

Quantum-classical transition for an analog of the double-slit experiment in complex collisions: Dynamical decoherence in quantum many-body systems

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We study coherent superpositions of clockwise and anticlockwise rotating intermediate complexes with overlapping resonances formed in bimolecular chemical reactions. Disintegration of such complexes represents an analog of a famous double-slit experiment. The time for disappearance of the interference fringes is estimated from heuristic arguments related to fingerprints of chaotic dynamics of a classical counterpart of the coherently rotating complex. Validity of this estimate is confirmed numerically for the H+D₂ chemical reaction. Thus we demonstrate the quantum-classical transition in temporal behavior of highly excited quantum many-body systems in the absence of external noise and coupling to an environment.

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The famous double-slit experiment [1] provides the most vivid demonstration of quantum coherent superpositions. These manifest themselves in interference fringes of the intensity due to the interference between the matter waves emerging from different slits. The double-slit experiments have successfully demonstrated the wave nature of, e.g., electrons, neutrons, atoms, small molecules, noble gas clusters, and fullerenes [1]. A process of converting coherent superpositions into a classical sum of intensities manifests itself in the disappearance of the interference fringes. This fundamental physical process of the emergence of classical dynamics from the quantum-mechanical description is referred to as the quantum-classical transition (QCT).

There are two possible roots to describe QCT. The first one is decoherence [2] due to the external noise or coupling to the environment. This mechanism of QCT results from a nonunitary evolution of the system. The QCT due to the decoherence for the double-slit experiment with fullerenes has been demonstrated [3]. On the contrary, dynamical decoherence [4] describes the QCT without coupling to environment and only due to the intrinsic unitary evolution of a pure quantum state [5]. This process, unlike decoherence [2], describes the QCT on a finite time scale shorter than Heisenberg time, which diverges in the macroscopic limit [4].

In this paper we address the problem of quantum-classical transition in the absence of any coupling to the environment, revealing an effect analogous to dynamical decoherence [4,5]. We focus on the QCT in a temporary quantum evolution. This problem is motivated by the work [5], where time-integration appeared to be a precondition for the quantum-classical transition. However, without disappearance of the interference fringes at a fixed moment of time the QCT is clearly incomplete. This raises the question whether dynamical decoherence [4] can lead to a QCT in a way that decoherence does [2].

Instead of a single-particle problem [5], we consider coherent superpositions of clockwise and anticlockwise rotat-

ing many-body intermediate complexes (ICs) with strongly overlapping resonances. Such ICs can be created in atomic cluster collisions, bimolecular chemical reactions, and heavy-ion collisions. The physical picture of rotational wave packets and their interference, originally revealed for heavy-ion collisions [6–8], has been strongly supported by numerical calculations for, e.g., the H+D₂ [9], F+HD [10], and He+H₂⁺ [11] state-to-state chemical reactions. The many-body aspect is of primary importance for the QCT since the macroscopic world consists of complex systems. We demonstrate here that isolated quantum systems can undergo a QCT and we obtain the characteristic time for such a transition.

We consider the spin-less collision partners in the entrance (*a*) and exit (*b*) channels. The energy fluctuating *S*-matrix elements are taken in the pole form [12]. This is a good approximation in the regime of overlapping resonances of the IC, $\Gamma/D \gg 1$, where Γ is the total width of resonance levels and D is average level spacing of the IC, provided $n_{ch} \gg \Gamma/D$ with n_{ch} being a number of open channels [12]. The high excitation energy of the IC, the smallness of the average level spacing of the IC, the strong overlap of the resonance levels, and the condition $n_{ch} \gg 1$ imply that we are deeply in the semiclassical region. In this regime we expect the quantum evolution to reveal fingerprints of classical dynamics of the classical counterpart of the system. We shall see that this is indeed the case.

Under the above stated conditions the time power spectrum for the time-delayed collision has the form $\Pi(t, \theta) = H(t) \exp(-t/t_H) |\mathcal{P}(t, \theta)|^2$, where

$$\mathcal{P}(t, \theta) = \frac{1}{N^{1/2}} \sum_J (2J+1) W(J)^{1/2} \exp(i\Phi_J) P_J(\theta) \times \sum_{\mu} \tilde{c}_{\mu}^J \exp(-iE_{\mu}^J t/\hbar). \quad (1)$$

Here, t is the time, θ is the scattering angle, J is the total spin

of the IC, and $H(t)$ is the Heaviside step function. The deflection angle Φ is given by the first J derivative of a sum of the potential phase shifts in the entrance and exit channels, and the $P_J(\theta)$ are Legendre polynomials. In the $\Pi(t, \theta)$, the factor $\exp(-t/t_l)$ accounts for the finite lifetime, $t_l = \hbar/\Gamma$, of the IC. Therefore its energy levels have a finite width, \hbar/t_l , corresponding to the continuous spectrum. On the other hand, even though the resonance levels are strongly overlapping, $P(t, \theta) = |\mathcal{P}(t, \theta)|^2$ describes the time evolution in discrete spectrum. Note that while $P(t, \theta)$ is the probability, except for the omitted factor $\exp(-t/t_l)$, for the IC to decay into an angle θ at the moment t , it also describes the time evolution of the angular orientation of the IC [13]. In Eq. (1), $\tilde{c}_\mu^J = c_\mu^J / [(c_\mu^J)^2]^{1/2}$, $c_\mu^J = \gamma_\mu^{J^a} \gamma_\mu^{J^b} - \gamma_\mu^{J^a} \gamma_\mu^{J^b}$ ($\overline{c_\mu^J} = 0$), $\gamma_\mu^{J^a(b)}$ and $\gamma_\nu^{J^a(b)}$ are real partial width amplitudes, and $E_\mu^J, E_\nu^{J'}$ are the resonance energies. The overbars stand for averaging over resonance states. The number of resonance states included in the sum in Eq. (1) is $N = \mathcal{I}/D \gg 1$, where the energy interval \mathcal{I} should be sufficiently long to resolve the interference fringes [14]. In Eq. (1), $W(J) = \langle |\delta S^J(E)|^2 \rangle$ is the average partial reaction probability taken in the Gaussian form, $\exp[-(J-I)^2/d^2]$, where I is the average total spin of the IC and d is the J -window width.

The intensity $|\mathcal{P}(t, \theta)|^2$ includes the sums $\frac{1}{N} \sum_{\mu\nu} \tilde{c}_\mu^J \tilde{c}_\nu^{J'} \exp[i(E_\nu^{J'} - E_\mu^J)t/\hbar]$. We calculate these sums by performing first a partial (μ, ν) -summation with $(E_\mu^J - E_\nu^{J'})$ fixed within an uncertainty of about a few units of D . Then, we get instead of the product $\tilde{c}_\mu^J \tilde{c}_\nu^{J'}$ its average over resonances, $\overline{\tilde{c}_\mu^J \tilde{c}_\nu^{J'}}$. The latter is given [6] by

$$\overline{\tilde{c}_\mu^J \tilde{c}_\nu^{J'}} = \frac{(1/\pi)D\beta|J - J'|}{[E_\mu^J - E_\nu^{J'} - \hbar\omega(J - J')]^2 + \beta^2(J - J')^2}, \quad (2)$$

where $\beta \gg D$ is the phase relaxation width and ω is the angular velocity of the coherent rotation of the highly excited IC. The spin diagonal contribution is evaluated by performing first a partial (μ, μ') -summation with fixed $(\mu - \mu')$ value. This yields $\overline{\tilde{c}_\mu^J \tilde{c}_{\mu'}^{J'}} = \delta_{\mu\mu'}$. The above approximation is justified if the time for formation of the many-body eigenstates of the effective Hamiltonian of the IC originated from the intramolecular energy relaxation due to, e.g., a strong anharmonic coupling between the normal vibrational modes, is faster than a characteristic rotation time of the IC. The diagonal approximation, for the states with the same J -values, is the standard assumption in the theory of quantum chaotic scattering [15]. The employed approximations imply that instead of the products $\tilde{c}_\mu^J \tilde{c}_\nu^{J'}$ we can use their average over resonances. This substitution does not violate the time quasiperiodicity of $|\mathcal{P}(t, \theta)|^2$, which is characteristic of a unitary evolution, since it is still given by a discrete superposition of a finite number of Fourier components with discrete frequencies $(E_\mu^J - E_\nu^{J'})/\hbar$.

We first calculate the sums

$$\frac{1}{N} \sum_{\mu\nu} \overline{\tilde{c}_\mu^J \tilde{c}_\nu^{J'}} \exp[i(E_\nu^{J'} - E_\mu^J)t/\hbar] \quad (3)$$

in the macroscopic limit $\hbar \rightarrow 0$. Since $D \propto \hbar^f$, where f is a number of degrees of freedom, the macroscopic limit corre-

sponds to a continuous spectrum approximation. Changing from the summation to integration in Eq. (3) we obtain

$$\exp[-i\omega t(J - J')] \exp[-\beta t|J - J'|/\hbar]. \quad (4)$$

Therefore the continuous spectrum approximation results in an irreversible decay of the spin off-diagonal correlations implying a nonunitary nonperiodic time evolution. In order to preserve a unitary time-periodic evolution we must evaluate the sums (3) for a discrete spectrum. We do so using first the equidistant spectrum approximation. Such an approximation is justified for $t \ll 2\pi\hbar/D$, when the spectrum is not resolved and thus the detailed spectral properties do not affect the time behavior. Then, for $\mathcal{I} \gg \hbar\omega, \beta$, the original sums (3) are reduced to

$$(1/\pi)D\beta|J - J'| \sum_{k=-\infty}^{\infty} \frac{\exp(-ikDt/\hbar)}{[kD - \hbar\omega(J - J')]^2 + \beta^2(J - J')^2}$$

with k being integers. We calculate these sums using the Poisson summation formula and obtain

$$\sum_{M=-\infty}^{\infty} \exp[-2\pi(\beta/D)|J - J'| |M - Dt/2\pi\hbar|] \times \exp[i2\pi(\hbar\omega/D)(J - J')(M - Dt/2\pi\hbar)], \quad (5)$$

where M are integers. The above expression is periodic in time, with the period $2\pi\hbar/D$, resulting in a periodicity of $|\mathcal{P}(t, \theta)|^2$ with this same period. However, we are interested in the time evolution of the IC with strongly overlapping resonances corresponding to relatively short time intervals $0 \leq t \ll 2\pi\hbar/D$. Then, the above sums are dominated by the $M=0$ term since, for $\beta \gg D$, the $M \neq 0$ terms are exponentially small and can be neglected. Then, the $M=0$ term equals the expression (4) and we observe that, for $t < \pi\hbar/D$, the continuous and equidistant spectrum approximations yield identical results. Yet, while the continuous spectrum approximation violates a unitary periodic in time evolution, the equidistant spectrum approximation obviously leads to a unitary time-periodic evolution. We have also calculated the sums (3) numerically with E_μ^J and $E_\nu^{J'}$ discrete spectra being uncorrelated for $J \neq J'$ and each having the universal spectral fluctuations of random matrix theory (Wigner-Dyson statistics [16]). We have found that, for $D \ll \beta$ and $t \ll 2\pi\hbar/D$, these sums are accurately given by the expression (4), i.e., by the $M=0$ term in Eq. (5). Again, this is because for $t \ll 2\pi\hbar/D$ the spectrum is not resolved and the sums (3) are insensitive to the detailed spectral properties. Altogether, for $t \ll 2\pi\hbar/D$, we have

$$P(t, \theta) \propto \sum_{JJ'} (2J + 1)(2J' + 1) [W(J)W(J')]^{1/2} \times P_J(\theta) P_{J'}(\theta) \exp[i(\Phi - \omega t)(J - J') - \beta|J - J'|t/\hbar]. \quad (6)$$

Obviously, the above result can also be obtained within the continuous spectrum approximation which is valid only for $t \leq \hbar/D$ when the spectrum is not resolved. Clearly the appearance of the decaying factor $\exp(-\beta|J - J'|t/\hbar)$ for t

$< \hbar/D$ does not violate the unitary character of evolution of the IC. The problem of experimental determination of $P(t, \theta)$ has been discussed in [14,17].

We employ the near-side far-side decomposition [11] of the collision amplitude, $\mathcal{P}(t, \theta) = \mathcal{P}^{(-)}(t, \theta) + \mathcal{P}^{(+)}(t, \theta)$, so that $P(t, \theta) \propto |\mathcal{P}^{(+)}(t, \theta)|^2 + |\mathcal{P}^{(-)}(t, \theta)|^2 + 2 \text{Re}[\mathcal{P}^{(+)}(t, \theta)\mathcal{P}^{(-)}(t, \theta)^*]$. The amplitudes $\mathcal{P}^{(\pm)}(t, \theta)$ have the form of Eq. (1) but with $Q_J^{(\pm)}(\theta) = (1/2)[P_J(\theta) \mp (2i/\pi)Q_J(\theta)]$ instead of $P_J(\theta)$. Here $Q_J(\theta)$ are Legendre functions of the second kind of degree J and the traveling Legendre functions have the semiclassical asymptotics $Q_J^{(\pm)}(\theta) \sim [1/2\pi J \sin \theta]^{1/2} \exp\{\pm i[(J+1/2)\theta - \pi/4]\}$.

It is expected that the quantum superpositions originate from the interference between the $\mathcal{P}^{(-)}(t, \theta)$ and $\mathcal{P}^{(+)}(t, \theta)$ in a close vicinity of $\theta=0, \pi$, where the wave packets overlap. However, for these angle ranges, the asymptotic form of $Q_J^{(\pm)}(\theta)$ is not valid. Yet, t_{q-cl} may be conjectured from the time dependence of the near-side and far-side intensities, $|\mathcal{P}^{(\pm)}(t, \theta)|^2$, for the intermediate angle interval, where the asymptotic form of $Q_J^{(\pm)}(\theta)$ is an accurate approximation.

We calculate $|\mathcal{P}^{(\pm)}(t, \theta)|^2$ using the same approximations as those employed for the calculation of $P(t, \theta)$. The $|\mathcal{P}^{(\pm)}(t, \theta)|^2$ have the form of Eq. (6) but with the asymptotics of $Q_J^{(\pm)}(\theta)$ instead of $P_J(\theta)$. Calculations analogous to those of [6] yield that, for $t \ll \hbar/d\beta$, $|\mathcal{P}^{(\pm)}(t, \theta)|^2$ describe Gaussian rotational wave packets having a constant width (angular dispersion) $\approx 1/d$. Thus for $t \ll \hbar/d\beta$, there is no spreading of the wave packets and the memory about their width at $t=0$ remains. This is characteristic of purely quantum integrable-like evolution while the classical counterpart of the many-body IC is clearly chaotic. On the contrary, for $t \gg \hbar/d\beta$, the width of the wave packets becomes much greater than that at $t=0$ and the memory about the angular dispersion at $t=0$ is lost. In addition, for $t \gg \hbar/d\beta$ and $\beta t/\hbar > 1$, $|\mathcal{P}^{(\pm)}(t, \theta)|^2$ approach a uniform distribution exponentially fast [6], $|\mathcal{P}^{(\pm)}(t, \theta)|^2 \propto \exp[2 \cos(\Phi \pm \theta - \omega t)/\exp(\beta t/\hbar)]$. We conjecture that this long-time behavior may be considered as a fingerprint of chaotic dynamics of the classical counterpart of the IC. Since the crossover from the integrablelike to the chaoticlike dynamics of the quantum evolution occurs between $t_1 \ll \hbar/\beta d$ and $t_2 \gg \hbar/\beta d$, the characteristic time for the QCT can be evaluated as $t_{q-cl} \sim \hbar/\beta d$; but then another feature of the QCT should be the disappearance of the interference between $\mathcal{P}^{(+)}(t, \theta)$ and $\mathcal{P}^{(-)}(t, \theta)$. This transformation of the quantum superposition into an incoherent sum of the intensities should also happen at $t_{q-cl} \sim \hbar/\beta d$.

To test this we use Eq. (6) to describe rotational wave packets and their interference revealed [9] for the chemical reaction $\text{H} + \text{D}_2(v_i=0, j_i=0) \rightarrow \text{HD}(v_f=3, j_f=0) + \text{D}$, where v_i, j_i and v_f, j_f are vibrational and rotational quantum numbers for the initial and final states, respectively. In this reaction, the wave packets likely originate from interference of the overlapping resonances of the IC [18]. From the numerical calculations [9] we deduce $\Phi = 135^\circ$ and $\hbar\omega = 0.045$ eV. It was found that the main contribution to the time-delayed reaction mechanism comes from the total angular momenta $J=15-20$. Accordingly, we choose $I=18$. To reproduce the width of the wave packets, $\approx 25^\circ$, at an early stage, when

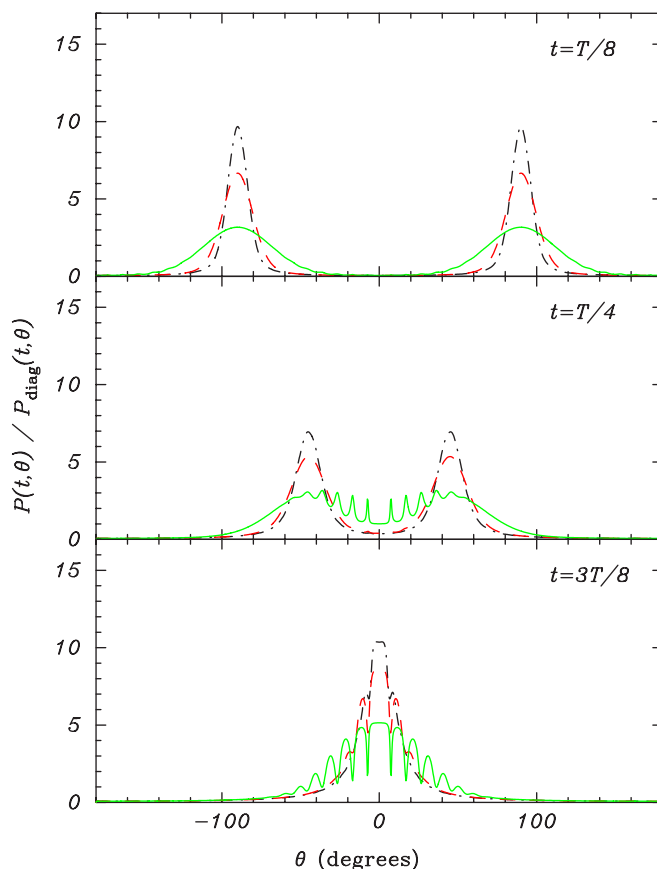


FIG. 1. (Color online) Time power spectra related to the $\text{H} + \text{D}_2(v_i=0, j_i=0) \rightarrow \text{HD}(v_f=3, j_f=0) + \text{D}$ chemical reaction for different moments of time. Solid lines correspond to $d=2$ and $\beta=0.003$ eV, dashed lines $d=5$ and $\beta=0.003$ eV, and dashed-dotted lines $d=10$ and $\beta=0.003$ eV.

$\theta \approx \Phi$, we take $d=2$. To account for the strong interference contrast in the forward direction we choose $\beta=0.003$ eV.

In Fig. 1 we present $P(t, \theta)/P_{diag}(\theta)$, where $P_{diag}(\theta)$ is a time-independent quantity given by the diagonal $J=J'$ terms in Eq. (6). The reason for this scaling is that amplitudes of individual wave packets are strongly and abruptly enhanced in the vicinity of $\theta=0, \pi$ due to the azimuthal symmetry of the problem [8]. However, $P_{diag}(\theta)$ has the similar enhancement for $\theta \sim 0, \pi$, so that the quantity $P(t, \theta)/P_{diag}(\theta)$ permits us to probe the interference between the wave packets whose amplitudes depend smoothly on θ . Another advantage of this scaling is that $P_{diag}(\theta)$ corresponds to the very quick phase-relaxation ($\hbar/\beta \rightarrow 0$) yielding the universal limit of the theory of quantum chaotic scattering [15]. In this limit, wave packets are uniformly spread and their interference is completely destroyed due to the absence of the spin off-diagonal contributions. Thus a deviation of $P(t, \theta)/P_{diag}(\theta)$ from a constant unity is a quantitative measure of the deviation of the collision process from the universal limit of the quantum chaotic scattering theory.

In Fig. 1 we present $P(t, \theta)/P_{diag}(\theta)$ for three moments of time. One can see that the wave packets, initially oriented along $\theta=\Phi, 2\pi-\Phi$, rotate towards each other. Interference fringes are produced when the wave packets start to overlap.

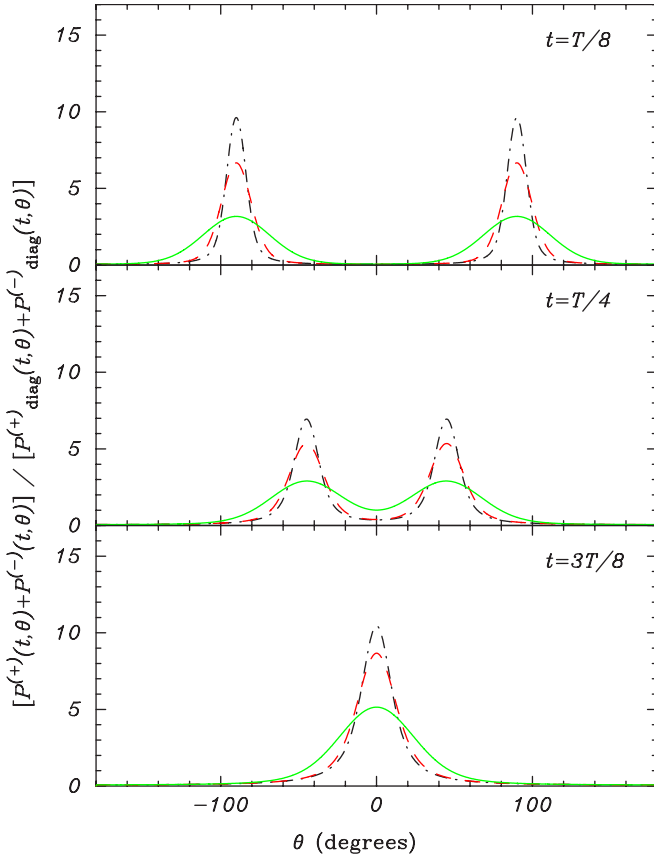


FIG. 2. (Color online) Classical limits for the time power spectra presented in Fig. 1 (see text).

For $d=2$, $\beta=0.003$ eV we have $t_{q-cl} \approx \hbar/\beta d = 220$ fs. This time is longer by about a factor of 6.5 than the longest time $3T/8$ in Fig. 1, where $T=2\pi/\omega=90$ fs. We observe a strong contrast of the interference fringes. Now we decrease t_{q-cl} by taking $d=5$ and keeping β the same. Then, for $t=3T/8 \approx 0.38t_{q-cl}$, the interference fringes are suppressed but still visible in the bottom panel of Fig. 1. We further decrease t_{q-cl} by taking $d=10$ with the value of β unchanged. Then, for $t=3T/8 \approx 0.77t_{q-cl}$, the interference fringes practically disappear manifesting a transformation of the coherent superpositions into the incoherent mixture of the intensities. Thus we have confirmed the validity of our estimate for the t_{q-cl} . This estimate has also been confirmed by calculations where we have increased β keeping $d=2$ fixed.

To test that the interference fringes do originate from the interference between the $\mathcal{P}^{(+)}(t, \theta)$ and $\mathcal{P}^{(-)}(t, \theta)$ we calculate $P^{(+)}(t, \theta) + P^{(-)}(t, \theta)$, where $P^{(\pm)}(t, \theta) = |\mathcal{P}^{(\pm)}(t, \theta)|^2$. We employ the same approximations used in calculating $P(t, \theta)$, Eq. (6). We obtain $P^{(\pm)}(t, \theta)$ having the same form as Eq. (6) but with $Q_j^{(\pm)}(\theta)Q_{j'}^{(\pm)*}(\theta)$ instead of $P_j(\theta)P_{j'}(\theta)$. In Fig. 2 we present $[P^{(+)}(t, \theta) + P^{(-)}(t, \theta)] / [P_{diag}^{(+)}(t, \theta) + P_{diag}^{(-)}(t, \theta)]$ for the same values of parameters as used in Fig. 1. The time independent quantity $[P_{diag}^{(+)}(t, \theta) + P_{diag}^{(-)}(t, \theta)]$ contains the diago-

nal $J=J'$ terms only. The reason for this scaling is the same as that employed in Fig. 1. In addition, $Q_j^{(\pm)}(\theta)$ have a logarithmic divergence at $\theta=0, \pi$ resulting in an unphysical increase of $P^{(\pm)}(t, \theta)$ in a close vicinity of $\theta=0, \pi$. The presentation in Fig. 2 allows one to scale out these undesirable features enabling a meaningful comparison with Fig. 1. We see that the interference fringes in Fig. 1 do originate from the interference between the $\mathcal{P}^{(+)}(t, \theta)$ and $\mathcal{P}^{(-)}(t, \theta)$ while Fig. 2 shows a classical sum of the near-side and far-side intensities.

Note that the linear size of the chaotic billiard [5] is only about twice as big as the spatial width of the wave packet at $t=0$. Therefore, at any moment of time, the width of the wave packet cannot exceed its width at $t=0$ more than by about a factor of 2. From the obtained criterion the interference fringes wash out when the width of the spreading wave packets begins to exceed its initial value at $t=0$ by a factor ≥ 2 . It could be of interest to check if the interference fringes, at fixed moments of time, for chaotic billiard would disappear upon increasing the size of the billiard by a factor of 2 to 3 keeping the width of the wave packet at $t=0$ the same as in [5].

Our approach does not intend to substitute rigorous numerical calculations [9]. Yet, it does show that the complicated many-body collision problem can be accurately represented by a much simpler physical picture of a weakly damped ($\beta \ll \hbar\omega$) quantum rotator. This mapping has enabled us to address a problem of the quantum-classical transition in complex collisions and evaluate a characteristic time for such a transition. Remarkably, this simple picture of a weakly damped coherent rotation has also emerged from the numerical calculations for the F+HD [10] and He+H₂⁺ [11] chemical reactions. Originally this same physical picture of stable rotational wave packets was revealed for heavy-ion collisions [7,19]. Interestingly, this is in spite of the fact that a characteristic rotation time of the IC formed in bimolecular reactions is ~ 8 orders of magnitude longer than that for the IC formed in heavy-ion collisions.

The obtained criterion yields $t_{q-cl} \approx t_{spr}/d$, where $t_{spr} = \hbar/\beta$ is a characteristic time for the spread of a wave packet [6]. For classical macroscopic systems the wave packet spreading may be associated with the chaotic dynamics and t_{spr} could be macroscopically large. However, since d is in \hbar units and, in the macroscopic limit, \hbar vanishes, then $t_{q-cl} \propto \hbar \rightarrow 0$. This provides an alternative to the explanation in [2] of the practically instantaneous collapse of coherent superpositions into a classical sum of intensities for macroscopic systems. Unlike decoherence [2], in our approach “the quantum origins of the classical” are obtained in the absence of external noise or coupling to an environment and only due to the unitary evolution of a pure quantum state.

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