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Terahertz radiation from crystals of nanomagnets

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Abstract. Certain crystals, consisting of molecules with unusually large spin, exhibit macroscopically observable signatures of quantum tunneling, when a slowly varying external magnetic field is applied parallel to the easy axis of the crystal. Recently it has been observed that jumps in the magnetization are sometimes accompanied by the emission of infrared radiation. We discuss the connection of the tunneling with the electromagnetic transition, and we address the questions: to what extent can the radiation be considered as a collective, superradiant emission, and what is the role played by the cavity in the experiments? Our conclusion is that among the reported experimental conditions the radiation is not superradiance, but rather a maserlike effect.

An increasing attention has been attracted in the last few years by a class of complex molecular crystals also called nanomagnets, which at sufficiently low temperatures exhibit hysteresis steps [1–3]. The observations can be explained by assuming that the molecules in the crystal have an unusually large spin, in case of a typical representative of these materials, usually abbreviated as Mn₁₂Ac [1–3], the value of $S = 10$. This crystal also shows an uniaxial magnetic anisotropy: in the presence of an external magnetic field of the same direction as the preferred easy (z) axis, the properties of the molecules in the crystal can be described in the first approximation by an effective magnetic Hamiltonian of the form:

$$H_0 = -DS_z^2 - FS_z^4 - \tilde{\mu}B_0S_z. \quad (1)$$

H_0 is diagonal in the eigenbasis $\{|m\rangle\}$ of the (dimensionless) z component of the spin operator, S_z : $E_m = -Dm^2 - Fm^4 - \tilde{\mu}B_0m$, where $\tilde{\mu} = g\mu_B$, the gyromagnetic factor ($g = 1.94$ for Mn₁₂Ac) times the Bohr magneton. In the case of Mn₁₂Ac the values of the parameters are $g = 1.94$, $D/k_B = 0.55K$, $F = 1.2 \cdot 10^{-3}K$. (We note that there is about a 5% uncertainty in these values depending on the individual samples.) This means that in the absence of B_0 the two lowest degenerate eigenvalues with $m = -m' = 10$ are separated by a barrier of height $100D$ which is insurmountable at low temperatures. This remains valid also for moderate values of B_0 , so there is no transition between the two valleys. Simple algebra shows that eigenvalues become doubly degenerate with given m and m' at the following values of B_0 :

$$\tilde{\mu}B_0 = -D(m + m') \left(1 + \frac{F}{D} [m^2 + m'^2] \right). \quad (2)$$

The hysteresis jumps can be explained by transitions between equal energy levels at these crossing points, which must be induced by a coupling between the levels. There is experimental evidence that the coupling does exist and can be described by a Hamiltonian:

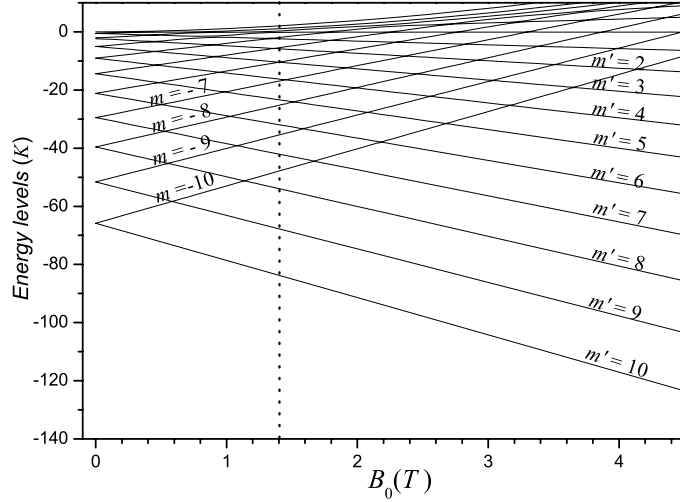


Figure 1. The energy levels of Hamiltonian $H_0 + H_1$ as a function of the external magnetic field B_0 . The vertical line at $B_0 = 1.4$ T indicates the field value where microwave radiation were observed.

$$H_1 = C(S_+^4 + S_-^4) + E(S_+^2 + S_-^2)/2 + K(S_+ + S_-)/2. \quad (3)$$

In contrast to thermally activated processes, the transition generated by H_1 is a quantum tunneling effect. If one diagonalizes the Hamiltonian $H_0 + H_1$ in the vicinity of the crossing points one obtains in each two-dimensional subspace spanned by the crossing levels $\{|m\rangle, |m'\rangle\}$ the following matrix for the effective Hamiltonian:

$$H_e(t) = \begin{pmatrix} \varepsilon_0 + w/2 & \Delta_0/2 \\ \Delta_0/2 & \varepsilon_0 - w/2 \end{pmatrix}_{m,m'} \quad (4)$$

Here ε_0 is the energy where the given crossing would occur, w is proportional to the time dependent external field in the z direction, while the offdiagonal element Δ_0 is the level splitting responsible for the effective coupling between the levels. This means, as usual, that instead of the crossing one has a splitting of the levels, also called anticrossing. The magnetic field B_0 is assumed to be linear in time with constant \dot{B}_0 , yielding $w = -\tilde{\mu}\dot{B}_0(t - t_0)(m - m')$ with t_0 being the time instant when the crossing would occur. This linear approximation corresponds to the usual Landau-Zener-Stückelberg (LZS) model [4–6], by the aid of which we can calculate the probability of a given $m \rightarrow m'$ transition, $P_{mm'} = 1 - \exp(-\pi\Delta_0^2/2\hbar w)$. The situation is similar to crossing points of molecular vibrations where transitions and wave packet dynamics have been extensively studied [7,8].

In accordance with the experiments we shall assume that initially all the atoms are on one side of the potential barrier. It turns out that for low lying levels $|m|, |m'| > 6$, the transition probabilities $P_{mm'}$ are very small, therefore after the crossing points a significant inversion may appear between certain levels. This fact raised the possibility that the inversion emerging in this way can lead to an electromagnetic emission [9], and the question was put whether this emission could be one where the phases of all the individual emitters are coupled leading to a very fast superradiant pulse [9]. Experiments where samples of crystals were put into a metallic cavity show that such emission really takes place in the frequency domain around 100 GHz [10–12]. The effect is much more involved than proton spin resonance in a cavity [13–16], where the

spin in question has only two stationary states. Studies in SR with other physical systems show that the presence of a resonant cavity may enhance the collectivity of the radiating individual dipoles, as first proposed by Bloembergen and Pound [17], and which seems to be necessary to obtain radiation in the case of molecular magnets, as well. [18, 19]. This can be the reason why in an another experiment [20], no sign of radiation was found.

In order to be able to describe the experimental situation we need to take into account effects caused by the cavity as well. For the sake of simplicity we consider the microwave cavity field as a single transverse (TM) mode being perpendicular to the z axis and having a frequency Ω and equal amplitudes in the x and y directions. A given molecule interacts with the magnetic part of this classical field the amplitude of which is denoted by \mathcal{B} via the Hamiltonian $H_I = -\tilde{\mu}\mathcal{B}(\tilde{S}_x + \tilde{S}_y)/\sqrt{2}$, where \tilde{S}_x and \tilde{S}_y are the spin operators acting in the corresponding two dimensional subspace determined by the chosen m and m' [21]. Without relaxation the time evolution of a molecular density matrix ϱ corresponding to the actual level pair is given by

$$\frac{\partial \varrho}{\partial t} = -\frac{i}{\hbar} [H', \varrho], \quad H' = H_e + H_I = \begin{pmatrix} \varepsilon_0 + \hbar\omega/2 & \Delta/2 \\ \Delta^*/2 & \varepsilon_0 - \hbar\omega/2 \end{pmatrix}_{mm'}, \quad (5)$$

were $\hbar\omega(t) = w(t) - 2\tilde{\mu}\mathcal{B}s'$ and $\Delta = \Delta_0 - 2\tilde{\mu}\mathcal{B}s$ with s' and s being the diagonal and offdiagonal elements resulting from the operator $(\tilde{S}_x + \tilde{S}_y)/\sqrt{2}$ that couples the molecular system to the cavity mode.

The dominant relaxation effect in this system is due to spin phonon coupling, i.e, the oscillation of the atoms in the lattice. There is another important depahasing mechanism, near field dipole-dipole coupling, which is always present independently from the temperature. We note that at very low temperatures the near field dipole-dipole interaction can be the main source of decoherence, but the temperature range around 2 K, where the experiments were performed, is well above this regime. Similarly to numerous system-environment interaction models [22–25], the coupling to the phonon modes leads to both dissipation and phase relaxation, but the former process has a much longer time scale thus it can be omitted here: We consider only the transversal relaxation leading to the decay of the offdiagonal elements of the molecular density matrix ϱ . In the present approach, it will be taken into account by assuming a simple exponential decay with a time constant T_2 .

We also treat the mode amplitude of the cavity as a dynamical quantity. In semiclassical approximation, the time varying cavity field \mathcal{H} will be described here to be generated by the magnetic dipole moment density of the crystal, \mathcal{M} as a source. Maxwell's equations lead

$$\Delta\mathcal{H} - \dot{\mathcal{H}}/(c^2T_c) - \ddot{\mathcal{H}}/c^2 = \ddot{\mathcal{M}}/c^2, \quad (6)$$

where T_c is the cavity lifetime. Now, as Ω is in the terahertz domain, we can assume that oscillations with this frequency are faster than any other time scale in the system, i.e., we can apply rotating wave approximation (RWA) to obtain dynamical equations for the slowly varying quantities: H , R and Z , where $\varrho_{mm'} = Re^{-i\Omega t}$, $Z = \varrho_m - \varrho_{m'}$ and $\mathcal{H} = \left(\frac{1}{2}H^{-i\Omega t} + c.c.\right) u_k(z)$ with $u_k(z)$ being the mode function. The characteristic time of the problem is given by

$$T_0 = \left(\frac{2\hbar}{\eta N_0 \Omega \mu_0 \tilde{\mu}^2 |s|^2}\right)^{1/2}, \quad (7)$$

where the cavity filling factor η is essentially the ratio of the length of the sample and the length of the cavity to a very good approximation. By the aid of T_0 we can finally write the dynamical equations under RWA in the following dimensionless form:

$$\frac{d}{d\tau} Z = -i(b^* R e^{-i\psi} - b R^* e^{i\psi}), \quad (8)$$

$$\frac{d}{d\tau}R = -iT_0(\omega(\tau) - \Omega)R - ibe^{i\psi}Z - \gamma R, \quad (9)$$

$$\frac{d}{d\tau}h = -\frac{\kappa}{2}h + iRe^{-i\psi}, \quad (10)$$

where $\tau = t/T_0$, $\gamma = T_0/T_2$, $\kappa = T_0/T_c$ and the dimensionless field amplitude is given by:

$$h = \left(\frac{\mu_0}{N_0\hbar\Omega} \right)^{1/2} H. \quad (11)$$

We note that the dimensionless B and H fields have been identified in Eq. (8,9) which is a very good approximation in this case. The field intensity can be measured as the energy density averaged out over the time and space period. Its dimensionless form $I = |h|^2/2$ gives the number of emitted photons of energy $\hbar\Omega$ per number of molecules participating in the given transition. We also introduce the dimensionless sweep rate via

$$T_0(\omega(\tau) - \Omega) = v\tau, \quad (12)$$

where the origin of the time axis is chosen so that $\tau = 0$ corresponds to exact resonance: $\omega(0) = \Omega$.

Now we shall analyze the model described above in comparison with experimental findings. We solve the differential equations (8,9,10) that describe the time evolution of the molecular system coupled to the cavity mode. Crucial roles are played here by the relaxation rate γ , the coupling coefficient s and the sweep rate v . The parameter γ determines the nature of the emitted radiation, while the time spent by the system around resonance depends on v . Note that for a given physical setup T_0 is inversely proportional to $|s|$. The intensity I of the emitted radiation as well as the inversion Z can be seen in Fig. 2 as a function of time for different dephasing rates γ . As we can see, the emission process starts around resonance $\tau = 0$, and for weak dephasing several oscillations in Z and I can be seen indicating energy exchange of the molecular system and the cavity field. The frequency of these oscillations increases, reflecting the time dependent separation of the levels. Finally, due to the relaxation effects, the amplitude of the oscillations diminishes, Z reaches its stationary value and the emission stops. As it can be expected, for strong dephasing molecular coherence plays no role, rate equations with time dependent detuning can be used to describe the dynamics [21]. Analyzing the time scales characteristic to the experiments (T_0 has the order of $10^{-4}s$ and T_2 is assumed to be around 10^{-5} – 10^{-6} s, see Ref. [26]), we find that this kind of maser-like radiation can be the reason for the observed bursts of radiation.

The magnitude of the emitted energy was found to be around $3 nJ$ in a recent experiment [12], where radiative bursts of duration of a few milliseconds were observed at $B_0 = 1.4$ T and temperature of 2 K. Using the parameters of this experiment we investigated all the possible transitions at $B_0 = 1.4$ T and found the best agreement with the experimental data for the transition $m = -8 \rightarrow m' = 6$, giving a value of T_0 in the ms range and a total emitted energy to be around $1.5 nJ$. Note that for initial states below $m = -8$ the time scale of the process turns out to be too long, while for $m > -8$ the number of active molecules is too small. Thus our model predicts that the process having the most important role in producing the observed radiation is the transition $m = -8 \rightarrow m' = 6$.

In conclusion, we investigated the interaction of a crystal of molecular magnets with the magnetic field of a surrounding cavity. The sample itself generates this transversal field, while it also acts back on the molecules. The most important point of our treatment is that the cavity mode with fixed frequency Ω comes to resonance with a magnetic transition at a given value of the external longitudinal magnetic field. Around this resonance the interaction of the

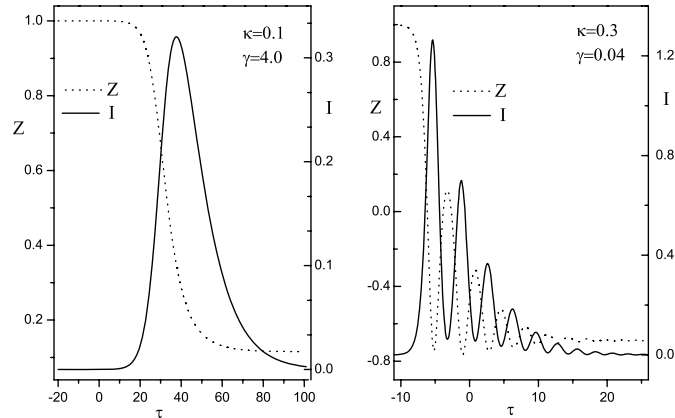


Figure 2. Population difference Z and the dimensionless intensity I of the radiation emitted by the molecular system as a function of τ for different relaxation rates. The dimensionless sweep rate corresponding to these plots is $v = 0.1$.

molecules with the mode significantly increases leading to an observable burst of electromagnetic radiation. Comparing the parameters of our model to those of the experiments, we concluded that the observed radiation at 2 K is not superradiance but similar to a maser effect, where the detuning of the transition and the field mode is time dependent. We identified the transition that plays the most important role in the emission process. It has been found that our model gives satisfactory agreement with experimental results.

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