

Effect of phase relaxation on quantum superpositions in complex collisionsL. Benet,¹ S. Yu. Kun,^{1,2,3} and Wang Qi⁴¹*Centro de Ciencias Físicas, National University of Mexico (UNAM), 62210-Cuernavaca, Morelos, Mexico*²*Nonlinear Physics Center, RSPHysSE, Australian National University, Canberra ACT 0200, Australia*³*Department of Theoretical Physics, RSPHysSE, Australian National University, Canberra ACT 0200, Australia*⁴*Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China*

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We study the effect of phase relaxation on coherent superpositions of rotating clockwise and anticlockwise wave packets in the regime of strongly overlapping resonances of the intermediate complex. Such highly excited deformed complexes may be created in binary collisions of heavy ions, molecules, and atomic clusters. It is shown that phase relaxation leads to a reduction of the interference fringes, thus mimicking the effect of decoherence. This reduction is crucial for the determination of the phase-relaxation width from the data on the excitation function oscillations in heavy-ion collisions and bimolecular chemical reactions. The difference between the effects of phase relaxation and decoherence is discussed.

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I. INTRODUCTION

For highly excited strongly interacting many-body systems, the independent particle picture has very limited validity [1]. For high excitations, the interaction results in a quick decay of single-particle [2–4] as well as collective modes [2,4], which are not eigenstates of the Hamiltonian of the system. This decay leads to the formation of complicated many-body configurations. Each of these many-body states is characterized by a uniform occupation of the accessible phase space and by the sharing of energy between many particles of the system. The characteristic time for the formation of such spatially extended ergodic many-body states, independent of the initial conditions, is given by the inverse spreading width, $\tau_{\text{erg}} = \hbar/\Gamma_{\text{spr}}$ [4]. Introduced by Wigner [5], Γ_{spr} also characterizes the width of the distribution of the expansion coefficients of the many-body eigenstates over a noninteracting mean-field basis. For sufficiently high excitation energy, the decay of single-particle modes creates a linear superposition of a very large number of many-body configurations. The question is whether the phase relations between these individually ergodic many-body states in the superposition may still preserve a memory of the way the system was excited. This question is of fundamental importance for the study of relaxation phenomena in nuclear, atomic, molecular, and mesoscopic many-body systems, and for many-qubit quantum computation. In particular, if the phase relaxation is longer than the energy relaxation time $\hbar/\Gamma_{\text{spr}}$, this can extend the time for quantum computing [6,7] beyond the quantum-chaos border [8,9].

To answer this question from first principles is difficult because of computational limitations. Indeed, in order to solve the full quantum many-body problem, one would require a many-qubit quantum computer. Therefore, the only currently available way to search for possible manifestations of long phase relaxation is the experiment and a careful data analysis. Nuclear systems are an ideal laboratory to study many-body systems, since nuclear interactions are so strong that external perturbations can often be neglected. In particular, the analysis

of Refs. [6,7,10] of the data on asymmetry around 90° c.m. in angular distributions of evaporated protons from heavy nuclei in nucleon-induced and photonuclear reactions clearly indicates that phase-relaxation time in highly excited strongly interacting many-body systems can be up to eight orders of magnitude [7] longer than the energy-relaxation time $\hbar/\Gamma_{\text{spr}}$. This reveals a new form of matter—thermalized non-equilibrated matter introduced in Refs. [11–13].

A more subtle indication of a slow phase relaxation is found in light heavy-ion scattering [14–17]. It manifests itself in the formation of rotating wave packets whose spreading time, which is given by the inverse phase-relaxation width, is much longer than the energy-relaxation time $\hbar/\Gamma_{\text{spr}}$. The approach [15–17] treats time-delayed light heavy-ion scattering in terms of the formation and decay of quasimolecular resonances [18]. The highly excited coherently rotating intermediate system has the energy of the intrinsic excitation ≥ 10 MeV. The intermediate system is in the regime of strongly overlapping resonances. Therefore, this coherent rotation does not originate from the discreteness of the spectrum, which is not resolved, but is due to the spin off-diagonal correlations between partial width amplitudes. Indeed, the period of the coherent rotation is much shorter than the inverse level spacing of the intermediate system. This reveals a new root, as compared to Bohr's correspondence principle, for a quantum-classical transition in highly excited many-body systems.

The width of the rotating wave packets is about $1/d + \beta t/\hbar \approx 1/d + \beta \vartheta/\hbar\omega$, where d is the effective number of partial waves, β is the spin off-diagonal phase-relaxation width, ω is the angular velocity, and t and ϑ are the time and angle of the rotation, respectively. This seemingly allows us to determine the wave-packet spreading rate from the time power spectra at different scattering angles, which can be reconstructed, for binary collisions, from the data on energy fluctuations of the cross sections [16,17]. These fluctuations originate from the energy-fluctuating collision amplitude corresponding to a resonance time-delayed process. However, for elastic heavy-ion scattering, such energy fluctuations can be typically observed only at backward

angles $140^\circ \leq \theta \leq 180^\circ$, since for smaller θ the direct reaction contribution grows exponentially and becomes much greater than the time-delayed one. Therefore, since at the initial moment of time the deformed intermediate system is oriented in the forward direction $\theta \approx 0^\circ$, the spreading of the wave packets within the backward angular interval $140^\circ \leq \theta \leq 180^\circ$ can hardly be detected reliably. Moreover, the width of the wave packets at this backward-angle range does not allow us to determine β , since the angular dispersion $1/d$ at $t = 0$ is also unknown. In this paper, we show that both d and β can indeed be determined unambiguously. This results from the strong sensitivity of the interference fringes between the rotating clockwise and anticlockwise wave packets for the backward angle range to the phase-relaxation width: As β grows, the interference fringes are suppressed more strongly. The effect is considered in relation to heavy-ion scattering and bimolecular chemical reactions. The difference between the effects of phase relaxation and decoherence is discussed.

II. TIME POWER SPECTRUM OF TIME-DELAYED COLLISIONS

Following Ref. [19], we consider spinless collision partners in the entrance and exit channels. The time and angle dependent intensity of the decay (the time power spectrum) $P(t, \theta)$ is given by the modulus square of the Fourier component of the energy-fluctuating collision amplitude. It can be also expressed as the Fourier component of the amplitude energy autocorrelation function. $P(t, \theta)$ has been obtained in [19] by summing over a very large number of strongly overlapping resonance levels, $\Gamma/D \gg 1$, where Γ is the total decay width and D is the average level spacing of the intermediate complex. As a result, after changing from summation over the resonance levels to integration, which is a good approximation for $t \ll \hbar/D$, $P(t, \theta)$ takes the form

$$\begin{aligned}
 P(t, \theta) &\propto H(t) \exp(-\Gamma t/\hbar) \\
 &\times \sum_{JJ'} (2J+1)(2J'+1) \times [W(J)W(J')]^{1/2} \\
 &\times \exp[i(\Phi - \omega t)(J - J') - \beta|J - J'|t/\hbar] P_J(\theta) P_{J'}(\theta).
 \end{aligned} \tag{1}$$

Here, $H(t)$ is the Heaviside step function, β is the spin phase-relaxation width, ω is the angular velocity of the coherent rotation, Φ is the deflection given by the total spin J (in \hbar units) derivative of the potential phase shifts (direct reaction), and $P_J(\theta)$ are the Legendre polynomials. The physical meaning of the inverse spin phase-relaxation width, \hbar/β , is the characteristic time for the angular spreading of the clockwise and anticlockwise rotating wave packets. The partial average reaction probability is taken in the J -window form, $W(J) = \langle |\delta S^J(E)|^2 \rangle \propto \exp[-(J - \bar{J})^2/d^2]$, where \bar{J} is the average spin and d is the J -window width.

It should be noted that a similar effect of coherent rotation of the nuclear molecule, as described by Eq. (1) with $\beta = 0$, was found in Ref. [20] for a scattering of heavy nuclei. However, while Eq. (1) has been obtained for a large number of open channels and strongly overlapping resonances of the intermediate system [19], the time power spectrum in Ref. [20]

was derived from one open channel and one resonance pole form of the S -matrix elements. Therefore, the latter approach is not applicable for the forthcoming analysis of the $^{12}\text{C}+^{24}\text{Mg}$ system since, for this system, the number of open channels is much greater than 1. Furthermore, the one-resonance pole form of the S -matrix elements employed in Ref. [20] results in isolated resonances with the energy spacing between them to be about $\hbar\omega$. This is not the case for the $^{12}\text{C}+^{24}\text{Mg}$ elastic scattering analyzed in this paper. Indeed, the data on the excitation function for the $^{12}\text{C}+^{24}\text{Mg}$ elastic scattering at $\theta = 180^\circ$ reveal about 15 local maxima on the energy interval $\Delta E_{\text{c.m.}} = 13\text{--}22$ MeV [21]. Then the interpretation [20] would mean that average level spacing, ≈ 0.6 MeV in our case, between the local maxima is given by $\hbar\omega$. This, in turn, would reflect the quasiperiodic behavior of the cross section energy autocorrelation functions with the period of about 0.6 MeV. This is inconsistent with the data [21] showing the quasiperiodicity of the cross section with the period of about 2.9 MeV yielding $\hbar\omega = 1.35\text{--}1.45$ MeV [16,17]. Moreover, the interpretation [20] implies that these local maxima in the excitation function should be approximately equidistant. Instead, the data [21,22] show big fluctuations, from 0.3 to 1.3 MeV, for the energy spacing between the nearest-neighboring local maxima in the excitation function. Note that the indication against the interpretation in terms of isolated resonances comes from the statistically insignificant channel-channel correlations for the $^{12}\text{C}+^{24}\text{Mg}$ elastic and inelastic scattering at $\theta = 180^\circ$ [16]. Finally, the effect of coherent rotation [20] does not survive a generalization [23] by taking into account intermediate structure by means of inclusion of many isolated resonances in the S -matrix elements. On the basis of the above arguments, we believe that the interpretation of the fine structure in the excitation functions for the $^{12}\text{C}+^{24}\text{Mg}$ scattering at $\theta = 180^\circ$ in terms of overlapping, rather than isolated, resonances of the intermediate system is a justified approach.

First, we calculate $P(t, \theta)$ for the set of parameters obtained from the description [17] of the experimental cross section energy autocorrelation functions [21] for $^{12}\text{C}+^{24}\text{Mg}$ elastic and inelastic scattering at $\theta = 180^\circ$ [22]. For these collisions, the analysis of the oscillations in the cross section energy autocorrelation functions [16,17] indicates the formation of stable rotational wave packets, in spite of the strong overlap of resonance levels in the highly excited intermediate molecule. The set of parameters [17] is $\Phi = 0$, $d = 3$, $\bar{J} = 14$, $\beta = 0.01$ MeV, $\hbar\omega = 1.45$ MeV, and $\Gamma = 0.3$ MeV.

In Fig. 1, we plot the quantity $AP(t, \theta)/\langle \sigma(E, \theta) \rangle$ at four moments of time, with $T = 2\pi/\omega$ being the period of one complete revolution. Here, $\langle \sigma(E, \theta) \rangle \propto \int_0^\infty dt P(t, \theta)$ is the energy-averaged differential cross section for the time-delayed collision. In Fig. 1, $P(t, \theta)$ is scaled with $\langle \sigma(E, \theta) \rangle$ for the reasons discussed in Ref. [19]. The constant A is determined by the condition $AP(t=0, \theta=0)/\langle \sigma(E, \theta=0) \rangle = 1$. Figure 1(a) shows the two slightly overlapping wave packets rotating toward each other in the backward direction. In panels (b) and (c), the wave packets strongly overlap around $\theta = 180^\circ$, producing interference fringes. Finally, in panel (d) the wave packets have passed each other and move apart rotating in the forward direction.

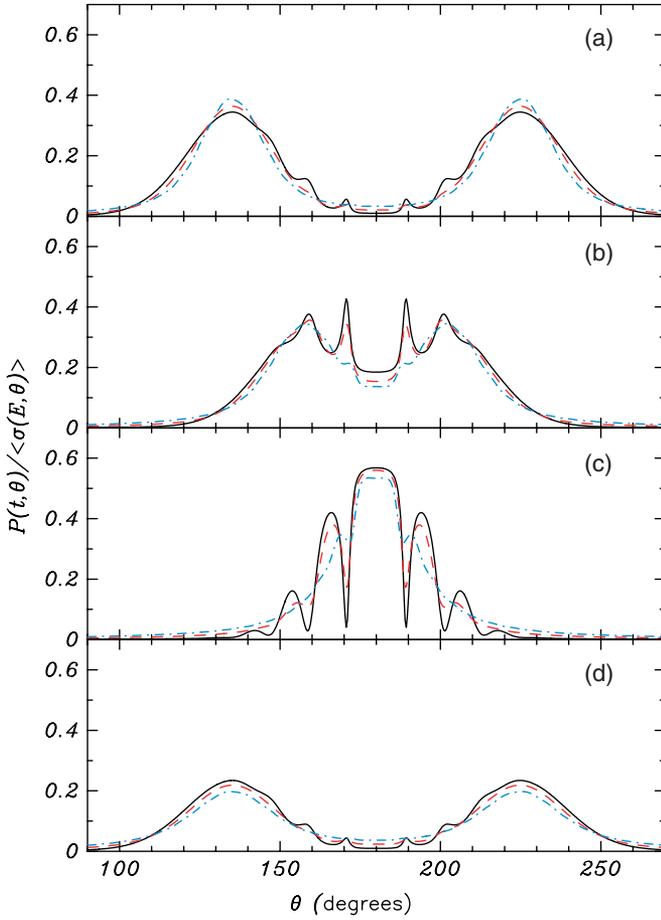


FIG. 1. (Color online) Angular dependence of the decay intensity of the highly excited intermediate complex at different moments of time for different combinations of β and d . Panel (a) corresponds to time $t = 3T/8$; (b) $t = 7T/16$; (c) $t = T/2$; (d) $t = 5T/8$. Here, T is the period of one complete revolution of the complex. Solid lines (black) correspond to $\beta = 0.01$ MeV and $d = 3$; dashed (red) to $\beta = 0.04$ MeV and $d = 4$; dashed-dotted (blue) to $\beta = 0.075$ MeV and $d = 8$. As β gets bigger, the interference fringe contrast becomes smaller.

In Fig. 1, we also plot $AP(t, \theta)/\langle \sigma(E, \theta) \rangle$ with the same constant A and the set of parameters as before, except for d and β . We consider two other cases: $d = 4$, $\beta = 0.04$ MeV, and $d = 8$, $\beta = 0.075$ MeV. One can see that when the wave packets overlap only slightly [panels (a) and (d)] one can hardly distinguish between the three combinations of d and β , even though β changes by almost one order of magnitude. On the other hand, from panels (b) and (c), we observe that a fringe contrast due to the interference of the wave packets is very sensitive to absolute values of β and d . This result is further illustrated in Fig. 2, where we plot $P(t, \theta)$ calculated for the three sets of β and d values, as a function of t for $\theta = 170.6^\circ$. One observes that the smaller β is, the deeper the minima are, because of the destructive interference between the wave packets at $t = T/2$. However, for $\theta = 180^\circ$, $P(t, \theta)$ in Fig. 2 is insensitive to the three sets of β and d .

Note that to reproduce the width of the wave packets in Figs. 1 and 2 for the maximal possible $d = I$ value, one has

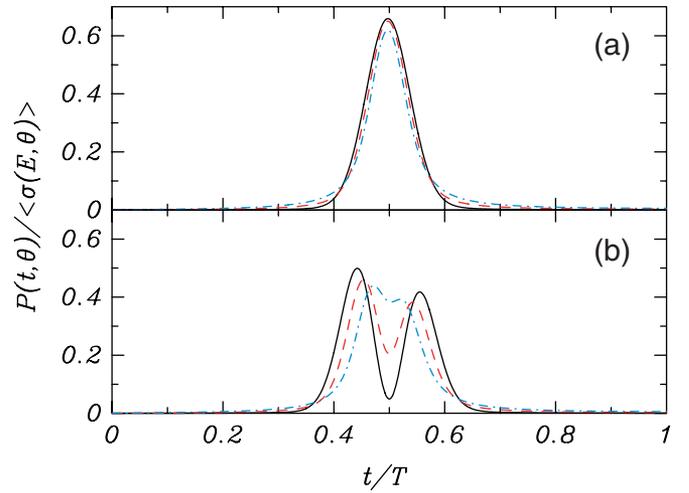


FIG. 2. (Color online) Time dependence of the decay intensity of the highly excited intermediate complex at (a) $\theta = 180^\circ$ and (b) $\theta = 170.6^\circ$, for the different sets of β and d . Solid lines (black) correspond to $\beta = 0.01$ MeV and $d = 3$; dashed (red) to $\beta = 0.04$ MeV and $d = 4$; dashed-dotted (blue) to $\beta = 0.075$ MeV and $d = 8$.

to take $\beta = 0.1$ MeV. Therefore, even the largest possible β value is more than one order of magnitude smaller than Γ_{spr} . The latter is usually estimated from the width of giant resonances [2,4]: $\Gamma_{\text{spr}} \simeq 5$ MeV. This indicates that energy relaxation, i.e., the process of formation of ergodic many-body configurations, in highly excited nuclear systems can be much faster than phase relaxation.

III. DETERMINATION OF THE TIME POWER SPECTRUM FROM DATA ON EXCITATION FUNCTION OSCILLATIONS

We consider a collision of the spinless reaction partners in the entrance and exit channels. The cross section of the collision is given by $\sigma(E, \theta) = |f(E, \theta)|^2$. Here E is the energy in the entrance channel and θ is the scattering angle. The reaction amplitude $f(E, \theta)$ is a linear combination of S -matrix elements $S^J(E)$ corresponding to the different total spin values J . Suppose that, for a fixed energy E_1 , a measurement of detailed angular distribution is performed. Then, using, e.g., the method [24], one can find $S^J(E_1)$ and, therefore, $f(E_1, \theta)$. Measuring the angular distributions for different energies on the energy interval $I = E_{\text{min}} - E_{\text{max}}$ with the energy step ΔE and energy resolution smaller or about the minimal characteristic energy scale of a variation of $f(E, \theta)$, one obtains detailed energy and angle dependencies of the collision amplitude. Its Fourier component,

$$\begin{aligned} \mathcal{P}(t, \theta) &\propto \int_{E_{\text{min}}}^{E_{\text{max}}} dE \exp(-iEt/\hbar) f(E, \theta) \\ &\approx \Delta E \sum_{n=0}^N \exp[-i(E_{\text{min}} + n\Delta E)t/\hbar] \\ &\quad \times f(E_{\text{min}} + n\Delta E, \theta), \end{aligned} \quad (2)$$

is the instantaneous decay amplitude received at a detector at time t with the time uncertainty of about \hbar/I , for a collision initiated at $t = 0$. In the sum in Eq. (2), $N = I/\Delta E$ and $(N + 1)$ is the number of equidistant energy values for which the detailed angular distributions should be measured. The summation in the r.h.s. of Eq. (2) may be changed to the integration if we use a linear interpolation for $f(E, \theta)$ in between the consequent energies E_n and E_{n+1} . This is justified provided the energy step ΔE is smaller or about the minimal characteristic energy scale of a variation of the cross section. For example, in Ref. [22] the energy step is $\Delta E_{c.m.} = 133$ keV, while the minimal characteristic scale of the variation of $\sigma(E, \theta = 180^\circ)$ is $\Gamma = 0.3$ MeV [16,17].

Having reconstructed $\mathcal{P}(t, \theta)$ from the excitation function data, one obtains the time power spectra $P(t, \theta) = |\mathcal{P}(t, \theta)|^2$ for any angle. The latter quantity is equivalent to that directly measured using real-time methods of femtochemistry, with the energy bands of I for the pump and probe laser pulses, to monitor chemical reaction dynamics [25,26]. Due to its dependence on θ , $P(t, \theta)$ determined by the proposed method could provide even more detailed information on the collision dynamics, as compared with the femtochemistry experiments, since latter measurements often allow one to obtain $P(t, \theta)$ averaged over θ .

For the energy interval I being long enough to provide a sufficient time resolution \hbar/I to resolve interference fringes [27], the proposed experiment would allow us to test our theoretical predictions. If these were confirmed by the data, then the experimentally determined $P(t, \theta)$ could be compared with our calculations presented in Figs. 1 and 2 with the purpose of evaluating β and d . In particular, the energy interval of 10.5 MeV, over which the excitation function for the $^{12}\text{C}+^{24}\text{Mg}$ elastic scattering at $\theta = 180^\circ$ was measured [22], should be sufficient to resolve the interference fringes in $P(t, \theta)$ [27].

It should be noted that the method proposed above allows the reconstruction of $P(t, \theta)$, which includes both potential scattering, i.e., fast processes, and the time-delayed mechanism. Since the previous analysis [16,17] demonstrated the presence of a strong potential scattering component, we expect that this method will yield a strong potential scattering contribution in $P(t, \theta)$. However, this contribution is expected to be restricted to relatively short times $t_{ps} \leq \hbar/\Delta E_{ps}$, where ΔE_{ps} is a characteristic energy interval for the variation of the potential scattering amplitude. For $\Delta E_{ps} \simeq 2-3$ MeV [18], $t_{ps} \simeq 3-2 \times 10^{-22}$ sec. Therefore, this potential scattering contribution is expected to be restricted to $t/T \leq 0.1$ in Fig. 2 and should not show up at all in Fig. 1.

The above method to obtain $P(t, \theta)$ from the data is model independent and, therefore, a most reliable one. However, it is very demanding experimentally since $\sigma(E, \theta)$ must be measured for a very large number of angles [24]. There is another way to extract $P(t, \theta)$ from the data even if the excitation function is measured for a single angle. This method can be applied if the direct reaction process provides a major contribution to the cross section [17]. We consider again a case of spinless collision partners in the entrance and exit channels. The main idea is based on the decomposition of the collision amplitude, $f(E, \theta) = f_{\text{dir}}(E, \theta) + \delta f(E, \theta)$. Here $f_{\text{dir}}(E, \theta)$

is the energy smooth amplitude corresponding to potential scattering or direct fast process, and $\delta f(E, \theta)$ is the energy fluctuating amplitude, $\langle \delta f(E, \theta) \rangle = 0$, corresponding to the time-delayed collision. Consider first an idealized case of energy-independent amplitude $f_{\text{dir}}(E, \theta)$ when it coincides with the energy-averaged amplitude $\langle f(E, \theta) \rangle$. The collision cross section has the form $\sigma(E, \theta) = |f(E, \theta)|^2 = \sigma_{\text{dir}}(E, \theta) + \sigma_{\text{fl}}(E, \theta) + 2\text{Re}[\delta f(E, \theta)f_{\text{dir}}(E, \theta)^*]$. Here, the potential scattering or direct reaction cross section $\sigma_{\text{dir}}(E, \theta) = |f_{\text{dir}}(E, \theta)|^2$ is energy independent, and $\sigma_{\text{fl}}(E, \theta) = |\delta f(E, \theta)|^2$. For a relative contribution of the potential scattering or direct reaction cross section to be about 70% or more of the total cross section, $\Delta\sigma(E, \theta) = \sigma(E, \theta) - \langle \sigma(E, \theta) \rangle \simeq 2\text{Re}[\delta f(E, \theta)f_{\text{dir}}(E, \theta)^*]$, where $\langle \sigma(E, \theta) \rangle = \sigma_{\text{dir}}(E, \theta) + \langle \sigma_{\text{fl}}(E, \theta) \rangle$ is the energy-averaged cross section. This means that rapid energy variations of the cross section originate mostly from interference between the energy fluctuating $\delta f(E, \theta)$ and the energy-independent amplitude $f_{\text{dir}}(E, \theta)$. Suppose that the excitation function is measured on the energy interval $I = E_{\text{max}} - E_{\text{min}}$ with energy step ΔE so that the total number of steps is $N + 1$, where $N = I/\Delta E$. Then, for $t > 0$, we have [17]

$$\begin{aligned} & \int_{E_{\text{min}}}^{E_{\text{max}}} dE \exp(-iEt/\hbar) \Delta\sigma(E, \theta) \\ & \propto \int_{E_{\text{min}}}^{E_{\text{max}}} dE \exp(-iEt/\hbar) \delta f(E, \theta) \\ & \approx \Delta E \sum_{n=0}^N \exp[-i(E_{\text{min}} + n\Delta E)t/\hbar] \\ & \quad \times \delta f(E_{\text{min}} + n\Delta E, \theta) \propto \mathcal{P}(t, \theta), \end{aligned} \quad (3)$$

with $P(t, \theta) = |\mathcal{P}(t, \theta)|^2$. In Eq. (3), we employed a causality condition, $\mathcal{P}(t < 0, \theta) = 0$. This condition implies that the molecule cannot decay before it is formed at $t = 0$. Again, the above information on $P(t, \theta)$ is equivalent to that obtained directly using real-time methods of femtochemistry to monitor unimolecular chemical reactions, with the energy bands of $I = (E_{\text{max}} - E_{\text{min}})$ for the pump and probe laser pulses [25].

The above consideration can be extended to a case when the potential scattering or direct reaction amplitude $f_{\text{dir}}(E, \theta)$ depends on energy, but this dependence is considerably smoother than the energy dependence of the amplitude $\delta f(E, \theta)$ for the time-delayed collision. We again assume that a relative contribution of direct processes into the energy-averaged cross section is about 70% or more. We define the characteristic energy interval I_d of a variation of $f_{\text{dir}}(E, \theta)$ with $I_d \gg \Gamma$, where Γ is the characteristic energy interval of variation of $\delta f(E, \theta)$. The energy interval I_d can be evaluated using, e.g., the trend reduction method [28]. In particular, for the $^{12}\text{C}+^{24}\text{Mg}$ scattering [22] analyzed in Sec. II, this interval was evaluated to be about 4–5 MeV [21], i.e., much greater than $\Gamma = 0.3$ MeV. Then one can find the best polynomial fit of the energy dependence of the cross section $\sigma(E, \theta) = \sigma_d(E, \theta) + \sigma_{\text{fl}}(E, \theta) + 2\text{Re}[\delta f(E, \theta)f_{\text{dir}}(E, \theta)^*]$ with the order of polynomial being $[I/I_d] + 1$, where $[I/I_d]$ is integer part of I/I_d . The resulting energy-smooth cross section is denoted as $\tilde{\sigma}(E, \theta) \approx \sigma_d(E, \theta) + \langle \sigma_{\text{fl}}(E, \theta) \rangle$ with $\langle \sigma_{\text{fl}}(E, \theta) \rangle$ being

the energy-averaged, i.e., energy-independent, cross section of the time-delayed processes. It can be found using the standard procedure employed for the analysis of Ericson fluctuations (see, e.g., Refs. [21,22]). As a result, one finds energy-smooth direct-reaction cross section $\sigma_d(E, \theta)$. Consider $\sigma(E, \theta) - \tilde{\sigma}(E, \theta) \simeq 2\text{Re}[\delta f(E, \theta) f_{\text{dir}}(E, \theta)^*]$. Assume

first that energy dependence of the phase of $f_{\text{dir}}(E, \theta)$ is negligible, $\arg[f_{\text{dir}}(E, \theta)] = \text{constant}$, and the smooth energy dependence of $\sigma_d(E, \theta)$ is due to the energy dependence of $|f_{\text{dir}}(E, \theta)| = \sigma_d(E, \theta)^{1/2}$. Then, applying the same arguments as those used to obtain Eq. (3), for $t > 0$, we have

$$P(t, \theta) \propto \sum_{n=0}^N \frac{\exp[-i(E_{\text{min}} + n\Delta E)t/\hbar][\sigma(E_{\text{min}} + n\Delta E, \theta) - \tilde{\sigma}(E_{\text{min}} + n\Delta E, \theta)]}{\sigma_d(E_{\text{min}} + n\Delta E, \theta)^{1/2}} \quad (4)$$

with $P(t, \theta) = |\mathcal{P}(t, \theta)|^2$.

To generalize Eq. (4) by taking into account the energy dependence of $\arg[f_{\text{dir}}(E, \theta)] \equiv \phi(E, \theta)$ ($f_{\text{dir}}(E, \theta) = |f_{\text{dir}}(E, \theta)|\exp[i\phi(E, \theta)]$), we use the linear approximation $\phi(E, \theta) = \phi(\bar{E}, \theta) + (E - \bar{E})d\phi(E, \theta)/dE|_{E=\bar{E}}$, where $\bar{E} = (E_{\text{max}} - E_{\text{min}})/2$ and $d\phi(E, \theta)/dE|_{E=\bar{E}} = t_{\text{dir}}/\hbar$ with $t_{\text{dir}} \ll \hbar/\Gamma$ being the time delay due to the potential scattering or direct reactions [29]. Then, it is easy to see that for $t > t_{\text{dir}}$, the r.h.s. of Eq. (4) is changed to $\mathcal{P}(t - t_{\text{dir}}, \theta)$. This manifestly demonstrates that the interference between time-delayed and fast direct processes in the cross section is a precondition for monitoring the time evolution; the fast process switches on the clock at the initial moment of time t_d playing the role of the pump pulse. In the absence of direct processes, the initial moment of time is not defined.

Note that $P(t, \theta)$ can also be expressed as a cosine half-Fourier transform of the cross section energy autocorrelation function $C(\varepsilon, \theta)$, provided the relative contribution of potential scattering is larger than 70% [16,17]. In Fig. 3, we plot $C(\varepsilon, \theta)/C(\varepsilon = 0, \theta) = [\int_0^\infty dt \cos(\varepsilon t/\hbar)P(t, \theta)]/(\sigma(E, \theta))$ for (a) $\theta = 180^\circ$ and (b) $\theta = 170.6^\circ$, for the three combinations of β and d . Since the $P(t, \theta = 180^\circ)$ are close for the different sets of β and d , the corresponding $C(\varepsilon, \theta = 180^\circ)$ can hardly be distinguished reliably. This is the reason that the analysis at $\theta = 180^\circ$ [16,17] is not sufficient to determine unambiguously the values of β and d . However, $C(\varepsilon, \theta = 170.6^\circ)$ is more sensitive to the different sets of β and d . In particular, for $\varepsilon \geq 4$ –5 MeV, the oscillations in $C(\varepsilon, \theta = 170.6^\circ)$ for $\beta = 0.01$ MeV, $d = 3$ and $\beta = 0.075$ MeV, $d = 8$ are out of phase, with the absolute value of their difference being up to 0.4. Still, from the comparison of Figs. 1(c), 2(b), and 3, we observe that the sensitivity of $P(t, \theta)$ to different values of β and d is considerably stronger than the sensitivity of $C(\varepsilon, \theta)$ to these values. Also $P(t, \theta)$ obtained using Eqs. (3) and (4) does not acquire statistical errors due to the finite energy interval I . Indeed, such a method provides information on $P(t, \theta)$ equivalent to that obtained in the femtochemistry experiments. Therefore, the only uncertainty of this method is a finite time resolution, $\Delta t \approx \hbar/I$, which is the uncertainty in the femtochemistry experiments with the finite energy band I of the pump and probe laser pulses [25,26]. On the other hand, a reconstruction of $P(t, \theta)$ from $C(\varepsilon, \theta)$ may result in additional errors due to possible statistical

uncertainties in $C(\varepsilon, \theta)$ related to the finite energy range I [30]. Such additional statistical uncertainties may not be excluded even though the effects studied in this paper can clearly not be associated with Ericson fluctuations. Indeed, unlike, e.g., oscillating structures in Fig. 3, Ericson theory predicts angle-independent Lorentzian shapes for the $C(\varepsilon, \theta)$. Therefore, for the reasons described above, we suggest that a reconstruction of $P(t, \theta)$ directly from the excitation functions [Eqs. (3) and (4)] is more reliable than from $C(\varepsilon, \theta)$.

IV. FROM HEAVY-ION COLLISIONS TO BIMOLECULAR CHEMICAL REACTIONS

An important question is whether the rotating wave packets and their interference can occur in colliding systems for

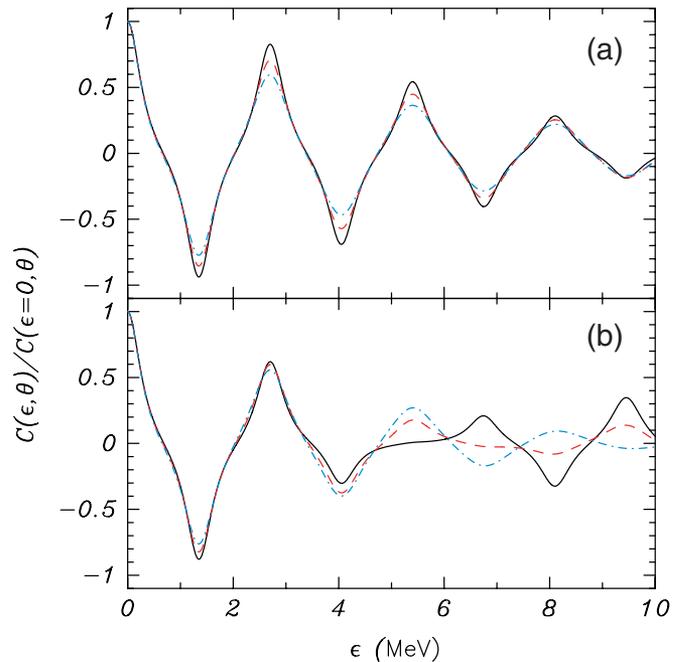


FIG. 3. (Color online) Cross section energy autocorrelation functions calculated at (a) $\theta = 180^\circ$ and (b) $\theta = 170.6^\circ$ for the different sets of β and d . Solid lines (black) correspond to $\beta = 0.01$ MeV and $d = 3$; dashed (red) to $\beta = 0.04$ MeV and $d = 4$; dashed-dotted (blue) to $\beta = 0.075$ MeV and $d = 8$.

which more rigorous approaches can be implemented. The encouraging answer has been given by the calculations of the time evolution of the $H + D_2 \rightarrow HD + D$ bimolecular chemical reaction [31–33]. The evidence for rotational wave packets has been demonstrated (see, e.g., Fig. 3 in [31]) because of the interference of the overlapping resonances [34] of the quasibound complex with $J = 15\text{--}20$ total spin values (i.e., within our approach, $d \approx 5$). In this peripheral time-delayed collision, the rotational wave packets are stable showing a moderate spreading during half a period of rotation. Within our description, this reveals a small β value in the time-delayed peripheral reaction mechanism, i.e., $\pi\beta/\hbar\omega \ll 1/d$. Then, in accordance with our prediction, the coherent superposition of the rotating wave packets should produce a strong interference contrast. This is indeed the case as clearly seen in Fig. 3 of Ref. [31]. Similar effects of the stable rotating wave packets and their coherent superpositions with strong interference contrast have also been found in the calculations for $F + HD \rightarrow HF + D$ bimolecular reaction [35,36].

The above results should allow us to test our suggestion for the determination of the phase-relaxation time and, in general, for the time evolution of the collision in bimolecular chemical reactions. For example, in [32] a total time-dependent amplitude was calculated for the $H + D_2 \rightarrow HD + D$ reaction. Then it was split into a short-time direct reaction component and the time-delayed one. Taking the half-Fourier transform, explicit energy and angle dependencies of both the direct and time-delayed amplitudes were obtained. It was found that in the region of backward angles $\theta \leq 70^\circ$ and for the c.m. energy interval 1.6–2 eV, the direct reaction cross section is considerably bigger than the time-delayed one. Then one can apply our method (Sec. III) to obtain $P(t, \theta)$ of the time-delayed reaction mechanism from the energy dependence of the double-differential cross sections. This extracted $P(t, \theta)$ can be compared with its actual exact form since the latter is known and was used to generate the energy dependence of the double-differential cross sections.

The calculations [32,33] suggest that in the forward angle range, where the Schrödinger cat states originated from the interference of the rotating toward each other wave packets are observed, contribution of the direct reactions is negligible. Therefore, for this particular $H + D_2 \rightarrow HD + D$ reaction for the c.m. energy less than 2 eV, our method cannot be applied. Yet one can still check the sensitivity of the method by artificially adding an energy-smooth background amplitude to the calculated one for the time-delayed reaction mechanism. The energy-smooth amplitude would play a role of the direct reaction amplitude. This would enable one to directly test the accuracy of the determination of the interference fringe contrast from the data on the double differential cross sections generated with the artificially added energy-smooth direct reaction amplitude. Such a test may be important since for the higher energy and/or for some other bimolecular chemical reactions, a strong contribution of direct reactions is possible for the same angle range at which the Schrödinger cat states appear.

Note that pure energy resolution measurements of the cross sections of bimolecular chemical reactions, in particular

$H + D_2 \rightarrow D + HD$ [37] and $F + HD \rightarrow HF + D$ [38] reactions, have recently become possible. Our results suggest that this should enable experimentalists to extract information on time dependence, in particular on the rotational coherent dynamics and possible wave packet interference, of the bimolecular chemical reactions which previously could only be studied by the pump probe laser pulses technique to monitor unimolecular chemical reactions [25,26].

V. PHASE RELAXATION VERSUS DECOHERENCE

Since phase relaxation results in washing out the interference fringes and eventually, for a sufficiently large β , it can destroy the Schrödinger cat states of Figs. 1 and 2, this effect appears to be quite similar to decoherence [39]. Yet, there is an essential difference between the two effects. It is instructive to compare the extreme case of very fast phase relaxation (very large β) with very fast decoherence. Very fast phase relaxation corresponds to the regime of random matrix theory, i.e., to random phase relations between all partial width amplitudes corresponding to different strongly overlapping resonance states [4]. This yields angle-independent exponential form for the time power spectrum $P(t, \theta)$ and unity for the normalized variance of the fluctuating cross section. Analogous to the fast phase relaxation would be quick decoherence between all many-body quasistationary resonance states, similar to the decoherence of Fock states due to the coupling of the system to a phase reservoir [39]. Such a decoherence would destroy interference terms between all the many-body eigenstates, leading to the vanishing of interference between different partial width amplitudes. It can be shown that in this case, the time power spectrum would have the same angle-independent exponential form as for fast phase relaxation. However, in contrast to fast relaxation, quick decoherence would result in washing out the cross section energy fluctuations, reducing its normalized variance to $D/\Gamma \ll 1$ [40]. It should be noted that the suppression of electron transmission intensity fluctuations through nanostructures due to dephasing, an effect similar to decoherence, can be described on the basis of different arguments allowing the evaluation of dephasing rates from the amplitude of conductance fluctuations [41].

VI. DISCUSSION AND CONCLUSIONS

Our approach is based on a new idea of slow phase relaxation between partial width amplitudes in a regime of strongly overlapping resonances. Contrary to the conventional idea of random phase relations in this regime [4,42,43], our analysis invokes spin off-diagonal phase correlations and their slow decay for resonance time-delayed collisions. Though these effects may occur for atomic cluster collisions [19] and are already revealed for the bimolecular chemical reactions [31–33,35,36], the present consideration is essentially motivated by the pure energy resolution data on fluctuations in $^{12}\text{C}+^{24}\text{Mg}$ elastic and inelastic scattering at $\theta = 180^\circ$ [21,22], demonstrating both the fine energy structure of $\simeq 0.3$ MeV and quasiperiodicity with period $\simeq 3$ MeV [16,17]. It is these peculiar features that led us to the interpretation of the $^{12}\text{C}+^{24}\text{Mg}$ scattering in terms of highly excited

quasimolecular states with strongly overlapping resonances, stable rotational wave packets, and strong sensitivity of their coherent superpositions to the phase-relaxation width. Note that quasiperiodic structures in the excitation functions are present also for other heavy-ion colliding systems [15]. We have also described thoroughly how to extract the time power spectrum from the data on the excitation function oscillations.

It should be mentioned that a conceptually different optical model description of the $^{12}\text{C}+^{24}\text{Mg}$ elastic scattering at backward angles was presented in Ref. [44], where the quasimolecular structures were ruled out. This conclusion was based on a qualitative description of angular oscillations at backward angles for only three energies within the same energy range which is the subject of our study in this paper. Unfortunately, that analysis is inconsistent with the presence of the fine energy structure of ≈ 0.3 MeV at $\theta = 180^\circ$ [21], which unambiguously reveals the presence of time-delayed processes at backward angles. On the contrary, the optical model description relies on the energy average S -matrix and therefore is expected to reproduce broad $\simeq 2\text{--}3$ MeV energy structures in the cross section. From our point of view, the fact that the presence of the fine energy structures was not taken into account led to an incorrect qualitative and then quantitative

identification of the physical picture of the collision process in Ref. [44].

In conclusion, we have demonstrated a strong sensitivity of Schrödinger cat states to the phase-relaxation width in complex quantum collisions. This should permit to determine the phase-relaxation time from measurements of the excitation functions for $^{12}\text{C} + ^{24}\text{Mg}$ elastic scattering at backward angles. Such an experiment would be desirable since the Schrödinger cat states predicted in [19] for $^{12}\text{C}+^{24}\text{Mg}$ scattering involve $\sim 10^3\text{--}10^4$ many-body configurations of the highly excited intermediate complex. To the best of our knowledge, the internal interactive complexity of these quantum macroscopic superpositions dramatically exceeds [19] all those previously experimentally realized. The proposed method can also be applied for determination of the phase-relaxation time from the data on excitation functions for bimolecular chemical reactions.

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