

## Neutron Emission from Fission Fragments during Acceleration

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Fission-neutron angular correlations following fusion of  $^{19}\text{F}$  and  $^{232}\text{Th}$  have been measured. Conventional analysis, based on the approximation that post-fission neutrons originate only from fully accelerated fission fragments, gives unexpectedly large numbers of "prefission" neutrons. Comparison with the considerably less fissile system  $^{200}\text{Pb}$  gives the first convincing evidence that this approach is inadequate. Consideration of neutron emission from the accelerating fragments gives results consistent with expectations.

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The investigation of heavy-ion fusion-fission has recently become a field of great activity, centered on measurements of fission and evaporation-residue excitation functions,<sup>1</sup> and their analysis with use of the statistical model. The principal parameters varied to fit the data are (i) the ratio of the level-density parameters at the saddle-point and equilibrium deformations ( $a_f/a_n$ ), and (ii) the scaling factor ( $k_f$ ) of the rotating liquid-drop model fission barrier. Good fits have been obtained, though a large correlated range of these parameters may be allowed.<sup>2</sup>

As the compound nucleus (CN) cools by neutron evaporation from its initial excitation energy ( $E_x$ ), fission may occur at any stage. For a given angular momentum, the competition between neutron emission and fission as a function of thermal excitation energy ( $U_x$ ) determines the multiplicity of neutrons evaporated before fission ( $\nu_{\text{pre}}$ ), and calculated values of  $\nu_{\text{pre}}$  are governed principally by  $a_f/a_n$ .<sup>3</sup> The average prefission neutron multiplicity (and thus  $a_f/a_n$ ) can be determined by measuring the angular correlation between neutrons and fission fragments; the post-fission neutrons, emitted by the fast-moving fragments, have strong kinematic focusing along the fragment direction, while the prefission neutrons do not. Measurements of  $\nu_{\text{pre}}$  following the fusion of  $^{19}\text{F}$  with  $^{181}\text{Ta}$  (to give  $^{200}\text{Pb}$ ) have been made<sup>3</sup> at low CN excitation energies ( $E_x = 65$  to  $82$  MeV). Agreement between experiment and calculations was obtained with  $a_f/a_n = 1.02$ , in accordance with expectations. However, measurements of this type for reactions of  $^{20}\text{Ne}$  on  $^{150}\text{Nd}$ <sup>4</sup> and  $^{165}\text{Ho}$ ,<sup>5</sup> producing CN with high  $E_x$  (135 to 325 MeV), have given values of  $\nu_{\text{pre}}$  much larger than predicted by the statistical model.

The analyses of these experiments use the approximation that neutrons are emitted isotropically in the rest frame of the source. This approximation

is unlikely to cause major error. They also include the conventional assumption that the neutron angular correlation has only *two* components, due to neutrons emitted from the compound nucleus (either before or after equilibrium is attained),  $\nu_{\text{pre}}$ , and from the *fully accelerated* fission fragments,  $\nu_{\text{post}}$ . If a significant number of neutrons were emitted *during* the acceleration of the fission fragments to their asymptotic velocity ( $V_\infty$ ),<sup>6</sup> this conventional analysis would be in error, tending to give too many prefission neutrons. The acceleration time is almost independent of the fissioning system, taking  $\sim 10^{-20}$  s to reach  $0.9V_\infty$ . For several neutrons to be emitted from the accelerating fragments, the lifetime for neutron evaporation should be small compared to this time, requiring high thermal excitation energy  $U_x$  and/or low neutron binding energy  $B_n$  in the fragments. Thus this mechanism may help to explain the anomalously large values of  $\nu_{\text{pre}}$  inferred from measurements at high  $E_x$ .

We have attempted to isolate any effect of neutron evaporation from the fragments during acceleration by studying two different compound nuclei,  $^{200}\text{Pb}$  and  $^{251}\text{Es}$ , produced with similar excitation energy and angular momentum distributions.  $^{251}\text{Es}$  has a much lower fission barrier, and so is more likely than  $^{200}\text{Pb}$  to undergo fission before emitting neutrons, and  $\nu_{\text{pre}}$  would be expected to be considerably smaller. However, the  $Q$  values for fission are such that for  $^{200}\text{Pb}$  there is almost no gain in  $U_x$  at scission, while for  $^{251}\text{Es}$ , the gain is  $\sim 40$  MeV. Thus more neutrons should be emitted during acceleration of the hotter fragments of  $^{251}\text{Es}$ .

Neutron-fission-fragment angular correlations were measured following the bombardment of targets of  $^{181}\text{Ta}$  ( $0.9$  mg  $\text{cm}^{-1}$ ) and  $^{232}\text{Th}$  ( $1.5$  mg  $\text{cm}^{-2}$ ), respectively, by 105- to 138-MeV beams of  $^{19}\text{F}$  from the Australian National University's model 14UD Pelletron accelerator. Complete

fusion is expected to be dominant at these energies. The apparatus<sup>3</sup> consisted of two fission detectors, at 0° and 90° to a neutron detector (NE 213), all perpendicular to the beam axis. The experimental data were first analyzed in the conventional way (see, for example, Ref. 3). Good agreement with previous measurements<sup>3</sup> for <sup>200</sup>Pb was obtained. For <sup>251</sup>Es, the experimental and fitted neutron velocity spectra at 124 MeV bombarding energy are shown in Fig. 1(a). From such fits, the multiplicities shown in Fig. 2(a) were obtained. The values of  $\nu_{pre}$  for <sup>251</sup>Es are at least as large as for <sup>200</sup>Pb at the same  $E_x$ . In contrast, statistical model calculations by ALERT1,<sup>7</sup> with  $a_f/a_n$  and  $k_f=1.0$ , predict  $\nu_{pre}=0.65$ , almost independent of  $E_x$  (Fig. 2), while for  $a_f/a_n > 1.0$ , or  $k_f < 1.0$ ,  $\nu_{pre}$  is even smaller. This result suggests that neutron emission from the accelerating fragments is an important process in the fission of <sup>251</sup>Es.

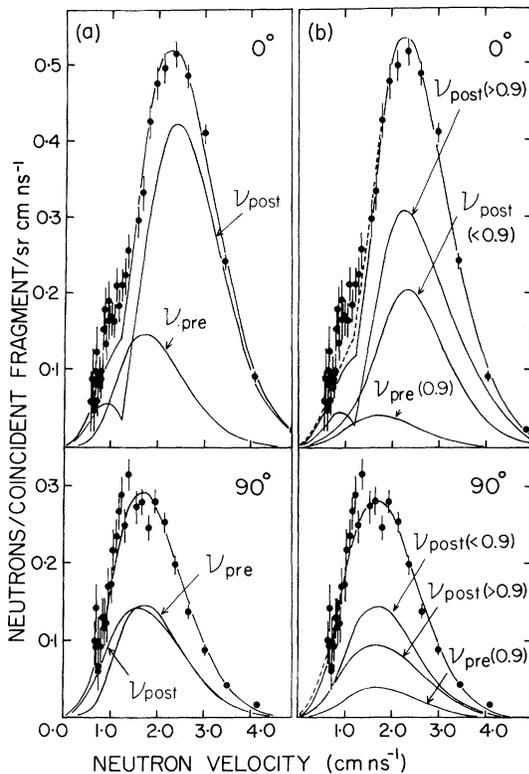


FIG. 1. Experimental and fitted laboratory neutron velocity spectra at 0° and 90° to the detected fragment for <sup>251</sup>Es at 124 MeV bombarding energy: (a) for a conventional analysis allowing only two components,  $\nu_{pre}$  and  $\nu_{post}$ , (b) including neutron emission during fragment acceleration, using the spectrum of  $\nu_{pre}$  to calculate  $\nu_{post}(<0.9)$  (see text). The dashed line shows the fit obtained with use of  $\nu_{post}$  rather than  $\nu_{pre}$ .

We have made a quantitative calculation to test the above hypothesis. The following approximations were used: (a) Fission of <sup>251</sup>Es occurs at the average CN angular momentum, half being at the initial excitation energy, and half at the mean energy available after emission of one neutron. The calculation is not sensitive to small changes in this distribution. (b) Neutrons emitted after the saddle-point deformation is reached are defined as post-fission neutrons, and are emitted isotropically in the rest frame of the source. (c) From saddle to scission,  $U_x$  remains constant. (d) At scission, the excitation energy is divided in proportion to the mass of the fragments. (e) The calculation is made for zero fragment velocity at scission, since the actual value is unknown,<sup>8</sup> particularly at high  $U_x$ . The effect of a large velocity at scission is simulated by suppressing neutron emission up to  $0.5V_\infty$ . (f) The neutron lifetime ( $\tau_n$ ) is given by<sup>9</sup>

$$\tau_n = 2\pi\hbar\rho(U_x)(A')^2/0.189A^{2/3}\rho(U_x - B_n),$$

$$A' = d \ln \rho(U_x - B_n) / d(U_x - B_n),$$

where the level density  $\rho(U_x) \propto U_x^{-2} \exp[2(a_n \times U_x)^{1/2}]$ ;  $a_n$  has values between  $A/8$  and  $A/10$ . The value of  $B_n$  is taken to be the average of the liquid-drop binding energies of the first and second neutrons for <sup>251</sup>Es, and that of the second neutron for each fragment. (g) The fragment mass distribu-

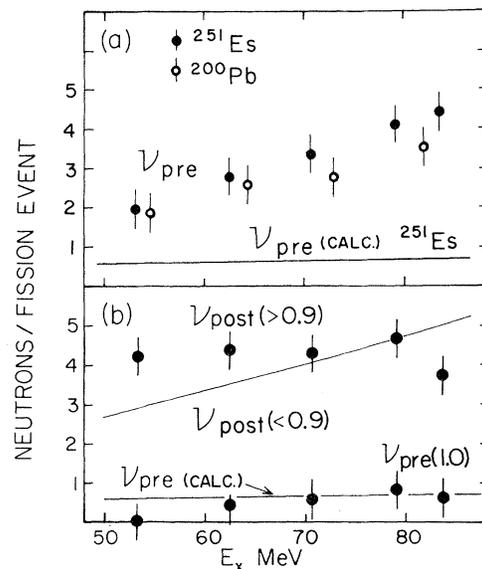


FIG. 2. Neutron multiplicities as a function of CN excitation energy for  $a_n = A/10$ . (a), (b) As in Fig. 1. Experimental values of  $\nu_{pre}$  for <sup>200</sup>Pb are also shown in (a). The predicted prefission neutron multiplicity for <sup>251</sup>Es is indicated by  $\nu_{pre}(CALC.)$ .

tion is simulated by two mass splits, the heavy primary fragments being  $^{131}\text{Sb}$  and  $^{142}\text{Cs}$ , weighted 60% and 40%, respectively. (h) The center-of-mass velocity distribution of neutrons emitted from the fragments is taken to be that of either the prefission or post-fission neutrons from the conventional analysis. The velocity distribution of the latter gives a slightly better fit at  $0^\circ$  [see Fig. 1(b)].

The probability of neutron emission was evaluated [see (f) above] for a saddle-to-scission time of  $4 \times 10^{-21}$  s, and subsequently at intervals of  $0.5 \times 10^{-21}$  s. At the end of each interval, the distribution in  $U_x$  was adjusted to account for the neutrons emitted; the  $0^\circ$  and  $90^\circ$  laboratory-frame velocity spectra of these neutrons were obtained with use of the average velocity vector of the emitting fragment, evaluated for that interval. The calculation was performed up to a velocity  $f_v V_\infty$ , and cumulative  $0^\circ$  and  $90^\circ$  spectra were obtained and subtracted from the experimental data. The residual spectra were then analyzed in the conventional manner, yielding the multiplicities of prefission neutrons [ $\nu_{\text{pre}}(f_v)$ ], and of post-fission neutrons emitted after  $f_v V_\infty$  was reached [ $\nu_{\text{post}}(> f_v)$ ].

The results of this calculation, with  $f_v = 0.9$ , are shown in Fig. 1(b) for the same data as in Fig. 1(a). The component labeled  $\nu_{\text{post}}(< 0.9)$  is the calculated spectrum of post-fission neutrons emitted in the time taken to achieve  $f_v = 0.9$ . For  $^{251}\text{Es}$ , only  $\sim 5\%$  of  $\nu_{\text{post}}(< 0.9)$  occurs during the transition from saddle to scission; thus the assumptions made concerning this period do not seriously affect the calculated multiplicities. However, this proportion varies strongly for different reactions, depending on the relative values of excitation energy and neutron binding energies in the CN and fission fragments.

The value of  $\nu_{\text{pre}}(1.0)$  (corresponding to extending the calculation to  $f_v = 1.0$  and thus to infinite time) was estimated by a linear extrapolation of  $\nu_{\text{pre}}(f_v)$  for  $f_v$  between 0.80 and 0.95. Typically,  $\nu_{\text{pre}}(1.0)$  was 0.4 neutron less than  $\nu_{\text{pre}}(0.9)$ . The emission of scission neutrons, produced during the snapping of the neck,<sup>10</sup> must be considered. Conventional analysis of  $^{252}\text{Cf}$  spontaneous-fission data gives  $\nu_{\text{pre}} \approx 0.4$ ,<sup>3</sup> which has been attributed to scission neutrons. However, from analysis<sup>11</sup> similar to that described here, it was concluded that there is no convincing experimental evidence for scission neutrons; our preliminary calculations for  $^{252}\text{Cf}$  support this conclusion. Thus we do not subtract a contribution from scission neutrons. Figure 2(b) shows the neutron multiplicities as a function of  $E_x$ , for  $a_n = A/10$ . The values of  $\nu_{\text{pre}}(1.0)$  are smaller than those conventionally determined, by as many

TABLE I. Neutron multiplicities per fission event from 124-MeV  $^{19}\text{F} + ^{232}\text{Th}$ . The prefission multiplicity derived from the conventional analysis is 3.3.

$a_n$	$\nu_{\text{pre}}(1.0)$	$\nu_{\text{pre}}(0.9)$	$\nu_{\text{post}}(< 0.9)$	$\nu_{\text{post}}(> 0.9)$
$A/8$	1.62	1.91	2.53	4.90
$A/9$	1.11	1.46	3.33	4.59
$A/10$	0.57	0.98	4.10	4.28
$A/10$ $v > 0.5 V_\infty$	1.27	1.67	3.54	4.14

as three neutrons. The sensitivity of the results to the parameter  $a_n$  is illustrated in Table I; also shown is the effect of suppressing neutron emission until  $f_v = 0.5$ . In view of the uncertainties involved in our calculations, the agreement between the various possibilities and the statistical-model predictions of  $\nu_{\text{pre}}$  is good, particularly when compared with the value obtained by conventional analysis. The best agreement is obtained for  $a_n = A/10$ , a value which is not unreasonable. The data for  $^{200}\text{Pb}$  were analyzed by the new method for  $a_n = A/10$ ;  $\nu_{\text{pre}}$  was reduced by only 0.7 even at the highest value of  $E_x$ . Thus in this case the correction is small as expected, but for reactions giving higher excitation energies, large corrections may have to be made.

We have measured fission-neutron angular correlations following fission of  $^{251}\text{Es}$ . Conventional analysis indicated large number of "prefission" neutrons, which are not expected on the basis of fission probability systematics or theory. Comparison with the less fissionable  $^{200}\text{Pb}$  at the same  $U_x(\text{CN})$  and similar angular momentum is very strong evidence, presented for the first time, that such analysis is inadequate. Analysis including neutron emission from the accelerating fragments can resolve the conflict. Other possible processes<sup>12</sup> are not excluded, but we emphasize that for systems with high  $E_x$ , or favorable  $Q$  values for fission, analysis must account for neutron evaporation during the transition from saddle to scission and during the fission fragment acceleration period. Incorrect conclusions may otherwise be drawn from the experimental data.

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