Matter-wave interference originates mass-angle correlation in fusion-fission

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Mass-angle correlation of fission fragments has been understood as manifestation of quasifission. We show that this is not so: the effect can originate from correlation between fusion-fission amplitudes with different total spins signifying matter-wave interference in compound nucleus processes. This resolves the well-known puzzle with the mass-angle correlation in the complete fusion sub-barrier reaction \(^{16}\text{O} + ^{238}\text{U}\). Our finding is important for more reliable predictions of production cross sections for superheavy elements. Matter-wave interference also produces quantum-classical transition to the time-orientation localization of the coherently rotating dinucleus in quasifission.

In classical physics an individual binary collision is characterized by a single orbital momentum and, therefore, by a single impact parameter corresponding to precisely defined transverse spatial position - spacial localization - of the incoming material particle. In contrast, in quantum mechanics each single particle of the incident beam is represented by a coherent superposition of partial waves with different orbital momenta. Interference between the states with different orbital angular momenta carried at once by the same incoming particle signifies matter-wave interference in the entrance channel. Manifestation of matter-wave interference between spatially distinct reaction paths - different impact parameters - in chemical reactions has been demonstrated \cite{1} by mapping the quantum collision problem onto the famous Young double-slit experiment \cite{2}.

We ask the question: Is matter-wave interference necessarily destroyed in the compound nucleus (CN) reactions? The modern understanding, based on the Bohr’s independence hypothesis, assumes absence of the \(S\)-matrix total spin \((J)\) and parity \((\pi)\) off-diagonal correlations \cite{3}. This means that the CN formation produces effect of decoherence similar to that originated from detection of the which-path - which impact parameter - in the double-slit experiment \cite{2} thereby destroying matter-wave interference and leading to the fore-aft symmetry in CN reactions \cite{4}. However this conventional understanding is inconsistent with many data sets demonstrating a strong fore-aft asymmetry of the evaporation spectra. The first well-known example of a strong fore-aft asymmetry of the evaporation spectra was reported in \cite{1} for the Pt(p,p') process. Examples of strong fore-aft asymmetry of the evaporation proton spectra in nucleon-induced reactions with the targets \(^{197}\text{Au}, ^{208}\text{Pb}, ^{209}\text{Bi}\) and \(^{nat}\text{U}\) are displayed in \cite{5}. Some representative data on the strong fore-aft asymmetry and anomalous, significantly stronger than \((1/\sin \theta)\), back-angle peaking of the proton and \(\alpha\)-particle evaporation spectra demonstrating matter-wave interference in CN processes will be displayed in the extended version of this Letter.

Here we refer to the conclusive experimental evidence for the mass-angle correlation (MAC) for the sub-barrier reaction \(^{16}\text{O} + ^{238}\text{U}\) \cite{6}. This MAC and anomalously large anisotropy of the fission fragments were interpreted \cite{6} as the orientation-dependent quasifission \cite{7}. The idea \cite{7} was tested by measuring the evaporation residue cross section for the reaction \(^{16}\text{O} + ^{238}\text{U}\) at the sub-barrier energies \cite{8}. The interpretation \cite{9, 10} has been refuted by the experimental evidence that the complete fusion is the main process in the sub-barrier region while quasifission contribution is insignificant \cite{11}. Therefore MAC in the reaction \(^{16}\text{O} + ^{238}\text{U}\) \cite{6} originates from matter-wave interference in fusion-fission and not in quasifission.

A correct understanding of competition between fusion-fission and quasifission in reactions involving prolate deformed actinide nuclei is important for reliable predictions of production cross sections for superheavy elements \cite{11, 12}. For example, the insignificance of quasifission, in particular, the orientation-dependent quasifission \cite{10} in the complete fusion sub-barrier reaction \(^{26}\text{Mg} + ^{248}\text{Cm}\) led to the observation of the new superheavy nuclide \(^{271}\text{Hs}\) \cite{11}.

In order to describe MAC one can consider fission as a complex particle evaporation \cite{13, 14}. Then the result \cite{15, 14} for the CN \((J_1 \neq J_2)\)-correlation is directly applicable to reproduce MAC and fore-aft asymmetry of fusion-fission angular distributions (FFAD).

In this Letter we extend the more popular treatment (the transition-state statistical model (TSSM)) of the FFAD \cite{16} by introducing the CN \((J_1 \neq J_2)\)-correlation. We represent FFAD in the form \(W(\theta) = \sum_K <J_2^K\sum_J F_{a,c,k}^J(\theta,E)|^2 > \). Here \(F_{a,c,k}^J(\theta,E) = (2J + 1) S_{a,c,k}^{J}(E) d_{0k}^J(\theta), < ... > \) stands for the energy averaging, \(d_{0k}^J(\theta) = D_{0k}^J(\alpha = 0, \theta, \gamma = 0)\), \(D\)-functions are the wave functions of the axially symmetric top, \(a\) is index for the entrance channel, \(K\) is projection of \(J\) on the symmetry axis of the nucleus at the saddle-point and \(c\) stands for the rest of indices of the fission channels.

Neglecting \(J\)-dependence of the potential phase shifts in the fission channels we take \(S\)-matrix in the form \(S_{a,c,k}^{J}(E) = -i \exp(i \nu^c_{a,J}) \delta S_{a,c,k}^{J}(E)\), where \(\nu^c_{a,J}\) is the potential phase shift in the entrance channel while \(\delta S_{a,c,k}^{J}(E)\) is given by the pole expansion \cite{17} with the residues \(\gamma_{a,J} F_{\alpha,J}^{a,J} c_{a,J} K\). Here, \(\gamma\)’s are the real partial width.
amplitudes in the entrance and exit (fission) channels at the saddle-point with the $\mu$-index denoting the CN resonance level. We assume that statistical properties of $\gamma$'s with fixed $J$-value are described by statistics of Gaussian Orthogonal Ensemble. Note that, for $J \geq 1$, inclusion of $K$-projections into the fission channel indices takes us to a regime of the weak coupling to the continuum, $(\gamma_{\mu}^{j,K}/D_J \approx 1/2\pi(2J + 1)) \ll 1 (D_J$ is the CN average level spacing). Therefore, the pole expansion is an accurate approximation to the $S$-matrix unitary representation in a regime of the strongly overlapping resonances, $\Gamma/D_J \gg 1$, with $\Gamma$ being the CN total decay width. The $S$-matrix spin off-diagonal correlations result from the correlation between $\gamma_{\mu_1}^{j_1,a_1} \sum_{\gamma_{\mu_2}} \gamma_{\mu_2}^{j_2,c_2}$ and $\gamma_{\mu_2}^{j_2,a_2} \sum_{\gamma_{\mu_2}} \gamma_{\mu_2}^{j_2,c_2}$ with $|J_1 - J_2|/\beta$ being the correlation length in the $(E_{\mu_1}^{j_1} - E_{\mu_2}^{j_2})$-space ($E_{\mu}^{j}$ are the resonance energies) playing a role of a quantum analog of imaginary parts of the Ruelle-Pollicott resonances (to be reported elsewhere). In turn, this correlation originates from the correlation between the CN resonance eigenstates $\phi^j_{\mu}$ of the type

$$
\int dR_1 dR_2 \left[ \phi_{\mu_1}^{j_1}(r, R_1) \phi_{\mu_1}^{j_1}(r, R_2) \phi_{\mu_2}^{j_2}(r, R_1) \phi_{\mu_2}^{j_2}(r, R_2) \right] \ .
$$

Here $r = (r_1, r_2, ..., r_n)$ are coordinates of $n$ nucleons and $R_{1,2}$ are coordinates of the rest of the $(A - n)$ nucleons. In Eq. (1) (...) stand for the spatial averaging. Note that upon the partial $(\mu_1, \mu_2)$-averaging (keeping $(E_{\mu_1}^{j_1} - E_{\mu_2}^{j_2})$ fixed with accuracy $\ll \beta$) the main contribution to Eq. (1) is produced with $R_1 \rightarrow R_2$. Seemingly paradoxically, the problem of the cross-symmetry correlation relates to the Wigner dream to develop a theory of correlations between reduced widths within a single symmetry sector (to be reported elsewhere).

The final result, for the fission fragments sufficiently lighter than the average fragment mass, reads

$$
W(\theta) \propto \sum_{J_1, J_2} (2J_1 + 1)(2J_2 + 1)(T_a^{J_1} T_a^{J_2})^{1/2} \exp(i(\varphi_{a_1}^{j_1} - i\varphi_{a_2}^{j_2}))[1 + (\beta/\Gamma)|J_1 - J_2|]^{-1}
$$

min $J_1, J_2$ $K = -min(J_1, J_2)$

$$
\sum_{K = -min(J_1, J_2)} [B_{J_1}(K) B_{J_2}(K)]^{1/2} d_{10}^{J_1}(\theta) d_{10}^{J_2}(\theta)
$$

(2)

with $B_J(K > J) = 0$, $B_J(K \leq J) = p_K / \sum_{K = -J}^{+J} p_K$. Here, $p_K = \frac{J_{eff}}{T_a/\hbar^2} J_{eff}^{-1} = \frac{J_{perp}}{J_{par}}$, $J_{par}$ and $J_{perp}$ are the nuclear moments of inertia for rotations around the symmetry axis and a perpendicular axis, $T_a = [8(E - B_f - E_{\text{res}})/A]^{1/2}$ is the nuclear temperature at the saddle-point, $B_f$ is the fission barrier, $E$ is the excitation energy and $E_{\text{res}}$ is the rotational energy. For $\beta/\Gamma \gg 1$ the $(J_1 \neq J_2)$-correlations decay much faster than the CN average lifetime thereby leading to the TSSM result. The time power spectrum, $P(\theta, t)$, is given by Eq. (2) with $\exp[-(\Gamma - \beta)|J_1 - J_2|/\hbar]$ instead of $[1 + (\beta/\Gamma)|J_1 - J_2|]^{-1}$. The time-dependent FFAD is $\propto \int_0^\infty d\tau P(\theta, \tau)$.

Eq. (2) reproduces fore-aft symmetry and MAC for the mass-asymmetric fission due to the $(J_1 \neq J_2)$-contributions with odd values of $(J_1 + J_2)$, i.e., $\sigma_1 \neq \sigma_2$. The fore-aft symmetry is restored destroying MAC for $\beta_{\pi_1 \neq \pi_2} \gg 1$ leading to $W(\theta) \rightarrow W(\theta) + W(\pi - \theta)$, like for the mass-summed FFAD. However, such a fore-aft symmetry does not necessarily mean a complete mass-symmetrization but can also imply that the mass-asymmetric deformed system at the saddle-point is oriented in opposite directions with equal probabilities. Yet, weakening of MAC with a decrease of the mass-asymmetry can be reproduced by increasing $\beta_{\pi_1 \neq \pi_2}$ with $\beta_{\pi_1 \neq \pi_2} \rightarrow \infty$ for the mass-symmetric fission.

On the initial stage the colliding ions form a dinuclear system. A distribution of its orientations, $P_{\theta} = (\theta)$, is identified with the time power spectrum at $t = 0$ for dissipative heavy-ion collisions or for heavy-ion scattering. It follows that $P_{\theta} = (\theta)$ is formally given by Eq. (2) with $\beta = 0$ and $K_0^2 = 0$ ($B_J/K = \delta_0\kappa$). We expand $\varphi_{\theta}^{j_1} = (\Phi(J - j_1) + \Phi(J - j_2)/2$, where $J = \sum_{j = 0}^\infty T_a^j / \sum_{j = 0}^\infty T_a^j$. Then, for $\Delta_{\theta} = 0 \leq 1$, we are dealing with a fusion of the dinuclear system preferentially orientated along the $\Phi$-direction. It follows from Eq. (2) that the pre-fusion phase relations (initial conditions) in the entrance channel, $(\varphi_{\theta_1}^{j_1} - \varphi_2^{j_2})$, are not forgotten even for the complete $K$-equilibration, $K_0^2 \gg J^2 >$, providing $\beta/\Gamma$ is a finite number. This means that if the same CN is formed but with different phase relations in the entrance channel (different collision partners) the FFADs will be different for a finite value of $\beta/\Gamma$. For $\Delta_{\theta} = 0 > \pi$, the $(J_1 \neq J_2)$-correlation and MAC are strongly suppressed already on the pre-fusion stage even for a finite value of $\beta/\Gamma$.

The sub-barrier $^{18+238}_{\text{U}}$ fusion-fission demonstrates both MAC and anomalous anisotropy. To illustrate the effect of the orientation-dependent fusion-fission we focus on the energy interval $E_{\text{c.m.}} = 72.8 - 75.6$ MeV ($E = 34.5 - 37.3$ MeV, $\Gamma/D_J \approx 10^{-5}$) for which the interpretation suggested absence of the fusion-fission. Employing a standard statistical model we find that to reproduce the experimental value of $\sigma_{\text{ER}}(4n)/\sigma_{\text{fiss}} \approx 6 \times 10^{-5}$ for $E_{\text{c.m.}} = 72$ MeV it is necessary to take $B_f = 4.15$ MeV instead of $B_f = 1.5$ MeV. Here $\sigma_{\text{fiss}}$ and $\sigma_{\text{ER}}(4n)$ are the fission and 4n evaporation residue cross sections. For $B_f = 1.5$ MeV, a statistical model predicts $\sigma_{\text{ER}}(4n)/\sigma_{\text{fiss}} \approx 10^{-11}$ for $E_{\text{c.m.}} = 72$ MeV instead of the experimental value of $\approx 6 \times 10^{-5}$. For $B_f \approx 1$ MeV and $B_f = 4.15$ MeV, the pre-equilibrium fission [24, 23, 20], which anyway can...
not reproduce the fore-aft asymmetry and the associated MAC, is insignificant.

With a decrease of \( E_{cm} \) from 75.6 MeV to 72.8 MeV (\( A_{exp} \rightarrow 1 \)) increases from \( \approx 0.85 \) to \( \approx 1.6 \) (\( A = W(\pi)/W(\pi/2) \)). In contrast, the TSSM predicts a decrease of \( A_{TSSM} \rightarrow 1 \) from \( \approx 0.16 \) to \( \approx 0.1 \) (\( A = J_2(75.6 \text{MeV}) / J_2(72.8 \text{MeV}) \)). With \( K_0 = 200 \), \( < J^2(72.8 \text{MeV}) > = 80 \) and \( < J^2(75.6 \text{MeV}) > = 128 \). Then, to reproduce \( A_{exp} \) within the TSSM one has to take \( K_0(75.6 \text{MeV}) \approx 37 \) and \( K_0(72.8 \text{MeV}) \approx 12 \) instead of \( K_0 \approx 200 \) with \( \langle J_f \rangle \) from \( 23 \).

In our interpretation we take into account the orientation dependence of the sub-barrier fusion for the target having a prolate deformation \( K = 0 \). Then we deal with a fusion of the strongly mass asymmetric dinuclear system with \( K = 0 \) which is preferably oriented parallel to the beam axis with its projectile-like tip pointing opposite to the beam direction. Therefore we take \( \Phi = \pi \) for both \( E_{cm} = 72.8 \) MeV and 75.6 MeV.

To minimize a width of \( P_{K=0}(\theta) \) for the smaller sub-barrier energy \( E_{cm} = 72.8 \text{MeV} \) we take \( \Phi = 0 \). Then, for \( T_u \propto \exp[-J^2(20) \theta^2] \) with \( < J^2(20) > = 80 \), \( P_{K=0}(\theta) \) is peaked at \( \theta = \pi \) having a width of \( \Delta K=0 \approx 6^\circ \). Admixture of the states with \( K = 1,2 \) will increase dispersion of the pre-fusion dinuclear orientation.

Since \( < J^2 > / (4K^2_0) = 0.1 - 0.16 \ll 1 \) we take \( K_0^2 = \infty \) (\( B_2(K) = 1/(2J + 1) \)) resulting in isotropic FFAD in the absence of the \( (J_1 \neq J_2)-\)correlation. Now the mass-summed FFAD, obtained by symmetrizing Eq. (2) about \( \theta = \pi/2 \), depends on a single parameter \( \beta/\Gamma \). In Fig. 1, \( A = 2.6 \) is reproduced with \( \beta/\Gamma = 1.66 \). For \( < J^2(75.6 \text{MeV}) > = 128 \) and this same value of \( \beta/\Gamma = 1.66 \), \( A_{exp}(75.6 \text{MeV}) = 1.85 \) is reproduced in Fig. 1 with \( |\bar{\Phi}| = 3.6^\circ \) corresponding to a width of \( P_{K=0}(\theta) \) of \( \approx 8^\circ \) with its tail extended up to \( \approx 135^\circ \). This demonstrates that a fast growing of \( (A-1) \) with the energy decrease is due to the moderate reduction of a dispersion of the pre-fusion dinuclear orientation with a decrease of \( E_{cm} \). With \( \Gamma_g \approx T_g \exp[-B_2(T_b)/(2\pi)] \approx 2.5 \text{ keV} \) (\( T_b = 1 \text{ MeV}, B_2 = 4.15 \text{ MeV} \)) we obtain \( \beta \approx 4 \text{ keV} \) and \( h/\beta \approx 1.65 \times 10^{-19} \text{ sec} \). Therefore, the characteristic time for a decay of the CN \( (J_1 \neq J_2)-\)correlation is about 3 orders of magnitude longer than the CN thermalization time, \( h/\Gamma_{spr} \approx 1.3 \times 10^{-22} \text{ sec} \), where \( \Gamma_{spr} \approx 5 \text{ MeV} \) is a width of the giant resonances.

MAC has been revealed in \( \bar{\Phi} \) from asymmetry of the fragment mass distribution at the back-angles. For the lowest energy reported, \( E_{cm} = 72.8 \text{ MeV} \), it has been found that the light fragment yield, \( M_L \), integrated over the angular range \( 110^\circ \leq \theta_{cm} \leq 155^\circ \) exceeds that for the heavy fragments, \( M_H \leq 150 \), by a factor of 1.8±0.3. To reproduce this experimental value we need \( r = R_1/R_3 = 1.8 \), where

\[
R_{k}(\theta) = \int_{110^\circ}^{70^\circ} d\theta \sin \theta W(\theta)
\]

We calculate \( r \) with \( K_0^2 = \infty \) and the previously fixed \( \Phi = \pi \), \( \bar{\Phi} = 0 \), \( < J^2 > = 80 \) first with \( \beta_1=\beta_2/\Gamma = 1.66 \). We obtain \( r = 2.7 \) instead of the experimental value 1.8. To reduce the calculated value of \( r \) we increase \( \beta_1,\beta_2/\Gamma \) but keep \( \beta_1=\beta_2/\Gamma = 1.66 \) unchanged (to have the unchanged mass-summed FFAD). The value \( r = 1.8 \) is obtained with \( \beta_1=\beta_2/\Gamma = 3.36 \). The corresponding FFADs calculated with \( \beta_1=\beta_2/\Gamma = 3.36 \) for both the energies \( E_{cm} = 72.8 \text{ MeV} \) and 75.6 MeV are displayed in Fig. 1. FFADs for the heavy fragments, \( M_H \geq 150 \), can be obtained by the reflection about \( \pi/2 \), \( W_H(\theta) = W_L(\pi-\theta) \), reproducing the significant MAC \( \bar{\Phi} \). Weakening of the fore-aft asymmetry and MAC for \( E_{cm} = 72.8 \text{ MeV} \) with increase of \( \beta_1=\beta_2/\Gamma \) is demonstrated in Fig. 1. This weakening also illustrates evolution of FFADs for a transition from mass-asymmetric to mass-symmetric fission.

While the analysis \( \bar{\Phi} \) produced a conclusive evidence for MAC the mass-angle distribution is even more sensitive, informative and visually transparent experimental observable to demonstrate the effect. For example, MAC is unnoticeable in the fragment mass distribution for the \( ^{19}F+^{238}U \) reaction \( \text{[28]} \). Yet MAC is clearly seen in the mass-angle distribution, Fig. 4 in \( \text{[28]} \).

Suppose that, on the pre-fusion stage, the prolate shape of the target nucleus becomes unstable changing to the oblate shape with the symmetry axis oriented along and/or perpendicular to the beam direction. One may imagine a coexistence of the three pre-fusion configurations coherently contributing into a formation of the same CN. Can interference between the three fusion-fission amplitudes produce oscillations in the FFAD?

In case of the noticeable suppression of the evapor-
tion residue yield indicating quasi-fission we would be led to deal with the \((J_1 \neq J_2)\)-correlation at the conditional saddle-point. The corresponding expression for the angular distribution can be envisaged from the consideration of the angular distributions in dissipative heavy-ion collisions \([21]\). The result for the time power spectrum is given by Eq. \(2\) with exp\((-\Gamma t/\hbar)\exp(-i\omega t/(J_1 - J_2) - \beta(J_1 - J_2)/\hbar)\) instead of \([1 + (\beta/\Gamma)(J_1 - J_2)]^{-1}\), where \(\omega\) is a real part of the angular velocity of the coherent rotation of the dinuclear system. In this resulting expression possible time-dependencies of \(\omega\) and \(K_2^J\) can be taken into account \([29]\). The time power spectrum for quasi-fission describes the classical-like rotation of the deformed system having a classically single \(J\)-value \([21]\). Yet, this classical-mechanics picture originates from a coherent superposition of the dinuclear states with different quantum-mechanical \(J\)-values. Determination of the impact parameter of the incident particle using a which-path detector destroys the coherent superpositions \([1, 2]\) of the dinuclear states with different quantum-mechanical \(J\)-values disabling the quantum-classical transition to the macroscopic-like time-orientation localization and the classical-like rotation. Therefore it is matter-wave interference that produces quantum-classical transition to the time-orientation localization/correlation for the macroscopic-like rotation of the deformed system. This interpretation is complementary to the traditional understanding that the emergence of macro-world described by classical physics originates from decoherence destroying matter-wave interference \([2]\). For \(\beta \ll \Gamma\) and \(\Phi \approx 0\) the fore-aft asymmetry factor for quasi-fission fragment angular distribution is \(\propto \cosh \left(\pi/\sqrt{\Gamma \omega} \right)\) demonstrating yet again that matter-wave interference produces a classical-like rotation of the osculating complex with a classically single \(J\)-value (fixed \(\omega\)), Fig. 14 in \([31]\).

The wave nature of the heated organic macromolecules has been firmly established, in a model-independent way, raising a question of matter-wave interference for biologically functioning entities of elevated temperature carrying the code of self-replication such as viruses and bacteria \([31]\). In this Letter we have proposed that matter-wave interference plays an important role in nuclear fission. We have demonstrated that, contrary to the conventional understanding, MAC does not necessarily indicate hindrance of the CN formation due to quasi-fission but can originate from the CN spin off-diagonal phase correlation signifying matter-wave interference in fusion-fission. One of the central conventionally counter-intuitive problems of the proposed interpretation, which has not been explicitly addressed in the study of matter-wave interference with complex molecules \([2, 31]\), is to justify the anomalously slow cross-symmetry phase relaxation in classically chaotic many-body systems as compared to the relatively fast phase and energy relaxation (thermalization) within a single symmetry sector.

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