Rotating Morse wave packet dynamics of diatomic molecule

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We investigate the dynamics of a rotating Morse wave packet, appropriate for a ro-vibrating diatomic molecule. The coupling between vibrational and rotational degrees of freedom is explicated in real position space as well as in phase space Wigner distribution of a SU(2) coherent state at various dynamically evolved times. We choose the well studied $I_2$ molecule with the parameter values in good agreement with experiments. A quantitative measure of the angles of rotation for different angular momenta is also given.

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I. INTRODUCTION

The Morse potential \[^1\] is extensively studied in the literature and has been a subject of considerable attention. It involves the vibrational degrees of freedom of a diatomic molecule. On the other hand, rigid rotor model is described to take into account only the rotational degrees of freedom, where on-axis rotation is usually neglected and end-over-end rotations are identical for a homo-nuclear molecule. The rotational and vibrational energy scales are far apart and it has been a customary to treat them independently. However, in reality, this separation is inadequate and the internuclear distance is affected by the centrifugal force resulting from rotation. Thus, one should consider both degrees of freedom of a diatomic molecule. A combined form of the Morse potential and the rotational centrifugal term is known as the rotating Morse potential. It leads a more complex energy spectrum reflecting the coupling among the two intrinsic properties of nuclear molecule. It is known that the Schrödinger equation for this potential can be exactly solved only when the orbital angular momentum quantum number $j$ is equal to zero. Since, there is no exact analytical solution for the rotating Morse potential in general, some semiclassical and/or numerical solutions have been obtained by using various approximate methods. The original approach developed by Pekeris \[^2\] is based on the expansion of the centrifugal barrier in a series of exponentials depending on the internuclear distance up to second order. A more appropriate expansion has been proposed by Duff and Rabitz \[^3\] and Elsum and Gordon \[^4\]. These semi-analytical models are based on the idea of building up Morse potentials with parameters dependent on the rotational quantum number, which represent adequately the effective potential curves of the rotating Morse oscillator. There are various other numerical and proposed methods to study the rotating Morse potential \[^5\] \[^6\] \[^7\] \[^8\] \[^9\] \[^10\] \[^11\] \[^12\] \[^13\]. These studies are very useful to obtain more accurate energy eigen values.

Recent development of ultrashort pulses opened a new research field which allows to study the wave packet dynamics of diatomic molecule in real time scales. Pulses of a few tens of femtosecond allow a coherent broadband excitation with preparation and detection of rovibrational dynamics. In all previous works, the wave packet dynamics have been studied in diatomic molecule either considering the vibration \[^14\] \[^15\] \[^16\] and rotation \[^17\] \[^18\] \[^19\] separately. There are few works, which considers the both in two-time scale configuration \[^20\] \[^21\] \[^22\].

In this work, we study the wave packet dynamics, considering the coupling among the vibrational and rotational degrees of freedom of nuclear motion. Our analytical approach for the study of the rotating Morse potential is inspired by Gordon’s method \[^4\], where the effective potential becomes minimum around a new equilibrium internuclear distance $r_j$, which is a function of angular momentum quantum number $j$. This model, so far, most appropriately manifests the coupling between two degrees of freedom, vibration and rotation. To the authors’ knowledge, there is no study about the wave packet dynamics of this appropriate system. We show that this 1D model is accurate up to a reasonably high angular momentum ($j = 200$), accessible to experiments. We study rovibrational interplay through a coherent state (CS) dynamics, not only in coordinate space, but also in phase space Wigner distribution. It is shown that rovibrational interplay is better explicated in phase space rotation of the CS for different angular momenta. When rovibrational dynamics is seen in 3D configuration space, the corresponding phase space is six dimensional, which is not possible to visualize. Avoiding this 3D intricacy, this 1D rotating Morse potential can well describe the coupling between the two degrees of freedom, hence can nicely manifest the rovibrational interplay for this realistic system in phase space. It also shows the effects of rotation-vibration in the quantum interference regime. We have chosen the iodine molecule ($I_2$) which is an unique suited seed molecule for laser induced fluorescence. Additionally, we give a numerical method, which can estimate the angles of rotation, are in good agreement with the Wigner plots.

The paper is organized as follows. In the following section, we present a brief overview and justification of the
1D rotating Morse system. We also evaluate the corresponding SU(2) CS wave packet. In Sec. III, we analyze the dynamics of the CS of a diatomic molecule and explain the effect of rovibrational coupling in this. After showing the effect in the configuration space, we move to the phase space picture for a better representation, which clearly displays the rotational effect. A quantitative analysis of the angle of rotation is also given. This numerical analysis confirms the values of rotation angle, corresponding to various angular momentum quantum number. Finally, we end with some conclusions in Sec. IV.

II. ROTATING MORSE WAVE PACKET

The Schrödinger equation for the rotating Morse oscillator reads as follows,

\[ \frac{h^2}{2\mu} \frac{d^2}{dr^2} + V_{\text{eff}}(r) \psi_{n,j}(r) = E_{n,j}(r), \quad (1) \]

where \( \mu \) is the reduced mass of the diatomic molecule and \( n, j \) are the vibrational and rotational quantum numbers, respectively. The effective potential \( V_{\text{eff}}(r) \) has the form

\[ V_{\text{eff}}(r) = D[e^{-2\beta(r-r_0)} - 2e^{-\beta(r-r_0)}] + \frac{j(j+1)\hbar^2}{2\mu r^2}. \quad (2) \]

The first part describes the well known Morse potential, where \( D \) is the dissociation energy of the Morse potential, \( r_0 \) is the equilibrium internuclear separation and \( \beta \) is the range parameter. The second part of the above equation stands for the centrifugal contribution of rotation. More accurate description of the system can be obtained with a modified equilibrium internuclear distance \( r_j \) and a dissociation energy \( D_j \) [4]. Using a semi-analytical method [23], one can find

\[ r_j = r_0 \left[ 1 + \frac{A}{\beta^2 r_0^2 D} \right]; \quad D_j = D - A \left( 1 - \frac{A}{\beta^2 r_0^2 D} \right), \quad (3) \]

where \( A = \frac{j(j+1)\hbar^2}{2\mu r_0^2} \). Alternatively, \( r_j \) can also be computed numerically by solving the transcendental equation

\[ \frac{dV_{\text{eff}}(r)}{dr} \bigg|_{r=r_j} = 0. \quad (4) \]

These two sets of \( r_j \)’s are plotted in Fig. 1(a), which shows very good agreement for \( j < 200 \). It shows that the rotational potency results into the increasing value of the equilibrium distance [Fig. 1(a)] and decreasing nature of dissociation energy [Fig. 1(b)]. Physically, in presence of the rotational centrifugal force, the two constituent atoms of a diatomic molecule tend to settle in a larger distance and they are more prone to dissociate, reducing the amount of energy required to make them independent. We define \( A_j = \frac{j(j+1)\hbar^2}{2\mu r_0^2} \) and expand the centrifugal term of Eq. (2) around \( r = r_j \). Keeping terms up to second order, the effective potential becomes

\[ V_{\text{eff}}(y) = \frac{c_2}{4\lambda_j^2} y^2 - \frac{c_1}{\lambda_j} y + c_0, \quad (5) \]

where \( y = 2\lambda_j e^{-\beta(r-r_0)} = 2\lambda_j e^{-\beta(r-r_j)} \) and \( u = e^{-\beta(r-r_j)} \).

The parameter, \( \lambda_j = \sqrt{\frac{2\mu c_2}{\beta^2 D_j}} \), where the constants, \( c_0, c_1 \), and \( c_2 \) are dependent on quantum number \( j \):

\[ c_0 = 3A_j b_j^2 - 3A_j b_j + A_j, \]

\[ c_1 = (3A_j b_j^2 - 2A_j b_j + A_j + uD)/u, \]

and

\[ c_2 = (3A_j b_j^2 - A_j b_j + A_j + u^2 D)/u^2, \quad (6) \]

with \( b_j = (\beta r_j)^{-1} \). Under this approximation, the Schrödinger equation (1) is solved to yield the eigen functions of this rotating Morse system:

\[ \psi_{n,j}(y) = N_{n,j} e^{-y^2/2} y^s L_n^{2s}(y), \quad (7) \]

where the variable \( y \): \( 0 < y < \infty \), \( n \) is the vibrational quantum number, \( L_n^{2s}(y) \) stands for associate Laguerre polynomial and \( s = \sqrt{-\frac{(E_{n,j} - c_0)/\lambda_j^2}{c_2}} \). The number of
bound states in the system is $[\tilde{\lambda}_j - 1/2]$, where $[\rho]$ denotes the largest integer smaller than $\rho$. The parameter $\tilde{\lambda}_j$ is related as $\tilde{\lambda}_j = \frac{2\lambda_j}{\lambda_j}$.

They satisfy the constraint condition $s + 2n = 2\tilde{\lambda}_j - 1$. $N_{n,j}$ is the normalization constant:

$$N_{n,j} = \left[ \frac{\beta(2\lambda_j - 2n - 1)\Gamma(n + 1)}{\Gamma(2\lambda_j - n)} \right]^{1/2}.$$  \hfill (8)

The rovibrational energy eigen values $E_{n,j}$ turns out as

$$E_{n,j} = 2\lambda_j(1 + 1/2) - \frac{c_2}{\lambda_j}(n + 1/2)^2 + c_0 - \frac{c_1^2}{c_2}.$$  \hfill (9)

It is worth pointing out that in absence of rotation $c_0 = 0$, $c_1 = c_2 = D$, the system only describes a vibrating diatomic molecule.

III. DYNAMICS OF A ROTATING MORSE WAVE PACKET OF A DIATOMIC MOLECULE

Ultrashort laser pulses can probe the molecular dynamics in real time scale. Time-resolved measurements on the femtosecond scale have made it possible to know the evolution of a coherently prepared superposition of ro-vibrational states. Here, we consider a rovibrational wave packet of $I_2$ molecule, which is a coherent state, dependent on particular rotational quantum number. Many theoretical and experimental investigations have been carried out on this molecule and has been a subject of many femtosecond studies, particularly in wave packet interferometer and detection purpose, probe pulses of 310 nm and 330 nm are used. These parameters allow the probe excitation mainly between the $B0^a_2$ and $f0^a_0$ states. Under the laser polarizations magic angle conditions, it takes into account the vibrational as well as the rotational motions.

The rotational effect on the effective potential of $I_2$ molecule is depicted in Fig. 2. Without rotation i.e., $j = 0$ (solid line), it is nothing but the Morse potential. Once rotation is considered, e.g., $j = 60$ (dashed line) or $j = 81$ (dotted line), the potential changes, i.e., the potential-minima slowly shifts towards right (from $r_e = 5.03$ a.u. to $r_j = 5.0323$ a.u. and $r_j = 5.034$ a.u. respectively) and dissociation energy diminishes. The rovibrational coupling changes the effective potential and hence energy levels are shifted upwards. The 10th energy level, where the coherent state is excited, is shifted from $E_{10} = -0.04719$ a.u. to $E_{10} = -0.046595$ a.u. and $E_{10} = -0.046108$ a.u respectively. In our further study, in particular, we consider these effective potentials and the initial wave packet as a coherent state involving few lower levels of $I_2$ molecule centered around $n = 10$. If we take only bound states of the potential, the dynamical symmetry group is $SU(2)$. In this case, the displacement operator coherent state can be constructed and written as

$$\Phi(y, t) = \sum_{n=0}^{N} d_n^* \psi_{n,j}(y)e^{-iE_{n,j}t}.$$  \hfill (10)

![FIG. 2: Effective potentials for $j = 0$ (solid line), $j = 60$ (dashed line) and $j = 81$ (dotted line) are depicted. The corresponding 10th energy levels are zoomed in the inset plot. The wave packets at $t = 0.25 T_{rev}$ are also shown in the inset as dark filled ($j = 0$), dashed ($j = 60$) and light filled ($j = 81$) plots, respectively. Changes of $j$ value cause slight changes in the effective potential, which result in rotational effect on the wave packet. The two splitted wave packets in co-ordinate space come closer with increasing $j$ and superpose each other for $j = 81$, producing interference ripples. The potential and the internuclear distance are in atomic units.](image)

Here, the group generators are $j$ dependent and the weighting coefficient becomes

$$d_n^j = \left\langle \alpha \right| \left\langle n' | \Gamma(2\lambda_j - n) \right\rangle^{1/2} \left\langle n' | \Gamma(2\lambda_j - n') \right\rangle.$$  \hfill (11)

It can be easily verified that for $j = 0$, the above coherent state (CS) is the same as reference 30. The $SU(2)$ generators satisfy the commutator relations, $\left[ J_+ , J_- \right] = 2J_0$, $\left[ J_0 , J_{\pm} \right] = \pm J_{\pm}$ and also satisfy

$$\left[ J_+ , J_- \right] \psi_{n,j}(y) = 2j_0^j \psi_{n,j}(y).$$  \hfill (12)
at the level of wave function, where \( j_0 = n - \lambda_j + 1/2 \). \( \hat{J}_0 \) is the projection operator of the angular momentum \( m: m = n - \lambda_j + 1/2 \). Although, the CS of a physical system is never ideal, like that of harmonic oscillator, we find that our chosen CS is very well localized and remains coherent for a reasonable time duration. Presence of quadratic term in energy expression, leads some interesting phenomena, called fractional revivals, which occur at some specific instances between two full revivals \((T_{rev})\) \([31, 32]\). The short time evolution displays a classical periodicity \((T_{cl})\). The classical and revival time periods are respectively given by

\[
T_{cl} = \frac{2\pi\lambda_j}{2c_1-c_2\lambda_j} \quad \text{and} \quad T_{rev} = 2\pi\lambda_j^2/c_2. \tag{13}
\]

For \( I_2 \) molecule, these time scales are \( T_{cl} = 0.156 \) picosecond and \( T_{rev} = 36.2 \) picosecond. At fractional revival times (rational fraction \( r/s \) of the revival time \( T_{rev} \)), the wave packet breaks into several number of subsidiary wave packets, where \( r \) and \( s \) are mutually prime integers. For even values of \( s \) the wave packet breaks into \( s/2 \) in number, otherwise breaks into \( s \) parts. In the course of time evolution, one obtains the Schrödinger cat like state at one fourth of the revival time. Four-way break up or the compass-like state emerges at one-eight of the revival time. Latter produces sub-Planck scale structures in phase space Wigner distribution. In the inset of Fig. 2 the 10th energy levels for different angular momenta are zoomed and the rotational coupling effect is shown at \( t = 0.25 T_{rev} \), when CS is splitted into two parts. Initially \((j = 0)\), the two parts are situated at \( r = 4.7 \) a.u. and \( 5.58 \) a.u. (dark filled plot). After the introduction of rotational coupling \((j = 60)\), they approach each other, while in the next quarter, they recede. At halfway of the oscillation, they reflect from the potential well with a phase change \( \pi \) and become again counter-propagating. For the angular momentum \( j = 81 \), they overlap each other in the course of their oscillation and produce the oscillatory ripples, clearly visible in the inset of Fig. 2 and in Fig. 3(a).

Until now we have confined our study in position space explanation. This explanation does not reveal the full description of rotational coupling of the system. Hence, we enlarge our view towards the phase space description. Wigner function \([33]\) is very useful tool to visualize the quantum dynamics in phase space and is defined as

\[
W(r, p, t) = \frac{1}{\pi\hbar} \int_{-\infty}^{+\infty} \Phi^*(r - r', t) \Phi(r + r', t)e^{-2ipr'/\hbar} dr'. \tag{14}
\]

This representation can reveal interesting structures of the CS at different evolved times. As \( I_2 \) molecules has large number of bound states (117), we choose the coherent state parameter \( \alpha = 2.15 \), which involves few lower levels of the potential. In Fig. 3 first row shows the time evolution of the CS with \( j = 0 \). It represents the Morse vibrational dynamics at different fractional revival times. The initial CS is situated near the left potential boundary and is well localized [Fig. 3 (a)]. Owing to the anharmonicity of the potential, the probability

\[
W(r, p, t) = \frac{1}{\pi\hbar} \int_{-\infty}^{+\infty} \Phi^*(r - r', t) \Phi(r + r', t)e^{-2ipr'/\hbar} dr'. \tag{14}
\]
densities near the left boundary are stiffer compare to those near right boundary. From the previous explanation, it is evident that the compass-like state appears at \( t = T_{\text{rev}}/8 \) [Fig. 3(b)], which shows the smallest interference structures, i.e., the well known sub-Planck scale structures \([32, 34, 35, 36]\). Origin of these smallest structures is the superposition of the two diagonal interference ripples. The Schrödinger cat-state \([37]\) appears at \( t = T_{\text{rev}}/4 \) [Fig. 3(c)], where the interference ripples appear in the middle as alternate positive and negative values, along the line joining the two component CSs. All these negative phase space interference structures are the strong signature of quantum behavior. At half revival time the CS revives near the right boundary of the potential well [Fig. 3(d)]. Upon introducing the angular momentum contribution, we show the phase space structures at the same times as above in Fig. 3(e)-(h). The initial CS is similar because it does not depend on rotation. One can observe the rotational effect on the other structures. For heavy molecules, the rotational effect is less for smaller values of \( j \). We choose \( j = 60 \) as it is reasonable and also appropriate for experiment. Interestingly, the revival time \( T_{\text{rev}} \), does not change with \( j \) values, because the ratio \( \lambda_j^2/c_2 \) remains same. Whereas, the classical period \( T_c \) alters due to the rovibrational coupling. The rotation is anti-clockwise, thereby making the angle of rotation positive in our convention. Fig. 3(f) describes the rotational effect at the sub-Planck interference structures. It is established that these structures are very sensitive to external perturbation or rotation, which can be utilized for quantum parameter estimations \([38]\). These sub-Planck structures are also found most sensitive against decoherence in diatomic molecular system of hydrogen iodide molecule \([39]\). In our model, it is worth to study the rotational sensitivity due to a infinitesimal rovibrational coupling in this physical system. As this is not the present goal of this work, we are studying it in another context \([39]\). Fig. 3(g) shows the rotational effect in the cat state. The CS is revived at \( t = 0.5 T_{\text{rev}} \), but shifted in its orbit due to rotation.

We would like to explain another specific case of the phase space structures to reveal the intricacies in a better way. Fig. 4 shows the cat state for higher values of \( j = 81 \). This cat state is separated in momentum space, but exactly superposed in real position space. A single interference ripple in Fig. 4(a) has dimension 0.07 a.u. or 3.7 picometer. Although the experimental observation of small quantum interference structures is very challenging, similar interference ripples in picometer scale is recently visualized experimentally for \( \text{I}_2 \) molecule \([26]\). They observed it in the classical time evolution of a vibrational wave packet at time \( t = 0.25 T_{\text{rev}} + 0.25 T_c \) or \( t = 0.25 T_{\text{rev}} + 0.75 T_c \). The same picometer structures can be observed for a rovibrating CS of \( \text{I}_2 \) molecule at a fixed time \( t = 0.25 T_{\text{rev}} \) with angular momentum around \( j = 81 \). This structure remains stationary in their full crossing pathway in the range \( 74 < j < 88 \), which keeps the flexibility of experiments.  

Now a natural question can be asked whether there is a way to know the orientation \( \phi \) corresponding to a \( j \) value. Here, we provide a numerical estimation of this rotation angle. We operate an operator \( U = e^{i\hat{J}_\phi \omega} \), where \( \hat{J}_\phi \) is the angular momentum operator, on the initial wave packet and obtain

\[
U \Phi(y, t)_{j=0} = \sum_{n=0}^{n'} \phi_n e^{i(n - \bar{X}_j + 1/2)\phi} \psi_{n,0} e^{-iE_{n,0}t} = \chi(y, t).
\]  

(15)

Then we compute the overlap \( | \chi(y, t)|\Phi(y, t) |^2 \), numerically and take the value of \( \phi \) for the largest overlap \( \sim 1 \). We repeat it for various angular momenta and find the corresponding angles of rotation. In particular, we have chosen the rotation of cat state, i.e., at time \( 0.25 T_{\text{rev}} \). The result is depicted in Fig. 5 up to a full period of rotation (2\( \pi \) radians). Two particular rotations \( (j = 60 \text{ and } j = 81) \) are indicated in the figure by dotted lines. From the nature of the plot, one can infer that the wave packet is more phase sensitive at larger \( j \) val-
ues. We have checked that these rotation angles are in good agreement with the phase space pictures. To be more precise, let us take a particular case for $j = 81$. We obtained an angle of rotation $\phi = 0.573\pi$ radians for $j = 81$, but the cat state is exactly vertically situated (0.5r radians) with respect to the $p = 0$ line (Fig. 1b). This is because, the initial cat state ($j=0$, Fig. 3c) is not exactly horizontal due to the $T_{\text{rev}}/T_{\text{cl}}$ ratio, which is not exactly an integer and imprinting a small phase or rotation equivalent to $\phi = -0.073\pi$ radians. Thus, for a perfect vertical cat state, one needs the rotation of $0.573\pi$ radians, which is exactly the obtained value in our analysis.

IV. CONCLUSION

We have showed the time evolution of a CS wave packet of rotating Morse potential. We are able to manifest the rovibrational coupling of a diatomic molecule through this CS dynamics in the experimentally allowed parameter regime. We considered a well behaved and appropriate SU(2) coherent state, including few bound states of $I_2$ molecule. This can be generated with an available pump beam excited at the 10th energy level and for angular momenta $j = 60$ or 81. The rovibrational coupling is explored in position space, where the physical rotation of the CS is not so transparent. Phase space structure clearly showed the rotational influences on the cat state as well as the compass-like state. Our study deprecates the complication of six-dimensional physics in phase space for ro-vibrational dynamics. The 1-D rotating Morse potential can well capture the rotational effect throughout the time evolution in phase space. Moreover, we provide a way to identify the amount of rotation of the rovibrating CS at arbitrary time and angular momentum, which can be very useful to know the behavior of the system at certain extend, without going into the phase space description.

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