New procedure to determine the mass-angle correlation of quasifission

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An innovative procedure to determine the mass-angle distribution of fragments from quasifission is proposed. It is based on the subtraction of the contribution from compound-nucleus fission from the experimental correlation spectrum which is in most cases a mixture of fusion-fission and quasifission. The former is calculated with a four-dimensional dynamical model of fission within the stochastic Langevin theory. The proposed approach is benchmarked using measurements for different entrance channels, ranging from below to above the onset of quasifission, all leading to the same ^{202}Po composite system. The sensitivity of the procedure is evaluated. Its potential for isolating the fragment-mass and emission-angle information pertaining to the sole quasifission process is discussed.

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I. INTRODUCTION

Understanding the dynamics of heavy-ion collisions around the Coulomb barrier has become a major focus of nuclear research. Intense experimental and theoretical efforts have in particular been devoted to the competition between the fusion and quasifission mechanisms due to its impact on probing the limit of nuclear stability.While the former leads to a fully equilibrated compound nucleus (CN) in which decay is essentially independent of the entrance-channel phase, the latter is an out-of-equilibrium process with the reseparation of the colliding partners prior to a CN being formed. The motivation for understanding quasifission was first "practical," as it constitutes a strong hindrance to the synthesis of superheavy elements. However, studies have shown that quasifission investigations are important also from a fundamental point of view because they provide insight into various general aspects, among which are mass-equilibration and energy-dissipation timescales, the influence of shell effects on nuclear reactions, the nuclear equation of state, etc.

The quasifission mechanism still represents a challenge for both experiment and theory. A major difficulty that experimental investigations have to face is the proper separation of fusion-fission (CNF) and quasifission (QF) events, due to the overlap in their observables. In particular, slow quasifission can exhibit fragment properties that are very similar to those from CNF, with large mass exchange between the colliding partners populating up to symmetric fission. Dedicated work during the past decade has established that the most discriminant observable between the two reaction mechanisms is the correlation between the fragment mass and the emission angle, commonly referred to as the mass-angle distribution (MAD) [1,2]. On the theoretical front, dealing with the QF process is a difficult task as well. A proper modeling requires the description of the dynamical evolution of the dinuclear system from the approach to the separation stage, with a detailed account of the variety of possible shapes, the macroscopic and microscopic forces at work, and the dissipation of kinetic and rotational energies. While the pioneering experimental [3,4] and theoretical [5] studies have emphasized the leading role played by the entrance-channel Coulomb repulsion [$\approx Z_P Z_T$ with $Z_{P(T)}$ the projectile (target) charge], the systematic investigations of the past years have demonstrated the intricate dependence of the competition between fusion and quasifission on beam energy, entrance- and exit-channel fissility, reaction partner deformation, shell effects, and neutron-richness (see Refs. [1,2] and references therein). According to this complex interplay, and the uncertainty regarding some fundamental nuclear properties, theory still lacks satisfactory predictive power despite the recent huge progress within macro-microscopic [6,7] and purely microscopic [8] approaches.

To get deeper insight into QF, the fragment mass-angle distribution is an observable of primary interest because it is directly related to the time the colliding partners stick together, during which they rotate as a whole and exchange nucleons [9]. Few measurements were available until recently. An intense experimental program was therefore initiated at the Australian National University, Australia, allowing a rich set of information to be collected over the past years. Thanks to the systematic and detailed character of these studies, tremendous progress in the understanding of QF occurrence has been achieved (see Ref. [2] and references therein). Unfortunately, the experimentally measured MAD usually consists of a mixture of QF and CNF events, and no method exists to separate the individual components. That so far hampers the extraction of accurate quantitative information on the QF

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fragment properties and thus the associated dynamics. It is the goal of the present work to propose an innovative procedure to determine the MAD specific to the QF channel from experimental data. This paper focuses on the presentation of the method and proof-of-principle. Application to the most recent dedicated measurements will be presented in a follow-up publication [10].

II. STRATEGY TO INFER THE MAD FOR QUASIFISSION

A. Methodology

As noted in the Introduction, while experimental signatures of the occurrence of QF have been identified, such as anomalously low evaporation-residue cross sections, large fragment mass distribution widths [11], and anisotropies [12], the data set collected during a specific measurement is a mixture of QF and CNF, with no event-by-event discrimination between the two processes. We propose here to extract an experimental estimate of the QF mass-angle distribution by subtracting from the measured MAD the contribution from CNF as obtained from a suited model. From the inferred MAD, the fragment mass distribution and angular distribution characteristic of the sole QF mechanism can be derived. To the best of our knowledge, our proposal is the first attempt to isolate the QF component.

Elaborate microscopic calculations within the time-dependent Hartree-Fock (TDHF) approach are available for the mean QF properties and as computed for specific initial conditions (see, e.g., Ref. [13]). An estimate of the MAD from TDHF is given in Ref. [14] by folding the mean TDHF mass and angle with Gaussian functions. A full calculation—including all impact parameters, orientation angles, and fluctuations—is still to be developed [15]. In addition, going beyond the TDHF approach is necessary to compute the properties of the CNF channel. Consequently, comparison of the TDHF approach with experiments is still limited, namely when a mixture of CNF and QF is present. Monte Carlo simulations based on classical Coulomb trajectories for the incoming and outgoing nuclei and some parametrizations of the QF properties are presented in Ref. [9], but again only the average tendency for the QF channel can be obtained, and fusion-fission is not calculated. These simulations were thoroughly pushed and exploited by Prasad *et al.* [16] and the average sticking time of fast quasifission was extracted. Unfortunately, such simulations cannot be used for slow quasifission which, together with CNF, populates the symmetric region, and there is no way at present day to separate the two components. This shows the need for a procedure like the one proposed here.

B. Theoretical framework

The model used in this work is based on the stochastic classical approach of fission dynamics [17]. Dynamical calculations were performed with the four-dimensional (4D) Langevin code developed by Nadtochy and collaborators [18]. A brief description of the model is given here below, with emphasis on the aspects important for the present work. Further details can be found in the quoted references.

In the stochastic approach, fission is modeled considering the most relevant degrees of freedom as collective coordinates. Their evolution with time is treated as the motion of Brownian particles, which interact stochastically with the larger number of internal degrees of freedom constituting the surrounding "heat bath." In the present model, four collective coordinates are considered. Three variables describe the shape of the nucleus along its path to fission, and a fourth one corresponds to the orientation of its angular momentum relative to the symmetry axis. The shape coordinates $\mathbf{q}(q_1, q_2, q_3)$ are derived from the (c, h, α) parametrization [19], representing elongation, neck constriction, and left-right asymmetry, respectively. The projection K of the total angular momentum onto the symmetry axis of the fissioning nucleus is chosen for the fourth so-called tilting coordinate as originally proposed by Lestone [20]. Following Ref. [20], the motion in the K direction is assumed to be overdamped. A main advantage of the model for the present work, dedicated to an as proper as possible modeling of the mass-angle correlation, is the fact that the time evolutions of the shape and K coordinates are treated simultaneously and in a consistent manner. This is done by solving in parallel the corresponding Langevin equations of motion.

Several ingredients enter the above-outlined modeling. In the calculations of the present work, we employ the "standard" set of ingredients and parameters [18]. The driving potential is given by the Helmholtz free energy $F(\mathbf{q}, K) = V(\mathbf{q}, K) - a(\mathbf{q})T^2$, with $V(\mathbf{q}, K)$ being the potential energy, $a(\mathbf{q})$ the level-density parameter, and T the temperature of the system. The potential energy $V(\mathbf{q}, K)$ is calculated within the framework of the macroscopic model FRLDM accounting for the finite range of nuclear forces [21], and the prescription of Ignatyuk *et al.* [22] is used for the level-density parameter. The moment of inertia required to calculate the rotational part of $V(\mathbf{q}, K)$ accounts for the diffuseness of the nuclear surface [23]. Calculation of the mass tensor in the equation of motion uses the Werner-Wheeler approximation of an incompressible irrotational flow [23]. The friction tensor is derived assuming the chaos-weighted one-body dissipation formalism for the shape variables [24] and using the prescription of Refs. [25,26] for the K coordinate. Deexcitation by evaporation of light particles by the compound system prior to scission is taken into account employing the Monte Carlo approach. Particle-decay widths are calculated within the Hauser-Feschbach theory. The initial angular momentum of the compound nucleus is related to the entrance-channel reaction system according the the prescription of Ref. [27], with some adjustment (see below). Note that the ingredients entering the model used in this work are based on macroscopic concepts, which restricts its validity to fission at moderate-to-high compound-nucleus excitation energy. This limitation has no consequence for the present purpose, because for reasons given in the next section we consider here systems with E^* above about 55 MeV.

The 4D model employed in this work has been shown to be able to explain a large variety of observables for fission over a wide range of medium- and heavy-mass compound nuclei [18,28–30], including fragment mass distribution widths σ_M and angular anisotropies $W(0^\circ)/W(90^\circ)$. Models based on the Langevin approach with a larger number of shape degrees of freedom and including microscopic effects (see, e.g., Refs. [31–33]) do exist. However, none of them treat the dynamics of K, and therefore a statistical-model picture, along with the assumption of a specific transition state, has to be used when it comes to the prediction of fission-fragment angular distributions and anisotropies. The dynamics of the K mode was explicitly coupled to the dynamical treatment of shape evolution by Karpov *et al.* [34] and Eremenko *et al.* [35]. The former work considers three shape coordinates as we do, but the evolution of K is governed by energetics arguments, only, in a Metropolis-like algorithm. In particular, friction in the K mode is not included. The work of Eremenko *et al.* [35] uses an approach similar to that of Ref. [34], but for a single collective shape variable. In the field of heavy-ion collisions, for excitation energies above 40 MeV or so, the theoretical framework that we propose therefore constitutes *a priori* an ideally suited approach for the calculation of reliable MAD for CNF, which can further be used to estimate the MAD characteristic of QF in comparison with experiment.

III. RESULTS

A. Benchmark of the model

To illustrate the proposed method, we consider the data set of Rafiei *et al.* [36]. This measurement is particularly interesting for the present demonstration, because it consists of the MAD for four different reactions ($^{16}O + {}^{186}Os$, $^{24}Mg + {}^{178}Hf$, $^{34}S + {}^{168}Er$, $^{48}Ti + {}^{154}Sm$) all leading to the same compound nucleus ^{202}Po over a wide excitation-energy E^* range, which is important to reveal robust trends. Furthermore, the ^{202}Po compound nucleus being moderately fissile, its angular momentum L can crucially influence its decay and fragment properties [29,37]. In other words, the intricate interplay between the various effects important in CNF (i.e., Z^2_{CN}/A_{CN}, E^*, and L), and as supposed to be accounted for in the model employed, should be most effective.

The experimental and calculated fission-fragment mass distributions for $^{24}Mg + {}^{178}Hf$ at $E_{lab} = 145$ MeV (equivalently, $E^* = 79$ MeV, or energy above the barrier $E_{cm}/V_b = 1.26$) are compared in Fig. 1(a). According to the analysis of Rafiei *et al.* [36], only CNF contributes in this system. A good description by the model is indeed observed. Similar agreement is obtained at other energies for this reaction, as well as for $^{16}O + {}^{186}Os$ where QF is not present neither. The fission-fragment angular distribution as predicted for the same reaction is shown in Fig. 1(b). Angular distributions are not explicitly presented in Ref. [36], and no comparison is possible. Though, we note that the anisotropies computed for similar systems, and for which the QF contribution is negligible, were found consistent with experimental data [28].

B. MAD and evidence of QF

Having validated the model in the CN region of interest and for the observables of importance, we are able to address mass-angle correlations. The calculated MAD for $^{24}Mg + {}^{178}Hf$ at $E_{lab} = 145$ MeV is displayed in Fig. 2. Similarly to the experimental representation [36], the MAD is given in terms of the $d^2\sigma/d(\theta_{CM})d(M_R)$ intensity profile, where the mass ratio M_R is related to the absolute fragment mass M (before neutron evaporation) by $M_R = M/A_{fiss}$, with A_{fiss} being the mass of the fissioning nucleus. The two-dimensional spectrum is here shown normalized to the number of fission events. Absolute differential cross-section values can be straightforwardly obtained by normalizing the z scale with either the calculated or the experimental (wherever available) fusion-fission cross section. As expected within a theoretical framework of sole CNF, no correlation between mass and angle is observed. With the term absence of correlation we mean that the intensity pattern in the MAD of Fig. 2 is strictly vertical. The calculated MAD is compared to Fig. 2, second panel from the top, of Ref. [36]. Note though that comparison is possible only within the rectangular box depicted in Ref. [36] due to limited experimental coverage and additional bias inherent to the experiment. This box is reproduced on top of the theoretical calculation in Fig. 2. In this angular domain, the patterns of the calculated and measured MAD look very similar, giving us confidence in the potential of the proposed method to extract the QF MAD.

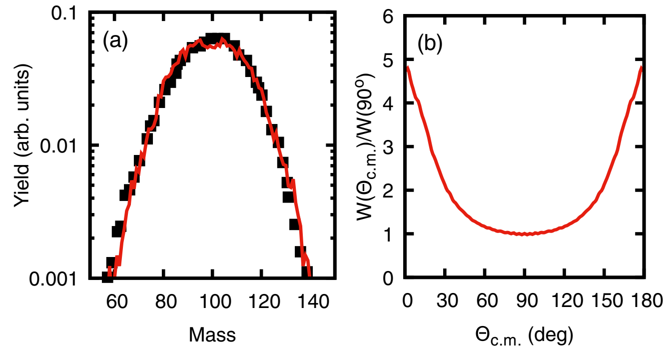

FIG. 1. Fission-fragment preneutron mass (a) and angular (b) distributions as calculated for ^{24}Mg ($E_{lab} = 145$ MeV) $+{}^{178}Hf$ (solid red lines). The experimental mass distribution (black squares) is from Ref. [36].

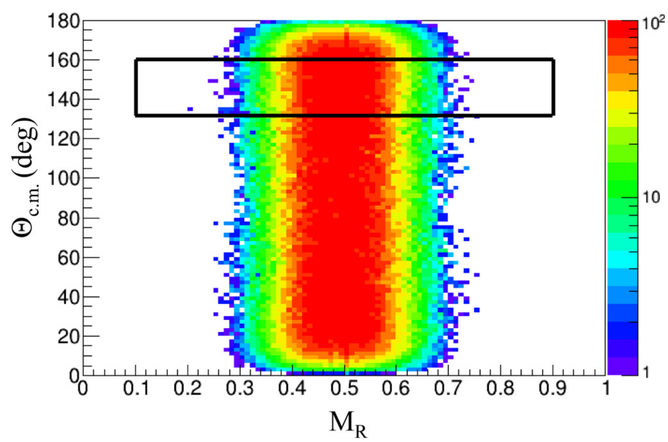

FIG. 2. Calculated MAD of CNF for ^{24}Mg ($E_{lab} = 145$ MeV) $+{}^{178}Hf$. The spectrum is normalized to the number of calculated fission events.

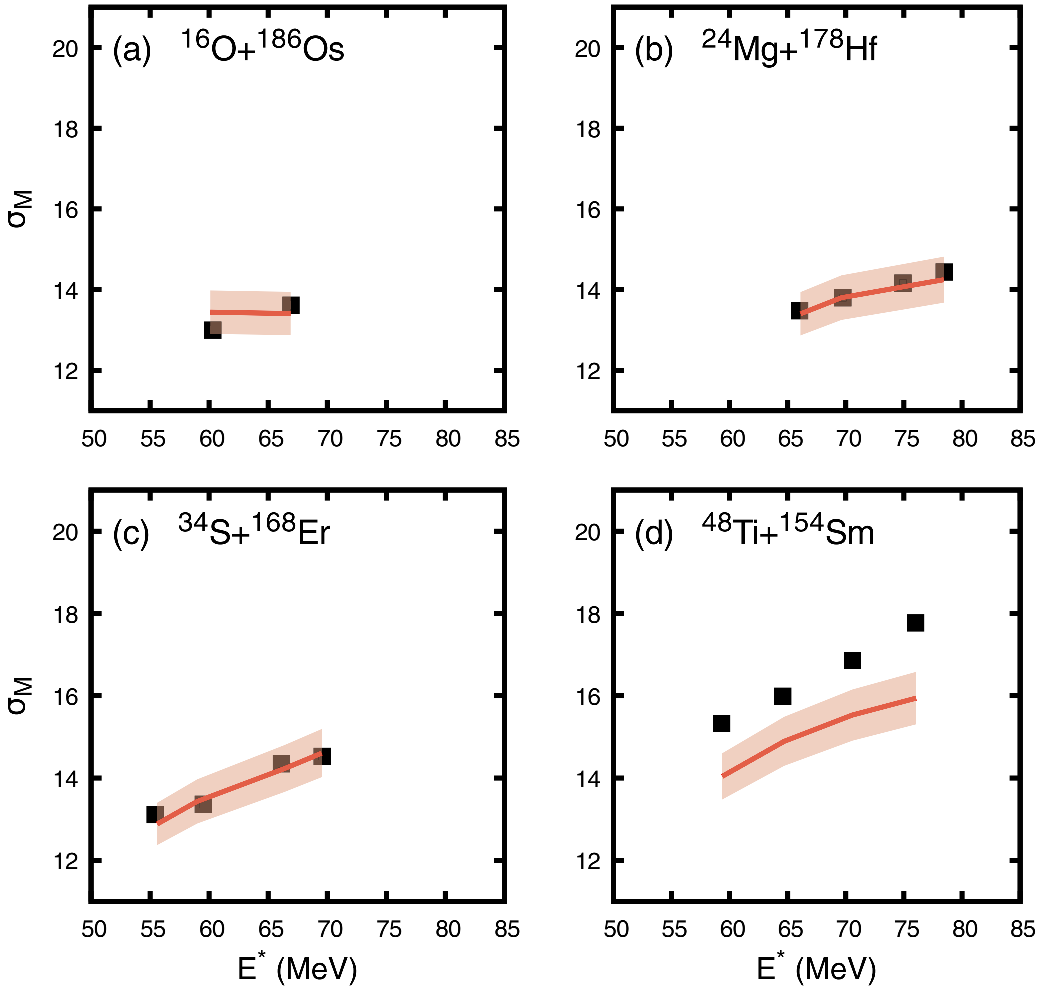

FIG. 3. Comparison between the calculated (solid red lines) and experimental (black squares) [36] fragment mass distribution widths as a function of CN excitation energy for ^{16}O + ^{186}Os (a), ^{24}Mg + ^{178}Hf (b), ^{34}S + ^{168}Er (c), and ^{48}Ti + ^{154}Sm (d). The shaded bands give the theoretical uncertainty.

As the survey of the two-dimensional MAD remains qualitative to some extent, Rafiei *et al.* [36] used the one-dimensional σ_M observable to get a more "quantitative" idea of the presence of QF depending on the reaction system. As a reference point for CNF, they employed an empirical parametrization of σ_M [38]. Here, we propose instead to compare the measured width with the dynamical prediction as obtained from the 4D Langevin code. Experiment and theory are overlaid for all four systems in Fig. 3. The comparison is restricted to E_{cm}/V_b in excess of 1.05 or so to avoid sub-barrier effects [39] as also suggested in Ref. [36]. This restriction at the same time matches the validity range of the model, implying E^* above 55 MeV for all reactions. Note that absolute σ_M values are given here, while the experimental publication [36] presented the width in terms of the mass ratio M_R; both are related by $\sigma_M = A_{\mathrm{fiss}}\sigma_{M_R}$. The shaded band around the calculated points represents what we estimate being the theoretical uncertainty coming from the more or less robust knowledge of specific ingredients entering the modeling (mainly mass table, parametrization of the driving potential, L distribution prescription, dissipation strength in shape, and K variables). For both the σ_M and angular anisotropy observables, the theoretical uncertainty as extracted from the overview of the model achievement over a wide range of systems and energies (see Refs. [18,28–30] and therein) is typically between 5% and 10%. Note that, in the present work, the L distribution was slightly adjusted (it was restricted to values given by the maximum from Ref. [27] minus 5%) to best describe the most asymmetric reaction for which only CNF contributes. The same adjustment was then consistently applied to all systems. It is observed that the experimental width is properly described for ^{16}O + ^{186}Os and ^{24}Mg + ^{178}Hf. In contrast, the width for ^{48}Ti + ^{154}Sm is substantially underestimated. The failure of the model in this case is attributed to the presence of a sizable contribution from QF that is characterized by a mass distribution broader than that from CNF [36]. The ^{34}S + ^{168}Er system is observed to be properly described, although the experimental analysis concluded that there is some QF component. Actually, for reactions leading to ^{202}Po, Rafiei *et al.* [36] set the onset of QF for an entrance-channel asymmetry around ^{34}S + ^{168}Er. The present observation (of a reasonable description where one

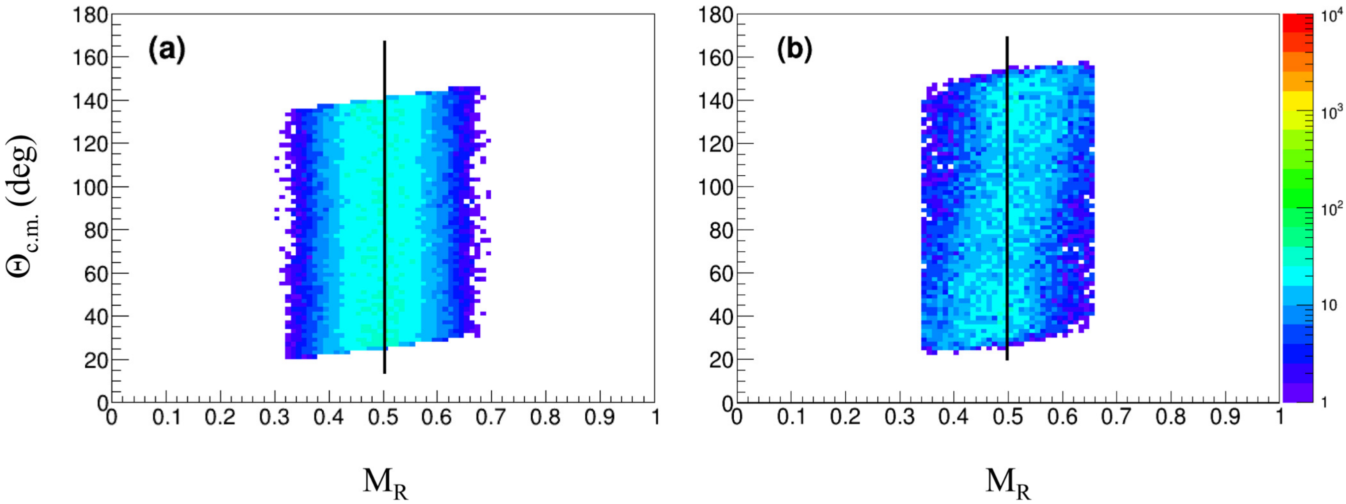

FIG. 4. Calculated MAD of CNF for ^{48}Ti ($E_{lab} = 205$ MeV) $+^{154}$Sm (a). The measured spectrum from Ref. [36] and after removal of the quasielastic and deep-inelastic contributions is shown in panel (b). The calculated spectrum is normalized to the experimental number of counts. Geometrical cuts similar to those present in the experiment have been applied. Lines are used to schematically materialize the situation of an absence of so-called mass-angle correlation.

would expect some deviation) suggests that we are reaching the accuracy of the theoretical framework. The consequence of this is discussed in Sec. III D.

C. About extracting the quasifission MAD

As noted above, the data collected in heavy-ion collision experiments usually contain a mixture of QF and CNF. To isolate the contribution specific to QF for the MAD observable, we propose to subtract from the measured MAD the mass-angle correlation characteristic of CNF as obtained by a dynamical CNF model. From the extracted differential MAD, the fragment mass distribution and angular distribution relevant for quasifission alone can then be derived. This "clean" information is crucial for a proper modeling of the QF dynamics and improving the predictive power of current theories [40].

A feeling about the expected outcome of our proposal can be gotten from Fig. 4 where we show the calculated MAD for CNF [panel (a)] next to the experimental spectrum [panel (b)] [36] for ^{48}Ti ($E_{lab} = 205$ MeV) + ^{154}Sm. The quasielastic and deep-inelastic components at extreme mass ratios have been removed from the experimental figure, as they are irrelevant for the present concern. Geometrical cuts similar to those of the experimental coverage have been applied to the calculated spectrum. So far, the calculated spectrum is normalized to the number of counts in the experimental spectrum for the sake of comparison. For a meaningful quantitative subtraction, the calculated spectrum shall of course be normalized to the fusion-fission cross section, and the experimental MAD will be normalized to the capture cross section (being the sum of QF and fusion-fission). Note that, in most cases, the fusion-fission cross section would come from theory because it is usually hard to extract in experiments precisely due to the impossible discrimination between CNF and QF events.

Like in Fig. 2, the MAD calculated by the model for fusion-fission of ^{48}Ti + ^{154}Sm basically consists of a vertical band, with no so-called mass-angle correlation. In contrast, the experimental spectrum exhibits a clearly "tilted" trend. The MAD extracted from the here-proposed subtraction method is therefore anticipated to exhibit a finite correlation, running from the lower left (light QF fragment at forward angles) to the upper right (heavy QF fragment at backward angles). The exact magnitude of the correlation will depend on the cross sections for each process. The outcome of the subtraction procedure will give access for the first time to an estimate of the experimental QF mass and angular distributions. Quantitative application of the method to most recent measurements [41] will be presented in future work [10].

D. Discussion

Summarizing our observations, the dynamical model used in this work is able to consistently describe the fragment mass distribution width for ^{16}O + ^{186}Os, ^{24}Mg + ^{178}Hf, and ^{34}S + ^{168}Er and underestimates it for ^{48}Ti + ^{154}Sm. The experimental analysis [36] concluded the absence of QF for the former two reactions, some contribution for the third one, and a substantial amount of QF for the last case. In other words, the theoretical framework is not accurate enough to see a deviation from the CNF behavior for ^{34}S + ^{168}Er, at least for the σ_M observable. For this reaction, Rafiei *et al.* [36] obtained a difference of the order of 5% to 10% between the measured σ_M and the empirical parametrization in the energy range relevant here. This difference is similar to the uncertainty of our model, which limits its sensitivity and thus the ability to evidence the *very onset* of QF occurrence in this mass region. However, the present work shows that it is certainly well suited above that onset.

Most of the work on QF relies on (one-dimensional) mass or angular distributions. The occurrence or absence of QF in the vast majority of cases was determined by comparing either σ_M or the fragment anisotropy to calculations based on phenomenology or a statistical model (with the exception of Ref. [42] where dynamical Langevin calculations have been performed). The uncertainty of the parameters entering the phenomenological and statistical models, and even most important, their correlation, led to controversial conclusions regarding the occurrence of QF. The step forward with the advent of systematic measurements of the MAD [2] has proven to be essential, because it has provided a model-independent tool with which to evidence QF. However, the availability of the experimental MAD alone does not solve the issue of the discrimination between CNF and QF. Hence, a method like the one outlined here is necessary to extract the properties pertaining to the sole QF events. Furthermore, for those most challenging cases where QF is very slow and does not exhibit a mass-angle correlation [1], the necessity of such an approach becomes even the more crucial. The theoretical model that we use is not free of some uncertain ingredients, limiting its sensitivity as discussed above. However, it provides a first step in the direction of extracting QF properties from experimental data. It is hoped that further extension of the method can improve its sensitivity and accuracy.

IV. CONCLUSION

Interest in the out-of-equilibrium quasifission process occurring in heavy-ion collisions at barrier energies has grown tremendously over the past decade. The motivation of the intense worldwide experimental and theoretical efforts is both fundamental and practical, because QF is, respectively, a rich laboratory for studying nuclear dynamics and the main obstacle to the synthesis of superheavy elements. Fission-fragment mass-angle correlations are now established as the most powerful, model-independent way to evidence and study quasifission. However, the insight provided by the MAD is hampered by the impossibility to resolve the mixture of fusion-fission and quasifission in the collected data set. We propose a new procedure to extract the MAD characteristic of the sole QF component. It consists of subtracting from the measured correlation the contribution due to fusion-fission as calculated by a dynamical model. The latter is based on a four-dimensional Langevin code, with three dimensions for the description of the dynamics of nuclear shape evolution and a fourth dimension for the dynamics of the angular momentum projection degree of freedom. Such a framework is expected to be particularly suited for modeling the dynamical correlation between fragment mass and emission angle.

This work illustrates our proposal by comparing the predicted CNF properties to experimental data both below and above the onset of quasifission for a ^{202}Po composite system produced by means of different entrance channels. Good agreement is obtained for the fragment mass distribution width between the measurement and the calculation wherever QF is absent, whereas a clear discrepancy is observed where QF is known to contribute substantially. The availability of various systems permits us to study the sensitivity of the method, demonstrating its potential to be used to learn about QF properties. It shows as well the limited achievable accuracy around the very onset of QF in the case of lowly fissile systems, calling for further improvement of the CNF model. However, the current procedure remains essential to advance into the field for those systems where QF is contributing significantly. The proposed innovative approach is therefore a promising first step in the direction of deepening our insight into quasifission. It is expected to provide essential "experimental" information to dedicated models of the QF process and more generally to heavy-ion collision dynamics.

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