

Mechanical rotation of nanomagnet through interaction with an electromagnetic wave

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Abstract. We report a theoretical study on the effect of mechanical rotation of nanomagnet about a fixed axis due to its interaction with an electromagnetic wave with a frequency approaching that of ferromagnetic resonance. A stationary mode of rotation is investigated. We show that the rotation speed magnitude is proportional to the squared amplitude of the wave's magnetic field, being maximum for a circularly polarized wave when the rotation direction changes for the opposite following a likewise change in the direction of polarization. The role of the magnetic anisotropy of nanomagnet is discussed. The effect is numerically estimated for molecular nanomagnet Fe₈.

1 Introduction

The phenomena arising from interaction between spin and mechanical degrees of freedom have always attracted attention of both theoretical and experimental physicists. Of the longest and most widely known ones are the Einstein-de Haas [1,2] and Barnett [3] gyromagnetic effects observed in macroscopic samples (see also Ref. [4]). For a fairly long time now researchers have been conducting studies on small free magnetic clusters (Co_N, Fe_N, Gd_N) in a beam, that also feature strong interaction between spin and mechanical degrees of freedom (magnetic deflection experiments of Stern-Gerlach type) [5–10].

It is obvious that the physical mechanisms behind the Einstein-de Haas and Barnett effects are universal and, hence, should be at work in small-size (non-macroscopic) samples, as well. This fact is pointed out in references [11,12]. Of active research interest, therefore, are studies of the interaction between spin and mechanical rotational degrees of freedom in nanoparticles. Thus, in reference [13] the authors report observation of how free rotational movement of magnetic nanoparticles CoFe₂O₄ confined within a polymer cavity affects the frequency shift of a ferromagnetic resonance in these particles (see also Ref. [14]). Among the noteworthy theoretical works are researches addressing interaction between the spin and mechanical rotational degrees of freedom in molecular nanomagnets Mn₁₂ and Fe₈ [15,16] (see also Refs. [17,18]). It is shown therein that such interaction is related with the quantum tunnelling process. The results obtained in references [15–18] are important, since molecular nanomagnets are viewed currently as an essential structural element in various devices of molecular spintronics [19–23].

We should emphasize one important issue involved in the study of interaction between spin and mechanical rotational degrees of freedom: magnetic nanoparticles [13] and molecular nanomagnets [15,16] in particular can be regarded as promising candidates for magnetic nanorotors, as it is possible to control their rotation with a rotating magnetic field. Important data to this effect were obtained in the experimental study reference [24], reporting creation of molecular nanorotors on the basis of zinc phthalocyanine molecules on gold surface. The rotation axis of such nanorotors remains fixed, which is of critical importance. Although these devices are non magnetic, they can acquire magnetic properties through implanting magnetic atoms in them and thus function as magnetic nanorotors controllable with a rotating magnetic field.

So, the behavior of a magnetic nanorotor (specifically, molecular nanomagnet) in an alternating magnetic field is a problem of active research interest currently. In this paper we offer a theoretical study into the effect of molecular nanomagnet rotation about a fixed axis under an alternating magnetic field. Such a problem has never been tackled before. Moreover, we focus on the most promising situation, when interaction between the alternating field and molecular magnet is of a resonance character. The problem is solved in the framework of quantum mechanics. A dependence of the molecular nanomagnet rotation speed on the polarization and frequency of magnetic field is established. The effect is numerically estimated. In what follows we will, for brevity, use the term “nanomagnet”.

The paper is organized as follows.

In Section 2 we find the quantum mechanical states of the fundamental and first excited doublets of nanomagnet having several axes of magnetic anisotropy and one fixed axis of mechanical rotation.

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Section 3 deals with solution of the quantum mechanical equations of motion, describing the nanomagnet behavior in an alternating magnetic field of electromagnetic wave. The expression for the speed of mechanical rotation of nanomagnet is obtained.

In Section 4 the effect in question is evaluated.

Section 5 is a summary of the obtained results.

Finally, the Appendix deals with the problem on the degeneracy of the energy levels of a freely rotating nanomagnet.

2 Free rotating nanomagnet

Consider a nanomagnet with a constant spin $|\mathbf{S}| = S = \text{const.}$, having hard, medium and easy axes of magnetic anisotropy (a likely example is molecular cluster Fe_8 [16,25,26], where $S = 10$). When it does not move, its Hamiltonian can be written as:

$$\hat{H}_s = -D\hat{S}_z^2 - K\hat{S}_x^2, \quad (1)$$

yet, when such a nanomagnet starts rotating about axis $0Z$, its Hamiltonian takes the form:

$$\hat{H} = \frac{(\hbar\hat{L}_z)^2}{2I} - D\hat{S}_z^2 - K(\cos\varphi\hat{S}_x + \sin\varphi\hat{S}_y)^2. \quad (2)$$

Here x, y, z are the Cartesian coordinates in the laboratory frame, $\hat{S}_{x,y,z}$ are the operators of spin projections, D and K are the magnetic anisotropy constants for the easy and medium axes, respectively, φ is the angle of nanomagnet rotation about axis $0Z$, I is the nanomagnet moment of inertia with respect to the $0Z$ axis, $\hbar\hat{L}_z = -i\hbar\frac{\partial}{\partial\varphi}$ is the operator of the angular momentum with respect to axis $0Z$, that is associated with the mechanical rotation of nanomagnet. Rotating magnet is shown schematically in Figure 1.

We assume relation

$$D > K \gg \frac{\hbar^2}{I}, \quad (3)$$

to be valid for the constants entering in equation (2). With an accuracy to a non-essential constant member it is convenient to write Hamiltonian (2) in the form:

$$\begin{aligned} \hat{H} &= \hat{H}^{(0)} + \hat{H}_\perp, & \hat{H}^{(0)} &= \frac{(\hbar\hat{L}_z)^2}{2I} - \left(D - \frac{K}{2}\right)\hat{S}_z^2, \\ \hat{H}_\perp &= \frac{K}{4}(e^{2i\varphi} + e^{-2i\varphi})(\hat{S}_y^2 - \hat{S}_x^2) \\ &+ \frac{iK}{4}(e^{2i\varphi} - e^{-2i\varphi})(\hat{S}_x\hat{S}_y + \hat{S}_y\hat{S}_x). \end{aligned} \quad (4)$$

Here $\hat{H}^{(0)}$ describes the mechanical rotation about the $0Z$ axis and the rotation-independent precession of spin about the $0Z$ axis in the uniaxial anisotropy field (anisotropy constant $(D - K/2)$), \hat{H}_\perp defines the interaction between

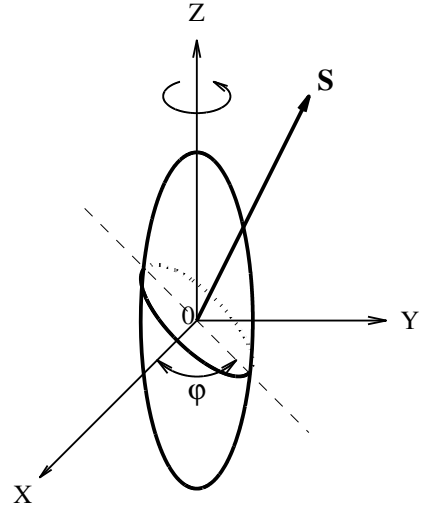


Fig. 1. Nanomagnet rotating about a fixed axis $0Z$ (laboratory frame of reference). Easy axis of magnetic anisotropy (anisotropy constant D) is parallel to axis $0Z$. Medium axis of magnetic anisotropy (anisotropy constant K) is shown by dashed line in plane XY .

the spin- and rotational degrees of freedom. \hat{H}_\perp can be regarded as small perturbation with respect to $\hat{H}^{(0)}$.

We now seek the eigenfunctions and eigenvalues of Hamiltonian \hat{H} within the perturbation theory. To this effect we will use the eigenfunctions and eigenvalues of operator $\hat{H}^{(0)}$, that is, the eigenfunctions and eigenvalues of operators $(\hbar\hat{L}_z)^2/(2I)$ and $-(D - K/2)\hat{S}_z^2$. Since operators \hat{S}_z and $-(D - K/2)\hat{S}_z^2$ commute, they have common eigenfunctions; we will denote them as $|m\rangle$ and thus arrive at the following:

$$\begin{aligned} \hat{S}_z|m\rangle &= m|m\rangle, \\ -\left(D - \frac{K}{2}\right)\hat{S}_z^2|m\rangle &= -\left(D - \frac{K}{2}\right)m^2|m\rangle \\ &\equiv E_m^{(0)}|m\rangle, \end{aligned} \quad (5)$$

where $m = -S, -S + 1, \dots, S - 1, S$.

Likewise, since operators $\hbar\hat{L}_z$ and $(\hbar\hat{L}_z)^2/(2I)$ commute, we denote their common eigenfunctions as $|l\rangle$ to obtain

$$\begin{aligned} \hbar\hat{L}_z|l\rangle &= \hbar l|l\rangle, \\ \frac{(\hbar\hat{L}_z)^2}{2I}|l\rangle &= \frac{(\hbar l)^2}{2I}|l\rangle \equiv E_l^{(0)}|l\rangle, \end{aligned} \quad (6)$$

where $l = 0, \pm 1, \dots$, and $|l\rangle \equiv e^{il\varphi}/\sqrt{2\pi}$.

So, the eigenfunctions of operator $\hat{H}^{(0)}$ can be represented in the form:

$$|m, l\rangle = |m\rangle \otimes |l\rangle, \quad (7)$$

and the corresponding eigenvalues $\hat{H}^{(0)}$ are

$$E_{m,l}^{(0)} = E_m^{(0)} + E_l^{(0)} \equiv \frac{(\hbar l)^2}{2I} - \left(D - \frac{K}{2}\right)m^2. \quad (8)$$

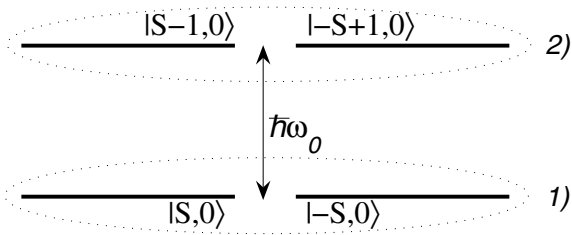


Fig. 2. Fundamental (1) and first excited (2) doublets of nano-magnet in the absence of mechanical rotation.

It follows from equations (7) and (8) that at zero-order approximation and $l = 0$, i.e., in the absence of mechanical rotation, the energy

$$E_{\pm S,0}^{(0)} = - \left(D - \frac{K}{2} \right) S^2 \quad (9)$$

corresponds to fundamental doublet ($|S, 0\rangle$ and $|-S, 0\rangle$), and the energy

$$E_{\pm(S-1),0}^{(0)} = - \left(D - \frac{K}{2} \right) (S-1)^2 \quad (10)$$

corresponds to first excited doublet ($|S-1, 0\rangle$ and $|-S+1, 0\rangle$). Both these doublets are schematically shown in Figure 2, using the notation

$$\hbar\omega_0 = E_{\pm(S-1),0}^{(0)} - E_{\pm S,0}^{(0)} = \left(D - \frac{K}{2} \right) (2S-1).$$

Note that frequency ω_0 is essentially the frequency of the ferromagnetic resonance.

Now, following the standard procedure (see, e.g., Ref. [27]), we can easily find the wave functions of the fundamental and first excited doublets in the first approximation. Using notations (7) and equation (8) instead of $|S, 0\rangle$ and $|-S, 0\rangle$ for the wave functions of the fundamental doublet, we arrive at the following equations:

$$\begin{aligned} \Phi_S &\simeq (1 + \mathbb{C}_S^2)^{-1/2} (|S, 0\rangle + \mathbb{C}_S |S-2, 2\rangle), \\ \Phi_{-S} &\simeq (1 + \mathbb{C}_S^2)^{-1/2} (|-S, 0\rangle + \mathbb{C}_S |-S+2, -2\rangle), \end{aligned} \quad (11)$$

and instead of $|S-1, 0\rangle$ and $|-S+1, 0\rangle$ for the wave functions of the first excited doublet we have:

$$\begin{aligned} \Phi_{S-1} &\simeq (1 + \mathbb{C}_{S-1}^2)^{-1/2} (|S-1, 0\rangle + \mathbb{C}_{S-1} |S-3, 2\rangle), \\ \Phi_{-S+1} &\simeq (1 + \mathbb{C}_{S-1}^2)^{-1/2} (|-S+1, 0\rangle \\ &\quad + \mathbb{C}_{S-1} |-S+3, -2\rangle). \end{aligned} \quad (12)$$

In equations (11) and (12)

$$\begin{aligned} \mathbb{C}_S &= \frac{\langle S-2, 2 | H_{\perp} | S, 0\rangle}{E_{S,0}^{(0)} - E_{S-2,2}^{(0)}} \\ &= \frac{\langle -S+2, -2 | H_{\perp} | -S, 0\rangle}{E_{-S,0}^{(0)} - E_{-S+2,-2}^{(0)}} \\ &= \frac{IK\sqrt{S(2S-1)}}{4I(2D-K)(S-1) + 4\hbar^2}, \\ \mathbb{C}_{S-1} &= \frac{\langle S-3, 2 | H_{\perp} | S-1, 0\rangle}{E_{S-1,0}^{(0)} - E_{S-3,2}^{(0)}} \\ &= \frac{\langle -S+3, -2 | H_{\perp} | -S+1, 0\rangle}{E_{-S+1,0}^{(0)} - E_{-S+3,-2}^{(0)}} \\ &= \frac{IK\sqrt{3(S-1)(2S-1)}}{4I(2D-K)(S-2) + 4\hbar^2}. \end{aligned} \quad (13)$$

It is obvious that the right-hand parts of equations (11) and (12) are basically superpositions of states with $l = 0$ and $l = \pm 2$, which corresponds to inclusion of the interaction between spin and rotational degrees of freedom in the first approximation.

One can easily check that wave functions $\Phi_{\pm S}$ (11) and $\Phi_{\pm(S-1)}$ (12) are eigenfunctions of the operator of the total angular momentum projection on axis OZ :

$$\hbar\hat{M}_z = \hbar\hat{L}_z + \hbar\hat{S}_z. \quad (14)$$

This fact naturally follows from condition

$$[\hbar\hat{M}_z, \hat{H}] = 0 \quad (15)$$

hereinafter the square brackets denote the commutator.

Note that in the first approximation the pair of states (11) still corresponds to energy $E_{\pm S,0}^{(0)}$ (see Eq. (9)), and (12) to energy $E_{\pm(S-1),0}^{(0)}$ (see Eq. (10)), i.e., the degeneracy is not removed ((11) and (12) are the states with $M_z \neq 0$). It is easily seen that double degeneracy occurs at any order of the perturbation theory: each, of the energy levels in question ($M_z \neq 0$), corresponds to two states that differ in the sign of the total angular momentum projection on axis OZ (this issue is considered in ample detail in Appendix). In reality there is always a perturbation whose symmetry differs from symmetry \hat{H} , like, for example, perturbation caused by a magnetic field that is non parallel to the OZ axis. By taking into account such, however small, perturbation one can remove the degeneracy. We assume this to be the case in our study. Then the value of splitting of the fundamental doublet is Δ_S , i.e., the energies of the states take the following form:

$$\begin{aligned} E_S^{(+)} &\simeq E_{\pm S,0}^{(0)} + \frac{\Delta_S}{2}, \\ E_S^{(-)} &\simeq E_{\pm S,0}^{(0)} - \frac{\Delta_S}{2}, \end{aligned} \quad (16)$$

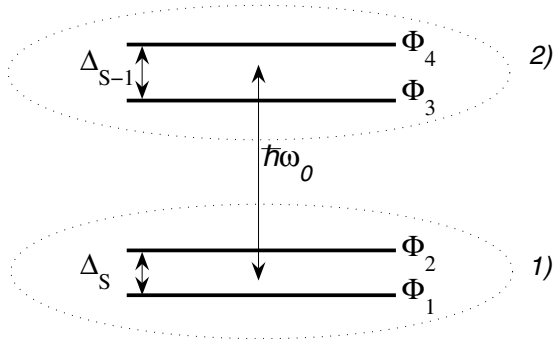


Fig. 3. Fundamental (1) and first excited (2) doublets of a mechanically rotating nanomagnet.

where the state with energy $E_S^{(-)}$ is described by a corresponding symmetric combination of functions $\Phi_{\pm S}$, that we denote as:

$$\Phi_1 = \frac{\Phi_S + \Phi_{-S}}{\sqrt{2}}, \quad (17)$$

and the state with energy $E_S^{(+)}$ corresponds to antisymmetric combination of functions $\Phi_{\pm S}$; we will denote it as:

$$\Phi_2 = \frac{\Phi_S - \Phi_{-S}}{\sqrt{2}}. \quad (18)$$

In a similar way, the splitting value of the first excited doublet is Δ_{S+1} , i.e., the energies of the states corresponding to this doublet will be defined as:

$$\begin{aligned} E_{S-1}^{(+)} &\simeq E_{\pm(S-1),0}^{(0)} + \frac{\Delta_{S-1}}{2}, \\ E_{S-1}^{(-)} &\simeq E_{\pm(S-1),0}^{(0)} - \frac{\Delta_{S-1}}{2}; \end{aligned} \quad (19)$$

here the state with energy $E_{S-1}^{(-)}$ corresponds to a symmetric combination of functions $\Phi_{\pm(S-1)}$, denoted as:

$$\Phi_3 = \frac{\Phi_{S-1} + \Phi_{-S+1}}{\sqrt{2}}, \quad (20)$$

while the state with energy $E_{S-1}^{(+)}$ corresponds to an antisymmetric combination of functions $\Phi_{\pm(S-1)}$, we will define it as:

$$\Phi_4 = \frac{\Phi_{S-1} - \Phi_{-S+1}}{\sqrt{2}}. \quad (21)$$

So, the states (Φ_1, Φ_2) and (Φ_3, Φ_4) are, respectively, the states of fundamental and first excited doublets of a rotating nanomagnet (Fig. 3).

Note that the degeneracy is removed in high orders of the perturbation theory, hence, the relation below is valid:

$$\Delta_S, \Delta_{S-1} \ll \hbar\omega_0. \quad (22)$$

In our further consideration it is (17), (18), (20) and (21) that will be used as the base functions.

Before proceeding to the next section we would like to emphasize the obvious circumstance that is important

to our further consideration of the subject. Specifically, from (4) we have:

$$\begin{aligned} \frac{\partial \hat{S}_z}{\partial t} &= -\frac{\partial \hat{L}_z}{\partial t} = \frac{K}{2\hbar} \left(i(e^{2i\varphi} - e^{-2i\varphi}) (\hat{S}_y^2 - \hat{S}_x^2) \right. \\ &\quad \left. - (e^{2i\varphi} + e^{-2i\varphi}) (\hat{S}_x \hat{S}_y + \hat{S}_y \hat{S}_x) \right). \end{aligned} \quad (23)$$

It follows from equation (23) that in the case of interest the interaction between mechanical rotational degrees of freedom and spin degrees of freedom occurs at $K \neq 0$.

3 Rotation of nanomagnet in alternating magnetic field

Let us now consider a rotating nanomagnet in an alternating magnetic field of an electromagnetic wave. The alternating magnetic field acts directly on the nanomagnet spin, changing its direction, which, in turn, causes a change in the rotational motion of nanomagnet. This is basically the physical mechanism underlying the effect we discuss below.

Consider an alternating elliptically polarized magnetic field that is perpendicular to axis OZ (i.e., the electromagnetic wave propagates along the OZ axis). The operator of nanomagnet interaction with such a field has the form:

$$\hat{V} = g\mu_B \hat{S}_x H_0 \sin(\omega t) + g\mu_B \hat{S}_y H_0 \sin(\omega t + \alpha), \quad (24)$$

where H_0 is the magnetic field amplitude, ω the frequency of the magnetic field ($\omega > 0$), α is the phase, g is the Lande factor, μ_B is the Bohr magneton.

We will study the nanomagnet behavior, using the density matrix formalism and considering only four states that correspond to fundamental doublet (Φ_1, Φ_2) and first excited doublet (Φ_3, Φ_4) . That is, we actually treat nanomagnet as a four-level system. Such an approach is justified, since we reduce our consideration to the case of fairly low temperatures, when in the state of thermodynamic equilibrium only fundamental doublet (Φ_1, Φ_2) is populated, and because the only transitions under the alternating magnetic field are those between the states of the fundamental and first excited doublets: $(\Phi_1, \Phi_2) \leftrightarrow (\Phi_3, \Phi_4)$.

In the common relaxation-time τ model the Schrodinger representation of equations for the density matrix elements will have the form [28]:

$$\begin{aligned} \frac{\partial \rho_{kk}(t)}{\partial t} &= i\hbar^{-1} \left[\hat{\rho}(t), \hat{H} + \hat{V} \right]_{kk} + \left(\rho_{kk}^{(0)} - \rho_{kk}(t) \right) \tau^{-1}, \\ \frac{\partial \rho_{kj}(t)}{\partial t} &= i\hbar^{-1} \left[\hat{\rho}(t), \hat{H} + \hat{V} \right]_{kj} - \rho_{kj}(t) \tau^{-1}, \quad (k \neq j). \end{aligned} \quad (25)$$

Here indices $k, j = 1, 2, 3, 4$ correspond to functions $\Phi_1, \Phi_2, \Phi_3, \Phi_4$. The matrix elements of operator \hat{V} are readily found, since we can easily find the matrix elements of operators $\hat{S}_{x,y}$ (in the zeroth approximation):

$$\begin{aligned} (S_x)_{13} &= (S_x)_{31} = (S_x)_{24} = (S_x)_{42} = \sqrt{2S}, \\ (S_y)_{14} &= -(S_y)_{41} = (S_y)_{23} = -(S_y)_{32} = -i\sqrt{2S}. \end{aligned}$$

In equation (25) $\rho_{kk}^{(0)}$ are the values of the diagonal elements of density matrix at thermodynamic equilibrium. Following from the assumption of only fundamental doublet being populated at thermodynamic equilibrium state, for $\rho_{kk}^{(0)}$ we derive:

$$\rho_{kk}^{(0)} = \begin{cases} \frac{1}{2}, & k = 1, 2, \\ 0, & k = 3, 4. \end{cases} \quad (26)$$

It is obvious that equation (34) is valid at temperatures satisfying the condition

$$\Delta_S \ll kT < \hbar\omega_0, \quad (27)$$

(see Fig. 3 and Eq. (22)). For estimations we can take $\Delta_S \leq 10^{-6}$ K, $\hbar\omega_0 \sim 5$ K (see, e.g., Refs. [26,29]), so that condition (27) is fulfilled at $T \sim 1$ K.

Our goal is to calculate the angular speed of rotation of nanomagnet about the 0Z axis. Since operator of the angular speed is $\hat{\Omega} = I^{-1}\hbar\hat{L}_z$, in accordance with the mean value definition we obtain:

$$\langle \hat{\Omega} \rangle = \text{Sp}(\hat{\rho}I^{-1}\hbar\hat{L}_z). \quad (28)$$

We are interested in the stationary rotation mode, which is the case at $t \gg \tau$. Therefore, we seek a stationary (corresponding to $t \gg \tau$) solution to system (25). It can be easily found based on some simplifying assumptions. As a first step, we use a resonant approximation (the rotating wave approximation, see Refs. [27,30]). This approximation imposes a restriction on frequency: $\omega - \omega_0 \ll \omega_0$. Secondly, we assume $\Delta_S = \Delta_{S-1} = 0$. This assumption makes sense because the effect in question is basically unrelated to the condition $\Delta_S \neq 0$ and $\Delta_{S-1} \neq 0$ and, besides, relation (22) holds here. Based on these assumptions we derive from (25):

$$\begin{aligned} \rho_{34} + \rho_{43} &= -(\rho_{12} + \rho_{21}) \\ &= \frac{2g^2\mu_B^2\hbar^{-2}SH_0^2\tau^2 \sin \alpha}{1 + (\omega - \omega_0)^2\tau^2 + 8g^2\mu_B^2\hbar^{-2}SH_0^2\tau^2(1 + 4\cos^2 \alpha)}. \end{aligned} \quad (29)$$

Next, from equations (17), (18) and (20), (21) given (3) it follows:

$$\begin{aligned} (L_z)_{12} &= (L_z)_{21} \simeq \frac{1}{16} \left(\frac{K}{D} \right)^2, \\ (L_z)_{34} &= (L_z)_{43} \simeq \frac{3}{16} \left(\frac{K}{D} \right)^2. \end{aligned} \quad (30)$$

Then, using equations (28)–(30) we find for the mean value of the angular speed of nanomagnet rotation about the 0Z axis:

$$\begin{aligned} \Omega \equiv \langle \hat{\Omega} \rangle &\simeq \frac{\hbar}{4I} \left(\frac{K}{D} \right)^2 \\ &\times \frac{g^2\mu_B^2\hbar^{-2}SH_0^2\tau^2 \sin \alpha}{1 + (\omega - \omega_0)^2\tau^2 + 8g^2\mu_B^2\hbar^{-2}SH_0^2\tau^2(1 + 4\cos^2 \alpha)}. \end{aligned} \quad (31)$$

In deriving equation (31) we have taken into account that $S \gg 1$.

First of all, note the conclusion that follows from equation (31): the effect of mechanical rotation occurs only at $K/D \neq 0$. This result is fully consistent with the statement that, in the case of interest, interaction between the mechanical rotational- and spin degrees of freedom takes place at $K \neq 0$ (see end of Sect. 2). It is also seen that the angular speed of rotation is proportional to the squared amplitude of the magnetic field. The speed is maximum for a circular polarization of magnetic field ($\alpha = \pm\pi/2$), when the direction of rotation of nanomagnet changes for the opposite, following a change in the field circular polarization from right to left-hand and vice versa. In the case of linear polarization ($\alpha = 0$) the rotation speed is zero. The rotation speed value depends on relaxation time τ : the smaller the relaxation time (i.e., the higher the relaxation process intensity), the lower the speed of rotation.

4 Numerical estimation

It should be noted, in the first place, that the resulting estimates highly depend on quantity τ . Unfortunately, there is no reasonably reliable data on the significance of this quantity for freely rotating nanomagnets. Yet, we assume that τ should not be smaller than the time of spin relaxation in crystals of magnetic molecules. According to spectroscopic measurements data [29], the spin relaxation time in Fe₈ crystals is on the order of 10^{-9} s. We assume $\tau \sim 10^{-9}$ s, which is certain to not lead us to overestimation of the effect value. Therefore, at reasonable values of the radiation power on the order of 1 W/cm², which corresponds to the magnetic field amplitude $H_0 \sim 6 \times 10^{-2}$ G, we have:

$$g^2\mu_B^2\hbar^{-2}SH_0^2\tau^2 \ll 1. \quad (32)$$

Then, instead of equation (31) we will have:

$$\Omega \simeq \frac{\hbar}{4I} \left(\frac{K}{D} \right)^2 \frac{g^2\mu_B^2\hbar^{-2}SH_0^2\tau^2 \sin \alpha}{1 + (\omega - \omega_0)^2\tau^2}. \quad (33)$$

The estimations were made for a molecular cluster Fe₈: based on the data from reference [25] we arrived at $K = 0.092$ K, $D = 0.402$ K. To avoid confusion, note that we here use the notations for the anisotropy axes and anisotropy constants (see Eq. (1)), that are different from those in reference [25]. For a molecule inertia moment, according to reference [16], we may take $I \sim 10^{-35}$ g cm² (thus making the use of relation (3) justified). In result, for a circular polarization of the wave, at resonance $\omega = \omega_0$ (according to Ref. [29] $\omega_0 \sim 2\pi \times 10^2$ GHz) and power of the order of 1 W/cm² we obtained: $\Omega \simeq 10$ rad/s.

5 Discussion and conclusion

Our consideration is based on usage of the density matrix formalism; yet, there is a fairly simple treatment of the physics behind the effect in question. It involves using

the classical equations of motion for spin degrees of freedom with minimum of quantum mechanics to describe the mechanical rotational degrees of freedom and their interaction with the spin degrees of freedom. First of all, we recall that interaction between the spin degrees of freedom and mechanical rotational degrees of freedom takes place only at $K \neq 0$. Therefore, in a zero approximation ($K = 0$) the behavior of spin in an external magnetic field in no way is related with nanomagnet rotation and, due to $K = 0$, again, there are no effects of magnetic quantum tunnelling. Further, as we consider large spin values ($S \gg 1$) and neglect the quantum tunnelling effects, we can use the classical Landau-Lifshitz-Gilbert equation for the spin \mathbf{S} . For definiteness, we look at the situation when the external field is circularly polarized, $\alpha \equiv \pi/2$. Such a field interacts (under the resonance approximation) only with nanorotors having $S_z > 0$. For components $S_{x,y,z}$ we easily derive the equations:

$$\begin{aligned}\frac{\partial S_x}{\partial t} &= \frac{2}{\hbar} DS \cdot S_y - \frac{1}{\hbar} g\mu_B S_z H_0 \sin \omega t - \frac{1}{\tau} S_x, \\ \frac{\partial S_y}{\partial t} &= -\frac{2}{\hbar} DS \cdot S_x - \frac{1}{\hbar} g\mu_B S_z H_0 \cos \omega t - \frac{1}{\tau} S_y, \\ \frac{\partial S_z}{\partial t} &= \frac{1}{\hbar} g\mu_B (S_x \sin \omega t + S_y \cos \omega t) + \frac{2}{\tau} (S - S_z). \quad (34)\end{aligned}$$

Here τ is the relaxation time. In writing (34) we used the approximation $|S_z - S| \ll S_x, S_y$ for the smallness of deflection \mathbf{S} from equilibrium. The stationary ($t \gg \tau$) solution of equation (34) is easily found, so, if (32), we derive from equation (34):

$$S_z - S \simeq -\frac{1}{2} \frac{g^2 \mu_B^2 \hbar^{-2} S H_0^2 \tau^2}{1 + (\omega - 2DS\hbar^{-1})^2 \tau^2}. \quad (35)$$

Note that in terms of quantum mechanics the result (35) corresponds to the resonance interaction of a circularly polarized field with nanorotors in the states Φ_S and Φ_{S-1} (see Eqs. (11) and (12)).

Now we take into account the mechanical rotational degrees of freedom and their interaction with the spin degrees of freedom, i.e., allow for $K \neq 0$. This interaction, as seen from equations (11) and (12), manifests itself through state Φ_S being a superposition of $|S, 0\rangle$ and $|S-2, 2\rangle$, and state Φ_{S-1} being a superposition of $|S-1, 0\rangle$ and $|S-3, 2\rangle$. From expressions (11)–(13), considering equation (3), it follows that in transition from state Φ_S to state Φ_{S-1} the quantum mechanical mean value of the projection of spin S_z changes roughly by -1 , while the quantum mechanical mean value of the mechanical moment $I\Omega$ changes by about $\hbar/8(K/D)^2$. With this in mind, we can find the change in the mechanical moment, corresponding to variation of the spin projection by the value (35), which makes it easy to obtain the expression for the angular speed:

$$\Omega \simeq \frac{\hbar}{16I} \left(\frac{K}{D} \right)^2 \frac{g^2 \mu_B^2 \hbar^{-2} S H_0^2 \tau^2}{1 + (\omega - 2DS\hbar^{-1})^2 \tau^2}. \quad (36)$$

Compare equations (36) and (33). Since equation (36) corresponds to $\alpha \equiv \pi/2$, and in equation (33) $\omega_0 \approx 2DS/\hbar$

($D \gg K, S \gg 1$), equations (36) and (33) differ only by an insignificant numerical factor. The above calculations and our treatment of the problem are fairly simple; yet, as easily seen, they provide an insight into the physics of the problem.

Hypothetically, this problem could have been solved entirely as the classical one, which is beyond our consideration. It is doubtful, though, that in the classical model the calculations would have been less complicated than in our quantum mechanical approach that is based on the density matrix formalism. We stress again that this approach is quite simple in terms of mathematical calculations.

Now, let us sum up.

Based on the quantum mechanical approach we have found a solution to the problem of a molecular nanomagnet rotating about a fixed axis under the action of an elliptically polarized wave propagating along the rotation axis. The wave frequency corresponds to the frequency of ferromagnetic resonance. A stationary mode of rotation was considered, that is realized on a time scale largely in excess of the characteristic time of relaxation. It was shown that the speed of rotation is proportional to the squared amplitude of the magnetic field of the wave and is maximum for a circular polarization of the latter. A change in the circular polarization direction leads to a change in the direction of rotation. Estimations of the effect, made for molecular cluster Fe_8 have shown that at temperature ~ 1 K and fairly modest values of power ~ 1 W/cm² the speed of rotation may reach 10 rad/s.

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Appendix: Degeneracy of the energy levels of a freely rotating nanomagnet

Here we show the energy levels corresponding to Hamiltonian (4) to be degenerate in the case $M_z \neq 0$. Operators \hat{H} (Eq. (4)) and $\hbar\hat{M}_z$ (Eq. (14)) are commutative (see Eq. (15)), we will define their common eigenfunctions as Φ_{E, M_z} . These functions correspond to the states with certain values of energy E and total angular momentum $\hbar M_z = \hbar(m+l)$.

$$\hat{H}\Phi_{E, M_z} = E\Phi_{E, M_z}, \quad (A.1)$$

$$\hat{M}_z\Phi_{E, M_z} = M_z\Phi_{E, M_z}. \quad (A.2)$$

We now represent functions Φ_{E, M_z} as a series that is bound to satisfy equation (A.2):

$$\Phi_{E, M_z} = \sum_m C_{m, M_z - m} |m, M_z - m\rangle, \quad (A.3)$$

where $|m, M_z - m\rangle \equiv |m, l\rangle = |m\rangle \otimes |l\rangle$ (see Eq. (7)).

The process of finding Φ_{E, M_z} , i.e., the values of $C_{m, M_z - m}$ is reduced to solving equation (A.1) in the matrix form. For the basis functions we use $|m, M_z - m\rangle$. Then the matrix elements of operator \hat{H} are:

$$\begin{aligned} H_{m, M_z - m; m, M_z - m} &= H_{m, M_z - m; m, M_z - m}^{(0)} \\ &= E_{m, M_z - m}^{(0)}, \end{aligned} \quad (\text{A.4})$$

see equation (8), and

$$\begin{aligned} H_{m, M_z - m; m-2, M_z - m + 2} &= (H_{\perp})_{m, M_z - m; m-2, M_z - m + 2} \\ &= -\frac{K}{4} \sqrt{(S+m)(S-m+1)(S+m-1)(S-m+2)}. \end{aligned} \quad (\text{A.5})$$

Using equations (A.4) and (A.5) from (A.1), we will arrive at a set of $2S+1$ equations for $C_{m, M_z - m}$:

$$\begin{aligned} &\left(E_{m, M_z - m}^{(0)} - E \right) C_{m, M_z - m} \\ &- \frac{K}{4} \sqrt{(S+m)(S-m+1)(S+m-1)(S-m+2)} \\ &\quad \times C_{m-2, M_z - m + 2} \\ &- \frac{K}{4} \sqrt{(S+m+2)(S-m-1)(S+m+1)(S-m)} \\ &\quad \times C_{m+2, M_z - m - 2} = 0. \end{aligned} \quad (\text{A.6})$$

It is easily seen that, if S is an integer, the system (A.6) splits into two independent subsystems: one containing $S+1$ equations for $m = S, S-2, \dots, -S+2, -S$, the other having S equations for $m = S-1, S-3, \dots, -S+3, -S+1$. If S is a half-integer, one subsystem will include $S+1/2$ equations for $m = S, S-2, \dots, -S+3, -S+1$, while the other will also have $S+1/2$ equations for $m = S-1, S-3, \dots, -S+2, -S$.

Let us change $M_z \rightarrow -M_z$ in (A.6), i.e., use the equations for $C_{m, -M_z - m}$. Quantities $C_{m, -M_z - m}$ correspond to the states with the value of the total angular momentum $-\hbar M_z$ (see Eq. (A.3)). We now make one more change of $m \rightarrow -m$ in (A.6) simply for convenience, since both m and $-m$ have the same values from $-S$ to S . Also, we make use of the fact that $E_{m, M_z - m}^{(0)} = E_{-m, -M_z + m}^{(0)}$ (see Eq. (8)). Thus, instead of (A.6) we will eventually arrive at:

$$\begin{aligned} &\left(E_{m, M_z - m}^{(0)} - E \right) C_{-m, -M_z + m} \\ &- \frac{K}{4} \sqrt{(S+m)(S-m+1)(S+m-1)(S-m+2)} \\ &\quad \times C_{-m+2, -M_z + m - 2} \\ &- \frac{K}{4} \sqrt{(S+m+2)(S-m-1)(S+m+1)(S-m)} \\ &\quad \times C_{-m-2, -M_z + m + 2} = 0. \end{aligned} \quad (\text{A.7})$$

Comparing between equations (A.6) and (A.7) one can see that the coefficients matrices of those systems, coincide. This implies that solution of the sets (A.6) and (A.7) will

yield the same values for energy E_n (n is their index number). We will denote solutions of the system (A.6), corresponding to a certain value of E_n , as $C_{m, M_z - m}(E_n)$, and those for system (A.7), that correspond to the same E_n , as $C_{-m, -M_z + m}(E_n)$. Now, in compliance with (A.3), we write the eigenfunction corresponding to energy E_n and angular momentum M_z :

$$\Phi_{E_n, M_z} = \sum_m C_{m, M_z - m}(E_n) |m, M_z - m\rangle \quad (\text{A.8})$$

and the eigenfunction corresponding to energy E_n and angular momentum $-M_z$:

$$\Phi_{E_n, -M_z} = \sum_m C_{-m, -M_z + m}(E_n) |-m, -M_z + m\rangle. \quad (\text{A.9})$$

In equations (A.8) and (A.9), m lies within the same interval of values. We consider the case $M_z \neq 0$, when the basis functions $|m, M_z - m\rangle$ and $|-m, -M_z + m\rangle$, once M_z is fixed, do not coincide even at different values of m for both the functions. Thus, $\Phi_{E_n, M_z} \neq \Phi_{E_n, -M_z}$, which testifies to the occurrence of the energy levels degeneracy.

It should be emphasized that the approach used in this study is quite common, and the obtained result is in perfect agreement with the findings in reference [16], in which a rotating nanomagnet is regarded as a mechanically rigid two-state spin system. Also note that, as shown in reference [16], condition $M_z \neq 0$ corresponds to the low-energy states of a molecular cluster Fe_8 , which are of interest to this study. Finally, note that there is no degeneracy in the case $M_z = 0$, yet, the issue is beyond the scope of our present research. This fact is proved in reference [16], based on approximations.

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