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Quantum dynamics of crystals of molecular magnets inside microwave resonators

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Abstract

It is shown that crystals of molecular nanomagnets exhibit enhanced magnetic relaxation when placed inside a resonant cavity. Strong dependence of the magnetization curve on the geometry of the cavity has been observed, providing evidence of the coherent microwave radiation by the crystals. These observations open the possibility of building a nanomagnetic microwave laser pumped by the magnetic field.

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Molecular clusters of Mn_{12} and Fe_8 exhibit spectacular magnetic effects related to the quantization and quantum tunneling of the magnetic moment. The current great interest in these systems was ignited by the discovery of resonant spin tunneling in Mn_{12}-acetate (Mn_{12}Ac) [1]. This phenomenon is understood considering the Hamiltonian that describes the tetragonal symmetry of this high-spin ($S = 10$) molecule [2]. The strong uniaxial anisotropy of this molecule yields a double well configuration, the two energy minima of which correspond to the spin up and down orientations and are separated by the magnetic anisotropy barrier. In zero magnetic field, the ground state is doubly degenerate. In non-zero magnetic field, the degeneracy is broken and transitions between spin states in the two wells occur at certain resonant intensities by the combination of thermal activation and quantum tunneling [3,4]. The activation of molecules is believed to be due to phonon absorption, but in certain experimental conditions EPR experiments have demonstrated that photon absorption can also be responsible for that [5,6].

Most recently, Chudnovsky and Garanin have suggested that a crystal of molecular nanomagnets can also emit coherent microwave radiation during magnetic relaxation, what is called superradiance [7]. Experimental support to this suggestion is given here by demonstrating the strong interaction between a Mn_{12} crystal and a microwave cavity.

We used in our experiments 2-mm-long 0.5-mm-wide Mn_{12}Ac single crystals that were fixed in a vertical position at the bottom of a 99.99%-pure-copper-made cylindrical cavity of 1.6 mm diameter and adjustable length. A micrometric stepping motor control system that had a spatial resolution of 1μm was used for this purpose. The inner lateral and planar surfaces of the cavity were polished to a roughness of less than 10 μm to achieve a quality factor Q between 10^3 and 10^4. The cavity containing the crystal was immersed in a helium gas inside an MPMS SQUID magnetometer. The temperature inside and outside the cavity was monitored by carbon thermometers.

Before placing the crystals inside the cavity the magnetic signal from each was measured independently.

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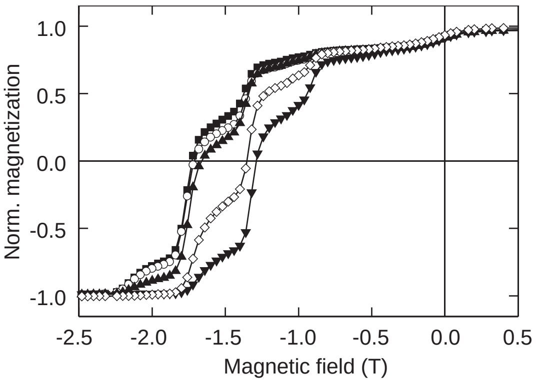

Fig. 1. Demagnetization curves of a $Mn_{12}Ac$ single crystal inside a 1.6 mm diameter resonant cavity at 2 K for five different lengths ranging from 21.0 mm (topmost curve) to 19.5 mm (lowest curve).

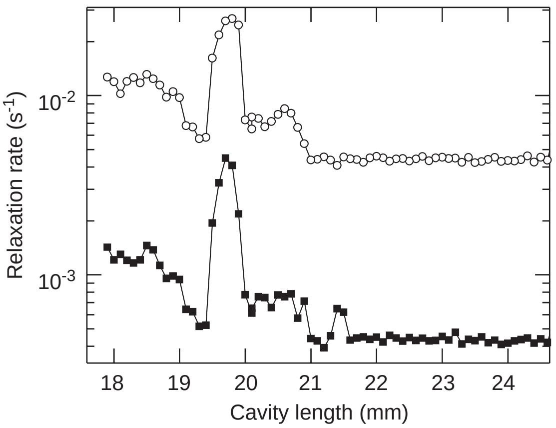

Fig. 2. Relaxation rate of a $Mn_{12}Ac$ single crystal inside a 1.6 mm diameter resonant cavity at 2 K as a function of the cavity length at the second (lower curve) and third (upper curve) resonant fields.

The signal from the crystal was always two orders of magnitude greater than the paramagnetic signal from the cavity. We then proceeded to measure the crystals inside the cavity. The system was first saturated by a 50 kOe field applied along the *c*-axis of the crystal. Then the field was swept in the opposite direction at the rate 40 Oe/s. Typical demagnetization curves thus obtained, with resonances (large jumps) at $H_R \sim 0, 9$ and 14 kOe, are shown in Fig. 1. The dependence of the relaxation rate calculated as $\Gamma = |M(H) - M_{eq}(H)|^{-1}(dM/dH)$ (where M_{eq} is the equilibrium magnetization), on the length of the cavity at the second ($H_R \sim 9$ kOe) and third ($H_R \sim 14$ kOe) resonance fields is shown in Fig. 2. All these data were reproducible within a 6% experimental error.

The experimental data demonstrate the dependence of the magnetic relaxation in $Mn_{12}Ac$ crystals on the geometry of the cavity. We verified that this phenomenon appears only in cavities of high quality factor. To disregard possible effects due to thermal heating the temperature of the cavity was carefully monitored. The temperature variation was found not to exceed a 0.3%, which is a small number that cannot account for any measurable change in the relaxation rate. Strong dependence of the relaxation rate on the geometry of the confinement was also observed when the crystals were placed between superconducting mirrors and so the helium gas circulated freely around the crystal [2]. The observed phenomenon had therefore nothing to do with thermal effects and was actually due to the microwave properties of the cavity.

According to this, in order to affect the magnetic relaxation of the $Mn_{12}Ac$ crystal, the cavity should acquire a very large number of non-thermal photons. There is only one source of these photons, the crystal itself. To change the orientation of the magnetic moment, the molecule must first go up the staircase of discrete spin levels lying in one of the two energy minima and then go down a similar staircase in the second energy minimum. The magnetic relaxation in the crystal creates therefore a massive inversed population of the energy levels. These excited states decay via emission of phonons or photons. Without cavity, the corresponding photon transitions would have a negligible probability compared to the probability of phonon transitions. Consequently, the relaxation towards thermal equilibrium would occur via the emission of phonons.

Inside the cavity, however, a maser-like effect can take place if some of the frequencies of the emitted photons coincide with resonances of the cavity. The photon emitted by one molecule remains in the cavity and stimulates the emission of photons by other molecules. The shortest photon wavelength, λ, corresponds to the decay transition between the two lowest spin levels in the spin down energy minimum. For $Mn_{12}Ac$ in zero field, this wavelength is about 1 mm, but the wavelengths of photons emitted in other transitions are longer. Consequently, for a crystal of size 2 mm, the phase of the emitted photons is the same for a macroscopically large number of molecules, $N = N_{SR} \sim (\lambda/2)^3$. In this case, the photon emission rate increases by a factor N_{SR}, the emission of photons by different molecules becomes correlated and the superradiance may occur [7–9].

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