



Magnetic rigid rotor in the quantum regime: Theoretical toolbox

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We describe the quantum dynamics of a magnetic rigid rotor in the mesoscopic scale where the Einstein-De Haas effect is predominant. In particular, we consider a single-domain magnetic nanoparticle with uniaxial anisotropy in a magnetic trap. Starting from the basic Hamiltonian of the system under the macrospin approximation, we derive a bosonized Hamiltonian describing the center-of-mass motion, the total angular momentum, and the macrospin degrees of freedom of the particle treated as a rigid body. This bosonized Hamiltonian can be approximated by a simple quadratic Hamiltonian that captures the rich physics of a nanomagnet tightly confined in position, nearly not spinning, and with its macrospin antialigned to the magnetic field. The theoretical tools derived and used here can be applied to other quantum mechanical rigid rotors.

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I. INTRODUCTION

A rigid body is described by its center-of-mass position and linear momentum as well as its orientation and rotational angular momentum [1,2]. A rigid body can also contain internal degrees of freedom. Particularly interesting is a magnetic rigid body such that the internal spin can couple to the external degrees of freedom via the Einstein-De Haas effect [3]. The physics of such a magnetic rigid rotor is very rich specially at mesoscopic scales where the Einstein-De Haas effect is enhanced due to a smaller moment of inertia. Quantum effects have to be considered [4,5] when thermal fluctuations of the degrees of freedom are small. The quantum mechanical theory of the rigid rotor has been applied to study rotational spectra of molecules [6,7], to model the structure of the atomic nucleus [8,9], and to control the rotational motion of molecules [10–13]. Magnetic rigid rotors in the quantum regime have been considered to study spin tunneling in single-molecule magnets and magnetic single-domain nanoparticles [14–20].

Motivated by the possibility to bring mesoscopic systems to the quantum regime, see [21] and references therein, here we use well-known results in quantum angular momentum theory [22,23] to develop a theoretical toolbox to describe the quantum dynamics of a levitated magnetic rigid rotor. We consider a single-domain magnetic nanoparticle (we call it nanomagnet hereafter) in a magnetic trap to derive, from first principles, a relatively simple quadratic Hamiltonian able to describe the rich dynamics of the system. A quantum mechanical description of the system has to be used when the thermal fluctuations of the degrees of freedom have been cooled to the limit where quantum fluctuations dominate. In the context of quantum nanomechanics many techniques have been devised to reach this regime [21]. An experimental proposal to bring a levitated nanomagnet in the quantum regime will be analyzed elsewhere [24].

The article is organized as follows. In Sec. II, we introduce the system, its degrees of freedom, the basic Hamiltonian in the laboratory and body frame, and the Hilbert space with the relevant basis. In Sec. III, we perform a unitary transformation that diagonalizes the magnetic dipole interaction term. In Sec. IV, we perform the Lamb-Dicke approximation by

assuming the nanomagnet to be tightly confined in the magnetic trap. In Sec. V, we show how to map the angular momentum operators of the Hamiltonian into bosonic creation and annihilation operators. This allows us to perform a Holstein-Primakoff approximation by assuming the nanomagnet to be well antialigned to the external magnetic field and nearly not rotating. In Sec. VI, we apply these approximations to derive a quadratic Hamiltonian and discuss its validity. In Sec. VII, we draw our conclusions and provide some further directions. We leave to the appendices the discussion of some required tools of quantum angular momentum theory (Appendix A), an example of magnetic field trap (Appendix B), and some details about the derivation of the quadratic Hamiltonian (Appendices C and D).

II. DESCRIPTION OF THE SYSTEM

We consider a nanomagnet of mass M , moment of inertia I , and magnetic moment $\hat{\boldsymbol{\mu}}$. The nanomagnet is interacting in free space with an external static B field $\mathbf{B}(\mathbf{r})$. Being a rigid body, the position of the nanomagnet in space is specified by its center of mass position and its orientation. The latter is equivalent to specifying the orientation of the coordinate frame $O\mathbf{e}_1\mathbf{e}_2\mathbf{e}_3$, attached to the body and centered at the position of its center of mass, with respect to the position of the coordinate frame $O\mathbf{e}_x\mathbf{e}_y\mathbf{e}_z$ fixed in the laboratory. The mutual orientation between the two frames is described through the Euler angles $\Omega = \{\alpha, \beta, \gamma\}$ defined in Fig. 1(a). Hereafter, latin indexes $i, j, k, \dots = 1, 2, 3$ label the body frame axis while Greek indexes $\mu, \nu, \lambda, \dots = x, y, z$ label the laboratory frame axes.

The nanomagnet is characterized by the following degrees of freedom, see Fig. 1(b). (1) The center of mass described by its position $\hat{\mathbf{r}}$ and momentum $\hat{\mathbf{p}}$. (2) The rotational momentum and orientation of the particle described by the angular momentum $\hbar\hat{\mathbf{L}}$ and the Euler angles $\hat{\Omega} = \{\hat{\alpha}, \hat{\beta}, \hat{\gamma}\}$. (3) The magnetic moment $\hat{\boldsymbol{\mu}}$.

The operator $\hbar\hat{\mathbf{L}}$ is the angular momentum associated to the rotational motion of the particle. The magnetic moment of the nanomagnet $\hat{\boldsymbol{\mu}} = \hbar\gamma\hat{\mathbf{F}}$, with $\hat{\mathbf{F}} = \sum_{i=1}^N \hat{\mathbf{F}}_i$, is the sum of the single magnetic moments of its N constituents, where $\gamma > 0$ is the gyromagnetic ratio and $\hat{\mathbf{F}}_i$ the total spin of the

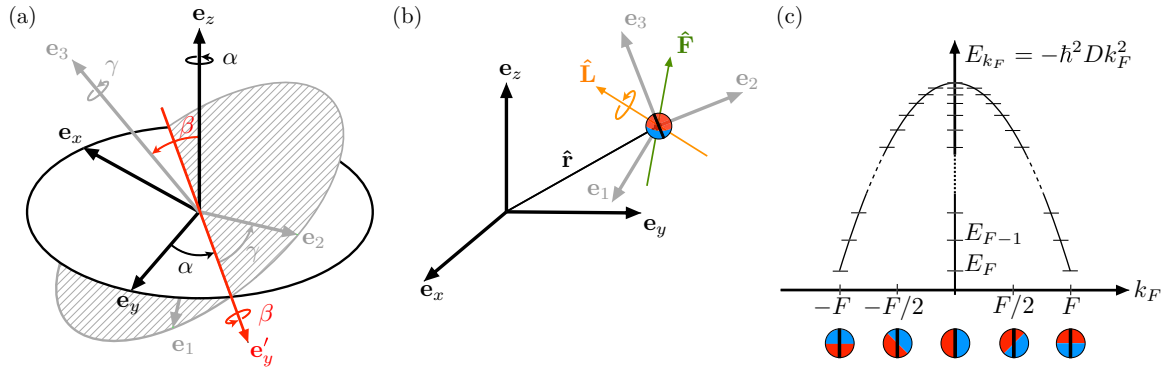


FIG. 1. (a) Definition of the Euler angles in the ZYZ convention. The Euler angles are defined by three successive rotations that one needs to apply in order to align $O\mathbf{e}_x\mathbf{e}_y\mathbf{e}_z$ with $O\mathbf{e}_1\mathbf{e}_2\mathbf{e}_3$. (i) Rotation of the frame $O\mathbf{e}_x\mathbf{e}_y\mathbf{e}_z$ of an angle $\alpha \in [0, 2\pi]$ about the axis \mathbf{e}_z into the new frame $O\mathbf{e}'_x\mathbf{e}'_y\mathbf{e}'_z$. (ii) Rotation of the frame $O\mathbf{e}'_x\mathbf{e}'_y\mathbf{e}'_z$ of an angle $\beta \in [0, \pi]$ about the axis \mathbf{e}'_y into the new frame $O\mathbf{e}''_x\mathbf{e}''_y\mathbf{e}''_z$. (iii) Rotation of the new frame $O\mathbf{e}''_x\mathbf{e}''_y\mathbf{e}''_z$ of an angle $\gamma \in [0, 2\pi]$ about the axis \mathbf{e}''_z into the final frame $O\mathbf{e}_1\mathbf{e}_2\mathbf{e}_3$. (b) Description of the system and its degrees of freedom. The nanomagnet is represented as a sphere divided in two parts: a red and a blue half that stays, respectively, for the north and south poles of the magnet, and thus give the direction of the spin $\hat{\mathbf{F}}$. The anisotropy axis is along the direction of \mathbf{e}_3 and it is represented by a black line in the nanomagnet, which need not to be aligned with the magnetization. Finally, the angular momentum $\hat{\mathbf{L}}$ relative to the mechanical rotational motion lies, in general, along a different direction. (c) Anisotropy of the nanomagnet. We consider a nanomagnet with a single axis anisotropy: the magnetic moment will preferably align parallel or antiparallel to the anisotropy direction. The \hat{F}_3^2 dependence produce a non linear level structure.

i th constituent. The individual spins inside the nanomagnet are subjected to two different interactions: (i) the exchange interaction, which tends to align all the single magnetic moments together, and (ii) the anisotropy interaction, which tends to align independently each magnetic moment to a given direction in the crystalline structure of the particle. When the first coupling dominates over the second coupling (weak anisotropy limit), one can perform the so-called macrospin approximation [26], which consists in projecting the total spin into the subspace with $\hat{\mathbf{F}}^2 = Nf(Nf + 1) \equiv F(F + 1)$, where f is the total spin of a single constituent (assumed to be identical for simplicity). Finally, note that the center-of-mass motion, described by $\hat{\mathbf{r}}$ and $\hat{\mathbf{p}}$, can yield angular momentum $\hat{\mathbf{r}} \times \hat{\mathbf{p}}$. This should not be confused with $\hbar\hat{\mathbf{L}}$.

For a solid nanosphere, the internal vibrations have frequencies of the order of the speed of sound divided by the size of the object. For a nanosphere, these frequencies are many orders of magnitude higher than any other frequency in the system. Hence internal vibrations are effectively decoupled [25] and can be safely ignored.

A. Hamiltonian in the laboratory frame

The dynamics of a rigid body can be studied both in the laboratory frame $O\mathbf{e}_x\mathbf{e}_y\mathbf{e}_z$ and in the body frame $O\mathbf{e}_1\mathbf{e}_2\mathbf{e}_3$. In the laboratory frame, the dynamics of a spherical nanomagnet of radius R (moment of inertia $I = 2MR^2/5$) is described by the Hamiltonian

$$\hat{H} = \frac{\hat{\mathbf{p}}^2}{2M} + \frac{\hbar^2}{2I} \hat{\mathbf{L}}^2 - \hbar^2 D [\hat{\mathbf{F}} \cdot \mathbf{e}_3(\hat{\Omega})]^2 - \hbar\gamma \hat{\mathbf{F}} \cdot \mathbf{B}(\hat{\mathbf{r}}). \quad (1)$$

Note that when a nonspherical shape is considered, the moment of inertia will be a tensor depending on the Euler angles [1,2]. The first (second) term represents the kinetic energy of the center of mass (rotational) motion. The third term represents the uniaxial anisotropy energy term in the macrospin

approximation with the preferred axis \mathbf{e}_3 , see Fig. 1(c). The uniaxial anisotropy is common in nanomagnets [14] but other anisotropies could be straightforwardly included in the analysis. In the laboratory frame, the unit vector \mathbf{e}_3 depends on the Euler angles and it is therefore an operator, $\mathbf{e}_3 = \mathbf{e}_3(\hat{\Omega})$. The anisotropy constant D depends on the material and is related to the blocking temperature of the nanomagnet $T_b \equiv \hbar^2 D F^2 / K_b$ [27], where K_b is the Boltzmann's constant. The fourth term represents the magnetic dipole interaction of the macrospin with the external static field $\mathbf{B}(\mathbf{r})$, which we describe as a classical field.

For later convenience we define $\hat{\mathbf{J}} \equiv \hat{\mathbf{L}} + \hat{\mathbf{F}}$, which is the total angular momentum of the system when $\hat{\mathbf{r}} \times \hat{\mathbf{p}} = 0$. The operators $\hat{\mathbf{J}}$, $\hat{\mathbf{L}}$, and $\hat{\mathbf{F}}$ fulfill the usual commutation relations of angular momenta, see Table I. Moreover, we assume $[\hat{L}_\nu, \hat{F}_\mu] = 0, \forall \mu, \nu$, namely that the spin angular momentum is independent of the Euler angles. This is commonly assumed in molecular quantum mechanics [6,28], and corresponds to neglecting the spin-orbit interaction between the individual spins of the nanomagnet and the electronic rotational motion within the Born-Oppenheimer approximation.

B. Hamiltonian in the body frame

The dynamics of a rigid body is more conveniently described in the body frame $O\mathbf{e}_1\mathbf{e}_2\mathbf{e}_3$ where the inertia tensor of a nonspherical body does not depend on the Euler angles [1,2]. The operators in the body frame are obtained by the following change of variables:

$$\begin{aligned} \hat{J}_i &= \sum_\nu R_{i\nu}(\hat{\Omega}) \hat{J}_\nu, & \hat{L}_i &= \sum_\nu R_{i\nu}(\hat{\Omega}) \hat{L}_\nu, \\ \hat{F}_i &= \sum_\nu R_{i\nu}(\hat{\Omega}) \hat{F}_\nu, & B_i(\mathbf{r}, \hat{\Omega}) &= \sum_\nu R_{i\nu}(\hat{\Omega}) B_\nu(\mathbf{r}), \end{aligned} \quad (2)$$

TABLE I. Commutation rules of the components of the angular momentum operators $\hat{\mathbf{L}}$, $\hat{\mathbf{S}} = -\hat{\mathbf{F}}$ and $\hat{\mathbf{J}} = \hat{\mathbf{F}} + \hat{\mathbf{L}} = -\hat{\mathbf{S}} + \hat{\mathbf{L}}$ both in the laboratory frame (left column) and in the body fixed frame (right column). The ladder operators in the body frame are $\hat{J}_{\uparrow(\downarrow)} = \hat{J}_1 \mp i\hat{J}_2$ ($\hat{S}_{\uparrow(\downarrow)} = \hat{S}_1 \mp i\hat{S}_2$), while in the laboratory are $\hat{J}_{\pm} = \hat{J}_x \pm i\hat{J}_y$ ($\hat{F}_{\pm} = \hat{F}_x \pm i\hat{F}_y$). Summation over repeated indexes is here assumed.

Laboratory frame $O\mathbf{e}_x\mathbf{e}_y\mathbf{e}_z$	Body frame $O\mathbf{e}_1\mathbf{e}_2\mathbf{e}_3$
$[\hat{L}_\mu, \hat{L}_\nu] = i\epsilon_{\mu\nu\lambda} \hat{L}_\lambda$	$[\hat{L}_i, \hat{L}_j] = -i\epsilon_{ijk} \hat{L}_k$
$[\hat{L}_z, \hat{L}_\pm] = \pm \hat{L}_\pm$	$[\hat{L}_3, \hat{L}_{\uparrow(\downarrow)}] = (-)\hat{L}_{\uparrow(\downarrow)}$
$[\hat{L}_+, \hat{L}_-] = 2\hat{J}_z$	$[\hat{L}_\uparrow, \hat{L}_\downarrow] = 2\hat{L}_3$
$[\hat{F}_\mu, \hat{F}_\nu] = i\epsilon_{\mu\nu\lambda} \hat{F}_\lambda$	$[\hat{S}_i, \hat{S}_j] = -i\epsilon_{ijk} \hat{S}_k$
$[\hat{F}_z, \hat{F}_\pm] = \pm \hat{F}_\pm$	$[\hat{S}_3, \hat{S}_{\uparrow(\downarrow)}] = (-)\hat{S}_{\uparrow(\downarrow)}$
$[\hat{F}^+, \hat{F}^-] = 2\hat{F}_z$	$[\hat{S}_\uparrow, \hat{S}_\downarrow] = 2\hat{S}_3$
$[\hat{J}_\mu, \hat{J}_\nu] = i\epsilon_{\mu\nu\lambda} \hat{J}_\lambda$	$[\hat{J}_i, \hat{J}_j] = -i\epsilon_{ijk} \hat{J}_k$
$[\hat{J}_z, \hat{J}_\pm] = \pm \hat{J}_\pm$	$[\hat{J}_3, \hat{J}_{\uparrow(\downarrow)}] = (-)\hat{J}_{\uparrow(\downarrow)}$
$[\hat{J}_+, \hat{J}_-] = 2\hat{J}_z$	$[\hat{J}_\uparrow, \hat{J}_\downarrow] = 2\hat{J}_3$
$[\hat{L}_\mu, \hat{F}_\nu] = 0$	$[\hat{L}_i, \hat{S}_j] = -i\epsilon_{ijk} \hat{S}_k$
$[\hat{J}_\mu, \hat{F}_\nu] = i\epsilon_{\mu\nu\lambda} \hat{F}_\lambda$	$[\hat{J}_i, \hat{S}_j] = 0$
$[\hat{J}_\mu, \hat{L}_\nu] = i\epsilon_{\mu\nu\lambda} \hat{L}_\lambda$	$[\hat{J}_i, \hat{L}_j] = -i\epsilon_{ijk} \hat{J}_k$
$[\hat{L}_\mu, R_{i\nu}(\hat{\Omega})] = i\epsilon_{\mu\nu\lambda} R_{i\lambda}(\hat{\Omega}) \quad \forall i$	$[\hat{L}_i, R_{j\mu}(\hat{\Omega})] = -i\epsilon_{ijk} R_{k\mu}(\hat{\Omega}) \quad \forall \mu$
$[\hat{J}_\mu, R_{i\nu}(\hat{\Omega})] = i\epsilon_{\mu\nu\lambda} R_{i\lambda}(\hat{\Omega}) \quad \forall i$	$[\hat{J}_i, R_{j\mu}(\hat{\Omega})] = -i\epsilon_{ijk} R_{k\mu}(\hat{\Omega}) \quad \forall \mu$
$[\hat{F}_\mu, R_{i\nu}(\hat{\Omega})] = 0$	$[\hat{S}_i, R_{j\mu}(\hat{\Omega})] = 0$
$[R_{i\mu}(\hat{\Omega}), R_{j\nu}(\hat{\Omega})] = 0 \quad \forall i, j, \mu, \nu$	$[R_{i\mu}(\hat{\Omega}), R_{j\nu}(\hat{\Omega})] = 0 \quad \forall i, j, \mu, \nu$

where the orthogonal matrix $R(\hat{\Omega})$ is given by

$$R(\hat{\Omega}) = \begin{pmatrix} \cos \hat{\gamma} & \sin \hat{\gamma} & 0 \\ -\sin \hat{\gamma} & \cos \hat{\gamma} & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \cos \hat{\beta} & 0 & -\sin \hat{\beta} \\ 0 & 1 & 0 \\ \sin \hat{\beta} & 0 & \cos \hat{\beta} \end{pmatrix} \times \begin{pmatrix} \cos \hat{\alpha} & \sin \hat{\alpha} & 0 \\ -\sin \hat{\alpha} & \cos \hat{\alpha} & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (3)$$

The change of variables (2) does not preserve the commutation relations of the angular momenta. Indeed, by writing the operator $\hat{\mathbf{L}}$ in the laboratory frame in the Euler angles representation [23]

$$\begin{pmatrix} \hat{L}_x \\ \hat{L}_y \\ \hat{L}_z \end{pmatrix} = \frac{-i}{\sin \beta} \begin{pmatrix} \cos \alpha \cos \beta & \sin \beta \sin \alpha & \cos \alpha \\ \sin \beta \sin \alpha & \sin \beta \cot \beta & \sin \alpha \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \partial_\alpha \\ \partial_\beta \\ \partial_\gamma \end{pmatrix}, \quad (4)$$

one can show that using Eq. (3)

$$[\hat{L}_\mu, R_{i\nu}(\hat{\Omega})] = i \sum_\lambda \epsilon_{\mu\nu\lambda} R_{i\lambda}(\hat{\Omega}). \quad (5)$$

Note, however, that this is not the case for the spin angular momentum since, as already discussed in Sec. II A, it commutes with the Euler angles

$$[\hat{F}_\mu, R_{i\nu}(\hat{\Omega})] = 0. \quad (6)$$

The results in Eqs. (5) and in (6) together with the property

$$\sum_{\mu, \nu} \epsilon_{\mu\nu\lambda} R_{i\mu}(\hat{\Omega}) R_{j\nu}(\hat{\Omega}) = \sum_k \epsilon_{ijk} R_{k\lambda}(\hat{\Omega}), \quad (7)$$

valid for an orthogonal matrix, allow to derive the commutation relations of the operators in the body frame, as given in Table I. Note that the body frame components of the spin commute as in the laboratory frame, while the commutators for the body frame components of $\hat{\mathbf{L}}$ and $\hat{\mathbf{J}}$ acquire a minus sign, namely $[\hat{L}_i, \hat{L}_j] = -i \sum_k \epsilon_{ijk} \hat{L}_k$ and $[\hat{J}_i, \hat{J}_j] = -i \sum_k \epsilon_{ijk} \hat{J}_k$. It is convenient to introduce the operator $\hat{\mathbf{S}} = -\hat{\mathbf{F}}$ to force the three angular momentum to have the same commutation rules. Note that $\hat{\mathbf{S}}^2 = \hat{\mathbf{F}}^2$ and we define $S \equiv F$. The body frame ladder operators are given by $\hat{J}_\uparrow = (\hat{J}_\downarrow)^\dagger = \hat{J}_1 - i\hat{J}_2$ and $\hat{S}_\uparrow = (\hat{S}_\downarrow)^\dagger = \hat{S}_1 - i\hat{S}_2$, where $\hat{J}_\uparrow, \hat{S}_\uparrow$ ($\hat{J}_\downarrow, \hat{S}_\downarrow$) increase (lower) the value of \hat{J}_3, \hat{S}_3 , see Table I.

With the change of variables given in Eq. (2), the Hamiltonian \hat{H} in the body frame reads

$$\hat{H} = \frac{\hat{\mathbf{P}}^2}{2M} + \frac{\hbar^2}{2I} (\hat{\mathbf{J}}^2 + 2\hat{S}_3 \hat{J}_3 + \hat{J}_\uparrow \hat{S}_\downarrow + \hat{J}_\downarrow \hat{S}_\uparrow) - \hbar^2 D \hat{S}_3^2 + \hbar \gamma \hat{\mathbf{S}} \cdot \mathbf{B}(\hat{\mathbf{r}}, \hat{\Omega}). \quad (8)$$

The term with $\hat{\mathbf{S}}^2$ has been dropped since under the macroscopic approximation $\hat{\mathbf{S}}^2 = S(S+1)$ is a c number. The external magnetic field depends in this frame on the Euler angles. So far, only a change of variables has been performed without any approximation.

C. Hilbert space structure of the system and suitable basis

The Hilbert space \mathcal{H} associated to the system is given by $\mathcal{H} = \mathbb{L}^2(\mathbb{R}^3) \otimes \mathcal{H}_{am}$. $\mathbb{L}^2(\mathbb{R}^3)$ is the Hilbert space associated to the motion of a pointlike particle in \mathbb{R}^3 , and it is spanned by the general basis $\{\psi_n(\mathbf{r})\}_n$. \mathcal{H}_{am} is the Hilbert space associated to the angular momenta of the particle. From the commutation relations in Table I, it is possible to find three different complete

sets of commuting observables (CSCO) for the particle angular momenta [7]. (1) Laboratory frame uncoupled representation $\{\hat{\mathbf{F}}^2, \hat{F}_z, \hat{\mathbf{L}}^2, \hat{L}_z, \hat{L}_3\}$ with eigenstates

$$|\varphi_{\text{am}}\rangle_1 = |Fm_F, Lm_L k_L\rangle_1, \quad (9)$$

such that

$$\begin{pmatrix} \hat{\mathbf{F}}^2 \\ \hat{F}_z \\ \hat{\mathbf{L}}^2 \\ \hat{L}_z \\ \hat{L}_3 \end{pmatrix} |\varphi_{\text{am}}\rangle_1 = \begin{pmatrix} F(F+1) \\ m_F \\ L(L+1) \\ m_L \\ k_L \end{pmatrix} |\varphi_{\text{am}}\rangle_1. \quad (10)$$

The Hilbert space is $\mathcal{H}_{\text{am}}^1 = \mathbb{C}^{2F+1} \otimes \left(\bigoplus_{L=0}^{\infty} \mathbb{C}^{(2L+1)^2}\right)$.

(2) Body frame uncoupled representation $\{\hat{\mathbf{J}}^2, \hat{J}_z, \hat{J}_3, \hat{\mathbf{S}}^2, \hat{S}_3\}$ with eigenstates

$$|\varphi_{\text{am}}\rangle_2 = |Jm_J k_J, Sk_S\rangle_2, \quad (11)$$

such that

$$\begin{pmatrix} \hat{\mathbf{S}}^2 \\ \hat{S}_3 \\ \hat{\mathbf{J}}^2 \\ \hat{J}_z \\ \hat{J}_3 \end{pmatrix} |\varphi_{\text{am}}\rangle_2 = \begin{pmatrix} S(S+1) \\ m_S \\ J(J+1) \\ m_J \\ k_J \end{pmatrix} |\varphi_{\text{am}}\rangle_2. \quad (12)$$

The Hilbert space is $\mathcal{H}_{\text{am}}^2 = \mathbb{C}^{2S+1} \otimes \left(\bigoplus_{J=0}^{\infty} \mathbb{C}^{(2J+1)^2}\right)$.

(3) Coupled representation $\{\hat{\mathbf{J}}^2, \hat{J}_z, \hat{\mathbf{S}}^2, \hat{\mathbf{L}}^2, \hat{L}_3\}$ with eigenstates

$$|\varphi_{\text{am}}\rangle_3 = |Jm_J, S, Lk_L\rangle_3, \quad (13)$$

such that

$$\begin{pmatrix} \hat{\mathbf{J}}^2 \\ \hat{J}_z \\ \hat{\mathbf{S}}^2 \\ \hat{\mathbf{L}}^2 \\ \hat{L}_3 \end{pmatrix} |\varphi_{\text{am}}\rangle_3 = \begin{pmatrix} J(J+1) \\ m_J \\ S(S+1) \\ L(L+1) \\ k_L \end{pmatrix} |\varphi_{\text{am}}\rangle_3. \quad (14)$$

The Hilbert space is $\mathcal{H}_{\text{am}}^3 = \bigoplus_{L=0}^{\infty} (\mathbb{C}^{2L+1} \otimes \mathbb{C}^{d_j}) \cong \bigoplus_{J=0}^{\infty} (\mathbb{C}^{2J+1} \otimes \mathbb{C}^{d_l})$, where $d_j = \sum_{J=|L-F|}^{L+F} 2J+1$ and $d_l = \sum_{L=|J-S|}^{J+S} 2L+1$.

The set 1 (2) contains the additional commuting operator $\hat{J}_z = \hat{F}_z + \hat{L}_z$ ($\hat{L}_3 = \hat{S}_3 + \hat{J}_3$), which has not been included since it is determined by some of the operators within the set and thus it does not represent an additional degree of freedom. The three Hilbert spaces of the representation are isomorphic (they correspond to a different choice of basis), and the change of basis is achieved by a unitary transformation. To switch from 1 to 3, one uses the relation

$$|Fm_F, Lm_L k_L\rangle_1 = \sum_{J=|L-F|}^{L+F} C_{Fm_F, Lm_L}^{Jm_J} |Jm_J, S, Lk_L\rangle_3 \quad (15)$$

and from 2 to 3 the relation

$$|Jm_J k_J, Sk_S\rangle_2 = \sum_{L=|J-S|}^{J+S} C_{Jk_J, Sk_S}^{Lk_L} |Jm_J, S, Lk_L\rangle_3. \quad (16)$$

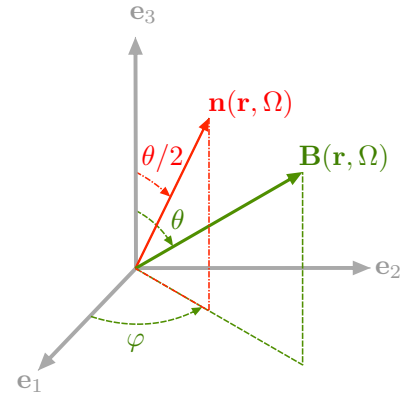


FIG. 2. Definition of the vector $\mathbf{n}(\mathbf{r}, \Omega)$ and $\mathbf{B}(\mathbf{r}, \Omega)$. The unitary \hat{U} is a rotation of an angle π around the local direction $\mathbf{n}(\mathbf{r}, \Omega)$. This transformation aligns \mathbf{e}_3 with $\mathbf{B}(\mathbf{r}, \Omega)$.

Here, $C_{Fm_F, Lm_L}^{Jm_J} = {}_3\langle Jm_J, S, Lk_L | Fm_F, Lm_L k_L \rangle_1$ and $C_{Jk_J, Sk_S}^{Lk_L} = {}_3\langle Jm_J, S, Lk_L | Jm_J k_J, Sk_S \rangle_2$ are Clebsch-Gordan coefficients under the Condon-Shortley convention (they are real numbers) [23]. $C_{J_1 m_1, J_2 m_2}^{J m}$ is nonzero provided $m = m_1 + m_2$ and $J \in \{J_1 + J_2, J_1 + J_2 - 1, \dots, |J_1 - J_2|\}$, which embodies the conservation of angular momentum.

The inspection of the Hamiltonian in the body frame, Eq. (8), suggests to use the representation 2 (body frame uncoupled representation). The basis of the total Hilbert space will thus be $\{|\psi_n\rangle \otimes |Jm_J k_J, Sk_S\rangle_2\}$.

III. DIAGONALIZATION OF THE MAGNETIC DIPOLE INTERACTION

In the context of magnetic trapping, it is convenient to apply a unitary transformation \hat{U} to the Hamiltonian such that the magnetic dipole interaction in Eq. (8) is transformed as follows:

$$\hat{U}^\dagger \mathbf{B}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{S}} \hat{U} = |\mathbf{B}(\hat{\mathbf{r}})| \hat{S}_3. \quad (17)$$

This can be achieved with the unitary transformation given by¹

$$\hat{U} \equiv \exp[-i\pi \mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{S}}], \quad (18)$$

where $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega}) = (n_1(\hat{\mathbf{r}}, \hat{\Omega}), n_2(\hat{\mathbf{r}}, \hat{\Omega}), n_3(\hat{\mathbf{r}}, \hat{\Omega}))^T$ is the unit vector that lies in the same plane as \mathbf{e}_3 and $\mathbf{B}(\hat{\mathbf{r}}, \hat{\Omega})$, and bisects the angle between them, see Fig. 2.

The body frame Hamiltonian (8) transformed by the unitary transformation (18) can be written as

$$\hat{H}' = \hat{U}^\dagger \hat{H} \hat{U} = \hat{H}_0 + \hat{H}_I + \hat{V}_D + \hat{V}_P, \quad (19)$$

where (1) \hat{H}_0 , which is diagonal in the basis $|Jm_J k_J, Sk_S\rangle_2$, is given by

$$\hat{H}_0 = \frac{\hat{\mathbf{p}}^2}{2M} + \frac{\hbar^2}{2I} (\hat{\mathbf{J}}^2 + 2\hat{S}_3 \hat{J}_3) - \hbar^2 D \hat{S}_3^2 + \hbar\gamma |\mathbf{B}(\hat{\mathbf{r}})| \hat{S}_3. \quad (20)$$

¹An analogous unitary transformation is also used in describing the magnetic trapping of atoms [29,30]. However, note that here the unitary operator (18) depends on $\hat{\Omega}$ and hence has to be treated more carefully.

Since local minima of $|\mathbf{B}(\hat{\mathbf{r}})|$ are allowed, the term $\hbar\gamma|\mathbf{B}(\hat{\mathbf{r}})|\hat{S}_3$ can trap the nanomagnet for macrospin states with $\langle\hat{S}_3\rangle > 0$.

(2) \hat{H}_I is given by

$$\hat{H}_I = \frac{\hbar^2}{2I}(\hat{J}_\uparrow\hat{S}_\downarrow + \hat{J}_\downarrow\hat{S}_\uparrow). \quad (21)$$

This term originates from the rotational kinetic energy of the nanomagnet, $\hbar(\hat{\mathbf{J}} + \hat{\mathbf{S}})^2/2I$ and couples the spin $\hat{\mathbf{S}}$ with the total angular momentum $\hat{\mathbf{J}}$.

(3) \hat{V}_D is given by

$$\hat{V}_D = \hbar^2 D \hat{S}_3^2 - \hbar^2 D [\hat{S}_3(1 - 2n_3(\hat{\mathbf{r}}, \hat{\Omega})) - n_3(\hat{\mathbf{r}}, \hat{\Omega})(n_\uparrow(\hat{\mathbf{r}}, \hat{\Omega})\hat{S}_\downarrow + n_\downarrow(\hat{\mathbf{r}}, \hat{\Omega})\hat{S}_\uparrow)]^2, \quad (22)$$

where we defined $n_\uparrow(\hat{\mathbf{r}}, \hat{\Omega}) = [n_\downarrow(\hat{\mathbf{r}}, \hat{\Omega})]^\dagger = n_1(\hat{\mathbf{r}}, \hat{\Omega}) - in_2(\hat{\mathbf{r}}, \hat{\Omega})$. This arises from the unitary transformation of the anisotropy interaction.

(4) \hat{V}_P is given by

$$\hat{V}_P = \frac{\hat{\mathbf{p}} \cdot \mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega}) + \mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{p}}}{2M} + \frac{\mathbf{A}^2(\hat{\mathbf{r}}, \hat{\Omega})}{2M}, \quad (23)$$

where $A_\nu(\hat{\mathbf{r}}, \hat{\Omega}) = -2\hbar[\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega}) \times \partial_\nu \mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})] \cdot \hat{\mathbf{S}}$. This term originates in the unitary transformation of the center of mass momentum $\hat{\mathbf{p}}$, which does not commute with \hat{U} .

$$R(\hat{\Omega}) = \frac{1}{2} \begin{pmatrix} (\hat{D}_{11}^1 + \hat{D}_{-1-1}^1 - \hat{D}_{1-1}^1 - \hat{D}_{-11}^1) & -i(\hat{D}_{11}^1 - \hat{D}_{-1-1}^1 + \hat{D}_{-11}^1 - \hat{D}_{1-1}^1) & \sqrt{2}(\hat{D}_{0-1}^1 - \hat{D}_{01}^1) \\ i(\hat{D}_{11}^1 - \hat{D}_{-1-1}^1 - \hat{D}_{-11}^1 + \hat{D}_{1-1}^1) & (\hat{D}_{11}^1 + \hat{D}_{-1-1}^1 + \hat{D}_{-1-1}^1 + \hat{D}_{-11}^1) & -i\sqrt{2}(\hat{D}_{0-1}^1 + \hat{D}_{01}^1) \\ \sqrt{2}(\hat{D}_{-10}^1 - \hat{D}_{10}^1) & i\sqrt{2}(\hat{D}_{10}^1 + \hat{D}_{-10}^1) & 2\hat{D}_{00}^1 \end{pmatrix}. \quad (26)$$

This allows to write \hat{V}_D and \hat{V}_P in terms of D matrices, since using Eq. (24) one has that

$$\begin{pmatrix} n_3(\hat{\mathbf{r}}, \hat{\Omega}) \\ n_\uparrow(\hat{\mathbf{r}}, \hat{\Omega}) \\ n_\downarrow(\hat{\mathbf{r}}, \hat{\Omega}) \end{pmatrix} = T(\hat{\Omega}) \begin{pmatrix} n_z(\hat{\mathbf{r}}) \\ n_+(\hat{\mathbf{r}}) \\ n_-(\hat{\mathbf{r}}) \end{pmatrix}, \quad (27)$$

where $n_\pm(\hat{\mathbf{r}}) \equiv n_x(\hat{\mathbf{r}}) \pm in_y(\hat{\mathbf{r}})$ and

$$T(\hat{\Omega}) \equiv \frac{1}{\sqrt{2}} \begin{pmatrix} \sqrt{2}\hat{D}_{00}^1 & \hat{D}_{-10}^1 & -\hat{D}_{10}^1 \\ -2\hat{D}_{01}^1 & -\sqrt{2}\hat{D}_{-11}^1 & \sqrt{2}\hat{D}_{11}^1 \\ 2\hat{D}_{0-1}^1 & \sqrt{2}\hat{D}_{-1-1}^1 & -\sqrt{2}\hat{D}_{1-1}^1 \end{pmatrix}. \quad (28)$$

As shown in Appendix A, the D matrices transform the state $|Jm_J k_J, S k_S\rangle_2$ in the following way:

$$\begin{aligned} & \hat{D}_{mk}^j |Jm_J k_J, S k_S\rangle_2 \\ &= \sum_{J'=|J-j|}^{J+j} \sqrt{\frac{2J+1}{2J'+1}} C_{jm, Jm_J}^{J'm'_j} C_{jk, Jk_J}^{J'k'_j} |J'm'_j k'_j, S k_S\rangle_2, \end{aligned} \quad (29)$$

The transformed Hamiltonian (19) is still exact. The terms \hat{V}_D and \hat{V}_P contain the components of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$, which are operators acting both on the center of mass subspace, through $\hat{\mathbf{r}}$, and on the angular momentum subspace, through the Euler angles $\hat{\Omega}$. One can show that

$$\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega}) = R(\hat{\Omega}) \mathbf{n}(\hat{\mathbf{r}}), \quad (24)$$

where $\mathbf{n}(\hat{\mathbf{r}})$ is the unit vector that lies in the same plane as $\mathbf{B}(\hat{\mathbf{r}})$ and \mathbf{e}_z and bisects the angle between the two. The operator $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$ acts on the angular momentum subspace through the matrix elements $R_{i\nu}(\hat{\Omega})$, which are a combination of trigonometric functions of the Euler angles, see Eq. (3). These can be expressed through the Wigner D -matrix tensor operators \hat{D}_{mk}^j [31], which are defined as follows. Consider the function $D_{mk}^j(\alpha, \beta, \gamma)$ as the matrix element of the general unitary rotation operator in quantum mechanics²

$$D_{mk}^j(\alpha, \beta, \gamma) \equiv \langle jk | e^{i\gamma \hat{J}_z} e^{i\beta \hat{J}_y} e^{i\alpha \hat{J}_z} | jm \rangle, \quad (25)$$

where $e^{i\gamma \hat{J}_z} e^{i\beta \hat{J}_y} e^{i\alpha \hat{J}_z}$ is the unitary representation of the Euler angles rotation matrix according to the convention adopted in Fig. 1. In Eq. (25), the angles α, β, γ are real parameters. The Wigner D -matrix tensor operator is then defined by $\hat{D}_{mk}^j \equiv D_{mk}^j(\hat{\alpha}, \hat{\beta}, \hat{\gamma})$, that is, by promoting the Euler angles to operators. Hereafter, we call the D -matrix tensor operator just D matrix. All the elements of the rotation matrix $R(\hat{\Omega})$ can be written as linear combination of \hat{D}_{mk}^j . The rotation matrix of Eq. (3) reads

where $m'_j = m_j + m$ and $k'_j = k_j + k$. For $j \neq 0$, the D -matrix operators couple subspaces of different J .

The Hamiltonian \hat{H}' , Eq. (19), which is derived from \hat{H} , Eq. (8), without any approximation, can be expressed as a function of the following operators: $\hat{\mathbf{r}}$, $\hat{\mathbf{p}}$, \hat{S}_3 , $\hat{S}_{\uparrow(\downarrow)}$, $\hat{\mathbf{J}}^2$, \hat{J}_3 , \hat{J}_z , $\hat{J}_{\uparrow(\downarrow)}$, and products of \hat{D}_{mk}^j (for $m, k = -1, 0, 1$).³ That is, it can be expressed without an explicit dependence on the Euler angles. We remark that in such a transformed frame, some of the mathematical operators appearing in \hat{H}' , Eq. (19), have a different physical meaning that those appearing in \hat{H} , Eq. (8). In particular note that (we define $\hat{M}' \equiv \hat{U}^\dagger \hat{M} \hat{U}$ for any operator \hat{M}),

$$\hat{\mathbf{r}}' = \hat{\mathbf{r}}, \quad (30)$$

$$\hat{\mathbf{p}}' = \hat{\mathbf{p}} + \mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega}), \quad (31)$$

$$\hat{\Omega}' = \hat{\Omega}, \quad (32)$$

$$\hat{S}'_3 = -\hat{S}_3 + 2n_3(\hat{\mathbf{r}}, \hat{\Omega})[\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{S}}], \quad (33)$$

²The ket $|jm\rangle$ is defined as $\hat{\mathbf{J}}^2|jm\rangle = j(j+1)|jm\rangle$ and $\hat{J}_z|jm\rangle = m|jm\rangle$, where $\hat{\mathbf{J}}$ is here a generic angular momentum operator.

³As shown in Appendix A, products of D matrices can always be written as a linear combination of D matrices \hat{D}_{mk}^j with $j \geq 1$.

$$\hat{J}'_3 = \hat{J}_3 + 2\hat{S}_3 - 2n_3(\hat{\mathbf{r}}, \hat{\Omega})[\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{S}}], \quad (34)$$

$$\hat{S}'_b = \hat{S}_3, \quad (35)$$

$$\hat{J}'_b = \hat{J}_b + \hat{S}_b - \hat{S}_3, \quad (36)$$

where we define the operators $\hat{J}_b = \mathbf{B}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{J}}/|\mathbf{B}(\hat{\mathbf{r}})|$ and $\hat{S}_b = \mathbf{B}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{S}}/|\mathbf{B}(\hat{\mathbf{r}})|$, which represent the projection along the local direction of the magnetic field. Note, for instance, that the mathematical operator \hat{S}_3 in the transformed frame has the physical meaning of the projection of the spin along the local magnetic field, as expected. However, this is not the case for \hat{J}_3 .

In the following sections, some assumptions are considered in the state of the nanomagnet to perform several approximations to the very rich, but complicated, Hamiltonian \hat{H}' given in Eq. (19).

IV. LAMB-DICKE APPROXIMATION

The first assumption is to consider that the state of the nanomagnet is such that $\langle \hat{S}_3 \rangle > 0$. This implies that the term $\gamma \hbar |\mathbf{B}(\hat{\mathbf{r}})| \hat{S}_3$ in \hat{H}_0 , see Eq. (20), can confine the nanomagnet around $\mathbf{r} \approx 0$ for a magnetic field with a local minima of $|\mathbf{B}(\mathbf{r})|$ at $\mathbf{r} = 0$ [32]. This allows to expand the functions of $\hat{\mathbf{r}}$ in the Hamiltonian (19) as a Taylor expansion. Note that only \hat{H}_0 , \hat{V}_D , and \hat{V}_P depend on $\hat{\mathbf{r}}$. The Lamb-Dicke approximation will consist in only keeping the lower orders of such expansions.

In \hat{H}_0 , this is done by expanding

$$|\mathbf{B}(\hat{\mathbf{r}})| \simeq B_0 + \sum_{\nu} \frac{B''_{\nu} \hat{r}_{\nu}^2}{2}, \quad (37)$$

where we have defined $B_0 \equiv |\mathbf{B}(0)|$ and $B''_{\nu} \equiv (\partial_{\nu}^2 |\mathbf{B}(\mathbf{r})|)_{\mathbf{r}=0}$. This expansion is valid provided

$$\left| \frac{\langle \hat{r}_{\nu}^2 \rangle B''_{\nu}}{B_0} \right| \ll 1 \quad \forall \nu. \quad (38)$$

In full generality the cross talking terms coupling different spatial directions can be made zero by choosing the laboratory frame axis where the symmetric matrix $(\partial_{\nu} \partial_{\mu} |\mathbf{B}(\mathbf{r})|)_{\mathbf{r}=0}$ is diagonal. Under the Lamb-Dicke approximation, the term \hat{H}_0 is approximated to

$$\begin{aligned} \hat{H}_0 \approx & \frac{\hat{\mathbf{p}}^2}{2M} + \frac{\hat{S}_3}{S} \sum_{\nu} \frac{M}{2} \omega_{\nu}^2 \hat{r}_{\nu}^2 + \hbar \omega_L \hat{S}_3 - \frac{\hbar \omega_D}{S} \hat{S}_3^2 \\ & + \frac{\hbar \omega_I}{2S} (\hat{\mathbf{J}}^2 + 2\hat{J}_3 \hat{S}_3), \end{aligned} \quad (39)$$

where we have defined the following frequencies:

$$\omega_{\nu} \equiv \sqrt{\frac{\hbar \gamma B''_{\nu}}{M_s}}, \quad (40)$$

$$\omega_I \equiv \frac{\hbar S}{I}, \quad (41)$$

$$\omega_D \equiv \hbar D S, \quad (42)$$

$$\omega_L \equiv \gamma B_0, \quad (43)$$

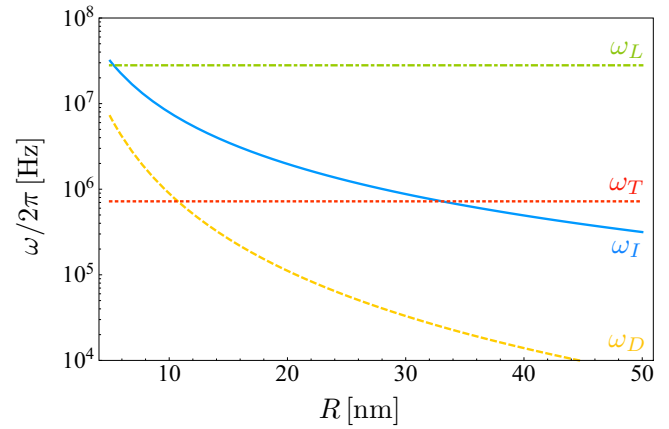


FIG. 3. The frequencies ω_L , ω_I , ω_D , and ω_T are plotted as a function of the radius R of the nanomagnet. We consider a Cobalt nanomagnet of density $\rho_{\text{Co}} = 8.9 \times 10^3 \text{ Kg/m}^3$, $S/N = 1.7$, and $T_b = \hbar^2 D S^2 / K_b = 30 \text{ K}$. The external Ioffe-Pritchard field is used with $B_0 = 10^{-3} \text{ T}$, $B' = 10^4 \text{ T/m}$, and $B'' = 10^6 \text{ T/m}^2$ [29,30].

with $M_s \equiv M/S$. Typical values and hierarchy of these frequencies depend on the size of the particle and on the external bias field B_0 , see Fig. 3. The center-of-mass position operator can be expressed as

$$\hat{r}_v = \frac{r_{0v}}{\sqrt{S}} (\hat{b}_v + \hat{b}_v^\dagger), \quad (44)$$

where $r_{0v} = [\hbar / (2M_s \omega_v)]^{1/2}$ and $[\hat{b}_v, \hat{b}_\mu^\dagger] = \delta_{v\mu}$.

The $\hat{\mathbf{r}}$ dependence of the terms \hat{V}_D and \hat{V}_P arise from the components of $\mathbf{n}(\hat{\mathbf{r}})$. Under the Lamb-Dicke approximation one can use the expansion

$$\begin{aligned} n_\mu(\hat{\mathbf{r}}) \approx & n_\mu(0) + \sum_{\nu} \frac{r_{0\nu}}{\sqrt{S}} (\hat{b}_\nu + \hat{b}_\nu^\dagger) \partial_\nu n_\mu \\ & + \frac{1}{2} \sum_{\nu\lambda} \frac{r_{0\nu} r_{0\lambda}}{S} (\hat{b}_\nu^\dagger + \hat{b}_\nu) (\hat{b}_\lambda^\dagger + \hat{b}_\lambda) \partial_\nu \partial_\lambda n_\mu, \end{aligned} \quad (45)$$

where we define $\partial_\nu n_\mu = \partial_\nu n_\mu(\mathbf{r})|_{\mathbf{r}=0}$ for shortness. Since the vector $\mathbf{n}(\mathbf{r})$ defined in Sec. III depends on \mathbf{r} only through the magnetic field $\mathbf{B}(\mathbf{r})$, this expansion is justified when the condition (38) is fulfilled. Recall that to obtain the Taylor expansion for the components of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$, it is sufficient to use Eqs. (27) and (45). This allows to expand \hat{V}_D and \hat{V}_P in powers of $\hat{\mathbf{r}}$, see Appendixes C and D.

V. HOLSTEIN-PRIMAKOFF APPROXIMATION: BOSONIZATION

Our aim is to describe the nanomagnet in the following regime:

$$\langle \hat{\mathbf{r}} \rangle \approx 0, \quad (46)$$

$$\langle \hat{S}_3 \rangle \lesssim S, \quad (47)$$

$$\langle \hat{\mathbf{J}}^2 \rangle \approx \langle \hat{\mathbf{S}}^2 \rangle = S(S+1), \quad (48)$$

$$\langle \hat{J}_3 \rangle \approx \langle \hat{J}_z \rangle \gtrsim -J \approx -S. \quad (49)$$

Recall that for a nanomagnet $S \gg 1$. We called this regime *highly polarized*, which corresponds to the nanomagnet being (1) highly confined in position, (2) with the macrospin highly antialigned to the external magnetic field (recall that $\hat{\mathbf{S}} = -\hat{\mathbf{F}}$), and (3) nearly not rotating, namely $\langle \hat{\mathbf{L}}^2 \rangle \gtrsim 0$. According to Eq. (36), we can approximate $\hat{J}'_b \approx \hat{J}_3$ when the nanomagnet is nearly not spinning.

We apply a Holstein-Primakoff (HP) boson mapping [33] to express the angular momentum operators and the D matrices as a function of a set of bosonic operators. This mapping allows us to express \hat{H}' , Eq. (19), in terms of bosonic operators. The exact mapping yields nonlinear bosonic terms to keep the angular momentum character of the original operators. Nevertheless, by assuming the highly-polarized regime of the nanomagnet, a quadratic Hamiltonian in bosonic operators can be obtained under the so-called HP approximation.

A. Holstein-Primakoff boson mapping

The HP boson mapping for the spin angular momentum is given by [33]

$$\hat{S}_3 = S - \hat{s}^\dagger \hat{s}, \quad \hat{S}_\uparrow = \hat{S}_\downarrow = (2S - \hat{s}^\dagger \hat{s})^{1/2} \hat{s}. \quad (50)$$

We have introduced the spin bosonic operator $[\hat{s}, \hat{s}^\dagger] = 1$. In the highly-polarized regime, one can perform the HP approximation, namely,

$$\hat{S}_3 = S - \hat{s}^\dagger \hat{s}, \quad \hat{S}_\uparrow = \hat{S}_\downarrow \approx \sqrt{2S} \hat{s}. \quad (51)$$

The boson mapping for the angular momentum operator $\hat{\mathbf{J}}$ and the D matrices has to be done more carefully since J is not fixed, as discussed in Eq. (29). The HP map can be generalized by promoting the quantum number J to the operator $\hat{J} \equiv \hat{d}^\dagger \hat{d} / 2$ with $[\hat{d}, \hat{d}^\dagger] = 1$ [9]. The exact mapping is given by

$$\begin{aligned} \hat{J}_3 &= -\frac{\hat{d}^\dagger \hat{d}}{2} + \hat{k}^\dagger \hat{k}, \\ \hat{J}_\uparrow &= \hat{J}_\downarrow = \hat{k}^\dagger \left(\frac{\hat{d}^\dagger \hat{d}}{2} - \hat{k}^\dagger \hat{k} \right)^{1/2}, \\ \hat{J}_z &= -\frac{\hat{d}^\dagger \hat{d}}{2} + \hat{m}^\dagger \hat{m}, \\ \hat{J}_+ &= \hat{J}_- = \hat{m}^\dagger \left(\frac{\hat{d}^\dagger \hat{d}}{2} - \hat{m}^\dagger \hat{m} \right)^{1/2}, \\ \hat{\mathbf{J}}^2 &= \frac{\hat{d}^\dagger \hat{d}}{2} \left(\frac{\hat{d}^\dagger \hat{d}}{2} + 1 \right). \end{aligned} \quad (52)$$

We have introduced the angular momentum bosonic operators $[\hat{m}, \hat{m}^\dagger] = [\hat{k}, \hat{k}^\dagger] = 1$. One can show that the mapping fulfills the commutation rules given in Table I. Therefore the general state $|Jm_J k_J, Sk_S\rangle_2$ can be obtained, using the bosonic mapping, from the vacuum of four bosonic modes, namely,

$$|Jm_J k_J, Sk_S\rangle_2 = \mathcal{N} (\hat{d}^\dagger)^{2J} (\hat{m}^\dagger)^{J+m_J} (\hat{k}^\dagger)^{J+k_J} (\hat{s}^\dagger)^{k_S} |0\rangle, \quad (53)$$

where $\mathcal{N} = [(2J)!(J+m_J)!(J+k_J)!k_S!]^{-1/2}$. In the highly polarized regime of the nanomagnet, it is useful to define $\hat{d} \equiv \sqrt{2J} + \hat{j}$, where $J \gg 1$. In this case, the HP approximation

of Eq. (52) reads

$$\begin{aligned} \hat{J}_3 &= -J - \frac{\sqrt{2J}}{2} (\hat{j}^\dagger + \hat{j}) - \frac{\hat{j}^\dagger \hat{j}}{2} + \hat{k}^\dagger \hat{k}, \\ \hat{J}_\uparrow &= \hat{J}_\downarrow \approx \sqrt{2J} \hat{k}^\dagger + \frac{\hat{j}^\dagger + \hat{j}}{2} \hat{k}^\dagger + O\left(\frac{1}{\sqrt{J}}\right), \\ \hat{J}_z &= -J - \frac{\sqrt{2J}}{2} (\hat{j}^\dagger + \hat{j}) - \frac{\hat{j}^\dagger \hat{j}}{2} + \hat{m}^\dagger \hat{m}, \\ \hat{J}_+ &= \hat{J}_- \approx \sqrt{2J} \hat{m}^\dagger + \frac{\hat{j}^\dagger + \hat{j}}{2} \hat{m}^\dagger + O\left(\frac{1}{\sqrt{J}}\right), \end{aligned} \quad (54)$$

and for the total angular momentum $\hat{\mathbf{J}}^2$ is

$$\begin{aligned} \hat{\mathbf{J}}^2 &= J(J+1) + J\sqrt{2J}(\hat{j}^\dagger + \hat{j}) \\ &+ J \left[\frac{(\hat{j} + \hat{j}^\dagger)^2}{2} + \hat{j}^\dagger \hat{j} \right] \\ &+ \frac{\sqrt{2J}}{2} [\{\hat{j}^\dagger + \hat{j}, \hat{j}^\dagger \hat{j}\} + (\hat{j}^\dagger + \hat{j})] \\ &+ \frac{\hat{j}^\dagger \hat{j}}{2} \left(\frac{\hat{j}^\dagger \hat{j}}{2} + 1 \right). \end{aligned} \quad (55)$$

Let us now address the boson mapping of the D matrices [34]. This can be done using Eqs. (29) and (53). Knowing how the single creation and annihilation bosonic operators act on $|Jm_J k_J, Sk_S\rangle_2$, we can derive an expression of \hat{D}_{mk}^q in terms of $\{\hat{j}, \hat{j}^\dagger, \hat{m}, \hat{m}^\dagger, \hat{k}, \hat{k}^\dagger\}$, such that when applied to $|Jm_J k_J, Sk_S\rangle_2$ using the mapping (53) gives exactly the right-hand side of Eq. (29). While this can be done exact, in Table II, we provide the mapping, using the HP approximation up to order $O(1/J)$, for the \hat{D}_{mk}^q appearing in the Hamiltonian.

VI. QUADRATIC BOSONIZED HAMILTONIAN

At this stage, one can use the Lamb-Dicke approximation and the HP mapping of the angular momentum operators and the D matrices to write the Hamiltonian \hat{H}' , Eq. (19), as a function of the bosonic modes of the system: $\{\hat{b}_x, \hat{b}_y, \hat{b}_z, \hat{j}, \hat{s}, \hat{k}, \hat{m}\}$. In principle, this Hamiltonian can be divided into two parts:

$$\hat{H}' = \hat{H}_G + \hat{H}_{NG}. \quad (56)$$

\hat{H}_G collects terms containing up to two bosonic operators (quadratic form), which lead to Gaussian physics. \hat{H}_{NG} contains normally ordered terms with more than two bosonic operators. Let us first obtain \hat{H}_G and then discuss when \hat{H}_{NG} can be neglected.

A. Quadratic terms

In order to obtain \hat{H}_G , one substitutes Eqs. (51), (54), (55), and Table II into the Hamiltonian (19) after doing the Lamb-Dicke approximation. After normal ordering the bosonic operators, one obtains the quadratic Hamiltonian of the system. The quadratic terms arising from \hat{H}_0 , Eq. (39), and \hat{H}_I , Eq. (21), are straightforward to obtain. The bosonization of the terms \hat{V}_D , Eq. (22), and \hat{V}_P , Eq. (23), is more involved, see details in Appendixes C and D.

In deriving the quadratic Hamiltonian, we assume the Ioffe-Pritchard configuration for the external magnetic field. This

TABLE II. Bosonized expression, up to order $O(1/J)$, for the relevant $\hat{\mathcal{D}}_{mk}^j$ appearing in the Hamiltonian.

 $j = 1:$

$$\begin{aligned}\hat{\mathcal{D}}_{00}^1 &= 1 - \frac{1}{J}[1 + \hat{k}^\dagger \hat{k} + \hat{m}^\dagger \hat{m} - \hat{k}^\dagger \hat{m}^\dagger - \hat{k} \hat{m}] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{01}^1 &= -[\hat{\mathcal{D}}_{0-1}^1]^\dagger = \frac{1}{\sqrt{J}}(\hat{k}^\dagger - \hat{m}) - \frac{1}{2\sqrt{2}}\frac{1}{J}[(\hat{j}^\dagger + \hat{j})\hat{k}^\dagger + (\hat{j} - 3\hat{j}^\dagger)\hat{m}] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{10}^1 &= -[\hat{\mathcal{D}}_{-10}^1]^\dagger = \frac{1}{\sqrt{J}}(\hat{m}^\dagger - \hat{k}) - \frac{1}{2\sqrt{2}}\frac{1}{J}[(\hat{j}^\dagger + \hat{j})\hat{m}^\dagger + (\hat{j} - 3\hat{j}^\dagger)\hat{k}] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{11}^1 &= [\hat{\mathcal{D}}_{-1-1}^1]^\dagger = 1 - \frac{\hat{j}^\dagger - \hat{j}}{\sqrt{2J}} - \frac{1}{2J}[2(\hat{j}^\dagger \hat{j} + 1) + \hat{k}^\dagger \hat{k} + \hat{m}^\dagger \hat{m} - 2\hat{m}^\dagger \hat{k}^\dagger + \hat{j}^2 - \frac{3}{4}(\hat{j}^\dagger + \hat{j})^2 - \frac{1}{4}(\hat{j}^\dagger - \hat{j})^2] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{-1-1}^1 &= [\hat{\mathcal{D}}_{-11}^1]^\dagger = \frac{1}{2J}(\hat{k}^2 - 2\hat{m}^\dagger \hat{k} + (\hat{m}^\dagger)^2) + O\left(\frac{1}{J\sqrt{J}}\right)\end{aligned}$$

 $j = 2:$

$$\begin{aligned}\hat{\mathcal{D}}_{00}^2 &= 1 - \frac{3}{J}[1 + \hat{k}^\dagger \hat{k} + \hat{m}^\dagger \hat{m} - \hat{k}^\dagger \hat{m}^\dagger - \hat{k} \hat{m}] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{01}^2 &= -[\hat{\mathcal{D}}_{0-1}^2]^\dagger = \sqrt{\frac{3}{J}}(\hat{k}^\dagger - \hat{m}) - \frac{\sqrt{3}}{2\sqrt{2}}\frac{1}{J}[(\hat{j}^\dagger + \hat{j})\hat{k}^\dagger + (\hat{j} - 3\hat{j}^\dagger)\hat{m}] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{10}^2 &= -[\hat{\mathcal{D}}_{-10}^2]^\dagger = \sqrt{\frac{3}{J}}(\hat{m}^\dagger - \hat{k}) - \frac{\sqrt{3}}{2\sqrt{2}}\frac{1}{J}[(\hat{j}^\dagger + \hat{j})\hat{m}^\dagger + (\hat{j} - 3\hat{j}^\dagger)\hat{k}] + O\left(\frac{1}{J\sqrt{J}}\right)\end{aligned}$$

 $j = 4:$

$$\begin{aligned}\hat{\mathcal{D}}_{00}^4 &= 1 - \frac{10}{J}[1 + \hat{k}^\dagger \hat{k} + \hat{m}^\dagger \hat{m} - \hat{k}^\dagger \hat{m}^\dagger - \hat{k} \hat{m}] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{01}^4 &= -[\hat{\mathcal{D}}_{0-1}^4]^\dagger = \sqrt{\frac{10}{J}}(\hat{k}^\dagger - \hat{m}) - \frac{\sqrt{10}}{2\sqrt{2}}\frac{1}{J}[(\hat{j}^\dagger + \hat{j})\hat{k}^\dagger + (\hat{j} - 3\hat{j}^\dagger)\hat{m}] + O\left(\frac{1}{J\sqrt{J}}\right) \\ \hat{\mathcal{D}}_{10}^4 &= -[\hat{\mathcal{D}}_{-10}^4]^\dagger = \sqrt{\frac{10}{J}}(\hat{m}^\dagger - \hat{k}) - \frac{\sqrt{10}}{2\sqrt{2}}\frac{1}{J}[(\hat{j}^\dagger + \hat{j})\hat{m}^\dagger + (\hat{j} - 3\hat{j}^\dagger)\hat{k}] + O\left(\frac{1}{J\sqrt{J}}\right)\end{aligned}$$

magnetic field is characterized by three parameters: the bias field B_0 , the field gradient along z , B' , and the field curvature along z , B'' , see Appendix B for details. With this field one has that $\omega_x = \omega_y \equiv \omega_T$, and the first-order terms in the Lamb-Dicke expansion of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$ can be characterized by a single expansion parameter $\eta = r_0 B' / (2B_0)$, where $r_0 \equiv r_{0x} = r_{0y}$.

After some effort, one obtains the following quadratic Hamiltonian:

$$\hat{H}_G = \hat{H}_0 + \hat{V}_s + \hat{V}_b, \quad (57)$$

where (1) \hat{H}_0 is given by

$$\begin{aligned}\frac{\hat{H}_0}{\hbar} &= \omega_r \hat{b}_r^\dagger \hat{b}_r + \omega_l \hat{b}_l^\dagger \hat{b}_l + \omega_z \hat{b}_z^\dagger \hat{b}_z - \Delta \hat{s}^\dagger \hat{s} \\ &+ \omega_j \hat{j}^\dagger \hat{j} + \xi_1 (\hat{j}^\dagger + \hat{j}) + \xi_2 (\hat{j}^\dagger + \hat{j})^2 \\ &+ \omega_m \hat{m}^\dagger \hat{m} + \omega_k \hat{k}^\dagger \hat{k} - \omega_m (\hat{m}^\dagger \hat{k}^\dagger + \hat{m} \hat{k}). \quad (58)\end{aligned}$$

The frequencies are given in Table III. We define the modes $\hat{b}_{r(l)} = (\hat{b}_x \mp i\hat{b}_y) / \sqrt{2}$ describing a counter clock wise (clock wise) circular motion of the center of mass. Note that Δ can be controlled via the strength of B_0 and can, in principle, be either positive or negative (see Fig. 3). As seen below, the mode \hat{j} is decoupled from the other modes in the quadratic Hamiltonian.

(2) \hat{V}_s is given by

$$\frac{\hat{V}_s}{\hbar} = g_k (\hat{s}^\dagger \hat{k}^\dagger + \hat{s} \hat{k}) - g_m (\hat{s}^\dagger \hat{m} + \hat{s} \hat{m}^\dagger). \quad (59)$$

This term describes the coupling between the spin degree of freedom and the total angular momentum. The coupling strengths are given in Table III. In essence, this term describes the Einstein-de Haas effect.

(3) \hat{V}_b is given by

$$\begin{aligned}\frac{\hat{V}_b}{\hbar} &= g_l \hat{b}_l (\hat{m}^\dagger - \hat{k}) + g_r \hat{b}_r^\dagger (\hat{m}^\dagger - \hat{k}) \\ &- g_s \hat{s}^\dagger (\hat{b}_r^\dagger - \hat{b}_l) + \text{H.c.} \quad (60)\end{aligned}$$

This term describes the coupling between the center-of-mass motion and the angular momenta. The coupling strengths are given in Table III. Note that the coupling is induced due to the

TABLE III. Definition of the frequencies appearing in Eqs. (58)–(60) in terms of the fundamental frequencies of the system, $\omega_l, \omega_L, \omega_D$, and ω_v . In the expression below, we neglected terms smaller than $O(\omega\eta)$, where $\omega = \omega_l, \omega_L, \omega_D$, or ω_v .

$\omega_r \hat{b}_r^\dagger \hat{b}_r$	$\omega_r \equiv \omega_T$
$\omega_l \hat{b}_l^\dagger \hat{b}_l$	$\omega_l \equiv \omega_T$
$-\Delta \hat{s}^\dagger \hat{s}$	$\Delta \equiv \omega_L - 2\omega_D - \omega_l \frac{J}{S}$
$\omega_k \hat{k}^\dagger \hat{k}$	$\omega_k \equiv \omega_l + 8\omega_D \frac{S}{J}$
$\omega_m \hat{m}^\dagger \hat{m}$	$\omega_m \equiv 8\omega_D \frac{S}{J}$
$\omega_j \hat{j}^\dagger \hat{j}$	$\omega_j = \frac{\omega_l}{2} \left(\frac{J}{S} - 1\right)$
$\xi_1 (\hat{j}^\dagger + \hat{j})$	$\xi_1 = \frac{\sqrt{2J}}{2} \omega_l \left(\frac{J}{S} - 1\right)$
$\xi_2 (\hat{j}^\dagger + \hat{j})^2$	$\xi_2 = \frac{\omega_l}{4} \frac{J}{S}$
$g_k \hat{s} \hat{k}$	$g_k \equiv \omega_l \sqrt{\frac{J}{S}} + 4\omega_D \sqrt{\frac{S}{J}}$
$g_m \hat{s} \hat{m}^\dagger$	$g_m \equiv 4\omega_D \sqrt{\frac{S}{J}}$
$g_r \hat{b}_r^\dagger (\hat{m}^\dagger - \hat{k})$	$g_r \equiv \eta(8\omega_D + 2\omega_T) \sqrt{\frac{S}{J}}$
$g_l \hat{b}_l (\hat{m}^\dagger - \hat{k})$	$g_l \equiv \eta(8\omega_D - 2\omega_T) \sqrt{\frac{S}{J}}$
$g_s \hat{s}^\dagger (\hat{b}_r^\dagger - \hat{b}_l)$	$g_s \equiv 2\omega_T \eta$

nonzero magnetic field gradient since $g_r = g_l = g_s = 0$ for $B' = 0$.

The derivation of the quadratic Hamiltonian \hat{H}_G is the main result of this article. The nonquadratic contributions given by \hat{H}_{NG} can be typically neglected, see below. \hat{H}_G describes the quantum dynamics of the nanomagnet in the magnetic trap provided it is in the highly-polarized regime defined by Eqs. (46)–(49). Under which physical parameters the highly-polarized regime is stable, the rich physics and wealth of applications that \hat{H}_G can describe, and how to implement it in an experimentally feasible scenario, are very interesting questions that lie beyond the scope of this paper and will be addressed elsewhere [24].

B. Nonquadratic terms: when can they be neglected?

The nonquadratic Hamiltonian \hat{H}_{NG} collects the terms with more than two normally ordered bosonic operators $\{\hat{b}_x, \hat{b}_y, \hat{b}_z, \hat{j}, \hat{s}, \hat{k}, \hat{m}\}$. In the highly-polarized regime, Eqs. (46)–(49) $J \approx S \gg 1$, one can realize that terms with an additional bosonic operator are a factor $1/\sqrt{S}$ smaller. All the energy terms that appear in the Hamiltonian $\hat{H}' = \hat{H}_G + \hat{H}_{NG}$ can be expressed in terms of the frequencies $\omega_T, \omega_z, \omega_D, \omega_I$, and ω_L . Establishing a hierarchy between these frequencies is needed to understand when the nonlinear terms can be neglected. The strength of these frequencies depends on the size of the nanomagnet and on the external magnetic field. As shown in Fig. 3, different regimes can be achieved but we consider the following one:

$$\omega_L \geq \omega_I \gg \omega_D \sim \omega_T, \omega_z. \quad (61)$$

In order to neglect the nonquadratic contribution in the bosonized Hamiltonian, one needs to compare the order of the strongest of the nonquadratic terms in \hat{H}_{NG} with the weakest contribution kept in \hat{H}_G taking into account Eq. (61). The smallest terms in \hat{H}_G are of the order $O(\omega_T \eta)$ and $O(\omega_D \eta)$. The strongest nonlinear term contribution comes from the bosonization of \hat{H}_I and reads

$$\hat{V}_{jkm} = \frac{\hbar \omega_I}{2\sqrt{2S}} (\hat{j}^\dagger + \hat{j})(\hat{k}^\dagger \hat{s}^\dagger + \hat{k} \hat{s}) \sim O\left(\frac{\omega_I}{\sqrt{S}}\right). \quad (62)$$

Therefore \hat{H}_{NG} can be neglected provided

$$\eta \gg \min\left\{\frac{\omega_I}{\omega_D \sqrt{S}}, \frac{\omega_I}{\omega_T \sqrt{S}}\right\}, \quad (63)$$

which can be easily satisfied under typical experimentally feasible parameters [24].

VII. CONCLUSION AND OUTLOOK

In conclusion, we have shown how to describe the quantum dynamics of a nanomagnet with uniaxial anisotropy in a magnetic trap. Starting from the basic Hamiltonian under the macroscopin approximation, we have performed (1) a change of variables to describe the dynamics in the body frame, (2) a unitary transformation to diagonalize the magnetic dipole coupling, (3) the Lamb-Dicke expansion around the trapping position of the nanomagnet, (4) a bosonic mapping of the angular momentum operators, and (5) the Holstein-Primakoff

expansion when the macrospin is well antialigned and the nanomagnet is nearly nonrotating (highly polarized regime).

This allows to derive a quadratic Hamiltonian depending on the bosonic operators that describes both the Einstein-de Haas coupling between the macrospin and the rotational angular momentum, and the magnetic-trap-induced-coupling between the center-of-mass motion and the rotational angular momentum. The nonquadratic terms can be neglected in the highly-polarized regime with typical experimental parameters.

The derivation of the Hamiltonian can be straightforwardly extended to cover more general situations. For instance, one could consider ellipsoidal objects instead of spherical and other forms of magnetic anisotropy (e.g., cubic anisotropy). This would change the rotational kinetic term and the magnetic anisotropy term, respectively, in the very initial Hamiltonian but the derivation to obtain the quadratic Hamiltonian would be analogous. Should one also be interested in describing the magnetic coupling of the nanomagnet with the quantum electromagnetic field, one could just add the quantized magnetic field operator to the magnetic dipole interaction term in the very initial Hamiltonian. This could be used to calculate vacuum forces and decay rates of the eigenstates of the Hamiltonian. Also note that the derivation of the quadratic Hamiltonian is constructive in the sense that one could consider next orders in both the Lamb-Dicke and the Holstein-Primakoff expansions to obtain the higher order nonquadratic contributions.

The unitary transformation in Sec. III is motivated by the goal of making the magnetic trapping potential appear explicitly in the Hamiltonian of the system. However, this transformation is not necessary to describe the dynamics of the quantum fluctuations around a stable state of the system. One could carry out the bosonization of the fluctuations in the Hamiltonian Eq. (8), before applying the unitary transformation. While this is mathematically equivalent, one should nevertheless be careful when performing the approximations after bosonizing since the physical meaning of the fluctuations, should one bosonize the same mathematical operators, would be different in the two cases. Recall the remark made in the end of Sec. III.

The derived quadratic Hamiltonian can be handled with the usual techniques of quantum optics and describes a very rich and original physical system. This opens many further directions that we are currently investigating [24]. Some of them are the following: (i) an experimental proposal where nanomagnets can be levitated in a Z-shaped atom chip and brought to the quantum regime by inductively coupling its motion to a flux-dependent quantum circuit. This coupling can be used to cool the center-of-mass motion to the ground state as well as to sympathetically cool the rotational angular momentum and the macroscopin degrees of freedom. (ii) The proposal and study of magnetically levitating the nanomagnet using the Meissner field induced on a superconductive film with a hole [35]. This permanent passive short-distance trap could be used to measure Casimir and short-distance gravitational forces; (iii) once in the quantum regime, exploit the nanomagnet as a nanogyroscope. (iv) Perform matter-wave interferometry by releasing the ground-state cooled nanomagnet from the trap. (v) Explore the regimes where the non-Gaussian physics given by the nonquadratic terms is

relevant, for instance, in the context of superradiance effects, etc.

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APPENDIX A: PROPERTIES OF THE WIGNER D -MATRIX TENSOR OPERATOR

In this Appendix, we list some useful properties of the D -matrix tensor operator. We will not prove them. The interested reader is referred to [22,23] for more details. These are the following. (1) The matrix elements of \hat{D}_{mk}^q between the eigenstates $|Lm_L k_L\rangle$ of the operators \hat{L}^2, \hat{L}_z , and \hat{L}_3 read

$$\langle L' m'_L k'_L | \hat{D}_{mk}^q | L m_L k_L \rangle = \frac{8\pi^2}{2L'+1} C_{qm, Lm_L}^{L' m'_L} C_{qk, Lk_L}^{L' k'_L}. \quad (\text{A1})$$

(2) The matrix elements of \hat{D}_{mk}^q on the basis $|Jm_J k_J, Sk_S\rangle_2$ read

$$\begin{aligned} & {}_2 \langle J' m'_J k'_J, S' k'_S | \hat{D}_{mk}^q | J m_J k_J, S k_S \rangle_2 \\ &= \delta_{SS'} \delta_{k_S k'_S} \sqrt{\frac{2J'+1}{2J'+1}} C_{qm, Jm_J}^{J' m'_J} C_{qk, Jk_J}^{J' k'_J} \end{aligned} \quad (\text{A2})$$

This result can be proved by changing the basis to $|Lm_L k_L, Sk_S\rangle_1$ through Eqs. (15) and (16) and then applying Eq. (A1).

(3) The product of two D -matrix operators reads

$$\hat{D}_{m_1 k_1}^{q_1} \hat{D}_{m_2 k_2}^{q_2} = \sum_{Q=|q_1-q_2|}^{q_1+q_2} \sum_{M, K=-Q}^Q C_{q_1 m_1, q_2 m_2}^{QM} C_{q_1 k_1, q_2 k_2}^{QK} \hat{D}_{MK}^Q. \quad (\text{A3})$$

(4) The D -matrix operators fulfill the following commutation relations with the total angular momentum of the system \hat{J} :

$$\begin{aligned} [\hat{J}_z, \hat{D}_{mk}^q] &= m \hat{D}_{mk}^q, \\ [\hat{J}_\pm, \hat{D}_{mk}^q] &= \sqrt{(q \mp m)(q \pm m + 1)} \hat{D}_{m\pm 1k}^q, \\ [\hat{J}_3, \hat{D}_{mk}^q] &= k \hat{D}_{mk}^q, \\ [\hat{J}_\uparrow, \hat{D}_{mk}^q] &= \sqrt{(q-k)(q+k+1)} \hat{D}_{mk+1}^q, \\ [\hat{J}_\downarrow, \hat{D}_{mk}^q] &= \sqrt{(q+k)(q-k+1)} \hat{D}_{mk-1}^q. \end{aligned} \quad (\text{A4})$$

The strategy to prove these relations is via the properties of the function $D_{mk}^j(\alpha, \beta, \gamma)$, defined in Eq. (25). It can be shown that $D_{mk}^j(\alpha, \beta, \gamma) \sim \langle \alpha, \beta, \gamma | L, m_L, k_L \rangle$. These eigenfunctions satisfy the following normalization condition:

$$\begin{aligned} & \int_0^{2\pi} d\alpha \int_0^\pi d\beta \sin \beta \int_0^{2\pi} d\gamma (D_{m_1 k_1}^{q_1}(\alpha, \beta, \gamma))^* D_{m_2 k_2}^{q_2}(\alpha, \beta, \gamma) \\ &= \frac{8\pi^2}{2q_1+1} \delta_{q_2, q_1} \delta_{m_2 m_1} \delta_{k_1 k_2}. \end{aligned} \quad (\text{A5})$$

Moreover, it can be shown that the product of two such eigenfunction can be written as a linear combination of single eigenfunctions with different indexes according to the following relation:

$$\begin{aligned} & D_{m_1 k_1}^{q_1}(\Omega) D_{m_2 k_2}^{q_2}(\Omega) \\ &= \sum_{Q=|q_1-q_2|}^{q_1+q_2} \sum_{M, K=-Q}^Q C_{q_1 m_1, q_2 m_2}^{QM} C_{q_1 k_1, q_2 k_2}^{QK} D_{MK}^Q(\Omega). \end{aligned} \quad (\text{A6})$$

Combining the results in Eqs. (A5) and (A6), one can prove the following identity involving the angular integral of three eigenfunction:

$$\begin{aligned} & \int_0^{2\pi} d\alpha \int_0^\pi d\beta \sin \beta \int_0^{2\pi} d\gamma [D_{M'K'}^{L'}(\Omega)]^* D_{mk}^q(\Omega) D_{MK}^L(\Omega) \\ &= \frac{8\pi^2}{2L'+1} C_{qm, LM}^{L' M'} C_{qk, LK}^{L' K'}. \end{aligned} \quad (\text{A7})$$

The integral in Eq. (A7) can be used to obtain Eq. (A2). See Refs. [22,23] for further details.

APPENDIX B: IOFFE-PRITCHARD MAGNETIC TRAP

Among the many different field configurations realizable in the magnetic traps, the Ioffe-Pritchard configuration is one of the most common. The shape of this magnetic field close to the vertical axis z reads [36]

$$\begin{aligned} \mathbf{B}(\mathbf{r}) &= \mathbf{e}_x \left(B'x - \frac{B''}{2}xz \right) - \mathbf{e}_y \left(B'y + \frac{B''}{2}zy \right) \\ &+ \mathbf{e}_z \left[B_0 + \frac{B''}{2} \left(z^2 - \frac{x^2 + y^2}{2} \right) \right], \end{aligned} \quad (\text{B1})$$

where $B_0 = |\mathbf{B}(0)|$, $B' \equiv \partial_z B_z(\mathbf{r})|_{\mathbf{r}=0}$, and $B'' \equiv \partial_z^2 B_z(\mathbf{r})|_{\mathbf{r}=0}$.

When the particle is confined at the center of the trap, namely,

$$\sqrt{\langle \hat{z}^2 \rangle} \ll \sqrt{\frac{B_0}{B''}}, \quad (\text{B2})$$

$$\sqrt{\langle \hat{x}^2 \rangle} \approx \sqrt{\langle \hat{y}^2 \rangle} \ll \sqrt{\frac{B_0^2}{B'^2 - B_0 B''}} \sim \frac{B_0}{B'}, \quad (\text{B3})$$

one can approximate

$$|\mathbf{B}(\mathbf{r})| \simeq B_0 + \frac{B''}{2} z^2 + \frac{B'^2 - B_0 B''/2}{2B_0} (x^2 + y^2). \quad (\text{B4})$$

By substituting this expansion into the Hamiltonian \hat{H}_0 in Eq. (20), one obtains a three-dimensional Harmonic potential

with trapping frequencies

$$\omega_x = \omega_y = \omega_T \equiv \sqrt{\frac{\hbar\gamma}{M_s B_0} \left(B'^2 - \frac{1}{2} B_0 B'' \right)},$$

$$\omega_z = \sqrt{\frac{\hbar\gamma B''}{M_s}}.$$
(B5)

We define $r_0 \equiv r_{0x} = r_{0y}$. It is useful to introduce the following Lamb-Dicke parameters that characterize the confinement of the particle in the Ioffe-Pritchard magnetic trap:

$$\eta = \frac{B' r_0}{2B_0},$$
(B6)

$$\eta' = \frac{B'' r_0^2}{4B_0}.$$
(B7)

Typically $\eta' \ll \eta \ll 1$. With such magnetic field, the Taylor expansion of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$ can be obtained by realizing that

$$\begin{aligned} n_z(\mathbf{r} = 0) &= 1, \\ \partial_x n_+|_{\mathbf{r}=0} &= \partial_x n_-|_{\mathbf{r}=0} = \frac{B'}{2B_0}, \\ \partial_y n_+|_{\mathbf{r}=0} &= -\partial_y n_-|_{\mathbf{r}=0} = -i \frac{B'}{2B_0}, \\ \partial_x^2 n_z|_{\mathbf{r}=0} &= \partial_y^2 n_z|_{\mathbf{r}=0} = -\left(\frac{B'}{2B_0}\right)^2, \\ \partial_{xz} n_+|_{\mathbf{r}=0} &= \partial_{xz} n_-|_{\mathbf{r}=0} = -\frac{B''}{4B_0}, \\ \partial_{yz} n_+|_{\mathbf{r}=0} &= -\partial_{yz} n_-|_{\mathbf{r}=0} = -i \frac{B''}{4B_0}. \end{aligned}$$
(B8)

The other components not shown are identically zero. To calculate the operators $n_3(\hat{\mathbf{r}}, \hat{\Omega})$, $n_\uparrow(\hat{\mathbf{r}}, \hat{\Omega})$, $n_\downarrow(\hat{\mathbf{r}}, \hat{\Omega})$, and their derivatives, we just need to substitute the results of Eq. (B8) into Eq. (27). The terms to zero order in Lamb-Dicke, $n_3(\hat{\Omega}) = n_3(0, \hat{\Omega})$, $n_\uparrow(\hat{\Omega}) = n_\uparrow(0, \hat{\Omega})$ and $n_\downarrow(\hat{\Omega}) = n_\downarrow(0, \hat{\Omega})$, read

$$\begin{aligned} n_3(\hat{\Omega}) &= \hat{D}_{00}^1, \\ n_\uparrow(\hat{\Omega}) &= -\sqrt{2} \hat{D}_{01}^1, \\ n_\downarrow(\hat{\Omega}) &= (n_\uparrow(\hat{\Omega}))^\dagger. \end{aligned}$$
(B9)

The first derivative of the components of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$ evaluated in $\mathbf{r} = 0$, i.e., the operators $\partial_\nu n_3(\hat{\Omega}) = \partial_\nu n_3(\hat{\mathbf{r}}, \hat{\Omega})|_{\mathbf{r}=0}$, $\partial_\nu n_\uparrow(\hat{\Omega}) = \partial_\nu n_\uparrow(\hat{\mathbf{r}}, \hat{\Omega})|_{\mathbf{r}=0}$, and $\partial_\nu n_\downarrow(\hat{\Omega}) = \partial_\nu n_\downarrow(\hat{\mathbf{r}}, \hat{\Omega})|_{\mathbf{r}=0}$, read

$$\begin{aligned} \partial_x n_3(\hat{\Omega}) &= -\frac{B'}{2\sqrt{2}B_0} (\hat{D}_{10}^1 - \hat{D}_{-10}^1), \\ \partial_x n_\uparrow(\hat{\Omega}) &= -\frac{B'}{2B_0} (\hat{D}_{-11}^1 - \hat{D}_{11}^1), \\ \partial_x n_\downarrow(\hat{\Omega}) &= -\frac{B'}{2B_0} (\hat{D}_{1-1}^1 - \hat{D}_{-1-1}^1), \end{aligned}$$
(B10)

for $\nu = x$, and

$$\begin{aligned} \partial_y n_3(\hat{\Omega}) &= -i \frac{B'}{2\sqrt{2}B_0} (\hat{D}_{10}^1 - \hat{D}_{-10}^1), \\ \partial_y n_\uparrow(\hat{\Omega}) &= i \frac{B'}{2B_0} (\hat{D}_{-11}^1 - \hat{D}_{11}^1), \\ \partial_y n_\downarrow(\hat{\Omega}) &= i \frac{B'}{2B_0} (\hat{D}_{1-1}^1 - \hat{D}_{-1-1}^1), \end{aligned}$$
(B11)

for $\nu = y$. The operators for $\nu = z$ are zero due to the axial symmetry of the magnetic field. The second derivatives of the components of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$ are obtained from Eq. (B8) as follows:

$$\begin{pmatrix} \partial_{\nu\mu} n_3(\hat{\Omega}) \\ \partial_{\nu\mu} n_\uparrow(\hat{\Omega}) \\ \partial_{\nu\mu} n_\downarrow(\hat{\Omega}) \end{pmatrix} = T(\hat{\Omega}) \begin{pmatrix} \partial_{\nu\mu} n_z \\ \partial_{\nu\mu} n_+ \\ \partial_{\nu\mu} n_- \end{pmatrix},$$
(B12)

where all the derivatives are evaluated in $\mathbf{r} = 0$.

APPENDIX C: BOSONIZATION OF \hat{V}_D

This section provides more details on the bosonization of $\hat{V}_D = \hat{V}_D^G + \hat{V}_D^{NG}$ into quadratic and nonquadratic terms assuming a the Ioffe-Pritchards field Eq. (B1).

To obtain \hat{V}_D^G , the procedure is the following. (1) Apply the Lamb-Dicke expansion on the components of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$ up to second order in $\hat{\mathbf{r}}$. (2) Substitute it into Eq. (22) and expand the square keeping only terms up to second order in $\hat{\mathbf{r}}$. After this procedure, the anisotropy energy reads $\hat{V}_D = \hat{V}_{D0} + \hat{V}_{D1} + \hat{V}_{D2}$, where \hat{V}_{Di} contains terms of order i in $\hat{\mathbf{r}}$. (3) Substitute the bosonic expressions for the spin angular momentum operators, Eq. (51), and for the D matrix, Table II. (4) Rearrange the operators in normal order and keep the terms that contain only up to a product of two bosons.

Following this procedure one can derive all the quadratic terms that appear in $\hat{V}_D^G = \hat{V}_{D0}^G + \hat{V}_{D1}^G + \hat{V}_{D2}^G$. Note that some of this quadratic terms appear from the normal ordering of nonlinear terms, and will thus lead to subleading contribution. Here, we report only the quadratic terms up to order $O(\hbar\omega_D\eta)$. (1) \hat{V}_{D0}^G collects the zero-order terms in the Lamb-Dicke expansion of $\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})$. It does not contain the center-of-mass bosons, and couples only the spin with the total angular momentum. It reads

$$\begin{aligned} \hat{V}_{D0}^G &= 32\hbar\omega_D \frac{S}{J} \hat{j}^\dagger \hat{j} - \frac{56}{5} \frac{S}{J} \hbar\omega_D (\hat{j}^\dagger + \hat{j})^2 \\ &+ 8\hbar\omega_D \frac{S}{J} (\hat{k}^\dagger \hat{k} + \hat{m}^\dagger \hat{m} - \hat{k}^\dagger \hat{m}^\dagger - \hat{k} \hat{m}) \\ &+ 4\hbar\omega_D \sqrt{\frac{S}{J}} [\hat{s}^\dagger (\hat{k}^\dagger - \hat{m}) + \text{H.c.}] + O\left(\frac{\hbar\omega_D}{J}\right). \end{aligned}$$
(C1)

(2) \hat{V}_{D1}^G collects the first-order Lamb-Dicke expansion and couples the center of mass motion with the total angular momentum $\hat{\mathbf{J}}$. It reads

$$\hat{V}_{D1}^G = 8\hbar\omega_D \eta \sqrt{\frac{S}{J}} [(\hat{b}_r^\dagger + \hat{b}_l)(\hat{m}^\dagger - \hat{k}) + \text{H.c.}] + O\left(\frac{\hbar\omega_D}{\sqrt{SJ}}\eta\right).$$
(C2)

(3) \hat{V}_{D2}^G collects the second-order expansion in Lamb-Dicke. It is a quadratic term in only the center-of-mass bosons but it is subleading, since $\hat{V}_{D2}^G \simeq O(\hbar\omega_T\eta^2)$.

The procedure to collect the nonlinear contributions is analogous, but the expansions in steps 2 and 3 are carried up to the order of interest. Following this approach, the strongest nonlinear term in \hat{V}_D^{NG} is a three-boson term that scales as $O(\hbar\omega_D\eta'/\sqrt{J})$.

APPENDIX D: BOSONIZATION OF \hat{V}_P

This section provides more details on the bosonization of $\hat{V}_P = \hat{V}_{P1} + \hat{V}_{P2}$ in Eq. (23), assuming the Ioffe-Pritchard field, where

$$\hat{V}_{P1} = \frac{\mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega}) \cdot \hat{\mathbf{p}} + \hat{\mathbf{p}} \cdot \mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega})}{2M}, \quad (\text{D1})$$

is the linear term in $\mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega})$ and

$$\hat{V}_{P2} = \frac{\mathbf{A}^2(\hat{\mathbf{r}}, \hat{\Omega})}{2M}, \quad (\text{D2})$$

is the quadratic term in $\mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega})$. Recall that

$$A_v(\hat{\mathbf{r}}, \hat{\Omega}) = -2\hbar[\mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega}) \times \partial_v \mathbf{n}(\hat{\mathbf{r}}, \hat{\Omega})] \cdot \hat{\mathbf{S}}. \quad (\text{D3})$$

We find the contribution to the final bosonic Hamiltonian \hat{H}_G and estimate the strongest nonlinear term in \hat{H}_{NG} .

The procedure to obtain the quadratic terms $\hat{V}_P^G = \hat{V}_{P1}^G + \hat{V}_{P2}^G$ is the following. (1) Apply the Lamb-Dicke expansion up to second order in $\hat{\mathbf{r}}$:

$$A_v(\hat{\mathbf{r}}, \hat{\Omega}) \simeq A_v + \sum_{\mu} \partial_{\mu} A_v \hat{r}_{\mu} + \frac{1}{2} \sum_{\mu\lambda} \partial_{\mu\lambda} A_v \hat{r}_{\mu} \hat{r}_{\lambda} + O(\mathbf{r}^3), \quad (\text{D4})$$

where

$$\begin{aligned} A_v &= -2\hbar(\mathbf{n} \times \partial_v \mathbf{n}) \cdot \hat{\mathbf{S}} \\ \partial_{\mu} A_v &= -2\hbar[(\mathbf{n} \times \partial_{v\mu} \mathbf{n}), + (\partial_{\mu} \mathbf{n} \times \partial_v \mathbf{n})] \cdot \hat{\mathbf{S}}, \\ \partial_{\mu\lambda} A_v &= -\hbar[(\mathbf{n} \times \partial_{v\mu\lambda} \mathbf{n}) + 2(\partial_{\mu} \mathbf{n} \times \partial_{v\lambda} \mathbf{n}) \\ &\quad + (\partial_{\mu\lambda} \mathbf{n} \times \partial_v \mathbf{n})] \cdot \hat{\mathbf{S}}. \end{aligned} \quad (\text{D5})$$

We define $\mathbf{n} \equiv \mathbf{n}(0, \hat{\Omega})$, $\partial_v \mathbf{n} \equiv \partial_v \mathbf{n}(\mathbf{r}, \hat{\Omega})|_{\mathbf{r}=0}$, $\partial_{v\mu} \mathbf{n} \equiv \partial_{v\mu} \mathbf{n}(\mathbf{r}, \hat{\Omega})|_{\mathbf{r}=0}$, $\partial_{v\mu\lambda} \mathbf{n} \equiv \partial_{v\mu\lambda} \mathbf{n}(\mathbf{r}, \hat{\Omega})|_{\mathbf{r}=0}$, and $r_{\mu} = x, y, z$ when $\mu = x, y, z$ respectively. (2) Substitute Eq. (D4) into Eqs. (D1) and (D2) keeping only terms up to second order in $\hat{\mathbf{r}}$. (3) Use Eqs. (B9)–(B12) to express $\mathbf{n}, \partial_v \mathbf{n}, \partial_{v\mu} \mathbf{n}$, and $\partial_{v\mu\lambda} \mathbf{n}$ in terms of D matrices, the components of $\mathbf{n}(\hat{\mathbf{r}})$, and its derivatives, Eq. (B8). (4) Take the expression for \hat{V}_{P1} and \hat{V}_{P2} obtained in the preceding step and substitute the bosonic expression for the angular momentum $\hat{\mathbf{S}}$, Eq. (51), and for the D matrix, Table II. (5) Rearrange the bosonic operators in normal order and keep only terms that contain a product of up to two bosonic operators.

The procedure allows to exactly calculate all the quadratic terms appearing in $\hat{V}_P^G = \hat{V}_{P1}^G + \hat{V}_{P2}^G$. Here, we will report terms up to $O(\hbar\omega_T\eta)$. The linear terms in \hat{V}_P^G read

$$\hat{V}_{P1}^G = 2\hbar\omega_T\eta\sqrt{\frac{S}{J}}[(\hat{m}^{\dagger} - \hat{k})(\hat{b}_r^{\dagger} - \hat{b}_l) + \text{H.c.}] + O(\hbar\omega_T\eta^2). \quad (\text{D6})$$

The contribution from the quadratic term in $\mathbf{A}(\hat{\mathbf{r}}, \hat{\Omega})$ is of order $\hat{V}_{P2}^G \simeq O(\hbar\omega_T\eta^2)$, and therefore can be neglected.

To evaluate the nonlinear terms, one proceeds as for the case of the quadratic terms, however, steps 1–3 must be done keeping all the bosonic terms up to the order of interest. Following this approach, one finds that the strongest nonlinear term in \hat{V}_P^{NG} is a three-bosons term that scales as $O(\hbar\omega_T\eta'/\sqrt{J})$.

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