Collective spin dynamics in magnetic nanomaterials

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Abstract. Magnetic nanomaterials are considered, formed by magnetic nanomolecules with high spins. The problem of spin reversal in these materials is analyzed, which is of interest for the possible use of such materials for quantum information processing and quantum computing. The fastest spin reversal can be achieved by coupling the spin sample to a resonant electric circuit and by an appropriate choice of the system parameters. A principal point is to choose these parameters so that to organize coherent spin motion. Dynamics of collective motion is modelled by computer simulations, which confirm the high level of dynamical coherence of molecular spins in the process of spin reversal.

1. Introduction
There exists a wide class of nanomolecules possessing large magnetic moments, reaching tens of the Bohr magnetons. Ensembles of such molecules form crystalline clusters, called molecular nanomagnets. The properties of these materials have been described in review articles [1–4]. Because of the high molecular spins and regular crystalline structure, molecular nanomagnets are considered as promising candidates for quantum information processing and quantum computing. The main problem, hindering their application for that purpose, is a very slow relaxation of the magnetic moment. While, for serving as a fast device for information processing, it is necessary that magnetic moments could be quickly manipulated between up and down directions.

In order to realize the spin reversal in molecular nanomagnets as an ultrafast process, a mechanism has been suggested [3–7] allowing one to reach the characteristic reversal time as short as $10^{-12}$ s. This can be achieved by connecting the molecular sample to a resonant electric circuit, providing feedback to the spin motion and organizing the spin relaxation as a collective process with a high level of coherence in spin dynamics.

There also exists another type of magnetic nanomaterials, formed by magnetic nanoclusters [3], whose spins can reach $s \sim 100 - 10^4$. In principle, ensembles of such nanoclusters could also be employed for realizing collective spin relaxation as an ultrafast process. There is, however, an important difference between nanomolecules and nanoclusters. A collection of nanomolecules forms a well-defined crystalline structure, with all molecules being exactly the same, possessing identical sizes and all properties. But nanoclusters do not form crystals, their collections
representing a kind of powder. The most important is that it is impossible to produce a large number of nanoclusters with identical properties. Sizes, shapes, spins, and all other parameters of magnetic nanoclusters vary in a wide range. A system of such different nanoclusters would form a strongly nonuniform matter, with a large inhomogeneous broadening, hindering the possibility of coherent spin motion. This is why magnetic nanomolecules are preferable for realizing coherent spin dynamics.

The aim of the present communication is to study spin dynamics of magnetic nanomolecules in the regime of their maximally fast spin reversal, which can be found by varying the system parameters. We concentrate our attention on the analysis of dynamical coherence of moving spins. For this purpose, we study the temporal behavior of the coherence factor. We demonstrate that, appropriately choosing the system parameters and tuning the Zeeman frequency of nanomolecules in resonance with the electric circuit, it is feasible to realize highly coherent spin dynamics.

2. Basic equations

The system of magnetic molecules is characterized by the Hamiltonian

$$\hat{H} = \sum_i \hat{H}_i + \frac{1}{2} \sum_{i \neq j} \hat{H}_{ij},$$

in which the index $i = 1, 2, \ldots, N$ enumerates molecules,

$$\hat{H}_i = -\mu_0 \mathbf{B} \cdot \mathbf{S}_i - D(S^z_i)^2,$$

with $\mu_0 = -2\mu_B$ being the electronic magnetic moment and $D$ being the magnetic anisotropy parameter, the total magnetic field

$$\mathbf{B} = B_0 \mathbf{e}_z + H \mathbf{e}_x,$$

includes an external static magnetic field $B_0$ and the resonator feedback field $H$; the interaction term is

$$\hat{H}_{ij} = \sum_{\alpha\beta} D^{\alpha\beta}_{ij} S^\alpha_i S^\beta_j,$$

with the dipolar tensor

$$D^{\alpha\beta}_{ij} = \frac{\mu_0^2}{r_{ij}^3} \left( \delta_{\alpha\beta} - 3n_{ij}^\alpha n_{ij}^\beta \right),$$

where $r_{ij} \equiv |\mathbf{r}_{ij}|$, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, $n_{ij} \equiv \mathbf{r}_{ij}/r_{ij}$. The resonator feedback field is produced by a magnetic coil surrounding the sample and can be described [8,9] by the Kirchhoff equation

$$\frac{dH}{dt} + 2\gamma H + \omega^2 \int_0^t H(t') dt' = -4\pi \eta \frac{dm_x}{dt},$$

in which $\gamma$ is the resonator damping, $\omega$ is the resonator natural frequency, $\eta$ is a filling factor, and

$$m_x = \frac{\mu_0}{V} \sum_j <S^x_j>$$

is the transverse magnetization density, with $V$ being the sample volume. The overall experimental setup is explained in detail in the review article [4].

The main parameters characterizing the system are the Zeeman frequency

$$\omega_0 \equiv -\frac{\mu_0}{\hbar} B_0 = \frac{2}{\hbar} \mu_B B_0,$$
the resonator natural frequency $\omega$, the anisotropy frequency

$$\omega_D \equiv (2S - 1) \frac{D}{\hbar}, \quad (9)$$

the spin-phonon attenuation $\gamma_1 \equiv 1/T_1$, the spin-dephasing attenuation $\gamma_2 \equiv 1/T_2$, and the resonator dumping $\gamma$. The frequencies are assumed to satisfy the resonance conditions

$$\left| \frac{\omega - \omega_0}{\omega_0} \right| \ll 1, \quad \frac{\omega_D}{\omega_0} \ll 1. \quad (10)$$

The equations of motion for the ladder spin operator $S_j^- \equiv S_j^x - iS_j^y$ and the longitudinal spin operator $S_j^z$ are obtained [3–6] from the Heisenberg equations for the spin operators, with allowance made for the attenuation terms. This yields the equations of motion

$$\frac{dS_j^-}{dt} = -i \left( \omega_0 + \xi_j^0 - i\Gamma_2 \right) S_j^- + f_j S_j^z + i \frac{\omega_D}{S} S_j^z S_j^-,$$

$$\frac{dS_j^z}{dt} = -\frac{1}{2} \left( f_j^+ S_j^- + S_j^+ f_j \right) - \gamma_1 \left( S_j^z - \zeta \right), \quad (11)$$

in which $\zeta$ is an equilibrium spin polarization and

$$f_j = -\frac{i}{\hbar} \mu_0 H + \xi_j, \quad \Gamma_2 = \gamma_2 \left( 1 - s^2 \right),$$

$$\xi_j^0 = \frac{1}{\hbar} \sum_{j(\neq i)} \left( a_{ij} S_j^z + c_{ij} S_j^- - c_{ij} S_j^+ \right),$$

$$\xi_j = \frac{i}{\hbar} \sum_{j(\neq i)} \left( 2c_{ij} S_j^z - \frac{1}{2} a_{ij} S_j^- + 2b_{ij} S_j^+ \right).$$

Here

$$s \equiv \frac{1}{SN} \sum_{j=1}^N < S_j^z > \quad (12)$$

is the reduced longitudinal spin polarization.

To characterize the level of coherence in spin motion, we consider the total magnetodipole radiation intensity

$$I(t) = I_{inc}(t) + I_{coh}(t), \quad (13)$$

which is separated into the incoherent and coherent parts,

$$I_{inc} = \frac{2\mu_0^2}{3c^3} \sum_j | < \tilde{S}_j | |^2, \quad I_{coh} = \frac{2\mu_0^2}{3c^3} \sum_{i \neq j} < \tilde{S}_i \tilde{S}_j >, \quad (14)$$

where the overdots imply the time differentiation. The level of dynamic spin coherence is described by the coherence factor

$$C(t) \equiv \frac{I_{coh}(t)}{I(t)}. \quad (15)$$

The system is, first, prepared in a nonequilibrium state, with a positive initial spin polarization $s_0 = s(0)$ in a positive external magnetic field $B_0$. According to Eq. (2), molecular spins tend to reverse to negative values corresponding to an equilibrium state.
3. Collective dynamics

We analyzed the system of equations (11) in two ways. One way is by treating the spin variables $S_{j}^{\alpha}$ as operators, averaging Eqs. (11), and employing the scale-separation approach [8,9]. The second way is by considering $S_{j}^{\alpha}$ as classical variables and accomplishing direct computer simulations of Eqs. (11). The scale-separation approach is more appropriate for the initial stage of spin relaxation, when spins fluctuate chaotically and their quantum nature is essential. At the stage of the coherent spin motion, both ways give close results, though the direct computer simulation is more accurate, exhibiting some fine-structure oscillations that are smoothed out in the averaging technique.

Accomplishing numerical calculations for different system parameters, we set the filling factor $\eta = 1$ and take into account that for typical magnetic nanomolecules $\gamma_1 \ll \gamma_2$. By varying other parameters, we find that the fastest spin reversal occurs for the largest $\omega_0$ and $s_0$, but for the smallest $\gamma$ and $\omega_D$. Dipole interactions stronger destroy spin coherence for larger spins. For lower spins, the spin reversal is more pronounced. This is illustrated in Fig. 1 for $S = 10$ and $S = 1/2$. In this and all following figures, we present the results of the direct computer simulation with $N = 125$ magnetic nanomolecules. The initial spin polarization (12) is taken as $s_0 = 0.9$. Time is measured in units of $T_2$ and all frequencies and damping parameters in units of $\gamma_2$. The resonance condition $\omega_0 = \omega$ is assumed. The anisotropy frequency is taken as $\omega_D = 20$.

Figures 2 to 4 show the temporal behavior of the coherence factor (15) for the varying Zeeman frequency $\omega_0$ and the resonator damping $\gamma$. These figures demonstrate that the collective spin motion is highly coherent. Such a high level of coherence is due to the presence of the resonator, without which spin dynamics could not be coherent [3–9], so that no superradiance could exist [10].

![Figure 1](image1.png)  
**Figure 1.** Reduced spin polarization $s = s(t)$ as a function of dimensionless time for $\omega_0 = 2000$ and $\gamma = 10$, with the molecular spin $S = 10$ (solid line) and $S = 1/2$ (dashed line).

![Figure 2](image2.png)  
**Figure 2.** Coherence factor (15) as a function of dimensionless time for $\omega_0 = 1000$, $\gamma = 10$, and $S = 10$.

In conclusion, we have considered spin dynamics in a strongly nonequilibrium system of magnetic nanomolecules. By varying the system parameters, we find the optimal configurations for the fastest spin reversal. For typical magnetic nanomolecules, the reversed time can be of order $10^{-12} - 10^{-11}$ s. Without a resonator, the spin motion cannot be made coherent, provided
no strong transverse fields are imposed. But in the presence of the resonator, producing a feedback field, spin dynamics can become essentially coherent. The ultrafast self-organized spin reversal can be employed for information processing.

Acknowledgments
The authors acknowledge financial support from the Russian Foundation for Basic Research: Grant 07-02-96026 (V.K.H. and P.V.K.) and Grant 08-02-00118 (V.I.Y. and E.P.Y.).

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