Generation of Fragment Angular Momentum in Fission

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A recent analysis of experimental data [J. Wilson *et al.*, Nature (London) **590**, 566 (2021)] found that the angular momenta of nuclear fission fragments are uncorrelated. Based on this finding, the authors concluded that the spins are therefore determined only *after* scission has occurred. We show here that the nucleon-exchange mechanism, as implemented in the well-established event-by-event fission model FREYA, while agitating collective rotational modes in which the two spins are highly correlated, nevertheless leads to fragment spins that are largely uncorrelated. This counterexample invalidates the conclusion in [J. Wilson *et al.*] that uncorrelated spins must necessarily have been generated after scission. Furthermore, it was reported [J. Wilson *et al.*] that the mass dependence of the average fragment spin has a sawtooth structure. We demonstrate that such a behavior naturally emerges when shell and deformation effects are included in the moments of inertia of the fragments at scission.

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Introduction.—A recent article by Wilson *et al.* [1] addressed the generation of angular momentum in nuclear fission. In particular, based on the analysis of unique and extensive experimental data taken at the ALTO facility of the IJC Laboratory in Orsay, the authors concluded that there is no significant correlation between the angular momenta (or "spins") of the two fission fragments. Because the authors assume that any spin generated prior to scission must result in strongly correlated fragment spins, they conclude that their observation implies that the fragment spins must be generated *after* the two fragments are no longer in contact.

Using the fission model FREYA [2], we demonstrate here that a fission treatment based on correlated rotational modes in the dinuclear complex *prior* to scission may in fact endow the fragments with spins that are nevertheless approximately independent. This fact invalidates the above assumption and hence casts doubt on the conclusion in Ref. [1] regarding the mechanism for the angular momentum generation.

Furthermore, the data analysis in Ref. [1] revealed that the average magnitude of the fragment spins has a sawtoothlike dependence on the mass number of the fragment. We show that when realistic moments of inertia are employed for the fledgling fragments at scission, the calculation naturally yields a sawtooth behavior of the mean spin. This in turn provides an explanation for the observation in Ref. [1] that the light fragment may carry more angular momentum than its heavy partner.

In the "Model" we recall the main relevant features of the theoretical treatment. Then, in the "Spin correlations," we

discuss the correlations between the fragment spin magnitudes as well as their directions. In the "Mass dependence," we address the mass-number dependence of the mean spin magnitude and we finally present a concluding discussion in the "Discussion."

Model.—The fission model FREYA [2,3] employs Monte Carlo techniques to select the mass, charge, and velocity of the primary fragments as well as to their subsequent decays by sequential neutron evaporation and photon radiation, thereby generating large samples of complete fission events. The model is in many respects similar to other fission simulation treatments, such as CGMF [4], FIFRELIN [5], and GEF [6], although these models differ in many details. For example, FREYA is the only one that conserves angular momentum throughout an event.

Of particular relevance is the selection of the fragment spins [7], which was recently discussed in some detail [8]. The procedure employed in FREYA is motivated by the Nucleon-Exchange Transport model [9,10] which considers the effect of multiple nucleon transfers between the two parts of a dinuclear complex. Because each nucleon carries baryon number (and possibly electric charge) as well as linear and angular momentum, the associated observables exhibit a diffusive evolution as a function of the number of transfers. This mechanism was found to be the dominant cause of dissipation in strongly damped nuclear reactions [11] and it is expected to also play an important role during the late stages of fission, as the system develops a binary character prior to scission [12].

A detailed study of the effect of nucleon exchange on the dynamical evolution of the fragment spins considered the

agitation of the six dinuclear rotational modes (the transverse modes wriggling and bending, which are doubly degenerate, and the coaxial modes twisting and tilting) [13,14]. In particular, expressions were derived for the associated mobility coefficients which determine the time scales. When the relaxation time for a particular mode is short compared with the collective evolution, the associated spin distribution quickly readjusts to the evolving geometry and so it remains in local equilibrium.

Invoking this idealized limit for all six dinuclear modes, Moretto and Schmitt [15] formulated a statistical equilibrium model for the fragment spins in fission and heavy-ion reactions. However, the mobility coefficients for the dinuclear rotational modes differ significantly in magnitude, causing the relaxation time for the wriggling mode to be very fast, whereas the tilting mode is agitated only quite slowly. As a rough way of taking this complexity into account, FREYA implements the effect of the nucleon-exhange mechanism by assuming full relaxation of the transverse modes (wriggling and bending), while the coaxial modes (twisting and tilting) are not agitated [7,16].

Generally, the fissioning complex has an overall angular momentum S_0 , of which the fragments receive their share, $\underline{S}_f = (\mathcal{I}_f / \mathcal{I}_{tot}) S_0$, where \mathcal{I}_f is the moment of inertia of the fragment at scission and $\mathcal{I}_{tot} = \mathcal{I}_L + \mathcal{I}_H + \mathcal{I}_R$ is the total moment of inertia, with \mathcal{I}_R being the moment of inertia for the relative fragment motion. These rigid-rotation contributions tend to be negligible relative to the contributions from the statistical fluctuations.

The sampling of those is complicated by the fact that the individual fragment spins are not independent, due to the coupling imposed by conservation. There are three angular momentum vectors at scission: the spins of the two dinuclear partners, S_L and S_H , and their relative angular momentum, L. Because the system is isolated, their sum is conserved, $S_L + S_H + L = S_0$, leaving then six rotational degrees of freedom. These are conveniently represented by the six dinuclear normal modes in terms of which the rotational part of the collective Hamiltonian is diagonal.

The modes considered (all six modes in the statistical treatment [15] and only the four transverse modes in FREYA) are then populated statistically. Thus the amplitude s_m of a given mode m is sampled from $P_m(s_m) \sim \exp(-s_m^2/2\mathcal{I}_m T_S)$, where \mathcal{I}_m is the moment of inertia for that mode [7,15]. The wriggling moment of inertia is $\mathcal{I}_+ = (\mathcal{I}_L + \mathcal{I}_H)(\mathcal{I}_L + \mathcal{I}_H + \mathcal{I}_R)/\mathcal{I}_R$, while that for bending is $\mathcal{I}_- = \mathcal{I}_L \mathcal{I}_H/(\mathcal{I}_L + \mathcal{I}_H)$. (The temperature T_S employed by FREYA takes account of the fact that the distortion of the fragments at scission reduces the available statistical energy somewhat [7,16].) Once the normal modes have been agitated, yielding s_+ for wriggling and s_- for bending, the individual fragment spins then readily follow [7], resulting in

$$\boldsymbol{S}_{f} = \underline{\boldsymbol{S}}_{f} + (\boldsymbol{\mathcal{I}}_{f}/\boldsymbol{\mathcal{I}}_{\text{tot}})\boldsymbol{s}_{+} \pm \boldsymbol{s}_{-}, \qquad (1)$$

and their variances are $\sigma_f^2 = 2(1 - \mathcal{I}_f / \mathcal{I}_{tot})\mathcal{I}_f T_s$.

Spin correlations.—As discussed above, the two fragment spins are not independent, due to angular momentum conservation. Indeed, the spin contributions to the two fragments from wriggling are perfectly parallel, while the contributions from bending are antiparallel. When these dinuclear modes are populated statistically, the resulting correlation coefficient for the individual fragment spins amounts to

$$c(\mathbf{S}_L, \mathbf{S}_H) \equiv [\langle \mathbf{S}_L \cdot \mathbf{S}_H \rangle - \langle \mathbf{S}_L \rangle \langle \mathbf{S}_H \rangle] / [\sigma_L \sigma_H]$$

= $- \{ \mathcal{I}_L \mathcal{I}_H / [(\mathcal{I}_R + \mathcal{I}_L) (\mathcal{I}_R + \mathcal{I}_H] \}^{1/2}.$ (2)

This quantity is generally rather small because the moment of inertia for the relative fragment motion, \mathcal{I}_R , is typically an order of magnitude larger than those of the individual fragments, \mathcal{I}_L and \mathcal{I}_H , $\mathcal{I}_R \gg \mathcal{I}_f$. Thus, even though the fragment spins are strongly coupled for each of the normal modes, the resulting spins are expected to be relatively independent. This expectation is indeed borne out by actual FREYA simulations [8], as illustrated in Table I and Fig. 1.

The magnitudes of the fragment spins, $S_f = |S_f|$, have rather wide distributions with average values $\bar{S}_f \equiv \langle S_f \rangle \approx 5-6\hbar$. The associated covariance is given by $\sigma(S_L, S_H) = \langle S_L S_H \rangle - \bar{S}_L \bar{S}_H$, so the spin magnitude correlation coefficient is $c(S_L, S_H) = \sigma(S_L, S_H)/[\sigma(S_L)\sigma(S_H)]$. Table I lists the average spin magnitudes and the correlation coefficient for four fission cases of frequent interest. The correlation coefficients are essentially zero, indicating that the primary spin magnitudes are largely uncorrelated, in accordance with the reported experimental finding [1].

While the experimental data [1] cannot yield information on the fragment spin directions, it is noteworthy that those

TABLE I. The average magnitudes of the primary fission fragment spins, \bar{S}_L and \bar{S}_H , and the associated correlation coefficients $c(\bar{S}_L, \bar{S}_H)$ for four fission cases: ²³⁵U(*n*, f), ²³⁸U(*n*, f), ²³⁹Pu(*n*, f), and ²⁵²Cf(sf), as obtained with FREYA using either monotonically increasing moments of inertia or moments of inertia with a refined A dependence (numbers in parantheses). Also shown are the amplitudes f_1 in the distribution of the spin opening angles, $P(\phi_{LH}) = 1 + f_1 \cos \phi_{LH}$.

Case:	235 U(<i>n</i> , f)	238 U(<i>n</i> , f)	239 Pu (n, f)	²⁵² Cf(sf)
$\bar{S}_L = \langle S_L \rangle$	4.27	4.43	4,58	5.08
	(6.08)	(6.59)	(6.86)	(7.48)
$ar{S}_H = \langle S_H angle$	5.66	5.80	5.93	6.33
	(5.31)	(5.49)	(5.60)	(6.80)
$c(S_L, S_H)$ (%)	0.2	0.2	0.1	0.1
	(-10.9)	(-10.8)	(-10.3)	(0.8)
f_1 (%)	-8.2	-8.3	-8.3	-8.4
	(-10.9)	(-11.3)	(-11.7)	(-13.5)



FIG. 1. The distribution of the opening angle ϕ_{LH} between the angular momenta of the two fission fragments from 238 U(n, f), as obtained by FREYA using either the standard moment of inertia, $\mathcal{I}_f(A_f) = 0.5\mathcal{I}_{rig}(A_f)$ (dots) or the modified form depending on the shapes of the fragments at scission, $\mathcal{I}'_f(A_f)$ (open circles). Also shown are the corresponding lowest-order Fourier approximations, $P(\phi_{LH}) = 1 + f_1 \cos \phi_{LH}$.

are also largely uncorrelated in FREYA. The degree of correlation between the fragment spin directions is brought out by the distribution of the opening angle between the two fragment spins, ϕ_{LH} , which is given by $\cos \phi_{LH} = S_L \cdot S_H / [S_L S_H]$. This function is shown in Fig. 1 for ²³⁸U(*n*, f). (The other cases look very similar.) As was shown recently [8], the undulation of the fragment spin opening angle is generally well represented by the first harmonic, $P(\phi_{LH}) \approx 1 + f_1 \cos \phi_{LH}$. Table I shows the amplitudes f_1 for the four cases considered. As seen, they are all rather small, being of the order of 10%, showing that the spin directions are also largely uncorrelated.

Mass dependence.—In addition to the qualitative finding that the fragment spins are largely uncorrelated, Ref. [1] presented data for the dependence of the average spin magnitude \bar{S}_f on the fragment mass number A_f , revealing a sawtoothlike structure of $\bar{S}_f(A_f)$ (see Fig. 2). We now show that such a behavior emerges naturally due to the variation of the moments of inertia of the fragments at scission.

When the dinuclear rotational modes are sampled from thermal distributions, as in FREYA, then $\langle S_f^2 \rangle \approx 2 \mathcal{I}_f T_S$ (see above), so the mean fragment spin scales approximately as the square root of the moment of inertia, \mathcal{I}_f . To illustrate the close connection between \bar{S}_f and \mathcal{I}_f , we first note that the standard version of FREYA [3] assumes that the moment of inertia of a fragment is given by $\mathcal{I}_f(A_f) = c_{\rm rot} \mathcal{I}_{\rm rig}(A_f)$, with the global reduction factor being $c_{\rm rot} = 0.5$. Thus the A_f dependence of the moment of inertia is monotonic and the calculated $\bar{S}_f(A_f)$ is rather featureless. This is illustrated in Fig. 2 where $\bar{S}_f(A_f)$ obtained with the standard version of FREYA is shown for ²³⁸U(*n*, f) and



FIG. 2. The average magnitude of the spin of a fragment as a function of its mass number, $\overline{S_f}(A_f)$, for ²³⁸U(*n*, f) and ²⁵²Cf(sf). The solid squares are the data [1]. The FREYA calculations using the standard moments of inertia, $\mathcal{I}_f(A_f) =$ $0.5\mathcal{I}_{rig}(A_f)$ are shown by the open circles, with the green squares showing the values after the completion of the neutron evaporation. In addition to the spin values (dots) calculated with the modified moments of inertia, $\mathcal{I}'_f(A_f)$, the dashes show the (rescaled) function $[\mathcal{I}'_f(A_f)]^{1/2}$.

 252 Cf(sf) together with the data reported in Ref. [1]. (The figure also shows that the effect of neutron evaporation on the spin magnitudes is negligible, thus lending support to the assumption made in the data analysis [1].)

However, as pointed out in Ref. [1], fragments near the doubly magic nucleus ¹³²Sn have rather rigid spherical shapes and abnormally low level densities, as a consequence of which they cannot accommodate very much angular momentum. Indeed, in order to match the measured \bar{S}_f values near ¹³²Sn, it is necessary to reduce the rigid moment of inertia by 0.2. On the other hand, away from this special region the fragments have deformed equilibrium shapes and are likely significantly further distorted at scission. Thus they have considerably larger moments of inertia and can acquire correspondingly larger angular momenta. (These expectations were indeed borne out by recent microscopic studies of scission configurations [17–19].) Such a mass dependence of the moments of

inertia will then be reflected in the spin magnitudes of the nascent fragments.

To illustrate this relationship, we have made calculations with a schematic moment of inertia, $\mathcal{I}'_f(A_f)$, that roughly incorporates those features (particularly the reduction near A = 132). [$\mathcal{I}'_f(A_f)$ was obtained by assuming that the nascent fragments have spheroidal shapes with deformations $\varepsilon_{\rm sc}(A_f)$ that are twice the equilibrium values, $\tilde{\varepsilon}(A_f)$, which in turn were obtained by making a simple fit to the ground-state values tabulated in Ref. [20]; the moments of inertia were then taken as $\mathcal{I}'_f(A_f) = 0.2\mathcal{I}_{rig}(A_f; 0) +$ $2[\mathcal{I}_{rig}(A_f; \epsilon_{sc}(A_f)) - \mathcal{I}_{rig}(A_f; 0)],$ where the factor of 2 was adjusted to ensure that the resulting spins are in rough overall agreement with the measured values.] The precise procedure for obtaining the schematic function $\mathcal{I}'_{f}(A_{f})$ is not important because the purpose is merely to demonstrate that the modulation of $\mathcal{I}_f(A_f)$, (i.e., the decrease around the doubly magic nucleus ¹³²Sn and the increase well away from there due to the large deformations of the nascent fragments) leads to a corresponding modulation of $\bar{S}_f(A_f)$ resembling the sawtooth feature of the data.

The resulting spin magnitudes are shown in Fig. 2. To bring out the close connection between the moment of inertia and the mean spin magnitude, the figure also shows $[\mathcal{I}'_f(A_f)]^{1/2}$ (rescaled for display purposes). The remarkably similar mass dependence of those two quantities suggests that measurements of $\bar{S}_f(A_f)$, such as those reported in Ref. [1], in effect provide information on the geometry of the scission configurations.

It should be noted that the fragment spins remain approximately uncorrelated after the introduction of the modulated moments of inertia $\mathcal{I}'_f(A_f)$, as illustrated in Table I and Fig. 1.

The average spin magnitudes obtained with the schematic moments of inertia (see Table I) are in reasonably good agreement with the measured values for the heavy fragments shown in Fig. 4 of Ref. [1], as is also apparent from Fig. 2. However, as Fig. 2 also shows, the schematic calculations overshoot the measurements for $A_L > 100$ for U and for 98 < $A_L < 110$ for Cf. We emphasize again that those calculations merely serve to illustrate the effect of the modulation of the moments of inertia on the spin magnitudes and should not be regarded as an attempt to reproduce the data presented in Ref. [1].

Finally it should be pointed out that, for moderate mass asymmetries the modulated moments of inertia tend to favor the light fragments (because of their deformations), so these may now have larger mean spins than their heavy partners, as appears to be the case experimentally [1]. (This is also consistent with recent microscopic studies [18,19].) [Specifically, the calculations with $\mathcal{I}'_f(A_f)$ yield $\bar{S}_L > \bar{S}_H$ for $A_L \ge 98$ for ²³⁸U(*n*, f) and for $A_L \ge 106$ for ²⁵²Cf(sf), while the data (which were obtained for a more limited mass range) suggest $\bar{S}_L > \bar{S}_H$ for $A_L \ge 100$ and $A_L \ge 108$, respectively.]

Discussion.—The mean-field character of low-energy nuclear collective motion implies that the associated dissipation is of one-body form [12]. For a single nucleus, the one-body dissipation results from particle-hole excitations generated by the evolving mean field. For binary systems, such as those encountered in nuclear reactions and fission, an additional one-body mechanism is the multiple transfer of individual nucleons between the two parts, a process that leads to a diffusive evolution of the mass and charge partition in the dinucleus as well as the linear and angular momenta of the two partners. Because the active nucleons reside in the Fermi surface they carry relatively large momenta and, as a consequence, the resulting dissipation is strong [12,21].

This general character of nuclear dynamics causes the shape evolution in fission to be rather slow, a feature FREYA seeks to incorporate by assuming that the transverse dinuclear rotational modes maintain full equilibrium up to scission, while the coaxial modes are insignificantly agitated. It is important to note that even though the contributions from each dinuclear normal mode to the individual fragment spins are strongly aligned the resulting fragment spins are largely uncorrelated. This is in accordance with the experimental finding in Ref. [1].

This finding demonstrates that the absence of spin correlation *cannot* be taken as evidence for the fragments having acquired their spins independently, contrary to what was proposed in Ref. [1]. (In fact, the independent population of rotational states in the fragments *after* their separation seems hard to reconcile with the principle of angular momentum conservation for isolated systems.)

We have furthermore demonstrated that the sawtoothlike mass dependence of the average fragment spin can be understood as a reflection of the moments of inertia of the fragments at the time of their formation. Such a behavior would arise also in other simulation codes (such as Refs. [4–6]) if similar moments of inertia were employed. (It should also be noted that $\bar{S}_f(A_f)$ would still display a sawtooth form even in the extreme scenario where only the wriggling modes were populated, causing the two fragment spins to be perfectly parallel in each event.) The finding, based on the observed sawtooth behavior of $\bar{S}_f(A_f)$, that the light fragment may carry more angular momentum than its heavy partner, was anticipated recently on the basis of microscopic studies [18,19].

Finally we have pointed out that the strong connection between the moments of inertia of the nascent fragments and their mean spin magnitudes suggests that measurements of $\bar{S}_f(A_f)$, such as those reported in Ref. [1], may provide unique experimental information on the fissioning system at the time of scission, which in turn would be very helpful for the further development of fission theory. This work was supported by the Office of Nuclear Physics in the U.S. Department of Energy under Contracts DE-AC02-05CH11231 (J. R.) and DE-AC52-07NA27344 (R. V.) and was supported by the DOE Office of Nuclear Nonproliferation and the LLNL LDRD Program under Project No. 20-ERD-031 (R. V.).

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