Letter

Spatial orientation of the fission fragment intrinsic spins and their correlations

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New experimental and theoretical results obtained in 2021 made it acutely clear that more than 80 years after the discovery of nuclear fission we do not understand the generation and dynamics of fission fragment (FF) intrinsic spins well, in particular their magnitudes, their spatial orientation, and their correlations. The magnitude and orientation of the primary FFs have a crucial role in defining the angular distribution and correlation between the emitted prompt neutrons, and subsequent emission of statistical (predominantly $E1$) and stretched $E2$ γ rays, and their correlations with the final fission fragments. Here, we present detailed microscopic evaluations of the FF intrinsic spins, for both even- and odd-mass FFs, and of their spatial correlations. These point to a well-defined three-dimensional FF intrinsic spin dynamics, characteristics absent in semiphenomenological studies, due to the presence of the twisting spin modes, which artificially were suppressed in semiphenomenological studies.

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The year 2021 started with the publication of a new and very accurate experimental measurement of the fission fragments (FFs) intrinsic spins [1], significantly extending the results of, almost 50-year-old, similar experiments [2,3]. At the same time, an independent flurry of theoretical activity, based on phenomenological and microscopic models, were directed at studying various properties of the FF intrinsic spins, which led to new insights into the mechanics of FF angular momenta formation and their correlations [4,5]. Other theoretical studies followed [6–14], and a very intense hybrid workshop was held to discuss the topic, attended by both theorists and experimentalists from all around the world, where many new and old ideas were actively dissected [15]. As Sobotka has recently discussed, in a talk at the Nuclear Chemistry Gordon conference in June 2023, we are now at a very unusual juncture in time, when it is high time to address experimentally, *Fragment spin generation in fission: What we know, cannot know, and should know* [16].

The case of spontaneous fission of ^{252}Cf is perhaps the simplest and cleanest nucleus to consider in order to appreciate the complexity of what we need to better understand the FF intrinsic spins, both theoretically and experimentally. In its ground state, ^{252}Cf has the spin and parity $S_\pi = 0^+$, and is a cold isolated quantum system. After the FFs separate, both are highly excited, with the heavy FF (HFF) typically being cooler than the light FF (LFF) [17–19]. At the same time, the average intrinsic spin of the HFF is smaller compared to the LFF, as recently demonstrated in a first fully microscopic study on the FF intrinsic spin distributions [5]. This was opposite to the prior consensus in literature, namely that the HFF has a larger average intrinsic spin than the LFF [2,4,20–23], to cite a few representative studies. This surprising result turned everything around making it clear that too much was taken for granted in modeling fission dynamics and the decay properties of prompt FFs, which require a more detailed analysis. Subsequent theoretical and phenomenological studies incorporated this new aspect [6,7]. Recently, the relative angular momenta of the FFs was also investigated microscopically [9]. Conservation of the total angular momentum then requires that

$$\hat{\boldsymbol{S}}_H + \hat{\boldsymbol{S}}_L + \hat{\boldsymbol{\Lambda}} = \boldsymbol{S}_0 \approx \boldsymbol{0}, \tag{1}$$

where $\hat{\boldsymbol{\Lambda}} = \hat{\boldsymbol{R}} \times \hat{\boldsymbol{P}}$ is the relative orbital angular momentum perpendicular to the fission axis, $\hat{\boldsymbol{R}}, \hat{\boldsymbol{P}}$ are the the relative separation between the FFs and their relative linear momentum respectively, $\boldsymbol{S}_0$ is the compound's spin, and $\Lambda_z = 0$. The above approximation is exact for ^{252}Cf, and reasonable for the induced fission with low-energy incident neutrons on ^{235}U, ^{239}Pu targets. Now, a very important question arises: are the FF intrinsic spins $\boldsymbol{S}_{H,L}$ also perpendicular to the fission axis? Clearly their sum $\hat{\boldsymbol{S}}_H + \hat{\boldsymbol{S}}_L$ is, in the case of spontaneous fission of ^{252}Cf. This particular aspect is not yet resolved experimentally or theoretically [11], and is related to the strong disagreements between TDDFT predictions [9,10] and the phenomenological predictions of the FREYA model [4,7,11,24]. The event-by-event orientation of the FF intrinsic spins has important consequences, as it will affect the direction of emission for prompt neutrons and is one of the most pressing questions experiment should now address [16]. In the present microscopic study, we will specifically address this aspect and make a clear statement about where the most advanced microscopic theory stands today, with a result starkly different from what the phenomenological model FREYA [4,7,11] predicts, which is the only other source of clear information available currently in literature.

The first indication that the angular distribution between the FF intrinsic spins is likely very different from previous models was reported in Refs. [9,10]. This result was at odds with phenomenology implemented in FREYA [4,7,11], where the angular distribution was almost uniform, while the microscopic results showed a clear nonuniformity. One major assumption adopted in FREYA, and which we demonstrate in the present study to be theoretically wrong, is that the FF intrinsic spins are perpendicular to the fission direction and as a result the twisting and tilting modes [25–27] are artificially excluded from the fission dynamics. At the time, when the theoretical study [9] was performed, an angular momentum projection on several angles was out of question. Today, two new technical developments have made such a full study possible, see Supplemental Material [28], with additional references [29,30], and presently one can easily evaluate the triple distribution $P(\Lambda, S_H, S_L)$ exactly, without any additional assumptions or approximations.

The FF angular momentum projection is performed using well-known and established projection techniques [31–35], illustrated here for a specific FF,

$$\hat{P}^S_{MK} = \frac{(2S+1)}{16\pi^2}\int d\Omega \mathcal{D}^{S*}_{MK}(\Omega)\, e^{i\alpha\hat{S}_z}e^{i\beta\hat{S}_y}e^{i\gamma\hat{S}_z}, \quad (2)$$

$$P(S_F, K_F) = \langle\Psi|\hat{P}^{S_F}_{K_F K_F}|\Psi\rangle \quad (3)$$

with $\Omega = (\alpha, \beta, \gamma)$ representing a separate set of the three Euler angles for each FF, $|\Phi\rangle$ representing the many-body wave function, and $P(S_F, K_F)$ the probability distribution for either light or heavy intrinsic spin $S_F = S_L, S_H$ with projection K_F on the fission direction. The angular momenta $\hat{S}_{x,y,z}$ are defined in a spatial region around a specific FF in its center-of-mass frame [5]. M and K are the projections of the angular momentum S in either the laboratory or body frame. Our goals are to evaluate $P(S_F, K_F)$ is probability distribution for K_F for each FF, and the triple angular momentum distribution

$$P(\Lambda, S_H, S_L) = \sum_{k_H k_L}\langle\Psi|\hat{P}^{\Lambda}_{0,0}\hat{P}^{S_H}_{K_H K_H}\hat{P}^{S_L}_{K_L K_L}|\Psi\rangle. \quad (4)$$

The triple distribution can be shown to be given exactly by the expression

$$P(\Lambda, S_H, S_L) = \sum_{K_H K_L K'_H K'_L}(-1)^{K'_H - K_H + K'_L - K_L} \times C^{\Lambda,0}_{S_H,-K_H,S_L,-K_L}C^{\Lambda,0}_{S_H,-K'_H,S_L,-K'_L} \langle\Psi|\hat{P}^{S_H}_{K_H K'_H}\hat{P}^{S_L}_{K_L K'_L}|\Psi\rangle \quad (5)$$

with $C^{J,M}_{j_1,m_1,j_2,m_2}$ the well-known Clebsch-Gordan coefficients, see Supplemental Material [28]. This formula shares some common elements with a formula suggested by Døssing during the *Workshop of Fission Fragment Angular Momenta* [15] and also discussed in Refs. [10,13,14]. The presence of the Clebsch-Gordan coefficients, which emerge naturally, ensure that the triangle constraint Eq. (1) is automatically satisfied. The numerical evaluations were performed using the LISE package [36] to evolve the time-dependent density functional theory equations extended to superfluid systems and determine the many-body wave function $|\Phi\rangle$ used in Eq. (4). In addition, at the end of the simulation, we performed a unitary transformation to the canonical quasiparticle states [37], as they provide the most economic representation of a many-body wave function. For the evaluations of the overlaps in Eq. (5), which involves computing Pfaffians [33,34], we used the algorithm described in Ref. [38].

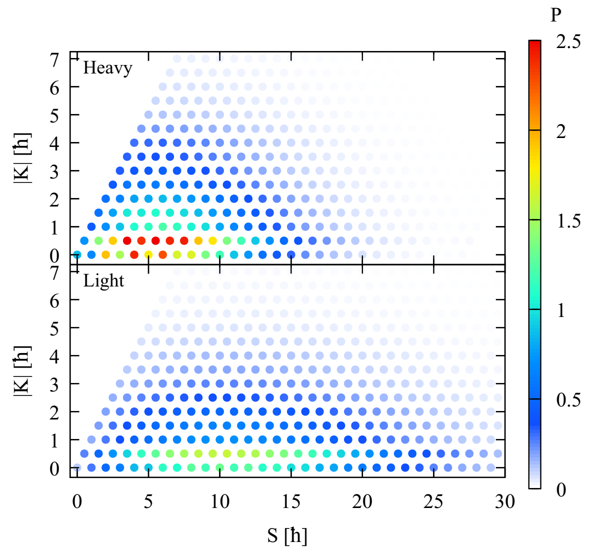

FIG. 1. Spin distribution in the heavy (upper panel) and light (lower panel) fragment obtained with Eq. (3). This calculation is done for a ^{236}U with the SeaLL1 functional. The states with the same integer K values (unlike the half-integer K values) for different $S_{H,L}$ are joined by a thin horizontal line for an easier visual identification. Since for both FFs $P(S, K) = P(S, -K)$ we show the distribution of $|K|$.

For each FF we define the angle between the $\boldsymbol{S}_{H,L}$ and the fission axis, as well as the angle between the two FF intrinsic spins [5]

$$\cos\theta_F = \frac{K_F}{\sqrt{S_F(S_F+1)}}, \text{ where } F = H, L, \quad (6)$$

$$\varphi_{HL} = \arccos\left(\frac{\Lambda(\Lambda+1) - S_H(S_H+1) - S_L(S_L+1)}{2\sqrt{S_H(S_H+1)S_L(S_L+1)}}\right). \quad (7)$$

Such angles can be defined if $S_{H,L} \neq 0$. With the triple distribution $P(\Lambda, S_H, S_L)$ one can straightforwardly evaluate the distributions $P(\theta_F)$ and $P(\varphi_{HL})$, which we will discuss now.

The distributions for the projections of each FF spin on the fission axis are shown in Figs. 1 and 2. Here, one sees the first new aspects of each FF K-spin distribution and also, as one might have expected, the presence of both integer and half-integer spins. If K is an integer, the corresponding FF is either an even-even (both Z_F and N_F are even) or an odd-odd nucleus (both Z_F and N_F are odd). In the case of half-integer FF spins and K values the corresponding A_F is odd. Additionally, the range of LFF intrinsic spins S_L is wider than the range of the HFF intrinsic spins S_H, in agreement with the results reported in Refs. [5,6]. Another noticeable aspect is that the probabilities to find even and odd mass FFs are almost equal. This is consistent with little or no odd-even staggering observed in experimental mass yields. Note that preneutron emission mass yields are corrected for neutron emission, correction that is subject to model dependence.

The most remarkable aspect of the FF spin distributions shown in Fig. 1 are illustrated in Fig. 2, where the probability

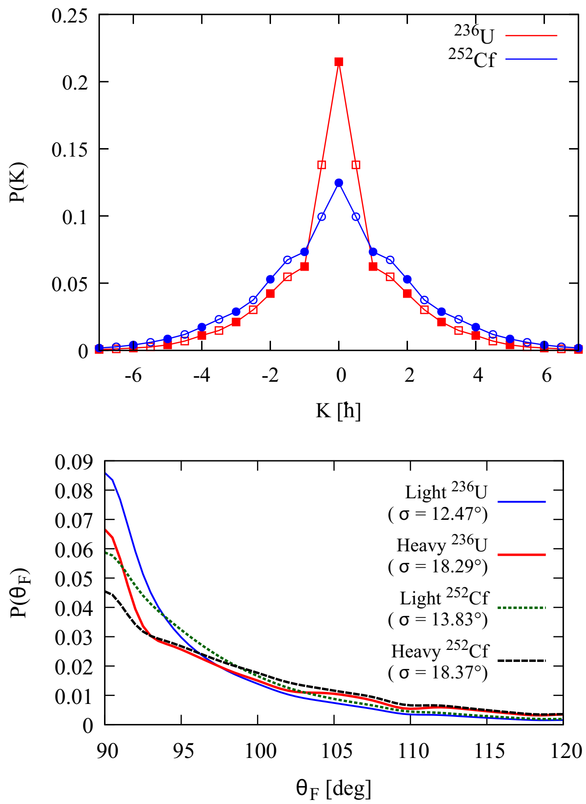

FIG. 2. Top: The distribution of the K quantum number. Since $K_H + K_L = 0$ the distributions for heavy and light FFs are identical. The integer K values are shown with filled symbols and with empty symbols for half-integer K values. The bottom panel shows the distribution $P(\theta_F)$ computed using Eq. (6), convoluted with a Gaussian of width 2° and shown here only for angles $\theta_F \geqslant 90°$, since $P(\theta_F)$ is symmetric with respect $\theta_F = 90°$.

distributions

$$P(K) = P_H(K) = P_L(K) = \sum_{S_{L,H}} P(S_{L,H}, K_{L,H}) \qquad (8)$$

show the presence of nonvanishing values of the projection of each FF intrinsic spin on the fission direction, an incontrovertible confirmation of that fact that the twisting spin modes are active. This feature is at stark odds with the almost 15 year old assumption made by the developers of FREYA [4,7,11,24] that the FF intrinsic spins are perpendicular to the fission axis and that the tilting and twisting modes of FF intrinsic spins are frozen and not active in the fission dynamics. Their justification is based on an argumentation used in the treatment of nucleon transfer in nuclear collisions [39,40]. This assumption played a key role in the claimed agreement [7] with the recent experimental results obtained by [1]. Since in FREYA the FF intrinsic spins are treated classically there is no distinction between integer and half-integer FF intrinsic spins and no statement can be made about whether even-odd staggering effects are present in their predictions.

For both fissioning nuclei ^{252}Cf and ^{236}U one observes a very large peak corresponding to $|K| \leqslant 1/2$, followed by

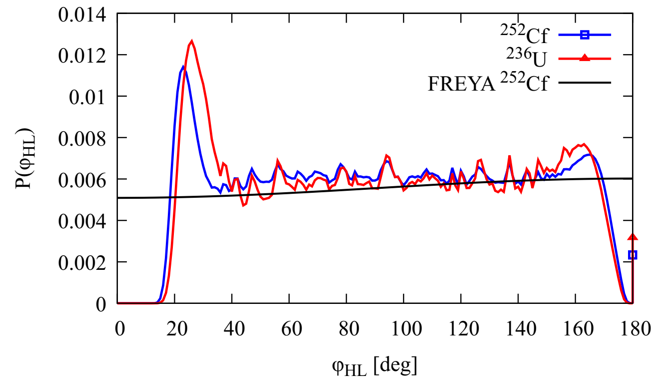

FIG. 3. The distribution $P(\varphi_{H,L})$, where φ_{HL} is the opening angle between the FF intrinsic spins, using Eqs. (7) and (5). This distribution was obtained after averaging with a 2° wide Gaussian function. The FF spin configuration at 180° is shown as a separate point, as when seen with higher resolution this configuration is clearly separated from the configuration at $\varphi_{HL} < 180°$, but not as distinctly as the configurations close to $\varphi_{HL} = 0°$.

quite long tails. In the cases of ^{252}Cf and ^{236}U, the sum $P(-1/2) + P(0) + P(1/2) \approx 0.33$ and $\approx$0.49, respectively. Correspondingly, this implies that the probability to find a FF with $|K| \geqslant 1$ is 0.67 for ^{252}Cf and 0.51 for ^{236}U. This is particularly important, as it points to the fact that the FF intrinsic spins are, with very large probability, not perpendicular to the fission axis. Instead, they are most likely to be found opposite to each other with respect to the fission axis (as $K_H + K_L = 0$). As a result, the plane defined by the triangle formed via the three angular momenta $\hat{\mathbf{S}}_H + \hat{\mathbf{S}}_L + \hat{\mathbf{\Lambda}} = \mathbf{0}$ forms an angle θ_F significantly different than 90° with the fission axis for very large fraction ($\geqslant$1/2) of fission events. The lower panel of Fig. 2 reinforces this conclusion. From the results reported in Ref. [5], specifically the expectation value of $K_F^2 \approx 1.6, \ldots, 4.4$ one obtains very similar values for θ_F.

The wide range of active $K_{H,L}$ values is particularly important, as they control the so-called FF twisting degrees of freedom, whose role was ignored in the FREYA model. In this respect one should also notice the role played by Coulomb reorientation effects of the separated FFs [13,14], which can lead to the increase of the FF intrinsic spins by 1–2 $\hbar$. Additionally, it contributes to the wriggling motion of the FFs, which otherwise is absent, since $K_H + K_L = 0$ before scission in case of ^{252}Cf(sf), unless the role of quantum fluctuations is taken into account explicitly [19,41].

In Fig. 3 we show the distribution of the opening angle between the FF intrinsic spins. This distribution, as reported in Refs. [9,10], was completely at odds with the results arising from FREYA simulations [4,7,11], and generated a lot of excitement and discussions at the fission workshop [15]. As mentioned above, at the time, due to technical difficulties in Refs. [9,10] we were not able to perform a full momentum projection and had to rely on the two-angle formulas. In the present study, this difficulty has been overcome and the exact triple angular momentum distribution is shown in Fig. 3. A comparison between the triple angular distributions $P(\Lambda, S_h, S_L)$ obtained in Refs. [9,10] and the triple distribution evaluated in this study is presented in the Supplemental Material [28]. In the present microscopic treatment of the FF intrinsic spins, the distribution $P(\varphi_{HL})$ shows an almost uniform distribution in the interval $\varphi \in (40^\circ, 160^\circ)$ with pronounced decays close to angles 0° and 180°. The probability that the angle φ_{HL} has values larger than 90° is about 0.53, thus pointing to a slight preference for the bending over wriggling modes.

As intuited by Døssing during the workshop [15], there was a need for a Clebsch-Gordan coefficient to enforce the triangle constraint, similar, but not identical to the combination of Clebsch-Gordan coefficients in Eq. (5). The final results, shown in the upper panel of Fig. 3, are closer to the FREYA almost uniform predictions [4,7,11,24] for angles $(50^\circ, 150^\circ)$. However, unlike FREYA predictions, the probabilities vanish at $\varphi_{HL} = 0^\circ$ and 180°, and obtain a prominent peak slightly above $\varphi_{HL} \approx 20^\circ$, and a smaller peak at $\approx 165^\circ$. These two peaks originate from the Clebsch-Gordan coefficients appearing Eq. (5) favoring angles φ_{HL} close to 0° and to a lesser extent 180°.

The emerging lesson from our microscopic calculations is that the FF intrinsic spins dynamics is indeed of a three-dimensional (3D) character and basically all FF intrinsic spin modes conjectured to be active almost six decades ago [25–27] are indeed present, see Figs. 1 and 2. These signals are clearest on the case of ^{252}Cf(sf), in which case the angle formed by the plane defined by the FF intrinsic spins with the fission axis or the distribution $P(K)$ has very wide fluctuations. The FF intrinsic spin dynamics is not restricted to the plane perpendicular to the fission axis, as in the classical treatment of the FF intrinsic spins of Randrup and Vogt [24], Vogt and Randrup [4], Randrup and Vogt [7], Randrup [11]. The FF collective twisting modes are clearly present, in agreement with the earlier conclusions in Ref. [5]. The FF spin dynamics is also not fully unrestricted in 3D, as initially assumed in Refs. [9,10]. The distribution of the angle between the FF intrinsic spins reported initially in Ref. [9], with the details clarified and reported here in Figs. 1–3, are what is expected to either emerge or to be refuted in future envisioned experiments [12,16]. The fact that the FF intrinsic spins are not perpendicular to the fission axis before the emission of prompt neutrons and γ rays, would likely lead to measurable effects [12,16].

Often, either in discussions or in literature [16], one can find the statements that the pair-breaking mechanism can lead to 3D dynamics of the FF intrinsic spins. This aspect requires some clarifications. In microscopic studies [9,17–19,41–43] pairing is treated explicitly and during the systems' evolution through the saddle-to-scission descent, as well as in studies where the excitation energy of the initial compound nucleus was increased, the *nn* and *pp* short-range correlations (SRCs) never vanished, even though the excitation energy of the nuclear system corresponds to a high temperature, where a pairing condensate does not exist. Instead, only the phase of the pairing condensate is lost, true also in collisions of heavy ions at rather large collision energies [37,42–44]. SRCs between either proton or neutron pairs survive to rather large excitation energies, an aspect that should not be conflated with pair breaking. Loss of long-range order, manifested as the loss of phase coherence of the pairing condensate, can be accompanied by the formation of new nucleon pairs with nonzero total spin. Nevertheless, the SRCs obviously survive in $L = 0$.

The semiphenomenological FREYA model [4,7,11,24], which is based on a number of fitting parameters and assumptions, is the only model which so far leads to predictions, which might be tested in experiments. FREYA and the microscopic treatment of fission dynamics lead to starkly different predictions. This difference will be addressed by future experiments, which will be hopefully interpreted in fully assumptions and parameter-free theoretical treatments. The microscopic framework adopted in this study is based on a nuclear energy density functional, which depends on only eight parameters: saturation density and energy of symmetric nuclear matter, spin-orbit and pairing couplings, proton charge, nuclear surface tension (related to the nucleon-nucleon interaction range), symmetry energy, and to a less extent its density dependence [45,46], whose values are well known for decades.

As Randrup [11] stressed: *In view of the large differences between the model calculations of the spin-spin opening angle distribution, experimental information on this observable is highly desirable as it could help to clarify the scission physics.* The assumption in FREYA the FF intrinsic (classical) spins are exactly perpendicular to the fission direction and that the twisting mode are inactive, is the origin of these large differences. As Sobotka [16] discussed, see slide 28, the angular distribution of FF stretched $\gamma(E2)$ with respect to the fission direction is the "smoking gun," which will discriminate between these two approaches and new experiments are planned to resolve discrepancies between old results [2,3].

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