statistical model. Upper curve assumes an axially asymmetric shape at the first saddle point while lower curve assumes axial symmetry. (from Ref. 5)

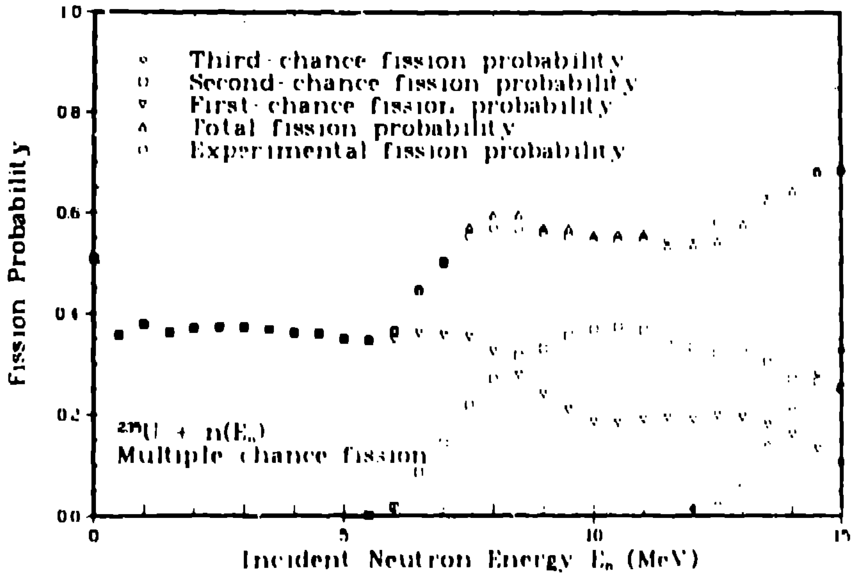

Fig. 2. Fission probabilities deduced from unfolding (n,f) cross sections for various uranium nuclei. (from Ref. 7)

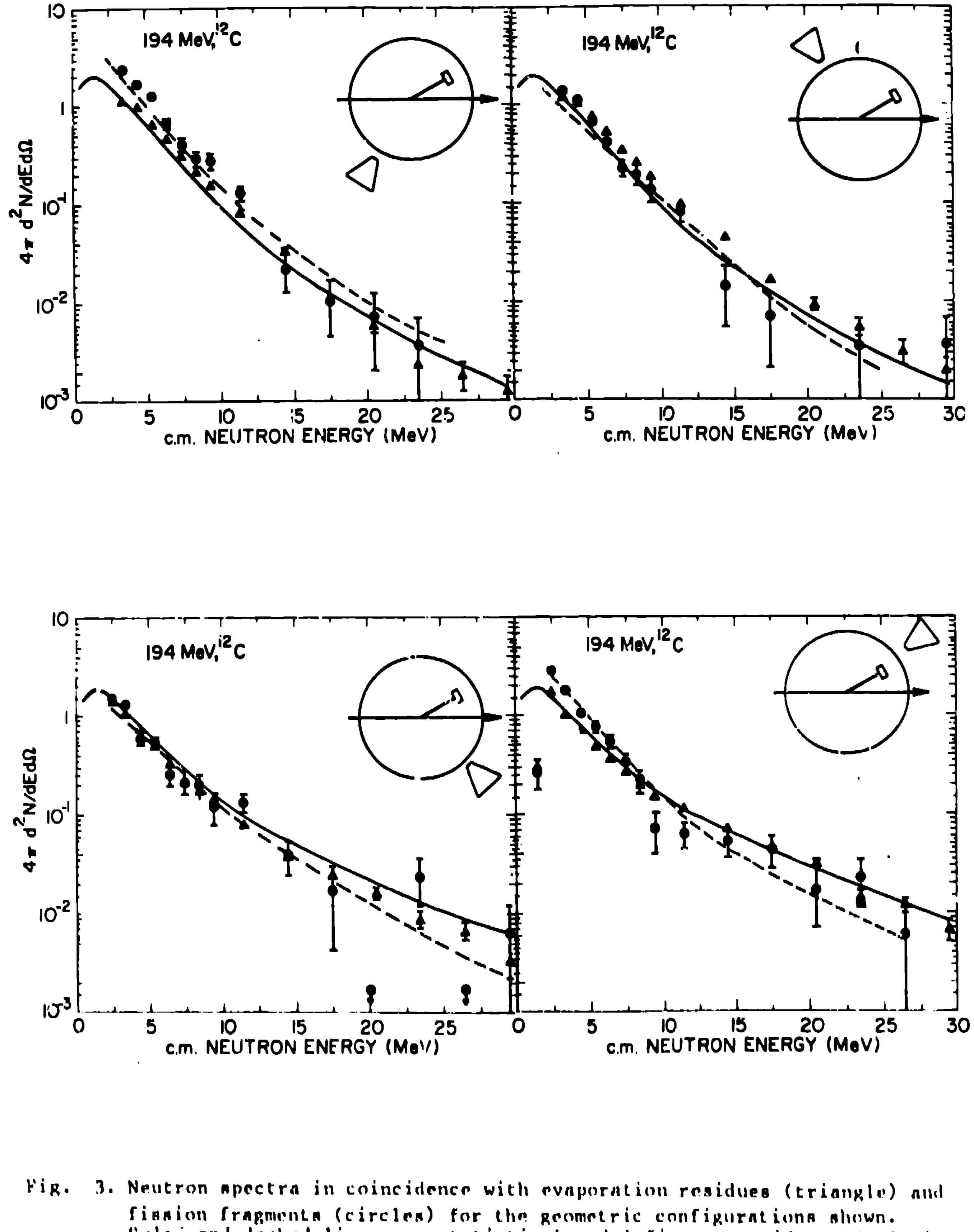

Fig. 3. Neutron spectra in coincidence with evaporation residues (triangle) and fission fragments (circles) for the geometric configurations shown. Solid and dashed lines are statistical model fits to residue and fission data, respectively. (from Ref. 11)

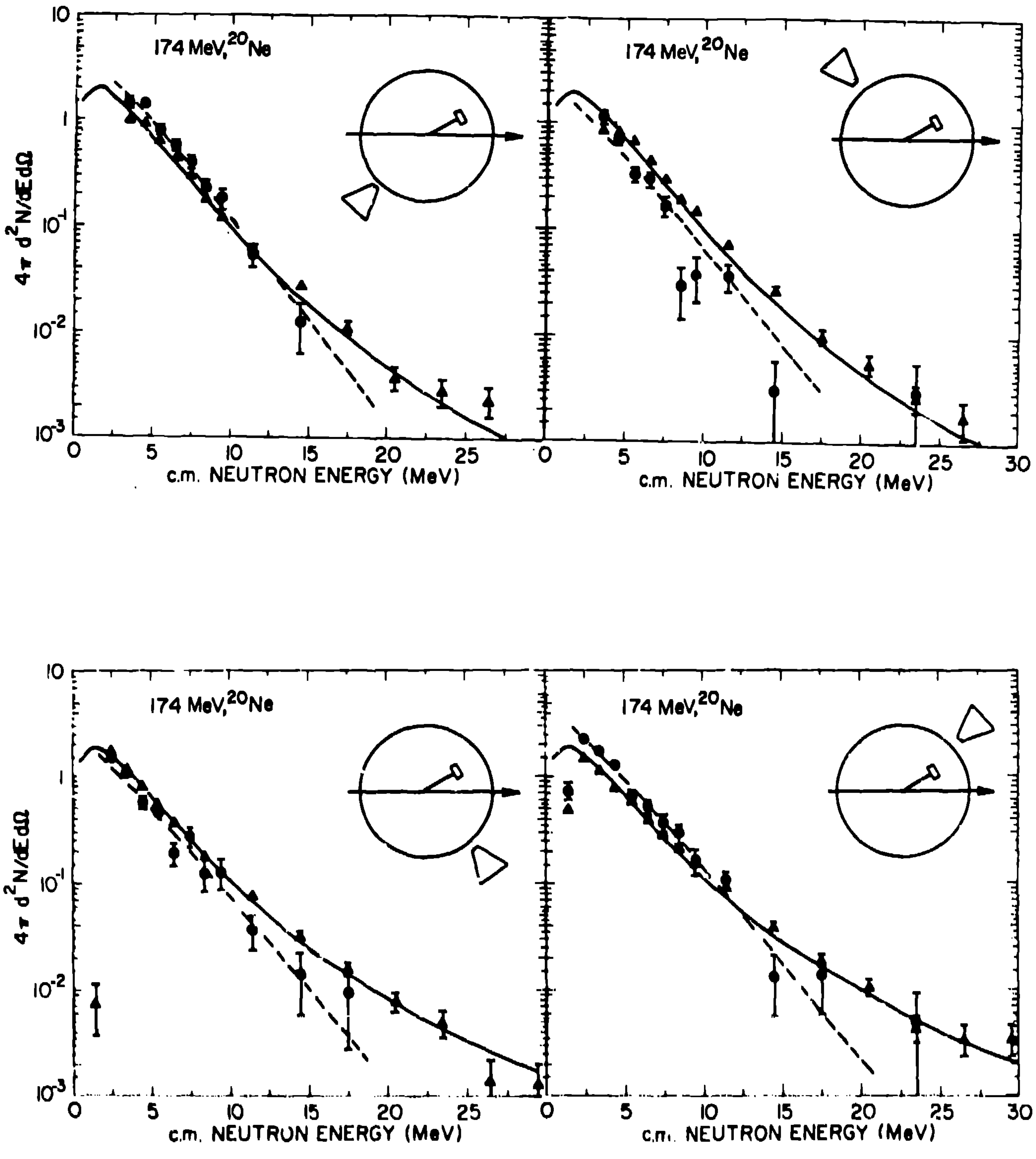

Fig. 4. Neutron spectra in coincidence with evaporation residues (triangle) and fission fragments (circles) for the geometric configurations shown. Solid and dashed lines are statistical model fits to residue and fission data, respectively. (from Ref. 11)

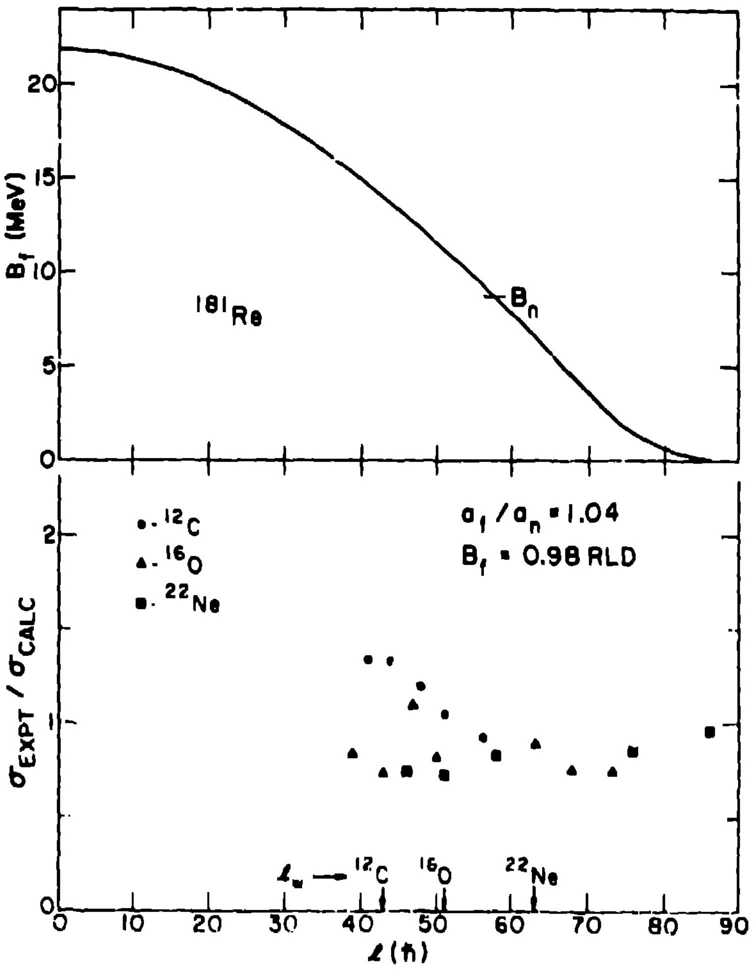

Fig. 5. Ratio experimental to calculated fission cross sections for the composite system ^{184}Re. Data is taken from Ref. 23,24. Parameters for statistical model calculation are shown. For lower half axis is ℓ_{crit}, the maximum angular momentum leading to fusion as calculated from a Bass model. Arrows indicate values for ℓ_w the cutoff expected due to incomplete fusion (Ref. 21) Upper half shows calculated fission barrier from rotating liquid drop model (Ref. 15).

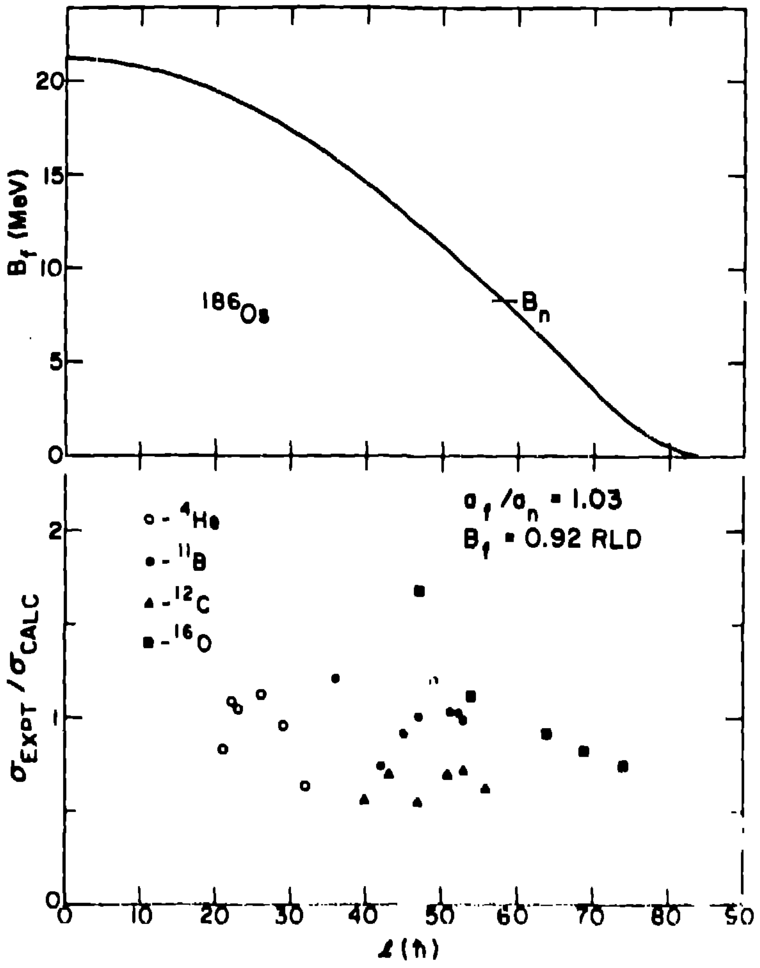

Fig. 6. Ratio experimental to calculated fission cross sections for the composite system ^{186}Os. Data is taken from Ref. 23-25 Parameters for statistical model calculation are shown. For lower half axis is ℓ_{crit} the maximum angular momentum leading to fusion as calculated from a Bass model. Upper half shows calculated fission barrier from rotating liquid drop model (Ref. 15).