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Angular momentum in spin-phonon processes

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Quantum theory of spin relaxation in the elastic environment is revised with account of the concept of a phonon spin recently introduced by Zhang and Niu [L. Zhang and Q. Niu, Phys. Rev. Lett. **112**, 085503 (2014)]. Similar to the case of the electromagnetic field, the division of the angular momentum associated with elastic deformations into the orbital part and the part due to phonon spins proves to be useful for the analysis of the balance of the angular momentum. Such analysis sheds important light on microscopic processes leading to the Einstein–de Haas effect.

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I. INTRODUCTION

The problem of conservation of angular momentum in systems containing magnetic moments has been around since the discovery a century ago of Einstein–de Haas [1] and Barnett [2] effects. The first effect demonstrated that the change in the magnetic moment of a freely suspended body generates mechanical rotation, while the second demonstrated that mechanical rotation induces magnetization. For some time, the Einstein–de Haas and Barnett effects were used to measure the gyromagnetic ratio of solids [3]. The significance of such measurements was diminished by the discovery of the electron-spin resonance and the ferromagnetic resonance that provided a more accurate determination of the gyromagnetic ratio. After that, the experiments on macroscopic magnetomechanical gyroscopic effects were largely abandoned. Surprisingly, however, microscopic mechanisms of the transfer of the spin angular momentum to the phonon system, and subsequently to the body as a whole, remain poorly understood even for a single spin in a crystal.

The tradition that goes back to the pioneering work on spin-phonon relaxation by Van Vleck [4] consists of ignoring conservation of angular momentum, with the excuse that the Hamiltonian of the system does not possess full rotational invariance. It is clear, however, that in theory (and in experiment) the angular momentum in a system of interacting spins and phonons is conserved. This has prompted a significant effort by a number of researchers to formulate the theory of magnetoelastic interactions in a rotationally invariant manner [5–11]. The advantage of such approach is that it is parameter free in the sense that spin-phonon rates can be expressed in terms of the well-known independently measured parameters.

The emergence of micro- and nanoelectromechanical devices (MEMS and NEMS) rejuvinated interest in the problem of angular momentum in magnetomechanical systems [12]. The Einstein–de Haas effect at the nanoscale has been experimentally studied in magnetic microcantilevers [13,14] and theoretically explained by the motion of domain walls [15]. The switching of magnetic moments by mechanical torques in nanocantilevers has been proposed [16–18]. Mechanical resonators containing single magnetic molecules have been studied by quantum methods [19–23]. Experiments have progressed to the measurement of the angular momentum exchange between a single molecular spin and a carbon nanotube [24,25].

In nanoresonators, the problem is somewhat simpler due to the finite number of resonant modes. For a single spin in a macroscopic body, however, the number of phonon degrees of freedom is practically infinite. In relation to the angular momentum, this problem has received significant recent attention in experiments with atomic spin-based qubits [26,27] and in application to spintronics [28]. To address this problem, Zhang and Niu recently introduced the concept of the phonon spin [29].

In this paper, we investigate this concept for the process of the relaxation of a single atomic spin in a macroscopic body. By developing an approach similar to that for photons, we find that within the elastic theory, the angular momentum of phonons can be naturally split into the orbital angular momentum $\mathbf{L}^{(1)}$ and the spin angular momentum $\mathbf{L}^{(2)}$. The orbital part corresponds to the rotation of the elastic medium around a certain point, while the spin part corresponds to small-radius circular shear displacements of points of the elastic media around their equilibrium positions; see Fig. 1.

The paper is structured as follows. The concept of the angular momentum in classical and quantum theories of elasticity is discussed in Sec. II. Conservation of the total angular momentum is studied in Sec. III by computing its commutator with the Hamiltonian. Quantum dynamics of the angular momentum of the relaxing spin and emitted phonons is investigated in Sec. IV. Section V contains a summary of the results and some final comments.

II. THE ANGULAR MOMENTUM

A. Angular momentum in the classical theory of elasticity

The angular momentum of the elastic solid is defined as

$$\mathbf{L} = \int d^3r(\mathbf{r} + \mathbf{u}) \times \mathbf{p}, \tag{1}$$

where time-independent $\mathbf{r}$ corresponds to the nondeformed body, $\mathbf{u}(\mathbf{r},t)$ is deformation, and $\mathbf{p}(\mathbf{r},t) = \rho\dot{\mathbf{u}}(\mathbf{r},t)$ is the momentum density. It consists of two parts,

$$\mathbf{L} = \mathbf{L}^{(1)} + \mathbf{L}^{(2)}, \tag{2}$$

where

$$\mathbf{L}^{(1)} = \int d^3r\rho\, \mathbf{r} \times \dot{\mathbf{u}}, \quad \mathbf{L}^{(2)} = \int d^3r\rho\, \mathbf{u} \times \dot{\mathbf{u}}. \tag{3}$$

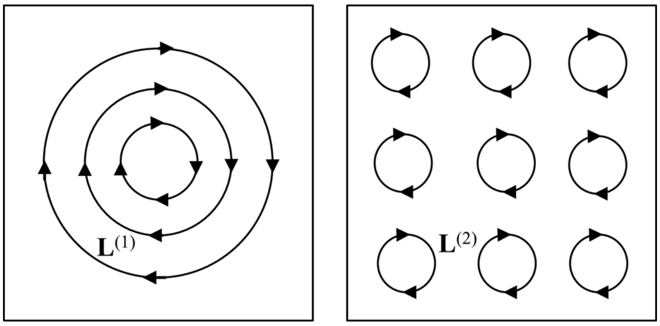

FIG. 1. Conceptual representation of the motion of the elastic medium that generates the orbital angular momentum $\mathbf{L}^{(1)}$ and the phonon-spin angular momentum $\mathbf{L}^{(2)}$.

The orbital part described by $\mathbf{L}^{(1)}$ corresponds to the rotation of the elastic medium around the origin, while the spin part described by $\mathbf{L}^{(2)}$ corresponds to small-radius circular shear displacements of points of the elastic media around their equilibrium positions; see Fig. 1.

Applying a time derivative to these expressions, one obtains

$$\dot{\mathbf{L}}^{(1)} = \int d^3r \rho\, \mathbf{r}\times\ddot{\mathbf{u}}, \quad \dot{\mathbf{L}}^{(2)} = \int d^3r \rho\, \mathbf{u}\times\ddot{\mathbf{u}}. \tag{4}$$

The dynamical equation for the displacement field is the Newton's equation,

$$\rho\frac{\partial^2 u_\alpha}{\partial t^2} = \frac{\partial\sigma_{\alpha\beta}}{\partial r_\beta}, \tag{5}$$

with the force on the right-hand side being a gradient of the stress tensor $\sigma_{\alpha\beta} = \delta\mathcal{H}/\delta e_{\alpha\beta}$. Here, $\mathcal{H}$ is the Hamiltonian density of the system and $e_{\alpha\beta} = \partial u_\alpha/\partial r_\beta$ is the strain tensor. After integrating by parts in Eqs. (4) and assuming zero elastic stress at the boundary of the body, one obtains

$$\dot{L}^{(1)}_\alpha = -\int d^3r \epsilon_{\alpha\beta\gamma}\sigma_{\gamma\beta}, \quad \dot{L}^{(2)}_\alpha = -\int d^3r \epsilon_{\alpha\beta\gamma} e_{\beta\delta}\sigma_{\gamma\delta}. \tag{6}$$

Within the linear elastic theory in the absence of internal torques (ignored by the conventional theory of elasticity [30]), the stress tensor $\sigma_{\alpha\beta}$ is symmetric, and thus $\dot{\mathbf{L}}^{(1)}$ is zero. However, $\dot{\mathbf{L}}^{(2)}$ is not vanishing in this approximation, and thus $\dot{\mathbf{L}} = 0$, expected on physical grounds, is not fulfilled.

To prove $\dot{\mathbf{L}} = 0$ for elastic systems, one has to take into account the intrinsic anharmonicity of the elastic theory due to the nonlinearity of the strain tensor [30],

$$u_{\rho\eta} = \tfrac{1}{2}(e_{\rho\eta} + e_{\eta\rho} + e_{\nu\rho}e_{\nu\eta}). \tag{7}$$

The fact that $\mathcal{H}$ must depend on $u_{\rho\eta}$ leads to

$$\sigma_{\gamma\delta} = \frac{\delta\mathcal{H}}{\delta e_{\gamma\delta}} = \frac{\delta u_{\rho\eta}}{\delta e_{\gamma\delta}}\frac{\delta\mathcal{H}}{\delta u_{\rho\eta}} = \frac{\delta\mathcal{H}}{\delta u_{\gamma\delta}} + e_{\gamma\rho}\frac{\delta\mathcal{H}}{\delta u_{\rho\delta}}, \tag{8}$$

which is nonsymmetric. Substituting this into Eq. (6), one can prove $\dot{\mathbf{L}} = \dot{\mathbf{L}}^{(1)} + \dot{\mathbf{L}}^{(2)} = 0$.

B. Spins as a source of internal torques

Anharmonicity, however, is not the only reason for $\sigma_{\alpha\beta}$ to be nonsymmetric. It also happens in the presence of spins because spin dynamics generates internal torques. Consider, e.g., a uniaxial spin Hamiltonian of the form

$$\hat{H}_S = -D(\mathbf{n}\cdot\mathbf{S})^2, \tag{9}$$

with $\mathbf{n}$ being the magnetic anisotropy axis. The corresponding Hamiltonian density is $\mathcal{H}_S = \hat{H}_S\delta(\mathbf{r})$. Elastic deformations of the body rotate the anisotropy axis $\mathbf{n}$ by a small angle $\boldsymbol{\phi}$,

$$\boldsymbol{\phi} = \tfrac{1}{2}\nabla\times\mathbf{u}(\mathbf{r}), \quad \phi_\alpha = \tfrac{1}{2}\epsilon_{\alpha\beta\gamma}e_{\gamma\beta}. \tag{10}$$

To the first order in $\boldsymbol{\phi}$, one has $\mathbf{n} = \mathbf{e}_z + [\boldsymbol{\phi}\times\mathbf{e}_z]$. Expanding $\hat{H}_S$ up to the linear terms in ϕ, we get $\hat{H}_S = \hat{H}_A + \hat{H}_{\text{s-ph}}$, where $\hat{H}_A = -DS_z^2$ and the spin-lattice coupling is given by [31]

$$\hat{H}_{\text{s-ph}} = -D(S_xS_z + S_zS_x)\phi_y + D(S_yS_z + S_zS_y)\phi_x. \tag{11}$$

The corresponding stress tensor $\sigma_{\alpha\beta} = \delta\mathcal{H}_{\text{s-ph}}/\delta e_{\alpha\beta}$ is nonsymmetric. Writing it as

$$\sigma_{\alpha\beta} = \frac{\delta\mathcal{H}_{\text{s-ph}}}{\delta e_{\alpha\beta}} = \frac{\delta\mathcal{H}_{\text{s-ph}}}{\delta\phi_\gamma}\frac{\delta\phi_\gamma}{\delta e_{\alpha\beta}} = \frac{1}{2}\frac{\delta\mathcal{H}_{\text{s-ph}}}{\delta\phi_\gamma}\epsilon_{\gamma\alpha\beta} \tag{12}$$

and using $\epsilon_{\alpha\beta\gamma}\epsilon_{\delta\beta\gamma} = 2\delta_{\alpha\delta}$, in Eq. (6), one obtains

$$\dot{\mathbf{L}}^{(1)} = -\int d^3r\frac{\delta\mathcal{H}_{\text{s-ph}}}{\delta\boldsymbol{\phi}} = -\frac{\partial\hat{H}_{\text{s-ph}}}{\partial\boldsymbol{\phi}}. \tag{13}$$

This explicitly expresses the internal mechanical torque in terms of rotation of the lattice and the spin with respect to each other in the presence of spin-lattice coupling. In what follows, we will show that $\mathbf{L}^{(2)}$ associated with the phonon spin is also generated in the problem of relaxation of the atomic spin, although we could not obtain for $\mathbf{L}^{(2)}$ a simple formula as for $\mathbf{L}^{(1)}$ above. The phonon-spin angular momentum $\mathbf{L}^{(2)}$ turns out to be important for the conservation of the total angular momentum, even in cases when the problem is solved with harmonic noninteracting phonons.

C. Quantum theory of phonon angular momentum

To obtain the second-quantized expression for the angular momentum, we use canonical quantization of phonons,

$$\mathbf{u}(\mathbf{r}) = \sqrt{\frac{\hbar}{2\rho V}}\sum_{\mathbf{k}\lambda}\frac{\mathbf{e}_{\mathbf{k}\lambda}e^{i\mathbf{k}\cdot\mathbf{r}}}{\sqrt{\omega_{\mathbf{k}\lambda}}}a_{\mathbf{k}\lambda} + \text{H.c.}, \tag{14}$$

where ρ is the mass density, V is the volume, $\mathbf{e}_{\mathbf{k}\lambda}$ are polarization vectors, $\omega_{\mathbf{k}\lambda}$ are phonon frequencies, and $a^\dagger$,a are creation and annihilation operators of phonons. One uses Eq. (14) as well as

$$\mathbf{p}(\mathbf{r}) = \rho\dot{\mathbf{u}}(\mathbf{r}) = -i\sqrt{\frac{\rho\hbar}{2V}}\sum_{\mathbf{k}\lambda}\mathbf{e}_{\mathbf{k}\lambda}\sqrt{\omega_{\mathbf{k}\lambda}}e^{i\mathbf{k}\cdot\mathbf{r}}a_{\mathbf{k}\lambda} + \text{H.c.} \tag{15}$$

The angular momentum of the body, given by Eq. (1), consists of two contributions, $\hat{\mathbf{L}} = \hat{\mathbf{L}}^{(1)} + \hat{\mathbf{L}}^{(2)}$, that have been discussed earlier. Here, $\hat{\mathbf{L}}^{(1)}$ is first order in phonon operators and it can be interpreted as the orbital angular momentum of the phonons. The term $\hat{\mathbf{L}}^{(2)}$ is second order in phonon operators and it can be interpreted as the spin of the phonons. Splitting the angular momentum into two parts is similar to that of photons. It will be shown below that the spin of a phonon is $\hbar$ and the phonon-spin eigenstates are circularly polarized phonons.

The operator of the orbital angular momentum becomes

$$\hat{\mathbf{L}}^{(1)} = \sqrt{\frac{\rho\hbar}{2V}} \sum_{\mathbf{k}\lambda} \sqrt{\omega_{\mathbf{k}\lambda}} [\mathbf{e}_{\mathbf{k}\lambda} \times \mathbf{j}_{\mathbf{k}}] a_{\mathbf{k}\lambda} + \text{H.c.}, \tag{16}$$

where $\mathbf{j}_{\mathbf{k}} \equiv i \int d^3r\, \mathbf{r} e^{i\mathbf{k}\cdot\mathbf{r}}$. As, by symmetry, $\mathbf{j}_{\mathbf{k}}$ can only be directed along $\mathbf{k}$, only transverse phonons contribute into $\hat{\mathbf{L}}^{(1)}$. In an infinite body, wave vectors are continuous, so that one can replace summation by integration,

$$\frac{1}{V}\sum_{\mathbf{k}} \dots \Rightarrow \int \frac{d^3k}{(2\pi)^3} \dots. \tag{17}$$

Then one can express $\mathbf{j}_{\mathbf{k}}$ as

$$\mathbf{j}_{\mathbf{k}} = (2\pi)^3 \partial_{\mathbf{k}} \delta(\mathbf{k}). \tag{18}$$

Dropping the terms aa and $a^\dagger a^\dagger$ in $\hat{\mathbf{L}}^{(2)}$ that do not conserve the number of phonon excitations, one obtains, after integration over the volume,

$$\hat{\mathbf{L}}^{(2)} = \frac{i\hbar}{2} \sum_{\mathbf{k}\lambda\lambda'} [\mathbf{e}_{\mathbf{k}\lambda} \times \mathbf{e}_{\mathbf{k}\lambda'}] a_{\mathbf{k}\lambda} a^\dagger_{\mathbf{k}'\lambda'} + \text{H.c.} \tag{19}$$

Keeping only transverse phonons, $\lambda\lambda' = 1,2$, and using $[\mathbf{e}_{\mathbf{k}1} \times \mathbf{e}_{\mathbf{k}2}] = \mathbf{k}/k$, one arrives at

$$\hat{\mathbf{L}}^{(2)} = i\hbar \sum_{\mathbf{k}} \frac{\mathbf{k}}{k} (a^\dagger_{\mathbf{k}2} a_{\mathbf{k}1} - a^\dagger_{\mathbf{k}1} a_{\mathbf{k}2}). \tag{20}$$

This operator becomes diagonal in terms of numbers of circularly polarized phonons $a_{\mathbf{k}\pm} \equiv (a_{\mathbf{k}1} \pm i a_{\mathbf{k}2})/\sqrt{2}$,

$$\hat{\mathbf{L}}^{(2)} = \hbar \sum_{\mathbf{k}} \frac{\mathbf{k}}{k} (-a^\dagger_{\mathbf{k}+} a_{\mathbf{k}+} + a^\dagger_{\mathbf{k}-} a_{\mathbf{k}-}). \tag{21}$$

Each such phonon carries an angular momentum $\hbar$ parallel or antiparallel to its wave vector that can be interpreted as the spin of the phonon.

III. CONSERVATION OF ANGULAR MOMENTUM

Let us now check conservation of the total angular momentum,

$$\mathbf{J} = \mathbf{L} + \hbar\mathbf{S}, \tag{22}$$

that implies that $\mathbf{J}$ must commute with the Hamiltonian. The dynamical change of the spin operator has to be absorbed by the angular momentum of the elastic matrix, whose evolution is given by

$$\dot{\hat{\mathbf{L}}} = \frac{i}{\hbar} [\hat{H}_{\text{s-ph}}, \hat{\mathbf{L}}]. \tag{23}$$

In particular, the precession of the spin around the anisotropy axis creates the cowiggling of the elastic matrix with the spin.

It turns out that by commuting operators, one can prove conservation of some parts of the angular momentum, whereas the complete proof of conservation requires a full quantum-mechanical solution for the relaxing spin and phonons created by its precession, presented in the next section. The situation is different for the angular momentum components perpendicular and parallel to the anisotropy axis.

We will study the spin-lattice model introduced above quantum mechanically ($\mathcal{H} \Rightarrow \hat{H}$). Introducing spin operators $S_\pm \equiv S_x \pm i S_y$ that follow commutation relations $[S_\pm, S_z] = \pm S_\pm$, one obtains, from Eq. (11),

$$\hat{H}_{\text{s-ph}} = -\frac{iD}{2} (S_+ S_z + S_z S_+) \phi_- + \text{H.c.}, \tag{24}$$

where $\phi_\pm \equiv \phi_x \pm i\phi_y$. Using Eqs. (10) and (14) with the atomic spin located at $\mathbf{r} = 0$, one obtains

$$\phi_\pm = \frac{1}{2} \sqrt{\frac{\hbar}{2\rho V}} \sum_{\mathbf{k}\lambda} \frac{\mathbf{e}_\pm \cdot [i\mathbf{k} \times \mathbf{e}_{\mathbf{k}\lambda}]}{\sqrt{\omega_{\mathbf{k}\lambda}}} (a_{\mathbf{k}\lambda} - a^\dagger_{\mathbf{k}\lambda}), \tag{25}$$

where $\mathbf{e}_\pm \equiv \mathbf{e}_x \pm i\mathbf{e}_y$.

We will need commutators

$$[\phi_\pm, \hat{\mathbf{L}}^{(1)}] = i \frac{\hbar}{2V} \sum_{\mathbf{k}\lambda} (\mathbf{e}_\pm \cdot [\mathbf{k} \times \mathbf{e}_{\mathbf{k}\lambda}]) [\mathbf{e}_{\mathbf{k}\lambda} \times \mathbf{j}_{\mathbf{k}}] \tag{26}$$

and

$$[\phi_\pm, \hat{\mathbf{L}}^{(2)}] = \frac{\hbar}{2} \sqrt{\frac{\hbar}{2\rho V}} \sum_{\mathbf{k}\lambda} \frac{\mathbf{k}}{\sqrt{\omega_{\mathbf{k}\lambda}}} (\mathbf{e}_\pm \cdot \mathbf{e}_{\mathbf{k}\lambda}) (a_{\mathbf{k}\lambda} - a^\dagger_{\mathbf{k}\lambda}) \tag{27}$$

that follow from Eqs. (25), (16), and (20).

Let us first consider dynamics of the transverse components of the angular momentum. The dominant source of spin precession around the anisotropy axis is the unperturbed spin Hamiltonian $\hat{H}_A$:

$$\dot{S}_x = \frac{i}{\hbar} [\hat{H}_{\text{A}}, S_x] = -\frac{i}{\hbar} D \left[S_z^2, S_x\right] = \frac{D}{\hbar} (S_z S_y + S_y S_z). \tag{28}$$

For the matrix, let us first consider the dynamics of the phonon orbital angular momentum $\hat{\mathbf{L}}^{(1)}$. From Eq. (26), with the help of the identity

$$\sum_{\lambda=1,2} (\mathbf{e}_{\mathbf{k}\lambda} \cdot \mathbf{A})(\mathbf{e}_{\mathbf{k}\lambda} \cdot \mathbf{B}) = \mathbf{A} \cdot \mathbf{B} - \left(\frac{\mathbf{k}}{k} \cdot \mathbf{A}\right)\left(\frac{\mathbf{k}}{k} \cdot \mathbf{B}\right) \tag{29}$$

and Eq. (18), one obtains

$$\begin{aligned}
\left[\phi_\pm, \hat{L}_x^{(1)}\right] &= i \frac{\hbar}{2V} \sum_{\mathbf{k}\lambda} (\mathbf{e}_\pm \cdot [\mathbf{k} \times \mathbf{e}_{\mathbf{k}\lambda}])(\mathbf{e}_x \cdot [\mathbf{e}_{\mathbf{k}\lambda} \times \mathbf{j}_{\mathbf{k}}]) \\
&= i \frac{\hbar}{2V} \sum_{\mathbf{k}\lambda} (\mathbf{e}_{\mathbf{k}\lambda} \cdot [\mathbf{e}_\pm \times \mathbf{k}])(\mathbf{e}_{\mathbf{k}\lambda} \cdot [\mathbf{j}_{\mathbf{k}} \times \mathbf{e}_x]) \\
&= i \frac{\hbar}{2V} \sum_{\mathbf{k}} [\mathbf{e}_\pm \times \mathbf{k}] \cdot [\mathbf{j}_{\mathbf{k}} \times \mathbf{e}_x] \\
&= i \frac{\hbar}{2V} \sum_{\mathbf{k}} \{(\mathbf{e}_\pm \cdot \mathbf{j}_{\mathbf{k}})(\mathbf{e}_x \cdot \mathbf{k}) - (\mathbf{e}_\pm \cdot \mathbf{e}_x)(\mathbf{k} \cdot \mathbf{j}_{\mathbf{k}})\} \\
&= i \frac{\hbar}{2} \int d^3k \left\{k_x \partial_{k_x} \delta(\mathbf{k}) - (\mathbf{k} \cdot \partial_{\mathbf{k}} \delta(\mathbf{k}))\right\} \\
&= i\hbar.
\end{aligned} \tag{30}$$

Now from Eqs. (23) and (24), one obtains

$$\dot{\hat{L}}_x^{(1)} = -D(S_y S_z + S_z S_y). \tag{31}$$

Combining this with Eq. (28), one obtains the conservation law

$$\hbar \dot{S}_x + \dot{\hat{L}}_x^{(1)} = 0. \tag{32}$$

In the same way, one can obtain $\hbar \dot{S}_y + \dot{\hat{L}}_y^{(1)} = 0$.

However, Eq. (32) is not the whole story. One has to consider $\dot{\hat{L}}_{x,y}^{(2)}$ using Eqs. (27) and (24). The resulting expression is a sum over **k**, linear in phonon operators. It is of the same order as the contribution to $\hbar\dot{S}_{x,y}$ due to the spin-phonon interaction, $i[\hat{H}_{\text{s-ph}},S_{x,y}]$, that was ignored above. Both terms discussed here are much smaller than the dominant terms in the angular momentum, conserved according to Eq. (32). These small terms are related to the spin-lattice relaxation of the spin. It is impossible to prove conservation of these terms without performing the full quantum-mechanical solution of the problem of spin relaxation.

Considering dynamics of the longitudinal component of the angular momentum, one can prove

$$\dot{L}_z^{(1)} = \frac{i}{\hbar}\left[\hat{H}_{\text{s-ph}},L_z^{(1)}\right] = 0 \tag{33}$$

by a calculation similar to that in Eq. (30). The terms $\dot{S}_z$ and $\dot{L}_z^{(2)}$ are related to spin-lattice relaxation and they are sums over **k**, linear in phonon operators. However, one cannot prove

$$\hbar\dot{S}_z + \dot{L}_z^{(2)} = 0 \tag{34}$$

without the full solution of the quantum problem that will be presented below.

IV. QUANTUM THEORY OF THE RELAXING SPIN

This problem resembles the problem of the relaxation of the excited state of an atom accompanied by the radiation of a photon. The atom is characterized by the discrete energy levels, while the electromagnetic field has a continuum of quantized photon states of an arbitrary energy. In a similar manner, a spin in the uniaxial crystal field has discrete energy levels characterized by the magnetic quantum number m, while phonons have a continuum of states characterized by energies $\hbar\omega_{\mathbf{k}\lambda}$.

A. General solution

To facilitate solving the problem of spin-lattice relaxation, we reduce the spin-phonon Hamiltonian to the rotating-wave approximation (RWA) form that conserves the energy. Consider transitions of the spin $|m-1\rangle \to |m\rangle$ for $m>0$ decreasing its energy and call the spin states $|1\rangle$ and $|0\rangle$, respectively. With the help of Eq. (24), one obtains the spin matrix element of this transition,

$$\langle m-1|\hat{H}_{\text{s-ph}}|m\rangle = \frac{iD}{2}(2m-1)l_{m-1,m}\phi_+, \tag{35}$$

where $l_{m-1,m} \equiv \sqrt{S(S+1)-m(m-1)}$. The explicit form of ϕ_+ in terms of the phonon operators, given by Eq. (25), then yields the following operator of the RWA coupling:

$$\hat{V} = \sum_{\mathbf{k}\lambda}\left(A_{\mathbf{k}\lambda}^* X^{01} a_{\mathbf{k}\lambda}^\dagger + A_{\mathbf{k}\lambda} X^{10} a_{\mathbf{k}\lambda}\right), \tag{36}$$

where

$$A_{\mathbf{k}\lambda} \equiv -\frac{D}{4}(2m-1)l_{m-1,m}\sqrt{\frac{\hbar}{2\rho V}}\frac{\mathbf{e}_+\cdot[\mathbf{k}\times\mathbf{e}_{\mathbf{k}\lambda}]}{\sqrt{\omega_{\mathbf{k}\lambda}}}, \tag{37}$$

and the X operators are defined by

$$X^{01}|1\rangle = |0\rangle, \quad X^{10}|0\rangle = |1\rangle. \tag{38}$$

The quantum state of the system can be specified by

$$\Psi = \left(cX^{10} + \sum_{\mathbf{k}\lambda} c_{\mathbf{k}\lambda} a_{\mathbf{k}\lambda}^\dagger\right)|00\rangle, \tag{39}$$

where $|00\rangle$ is the "vacuum" state. Ψ has only one excitation, spin or phonon. Considering the excited state of the spin as the reference-energy state, one obtains the Schrödinger equation for the coefficients,

$$\begin{aligned}\dot{c} &= -\frac{i}{\hbar}\sum_{\mathbf{k}\lambda} A_{\mathbf{k}\lambda}c_{\mathbf{k}\lambda},\\ \dot{c}_{\mathbf{k}\lambda} &= -i(\omega_{\mathbf{k}\lambda}-\omega_0)c_{\mathbf{k}\lambda} - \frac{i}{\hbar}A_{\mathbf{k}\lambda}^* c,\end{aligned} \tag{40}$$

where $\omega_0 \equiv (E_1-E_0)/\hbar$ is the frequency of the transition between the spin levels.

One can integrate the equations for the phonon modes $c_{\mathbf{k}}$ assuming the initial condition of the phonon vacuum:

$$\begin{aligned}c_{\mathbf{k}\lambda}(t) &= -\frac{iA_{\mathbf{k}\lambda}^*}{\hbar}\int_0^t dt' e^{-i(\omega_{\mathbf{k}\lambda}-\omega_0)(t-t')}c(t')\\ &= -\frac{iA_{\mathbf{k}\lambda}^*}{\hbar}\int_0^t d\tau e^{-i(\omega_{\mathbf{k}\lambda}-\omega_0)\tau}c(t-\tau),\end{aligned} \tag{41}$$

and insert the result into the equation for the spin c:

$$\frac{dc}{dt} = -\frac{1}{\hbar^2}\sum_{\mathbf{k}\lambda}|A_{\mathbf{k}\lambda}|^2\int_0^t d\tau e^{-i(\omega_{\mathbf{k}\lambda}-\omega_0)\tau}c(t-\tau). \tag{42}$$

In this integro-differential equation, $c(t-\tau)$ is a slow function of time, whereas the memory function $f(\tau) = \sum_{\mathbf{k}\lambda}|A_{\mathbf{k}\lambda}|^2 e^{-i(\omega_{\mathbf{k}\lambda}-\omega_0)\tau}$ is sharply peaked at $\tau=0$. Thus one can replace $c(t-\tau) \Rightarrow c(t)$, after which integration over τ and keeping only real contribution responsible for the relaxation yields the equation

$$\frac{dc}{dt} = -\frac{\Gamma}{2}c, \tag{43}$$

and thus

$$c = e^{-(\Gamma/2)t}, \tag{44}$$

where

$$\Gamma = \frac{2\pi}{\hbar^2}\sum_{\mathbf{k}\lambda}|A_{\mathbf{k}\lambda}|^2\delta(\omega_{\mathbf{k}\lambda}-\omega_0) \tag{45}$$

is the spin-relaxation rate. Now, adopting this solution in Eq. (41) and integrating over time, one obtains, for the phonons,

$$c_{\mathbf{k}\lambda}(t) = \frac{A_{\mathbf{k}\lambda}^*}{\hbar}\frac{e^{-i(\omega_{\mathbf{k}\lambda}-\omega_0)t} - e^{-(\Gamma/2)t}}{\omega_{\mathbf{k}\lambda}-\omega_0+i\Gamma/2}. \tag{46}$$

B. Dynamics of the phonon-spin angular momentum

Let us now compute the phonon-spin angular momentum $L_z^{(2)}$ resulting from the relaxation of the spin. Remember that $L_z^{(1)} = 0$ according to Eq. (33). It is not necessary to use circularly polarized phonons: one can work with linearly polarized phonons using Eqs. (21) and (39). For the quantum expectation value, one obtains

$$\mathbf{L}^{(2)} = i\hbar \sum_{\mathbf{k}} \frac{\mathbf{k}}{k}(c^*_{\mathbf{k}2}c_{\mathbf{k}1} - c^*_{\mathbf{k}1}c_{\mathbf{k}2}). \quad (47)$$

Using Eq. (46) and setting $\omega_{\mathbf{k}\lambda} \Rightarrow \omega_{\mathbf{k}}$, one obtains

$$\mathbf{L}^{(2)} = \frac{i}{\hbar}\sum_{\mathbf{k}} \frac{\mathbf{k}}{k}(A_{\mathbf{k}2}A^*_{\mathbf{k}1} - A_{\mathbf{k}2}A^*_{\mathbf{k}1}) \times \frac{1 + e^{-\Gamma t} - (e^{-i(\omega_{\mathbf{k}}-\omega_0)t} + e^{i(\omega_{\mathbf{k}}-\omega_0)t})e^{-(\Gamma/2)t}}{(\omega_{\mathbf{k}}-\omega_0)^2 + \Gamma^2/4}. \quad (48)$$

In the integration over $\omega_{\mathbf{k}}$, one goes to the upper and lower complex half plane for the two different oscillating terms. As the result, one obtains

$$\mathbf{L}^{(2)} = \frac{2\pi}{\hbar\Gamma}(1 - e^{-\Gamma t})\sum_{\mathbf{k}} \frac{\mathbf{k}}{k}\delta(\omega_{\mathbf{k}} - \omega_0)(iA_{\mathbf{k}2}A^*_{\mathbf{k}1} + \text{H.c.}). \quad (49)$$

It remains to show that the integral over $\mathbf{k}$ in this expression can be expressed through Γ so that Γ cancels and the result simplifies. Indeed, the combination that enters Eq. (45) after simplifications becomes

$$|A_{\mathbf{k}1}|^2 + |A_{\mathbf{k}2}|^2 = D^2[(2m-1)l_{m-1,m}]^2 \frac{\hbar}{4\rho V}\frac{k_z^2}{\omega_{\mathbf{k}}}. \quad (50)$$

On the other hand, in Eq. (49), one obtains

$$iA_{\mathbf{k}2}A^*_{\mathbf{k}1} + \text{H.c.} = -D^2[(2m-1)l_{m-1,m}]^2 \frac{\hbar}{4\rho V}\frac{kk_z}{\omega_{\mathbf{k}}}. \quad (51)$$

Note that in Eq. (49), only the longitudinal component $L_z^{(2)}$ is nonzero by symmetry. The latter is just the negative of Eq. (50) that enters Γ, given by Eq. (45). Thus, in Eq. (49), Γ cancels out and one obtains the simple behavior

$$L_z = L_z^{(2)} = -(1 - e^{-\Gamma t})\hbar, \quad (52)$$

as the spin undergoes a relaxational transition $|m-1\rangle \to |m\rangle$. This means that the total angular momentum in the system spin + phonons is conserved.

V. DISCUSSION

We have analyzed the transfer of the angular momentum from the atomic spin to the orbital and spin angular momentum of phonons. These two parts of the angular momentum of the phonon system are clearly distinguishable. The orbital part is first order on the phonon operators. Its classical counterpart is the twist of the elastic matrix around the position of the atomic spin, which is linear on the displacement field. The spin part of the phonon angular momentum is second order on phonon operators. Its classical counterpart corresponds to the rotational shear deformations that are quadratic on the displacement field.

Conservation of the angular momentum in the process of the relaxation of the atomic spin has been demonstrated by us explicitly. It turns out that the change in the transverse part of the atomic spin is balanced by the orbital part of the phonon angular momentum, while the change in the relaxing longitudinal part of the atomic spin is balanced by the spin part of the phonon angular momentum. These findings can be useful in schemes where individual atomic spins (e.g., used as qubits) are manipulated by phonons.

The solution of the full quantum many-body problem of the angular momentum conservation in a system of many relaxing spins in the elastic environment is missing at this time. It should be solved first for an isotropic elastic environment with the spin-phonon interaction of the simplest form (11) for which the mechanism of the transfer of the angular momentum between the spin and the elastic environment is apparent. Spins coupled by exchange interaction in a ferromagnet represent a many-body extension of the single-spin model studied in this paper. This problem can be readily solved only for small deviations of the spin system from equilibrium, described in terms of magnons and their interaction with phonons. However, theoretical description of magnetization reversal and the ensuing transfer of the angular momentum is a complicated problem that is beyond the scope of this paper. The problem of many paramagnetic spins interacting with the lattice is also nontrivial due to coherent and incoherent collective effects such as super-radiance and phonon bottleneck. Solutions of the Schrödinger equation have been obtained before in the context of phonon-laser [32] and phonon bottleneck [33] effects, but the analysis of the angular momentum conservation for these problems is significantly more complicated.

Another outstanding problem, not addressed in this paper, is how the orbital and spin angular momenta carried by phonons get transferred to the rotation of the body as a whole in the Einstein–de Haas effect. To answer this question, one must recall that in a typical Einstein–de Haas experiment, one induces rotational oscillations of a macroscopic body by the low-frequency ac magnetic field. The corresponding time scales are much greater than lifetimes of phonons emitted in atomic spin transitions. Consequently, such phonons fully equilibrate on the time scale of the transfer of the angular momentum from atomic spins to the body as a whole.

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[1] A. Einstein and W. J. de Haas, Experimenteller Nachweis der Ampereschen Molekularströme, Verh. Dtsch. Phys. Ges. **17**, 152 (1915).

[2] S. J. Barnett, Magnetization by rotation, Phys. Rev. **6**, 239 (1915).

[3] S. J. Barnett and G. S. Kenny, Gyromagnetic ratios of iron, cobalt, and many binary alloys of iron, cobalt, and nickel, Phys. Rev. **87**, 723 (1952); G. G. Scott and H. W. Sturner, Magnetomechanical ratios for Fe-Co alloys, *ibid.* **184**, 490 (1969), and references therein.

[4] J. H. Van Vleck, Paramagnetic relaxation times for titanium and chrome alum, Phys. Rev. **57**, 426 (1940).
[5] R. L. Melcher, Rotationally invariant theory of spin-phonon interactions in paramagnets, Phys. Rev. Lett. **28**, 165 (1972); Elastic properties of a paramagnet: Application to $NdVO_4$, Phys. Rev. B **19**, 284 (1979).
[6] V. Dohm and P. Fulde, Magnetoelastic interaction in rare earth systems, Z. Phys. B **21**, 369 (1975).
[7] P. A. Fedders, Resonant and nonresonant effects of paramagentic spins on acoustic modes, Phys. Rev. B **12**, 2045 (1975).
[8] L. Bonsall and R. L. Melcher, Rotational invariance, finite strain theory, and spin-lattice interactions in paramagnets; application to the rare-earth vanadates, Phys. Rev. B **14**, 1128 (1976).
[9] P. A. Fedders and R. L. Melcher, Rotational pseudospin-phonon interactions at finite frequencies, Phys. Rev. B **14**, 1142 (1976).
[10] P. A. Fedders, Effects of self-consistency on spin-lattice relaxation, Phys. Rev. B **15**, 3297 (1977).
[11] E. M. Chudnovsky, D. A. Garanin, and R. Schilling, Universal mechanism of spin relaxation in solids, Phys. Rev. B **72**, 094426 (2005).
[12] See, e.g., E. M. Chudnovsky and D. A. Garanin, Damping of a nanocantilever by paramagnetic spins, Phys. Rev. B **89**, 174420 (2014), and references therein.
[13] T. M. Wallis, J. Moreland and P. Kabos, Einstein - de Haas effect in a NiFe film deposited on a microcantilever, Appl. Phys. Lett. **89**, 122502 (2006).
[14] S-H. Lim, A. Imtiaz, T. M. Wallis, S. Russek, P. Kabos, L. Cai, and E. M. Chudnovsky, Magneto-mechanical investigation of spin dynamics in magnetic multilayers, Europhys. Lett. **105**, 37009 (2014).
[15] R. Jaafar, E. M. Chudnovsky, and D. A. Garanin, Dynamics of Einstein - de Haas effect: Application to magnetic cantilever, Phys. Rev. B **79**, 104410 (2009).
[16] A. A. Kovalev, G. E. W. Bauer, and A. Brataas, Nanomechanical magnetization reversal, Phys. Rev. Lett. **94**, 167201 (2005).
[17] L. Cai, R. Jaafar, and E. M. Chudnovsky, Mechanically assisted current-induced switching of the magnetic moment in a torsional oscillator, Phys. Rev. Appl. **1**, 054001 (2014).
[18] E. M. Chudnovsky and R. Jaafar, Electromechanical magnetization switching, J. Appl. Phys. **117**, 103910 (2015).
[19] R. Jaafar and E. M. Chudnovsky, Magnetic molecule on a microcantilever, Phys. Rev. Lett. **102**, 227202 (2009).
[20] R. Jaafar, E. M. Chudnovsky, and D. A. Garanin, Single magnetic molecule between conducting leads: Effect of mechanical rotations, Europhys. Lett. **89**, 27001 (2010).
[21] A. A. Kovalev, L. X. Hayden, G. E. W. Bauer, and Y. Tserkovnyak, Macrospin tunneling and magnetopolaritons with nanomechanical interference, Phys. Rev. Lett. **106**, 147203 (2011).
[22] D. A. Garanin and E. M. Chudnovsky, Quantum entanglement of a tunneling spin with mechanical modes of a torsional resonator, Phys. Rev. X **1**, 011005 (2011).
[23] M. F. O'Keeffe, E. M. Chudnovsky, and D. A. Garanin, Landau-Zener dynamics of a nanoresonator containing a tunneling spin, Phys. Rev. B **87**, 174418 (2013).
[24] M. Ganzhorn, S. Klyatskaya, M. Ruben, and W. Wernsdorfer, Strong spin-phonon coupling between a single-molecule magnet and a carbon nanotube nanoelectromechanical system, Nat. Nanotechnol. **8**, 165 (2013).
[25] M. Ganzhorn, S. Klyatskaya, M. Ruben, and W. Wernsdorfer, Carbon nanotube nanoelectromechanical systems as magnetometers for single-molecule magnets, ACS Nano **7**, 6225 (2013).
[26] L. C. Bassett, F. J. Heremans, D. J. Christle, C. G. Yale, G. Burkard, B. B. Buckley, and D. D. Awschalom, Ultrafast optical control of orbital and spin dynamics in a solid-state defect, Science **345**, 1333 (2014).
[27] D. J. Christle, A. L. Falk, P. Andrich, P. V. Klimov, J. Hassan, N. T. Son, E. Janzén, T. Ohshima, and D. D. Awschalom, Nat. Mater. **14**, 160 (2015).
[28] T. Qin, J. Zhou, and J. Shi, Berry curvature and the phonon Hall effect, Phys. Rev. B **86**, 104305 (2012).
[29] L. Zhang and Q. Niu, Angular momentum of phonons and Einstein - de Haas effect, Phys. Rev. Lett. **112**, 085503 (2014).
[30] L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon, New York, 1970).
[31] D. A. Garanin and E. M. Chudnovsky, Thermally activated resonant magnetization tunneling in molecular magnets: $Mn_{12}Ac$ and others, Phys. Rev. B **56**, 11102 (1997).
[32] E. M. Chudnovsky and D. A. Garanin, Phonon superradaiance and phonon laser effect in nanomagnets, Phys. Rev. Lett. **93**, 257205 (2004).
[33] D. A. Garanin, Towards a microscopic understanding of the phonon bottleneck, Phys. Rev. B **75**, 094409 (2007); Phonon bottleneck in the low-excitation limit, *ibid.* **75**, 024429 (2007).