

Desorption-induced decoherence of nanoparticle motion

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We derive the quantum master equation predicting how the translational and rotational dynamics of a nanoparticle is affected by the emission of surface adsorbates. This is motivated by recent experiments which prepared the motion of internally hot silica particles in the deep quantum regime. In the limit of a well localized nanoparticle the rotranslational dynamics can be characterized by diffusion rates in quantitative agreement with classical expectations. The theory is also suited to describe the decoherence effect of outgassing and sublimation.

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I. INTRODUCTION

Owing to their exceptional isolation from environmental noise, levitated nanomechanical systems are excellent platforms for metrology and for probing new physics [1]. Recent advances include ultraprecise torque sensing capabilities [2] as well as optical ground-state cooling of both the translational [3–8] and librational [9] degrees of freedom. This progress is enabled by increasing control over noise sources. The scattering of residual gas can be virtually eliminated by better vacua and laser phase noise introduced by optical trapping or manipulation may be reduced by better optical components and less sensitive control schemes [3,9,10]. The currently dominant noise source in the form of photon recoil heating is unlikely to be prohibitive for the creation of macroscopic superpositions, as hybrid traps [11–16] and accelerated expansion interferometric schemes [17–21] mature.

In view of the long coherence times targeted in superposition experiments [22–26], it is thus important to consider sources of noise that are potentially more difficult to eliminate, such as those associated with the internal makeup of the nanoparticle. While often approximated as rigid bodies, the nanoparticles are composed of a macroscopic number of constituents at a finite temperature [27,28] whose interaction with the environment and external forces can lead to additional decoherence of the center-of-mass and rotational motion. This includes the coupling to acoustic modes [29,30] and the emission of thermal radiation [21,31–34]. It is expedient to study how such noise affects both the center-of-mass and the rotational motion because these are in general coupled by the trapping potential. Moreover, the nonlinear quantum dynamics of rotations can offer inherent advantages [1,25,35,36].

In this article, we present the quantum master equation describing how the emission of constituent particles or surface contaminants decoheres the motional quantum state of the emitting bodies. Specifically, for the case of adsorbate desorption, we find the Markovian master equation to be entirely characterized by the spectral particle flux density per surface element. The corresponding Lindblad jump operators turn out to be a product of two contributions: the first is a linear and angular momentum recoil due to the emission event and the second amounts to a weak orientation measurement associated with anisotropic emission patterns. For particles well localized in both position and orientation the dynamics become diffusive in momentum space, with an additional thermophoresis-like Hamiltonian contribution. In this limit, our predictions agree with classical results.

We then provide the quantum description of more general outgassing processes, such as sublimation. It is characterized by complex emission amplitudes, which are defined in terms of the Green function associated with the dynamics of the escaping particle. We derive these master equations by casting the decay of a metastably bound state in the body-fixed frame in terms of the Møller operators of the corresponding scattering problem. To this end, the nanoparticle motion is assumed to be much slower than each emission process and the events to be independent.

II. DESORPTION-INDUCED DECOHERENCE

A. Quantum master equation

The position and orientation of the nanoparticle may be described by its center-of-mass vector $\mathbf{X}$ and its orientation tensor $\mathbf{R}$; the latter rotates body-fixed vectors from a reference orientation to the current one. The associated momenta are the vectors of linear momentum $\mathbf{P}$ and angular momentum $\mathbf{J}$. The corresponding quantum observables, denoted with a hat, fulfill the commutation relations $[\hat{\mathbf{X}}, \hat{\mathbf{m}} \cdot \hat{\mathbf{P}}] = i\hbar\mathbf{m}$ and $[\hat{\mathbf{R}}, \hat{\mathbf{m}} \cdot \hat{\mathbf{J}}] = i\hbar\mathbf{m} \times \hat{\mathbf{R}}$ for any vector $\mathbf{m}$.

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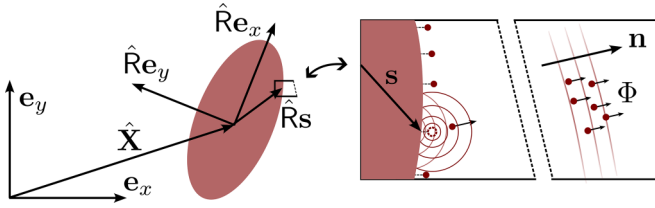


FIG. 1. Desorption of adsorbates from the surface of a nanoparticle, whose position $\hat{\mathbf{X}}$ and orientation $\hat{\mathbf{R}}$ are quantum degrees of freedom, leads to rotranslational decoherence. It is fully characterized by the adsorbate flux density $\Phi(\mathbf{n}, \mathbf{s}, E)$, a function of the emission direction $\mathbf{n}$, the (body-fixed) surface point $\mathbf{s}$, and the kinetic energy E .

The rotranslational state ρ of the nanoparticle evolves according to

$$\partial_t \rho = \frac{1}{i\hbar} \left[\frac{\hat{\mathbf{P}}^2}{2M} + \frac{1}{2} \hat{\mathbf{J}} \cdot \hat{\mathbf{R}}_0^{-1} \hat{\mathbf{R}}^T \hat{\mathbf{J}} + V_{\text{ext}}(\hat{\mathbf{X}}, \hat{\mathbf{R}}, t), \rho \right] + \mathcal{D}\rho, \quad (1)$$

where M is the nanoparticle mass and $\mathbf{l}_0$ is its tensor of inertia in the body-fixed frame, while the potential term accounts for (conservative) external forces and torques. In the following, we focus on the incoherent contribution to the Markovian dynamics, as described by the superoperator $\mathcal{D}$, resulting from the emission of nanoparticle constituents.

We first focus on the case of desorption, where scalar particles of mass $m \ll M$ are weakly bound to the nanoparticle surface ∂V at positions $\mathbf{s}$ relative to the center of mass and are released into the vacuum over time; see Fig. 1. From now on, these emitted particles will be referred to as *atoms*. For the nanoparticle in reference position and orientation this results in an atom flux density of $\Phi(\mathbf{n}, \mathbf{s}, E)$, which gives the rate of outgoing atoms per solid angle d^2n (i.e., into direction $\mathbf{n}$), per emitting surface element d^2s for an atom with energy E . For an arbitrary orientation the flux into direction $\mathbf{n}$ is then obtained by evaluating the reference flux at the inversely rotated direction $\hat{\mathbf{R}}^T \mathbf{n}$.

Assuming the nanoparticle dynamics to be much slower than the emission timescale (sudden approximation) we find that the incoherent part of the motional dynamics induced by the desorption of atoms is generated by a Lindbladian; see Sec. IV. It takes the form

$$\mathcal{D}\rho = \int_0^\infty dE \int_{\partial V} d^2s \int_{S^2} d^2n \\ \times [\hat{L}(\mathbf{n}, \mathbf{s}, E) \rho \hat{L}^\dagger(\mathbf{n}, \mathbf{s}, E) - |\hat{L}(\mathbf{n}, \mathbf{s}, E)|^2 \rho], \quad (2)$$

with jump operators

$$\hat{L}(\mathbf{n}, \mathbf{s}, E) = e^{-i\mathbf{n} \cdot (\hat{\mathbf{X}} + \hat{\mathbf{R}}\mathbf{s})p(E)/\hbar} \Phi^{1/2}(\hat{\mathbf{R}}^T \mathbf{n}, \mathbf{s}, E), \quad (3)$$

where $\mathbf{s} \in \partial V$ are the surface positions with respect to the center of mass, $p(E) = \sqrt{2mE}$, and S^2 is the unit sphere. Note that this equation is of Lindblad form because $\int_{S^2} d^2n |\hat{L}(\mathbf{n}, \mathbf{s}, E)|^2 = \int_{S^2} d^2n \Phi(\mathbf{n}, \mathbf{s}, E)$ is not operator-valued.

One can identify two conceptually distinct contributions to the jump operators. The unitary operator $e^{-in \cdot (\hat{\mathbf{X}} + \hat{\mathbf{R}}) p(E)/\hbar}$

imparts linear and angular momentum kicks compensating for the linear and orbital angular momentum of the outgoing atom. The square root of the flux, on the other hand, contributes to orientational decoherence because, for a fixed emission site, one could obtain information on the particle orientation from the solid angle distribution of the emission pattern. (Fixing a site is justified since the atoms are emitted independently.) Consequently, this factor may contribute to decoherence even if the total spectral flux $\int_{\partial V} d^2s \Phi(\mathbf{n}, \mathbf{s}, E)$ is isotropic.

B. Diffusive limit

For large particles one is interested mainly in the limit of small spatio-orientational delocalizations. One may then take the action of the jump operators on the orientational subspace to be dominated by the orbital angular momentum recoil.

As we shall see shortly, the nonunitary dynamics become diffusive in momentum space and may be characterized by a diffusion tensor. Following the classical treatment in [37], we denote this object as a matrix of tensors, where the diagonal blocks correspond to the linear and angular degrees of freedom, respectively, and the off-diagonal blocks describe correlated diffusion of the linear and angular momentum,

$$\begin{aligned} \underline{\underline{D_0}} = & \frac{1}{2} \int_0^\infty dE \int_{\partial V} d^2s \int_{S^2} d^2n \Phi(\mathbf{n}, \mathbf{s}, E) p^2(E) \\ & \times \begin{pmatrix} \mathbf{n} \otimes \mathbf{n} & \mathbf{n} \otimes (\mathbf{s} \times \mathbf{n}) \\ (\mathbf{s} \times \mathbf{n}) \otimes \mathbf{n} & (\mathbf{s} \times \mathbf{n}) \otimes (\mathbf{s} \times \mathbf{n}) \end{pmatrix}. \end{aligned} \quad (4)$$

In the following, multiplications of column vectors (of vectors) and matrices (of tensors) are to be understood as the corresponding operations in 6D.

If the nanoparticle state ρ is well oriented around an orientation $\hat{\mathbf{R}}$, and in absence of an external potential, $V_{\text{ext}} = 0$, the variance of the momentum increases linearly in time,

$$\frac{d}{dt} \text{Var} \begin{bmatrix} (\tilde{\mathbf{R}}^T \hat{\mathbf{P}}) \\ (\tilde{\mathbf{R}}^T \hat{\mathbf{J}}) \end{bmatrix} \approx 2 \mathbf{D}_0, \quad (5)$$

a defining characteristic of diffusion. The rotation tensors appear because $\underline{\underline{\mathbf{D}}}_0$ is defined for the reference orientation (body-fixed frame). (The variance of a vector is given by $\text{Var}[\mathbf{a}] = \langle \mathbf{a} \otimes \mathbf{a} \rangle - \langle \mathbf{a} \rangle \otimes \langle \mathbf{a} \rangle$.)

In addition, one gets Hamiltonian contributions corresponding to a thermophoresis-like force and torque, compare to the form of Eq. (38) in [37], which can be stated succinctly as

$$\underline{\mathbf{F}}_0 = - \int_0^\infty dE \int_{\partial V} d^2s \int_{S^2} d^2n \Phi(\mathbf{n}, \mathbf{s}, E) p(E) \begin{pmatrix} \mathbf{n} \\ \mathbf{s} \times \mathbf{n} \end{pmatrix}. \quad (6)$$

It leads to (again for $V_{\text{ext}} = 0$) a drift of the first moments of the momenta,

$$\frac{d}{dt} \left\langle \begin{pmatrix} \tilde{\mathbf{R}}^T \hat{\mathbf{P}} \\ \tilde{\mathbf{R}}^T \hat{\mathbf{J}} \end{pmatrix} \right\rangle \approx \underline{\mathbf{F}}_0. \quad (7)$$

To obtain the diffusive limit consider Eq. (2) for a state ρ well localized around $\hat{\mathbf{R}}$. The orientation tensor operator $\hat{\mathbf{R}}$ can then be approximated by means of the vector operator $\hat{\mathbf{w}} = \sum_{j=1}^3 \mathbf{e}_j \times \log(\hat{\mathbf{R}}^T \hat{\mathbf{R}}) \mathbf{e}_j / 2$, with orthonormal basis

vectors $(\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3)$. The length and direction of $\hat{\mathbf{w}}$ give the operator-valued rotation angle and axis of rotation relative to $\hat{\mathbf{R}}$. For small relative angles we have to lowest order $\hat{\mathbf{R}}\hat{\mathbf{w}} \approx \sum_{j=1}^3 (\hat{\mathbf{R}}\mathbf{e}_j) \times (\hat{\mathbf{R}}\mathbf{e}_j)/2$ and in turn $\hat{\mathbf{R}}\mathbf{e}_j \approx \hat{\mathbf{R}}[\mathbf{e}_j + \hat{\mathbf{w}} \times \mathbf{e}_j]$. This allows us to approximate the master equation (2) without introducing coordinates,

$$\begin{aligned} \mathcal{D}\rho \approx & \frac{1}{i\hbar} \left[- \left(\hat{\mathbf{R}}^T \hat{\mathbf{X}} \right) \cdot \underline{\mathbf{F}}_0, \rho \right] \\ & + \frac{2}{\hbar^2} \left[\left(\hat{\mathbf{R}}^T \hat{\mathbf{X}} \right) \cdot \underline{\mathbf{D}}_0 \rho \left(\hat{\mathbf{R}}^T \hat{\mathbf{X}} \right) \right. \\ & \left. - \frac{1}{2} \left\{ \left(\hat{\mathbf{R}}^T \hat{\mathbf{X}} \right) \cdot \underline{\mathbf{D}}_0 \left(\hat{\mathbf{R}}^T \hat{\mathbf{X}} \right), \rho \right\} \right]. \end{aligned} \quad (8)$$

From this one arrives at Eqs. (5) and (7) by keeping only the lowest order of the commutator $[\hat{\mathbf{R}}\mathbf{e}_j, \mathbf{e}_\ell \cdot \hat{\mathbf{J}}] \approx i\hbar \mathbf{e}_\ell \times (\hat{\mathbf{R}}\mathbf{e}_j)$, i.e., neglecting higher orders of $\hat{\mathbf{R}} - \hat{\mathbf{R}}$.

If the flux density is inversion symmetric, $\Phi(-\mathbf{s}, -\mathbf{n}, E) = \Phi(\mathbf{n}, \mathbf{s}, E)$, the linear and angular degrees of freedom decouple and the force vanishes. A vanishing torque however requires an additional symmetry.

For certain forms of $\Phi(\mathbf{n}, \mathbf{s}, E)$ and bodies of high symmetry, the integrals can be solved analytically. As an example, consider a flux density with cosine-shaped angular distribution, $\Phi(\mathbf{n}, \mathbf{s}, E) = \mathbf{n} \cdot \mathbf{n}_s \Theta(\mathbf{n} \cdot \mathbf{n}_s) \Phi_0(E)$, where $\mathbf{n}_s \equiv \mathbf{n}_s(\mathbf{s})$ is the local surface normal and Θ is the Heaviside step function. One can then directly carry out the solid angle integral. The force and torque vanish in this case, as may be seen by invoking the divergence theorem, while the diffusion tensor reads as

$$\begin{aligned} \underline{\mathbf{D}}_0 = & \frac{\pi}{8} \int_0^\infty dE \Phi_0(E) p^2(E) \int_{\partial V} d^2s \\ & \times \left[\begin{pmatrix} \mathbb{1} & -[\mathbf{s}]_\times \\ [\mathbf{s}]_\times & -[\mathbf{s}]_\times^2 \end{pmatrix} + \begin{pmatrix} \mathbf{n}_s \\ \mathbf{s} \times \mathbf{n}_s \end{pmatrix} \otimes \begin{pmatrix} \mathbf{n}_s \\ \mathbf{s} \times \mathbf{n}_s \end{pmatrix} \right]. \end{aligned} \quad (9)$$

Here, $[\mathbf{s}]_\times$ denotes the skew-symmetric tensor representation of the cross product with $\mathbf{s}$, i.e., $[\mathbf{s}]_\times \mathbf{a} = \mathbf{s} \times \mathbf{a}$ for any vector $\mathbf{a}$.

As a consistency check, we compare our results to those obtained by a classical model. Considering the flux obtained in the classical description of the desorptive part of diffusive gas scattering [37], we find that both the resulting diffusion tensor and the thermophoresis-like force and torque agree [38]. The classical model describes the reemission of previously adsorbed gas molecules after thermalization at the surface temperature.

III. GENERAL OUTGASSING MASTER EQUATION

We now consider general outgassing processes, where the source points of the ejected atoms are no longer confined to the particle surface. In this case, the potential imposed on the escaping atom by the delocalized nanoparticle imprints additional operator-valued phases which appear in the jump operators. The outgoing particle flux distribution then no longer suffices to determine the master equation, as the former does not contain phase information.

For general emission processes, the master equation is characterized by complex-valued amplitudes of emission. To

define the latter, we take the nanoparticle to be in reference position and orientation, and consider the retarded Green function $G^+(\mathbf{r}, \mathbf{s}, E)$ of the Schrödinger equation for the escaping atom. It describes the wave function at position $\mathbf{r}$ due to a source at position $\mathbf{s}$ at fixed energy E and yields the amplitude of emission in the limit

$$A^+(\mathbf{n}, \mathbf{s}, E) = \lim_{r \rightarrow \infty} -r e^{-ip(E)r/\hbar} G^+(r\mathbf{n}, \mathbf{s}, E). \quad (10)$$

This object may be understood as the analog to the scattering amplitude for an emission problem. The appearance of the radial limit $r \rightarrow \infty$ reflects the fact only the long-time asymptotics of the emitted particle state are relevant in a Markovian description.

To specify the general master equation, all that is needed in addition is the total emission rate due to a source at point $\mathbf{s}$, denoted by $\Gamma(\mathbf{s}, E)$. As shown in Sec. IV, the jump operators of the master equation then read

$$\begin{aligned} \hat{L}(\mathbf{n}, \mathbf{s}, E) = & \sqrt{\Gamma(\mathbf{s}, E)} \left[\int_{S^2} d^2n' |A^+(\mathbf{n}', \mathbf{s}, E)|^2 \right]^{-1/2} \\ & \times e^{-ip(E)\mathbf{n} \cdot \hat{\mathbf{X}}/\hbar} A^+(\hat{\mathbf{R}}^T \mathbf{n}, \mathbf{s}, E). \end{aligned} \quad (11)$$

The master equation takes the form of (2), with the surface integral $\int_{\partial V} d^2s$ replaced by the volume integral $\int_V d^3s$; see (45).

Furthermore, the jump operators give the outgoing particle flux density,

$$\Phi(\hat{\mathbf{R}}^T \mathbf{n}, \mathbf{s}, E) = |\hat{L}(\mathbf{n}, \mathbf{s}, E)|^2 \quad (12)$$

and they are normalized as $\int_{S^2} d^2n |\hat{L}(\mathbf{n}, \mathbf{s}, E)|^2 = \Gamma(\mathbf{s}, E)$. These expressions conform with physical expectations. Atoms are emitted independently from different sites at different energies and into varying directions. The information irreversibly lost to the environment is that contained in the asymptotic state of the atom.

IV. DERIVATION OF THE MASTER EQUATION

We proceed to derive the general Markovian outgassing master equation describing the impact of particle emission in terms of the Lindblad operators (11). This is done by combining the description of a metastably bound atom in the "weak-coupling limit" with the assumption of strongly separated motional and emission timescales ("sudden approximation") and utilizing the resolvent formalism for the escaping atom in the presence of the nanoparticle potential. This line of reasoning was already employed in the context of the emission of thermal radiation from dielectric particles [34]. Here we give a more detailed and self-contained account of the derivation.

A. Emission of metastably bound atoms

We fix the nanoparticle in reference position and orientation for the time being. To start with, consider an atom metastably bound in the state $|\varphi_0\rangle$ at energy E_0 . Ultimately, we are interested in the unbound part of the atomic motion, described by a wave function subject to the nanoparticle potential. To model the decay one may introduce a two-level system, which controls whether the atom feels a trapping potential, together with a coupling term inducing transitions

with a rate g ,

$$\begin{aligned}\tilde{H} = & E_0|\varphi_0\rangle\langle\varphi_0| \otimes |\uparrow\rangle\langle\uparrow| + H \otimes |\downarrow\rangle\langle\downarrow| \\ & + g\hbar|\varphi_0\rangle\langle\varphi_0| \otimes (|\uparrow\rangle\langle\downarrow| + |\downarrow\rangle\langle\uparrow|).\end{aligned}\quad (13)$$

Here, $H = p^2/2m + V$ is the Hamilton operator of the escaping atom (assumed to have no bound states). To solve the time evolution we decompose the state as

$$|\Psi(t)\rangle = e^{-iE_0t/\hbar}b(t)|\varphi_0\rangle|\uparrow\rangle + \int_0^\infty dE c_E(t)e^{-iEt/\hbar}|E\rangle|\downarrow\rangle, \quad (14)$$

taking the eigenstates $|E\rangle$ of H to be nondegenerate for the time being. Inserting (14) into the Schrödinger equation one finds the coefficients to obey

$$\dot{b}(t) = -ig \int_0^\infty dE e^{-i(E-E_0)t/\hbar} \langle\varphi_0|E\rangle c_E(t), \quad (15)$$

$$\dot{c}_E(t) = -ig e^{i(E-E_0)t/\hbar} \langle E|\varphi_0\rangle b(t). \quad (16)$$

We formally integrate the coefficient of the unbound part, insert it into the evolution of the bound part, and perform a Born-Markov-like approximation, by replacing $b(t') \rightarrow b(t)$ and extending the temporal integration to include the infinite past,

$$\begin{aligned}\dot{b}(t) = & -g^2 \int_0^\infty dE |\langle E|\varphi_0\rangle|^2 \int_0^t dt' e^{-i(E-E_0)(t-t')/\hbar} b(t') \\ \approx & -b(t)\hbar g^2 \int_0^\infty dE |\langle E_0|\varphi_0\rangle|^2 \\ & \times \left[\pi \delta(E-E_0) - i\mathcal{P} \frac{1}{E-E_0} \right].\end{aligned}\quad (17)$$

Neglecting the energy renormalization due to the principal value, the solution for the initial condition $b(0) = 1$ reads $b(t) = e^{-\Gamma_0 t/2}$, with the probability decay rate

$$\Gamma_0 = 2\pi\hbar g^2 |\langle E_0|\varphi_0\rangle|^2. \quad (18)$$

Reinsertion into the formal solution of the escaping part of the wave function $|\psi_\downarrow\rangle = \langle\downarrow|\Psi\rangle$ gives, for $\Gamma_0 t \ll 1$,

$$\begin{aligned}|\psi_\downarrow(t)\rangle = & e^{-iE_0t/\hbar}\hbar g \int_0^\infty dE (1 - e^{-i(E-E_0)t/\hbar}) \\ & \times \frac{|E\rangle\langle E|\varphi_0\rangle}{E_0 - i\hbar\Gamma_0/2 - E}.\end{aligned}\quad (19)$$

In the first term of the integrand one may identify the resolvent operator $G(z) = [z - H]^{-1}$ with $z = E_0 - i\hbar\Gamma_0/2$. To deal with the second term, we again consider the asymptotics of large t and take Γ_0 to be sufficiently small so that $\Gamma_0 t \ll 1$. In this case, the off-resonant contributions oscillate arbitrarily fast and average out, given that the remaining integrand is a smooth integrable function of E . This effectively allows one to replace the energy projector by $|E_0\rangle\langle E_0|$; the same argument allows extending the lower integration boundary to negative infinity. The remaining integral can then be solved using contour integration, $\int_{-\infty}^\infty dE e^{-iEt/\hbar}/(E + i\hbar\Gamma_0/2) = -e^{-\Gamma_0 t/2}2\pi i$, and the projector may also be expressed using resolvent operators since $-2\pi i|E\rangle\langle E| = G(E + i0^+) - G(E - i0^+)$. Since $\Gamma_0 t \ll 1$ this leaves us with the

approximate solution

$$|\psi_\downarrow(t)\rangle \sim \hbar g e^{-iE_0t/\hbar} G(E_0 + i\hbar\Gamma_0/2)|\varphi_0\rangle \quad (20)$$

as $t \rightarrow \infty$. It describes the arbitrarily slow leaking of a metastably bound state acting as a source term in the Schrödinger equation for the escaping part of the wave function,

$$(i\hbar\partial_t - H)|\psi_\downarrow\rangle = \hbar g \exp(-iE_0t/\hbar)|\varphi_0\rangle. \quad (21)$$

We note that the asymptotic solution (20) has unit norm since

$$\lim_{\Gamma_0 \rightarrow 0} \frac{\Gamma_0}{\pi} G(E - i\hbar\Gamma_0/2)G(E + i\hbar\Gamma_0/2) = |E\rangle\langle E|, \quad (22)$$

as follows from $(z' - z)G(z)G(z') = G(z) - G(z')$ (first resolvent identity [39]) and the Sokhotski-Plemelj formula.

B. Impact of emission events on the motional degrees of freedom

Let us now consider how the emission process acts back on the motional state of the emitting particle, assuming the latter to be static on the emission timescale. (In this approximation the kinetic energies of the atom in the body-fixed frame and the laboratory frame coincide.) The active transformation from the reference to the actual particle position and orientation is described by the unitary operator $\hat{D} \equiv D(\hat{\mathbf{X}}, \hat{\mathbf{R}})$, acting on the total nanoparticle-atom Hilbert space. Introducing the position-orientation basis as the common eigenvectors of $\hat{\mathbf{X}}$ and $\hat{\mathbf{R}}$, denoted $|\mathbf{X}, \mathbf{R}\rangle \equiv |\mathbf{X}\rangle|\mathbf{R}\rangle$, the transformation takes the form $\hat{D}|\mathbf{r}\rangle|\mathbf{X}, \mathbf{R}\rangle = |\mathbf{Rr} + \mathbf{X}\rangle|\mathbf{X}, \mathbf{R}\rangle$, with $|\mathbf{r}\rangle$ a position eigenstate of the atom.

The previous calculation is easily adapted to the transformed problem, i.e., with $V \rightarrow \hat{D}V\hat{D}^\dagger$ and $|\varphi_0\rangle \rightarrow \hat{D}|\varphi_0\rangle$, which shows that the state of the escaping atom is also transformed as $|\psi_\downarrow\rangle \rightarrow \hat{D}|\psi_\downarrow\rangle$. Expanding $\hat{D}$ in a position-orientation basis demonstrates that the escaping atom state is highly correlated with the nanoparticle state of motion. One may thus obtain the backaction on the reduced nanoparticle state conditioned on emission by tracing out the atomic state,

$$\rho' = \text{tr}_{\text{atom}}[\hat{D}|\psi_\downarrow\rangle\langle\psi_\downarrow| \otimes \rho\hat{D}^\dagger]. \quad (23)$$

To evaluate the trace we use the second resolvent identity [39], $G = G_0(1 + VG) = (1 + GV)G_0$, involving the free-space resolvent $G_0(z) = [z - H_0]^{-1}$, and identify the Møller operators $\Omega_\pm = \lim_{t \rightarrow \mp\infty} e^{iHt/\hbar} e^{-iH_0t/\hbar}$, where $H_0 = H - V$. The latter map asymptotically incoming and outgoing states to the corresponding scattering states and they are unitary since H has no bound states [40]. The time limit in the definition can be shown to be equivalent to the so-called adiabatic switching limit [40,41]

$$\Omega_\pm = \lim_{\epsilon \rightarrow 0^+} \pm \frac{\epsilon}{\hbar} \int_{\mp\infty}^0 dt e^{\pm\epsilon t/\hbar} e^{iHt/\hbar} e^{-iH_0t/\hbar}. \quad (24)$$

It will be useful to consider the action of the Møller operators on an eigenstate $|E\rangle_0$ of H_0 ,

$$\begin{aligned}\Omega_\pm|E\rangle_0 = & \lim_{\epsilon \rightarrow 0^+} \pm \frac{\epsilon}{\hbar} \int_{\mp\infty}^0 dt e^{\pm\epsilon t/\hbar} e^{iHt/\hbar} e^{-iEt/\hbar}|E\rangle_0 \\ = & \lim_{\epsilon \rightarrow 0^+} \pm i\epsilon G(E \pm i\epsilon)|E\rangle_0 \\ = & [1 + G(E \pm i0^+)V]|E\rangle_0,\end{aligned}\quad (25)$$

where the final line follows from the second resolvent identity.

Returning to Eq. (23), we perform the trace in momentum basis and use that $D^\dagger(\hat{\mathbf{X}}, \hat{\mathbf{R}})|\mathbf{p}\rangle = e^{i\mathbf{p}\cdot\hat{\mathbf{X}}/\hbar}|\hat{\mathbf{R}}^T\mathbf{p}\rangle$. Here, the operator-valued atomic momentum state is to be understood in terms of an expansion in the position-orientation basis of the nanoparticle. Together with the second resolvent identity we arrive at

$$\begin{aligned} \rho' &= \lim_{\Gamma_0 \rightarrow 0^+} \hbar^2 g^2 \int_{\mathbb{R}^3} d^3p e^{-i\mathbf{p}\cdot\hat{\mathbf{X}}/\hbar} \langle \hat{\mathbf{R}}^T\mathbf{p} | G_0(E_0 + i\hbar\Gamma_0/2) \\ &\quad \times [\mathbb{1} + VG(E_0 + i\hbar\Gamma_0/2)] |\varphi_0\rangle \langle \varphi_0| \otimes \rho \\ &\quad \times [\mathbb{1} + VG(E_0 + i\hbar\Gamma_0/2)]^\dagger G_0^\dagger(E_0 + i\hbar\Gamma_0/2) \\ &\quad \times |\hat{\mathbf{R}}^T\mathbf{p}\rangle e^{i\mathbf{p}\cdot\hat{\mathbf{X}}/\hbar}. \end{aligned} \quad (26)$$

Since G_0 is diagonal in momentum this yields a nascent delta function, which enforces the on-shell momentum $p_0 = \sqrt{2mE_0}$ and this allows one to identify the Møller operators for outgoing states based on (25),

$$\begin{aligned} \rho' &= \frac{1}{|\langle E_0|\varphi_0\rangle|^2} \lim_{\Gamma_0 \rightarrow 0^+} \int_{S^2} d^2n \int_0^\infty dp \frac{\hbar\Gamma_0/2\pi}{(E_p - E_0)^2 + \hbar^2\Gamma_0^2/4} \\ &\quad \times \langle p\mathbf{n} | \hat{D}[\mathbb{1} + VG(E_0 + i\hbar\Gamma_0/2)] |\varphi_0\rangle \langle \varphi_0| \otimes \rho \\ &\quad \times [\mathbb{1} + VG(E_0 + i\hbar\Gamma_0/2)]^\dagger \hat{D}^\dagger |p\mathbf{n}\rangle \\ &= \frac{mp_0}{|\langle E_0|\varphi_0\rangle|^2} \int_{S^2} d^2n \langle p_0\mathbf{n} | \hat{D}\Omega_-^\dagger |\varphi_0\rangle \rho \langle \varphi_0 | \Omega_- \hat{D}^\dagger |p_0\mathbf{n}\rangle. \end{aligned} \quad (27)$$

It is straightforward to check that the transformed state is still normalized; see the Appendix.

C. Dynamic equation for particle emission

To obtain the incoherent part $\mathcal{D}\rho$ of the nanoparticle equation of motion, we consider a time step dt small compared to both the free nanoparticle evolution and the lifetime of a metastably bound atom, but much greater than the timescale of the emission process of an escaping atom. Given the nanoparticle state $\rho(t)$, the time-evolved one is a mixture of the transformed state (27), weighted with the transmission probability $\Gamma_0 dt \ll 1$, and the unperturbed state, weighted with the probability for no emission having taken place, i.e., $\rho(t + dt) = \Gamma_0 dt \rho'(t) + (1 - \Gamma_0 dt)\rho(t)$. The differential quotient then takes the form

$$\begin{aligned} \mathcal{D}\rho &= \Gamma_0 \left[\frac{mp_0}{|\langle E_0|\varphi_0\rangle|^2} \int_{S^2} d^2n \langle p_0\mathbf{n} | \hat{D}\Omega_-^\dagger |\varphi_0\rangle \rho \langle \varphi_0 | \right. \\ &\quad \left. \times \Omega_- \hat{D}^\dagger |p_0\mathbf{n}\rangle - \rho \right]. \end{aligned} \quad (28)$$

It follows from (A1) that this equation is of Lindblad form.

For simplicity, we take the bound states $|\varphi_0\rangle$ to be well localized, such that we can approximate them in terms of position eigenstates $|\mathbf{s}\rangle$. Equation (28) can then be written as

$$\mathcal{D}\rho = \int_{S^2} d^2n \left[\hat{L}(\mathbf{n}) \rho \hat{L}^\dagger(\mathbf{n}) - \frac{1}{2} \{ \hat{L}^\dagger(\mathbf{n}) \hat{L}(\mathbf{n}), \rho \} \right], \quad (29)$$

with jump operators

$$\begin{aligned} \hat{L}(\mathbf{n}) &= \frac{\sqrt{\Gamma_0 mp_0}}{|\langle E_0|\mathbf{s}\rangle|} \langle p_0\mathbf{n} | \hat{D}\Omega_-^\dagger |\mathbf{s}\rangle \\ &= \sqrt{\Gamma_0} \frac{\langle p_0\mathbf{n} | \hat{D}\Omega_-^\dagger |\mathbf{s}\rangle}{\sqrt{\int_{S^2} d^2n' |\langle p_0\mathbf{n}' | \Omega_-^\dagger |\mathbf{s}\rangle|^2}}. \end{aligned} \quad (30)$$

The matrix elements in (30) can be related to a limit of the Green function $G^\pm(\mathbf{r}, \mathbf{r}'; E_0) = \langle \mathbf{r} | G(E_0 \pm i0^+) | \mathbf{r}' \rangle$ of the position-space Schrödinger equation for the atom. Specifically, they are given in terms of the amplitude of emission $A^\pm$, defined as

$$A^\pm(\mathbf{n}, \mathbf{s}, E_0) = - \lim_{r \rightarrow \infty} r e^{\mp i p_0 r / \hbar} G^\pm(r\mathbf{n}, \mathbf{s}, E_0). \quad (31)$$

(A^- may be understood as the corresponding amplitude of absorption.)

To show this connection, we first consider a Fourier-like solid-angle integral over a smooth function $g(\mathbf{n})$ for a real argument λ ,

$$\begin{aligned} I(\mathbf{n}; \lambda) &= \int_{S^2} d^2n' e^{\pm i\lambda \mathbf{n}' \cdot \mathbf{n}} g(\mathbf{n}') \\ &= 4\pi \int_{S^2} d^2n' \sum_{\ell, m} i^\ell j_\ell(\lambda) Y_{\ell m}^*(\mathbf{n}') Y_{\ell m}(\pm \mathbf{n}) g(\mathbf{n}'), \end{aligned} \quad (32)$$

where we expanded the exponential in terms of spherical harmonics. One may use the asymptotic form of the spherical Bessel functions as well as properties of the spherical harmonics (see 1.17.25 and 14.30.7 in Ref. [42]) to obtain that

$$\begin{aligned} I(\mathbf{n}; \lambda) &\sim -2\pi i \int_{S^2} d^2n' \sum_{\ell, m} i^\ell [e^{i\lambda} - (-1)^\ell e^{-i\lambda}] \\ &\quad \times Y_{\ell m}^*(\mathbf{n}') Y_{\ell m}(\pm \mathbf{n}) g(\mathbf{n}') \\ &= \mp i \frac{2\pi}{\lambda} [e^{\pm i\lambda} g(\mathbf{n}) - e^{\mp i\lambda} g(-\mathbf{n})] \end{aligned} \quad (33)$$

as $\lambda \rightarrow \infty$. With this we can now tackle the position-momentum matrix elements of the Møller operators by inserting a position basis into the adiabatic switching formulation (25)

$$\langle p_0\mathbf{n} | \Omega_\pm^\dagger | \mathbf{s} \rangle = \lim_{\epsilon \rightarrow 0^+} \mp i\epsilon \int_{\mathbb{R}^3} d^3r \langle p_0\mathbf{n} | \mathbf{r} \rangle \langle \mathbf{r} | G(E_0 \mp i\epsilon) | \mathbf{s} \rangle. \quad (34)$$

At this point, we incorporate the physical assumption that the emitted atom does not get trapped by the nanoparticle potential, by taking the Green function in (34) to be locally integrable. Since we consider the limit $\epsilon \rightarrow 0$, bounded domains do not contribute in the spatial integration, allowing us to introduce a lower cutoff radius R ,

$$\begin{aligned} \langle p_0\mathbf{n} | \Omega_\pm^\dagger | \mathbf{s} \rangle &= \lim_{\epsilon \rightarrow 0^+} \mp i\epsilon [2\pi \hbar]^{-3/2} \int_R^\infty dr r^2 \int_{S^2} d^2n' \\ &\quad \times e^{-ip_0 r \mathbf{n}' \cdot \mathbf{n} / \hbar} G(r\mathbf{n}', \mathbf{s}; E_0 \mp i\epsilon). \end{aligned} \quad (35)$$

The cutoff can be chosen arbitrarily large, so that we may use Eq. (32) as well as the asymptotic form of the Green function [as implied by Eq. (31)]. Because ϵ is still finite at this

point, the on-shell momentum takes the form $p(E_0 \pm i\epsilon) = \sqrt{2m(E_0 \pm i\epsilon)} \simeq p_0 \pm i\epsilon m/p_0$, so that

$$\begin{aligned} \langle p_0 \mathbf{n} | \Omega_{\pm}^{\dagger} | \mathbf{s} \rangle &= \lim_{\epsilon \rightarrow 0^+} \pm \epsilon \frac{2\pi \hbar}{p_0} [2\pi \hbar]^{-3/2} \int_R^{\infty} dr e^{-\epsilon m r / p_0 \hbar} \\ &\times [-e^{-ip_0 r(1 \pm 1)/\hbar} A^{\mp}(\mathbf{n}, \mathbf{s}, E_0) \\ &+ e^{ip_0 r(1 \mp 1)/\hbar} A^{\mp}(-\mathbf{n}, \mathbf{s}, E_0)]. \end{aligned} \quad (36)$$

The radial integration and the limit $\epsilon \rightarrow 0^+$ can now be computed,

$$\lim_{\epsilon \rightarrow 0^+} \epsilon \int_R^{\infty} dr e^{\pm i b r} e^{-a \epsilon r} = \begin{cases} 0, & b \neq 0, \\ a^{-1}, & b = 0. \end{cases} \quad (37)$$

It follows that the matrix elements of the Møller operators finally evaluate to

$$\langle p_0 \mathbf{n} | \Omega_{\pm}^{\dagger} | \mathbf{s} \rangle = 4\pi [2\pi \hbar]^{-3/2} \frac{\hbar^2}{2m} A^{\mp}(\mp \mathbf{n}, \mathbf{s}, E_0). \quad (38)$$

The jump operator (30) is thus given by

$$\hat{L}(\mathbf{n}) = \sqrt{\Gamma_0} e^{-ip_0 \mathbf{n} \cdot \hat{\mathbf{x}}/\hbar} \frac{A^+(\hat{\mathbf{R}}^T \mathbf{n}, \mathbf{s}, E_0)}{\sqrt{\int_{S^2} d^2 n' |A^+(\mathbf{n}', \mathbf{s}, E_0)|^2}}. \quad (39)$$

To illustrate its action, let us consider the idealized case of a transparent emitter, where the atom does not interact with the nanoparticle after release. The retarded Green function is then given by

$$G_0^+(\mathbf{r}, \mathbf{s}; E_0) = -\frac{2m}{\hbar^2} \frac{1}{4\pi} \frac{e^{ip_0 |\mathbf{r}-\mathbf{s}|/\hbar}}{|\mathbf{r}-\mathbf{s}|}, \quad (40)$$

so that it is straightforward to calculate the amplitude of emission (31),

$$A_0^+(\hat{\mathbf{R}}^T \mathbf{n}, \mathbf{s}, E_0) = \frac{m}{2\pi \hbar^2} e^{-ip_0 \mathbf{n} \cdot \hat{\mathbf{R}} \mathbf{s}/\hbar}, \quad (41)$$

by using $|\mathbf{r}\mathbf{n} - \mathbf{s}| = r - \mathbf{n} \cdot \mathbf{s} + O(r^{-1})$.

The appearance of the operator-valued complex phase factor can be understood in two complementary ways. Consider an emission into the laboratory-fixed direction $\mathbf{n}$. On the one hand, for two distinct particle orientations $\mathbf{R}$ and $\mathbf{R}'$ the associated paths traversed by the atom differ by $\mathbf{n} \cdot (\mathbf{R} - \mathbf{R}')\mathbf{s}$ and therefore the accumulated phase shifts may contain information on the particle orientation. On the other hand, the exponential in (41) effects an angular momentum kick, $e^{-ip_0 \mathbf{n} \cdot \hat{\mathbf{R}} \mathbf{s}/\hbar} \hat{\mathbf{J}} e^{ip_0 \mathbf{n} \cdot \hat{\mathbf{R}} \mathbf{s}/\hbar} = \hat{\mathbf{J}} - (\hat{\mathbf{R}} \mathbf{s}) \times p_0 \mathbf{n}$ [43]. The imparted angular momentum thus compensates the orbital angular momentum of the emitted atom with respect to the nanoparticle center of mass.

Finally, we note that Eq. (28) is closely related to the observable yielding the atom flux into the laboratory-frame direction $\mathbf{n}$. The latter is obtained from the correlated state of nanoparticle and atom by a partial trace over the atom Hilbert space,

$$\begin{aligned} \hat{\Phi}(\mathbf{n}) &= \lim_{r \rightarrow \infty} r^2 \mathbf{n} \cdot \text{tr}_{\text{atom}} [\hat{D}\rho \otimes |\psi_{\downarrow}\rangle \langle \psi_{\downarrow}| \hat{D}^{\dagger} \mathbf{j}(\mathbf{r}\mathbf{n})] \\ &= \lim_{r \rightarrow \infty} r^2 \mathbf{n} \cdot \frac{1}{m} \text{Re}[\langle \psi_{\downarrow} | \hat{D}^{\dagger} \delta(\mathbf{r}\mathbf{n} - \mathbf{x}) \mathbf{p} \hat{D} | \psi_{\downarrow} \rangle], \end{aligned} \quad (42)$$

where the first line involves the atomic probability current density operator $\mathbf{j}(\mathbf{r}) = \{\delta(\mathbf{x} - \mathbf{r}), \mathbf{p}\}/2m$. Using the general initial state (20), we insert a position basis and utilize that the retarded Green function in an unbounded domain fulfills the Sommerfeld radiation condition $\lim_{r \rightarrow \infty} r(\partial_r - ip_0/\hbar)G^+(\mathbf{r}, \mathbf{s}, E_0) = 0$ [44] to arrive at

$$\begin{aligned} \hat{\Phi}(\mathbf{n}) &= \lim_{r \rightarrow \infty} r^2 \frac{\hbar^2 g^2 p_0}{m} \langle \varphi_0 | G^+(E_0 + i\hbar\Gamma_0/2) \hat{D}^{\dagger} | \mathbf{r}\mathbf{n} \rangle \\ &\times \langle \mathbf{r}\mathbf{n} | \hat{D}G(E_0 + i\hbar\Gamma_0/2) | \varphi_0 \rangle. \end{aligned} \quad (43)$$

This limit can now be related to the momentum-position matrix elements of the Møller operators as follows by identifying the amplitudes of emission (31) and invoking (38). Taking note of the definition (18) of Γ_0 , the flux operator finally takes the form

$$\hat{\Phi}(\mathbf{n}) = \Gamma_0 \frac{|\langle p_0 \mathbf{n} | \hat{D} \Omega_{-}^{\dagger} | \varphi_0 \rangle|^2}{\int_{S^2} d^2 n' |\langle p_0 \mathbf{n}' | \Omega_{-}^{\dagger} | \varphi_0 \rangle|^2}. \quad (44)$$

Comparison with Eq. (30) shows that this expression is the modulus squared of the jump operators, as stated in Eq. (12). It demonstrates that the flux operator is related to the flux introduced in Sec. II by an operator-valued rotation, $\hat{\Phi}(\mathbf{n}) = \Phi(\hat{\mathbf{R}}^T \mathbf{n})$.

It is now straightforward to generalize the above treatment to the case of many independent emission sites. This amounts to taking a sum of dissipators of the form (28) for many atoms initially bound at positions $\mathbf{s}$ and energies E . In the continuum limit, the jump operators (30) then take the form (11) and the general master equation reads

$$\begin{aligned} \mathcal{D}\rho &= \int_0^{\infty} dE \int_V d^3 s \int_{S^2} d^2 n [\hat{L}(\mathbf{n}, \mathbf{s}, E) \rho \hat{L}^{\dagger}(\mathbf{n}, \mathbf{s}, E) \\ &- |\hat{L}(\mathbf{n}, \mathbf{s}, E)|^2 \rho]. \end{aligned} \quad (45)$$

In practice, the microscopic details of the emission process are typically unknown and only the atom flux density $\Phi(\mathbf{n}, \mathbf{s}, E)$ is available. Even in this case, the essential form of the jump operators can be specified by means of their polar decomposition. Equation (12) fixes their positive part, while it is clear from the above construction that the unitary must be of the form $e^{-ip\mathbf{n} \cdot \hat{\mathbf{x}}/\hbar} \hat{U}_{\text{rot}}$. In the absence of any further information, setting $\hat{U}_{\text{rot}} = e^{-ip(E)\mathbf{n} \cdot \hat{\mathbf{R}} \mathbf{s}/\hbar}$ as in (3) may be considered the natural choice in that it describes precisely the recoil to the emitted atoms orbital angular momentum, as seen above in Eq. (41).

D. Localization rate for desorption

The jump operators (3) and (11) are diagonal in the position-orientation basis and the linear motion is only subject to momentum kicks. This means that the spatio-orientational representation of the master equation takes the simple form $\langle \mathbf{X}, \mathbf{R} | \mathcal{D}\rho | \mathbf{X}', \mathbf{R}' \rangle = -F_{\mathbf{R}, \mathbf{R}'}(\mathbf{X} - \mathbf{X}') \langle \mathbf{X}, \mathbf{R} | \rho | \mathbf{X}', \mathbf{R}' \rangle$ specified by the complex rate $F_{\mathbf{R}, \mathbf{R}'}(\Delta \mathbf{X})$. Its real part is non-negative, describing an exponential decay of spatio-orientational coherences. It is referred to as the localization rate, since the loss of coherence implies that a delocalized state turns into a mixture of localized ones over time.

The localization rate can be simplified in the case of the desorption of atoms from the nanoparticle surface. By using $2ab^* - |a|^2 - |b|^2 = -|a - b|^2 + 2i \operatorname{Im}[ab^*]$, it reads

$$\begin{aligned} \operatorname{Re} F_{\mathbf{R}, \mathbf{R}'}(\Delta \mathbf{X}) &= \int_0^\infty dE \int_{\partial V} d^2 s \int_{S^2} d^2 n \frac{1}{2} \left[\Phi(\mathbf{R}^T \mathbf{n}, \mathbf{s}, E) \right. \\ &\quad + \Phi(\mathbf{R}'^T \mathbf{n}, \mathbf{s}, E) - 2\sqrt{\Phi(\mathbf{R}^T \mathbf{n}, \mathbf{s}, E)\Phi(\mathbf{R}'^T \mathbf{n}, \mathbf{s}, E)} \\ &\quad \left. \times \cos\left(\frac{p(E)}{\hbar} \mathbf{n} \cdot [\Delta \mathbf{X} + (\mathbf{R} - \mathbf{R}')\mathbf{s}]\right) \right]. \end{aligned} \quad (46)$$

In this expression one can again identify the two different contributions to the decoherence process discussed in Sec. II A. In the limit of negligible recoil, $p \rightarrow 0$, the integrand becomes $(\sqrt{\Phi_{\mathbf{R}}} - \sqrt{\Phi_{\mathbf{R}}})^2/2$, a measure of how discernible the two orientations are from the emitted flux density. On the other hand, even if the flux density were approximately independent of the emission direction, a nonzero recoil would lead to decoherence.

Finally, we note that the limit of an arbitrarily large recoil turns the decoherence rate into the total rate of atom emission because the cosine term becomes highly oscillatory. Moreover, the decoherence rate is bounded by twice the emission rate. Both of these statements also hold for the general outgassing master equation. To get a rough estimate for the strength of the effect, consider the specific outgassing rate $6.6 \times 10^{-9} \text{ Pa m}^3/\text{s m}^2$ [45] of bulk silica at room temperature 30 h after baking. For a spherical nanoparticle with a diameter of 150 nm this yields a total emission rate of 0.33 Hz. The rate for untreated bulk gold at room temperature is of the order of $8.5 \times 10^{-8} \text{ Torr l/cm}^2 \text{ s}$ [46]. Taking this at face value yields a total emission rate of about 2 kHz for a particle of the same size.

V. DISCUSSION AND CONCLUSION

In this article, we investigated how the stochastic emission of small constituents affects the motional quantum state of a nanoparticle. The emission characteristics were obtained by considering the escape of a scalar particle metastably bound to the emitting body. This allowed us to define amplitudes of emission and relate them to the Møller operators from scattering theory. To arrive at a general Markovian master equation (45), we assumed the nanoparticle motion to be slow on the emission timescale and the emission characteristics to be stationary on the scale of the decoherence dynamics.

As an important property of the master equation, we found that the squared modulus of the jump operators can be identified with the emission flux density operator; see Eq. (12). The latter can be readily obtained by promoting the emission rates from empirical data or phenomenological models to an operator. In principle, the jump operators (11) must be calculated from the Green function associated with the potential imposed by the nanoparticle on the escaping particle; see Eq. (10). Yet, their positive parts are fully determined by the emission flux density operator, see Eq. (12), and we have argued that their unitary parts can be approximated by the mechanically

expected linear and angular momentum recoil associated with an emission event.

For the case of the desorption of adsorbates from the particle surface, we presented a simplified master equation (8). It is valid in the case of small spatio-orientational delocalizations, as expected for large particles, describing diffusion and decoherence on equal footing. It is characterized by a matrix of momentum diffusion tensors and a thermophoresis-like force and torque, which agree with classical results.

The presented quantum master equations facilitate accurate estimation of a decoherence process which may become relevant in future quantum experiments in the field of levitated optomechanics. A notable feature of their derivation is the use of Møller operators to nonperturbatively describe the interaction of an emitted particle with the emitting body during the former's escape. This approach may find use beyond the problem discussed here, e.g., if the emitted particles exhibit internal structure or intrinsic angular momentum. Moreover, we expect the amplitudes of absorption defined in (31), see also (38), to permit describing decoherence due to absorptive processes, such as resublimation or absorption of heat radiation.

The presented treatment ceases to be valid for small emission energies, if the dwell time of the emitted particle becomes comparable to the motional timescale of the emitting body. In this case, one can no longer describe the particle degrees of freedom as entering parametrically through $\hat{D}$, but needs to calculate the Møller operators on the total nanoparticle-escaping particle Hilbert space. Moreover, we assumed the emission to take place into vacuum, an approximation bound to fail if the nanoparticle is very close to a surface, as might be the case in on-chip setups for levitation. How to include such structured environments into the presented approach is still an open problem.

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DATA AVAILABILITY

No data were created or analyzed in this study.

APPENDIX: NORMALIZATION OF THE TRANSFORMED STATE

We confirm that the transformed nanoparticle state ρ' obtained in Eq. (27) is normalized. From the intertwining relation $\Omega_{\pm} H_0 = H \Omega_{\pm}$ we have

$$\begin{aligned} |E_0\rangle\langle E_0| &= \delta(E_0 - H) = \int_{\mathbb{R}^3} d^3 p \, \Omega_- |\mathbf{p}\rangle\langle \mathbf{p}| \Omega_-^\dagger \delta\left(E_0 - \frac{p^2}{2m}\right) \\ &= m p_0 \int_{S^2} d^2 n \, \Omega_- |p_0 \mathbf{n}\rangle\langle p_0 \mathbf{n}| \Omega_-^\dagger. \end{aligned} \quad (A1)$$

Evaluating the trace in position-orientation basis then gives

$$\begin{aligned} \text{tr}[\rho'] &= \frac{mp_0}{|\langle E_0|\varphi_0\rangle|^2} \int_{\mathbb{R}^3} d^3X \int_{\text{SO}(3)} d\mu(\mathbf{R}) \langle \mathbf{X}, \mathbf{R} | \rho | \mathbf{X}, \mathbf{R} \rangle \\ &\times \int_{S^2} d^2n \langle \varphi_0 | \Omega_- | p_0 \mathbf{R} \mathbf{n} \rangle \langle p_0 \mathbf{R} \mathbf{n} | \Omega_-^\dagger | \varphi_0 \rangle \end{aligned}$$

$$= \frac{1}{|\langle E_0|\varphi_0\rangle|^2} \text{tr}[\rho] \langle \varphi_0 | E_0 \rangle \langle E_0 | \varphi_0 \rangle = 1. \quad (\text{A2})$$

The integration over rotation tensors covers all possible particle orientations such that $\int_{\text{SO}(3)} d\mu(\mathbf{R}) |\mathbf{R}\rangle \langle \mathbf{R}|$ is a resolution of identity on the orientational subspace.

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- [43] This can be seen by using $e^A B e^{-A} = \sum_{k=0} [A, B]_k / k!$, with $[A, B]_k$ the k -fold nested commutator, and noting that all matrix elements $\mathbf{e}_i \cdot \hat{\mathbf{R}} \mathbf{e}_j$ commute.
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Correction: Sign errors in Eq. (19) and in an inline equation in the text below Eq. (19) have been fixed.