We discuss the rotational cooling of diatomic molecules in a Bose-Einstein condensate (BEC) of ultra-cold atoms by emission of phonons with orbital angular momentum. Despite the superfluidity of the BEC there is no frictionless rotation for typical molecules since the dominant cooling occurs via emission of particle-like phonons. Only for macro-dimers, whose size becomes comparable or larger than the condensate healing length, a Landau-like, critical angular momentum exists below which phonon emission is suppressed. We find that the rotational relaxation of typical molecules is in general faster than the cooling of the linear motion of impurities in a BEC. This also leads to a finite lifetime of angulons, quasi-particles of rotating molecules coupled to phonons with orbital angular-momentum. We analyze the dynamics of rotational cooling for homo-nuclear diatomic molecules based on a quantum Boltzmann equation including single- and two-phonon scattering and discuss the effect of thermal phonons.

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I. INTRODUCTION

The physics of a quantum impurity in collective many-body environments is an important subject of condensed matter physics. It dates back to the classic problem of a polaron put forward by Landau and Pekar [1] and Fröhlich and Holstein [2–5] to explain charge transport in solids resulting from the dressing of a moving electron with phonon-like excitations of the surrounding material. In many systems internal degrees of freedom of the impurity can be disregarded as their characteristic energy scale is well separated from that of the environment and the impurity can be treated as point-like object. The Fermi and Bose polarons recently realized in ultra-cold quantum gases [6–17] are important examples providing a many-body model system where impurity problems can be analyzed very precisely. Also the dynamics of its formation can be studied, which is an equally important problem since collective properties such as the superfluidity of a BEC can strongly influence the equilibration dynamics [18–20]. Recently the concept of a polaron was extended to impurities with a more complex structure such as a molecule. It was shown that the coupling of rotation to collective excitations of a surrounding BEC can give rise to a new type of quasi-particles termed angulons [21–25]. In the present paper we discuss the cooling dynamics of the rotational degrees of freedom of a single, diatomic molecule immersed in a three-dimensional (3D) Bose-Einstein condensate, see Fig. 1 which is relevant both for the formation and the stability of angulons. To this end we use a microscopic quantum Boltzmann approach [26] based on a Bogoliubov theory of impurity-condensate interaction.

Emission and scattering of Bogoliubov phonons with orbital angular momentum off the molecule lead to a deceleration of the rotational motion and eventually to equilibration with the condensate. For typical sizes of molecules and weakly interacting condensates there is no

analogue of a Landau critical velocity, i.e. there is in general no critical value of angular momentum below which phonon emission and scattering is suppressed. This is because the spatial structure of the molecule can only be resolved by high-energy phonons, which have a particle-like character. Thus different from the case of polarons, i.e. point-like impurities dressed with Bogoliubov phonons, there are in general no stable states of angulons. The rotational relaxation rates are however smaller than the typical binding energies of angulons.

The situation is different if one considers macro-dimers, such as Rydberg molecules [27–30], where an atom is trapped in a high-lying Rydberg state of another atom. In this case molecular size and healing length can become comparable and the interaction with low-energy phonons becomes the most important one. The same
holds true for impurities trapped in shallow, rotationally symmetric potentials. In this limit the superfluidity of the condensate changes the relaxation dynamics and we recover a Landau critical behaviour. Below a certain angular momentum of the macro-dimer the emission of phonons is effectively suppressed and the rotational relaxation stops in a pre-thermalized state.

The paper is organized as follows: In Sec. II we will introduce the model of a rigid rotator coupled to Bogoliubov phonons of an atomic BEC. The quantum Boltzmann equation used to describe the relaxation dynamics is reviewed in Sec. III and the different contributions to the relaxation rates resulting from spontaneous and thermal single- and two-phonon processes are derived. The relaxation dynamics of macro-molecules will be discussed in Sec. IV and that of typical molecules in Sec. V.

II. MODEL

We here discuss the case of a diatomic molecule, which we describe as a rigid rotor of two point masses \( m \) with distance \( 2r_0 \), see Fig. I immersed in a three-dimensional (3D) weakly interacting Bose Einstein condensate of atoms, which we describe in Bogoliubov approximation. We assume that the center of mass (COM) of the molecule is at rest in the lab frame of the BEC and we disregard the COM kinetic energy of the molecule. The total hamiltonian of the systems

\[
H = H_0^m + H_0^{ph} + H_{int}
\]

consists of the free Hamiltonians of the diatomic molecule \( H_0^m \), and that of the Bogoliubov phonons \( H_0^{ph} \): \[2\]

\[
H_0^m = \frac{\hat{L}^2}{4m kr_0^2}, \quad H_0^{ph} = \sum_{k,\lambda,\mu} \omega_k \hat{b}_{k,\lambda,\mu} \hat{b}_{k,\lambda,\mu}.
\]

Here \( \hat{L} \) is the angular momentum operator of the rotating diatomic molecule and \( 2r_0 \) the molecule diameter. \( \omega_k = c \sqrt{1 + k^2 \xi^2 / 2} \) is the Bogoliubov dispersion relation of the phonons, with \( \xi = 1 / \sqrt{2m_B g n_0} \) being the condensate healing length. \( m_B \) is the mass of the BEC atoms, \( g \) is the strength of atom-atom interactions in the condensate in s-wave approximation. \( c = \sqrt{g n_0 / m_B} \) is the speed of sound of the phonons. The homogeneous condensate of density \( n_0 \) is assumed to be in an initial equilibrium state at temperature \( T \ll T_c \), \( T_c \) being the critical temperature of condensation, which for a non-interacting homogeneous condensate of density \( n_0 \) reads \( T_c = 2\pi n_0^{2/3} / (m_B \zeta(3/2)^{3/2}) \). If a rotating molecule is placed in the BEC we expect that its angular momentum thermalizes to an equilibrium distribution of quantum numbers \( j \) with characteristic value

\[
j_T(j_T + 1) = \frac{8\pi}{\zeta(3/2)^{2/3}} \frac{T}{T_c} \frac{m_I}{m_B} \left( r_0 n_0 / 3 \right)^2.
\]

For a typical molecule with size small compared to the average distance between atoms in the BEC, i.e. \( r_0 \ll n_0^{-1/3} \), we expect a cooling to the lowest angular momentum \( j \to 0 \).

The interaction of the homo-nuclear diatomic molecule with the BEC, \( H_{int} = H_1(\mathbf{r}) + H_2(-\mathbf{r}) \), is described as s-wave scattering interaction of the two atoms with the condensate. We assume that higher-order partial waves are not relevant for the scattering process, which sets some upper limit for the rotational quantum number. The interaction \( H_1(\mathbf{r}) \) of a point-like impurity at position \( \mathbf{r} \) with the BEC reads in terms of plane-wave Bogoliubov modes

\[
H_1(\mathbf{r}) = \int d^3k \frac{gBn_0^{1/2}}{(2\pi)^{3/2}} W_k e^{-i\mathbf{k} \cdot \mathbf{r}} \hat{b}_k^{\dagger} + \hat{b}_{-k}
\]

\[
+ \frac{gB}{2(2\pi)^{3/2}} \int d^3k \int d^3k' \left[ W_{k,k'}^{\infty} \hat{b}_{k}^{\dagger} \hat{b}_{k'} + \frac{1}{2} W_{k,k'}^{\infty} \left( \hat{b}_{k}^{\dagger} \hat{b}_{k'}^{\dagger} + \hat{b}_{-k'} \hat{b}_{-k} \right) \right] e^{-i(\mathbf{k} - \mathbf{k'}) \cdot \mathbf{r}}, \tag{4}
\]

where \( W_k = |k^2 \xi^2 / (2 + k^2 \xi^2)|^{1/4} = \sqrt{\epsilon_k / \omega_k} \), with \( \epsilon_k = k^2 / (2m_B) \) being the kinetic energy of the condensate atoms, and we used the abbreviations \( W_{k,k'}^{\infty} = W_{k} W_{k'} + W_{k}^{-1} W_{k'}^{-1} \) and \( W_{k,k'}^{\infty} = W_{k} W_{k'} - W_{k}^{-1} W_{k'}^{-1} \). Making use of the decomposition of plane waves into spherical ones

\[
e^{i\mathbf{k} \cdot \mathbf{r}} = 4\pi \sum_{\lambda,\mu} j_{\lambda}(kr) Y_{\lambda\mu}(\theta,\phi) Y_{\lambda\mu}^*(\theta,\phi)
\]

where \( j_{\lambda}(kr) \) is the spherical Bessel function, and the orthogonality relations of spherical harmonics, we can rewrite eq. (4) in terms of angular momentum modes

\[
\hat{b}_{k,\lambda,\mu} = k \int d\phi \sin\theta k^{i\lambda} Y_{\lambda\mu}(\theta,\phi) \hat{b}_{k,\lambda,\mu},
\]

\[
\hat{b}_{k} = \frac{1}{k} \sum_{\lambda,\mu} j^{-\lambda} Y_{\lambda\mu}(\theta,\phi) \hat{b}_{k,\lambda,\mu}.
\]

\( \lambda = 0,1,\ldots \) and \( \mu = -\lambda, -(\lambda-1), \ldots, \lambda-1, \lambda \) are the quantum numbers of the orbital angular momentum of the phonons in the rest frame of the center-of-mass of the molecule. The spherical-mode operators fulfill bosonic commutation relations \( [\hat{b}_{k,\lambda,\mu}, \hat{b}_{k',\lambda',\mu'}^\dagger] = \delta(k-k')\delta_{\lambda,\lambda'}\delta_{\mu,\mu'} \).

With this we find

\[
H_{int} = \sum_{k,\lambda,\mu} U_{\lambda}(k) \left[ Y_{\lambda\mu}(\theta,\phi) \hat{b}_{k,\lambda,\mu} + Y_{\lambda\mu}^*(\theta,\phi) \hat{b}_{k,\lambda,\mu}^\dagger \right] + \]

\[
+ \sum_{k,\lambda,\mu} U_{\lambda}(k, k') Y_{\lambda\mu}(\theta,\phi) Y_{\lambda\mu}'(\theta,\phi) \hat{b}_{k,\lambda,\mu}^\dagger \hat{b}_{k',\lambda,\mu'} + \]

\[
+ \sum_{k,\lambda,\mu} \frac{1}{2} U_{\lambda}(k, k') Y_{\lambda\mu}(\theta,\phi) Y_{\lambda\mu}'(\theta,\phi) \hat{b}_{k,\lambda,\mu}^\dagger \hat{b}_{k,\lambda,\mu} + h.a.
\]

where we made use of the fact that the distance of both atoms to the origin is the same and fixed to \( r = r_0 \). The
coupling constants for the single-phonon terms read

$$U_\lambda = \begin{cases} g_{1B} \sqrt{\frac{2m}{\pi}} k W_k j_\lambda(kr_0) & \lambda \text{ even} \\ 0 & \lambda \text{ odd} \end{cases}$$ (6)

and for the two-phonon terms

$$U_{\lambda\lambda'}^p = \begin{cases} \frac{2m}{\pi} k k' j_\lambda(kr_0) j_{\lambda'}(k'r_0) W_{k,k'}^{\mu} & \lambda + \lambda' \text{even} \\ 0 & \lambda + \lambda' \text{odd} \end{cases}$$ (7)

The vanishing of the coupling constants for odd values of $\lambda$ or $\lambda + \lambda'$ is due to the inversion symmetry of the molecules. For the hetero-nuclear case also odd terms would be nonzero. As a consequence the symmetric molecule can only emit and absorb single phonons with even orbital angular momentum or phonon-pairs which have an even total angular momentum. Rotational cooling will thus occur in a cascade with angular momentum steps of two.

### III. QUANTUM BOLTZMANN EQUATION

We now want to study the dynamics of a molecular impurity with finite initial angular momentum interacting with the BEC, described by the Hamiltonian [5]. The starting point is a master equation for the interacting with the BEC, described by the Hamiltonian.

The discussion can be substantially simplified if we consider only the probabilities for total angular momentum $j$, $p_j = \sum_{m=-j}^j p_{jm}$. Making use of the properties of Clebsch-Gordan coefficients we find that the total rates $\sum_m \Gamma_{jm \rightarrow j'm'} = \Gamma_{j \rightarrow j'}$ are independent of $m$ as expected from the rotational symmetry of the problem. Thus eq. (8) simplifies to

$$\frac{dp_j}{dt} = \sum_{j'} (p_{j'} \Gamma_{j' \rightarrow j} - p_j \Gamma_{j \rightarrow j'})$$ (12)

with the total rates

$$\Gamma_{j \rightarrow j'}^{1\text{ph},\text{sp}} = (2j'+1) \sum_{\lambda} \gamma_{\lambda} j' \left( C_{j0,j'0}^0 \right)^2 \Theta_{j,j'},$$

$$\Gamma_{j \rightarrow j'}^{1\text{ph},\text{T}} = (2j'+1) \sum_{\lambda} \gamma_{\lambda} j' \left( C_{j0,j'0}^0 \right)^2 \pi_{j,j'}.$$ (13)

Since for the Clebsch-Gordan coefficients holds $C_{j0,j'0}^0 = 0$, if $j' + j + \lambda$ is odd and $\gamma_{\lambda} j' = 0$ for odd $\lambda$, one recognizes that states with even (odd) initial angular momentum $j$ can only decay into states with even (odd) final angular momentum $j'$.

In order to get an impression on the dependence of the single-phonon decay rates on the angular momentum quantum numbers, we have plotted in Fig. 2 the spontaneous scattering rates $\Gamma_{j \rightarrow j'}^{1\text{ph},\text{sp}}$ as functions of $j$ and $j'$ for two different values of $r_0/\xi$. While for typical sizes of molecules, for which $r_0/\xi \ll 1$, shown in Fig. 2(a), there is a smooth dependence on $j$ and $j'$, one finds for macro-dimers, for which $r_0/\xi$ is of the order of or larger than unity, shown in Fig. 2(b), that the decay rates are strongly suppressed for $j$ below a critical value $j_c$. Also the final angular momentum that can be reached in a single-phonon process is limited by a second critical value $j_c^{(1)}$. This will be discussed in more detail in sec IV.
As discussed in the Appendix we find for the total transition rates corresponding to the scattering of a phonon the two relaxation cascades remain separated also when considering two-phonon processes.

IV. MACRO MOLECULES AND LANDAU CRITICAL ROTATION

As seen from Fig.2, the single-phonon rotational relaxation is very different in the two cases of a usual molecule with \( r_0 < \xi \) and a macro-molecule \( r_0 > \xi \) or an atom in a shallow rotationally symmetric trap. We thus will discuss these two cases separately in the following. We first consider macro-molecules with a radius \( r_0 > \xi \), the opposite limit is discussed in a subsequent section.

A. Relaxation rates and critical rotation

In the case of a macro-dimer the spontaneous single-phonon decay rate \( \Gamma_{j \rightarrow j'}^{1\text{ph,sp}} \) is the dominating one at low temperature and is plotted in Fig.2 (b). The checkerboard pattern evolves as a consequence of the two independent relaxation cascades for even (odd) angular momentum number. As noted above, transition rates are suppressed for low angular momentum states and the molecule cannot not decay to the lowest \( j \) value. This can be understood from analogy to linear motion of a single impurity through the condensate [19]. The impurity will not scatter phonons when its momentum is smaller than the Landau critical...
value $p_c = m_I c$ and for large radii the rotation of the molecule can be approximated as a translation.

One can determine a critical angular momentum $j_c$ below which the scattering of further phonons is strongly suppressed by simultaneous energy and angular momentum conservation. To this end we compare the energy of two linear moving impurities, each with momentum $p$, to one rotating molecule, identifying

$$\frac{j_c(j_c + 1)}{4m_I r_0^2} = 2 \frac{p^2}{2m_I}.$$  \hspace{1cm} (19)

The corresponding Landau critical angular momentum $j_c$ is then given by

$$j_c(j_c + 1) = 2 \frac{m_I^2 r_0^2}{m_B^2 \xi^2}. \hspace{1cm} (20)$$

We note that in order to have an integer $j_c \geq 1$ the size of the molecule $r_0$ has in general to be larger than the healing length $\xi$.

Furthermore we know that a linear moving impurity with $m_I > m_B$ can only decay into a state with momentum bigger than $p^{(1)}_c = p_c \sqrt{1 - m_B^2/m_I^2}$, when only single-phonon processes are considered. In analogy to the discussion above, one can derive the minimal angular momentum $j^{(1)}_c$ a rotating macro molecule can decay into:

$$j^{(1)}_c(j^{(1)}_c + 1) = 2 \frac{r_0^2}{\xi^2} \left( \frac{m_I^2}{m_B^2} - 1 \right).$$  \hspace{1cm} (21)

Both $j_c$ and $j^{(1)}_c$ fit very well to the rates calculated for the Boltzmann equation, see Fig.2(b).

In the inset of Fig.3 $\Gamma_{j}^{1\text{ph,sp}}$ is plotted for different ratios $r_0/\xi > 1$ against $j$. One clearly notices a sharp onset at $j_c$. When plotting the decay rates as function of angular momenta normalized to the critical value from eq. (20), all curves collapse to a single one when $r_0 \gg \xi$. This universal behavior can be understood by analogy to the case of two linear moving impurities: For a rotating macro molecule with angular momentum $j \gg 1$ and rotational energy equal to the kinetic energy of two linear moving impurities, each with momentum $p$, one finds

$$\frac{j}{j_c} = \frac{p}{p_c},$$  \hspace{1cm} (23)

independent on the ratio $r_0/\xi$.

### B. Cooling dynamics

Very similar to [19] one can show that the relaxation processes mediated by two-phonon processes are much slower than single-phonon terms in a weakly interacting 3D BEC, where $n_0 \xi^3 \gg 1$, since they scale as

$$\Gamma_{2\text{ph}}/\Gamma_{1\text{ph}} \sim (n_0 \xi^3)^{-1}.$$  \hspace{1cm} (24)

Furthermore also thermally induced two-phonon processes are very slow and not relevant below $T_c$. Note that the situation is markedly different in lower dimensions [20], where thermally-induced processes can become important due to the infra-red divergence of contributions by thermally occupied phonon modes.

---

**Figure 3.** (Color online) Single-phonon decay rate $\Gamma_{j}^{1\text{ph,sp}}$ for different molecule sizes $r_0/\xi$. As shown in the inset we find non vanishing decay rates only for $j > j_c$. Plotting the rates against $j$ normalized to $j_c$, the curves collapse to a single one if $r_0/\xi \gg 1$.

**Figure 4.** Relaxation of a macro dimer initially prepared in an angular-momentum eigenstate $j = 24$. A fast approach to a pre-thermalized state with $j \geq j_c$ is clearly visible. Here $r_0 = 10\xi$ and $m_I = m_B$ and $j_c = 12$ and we considered a finite temperature $T = 0.01T_c$. Due to the existence of a Landau critical angular momentum we expect a pre-thermalization to a non-equilibrium rotational state, which is visible unless $j_c \ll$...
\( j_T \), which only happens at high temperatures. In Fig. 4 we have plotted the time evolution of the occupation of angular momentum states starting at an eigenstate with \( j = 24 \). One clearly recognizes the formation of a prethermalized state with \( j \geq j_c^{(1)} \), while states with lower \( j \) will only be populated on a much larger time scales set by two-phonon processes.

V. SMALL MOLECULES

A. Single-phonon rates and angulon stability

Typical molecules have sizes much less than the healing length of the BEC \( r_0 \ll \xi \). In this case we can drastically simplify the effective single-phonon transition rates (11) which yields

\[
\gamma_{\lambda}^{j,j'} = \begin{cases} \frac{c}{2} \frac{g_B^2}{\pi} \frac{1}{n_0} \frac{\xi}{r_0} \sqrt{\frac{m_B}{m_1}} \Delta_{jj'} \lambda (\sqrt{\frac{m_B}{2m_1}} \Delta_{jj'})^2 \lambda & \text{even} \\ 0 & \text{else} \end{cases}
\]

(25)

where \( \Delta_{jj'} = |j(j+1) - j'(j'+1)| \).

Furthermore thermal contributions to the single-phonon rate can be completely disregarded as the energy spacing between adjacent rotational states is much larger than the thermal energy, \( E_{j,j'/k_BTc} > (m_B/m_1)(n_0^{1/3}r_0)^{-2} \). As a consequence \( \pi_{jj'} \ll 1 \).

In Fig. 2 (a), we plotted the transition rates \( \Gamma_{j\rightarrow j'}^{1ph,sp} \) in the limit of a small molecule. An important difference to the case of a macro molecule is that the molecule always decays into the lowest angular momentum states \( j = 0 \) or 1. The absence of a Landau critical rotation can be understood very simply from the following argument: Phonons can resolve the rotation of the molecule if their wavelength is comparable or smaller than the molecule size \( r_0 \). Thus the relaxation is dominated by scattering of high-energy, i.e. short wavelength phonons with \( k \geq r_0^{-1} \ll \xi^{-1} \). These short-wavelength phonons are however particle-like and there is no suppression of their emission or scattering by simultaneous energy-momentum conservation. As a consequence quasi-particles arizing from the dressing of rotating molecules with angular-momentum phonons are fundamentally unstable. Furthermore in the case of a linear motion of the impurity, it is known that the transition rates are on the order of \( \frac{c}{2} \frac{g_B^2}{\pi} \frac{1}{n_0} \frac{1}{r_0} \) [19]. In contrast \( \gamma_{\lambda}^{j,j'} \) shows that the typical transition rates for a rotating molecule are bigger by a factor \( \xi/r_0 \). This may raise concerns if angulons can be observed at all. However, the typical binding energies of angulons are sizable fractions of the rotational energy of the molecule. When we compare the single-phonon decay rate of angular-momentum states to the relevant energy scale, given by the rotational constant \( B = \frac{1}{4m_jr_0^2} \), we find

\[
\frac{\Gamma_{j\rightarrow j'}^{1ph,sp}}{B} \propto \frac{r_0}{\xi} \ll 1
\]

(26)

So while excited rotational states of a molecule in a BEC are not stable, their lifetime is still large compared to the energy of the angulon.

B. Thermal two-phonon contributions

For single-phonon processes thermal effects can be neglected. This no longer holds true for processes involving two phonons. The dominant two-phonon process is the one, were the state of the molecule decays, via absorption of a low-energy thermal phonon and subsequent (spontaneous) emission of a high energy phonon. For usually sized molecules, with \( r_0n_0^{1/3} \ll 1 \) the decay rates due to two-phonon processes are proportional to the spontaneous single phonon rates, with a proportionality factor which depends on the BEC temperature and \( n_0\xi^3 \), but not on \( j \) or \( j' \). In Fig. 5 we have plotted the ratio of thermal two-phonon to single-phonon decay rates from numerical calculations. One recognizes that they approach a universal curve (dashed line) when the gas parameter \( n_0\xi^3 \) increases.

As shown in the Appendix one finds

\[
\Gamma_{j\rightarrow j'}^{2ph,T} = \Gamma_{j\rightarrow j'}^{1ph,sp} \frac{\sqrt{2}}{4\pi^2} \frac{1}{n_0\xi^3} \int_0^\infty dk \xi k(k) \pi(k).
\]

(27)

Here \( \kappa \) is the energy of the thermal phonon in units of \( c/\xi \). \( k(k) \) is the phonon momentum and \( \pi(k) \) the thermal phonon number corresponding to this energy. For \( n_0\xi^3 \gg 1 \) this expression can be further simplified which
yields
\[ \Gamma^{2ph,T}_{j \rightarrow j'} \simeq \left( \frac{T}{T_c} \right)^{3/2} \Gamma^{1ph,sp}_{j \rightarrow j'} . \]

This simple relation holds, since the thermal phonon absorbed in the two-phonon process carries no angular momentum, and its energy is negligible compared to the transition energy \( E_{j,j'} \). Therefore the thermal contributions in the two-phonon scattering effectively lead to a renormalization of the single-phonon process.

C. Cooling dynamics

Finally we consider also the relaxation dynamics of small molecules. To this end we solve the Boltzmann equation \(^8\) numerically by calculating the spontaneous decay rates \(^{13}\) and their thermal equivalent \(^{10}\). In order to include two phonon processes given in eqs. \(^{13}\) and \(^{16}\) we focus on a subset of momenta up to \( j \leq 25 \). Fig. \(^7\) shows the angular momentum decay of an initial state with \( j = 24 \) into a final state with \( j = 0 \). Here we see no influence of two-phonon processes, but a smooth and fast relaxation to a thermal state for any initial distribution of a micro dimer.

\[
\rho_j(t) = \begin{cases} 1.0 & \text{if } j = 24 \\ 0.5 & \text{if } j = 0 \\ 0.0 & \text{otherwise} \end{cases}
\]

\[ \text{time } t/\left[ \xi/c \right] = 0.04, 0.1, 1.0, 25, 15, 5, 0 \]

\[ \text{ angular momentum } j = 24 \rightarrow 0 \]

Figure 6. Time evolution of angular momentum of a micro dimer starting at \( j = 24 \). The color encoding matches the occupation number \( \rho_j(t) \) plotted on the x-axis. For long times \( t \) we find a pumping to the final state with \( j = 0 \). Here we have \( r_0 = 0.1\xi \) and \( m_1 = m_B \) and a finite temperature \( T = 0.01T_c \).

VI. SUMMARY

In the present paper we have studied the rotational relaxation of diatomic molecules immersed in a Bose-Einstein condensate of atoms at a temperature much below the critical value of condensation. The BEC is assumed to be weakly interacting such that a description in terms of a homogeneous condensate and Bogoliubov phonons is valid. The molecule was modeled as rigid rotator of two point particles. A more accurate description of the interaction potential between molecule and condensate atoms is possible but only affects the quantitative value of the coupling constants. The relaxation dynamics was analyzed with a quantum Boltzmann approach, which is valid for weak BEC impurity interaction. The corresponding rates can be derived from Fermi-golden rule and describe spontaneous and thermally-induced creation or absorption of a single phonon by the impurity out of or into the condensate as well as spontaneous and thermal two-phonon processes. The rotational cooling is markedly different in the case of a macro molecule with a size \( r_0 \) exceeding the BEC healing length \( \xi \) and for a typical molecule, for which \( r_0 \ll \xi \). In the first case we found a universal behavior of the cooling rates and a Landau critical angular momentum \( j_c \) caused by the superfluidity of the condensate in analogy to the case of linear motion. An initially rotationally excited molecule will quickly evolve into a pre-thermalized state which contains only angular momenta above a certain value \( j_c \). The time scales of this evolution are comparable to that found in the case of linear motion. On the other hand for molecules of typical size, for which \( r_0 \ll \xi \), there is no effect of the superfluidity of the BEC since the cooling is dominated by short-wavelength phonons in the particle-like part of the Bogoliubov spectrum. Thus in contrast to polarons, angulons are in general not protected from decay by the superfluidity of the condensate. The typical relaxation rates are much larger than in the case of macro-dimers. They are however still smaller than the typical binding energies of angulons.

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APPENDIX

In order to calculate the single-phonon transition rates eq. \(^{9}\) and \(^{10}\) we first evaluate the matrix element in \( \Gamma_{m \rightarrow n} = 2\pi \delta(E_m - E_n) \langle m | H_{int} | n \rangle |^2 \), which yields

\[
\Gamma^{1ph,sp}_{jm \rightarrow jm'} = \frac{1}{2} \sum_{k,\lambda \mu} \delta(E_j - E_{jm} - \omega_k) U_\lambda(k)^2 G^m_{jm',\lambda \mu} \]

\[
\Gamma^{1ph,T}_{jm \rightarrow jm'} = \frac{1}{2} \sum_{k,\lambda \mu} U_\lambda(k)^2 \pi_k \left[ G^m_{jm',\lambda \mu} \delta(E_{jm'} - E_j - \omega_k) + G^m'_{jm',\lambda \mu} \delta(E_j - E_{jm'} - \omega_k) \right].
\]
We made the assumption that the phonon number $\pi_{k\lambda\mu}$ depends only on $k$, which is valid for thermal phonons. The integration over the absolute value of the phonon momentum $k$ can be carried out. Furthermore by using the symmetry $G^{j\eta} jk\mu\lambda = G^{j\mu\lambda} jk\eta\mu$, the thermal transition rates can be simplified. This yields

$$\Gamma^{1\text{ph}, sp}_{j m \to j' m'} = \sum_{\lambda\mu} \left. \frac{1}{2} \frac{d\omega}{d\omega} U_\lambda(k_\omega)^2 \Theta_{j j'} G^{j\eta} jk\eta\mu \right|_{\omega = \omega_{j j'}}$$

(31)

$$\Gamma^{1\text{ph}, th}_{j m \to j' m'} = \sum_{\lambda\mu} \left. \frac{1}{2} \frac{d\omega}{d\omega} U_\lambda(k_\omega)^2 n_{k_\omega} G^{j\eta} jk\eta\mu \right|_{\omega = \omega_{j j'}}$$

(32)

Where $k_\omega = \frac{1}{\xi} \sqrt{1 + 2\omega^2 \xi^2 \omega^2} - 1$ is the inverse of the dispersion relation $\omega_{k}$. The effective single phonon transition rates eq. (11) are then defined as

$$\gamma_{\lambda j}^{j'} = \left. \frac{1}{2} \frac{d\omega}{d\omega} U_\lambda(k_\omega)^2 \right|_{\omega = \omega_{j j'}}$$

(33)

The derivation of the two-phonon rates can be done in a similar way. In the following the term proportional to $U_\lambda^* (k, k')$ will be considered. The derivation of the rates proportional to $U_\lambda^* (k, k')$ follows analogously. When evaluating the Matrix element of $H_{int}$ proportional to $U_\lambda^* (k, k')$ one finds

$$\Gamma_{\lambda j \to j'}^\infty = \frac{1}{\delta \pi} \sum_{kk'\lambda\lambda'} U_{1\lambda\lambda'}^* (k, k')^2 \left( \bar{\pi}_{kk'} + 1 \right)$$

(34)

$$\pi_{k} \delta(E_j - E_j' - \omega_{k} + \omega_{k'}) \sum_{LL' MM'} G^{LM}_{\lambda\mu', \lambda\mu} G^{j\eta j'}_{j m, LL} G^{LM'}_{\lambda\mu', \lambda\mu} G^{j\eta j'}_{j m, L'M'}$$

(35)

where $m'$ is already summed over all final $m'$. The last sums over the $D$s can be simplified using properties of the Clebsch-Gordan coefficients [32].

$$\sum_{LL' MM'} G^{LM}_{\lambda\mu', \lambda\mu} G^{j\eta j'}_{j m, LL} G^{LM'}_{\lambda\mu', \lambda\mu} G^{j\eta j'}_{j m, L'M'} = (2j' + 1) \sum_{L} \frac{(\lambda + 1)(2\lambda + 1)}{2L + 1} C_{\lambda 0, \lambda' 0} C_{j 0, j' 0}$$

(36)

Furthermore the integral over $k'$ can directly be carried out and the one over $k$ is transformed into an integral over $\eta$, where $\eta E_{j j'}$ is the energy corresponding to the momentum $k$. This yields eq. (15), where the effective transition rate is given by

$$\gamma_{\lambda j j' j}^\infty (\eta) = \frac{E_{j j'}}{8\pi} \left| \frac{d\omega}{d\omega} \right|_{\omega = \omega_{j j'}} \left( \frac{d\omega}{d\omega} \right)_{\omega = (\eta + 1) E_{j j'}}$$

(37)

$$\left[ U_{\lambda j}^* (k_{j j'} (\eta), k_{j j'} (\eta + 1)) \right]^2$$

When a typical size molecule should be described this can be simplified further. In the following the essential steps to derive eq. (27), which gives the ratio between rates due to two- to single-phonon processes, are explained. Since $E_{j j'} \gg k_{B} T$, the molecule can not be excited, so $j > j'$. This yields

$$\Gamma_{j j' \to j}^\infty \propto \int_{0}^{\infty} d\eta \gamma_{\lambda j j' j}^\infty (\eta) \pi_{j j'} (\eta) \left[ \pi_{j j'} (\eta + 1) + 1 \right]$$

(38)

The thermal phonon $\pi_{j j'} (\eta)$ number at energy $\eta E_{j j'}$ decays exponentially fast with $\eta$. So only $\eta \ll 1$ must be considered in the integral, which is valid for $r_{0} \sqrt{n_{0}} \ll 1$. This has the physical meaning that the energy of the thermal absorbed phonon can be neglected when compared to the energy of the emitted one. In this approximation the waveform of the thermal phonon is much bigger than the molecule, so $r_{0} k_{j j'} (\eta) \ll 1$ which leads to

$$j_{\lambda} (r_{0} k_{j j'} (\eta))^{2} \approx \delta_{\lambda, 0}$$

(39)

Since all spherical Bessel function $j_{\lambda} (x)$ with $\lambda > 0$ are vanishing for small $x$. This leads to the conclusion, that the thermal absorbed phonon carries no angular momentum.

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