

Spin superradiance by magnetic nanomolecules and nanoclusters

V I Yukalov¹, V K Henner^{2,3} and E P Yukalova⁴

¹Bogolubov Laboratory of Theoretical Physics, Joint Institute for Nuclear Research, Dubna 141980, Russia

²Department of Physics, Perm State University, Perm 614190, Russia

³Department of Physics, University of Louisville, Louisville, Kentucky 40292, USA

⁴Laboratory of Information Technologies, Joint Institute for Nuclear Research, Dubna 141980, Russia

E-mail: yukalov@theor.jinr.ru

Abstract. Spin dynamics of assemblies of magnetic nanomolecules and nanoclusters can be made coherent by inserting the sample into a coil of a resonant electric circuit. Coherence is organized through the arising feedback magnetic field of the coil. The coupling of a magnetic sample with a resonant circuit induces fast spin relaxation and coherent spin radiation, that is, superradiance. We consider spin dynamics described by a realistic Hamiltonian, typical of magnetic nanomolecules and nanoclusters. The role of magnetic anisotropy is studied. A special attention is paid to geometric effects related to the mutual orientation of the magnetic sample and resonator coil.

1. Introduction

There exists a large class of magnetic nanomolecules and magnetic nanoclusters that can be considered as nanoparticles possessing high total spins (see review articles [1–9]). Below blocking temperature, the spin of such magnetic nanoparticles is frozen. For instance, the typical blocking temperature of magnetic nanomolecules is of order 1 – 10 K. The blocking temperature for nanoclusters is 10 – 100 K.

Magnetic properties of nanomolecules and nanoclusters are similar to each other. There are two main features distinguishing them. Magnetic molecules of the same chemical composition are identical and they can form crystals with almost ideal periodic lattice. While magnetic nanoclusters, even being made of the same element, say Fe, Ni, or Co, differ by their sizes, and they do not form periodic structures. Otherwise, the spin Hamiltonian for an ensemble of magnetic nanoparticles is of the same form for nanomolecules as well as for nanoclusters.

In the usual case, spin relaxation is due to spin-phonon interactions and, below the blocking temperature, is very slow. Thus for nanomolecules, the spin-phonon relaxation time is $T_1 \sim (10^5 - 10^7)$ s. But the spin relaxation time can be drastically shortened, if the magnetic sample is inserted into a coil of a resonant electric circuit. This is termed the Purcell effect [10]. In that case, the relaxation is caused by the resonator feedback field collectivizing moving spins and forcing them to move coherently. Coherent spin dynamics have been studied in several publications, e.g., [11–25].



Coherently moving spins produce coherent radiation, which, when it is self-organized, is called superradiance. It is worth stressing that spin superradiance is rather different from atomic superradiance. The latter is caused by the Dicke effect [26], while the cavity Purcell effect is secondary [27–29]. Contrary to this, spin superradiance is completely due to Purcell effect, with the Dicke effect playing no role [30]. The Purcell effect also enhances the signals of nuclear magnetic resonance [31,32] and of spin echo [33–35].

Here we consider the peculiarities of spin superradiance by magnetic nanomolecules and nanoclusters having strong magnetic anisotropy. We shall pay attention to the role of geometric effects related to the finiteness of the considered samples. Finite systems, as is known [36,37], can exhibit properties different from those of bulk systems. In the present case, we are interested in the geometric effects due to the mutual orientation of a finite magnetic sample and the resonator coil.

2. Spin Hamiltonian

An ensemble of magnetic nanomolecules or nanoclusters is described by the Hamiltonian

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

consisting of single-spin terms \hat{H}_i and spin-interaction terms \hat{H}_{ij} , with the index $i = 1, 2, \dots, N$ enumerating nanoparticles. The single-spin Hamiltonian

$$\hat{H}_i = -\mu_0 \mathbf{B} \cdot \mathbf{S} - D(S_i^z)^2 + D_2(S_i^x)^2 + D_4 [(S_i^x)^2(S_i^y)^2 + (S_i^y)^2(S_i^z)^2 + (S_i^z)^2(S_i^x)^2] \quad (2)$$

is a sum of the Zeeman energy and single-site magnetic anisotropy terms. The total magnetic field, acting on each spin,

$$\mathbf{B} = B_0 \mathbf{e}_z + H \mathbf{e}_x, \quad (3)$$

includes an external field B_0 and a resonator feedback field H . Spins interact with each other through dipolar forces characterized by the Hamiltonian

$$\hat{H}_{ij} = \sum_{\alpha\beta} D_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta, \quad (4)$$

with the dipolar tensor

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

where

$$r_{ij} \equiv |\mathbf{r}_{ij}|, \quad \mathbf{n}_{ij} \equiv \frac{\mathbf{r}_{ij}}{r_{ij}}, \quad \mathbf{r}_{ij} \equiv \mathbf{r}_i - \mathbf{r}_j.$$

The resonator feedback field is given by the Kirchhoff equation

$$\frac{dH}{dt} + 2\gamma H + \omega^2 \int_0^t H(t') dt' = -4\pi\eta \frac{dm_x}{dt}, \quad (5)$$

in which γ is resonator damping, ω is resonator natural frequency, η is filling factor, and

$$m_x \equiv \frac{\mu_0}{V} \sum_{j=1}^N \langle S_j^x \rangle \quad (6)$$

is the transverse magnetization density of the sample having volume V .

In addition to the resonator natural frequency ω , there are the following characteristic frequencies. The Zeeman frequency

$$\omega_0 \equiv -\frac{\mu_0}{\hbar} B_0 = \frac{2}{\hbar} \mu_B B_0 \quad (7)$$

and the anisotropy frequencies

$$\omega_D \equiv (2S - 1) \frac{D}{\hbar}, \quad \omega_2 \equiv (2S - 1) \frac{D_2}{\hbar}, \quad \omega_4 \equiv (2S - 1) \frac{D_4}{\hbar} S^2. \quad (8)$$

The resonator natural frequency has to be close to the Zeeman frequency, in order to satisfy the resonance condition

$$\left| \frac{\omega - \omega_0}{\omega} \right| \ll 1. \quad (9)$$

And the Zeeman frequency has to be larger than the anisotropy frequencies that freeze spin motion,

$$\left| \frac{\omega_D}{\omega_0} \right| \ll 1, \quad \left| \frac{\omega_2}{\omega_0} \right| \ll 1, \quad \left| \frac{\omega_4}{\omega_0} \right| \ll 1. \quad (10)$$

Among the anisotropy frequencies, the most important are ω_D and ω_2 that are close to each other. The frequency ω_4 , up to spins $S \sim 10^3$, is much smaller than ω_D .

We analyze the Heisenberg equations of motion for spins in two ways, by employing the scale separation approach [4, 5] and by directly solving the spin evolution equations in semiclassical approximation. Both ways give close results. Finding the average spins as functions of time, we can calculate the radiation intensity.

3. Radiation intensity

The intensity of radiation, induced by moving spins, can be calculated in two ways. One possibility is the classical formula

$$I(t) = \frac{2\mu_0^2}{3c^3} \left| \sum_j \langle \ddot{\mathbf{S}}_j \rangle \right|^2 \quad (11)$$

that should provide good approximation for high spins $S \gg 1$. The other way is to use the quantum formula [5, 38, 39], according to which the radiation intensity

$$I(t) = I_{inc}(t) + I_{coh}(t) \quad (12)$$

is the sum of the incoherent radiation intensity

$$I_{inc}(t) = 2\omega_0 \gamma_0 S N [1 + s(t)] \quad (13)$$

and the coherent radiation intensity

$$I_{coh}(t) = 2\omega_0 \gamma_0 S^2 N^2 \varphi_0 w(t), \quad (14)$$

where the natural width is

$$\gamma_0 \equiv \frac{2}{3} |\vec{\mu}|^2 k_0^3 = \frac{1}{3} \mu_0^2 k_0^3 \quad \left(k_0 \equiv \frac{\omega_0}{c} \right),$$

φ_0 is a form-factor, and

$$s(t) \equiv \frac{1}{NS} \sum_{j=1}^N \langle S_j^z(t) \rangle, \quad w(t) \equiv \frac{1}{N^2 S^2} \sum_{i \neq j}^N \langle S_i^+(t) S_j^-(t) \rangle. \quad (15)$$

If the wavelength is larger than the sample linear size, then $\varphi_0 \simeq 1$. But, when the wavelength is shorter than the system linear size, then the form-factor essentially depends on the sample shape [39, 40].

We have accomplished computer simulation for N magnetic nanomolecules possessing spin $S = 10$, such as Mn_{12} or Fe_8 , employing the parameters typical of these nanomolecules, for which D_2 and D_4 are negligible. The spin system is prepared in a nonequilibrium initial state, with the external magnetic field directed along the initial spin polarization, so that the spins tend to reverse to the opposite direction. It is convenient to consider a dimensionless radiation intensity, expressed through the units of

$$I_0 \equiv \frac{2\mu_0^2}{3c^3} \gamma_2^4 \quad \left(\gamma_2 \equiv \frac{1}{T_2} \right),$$

multiplied by the number of nanomolecules squared, N^2 , where $\mu_0 = -2\mu_B = 1.855 \times 10^{-20}$ erg/G and $\gamma_2 = 10^{10}$ 1/s, which gives $I_0 = 0.852 \times 10^{-38}$ W. All frequencies are measured in units of γ_2 . The resonance condition $\omega = \omega_0$ is assumed.

We have considered the influence of different factors on the radiation intensity. Thus, the role of the Zeeman frequency is exemplified in Fig. 1, showing that the larger the Zeeman frequency, the higher the radiation intensity. Figure 2 illustrates that the larger the initial spin polarization, the larger the radiation intensity. Figure 3 shows that increasing the magnetic anisotropy suppresses the radiation intensity. The role of dipole interactions is described in Fig. 4, demonstrating that they suppress the radiation intensity by a factor of 1.5. In Fig. 5, we study the role of the sample shape and its orientation, from where it follows that, under the same number of nanomolecules, the most favorable situation, with the highest radiation intensity, corresponds to the chain of nanomolecules along the resonator axis.

Calculations for the coherent radiation intensity (14) reduces to the solution of the evolution equation for the coherence function $w(t)$. Numerical solution yields the results close to the quasi-classical case, illustrated in Figs. 1 to 5. This is not surprising, since the coherent regime is known to be well represented by a quasi-classical approximation. The maximal number of coherently radiating spins can be estimated as $N_{coh} \sim \rho V_{coh}$, where ρ is the density of nanomolecules and V_{coh} is the coherence volume. The latter, for a cylindrical sample, is $V_{coh} \sim \pi R_{coh}^2 L$, where L is the cylinder length and R_{coh} is a coherence radius [39], which is of order $0.3\sqrt{\lambda L}$. This gives

$$N_{coh} \sim \rho \lambda L^2.$$

The typical density of magnets, formed by nanomolecules, is $\rho \approx 0.4 \times 10^{21}$ cm⁻³. For the Zeeman frequency $\omega \sim 2 \times 10^{13}$ 1/s, the wavelength is $\lambda \sim 10^{-2}$ cm. If $\lambda \sim L$, then $N_{coh} \sim 10^{14}$. The typical time of a superradiant pulse is 10^{-11} s.

In this way, magnetic nanomolecules and nanoclusters can be described by a similar macroscopic Hamiltonian. In the process of spin reversal from an initially prepared non-equilibrium state, there appears spin superradiance, due to the Purcell effect of the resonator feedback field. The radiation is mainly absorbed by the resonant coil surrounding the sample.

Acknowledgments

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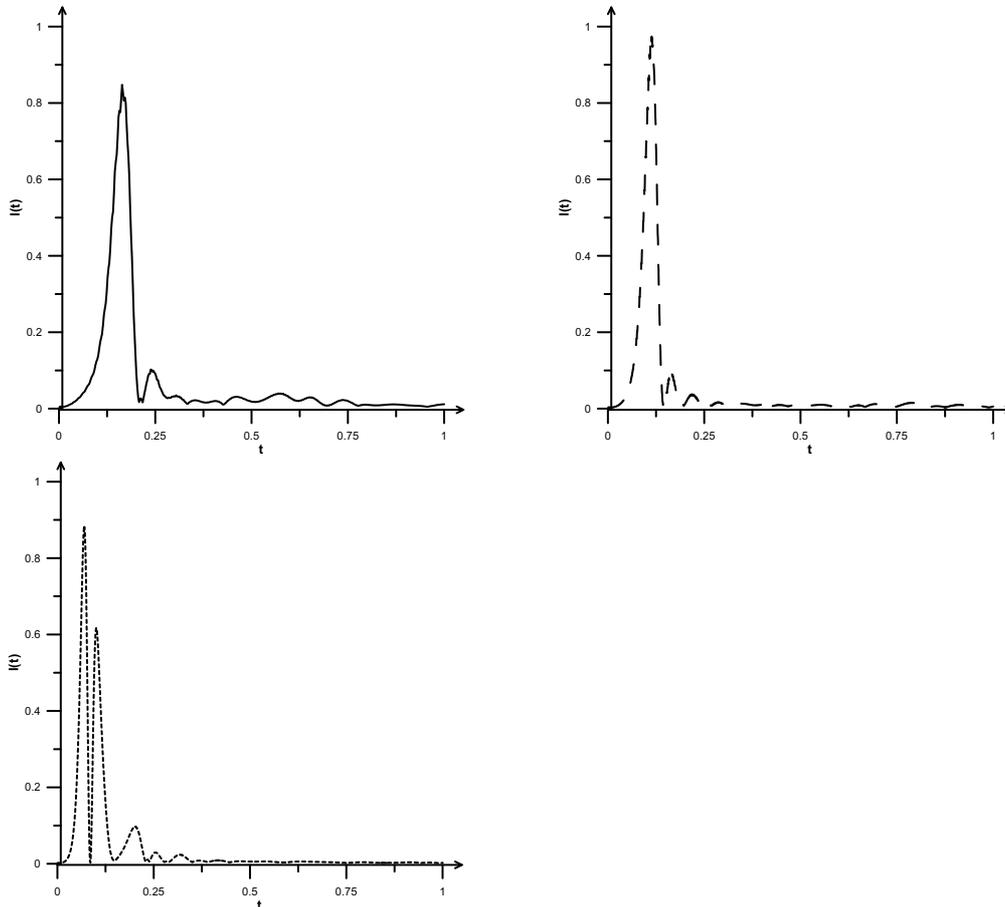


Figure 1. Radiation intensity (11) from $N = 125$ nanomolecules, with molecular spin $S = 10$, for a cubic sample. Initial reduced polarization is $s_0 = 0.9$, the anisotropy frequency is $\omega_D = 20$, and the resonator damping is $\gamma = 10$. The Zeeman frequency is $\omega_0 = 1000$ (solid line), with the intensity in units of $3.2 \times 10^{13} N^2 I_0$; $\omega_0 = 2000$ (long-dashed line), in units of $0.8 \times 10^{15} N^2 I_0$, and $\omega_0 = 5000$ (short-dashed line), in units of $5.1 \times 10^{16} N^2 I_0$.

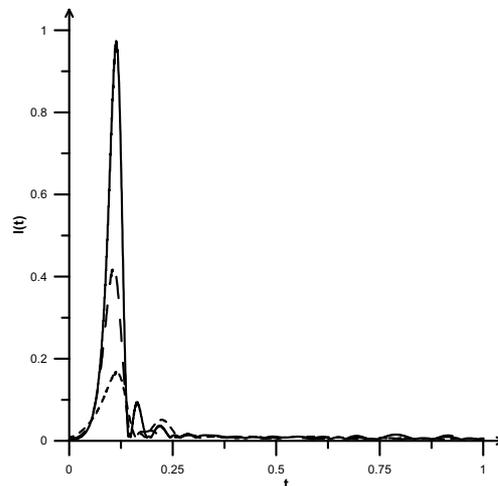


Figure 2. Radiation intensity (11) for a cubic sample of $N = 125$ nanomolecules, with spin $S = 10$, under the Zeeman frequency $\omega_0 = 2000$, anisotropy frequency $\omega_D = 20$, and the resonator damping $\gamma = 10$, for different initial polarizations: $s_0 = 0.9$ (solid line), $s_0 = 0.7$ (long-dashed line), and $s_0 = 0.5$ (short-dashed line). All intensities are in units of $0.8 \times 10^{15} N^2 I_0$.

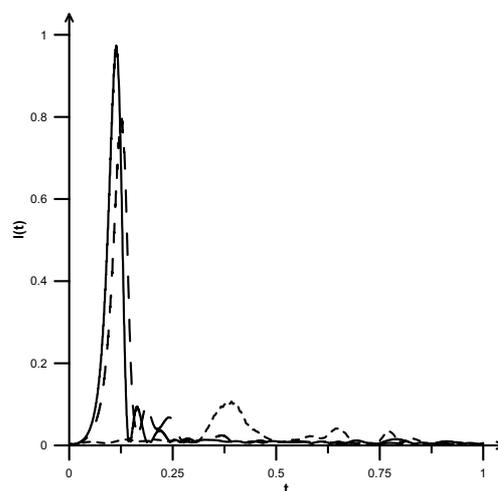


Figure 3. Radiation intensity (11) for a cubic sample of $N = 125$ nanomolecules, with spin $S = 10$, under the Zeeman frequency $\omega_0 = 2000$, resonator damping $\gamma = 10$, and the initial spin polarization $s_0 = 0.9$, for varying anisotropy frequency: $\omega_D = 20$ (solid line), $\omega_D = 50$ (long-dashed line), and $\omega_D = 100$ (short-dashed line). The values of the radiation intensity are in units of $0.8 \times 10^{15} N^2 I_0$.

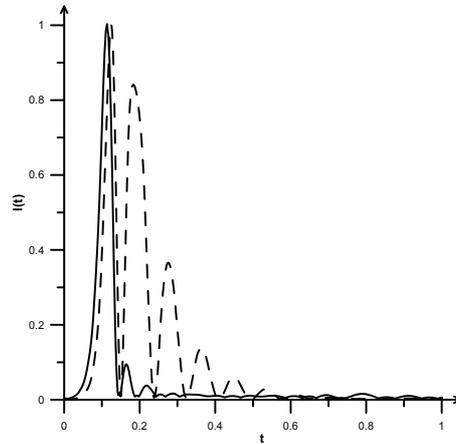


Figure 4. Radiation intensity (11) for a cubic sample of $N = 125$ nanomolecules, with spin $S = 10$, the Zeeman frequency $\omega_0 = 2000$, anisotropy frequency $\omega_D = 20$, resonator damping $\gamma = 10$, and the initial spin polarization $s_0 = 0.9$, for the cases with dipole interactions (solid line), in units of $0.8 \times 10^{15} N^2 I_0$, and without these interactions (dashed line), in units of $1.2 \times 10^{15} N^2 I_0$.

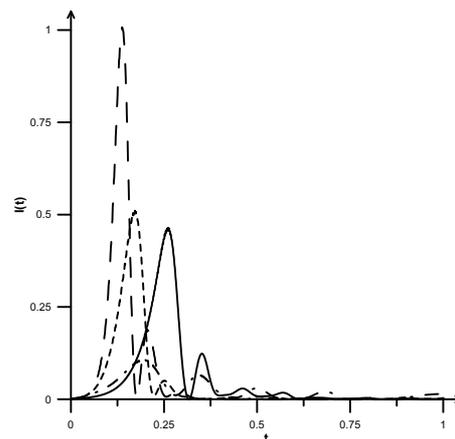


Figure 5. Radiation intensity (11) for a cubic sample of $N = 144$ nanomolecules, with spin $S = 10$, the Zeeman frequency $\omega_0 = 2000$, anisotropy frequency $\omega_D = 20$, resonator damping $\gamma = 30$, and the initial spin polarization $s_0 = 0.9$, for different sample shapes and orientations: the chain of molecules along the z - axis (solid line), the chain along the x - axis (long-dashed line), the $y - z$ plane of molecules (short-dashed line), and the $x - y$ plane of molecules (dotted-dashed line). The intensities are in units of $1.2 \times 10^{15} N^2 I_0$.

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