Rotational Friction and Diffusion of Quantum Rotors

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We present the Markovian quantum master equation describing rotational decoherence, friction, diffusion, and thermalization of planar, linear, and asymmetric rotors in contact with a thermal environment. It describes how an arbitrary initial rotation state decoheres and evolves toward a Gibbs-like thermal ensemble, as we illustrate numerically for the linear and the planar top, and it yields the expected rotational Fokker-Planck equation of Brownian motion in the semiclassical limit.

Introduction.—A quantum point particle moving in a thermal bath is subject to random interactions with the environmental degrees of freedom. They affect the particle in two ways: (i) initial superpositions of different positions quickly decohere and (ii) the particle gradually thermalizes with its surroundings. In many situations, the associated dynamics is well modeled by the Markovian master equation of quantum Brownian motion [1,2]. But what if the particle is not pointlike and hence able to rotate? How can the resulting rotational decoherence, friction, and diffusion be described quantum mechanically?

Beyond its conceptual significance, this question becomes increasingly relevant for state-of-the-art experiments. Numerous experimental studies demonstrate rotational manipulation and control of molecules [3–8] and recently also of nanoparticles [9–16]. Cooling the rotation state into the quantum regime was successfully implemented for small molecules [17–26], and is in reach for nano- to micrometer-sized objects [12,27,28]. Conceivable applications include orientation-dependent metrology [29–31], ultracold chemistry [32–34], highly sensitive torque sensors [12,15], realizations of a quantum heat engine [35], levitated nanomagnets [36], dissipative dynamics of angulons [37], tests of objective collapse models [38], and orientational quantum revivals [39,40]. The interpretation of such experiments will rely heavily on a theoretical assessment of the rotor dynamics in presence of an environment.

Here, we present the general Markovian quantum master equation describing rotational friction, diffusion, and thermalization of rigid rotors. It is the natural generalization of the master equation of Brownian center-of-mass motion [1,2,41], valid if the bath is sufficiently dilute or its temperature is high enough to warrant a Markovian description. Unlike the center-of-mass momentum, however, the angular momentum components do not commute, implying that the orientational degrees of freedom cannot be decoupled, and the friction and diffusion tensors depend necessarily on the particle orientation expressed in terms of rotation matrices. These characteristics of orientation and

rotation render their quantum dynamics substantially different from the center-of-mass motion, so that the presented master equation is not a straightforward extension thereof. As in the classical theory [42], it will turn out to be pertinent to use a coordinate-independent formulation in terms of rotation matrices and angular momentum vectors rather than the canonical phase space variables. The presented master equation reduces to pure orientational decoherence [43–45] in the high-temperature limit and to the expected Fokker-Planck equation [42] in the semiclassical limit.

Classical thermalization—It is useful to briefly review the classical description of rotational thermalization of a rigid body of orientation Ω (parametrized e.g., by the Euler angles) and angular momentum $\mathbf{J}$. In absence of an external torque, environment-induced friction and diffusion can be described by the stochastic differential equation $d\mathbf{J} = -\Gamma(\Omega)\mathbf{J}dt + d\mathbf{N}_t$. The first term accounts for rotational friction with the orientation-dependent friction tensor $\Gamma(\Omega) = R(\Omega)\Gamma_0 R^{\mathrm{T}}(\Omega)$. Here, the orthogonal matrix $R(\Omega)$ serves to rotate the particle from a reference orientation
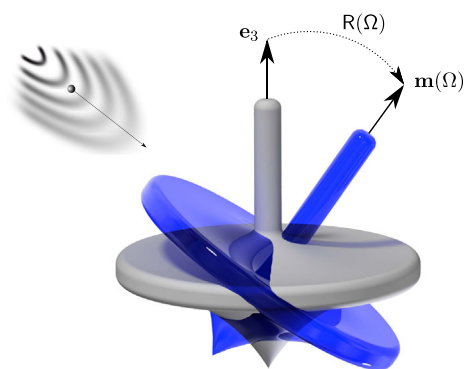

FIG. 1. A rigid rotor immersed in a thermal environment receives random angular momentum kicks analogous to the momentum kicks experienced by a Brownian particle. These interactions decohere an initial superposition of two different orientations and gradually thermalize the rotation state.

$\Omega = 0$ to the current one, see Fig. 1, and Γ_0 is the friction tensor at $\Omega = 0$. The strength and direction of the random angular momentum kicks $d\mathbf{N}_t = \sqrt{2D(\Omega)}d\mathbf{W}_t$ is determined by the diffusion tensor $D(\Omega)$ [45], while $d\mathbf{W}_t$ is a vector Wiener process. The angular momentum dynamics are complemented by the equation of motion for particle orientation, $dR(\Omega) = I^{-1}(\Omega)\mathbf{J} \times R(\Omega)dt$. Here, we introduced the tensor of inertia $I(\Omega)$ whose eigenvalues I_i are the moments of inertia. These two stochastic equations determine the Brownian rotation dynamics of an arbitrary particle.

The stochastic motion can be equivalently described by the deterministic evolution of the probability density $h_t(\Omega, \mathbf{J})$ [46–48]. It contains both the free rotational dynamics and a nonconservative part accounting for the interaction with the environment, $\partial_t h_t = \partial_t^{\text{rot}} h_t + \partial_t^{\text{nc}} h_t$. While the first part is determined by the Hamilton function $H = \mathbf{J} \cdot I^{-1}(\Omega)\mathbf{J}/2$, the second part takes the form of a Fokker-Planck equation,

$$\partial_t^{\text{nc}} h_t(\Omega, \mathbf{J}) = \nabla_{\mathbf{J}} \cdot [\Gamma(\Omega)\mathbf{J} h_t(\Omega, \mathbf{J})] + \nabla_{\mathbf{J}} \cdot D(\Omega)\nabla_{\mathbf{J}} h_t(\Omega, \mathbf{J}). \tag{1}$$

The dynamics of the first two moments of $\mathbf{J}$ follow directly as

$$\partial_t \langle \mathbf{J} \rangle = -\langle \Gamma(\Omega)\mathbf{J} \rangle, \tag{2a}$$

$$\partial_t \langle \mathbf{J}^2 \rangle = -2\langle \mathbf{J} \cdot \Gamma(\Omega)\mathbf{J} \rangle + 2\langle \text{Tr}[D(\Omega)] \rangle, \tag{2b}$$

$$\partial_t \langle \mathbf{J} \otimes \mathbf{J} \rangle = -\langle \Gamma(\Omega)\mathbf{J} \otimes \mathbf{J} + \mathbf{J} \otimes \mathbf{J}\Gamma^{\text{T}}(\Omega) \rangle + 2\langle D(\Omega) \rangle. \tag{2c}$$

As expected for Brownian motion, friction reduces the mean (angular) momentum (2a), while diffusion increases its variance and covariance with a constant rate determined by (2b) and (2c). [$\text{Tr}(\cdot)$ refers to the matrix trace, as opposed to the operator trace $\text{tr}(\cdot)$ used below.]

Using the fluctuation-dissipation relation $D(\Omega) = k_{\text{B}}T\Gamma(\Omega)I(\Omega)$ in Eq. (1), one finds that the rotor thermalizes toward the Gibbs state $\exp(-H/k_{\text{B}}T)/Z$ with mean energy

$$\langle H \rangle = \frac{1}{2}\langle \mathbf{J} \cdot I^{-1}(\Omega)\mathbf{J} \rangle = \frac{f}{2}k_{\text{B}}T, \tag{3}$$

where $f = \text{rank}[I(\Omega)]$ is the number of rotational degrees of freedom. For a given particle shape, the rotational friction tensor can be calculated microscopically from kinetic gas theory [42,49,50]. The Fokker-Planck description (1) allows general statements about thermalization [48], and is best suited for comparison with the quantized rotation dynamics.

Quantum-classical consistency demands that the quantum master equation of rotational friction and diffusion describes the same dynamics as Eq. (1) in the semiclassical limit. This means that the equations for the first and second moments of the angular momentum operator $\mathbf{J}$ (operators are denoted by sans-serif characters unless specified

otherwise) must coincide with their classical equivalents (2) up to corrections of order $\hbar$. Further, the steady state of the quantum master equation must approach the Gibbs state for large temperatures,

$$\rho_{\text{eq}} = \frac{1}{Z}e^{-H/k_{\text{B}}T} + \mathcal{O}(\hbar), \quad \text{with} \quad Z = \text{tr}(e^{-H/k_{\text{B}}T}), \tag{4}$$

implying the equipartition of energies (3) to lowest order in $\hbar$.

General master equation.—We now establish the quantum master equation $\partial_t \rho = -i[H, \rho]/\hbar + \mathcal{D}\rho$ describing rotational friction and diffusion through the dissipator $\mathcal{D}$. The latter can be heuristically derived from the Caldeira-Leggett equation [1,2] for N rigidly connected point particles. Denoting the position operators of the individual point particles by $\mathbf{r}_n$ and the momentum operators by $\mathbf{p}_n$, the master equation reads

$$\partial_t \rho = -\frac{i}{\hbar}\left[H + \frac{1}{2}\sum_{n=1}^{N}\gamma_n(\mathbf{r}_n \cdot \mathbf{p}_n + (\mathbf{r}_n \cdot \mathbf{p}_n)^{\dagger}), \rho\right]$$
$$+ \frac{2k_{\text{B}}T}{\hbar^2}\sum_{n=1}^{N}m_n\gamma_n\left(\mathbf{L}_n \cdot \rho\mathbf{L}_n^{\dagger} - \frac{1}{2}\{\mathbf{L}_n^{\dagger} \cdot \mathbf{L}_n, \rho\}\right), \tag{5}$$

with $\mathbf{L}_n = \mathbf{r}_n + i\hbar\mathbf{p}_n/4m_nk_{\text{B}}T$ and individual damping rates $\gamma_n > 0$.

In order to account for the fact that all point particles are rigidly connected, we introduce a quantum version of the rigid body approximation [51]. Denoting the orientation operator as Ω, it takes the center of mass to be at rest, $\mathbf{r}_n = R(\Omega)\mathbf{r}_n^{(0)}$, and replaces the momenta $\mathbf{p}_n$ by operator-valued classical expressions for the velocity of the nth particle multiplied by its mass, $-m_nR(\Omega)\mathbf{r}_n^{(0)}\times R(\Omega)I_0^{-1}R^{\text{T}}(\Omega)\mathbf{J}$, with $\mathbf{r}_n^{(0)}$ the nth particle position and I_0 the tensor of inertia at orientation $\Omega = 0$. While the new momenta are non-Hermitian, they ensure both that $\mathbf{J} = \sum_n \mathbf{r}_n \times \mathbf{p}_n$ still holds after the replacements and that the energy renormalization in (5) vanishes. Notwithstanding the heuristic nature of this quantum rigid body approximation, we will see that the resulting master equation has all desired properties.

Using the quantum rigid body approximation in (5) yields

$$\partial_t \rho = -\frac{i}{\hbar}[H, \rho] + \frac{2k_{\text{B}}T}{\hbar^2}\sum_{n=1}^{N}m_n\gamma_n\left[R(\Omega)\mathbf{K}_n \cdot \rho\mathbf{K}_n^{\dagger}R^{\text{T}}(\Omega)\right.$$
$$\left. -\frac{1}{2}\{\mathbf{K}_n^{\dagger} \cdot \mathbf{K}_n, \rho\}\right], \tag{6}$$

where $\mathbf{K}_n = \mathbf{r}_n^{(0)} + i\hbar I_0^{-1}R^{\text{T}}(\Omega)\mathbf{J} \times \mathbf{r}_n^{(0)}/4k_{\text{B}}T$. Subsuming the sum into the positive tensor

$$\widetilde{D}_0 = k_{\text{B}}T\sum_{n=1}^{N}m_n\gamma_n\mathbf{r}_n^{(0)} \otimes \mathbf{r}_n^{(0)} \equiv \sum_{k=1}^{3}\tilde{D}_k\mathbf{d}_k^{(0)} \otimes \mathbf{d}_k^{(0)}, \tag{7}$$

one obtains the master equation

$$\mathcal{D}\rho = \sum_{k=1}^{3} \frac{2\tilde{D}_k}{\hbar^2}\left[\mathbf{A}_k \cdot \rho\mathbf{A}_k^\dagger - \frac{1}{2}\{\mathbf{A}_k^\dagger \cdot \mathbf{A}_k, \rho\}\right]. \quad (8a)$$

It involves scalar products of the vectorial Lindblad operators

$$\mathbf{A}_k = \mathbf{d}_k(\Omega) - \frac{i\hbar}{4k_\mathrm{B}T}\mathbf{d}_k(\Omega) \times I^{-1}(\Omega)\mathbf{J}, \quad (8b)$$

with $\mathbf{d}_k(\Omega) = R(\Omega)\mathbf{d}_k^{(0)}$, where $\mathbf{d}_k^{(0)}$ are the three normalized orthogonal eigenvectors of $\tilde{D}_0$. Equation (9) below shows that the $\mathbf{d}_k(\Omega)$ are the eigenvectors of the diffusion tensor whose eigenvalues D_k fix the $\tilde{D}_k$.

Equations (8) specify the quantum Brownian rotation dynamics expected for asymmetric rigid tops. They give rise to the moment dynamics (2) to leading order in $\hbar$, and they ensure that ρ approaches the steady state (4) with energy expectation value (3) for small $\hbar^2/k_\mathrm{B}TI_\mathrm{min}$, with I_min the minimal moment of inertia. All this can be checked by straightforward but lengthy calculations taking into account that the $\mathbf{A}_k$ and their components do not commute, as explained in [52].

While the first term of the Lindblad operators (8b) represents the particle orientation through $\mathbf{d}_k(\Omega)$, the second is proportional to the quantized (but not hermitized) rate of change, $\dot{\mathbf{d}}_k(\Omega) = I^{-1}(\Omega)\mathbf{J} \times \mathbf{d}_k(\Omega)$. Equation (8b) is thus the rotational analog of the Lindblad operator $\mathsf{L} = \mathsf{x} + i\hbar\mathsf{p}/4mk_\mathrm{B}T$ of one-dimensional center-of-mass thermalization in quantum Brownian motion [1,2,41,53]. In contrast to the latter, the three Lindblad operators (8b) do not commute, accounting for the facts that the three principal axes of a rigid rotor cannot be rotated independently and that the components of the angular momentum vector do not commute.

The vectors $\mathbf{d}_k(\Omega)$ are the eigenvectors of the diffusion tensor $D(\Omega)$, as can be demonstrated by calculating the second moments (2) using Eq. (8). Comparison with (2) shows that

$$D(\Omega) = \sum_{k=1}^{3} \tilde{D}_k[\mathbb{1} - \mathbf{d}_k(\Omega) \otimes \mathbf{d}_k(\Omega)], \quad (9)$$

with eigenvalues $D_k = \tilde{D}_i + \tilde{D}_j$, so that $\tilde{D}_k = (D_i + D_j - D_k)/2$, where (i, j, k) are permutations of (1,2,3). This relation implies that the master equation (8) is completely positive ($\tilde{D}_k \geq 0$) only if $D_i + D_j \geq D_k$ (even though the localization rate is always positive). The same inequality is implied by the corresponding classical derivation of Brownian motion [52], where a more general diffusion tensor can be obtained if the diffusion of the individual point particles is not isotropic.

It remains an open question how to extend this to the quantum regime.

The semiclassical limit of (8) gives the rotational Fokker-Planck equation (1) with diffusion tensor (9) and friction tensor $\Gamma(\Omega) = D(\Omega)I^{-1}(\Omega)/k_\mathrm{B}T$. This can be shown by adopting the treatment in Ref. [45], i.e., first expressing (8) in the quantum phase space of the orientation state [54,55], approximating the discrete angular momentum quantum numbers by continuous variables, and then evaluating the limit $\hbar \to 0$.

Another limiting case is that the rotor is tightly aligned by an external potential, so that its dynamics are librational rather than rotational. If the angle coordinates can then be approximated harmonically, a linearization of the rotation matrix in the angle operators yields Lindblad operators reminiscent of center-of-mass Brownian motion with positions and momenta replaced by angles and their canonically conjugate momenta.

In what follows we will specialize the master equation (8), which is valid for general rotors, to the cases of the linear and planar rigid tops and illustrate their thermalization dynamics.

Linear rotors.—The orientation of a linear rigid rotor is specified by the direction of its symmetry axis $\mathbf{m}(\Omega)$, so that $I(\Omega) = I[\mathbb{1} - \mathbf{m}(\Omega) \otimes \mathbf{m}(\Omega)]$. Accordingly, friction and diffusion orthogonal to the symmetry axis, for instance due to specular gas scattering [42], are described by the tensors $\Gamma(\Omega) = \Gamma[\mathbb{1} - \mathbf{m}(\Omega) \otimes \mathbf{m}(\Omega)]$ and $D(\Omega) = D[\mathbb{1} - \mathbf{m}(\Omega) \otimes \mathbf{m}(\Omega)]$ with $D = k_\mathrm{B}T\Gamma I$. This implies that one eigenvalue of the diffusion tensor is zero, while the two eigenvalues associated with the two directions perpendicular to $\mathbf{d}_1(\Omega) = \mathbf{m}(\Omega)$ are D.

Calculating $\tilde{D}_k$ according to (9) yields the dissipator

$$\mathcal{D}\rho = \frac{2D}{\hbar^2}\left[\mathbf{A} \cdot \rho\mathbf{A}^\dagger - \frac{1}{2}\{\mathbf{A}^\dagger \cdot \mathbf{A}, \rho\}\right], \quad (10a)$$

with the vectorial Lindblad operators

$$\mathbf{A} = \mathbf{m}(\Omega) - \frac{i\hbar}{4k_\mathrm{B}TI}\mathbf{m}(\Omega) \times \mathbf{J}. \quad (10b)$$

Inserting (10b) into (10a) yields

$$\mathcal{D}\rho = -\frac{i\Gamma}{2\hbar}[\mathbf{m}(\Omega) \times \mathbf{J} \cdot \rho\mathbf{m}(\Omega) + \mathbf{m}(\Omega) \cdot \rho\mathbf{J} \times \mathbf{m}(\Omega)]$$
$$\frac{2D}{\hbar^2}[\mathbf{m}(\Omega) \cdot \rho\mathbf{m}(\Omega) - \rho] + \mathcal{O}\left(\frac{\hbar^2}{k_\mathrm{B}TI}\right). \quad (11)$$

Here, the first term is independent of temperature, linear in $\mathbf{J}$, and describes rotational friction. The second term is linear in T (since $D = k_\mathrm{B}T\Gamma I$) and describes angular momentum diffusion as well as an exponential decay of the orientational coherences $\langle\Omega|\rho|\Omega'\rangle$ with the rate

$$F(\Omega, \Omega') = \frac{2D}{\hbar^2}[1 - \mathbf{m}(\Omega) \cdot \mathbf{m}(\Omega')]. \qquad (12)$$

The remaining terms in (11) are proportional to $1/T$, quadratic in J and ensure complete positivity; like in the center-of-mass case, they can be neglected for sufficiently large temperatures. The special case that a symmetry in the environmental interaction prevents the complete localization (12) can also be accounted for, as discussed in the Supplemental Material [52].

In order to study the thermalization dynamics described by (10), we solve the master equation numerically with the free Hamiltonian $\mathsf{H} = \mathsf{J}^2/2I$ and calculate analytically the corresponding steady state ρ_{eq}. The latter can be determined by noting that the equation $-i[\mathsf{H}, \rho_{\text{eq}}]/\hbar + \mathcal{D}\rho_{\text{eq}} = 0$ implies that ρ_{eq} is diagonal in the angular momentum basis, $\rho_{\text{eq}} = \sum_{\ell m} \rho_{\text{eq}}^{\ell m}|\ell m\rangle\langle\ell m|$. Then, the unitary part vanishes and $\mathcal{D}\rho_{\text{eq}} = 0$ yields a set of coupled equations for the coefficients $\rho_{\text{eq}}^{\ell m}$. It can be solved explicitly [52],

$$\rho_{\text{eq}}^{\ell m} = \frac{1}{Z}\binom{2\xi}{\ell}^2\binom{2\xi + \ell + 1}{\ell}^{-2} \qquad (13)$$

in terms of $\xi = 2Ik_B T/\hbar^2$. The steady state approaches the Gibbs state (4) for large temperatures, $\rho_{\text{eq}}^{\ell m} \sim \exp[-\ell(\ell + 1)/\xi]/Z$ as $\xi \to \infty$, as can be checked using Stirling's formula. From the existence of the steady state (13) it follows that the relative entropy $S(\rho\|\rho_{\text{eq}}) = -\text{tr}[\rho(\log\rho - \log\rho_{\text{eq}})] \leq 0$ increases monotonically with time and vanishes only for $\rho = \rho_{\text{eq}}$ [2]. Thus, an arbitrary initial state converges toward ρ_{eq}.

We now simulate numerically the dynamics for the pure initial rotor state $\langle\Omega|\psi_0\rangle \propto \exp[-|\mathbf{e}_z \times \mathbf{m}(\Omega)|^2/2\sigma^2]$,

representing a superposition of pointing upwards and downwards along the z axis with width σ. Its time evolution is shown in Fig. 2. The initial superposition first decoheres into a mixture of the upwards and downwards orientation of the rotor, as is evident from the purity. On the longer timescale $1/\Gamma$, the rotor approaches thermal equilibrium, as indicated by the energy expectation value and the von Neumann entropy. The final state, given by Eq. (13), is already well approximated by the Gibbs state, even though the thermal occupation number $\bar{\ell}$, defined via $\bar{\ell}(\bar{\ell} + 1) = \xi$, is as low as $\bar{\ell} \simeq 2.7$.

Planar rotors.—If the rotor is confined to the xy plane, a single angle α suffices to describe the orientation, $\mathbf{e}_r(\alpha) = \mathbf{e}_x \cos\alpha + \mathbf{e}_y \sin\alpha$. The corresponding angular momentum operator points into the z direction, $\mathsf{J} = \mathbf{e}_z \mathsf{p}_\alpha$, and has discrete eigenvalues $\hbar m$, $m \in \mathbb{Z}$. The Lindblad operator takes on the form

$$\mathbf{A} = \mathbf{e}_r(\hat{\alpha}) + \frac{i\hbar}{4k_B TI}\mathbf{e}_\varphi(\hat{\alpha})\mathsf{p}_\alpha, \qquad (14)$$

where $\mathbf{e}_\varphi(\alpha) = \mathbf{e}_z \times \mathbf{e}_r(\alpha)$.

The action of the dissipator (10a) can be conveniently expressed in terms of the Wigner function $w_m(\alpha)$ [56,57],

$$\partial_t^{\text{nc}} w_m(\alpha) = \frac{\Gamma}{2}[(m + 1)w_{m+1}(\alpha) - (m - 1)w_{m-1}(\alpha)] + D\frac{w_{m+1}(\alpha) - 2w_m(\alpha) + w_{m-1}(\alpha)}{\hbar^2}. \qquad (15)$$

As in Eq. (11) we dropped the term proportional to $1/T$ (which vanishes in the semiclassical limit). Expression (15) is the discretized version of the Fokker-Planck equation,
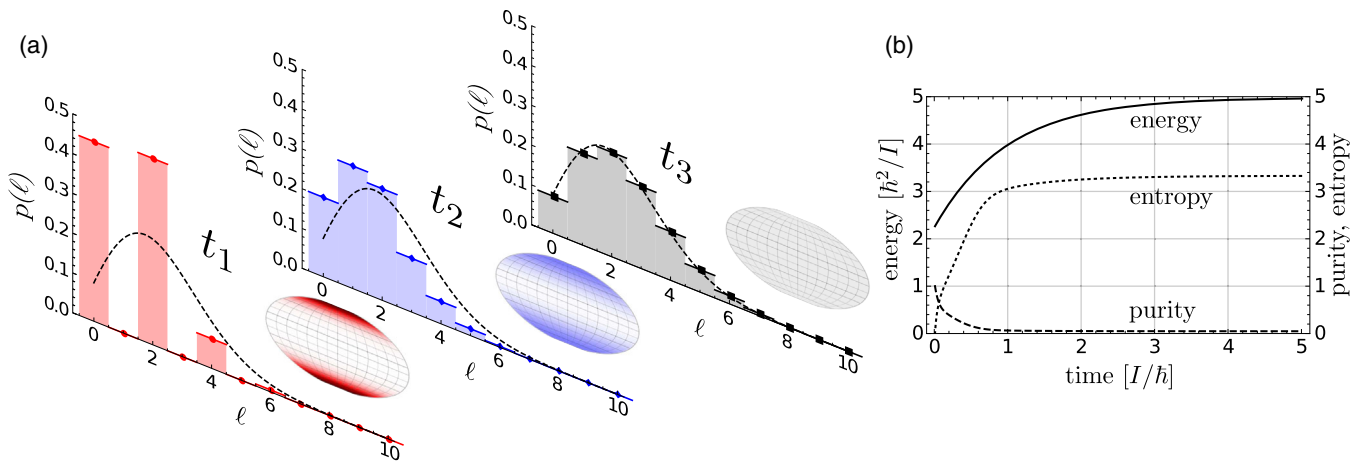

FIG. 2. Time evolution of the linear rigid rotor with free Hamiltonian $\mathsf{H} = \mathsf{J}^2/2I$ and dissipator (10) for an initial superposition of pointing upwards and downwards. (a) The histograms show the probability $p_\ell = \sum_{m=-\ell}^{\ell}\langle\ell m|\rho|\ell m\rangle$ of observing the total angular momentum ℓ for $t_1 = 0$, $t_2 = 0.5I/\hbar$, and $t_3 = 5I/\hbar$. The dashed line represents the Gibbs state, and the insets display polar density plots of the orientational distribution $\langle\Omega|\rho|\Omega\rangle$ (Mollweide projection). (b) Time dependence of the energy expectation value $\langle\mathsf{H}\rangle$ (solid line), the von Neumann entropy $-\text{tr}(\rho\log\rho)$ (dotted line), and the purity $\text{tr}(\rho^2)$ (dashed line). We use $\xi = 5$, $\Gamma = \hbar/I$, and $\sigma = 0.4$.
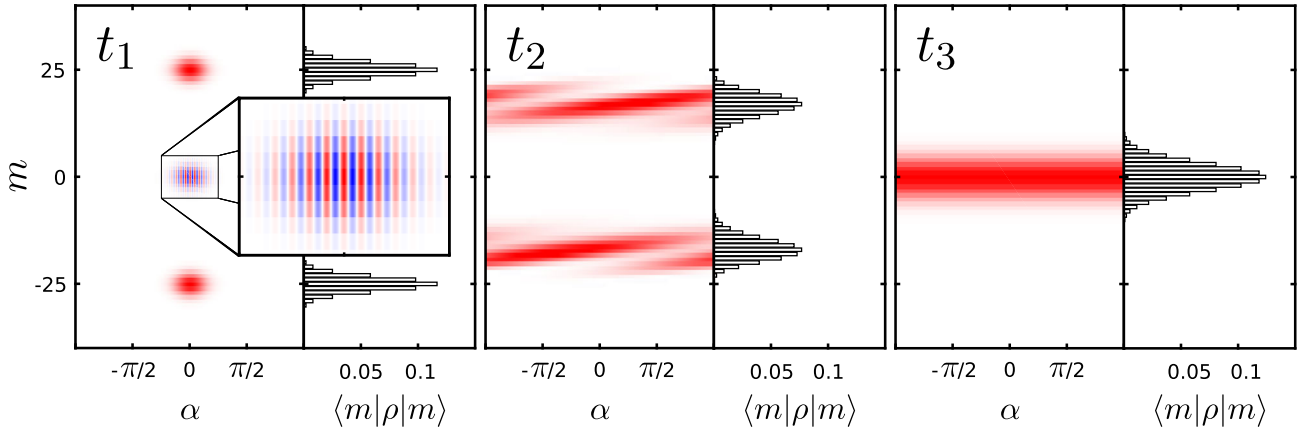
FIG. 3. Time evolution of the planar rotor. The left subpanels show the Wigner function $w_m(\alpha)$, and the right subpanels the momentum marginals $\langle m|\rho|m\rangle$ for $t_1 = 0$, $t_2 = 4\pi I/10\hbar$, and $t_3 = 4\pi I/\hbar$. The initial state is a superposition of two Gaussian states $\psi_0(\alpha) \propto \exp(im_0\alpha + \cos\alpha/4\sigma^2)$ with $m_0 = \pm 25$ and $\sigma = 0.2$. Its coherence is indicated by the fringe structure (with negative values in blue); see inset. The plots are obtained by computing the Wigner function [56,57] of the time evolved density operator. The parameters are chosen as $\xi = 20$ and $\Gamma = \hbar/\pi I$.

which is thus recovered in the semiclassical limit at fixed $p_\alpha = \hbar m$.

The stationary solution of the quantum planar rotor follows as $\rho_{\rm eq} = \sum_{m\in\mathbb{Z}} \rho_{\rm eq}^m |m\rangle\langle m|$ with [52]

$$\rho_{\rm eq}^m = \frac{1}{Z}\binom{2\xi}{|m|}^2 \binom{2\xi+|m|}{|m|}^{-2} \simeq \frac{1}{2^{2\xi}}\binom{2\xi}{\xi+m}. \qquad (16)$$

The first expression is the stationary solution of (10a) with the Lindblad operator (14); the second results if the term $\mathcal{O}(1/T)$ is dropped. Note that they approach the Gibbs state $\rho_{\rm eq}^m \sim \exp(-m^2/\xi)/Z$ as $\xi \to \infty$.

In Fig. 3 we show the phase space dynamics of an initial superposition of two Gaussian states centered at $\alpha = 0$ and $m = \pm 25$. It first decoheres into a mixture which then thermalizes with the environment. The final state is given by Eq. (16), which is well approximated by the Gibbs state.

Conclusion.—In summary, the master equation (8) established in this Letter applies to any rotating quantum object subject to linear friction and diffusion. The associated diffusion and friction tensors, which can be determined either by a dedicated measurement or by a microscopic calculation, serve to fully characterize the effect of the thermal environment. We found it instrumental to use a coordinate independent, tensorial formulation, rather than a specific parametrization of the rotation group and its generators. It reflects that the periodic and compact space of orientations cannot be linearized, precluding the use of standard quantum Brownian motion. Potential applications of rotational quantum Brownian motion range from ultra-cold chemistry with aligned molecules via torsional dynamics in molecular biophysics and optomechanics of levitated particles to quantum rotor heat engines.

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