Abstract

We analyze quasi-periodic oscillations in the angle-averaged ($\Delta \theta_m \approx 90^\circ \pm 25^\circ$) excitation functions for the $^{24}Mg + ^{24}Mg$ elastic-inelastic scattering and $\alpha$-transfer channels on the energy interval $E_{cm} = 44.86 - 47.76$ MeV. The period of the energy structures, $\approx 0.81$ MeV, is interpreted as inverse half-period ($\approx 5 \times 10^{-21}$ sec.) of coherent rotation of highly excited short-lived ($\approx 3.6 \times 10^{-21}$ sec.) chain of a length $\approx 30$ fm. The rotational wave packet coherence survives (i) the energy relaxation (fully mixing ergodic dynamics) for the strongly overlapping states with fixed total spins and (ii) the strong distortion of the motion upon a change of the total spin. The present discussion leads us to the question: Is rotational coherence of large molecules necessarily destroyed in the conventionally statistical limit of structureless (non-selective) continuum under the conditions of complete intramolecular energy redistribution and vibrational dephasing in the regime of strong ro-vibrational coupling?

Keywords: coherent rotation, phase relaxation, $^{24}Mg + ^{24}Mg$ collision, nuclear molecules, quantum macroscopic transition

Femtochemistry provides powerful tools in studying molecular structure and dynamics in chemistry and biology [1], [2]. Nowadays, the use of ultrashort femtosecond and attosecond laser pulses is a widely used technique for the real-time monitoring of chemical reactions. One of the successes of femtochemistry is its ability for direct probe of vibrational and rotational wave packets and their coherent evolution as a classical-like, essentially due to the quantum interference, system-specific trajectories of a transitional state of a given chemical reaction. Precondition for survival of the characteristic time and length scales for the system-specific wave packet dynamics is that the isolated intermediate complex is not in ergodic state. This means that phase relaxation (dephasing) or dynamical decoherence [3], responsible for a spread of the wave packets, is relatively slow, not faster than the life time of the intermediate complex. This regime of incomplete mixing is beyond applicability of random matrix theory [4] which addresses a featureless universal behavior of complex systems in the ergodic limit of complete lose of phase memory and uniform occupation of the available phase space (simple pictorial explanation of the domain of applicability of random matrix theory, for not experts in the field, is given in Sect. V of [5]).

Unfortunately, the femtochemistry experimental methods are inapplicable to study reactions which are not initiated by the laser pulses. In particular, time evolution of heavy ion collisions is not accessible for the direct monitoring. Additional experimental difficulty to measure time scales of reactions involving relatively light heavy ions is their short duration. Namely, time resolution of order of zeptosecond ($10^{-21}$ sec) would be needed for an accurate reconstruction of heavy ion collisional dynamics. Therefore, the only possible way to access time evolution of heavy ion reactions is provided by the detailed energy dependence of the cross sections, i.e. excitation functions. In this respect, the most favorable situation is the presence of both fast (direct) and time delayed processes [6], [7], [8], [9]. Then, under the condition of a major contribution of direct processes, the energy variations in the excitation functions are mainly given by the interference between the collision amplitudes corresponding to the direct and time delayed processes. Since the direct process amplitude is energy smooth function, this allows to obtain time dependent amplitude, $\mathcal{P}(t, \theta)$ of the time delayed process [6], [7], [8], [9]. Clearly, the direct process plays a role of the pump pulse switching on the clock at the initial moment of time [5], [9]. When, for that or another reason, the interference term between the amplitudes of the direct and time delayed processes does not contribute to the cross section, the energy variations in the excitation functions originate from the modulus square of the energy oscillating around zero amplitude, $F(E, \theta)$, corresponding to the time delayed processes. In this situation, an information on the time evolution, though not as complete as in the presence of direct processes, is still encoded in the incident energy dependence of the cross sections. Indeed, we represent the cross section $\sigma(E, \theta) = |F(E, \theta)|^2$ as

$$\sigma(E, \theta) \propto \int_{-\infty}^{\infty} dt \exp(iE\tau/\hbar) < \mathcal{P}(t + \tau/2, \theta) \mathcal{P}(t - \tau/2, \theta)^* >_t$$

(1)

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with

\[
\langle \mathcal{P}(t + \tau/2, \theta) \mathcal{P}(t - \tau/2, \theta) \rangle = \int_0^\infty dt \mathcal{P}(t + \tau/2, \theta) \mathcal{P}(t - \tau/2, \theta) \tau,
\]

where \( \mathcal{P}(t, \theta) \propto \int_0^\infty dt \exp(-iE \ell t) F(E, \theta) \) with \( \mathcal{P}(t \leq 0, \theta) = 0 \). In the above relations, \( E \) is the total energy, \( t \) is the time, and \( \theta \) is the scattering angle.

Suppose that a pronounced quasi-periodic component, with a period of \( \delta E \), is present in \( \mathcal{P}(E, \theta) \). From model independent Eqs. (1) and (2), this implies quasi-periodicity or recurrences, at least one recurrence, in \( \mathcal{P}(t, \theta) \) with a period of \( \approx 2 \pi \hbar/\delta E \). If, in addition, the quasi-periodic energy components strongly correlate for different exit channels, this implies presence of the significant channel independent quasi-periodic component or strong enhancement in \( \langle \mathcal{P}(t + \tau/2, \theta) \mathcal{P}(t - \tau/2, \theta) \rangle \) at \( \tau \approx 2 M \pi \hbar/\delta E \) with \( M = 1, 2, \ldots \), at least with \( M = 1 \).

Quasi-periodic structures in the excitation functions reveal presence of non-statistical effects. For example, the oscillating energy structures in the \( {}^{24}\text{Mg} + {}^{24}\text{Mg} \) elastic-inelastic scattering \([10], [11]\) were interpreted in terms of the highly excited coherently rotating hyper-deformed intermediate complex \([13]\). However, the identification of the non-statistical quasi-periodic structures from the analysis of the cross section energy autocorrelation functions in \([12]\) may look questionable for the following reason. In the statistical regime of random phases of the strongly overlapping resonance levels with different total spin values, the energy autocorrelation functions, \( C(\varepsilon) \), have a Lorentzian shape \([13]\). This result is obtained for very long energy interval on which excitation functions are measured. Yet, for the finite data range analysis of the concrete experimental data sets, one encounters deviations from the theoretical prediction, in particular, in the form of fluctuations around the Lorentzians \([14]\). Therefore, even though the oscillations in \( C(\varepsilon) \)'s have approximately the same period for the different exit channels \([11], [12]\), which is inconsistent with the statistical interpretation, additional supportive argumentation for the quasi-periodic non-chaotic behavior is highly desirable. The interpretation \([12]\) in terms of the relatively stable coherent rotation, due to the slow phase relaxation, was based on the assumption of the highly excited (\( \approx 10 - 15 \) MeV above Yrast line) intermediate complex with strongly overlapping resonances. Therefore, it should be insightful to also test this interpretation for transfer channels in the \( {}^{24}\text{Mg} + {}^{24}\text{Mg} \) collision. Do the energy oscillating structures, with the same quasi-period as that for the \( {}^{24}\text{Mg}^2{}^{24}\text{Mg}^2{}^{24}\text{Mg} \) elastic-inelastic scattering, present in the transfer channels? Affirmative answer to this question would provide additional supportive indication that both \( {}^{24}\text{Mg}^2{}^{24}\text{Mg}^2{}^{24}\text{Mg} \) elastic-inelastic scattering and the transfer reaction channels originate from the decay of the same highly excited coherently rotating hyper-deformed intermediate complex.

The reasonings presented above has motivated us to analyze the angle-averaged, \( \Delta \theta_{cm} \approx 90^\circ \pm 25^\circ \), excitation functions for the \( {}^{24}\text{Mg} + {}^{24}\text{Mg} \) elastic-inelastic scattering and \( \alpha \)-transfer channels measured on the energy interval \( E_{cm} = 44.86 - 47.76 \) MeV \([15]\), which is considerably smaller than that \( (E_{cm} = 42 - 56 \) MeV) on which the data were taken in \([10], [11]\). The analysis in \([15]\) confidently confirmed the non-statistical behavior of the energy structures in these processes. We extend the analysis \([15]\) concentrating on the quantitative interpretation of the pronounced, transparently visible in Figs. 2 and 3 of \([15]\), strongly correlated for different channels, quasi-periodic energy structures.

In Fig. \( \text{III} \) we present the normalized cross section energy autocorrelation functions \( C(\varepsilon) \)'s constructed from the summed deviation functions in Fig. 3 of \([15]\). In the upper panel of our Fig. \( \text{II} \) \( C(\varepsilon) \) is obtained from the three lowest transitions in the reaction \( {}^{24}\text{Mg}^2{}^{24}\text{Mg} + {}^{20}\text{Ne} \rightarrow {}^{28}\text{Si} \). In the lower panel of our Fig. \( \text{II} \) \( C(\varepsilon) \) is obtained from addition of the three lowest well resolved transitions in the reaction \( {}^{24}\text{Mg}^2{}^{24}\text{Mg} + {}^{20}\text{Ne} \rightarrow {}^{28}\text{Si} \) and the three lowest well resolved transitions in the \( {}^{24}\text{Mg}^2{}^{24}\text{Mg} + {}^{24}\text{Mg} \) elastic-inelastic scattering (see Fig. 3 in \([15]\)). The experimental \( C(\varepsilon) \)'s demonstrate oscillations with the period close to that in Fig. \( \text{II} \) of \([12]\). Therefore, we fit the data with the formula \([12]\)

\[
C(\varepsilon \geq 0)/C(\varepsilon = 0) \approx \text{Re}[\exp(i\varepsilon t)/(\varepsilon + i\beta)]/[1 - \exp(i\varepsilon t + i\Gamma)/(\varepsilon + i\beta)] \]

\[
\text{Re}[1/[1 - \exp(-\pi\Gamma/\varepsilon + i\beta)]].
\]

Here, \( \Gamma \) is total decay width of the intermediate complex, \( \omega \) is
a real part of the angular velocity of the coherent rotation and $\beta/\hbar$ has a physical meaning of its imaginary part with $\beta$ being the spin off-diagonal phase relaxation width $[16]$. One observes that $C(\epsilon)$ (3) oscillates with period $\approx 2\hbar\omega$. The oscillations are damped for a non-vanishing $\beta$-width. The fit (solid lines in Fig. 1) is obtained with $\hbar\omega = 0.41$ MeV, $\beta = 0.02$ MeV and $\Gamma = 0.2$ MeV. In the limit of short phase memory, $\beta \gg \Gamma$, $C(\epsilon)$ has a Lorentzian shape $[13, 4]$ (long-dashed lines in Fig. 1 with $\Gamma = 0.2$ MeV).

A few comments on the derivation and physical interpretation of Eq. (3), in relation to the analyzed data, are in order.

1) The starting point for the derivation of Eq. (3) is a standard pole expression for $S$-matrix elements $[16]$ which can be justified under the condition of the fast energy redistribution-relaxation within the states with fixed total spin values $[5]$. Then, we employ the relations (8.1) and (8.2) in $[17]$ between the product of the entrance $(a)$ and the exit $(b)$ channel partial width amplitudes for different $J$-values:

$$
\gamma_{\nu}^{I_a} \gamma_{\mu}^{I_b} = \sum_{\mu} \gamma_{\nu}^{I_a \mu} \gamma_{\mu}^{I_b} O_{\nu \mu}^{I} + R_{inc},
$$

where

$$
O_{\nu \mu}^{I} = (1/\pi)D\beta[J-I]/[(E_{\nu}^{I} - E_{\mu}^{I} - (J-I)\hbar\omega)^2 + \beta^2 (J-I)^2],
$$

with $E_{\nu}^{I}$ and $E_{\mu}^{I}$ being resonance energies for the states with total spin values $J$ and $I$, respectively. The relations (4) hold (but still $\beta \gg \Gamma$), $R_{inc}$ in Eq. (4) can be neglected $[17]$ for it produces insignificant contribution into the r.h.s. of Eq. (5). Then, in a formal analogy with the consideration of $[18]$, we obtain

$$
S^I_{ab}(E) = W[J - J(E)]d^{1/2} \sum_{\mu} \gamma_{\nu}^{I_a \mu} \gamma_{\mu}^{I_b} |E - E_{\mu}^{I}|^{1/2} + (i/2)\Gamma + i\beta[J - I]],
$$

where we have omitted energy smooth phase shifts which may originate from the potential mean field scattering taking place on the relatively short time intervals of formation and disintegration of the intermediate complex. In case of elastic scattering, $(a,b)$, the products $\gamma_{\nu}^{I_a \mu} \gamma_{\mu}^{I_b}$ and $\gamma_{\nu}^{I_a \mu} \gamma_{\mu}^{I_b}$ in Eq. (4) and Eq. (6) should be changed to $[\gamma_{\nu}^{I_a \mu} \gamma_{\nu}^{I_a \mu} - \gamma_{\nu}^{I_a \mu} \gamma_{\nu}^{I_a \mu}]$ and $[\gamma_{\nu}^{I_a \mu} \gamma_{\nu}^{I_a \mu} - \gamma_{\nu}^{I_a \mu} \gamma_{\nu}^{I_a \mu}]$, respectively $[5]$. In Eq. (6), $W[J - J(E)]d$ is the energy averaged decay probability. It was taken in a bell shaped form with the maximum at $J = J(E)$ and with the width $d$ of about an effective range of total spin values coherently excited in the collision process. For the data analyzed in this Letter this range is $\Delta J = 34 - 38$. In the derivation of Eq. (3), we took $J(E) = I + (E - \bar{E})/\hbar\omega$, where $\bar{E}$ is the average energy corresponding to $J(E) = I \gg 1$. Under the neglect of the interference term between the near and far side collision amplitudes due to the angle-averaging $[18]$, such a choice of $J(E)$ results in $C(\epsilon)$ (3) to be independent of $I$ and $d$. However, for the other choices, e.g. $J(E) = I = const$, the additional damping factor, $\approx W[-|\epsilon|/(d\hbar\omega)]$, would appear in the r.h.s. of Eq. (3).

2) The data fitted in Fig. 1 were obtained from the angle-averaged excitation functions, $\Delta \theta_{cm} = 90^\circ \pm 25^\circ$. Yet, the formula (3) was derived for the pure angle-resolution (though under the neglect of the interference term between the near and far side collision amplitudes as well as between direct and time-delayed processes $[12]$). Generalization for the angle-averaged excitation functions, $\Delta \theta_{cm} = 90^\circ \pm 25^\circ$, yields the additional damping factor, $\sin[\epsilon\Delta \theta/(2\hbar\omega)]/[\epsilon\Delta \theta/(2\hbar\omega)]$ with $\Delta \theta = 50^\circ = 0.88$, in the r.h.s. of Eq. (3). The corresponding fit of the data on the lower panel of Fig. 1 is obtained with $\hbar\omega = 0.404$ MeV, $\beta = 0.0205$ MeV and $\Gamma = 0.17$ MeV. Clearly, the additional damping factor, due to the poor angular resolution, does not affect period of the oscillations in $C(\epsilon)$.

3) The time dependent intensity, $|\mathcal{P}(t, \theta)|^2$, closely relates to the return probability for a finite time resolution, while modulus square of Fourier component of $\mathcal{P}(t, \theta)$, i.e. the cross section (1), is analogous to the low-resolution version the spectrum $[19]$. In our case the finite time resolution is $h/\Gamma$, and the spectrum is not resolved due to the strong overlap of the resonance levels, $\Gamma/D \gg 1$. Clearly, $C(\epsilon)$ has a physical meaning of the autocorrelation function of the low-resolution version of the spectrum $[19]$.

Stability of the coherent rotation may be appreciated by noticing that the spreading of the angular orientation during one revolution due to the finite $\beta$-width is $2\beta/\hbar\omega \approx 15^\circ$. Therefore, it takes about 10 complete revolutions, i.e. $\approx 10^{-19}$ sec., for the complex to loose the coherent nature of its rotation. Energy relaxation time scale for excited nuclei is $\approx h/\Gamma_{spr} = 10^{-22}$ sec., where $\Gamma_{spr} = 5$ MeV is the spreading width $[4]$. We observe that the coherent rotation persists for about three orders of magnitude larger than it takes to complete a process of energy relaxation.

The strong channel correlation between individual well-resolved channels in both the $^{24}$Mg ($^{24}$Mg, $^{22}$Mg)$^{22}$Mg elastic-inelastically scattering and $^{24}$Mg ($^{24}$Mg, $^{20}$Ne)$^{28}$Si transfer reaction, as well as between the channels for these two processes, is transparently visible in Fig. 2 of $[13]$. Therefore, it is obvious that strongly correlated regular oscillations with the approximately channel-independent quasi-period, $\approx 0.81$ MeV, are present in all the six channels. This rules out statistical origin of these strongly correlated quasi-periodic structures and, therefore, their interpretation in terms of Ericson fluctuations $[13]$. This is because Ericson fluctuations produce uncorrelated, for different channels, irregular energy structures. As a result, the deviations from the Lorentzian must be uncorrelated for different channels instead of demonstrating regular oscillations with the approximately channel-independent quasi-period for all the six channels. The fact that the present analysis of the data on the energy interval $E_{cm} = 44.86 - 47.76$ MeV revealed a value of the quasi-period very close to that obtained for the much longer energy interval $E_{cm} = 42 - 56$ MeV $[12]$ strongly supports our interpretation. A possibility of further test of our interpretation, e.g. for the $^{24}$Mg ($^{24}$Mg, $^{22}$Mg)$^{22}$Mg elastic scattering, is suggested by the expected dominance of direct processes (potential scattering) for $\theta \leq 60^\circ$ (see Figs.
6 and 7 in [20]). Then the energy variations in the excitation functions originate mainly from the interference between the collision amplitudes corresponding to the direct and time delayed processes. As a result, the characteristic quasi-periods of leading harmonics in $\sigma(E, \theta)$ become strongly $\theta$-dependent [7], [8], [9], which can be tested in experiments with pure angular resolution.

Angular velocity of coherent rotation, $\omega$, can not be defined within the states with fixed $J$-value. This transparently follows from the conjecture [5] which relates $Q_{12}^{ff}$ (5) to the spin off-diagonal correlations between squares of resonance states of the intermediate complex. Indeed, it is obvious that the distribution of spacial densities, including the radial extension, may well be different for the state with fixed $J$-value from that for the coherently rotating intermediate complex. Therefore, energy of the coherent rotation as well as the associated moment of inertia and, thus, the characteristic length scale of the coherently rotating intermediate complex are also undefinable within the states with fixed $J$-value.

Let us assume that the coherent rotation may be considered as the macroscopic motion. Then, an assumption of the spherical intermediate complex having rigid body moment of inertia is ruled out for, in this case, $\hbar \omega \approx 4$ MeV, resulting in the rotational energy ($\approx 70$ MeV) which exceeds the total excitation energy ($\approx 60 - 63$ MeV). Instead, the small value of $\hbar \omega \approx 0.4$ MeV, corresponding to the period of the rotation $\approx 10^{-20}$ sec, indicates an anomalously strong deformation of the coherently rotating complex with $\Gamma \approx 34 - 38$. We calculate moment of inertia of this complex and find that it corresponds to a chain-state of the length $\approx 30$ fm. This is close to the length $\approx 24$ fm of the chain of four touching carbon nuclei, calculated with $r_0 = 1.3$ fm. We evaluate deformation energy of the chain as a sum of Coulomb energy of the two touched $^{12}$C nuclei and double Coulomb energy of the two touched $^{24}$Mg nuclei. This gives 38 MeV for the upper limit, corresponding to $r_0 = 1.3$ fm, of the deformation energy. The energy of the coherent rotation, for $J \approx 34 - 38$, is $\approx 7$ MeV. Therefore, the energy of the intrinsic excitation (heat) is estimated $\geq 15$ MeV, i.e. our “carbon sausages” are really hot. Such a relatively high intrinsic excitation energy suggests the small average level spacing, $D$, for the states with fixed $J$-values from the interval $J \approx 34 - 38$. The standard statistical model evaluation in [21] yields $D \leq 10^{-5}$ MeV$< \Gamma \approx 0.2$ MeV for fixed $J$-value. This indicates a strong overlap of the resonance levels, $\Gamma/D \geq 10^4$.

Up to now the idea of rotational coherence of ergodic, with respect to the energy and phase relaxation within the states with fixed $J$-values, nuclear molecules in continuum has been employed to extract spectroscopic information ($\hbar \omega$, $J_{coh}$) from dynamics for a few colliding systems, e.g. $^{58}$Ni + $^{58}$Ni [22], $^{58}$Ni + $^{62}$Ni [23]. $^{60}$Ti + $^{58}$Ni [23], $^{12}$C + $^{24}$Mg [6], [7], [9], $^{24}$Mg + $^{24}$Mg and $^{28}$Si + $^{28}$Si [12]. $^{24}$Mg + $^{28}$Si [24]. Therefore, it may be of interest to apply this method for analysis of a large number of available data sets for many heavy ion colliding systems. On the other hand, the question is whether the stable coherent rotation survives violation of rotational symmetry and other external perturbations of the Hamiltonian? The conjecture [5] offers a possible specification of this question in terms of the correlation properties of eigenstates of the intermediate complex.

Experimental manifestation of rotational coherent motion in complex molecules was initially met with scepticism “because of the general belief that Coriolis interactions, anharmonicity and other interactions would destroy the coherence.” (see subsection “Changing a Dogma: Development of RCS” in [2]). The present discussion leads us to ask: Is rotational coherence of complex molecules necessarily destroyed in the conventionally statistical ergodic limit of structureless (non-selective) continuum, $\Gamma/D \geq 10^4$, under the conditions of complete intramolecular energy redistribution and vibrational dephasing in the regime of strong ro-vibrational coupling? Though challenging experimentally, the subject is of interest to search for fingerprints of transformation of the quantum coherent wave packet dynamics into macroscopic motion. More specifically, in the quantum regime, the spreading of the wave packets is determined by $\beta$-width, which has essentially quantum origin of the spin off-diagonal correlations [3]. Then what is a classical analog of $\beta$-width, for classically chaotic systems, when macroscopic non-linear dynamics takes over from the coherent rotation of the spreading quantum wave packets? Addressing the questions above should include detailed comparison of the nature of the dephasing analyzed in [1] with that discussed in this Letter (see also [5] and references therein).

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