

A general approach toward rotational decoherence

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(Dated: December 18, 2019)

Recent advances in levitated optomechanics provide new perspectives for the use of rotational degrees of freedom for the development of quantum technologies. As for the translational case, their use, especially in the quantum regime, is limited by environmental noises, whose characterization is thus fundamental to assess, control and minimize their effect, in particular decoherence. Here, we present a general analytic approach to quantify decoherence for a quantum system in a superposition of its rotational degrees of freedom. The specific case of the dipole-dipole interaction is solved explicitly, and we show that the rotational degrees of freedom decohere on a time scale that is longer than the translational one. Our scheme also provides a straightforward algorithm for computing the decoherence effect perturbatively, when calculation cannot be carried out exactly.

One of the main challenges of quantum technologies is to control, if not remove, the noise from quantum systems; the literature on this subject are vast [1, 2]. Environmental influences on the translational degrees of freedom of a material system [3, 4] were studied within the context of scattering theory [5–9] and Brownian motion [10–13], with important applications to molecular interferometry [14–16], cold atoms [17, 18] and optomechanics [19–25].

Recently, rotational degrees of freedom in levitated system have triggered the interest of the community for the new possibilities they offer both for research [26–38], as well as for technological applications [39–43]. With that comes the necessity to understand and characterize rotational decoherence from the surrounding environment. A first master equation was derived in [44], and then extended in [45] to include translational effects. However the formalism does not allow for a straightforward application to specific situations of interest.

In this letter, we approach the problem of quantifying decoherence effects on a quantum system initially prepared in a superposition of rotational degrees of freedom, from a more algorithmic point of view. We first develop a general and exact expression for the decoherence rate, which can be applied to every interaction potential between the system and its surrounding environment, and at the same time allows for a perturbative resolution. Then, we provide the explicit and exact form of the rotational decoherence rate due to a dipole-dipole interaction. The resulting formula will show that rotations can be far less affected by decoherence than translations. This means the rotational superpositions can be used to reach longer time-coherences for fundamental studies, as well as for technology development.

The model.— We consider a non-spherical particle of mass M , and we focus only on its rotational motion. Its orientation is described quantum mechanically in terms of the state $|\Omega\rangle$, representing the system in the angular con-

figuration Ω ; This can be obtained by starting from a reference configuration $|\mathbf{0}\rangle$ (e.g. with the anisotropy of the system along the x axis) then applying a rotation $\hat{D}_S(\Omega) = e^{-i\hat{L}_z\gamma/\hbar}e^{-i\hat{L}_x\beta/\hbar}e^{-i\hat{L}_z\alpha/\hbar}$ defined by the three Euler angles $\Omega = (\alpha, \beta, \gamma)$ with \hat{L}_i representing the angular momentum operator along the i -th axes [44, 46, 47]. The statistical operator describing the rotational state of the system is

$$\hat{\rho}_S = \int d\Omega \int d\Omega' \rho_S(\Omega, \Omega') |\Omega\rangle \langle \Omega'|, \quad (1)$$

where $\rho_S(\Omega, \Omega')$ are its matrix elements with respect to $|\Omega\rangle$ and $|\Omega'\rangle$. The system eventually couples to the surrounding environment through the interaction with environmental particles. The corresponding dynamics is described by the following master equation [44]

$$\frac{d\rho_S(\alpha, \alpha', t)}{dt} = -\Lambda_R \rho_S(\alpha, \alpha', t), \quad (2)$$

where, to be quantitative, we considered the case where the system is in a superposition of angular configurations obtained only from rotations around the z axis. Here, Λ_R is the decoherence rate due to the N particles of which the environment is made. The latter is defined as

$$\Lambda_R = n \int dk v(k) \rho(k) \frac{\int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}'}{8\pi} |\Delta f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2, \quad (3)$$

with $n = N/V$ the number density, $v(k) = \hbar k/m_{\text{gas}}$ the velocity of environment particle of mass m_{gas} , $\rho(k) = 4\pi k^2 \mu(k)$ the momentum distribution of the environmental particles and

$$\Delta f^\omega(\mathbf{k}', \mathbf{p}') = f(\mathbf{k}', \mathbf{p}') - f^\omega(\mathbf{k}', \mathbf{p}'), \quad (4)$$

where $f(\mathbf{k}', \mathbf{p}')$ is the scattering amplitude, where we defined $\omega = \alpha - \alpha'$. Here, $f^\omega(\mathbf{k}', \mathbf{p}')$ has the same form of $f(\mathbf{k}', \mathbf{p}')$ but with \mathbf{k}' and \mathbf{p}' replaced by \mathbf{k}'_ω and \mathbf{p}'_ω respectively, which are the same vectors rotated by the

angle ω . A derivation of Eq. (2), which differs from that in Ref. [44], is reported in Supplemental Material [48].

Rotations under the Born approximation.— To further investigate the properties of the decoherence rate Λ_R , we apply the Born approximation. In this case, $f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')$ is expressed as [44]:

$$f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}') = -\frac{m_{\text{gas}}}{2\pi\hbar^2} \int d\mathbf{r} V(\mathbf{r}) e^{-ik(\hat{\mathbf{k}}'_\omega - \hat{\mathbf{p}}'_\omega) \cdot \mathbf{r}}. \quad (5)$$

Being interested in rotations, we decompose $e^{-i\mathbf{k} \cdot \mathbf{r}}$ and the interaction potential in spherical harmonics $Y_{l,m}(\hat{\mathbf{r}})$: $e^{-i\mathbf{k} \cdot \mathbf{r}} = 4\pi \sum_{lm} (-i)^l j_l(kr) Y_{l,m}(\hat{\mathbf{r}}) Y_{l,m}^*(\hat{\mathbf{k}})$, where $j_l(x)$ are the spherical Bessel function of the first kind, and

$$V(\mathbf{r}) = \sum_{l''m''} d_{l'',m''}(r) Y_{l'',m''}(\hat{\mathbf{r}}), \quad (6)$$

where $d_{l'',m''}(r)$ denotes the radial part of the potential and $\hat{\mathbf{r}} = \mathbf{r}/r$. One obtains:

$$\begin{aligned} \Delta f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}') &= -\frac{8\pi m_{\text{gas}}}{\hbar^2} \sum_{lm} \sum_{l'm'} \sum_{l''m''} R_{l,l',l'',m''}(k) \\ &\quad \times Y_{l,m}^*(\hat{\mathbf{k}}') Y_{l',m'}(\hat{\mathbf{p}}') G_{l,m,l',m',l'',m''}(\omega), \end{aligned} \quad (7)$$

where

$$R_{l,l',l'',m''}(k) = \int dr r^2 j_l(kr) j_{l'}(kr) d_{l'',m''}(r), \quad (8)$$

contains the information on the radial part of the potential, and the angular part is encoded in

$$\begin{aligned} G_{l,m,l',m',l'',m''}(\omega) &= (-1)^{m'} i^{l'-l} \left(1 - e^{i\omega(m-m')}\right) \\ &\quad \times \sqrt{\frac{(2l+1)(2l'+1)(2l''+1)}{4\pi}} \begin{pmatrix} l & l' & l'' \\ m & -m' & m'' \end{pmatrix} \begin{pmatrix} l & l' & l'' \\ 0 & 0 & 0 \end{pmatrix}, \end{aligned} \quad (9)$$

with $\begin{pmatrix} p_1 & p_2 & p_3 \\ s_1 & s_2 & s_3 \end{pmatrix}$ denoting the Wigner 3-j symbol. The latter vanishes except when $s_1 + s_2 + s_3 = 0$ and the numbers p_j satisfy a triangle inequality: $p_i \leq p_j + p_k$, with $i, j, k = 1, 2, 3$ but different among them. In particular, due to the first Wigner 3-j symbol appearing in Eq. (9), we have always $m' = m + m''$.

Equation (8) simplifies when the radial dependence of the potential is of the form $d_{l'',m''}(r) = \tilde{d}_{l'',m''}/r^\chi$ with $\tilde{d}_{l'',m''}$ constant and $\chi \in \mathbb{N}$. In such a case one has

$$R_{l,l',l'',m''}(k) = \tilde{d}_{l'',m''} k^{\chi-3} \mathcal{I}_{\chi-2}^{l,l'}, \quad (10)$$

where we defined $\mathcal{I}_\beta^{l,l'} = \int dx j_l(x) j_{l'}(x) x^{-\beta}$ which is characterized by the following recurrence formula, inherited from the properties of the spherical Bessel functions:

$$\begin{aligned} \mathcal{I}_\beta^{l,l'} &= \frac{1}{(2l'+1)} \left(\mathcal{I}_{\beta-1}^{l,l'-1} + \mathcal{I}_{\beta-1}^{l,l'+1} \right), \quad \text{with} \\ \mathcal{I}_0^{l,l'} &= \int dx j_l(x) j_{l'}(x) = \frac{\sin\left[\frac{\pi}{2}(l-l')\right]}{(l-l')(l+l'+1)}. \end{aligned} \quad (11)$$

Moreover, here we notice that, in the perturbative treatment discussed below, the dominant terms $R_{l,l',l'',m''}(k)$ are those with l and l' small and with a small difference ($l' - l$); the others are suppressed by the larger values of the denominators in Eq. (11). Given Eq. (9) and Eq. (10), one has determined the terms of Eq. (7). Once one takes the square modulus of the latter and integrates it as in Eq. (3), one gets the rotational decoherence rate.

Since the decomposition in Eq. (6) is fully general and it can be applied to any potential, the expression in Eq. (7) for $\Delta f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')$ under the Born approximation is general as well. In particular, it provides a perturbative technique results for potentials whose exact expressions cannot be computed. Indeed, one can approximate the potential $V(\mathbf{r})$ to the first terms of the sum in Eq. (6):

$$V(\mathbf{r}) \simeq d_{0,0}(r) Y_{0,0}(\hat{\mathbf{r}}) + \sum_{m''=-1}^1 d_{1,m''}(r) Y_{1,m''}(\hat{\mathbf{r}}) + \dots, \quad (12)$$

and still obtain an analytical expression for Λ_R as well as the corresponding decoherence rate Λ_T for the translational case. For each term, one obtains the corresponding $R_{l,l',l'',m''}(k)$ from Eq. (8), which, together with Eq. (9), determines Eq. (7) and consequently the rotational decoherence rate in Eq. (3). In what follows, we study explicitly the first two contributions with two explicit cases.

0-th order: Spherical interaction.— Consider the simple case of an interaction exhibiting spherical symmetry. The potential is given only by the first term in Eq. (12). This means that $l'' = 0$ and $m'' = 0$ in the third sum in Eq. (7), and, correspondingly, Eq. (9) gives that $G_{l,m,l',m',0,0}$ is proportional to

$$\begin{pmatrix} l & l' & 0 \\ m & -m' & 0 \end{pmatrix} \begin{pmatrix} l & l' & 0 \\ 0 & 0 & 0 \end{pmatrix} = \frac{(-1)^m}{(2l+1)} \delta_{l,l'} \delta_{m,m'}. \quad (13)$$

Due to the presence of $(1 - e^{i\omega(m-m')})$ in Eq. (9), it follows that all $G_{l,m,l',m',0,0}$ are zero, leading to $\Lambda_R = 0$.

This result is not unexpected. Indeed, the symmetry of the interaction potential makes $f(\mathbf{k}'_\omega, \mathbf{p}'_\omega)$ independent from ω , giving $\Delta f^\omega(\mathbf{k}', \mathbf{p}') = 0$. Physically, the symmetrical interaction between the system and its environment means that the system interacts effectively as it was spherical, even if it is not.

1-st order: Dipole-dipole interaction.— The first non-zero contributions to Λ_R are given by the second term in Eq. (12). These are three contributions with $l'' = 1$ and $m'' = -1, 0, 1$. Thus, in Eq. (7), one can substitute $\sum_{l'',m''} \mathcal{S}_{l,m,l',m',l'',m''}$ with $\sum_{m''=-1}^1 \mathcal{S}_{l,m,l',m',1,m''}$, where \mathcal{S} denotes the terms of the sum. For each m'' , one determines the corresponding $G_{l,m,l',m',1,m''}(\omega)$ through Eq. (9). Due to the structure of the Wigner 3-j symbols, they are non vanishing only when $l' = l \pm 1$ and

$m' = m + m''$, thus imposing $\sum_{l'm'} \mathcal{S}_{l,m,l',m',1,m''} \rightarrow \sum_{s=-1,1} \mathcal{S}_{l,m,l+s,m+m'',1,m''} \theta_{l+s}$ in Eq. (7), where $\theta_{x \geq 0} = 1$ and $\theta_{x < 0} = 0$.

We see that, by using the expansion up to the first order in spherical harmonics of the potential, Eq. (7) is strongly simplified:

$$\int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}' |\Delta f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2 = \frac{64\pi^2 m_{\text{gas}}^2}{\hbar^4} \sum_{lm} \sum_{s=-1,1} \sum_{m''=-1}^1 |R_{l,l+s,1,m''}(k) G_{l,m,l+s,m+m'',1,m''}(\omega)|^2, \quad (14)$$

where we took into account that $\{Y_{l+s,m+m''}(\hat{\mathbf{p}}')\}$ and $\{Y_{l,m}^*(\hat{\mathbf{k}}')\}$ are two sets of orthonormal functions.

The corresponding coefficients $R_{l,l+s,1,m''}(k)$ depend on the radial behaviour of $d_{1,m''}(r)$ as in Eq. (8). As an explicit example, consider the interaction of a magnetic dipole system with an environment made of magnetic dipoles, whose form reads [49]

$$V(r, \hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2) = \frac{\mu_0 \gamma_1 \gamma_2}{4\pi r^3} \sum_{m''=-1}^1 a_{m''} Y_{1,m''}^*(\hat{\mathbf{r}}_1) Y_{1,m''}(\hat{\mathbf{r}}_2), \quad (15)$$

where γ_i is the modulus of the i -th dipole moment, $a_{\pm 1} = 1$, $a_0 = -2$, and r is the distance between the two dipoles. Here, $\hat{\mathbf{r}}_1$ identifies the orientation of the system, while $\hat{\mathbf{r}}_2$ that of the environmental dipole. The generalization to

$\sum_{l'=0}^{+\infty} \sum_{m'=-l'} \sum_{l''=0}^{+\infty} \sum_{m''=-l''} \mathcal{S}_{l,m,l',m',l'',m''}$ reduces to $\sum_{s=-1,1} \sum_{m''=-1}^1 \mathcal{S}_{l,m,l+s,m+m'',1,m''} \theta_{l+s}$. By merging these results with Eq. (7), taking its square modulus and performing the angular integration we find

other dipole-dipole interactions is straightforward. Such a potential can be expressed as in Eq. (6), with the only contributions given by $d_{1,m''}(r) = \tilde{d}_{1,m''}/r^3$ with $m'' = -1, 0, 1$ and

$$\tilde{d}_{1,m''} = (-)^{m''+1} \frac{\mu_0}{4\pi} \gamma_1 \gamma_2 Y_{1,m''}^*(\hat{\mathbf{r}}_1). \quad (16)$$

Correspondingly, Eq. (10) reduces to $R_{l,l+s,1,m''}(k) = \tilde{d}_{1,m''} \mathcal{I}_1^{l,l+s}$, giving

$$R_{l,l+s,1,m''}(k) = \frac{2 \cos(\frac{\pi}{2}s) \tilde{d}_{1,m''}}{(s-1)(s+1)(2l+s)(2l+s+2)}. \quad (17)$$

Once $R_{l,l+s,1,m''}(k)$ and $G_{l,m,l+s,m+m'',1,m''}(\omega)$ are determined, see Table I, we substitute them in Eq. (14) and get

$$\int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}' |\Delta f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2 = \frac{48\pi^3}{\hbar^4} m_{\text{gas}}^2 \sin^2(\frac{\omega}{2}) (|\tilde{d}_{1,1}|^2 + |\tilde{d}_{1,-1}|^2) \sum_{l,m} \frac{(l^2 + 2lm + l + m^2 + m + 1)}{[4l(l+2) + 3]^3}. \quad (18)$$

By summing the series and averaging the above expression over the possible angular configurations of the system, we find

$$\langle \int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}' |\Delta f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2 \rangle = \frac{m_{\text{gas}}^2 \mu_0^2 \gamma_1^2 \gamma_2^2}{\alpha \hbar^4} \sin^2(\frac{\omega}{2}), \quad (19)$$

with $\alpha \simeq 1.24$, and we used Eq. (16).

By assuming that the environment is in thermal equilibrium at the temperature T , then $\mu(k)$ is given by the Maxwell-Boltzmann distribution [4]

$$\mu(k) = \left(\frac{\hbar^2}{2\pi m_{\text{gas}} k_B T} \right)^{3/2} \exp\left(-\frac{\hbar^2 k^2}{2m_{\text{gas}} k_B T} \right). \quad (20)$$

Thus, by substituting $\rho(k) = 4\pi k^2 \mu(k)$ in Eq. (3), we get

$$\Lambda_R = \frac{m_{\text{gas}}^{3/2}}{(2\pi)^{3/2} \alpha \hbar^4} \mu_0^2 \gamma_1^2 \gamma_2^2 n \sqrt{k_B T} \sin^2(\frac{\omega}{2}). \quad (21)$$

This is an exact result. The rotational decoherence rate Λ_R depends on \sqrt{T} and the angular superposition distance through $\sin^2(\omega/2)$: it disappears for $\omega = 0$ and it is maximum for $\omega = \pi$, which are respectively the cases of an aligned and anti-aligned superposition.

Dipole-dipole interaction: translational case. – For a comparison, we compute the translational decoherence rate for dipole-dipole interaction. The translational rate can be obtained applying the following substitution in

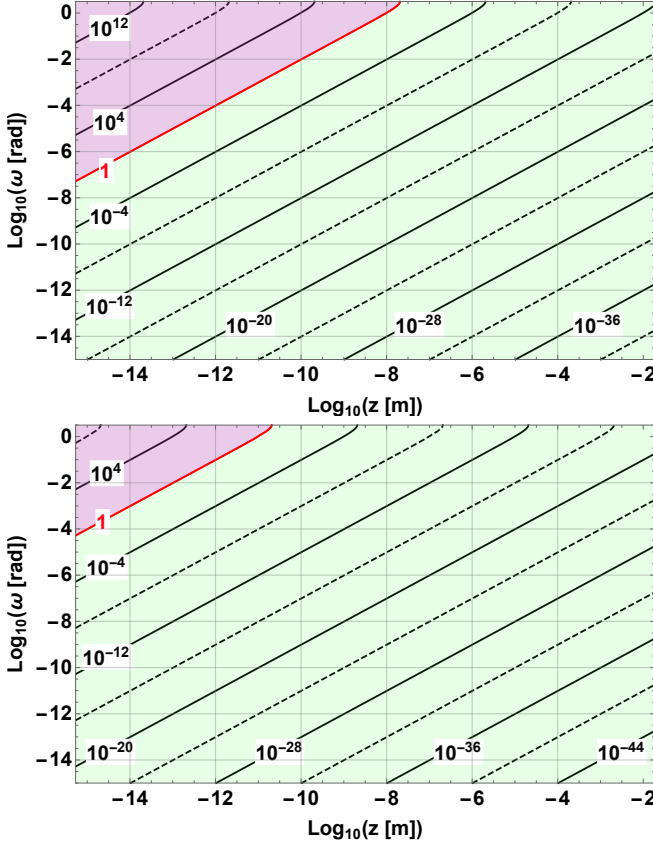


FIG. 1: Comparison of the rotational and translational decoherence rates. The level curves of Λ_R/Λ_T are reported as functions of the superposition angle ω and distance z . Here we took as reference values $T = 10^{-4}$ K (top panel) and $T = 100$ K (bottom panel). The gas mass is taken as $m_{\text{gas}} \simeq 10^{-26}$ kg (\sim light molecule).

TABLE I: Only non-vanishing terms $G_{l,m,l',m',1,m''}$ and corresponding values of $R_{l,l',1,m''}$ for the dipole-dipole interaction, as defined in Eq. (9) and Eq. (17) respectively. For the rotational case we have only the contributions from $m'' = +1$ and $m'' = -1$, while for the translational case we also have the contribution from $m'' = 0$. Here we have: $l' = l + s$, $m' = m + m''$. Moreover, $\theta_{x>0} = 1$ and $\theta_{x<0} = 0$.

s	m''	$(-)^m (1 - e^{i\omega}) G_{l,m,l+s,m+m'',1,m''}$	$R_{l,l+s,1,m''}$
-1	-1	$-\frac{i}{2} \sqrt{\frac{3(l+m-1)(l+m)}{2\pi(4l^2-1)}} \theta_{l+m-2} \theta_{l-1}$	$\tilde{d}_{1,-1} \frac{\pi}{8l^2-2}$
-1	0	0	$\tilde{d}_{1,0} \frac{\pi}{8l^2-2}$
-1	1	$\frac{i}{2} e^{-i\omega} \sqrt{\frac{3(l-m-1)(l-m)}{2\pi(4l^2-1)}} \theta_{l-m-2} \theta_{l-1}$	$\tilde{d}_{1,1} \frac{\pi}{8l^2-2}$
1	-1	$\frac{i}{2} \sqrt{\frac{3(l-m+1)(l-m+2)}{2\pi(4l(l+2)+3)}}$	$\tilde{d}_{1,-1} \frac{\pi}{8l^2+16l+6}$
1	0	0	$\tilde{d}_{1,0} \frac{\pi}{8l^2+16l+6}$
1	1	$-\frac{i}{2} e^{-i\omega} \sqrt{\frac{3(l+m+1)(l+m+2)}{2\pi(4l(l+2)+3)}}$	$\tilde{d}_{1,1} \frac{\pi}{8l^2+16l+6}$

Eq. (3):

$$\frac{1}{2} |\Delta f^\omega(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2 \rightarrow |f(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2 \left(1 - e^{ik(\hat{\mathbf{k}}-\hat{\mathbf{p}}') \cdot (\mathbf{x}-\mathbf{x}')}\right). \quad (22)$$

Following a similar strategy as described for the rotational case, in the short length limit ($kz \ll 1$ with $\mathbf{z} = (\mathbf{x} - \mathbf{x}')$ and $z = |\mathbf{z}|$) we obtain

$$\begin{aligned} & \langle \int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}' |f(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2 (1 - e^{ik(\hat{\mathbf{k}}-\hat{\mathbf{p}}') \cdot (\mathbf{x}-\mathbf{x}')}) \rangle \\ &= \frac{m_{\text{gas}}^2 \mu_0^2 \gamma_1^2 \gamma_2^2}{\alpha_1 \hbar^4} k^2 z^2, \end{aligned} \quad (23)$$

with $\alpha_1 \simeq 5.27$. We thus find the translational decoherence rate

$$\Lambda_T = \left(\frac{2}{\pi}\right)^{3/2} \frac{m_{\text{gas}}^{5/2}}{\alpha_1 \hbar^6} \mu_0^2 \gamma_1^2 \gamma_2^2 n (k_B T)^{3/2} z^2, \quad (24)$$

which is proportional to $T^{3/2}$ and depends on $|\mathbf{x} - \mathbf{x}'|^2$.

We can now compute the ratio of the two decoherence rates, which reads

$$\Lambda_R/\Lambda_T = \frac{\alpha_1}{\alpha} \frac{\hbar^2}{8m_{\text{gas}} k_B T} \frac{\sin^2(\omega/2)}{z^2}. \quad (25)$$

The latter depends on the superposition distances as $\sin^2(\omega/2)/z^2$, but most importantly, it scales with the inverse temperature. Figure 1 shows the ratio of the two decoherence rates for two values of the temperature of the environment (10^{-4} K and 100 K) varying ω and z . As it is clear from Figure 1, the ratio between the two decoherence rates decreases by increasing the temperature of the environment, while both rates increase with the temperature. Moreover, one can conclude that for a given temperature, the rotational decoherence time ($1/\Lambda_R$) can be much longer than the translational decoherence time ($1/\Lambda_T$). This corresponds to the green region in Fig. 1.

Conclusions.— Decoherence is an unavailable feature of quantum system and, ultimately, sets the limits to the applicability of quantum mechanics while moving towards the macroscopic realm. Various investigations, from quantum foundations to applications as quantum technologies, are inevitably influenced and disturbed by the environmental decoherence. Although one can try to minimize such decoherence effects, for example by developing experiments at low temperatures in ultra-high vacuum, the identification of alternative paths for obtaining longer decoherence time scales would be a game-changer. The avenue of levitated systems opens vast possibilities in this respect. Among them, the exploitation of rotational degrees of freedom could be a suitable solution for extending the decoherence time and thus enabling various possible applications of quantum mechanics to more macroscopic level than before.

In this work, we proposed a general approach to quantify the environmental decoherence effects on a quantum system prepared in a superposition of rotational degrees of freedom. We show that our proposed approach can be suitably applied to any potential that can be expressed in terms of spherical harmonics. For instance, we studied an explicit example: we considered a magnetic dipole system coupled to an environment made of magnetic dipoles. The electrical counterpart easily follows. We obtained the explicit form of the rotational decoherence rate for such a system. By applying the same approach, we also evaluated the translational decoherence rate for such interaction and made a comparison of the two. It results that rotational degrees of freedom are far less influenced by decoherence than translational ones. The advantage of rotational degrees of freedom is even stronger when moving to higher temperatures, as it is reported in Figure 1. Thus, our approach can be of strong interest for the calibration of decoherence effects also beyond what is usually considered the quantum realm.

Acknowledgments.— MC and AB acknowledge financial support from the H2020 FET Project TEQ (Grant No. 766900). HRN acknowledges financial support from the Ministry of Science, Research and Technology of IRAN, and hospitality from the University of Trieste, where part of this work was carried out. AB acknowledges financial support from the COST Action QTSspace (CA15220), INFN, and the University of Trieste.

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Supplemental Material

Derivation of the master equation

We assume that at time $t = 0$ the system and the environment are decoupled, and the total initial state is $\hat{\rho}_T = \hat{\rho}_S \otimes \hat{\rho}_E$, where $\hat{\rho}_E$ is the state of the environment. Starting from the configuration Ω , a scattering process at time t can be described as

$$|\Omega\rangle \otimes |\chi\rangle \xrightarrow{\text{scattering}} |\Omega\rangle \otimes \hat{S}_\Omega |\chi\rangle, \quad (26)$$

where $|\chi\rangle$ is a generic state of the environment and the recoil-less limit is considered ($M \gg m_{\text{gas}}$, where m_{gas} is the mass of the environmental particle). In this case, the scattering operator \hat{S}_Ω acts on the environmental state only. \hat{S}_Ω can be related to the standard unitary scattering operator \hat{S}_0 acting in $\Omega = \mathbf{0}$, through a rotation from the configuration $|\mathbf{0}\rangle$ to $|\Omega\rangle$: $\hat{S}_\Omega = \hat{D}_E(\Omega)\hat{S}_0\hat{D}_E^\dagger(\Omega)$.

At time t , after the scattering process has taken place, the system matrix elements change to $\rho_s(\Omega, \Omega')\eta(\Omega, \Omega')$, where

$$\eta(\Omega, \Omega') = \text{Tr}^{(B)} \left[\hat{\rho}_E \hat{S}_\Omega^\dagger \hat{S}_\Omega \right]. \quad (27)$$

To explicitly evaluate the partial trace over the degrees of freedom of the environment, we consider the total system as confined in a box of volume V , and we assume a thermal equilibrium for the environment, given by the state

$$\hat{\rho}_E = \frac{(2\pi)^3}{V} \int d k k^2 \mu(k) \sum_{l=0}^{+\infty} \sum_{m=-l}^l |k, l, m\rangle \langle k, l, m|, \quad (28)$$

where, we assumed that the momentum distribution of the environmental particles is invariant under rotations, thus $\mu(\mathbf{k}) = \mu(k)$. Here, $|k, l, m\rangle$ is the common eigenstate of the momentum $\hat{\mathbf{P}}^2$, the total angular momentum $\hat{\mathbf{L}}^2$ and its z component \hat{L}_z of the environmental particle. In particular, the relation between the usual momentum eigenstate $|\mathbf{p}\rangle$ and $|k, l, m\rangle$ is given by [47]:

$$\langle \mathbf{p} | k, l, m \rangle = \delta(p - k) Y_{l,m}(\hat{\mathbf{p}}) / p, \quad (29)$$

where $Y_{l,m}(\hat{\mathbf{p}})$ denotes the spherical harmonic and $\hat{\mathbf{p}} = \mathbf{p}/p$.

To be quantitative, let us consider the case where the system is in a superposition of angular configurations obtained only from rotations around the z axis. This will be also the case of interest in most experimental setups, where one focuses on one direction per time. The state of the system can be then identified by $|\alpha\rangle = \hat{D}_E(\alpha) |\mathbf{0}\rangle$, with $\hat{D}_E(\alpha) = \exp(-\frac{i}{\hbar} \hat{L}_z \alpha)$. Thus, we have

$$\eta(\alpha, \alpha') = \frac{(2\pi)^3}{V} \int d k k^2 \mu(k) \sum_{l,m} \langle k, l, m | \hat{S}_\alpha^\dagger \hat{S}_\alpha | k, l, m \rangle, \quad (30)$$

where the relation $\hat{D}_E(\alpha) |k, l, m\rangle = e^{-im\alpha} |k, l, m\rangle$ holds. We express the scattering matrix as

$$\hat{S}_\alpha = \hat{D}_E(\alpha) (1 + i\hat{T}) \hat{D}_E^\dagger(\alpha), \quad (31)$$

where \hat{T} is the T-matrix of scattering theory [47]. Due to the unitarity of \hat{S}_α , one has that $-i(\hat{T}^\dagger - \hat{T}) = \hat{T}^\dagger \hat{T}$. By exploiting these relations one finds

$$\eta(\alpha, \alpha') = 1 - \frac{(2\pi)^3}{V} \int d k k^2 \mu(k) \sum_{lm} \int d p p^2 \sum_{l'm'} \left(1 - e^{-i(m-m')(\alpha-\alpha')} \right) \langle k, l, m | \hat{T}^\dagger | p, l', m' \rangle \langle p, l', m' | \hat{T} | k, l, m \rangle, \quad (32)$$

where the matrix elements of \hat{T} can be expressed in the momentum space as [47]:

$$\langle \mathbf{k}'' | \hat{T} | \mathbf{p}'' \rangle = -\delta(k'' - p'') f(\mathbf{k}'', \mathbf{p}'') / (2\pi p''), \quad (33)$$

where $f(\mathbf{k}'', \mathbf{p}'')$ is the scattering amplitude. This, together with Eq. (29), brings to

$$\langle p, l', m' | \hat{T} | k, l, m \rangle = - \int d\hat{\mathbf{k}}'' \int d\mathbf{p}'' Y_{l,m}(\hat{\mathbf{k}}'') Y_{l',m'}^*(\hat{\mathbf{p}}'') f(k\hat{\mathbf{k}}'', k\hat{\mathbf{p}}'') \frac{\delta(p-k)}{2\pi}. \quad (34)$$

Consequently, one obtains

$$\begin{aligned} \eta(\alpha, \alpha') = & 1 - \frac{t}{V} \int dk \frac{\hbar k}{M} k^2 \mu(k) \sum_{lm} \sum_{l'm'} \left(1 - e^{-i(m-m')(\alpha-\alpha')} \right) \\ & \int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}' \int d\hat{\mathbf{k}}'' \int d\hat{\mathbf{p}}'' Y_{l,m}(\hat{\mathbf{k}}'') Y_{l',m}^*(\hat{\mathbf{k}}') Y_{l',m'}(\hat{\mathbf{p}}') Y_{l',m'}^*(\hat{\mathbf{p}}'') f^*(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}') f(k\hat{\mathbf{k}}'', k\hat{\mathbf{p}}''), \end{aligned} \quad (35)$$

where we exploited the normalization of the squared Dirac- δ :

$$(\delta(p-k))^2 \sim \frac{\hbar p t}{2\pi M} \delta(p-k), \quad (36)$$

which is valid under the assumption that the decoherence time is larger than the time of the collision [4] – usually considered instantaneous.

We now take into account that the spherical harmonics are of the form

$$Y_{l,m}(\hat{\mathbf{k}}) = (-)^m \sqrt{\frac{(2l+1)(l-m)!}{2\pi(l+m)!}} F_{l,m}(\theta_{\mathbf{k}}) e^{im\phi_{\mathbf{k}}}, \quad (37)$$

where $(\theta_{\mathbf{k}}, \phi_{\mathbf{k}})$ identify $\hat{\mathbf{k}}$ and $F_{lm}(\theta)$ is the Legendre polynomial. Consequently, we can rewrite

$$Y_{l,m}(\hat{\mathbf{k}}'') e^{-im\alpha} = Y_{l,m}(\hat{\mathbf{k}}''_{\alpha}), \quad (38)$$

where $\hat{\mathbf{k}}''_{\alpha}$ is obtained from $\hat{\mathbf{k}}''$ after a rotation α around z . Then, the phase in the parenthesis in Eq. (35) can be absorbed in the spherical harmonics, and we find that

$$e^{-i(m-m')(\alpha-\alpha')} Y_{l,m}(\hat{\mathbf{k}}'') Y_{l',m}^*(\hat{\mathbf{k}}') Y_{l',m'}(\hat{\mathbf{p}}') Y_{l',m'}^*(\hat{\mathbf{p}}'') = Y_{l,m}(\hat{\mathbf{k}}''_{\alpha}) Y_{l',m}^*(\hat{\mathbf{k}}'_{\alpha'}) Y_{l',m'}(\hat{\mathbf{p}}'_{\alpha'}) Y_{l',m'}^*(\hat{\mathbf{p}}''_{\alpha'}). \quad (39)$$

Now, by exploiting the orthonormality of the spherical harmonics [50]

$$\sum_{lm} Y_{l,m}(\hat{\mathbf{k}}) Y_{l,m}^*(\hat{\mathbf{k}}') = \delta(\hat{\mathbf{k}} - \hat{\mathbf{k}}'), \quad (40)$$

we obtain

$$\eta(\alpha, \alpha') = 1 - \frac{t}{V} \int dk \frac{\hbar k}{m_{\text{gas}}} k^2 \mu(k) \int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}' f(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}') \left(f^*(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}') - f^*(k\hat{\mathbf{k}}'_{\omega}, k\hat{\mathbf{p}}'_{\omega}) \right). \quad (41)$$

The final result is reported in Eq. (2), where one exploits $\eta(\alpha, \alpha') = 1 - t\Lambda_{\text{R}}/N$, and Λ_{R} is the N -particle decoherence rate reported in Eq. (3). To derive the latter, we also considered that the state is self-adjoint $\hat{\rho}_{\text{s}} = \hat{\rho}_{\text{s}}^{\dagger}$, which implies $\eta(\alpha, \alpha') = \eta^*(\alpha', \alpha)$.

Comparison with the translational case. – It is worth to notice that the expression for Λ_{R} in Eq. (3) has the same formal structure of the master equation describing decoherence for the translational degrees of freedom [44]:

$$\Lambda_{\text{T}}(\mathbf{x}, \mathbf{x}') = n \int dk v(k) \rho(k) \frac{\int d\hat{\mathbf{k}}' \int d\hat{\mathbf{p}}'}{4\pi} |f(\mathbf{k}, k\hat{\mathbf{p}}')|^2 \left(1 - e^{i(\mathbf{k}-k\hat{\mathbf{p}}') \cdot (\mathbf{x}-\mathbf{x}')} \right). \quad (42)$$

Such an equation is obtained by replacing $\frac{1}{2} |\Delta f^{\omega}(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}')|^2$ in Eq. (3) with $f(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}') \left(f^*(k\hat{\mathbf{k}}', k\hat{\mathbf{p}}') - f^*(k\hat{\mathbf{k}}'_{\mathbf{x}-\mathbf{x}'}, k\hat{\mathbf{p}}'_{\mathbf{x}-\mathbf{x}'}) \right)$. Here $\mathbf{k}_{\mathbf{x}-\mathbf{x}'}$ is the vector \mathbf{k} translated in space by the quantity $\mathbf{x} - \mathbf{x}'$. This result can be understood once we consider the expression for the scattering amplitude generated by the potential $V(\mathbf{r})$, under the Born approximation [44], which is given by Eq. (5) with $\omega = 0$, and substitute to $\hat{\mathbb{S}}_{\alpha}$ in Eq. (30) the scattering operator implementing the translation in space $\hat{\mathbb{S}}_{\mathbf{x}} = e^{-i\hat{\mathbf{p}} \cdot \mathbf{x}} \hat{\mathbb{S}}_0 e^{i\hat{\mathbf{p}} \cdot \mathbf{x}}$. In this way, one obtains the expression in Eq. (42).

Contributions to the dipole-dipole decoherence rate

Here we define

$$G_{l,m,l',m',l'',m''}^\alpha = (-)^{m'} i^{l'-l} \left(1 - \alpha e^{i\omega(m-m')}\right) \sqrt{\frac{(2l+1)(2l'+1)(2l''+1)}{4\pi}} \begin{pmatrix} l & l' & l'' \\ m & -m' & m'' \end{pmatrix} \begin{pmatrix} l & l' & l'' \\ 0 & 0 & 0 \end{pmatrix}. \quad (43)$$

With $\alpha = 1$, this reduces to Eq. (9); while with $\alpha = 0$, it gives the corresponding contribution for the translational case. The only non-vanishing contributions are reported in Table II. The terms there reported can be simplified by considering the following.

First, notice that for a generic function $F(l, m)$, the following relation holds

$$\sum_{l=0}^{+\infty} \sum_{m=-l}^l \theta_{l-m-2} F(l, m) = \sum_{l=1}^{+\infty} \sum_{m=-l}^{l-2} \theta_{l-m-2} F(l, m). \quad (44)$$

Then, by mapping $l \mapsto l_1 = l - 1$ and $m \mapsto m_1 = m + 1$, we find

$$\sum_{l=1}^{+\infty} \sum_{m=-l}^{l-2} \theta_{l-m-2} F(l, m) \mapsto \sum_{l_1=0}^{+\infty} \sum_{m_1=-l_1}^{l_1} \theta_{l_1-m_1} F(l_1 + 1, m_1 - 1), \quad (45)$$

where $l_1 - m_1 \geq 0$ and thus $\theta_{l_1-m_1} = 1$ for any choice of l_1 and m_1 . By applying this shift of the angular indices in the entries in Table II for $s = -1$ with $m'' = +1$ and $m'' = -1$, one finds that they are like the corresponding ones for $s = 1$. With the same idea one proves that the term with $s = -1$ and $m'' = 0$ is equal to that with $s = 1$ and $m'' = 0$.

TABLE II: Only non-vanishing terms $G_{l,m,l',m',1,m''}^{\alpha=1}$ and corresponding values of $R_{l,l',1,m''}$ for the dipole-dipole interaction, as defined in Eq. (43) and Eq. (17) respectively. For the rotational case we have only the contributions from $m'' = +1$ and $m'' = -1$, while for the translational case we also have the contribution from $m'' = 0$. Here we have: $l' = l + s$, $m' = m + m''$. Moreover, $\theta_{x>0} = 1$ and $\theta_{x<0} = 0$.

s	m''	$G_{l,m,l+s,m+m'',1,m''}^{\alpha=1}$	$G_{l,m,l+s,m+m'',1,m''}^{\alpha=0}$	$R_{l,l+s,1,m''}$
-1	-1	$\frac{1}{2} i \sqrt{\frac{3}{2\pi}} (-1)^m (-1 + e^{i\omega}) \sqrt{\frac{(l+m-1)(l+m)}{4l^2-1}} \theta_{l+m-2} \theta_{l-1}$	$-\frac{1}{2} i \sqrt{\frac{3}{2\pi}} (-1)^m \sqrt{\frac{(l+m-1)(l+m)}{4l^2-1}} \theta_{l+m-2} \theta_{l-1}$	$\tilde{d}_{1,-1} \frac{\pi}{8l^2-2}$
-1	0	0	$\frac{1}{2} i \sqrt{\frac{3}{\pi}} (-1)^m \sqrt{\frac{l^2-m^2}{4l^2-1}} \theta_{l-1} \theta_{l-m-1} \theta_{l+m-1}$	$\tilde{d}_{1,0} \frac{\pi}{8l^2-2}$
-1	1	$-\frac{1}{2} i \sqrt{\frac{3}{2\pi}} (-1)^m (1 - e^{-i\omega}) \sqrt{\frac{(l-m-1)(l-m)}{4l^2-1}} \theta_{l-m-2} \theta_{l-1}$	$-\frac{1}{2} i \sqrt{\frac{3}{2\pi}} (-1)^m \sqrt{\frac{(l-m-1)(l-m)}{4l^2-1}} \theta_{l-1} \theta_{l-m-2}$	$\tilde{d}_{1,1} \frac{\pi}{8l^2-2}$
1	-1	$\frac{1}{2} i \sqrt{\frac{3}{2\pi}} (1 - e^{i\omega}) (-1)^m \sqrt{\frac{(l-m+1)(l-m+2)}{4l(l+2)+3}}$	$i \sqrt{\frac{3}{\pi}} 2^{-l-\frac{3}{2}} (-1)^{m-l} \sqrt{\frac{(l-m+1)(l-m+2)}{4l(l+2)+3}}$	$\tilde{d}_{1,-1} \frac{\pi}{8l^2+16l+6}$
1	0	0	$\frac{1}{2} i \sqrt{\frac{3}{\pi}} (-1)^m \sqrt{\frac{(l-m+1)(l-m+1)}{4l(l+2)+3}}$	$\tilde{d}_{1,0} \frac{\pi}{8l^2+16l+6}$
1	1	$\frac{1}{2} i \sqrt{\frac{3}{2\pi}} (1 - e^{-i\omega}) (-1)^m \sqrt{\frac{(l+m+1)(l+m+2)}{4l(l+2)+3}}$	$i \sqrt{\frac{3}{\pi}} 2^{-l-1} (-1)^{m-l} \sqrt{\frac{(l+m+1)(l+m+2)}{2(2l+1)(2l+3)}}$	$\tilde{d}_{1,1} \frac{\pi}{8l^2+16l+6}$