

Superradiance Experiments In Magnetism

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In this talk we discuss low temperature demagnetization data in molecular magnets for which the most plausible interpretation is the emission of microwave superradiance (1-4).

Molecular clusters are nanomagnets showing important phenomena associated to the fact that it is possible to tune its quantum mechanics properties at low temperature and by applying an external magnetic field. The discovery ten years ago of the resonant spin tunneling between the degenerate spin levels at both sides of the magnetic anisotropy barrier height (5, 6) was the first sign that quantum mechanics can reveal itself in these magnetic units made of several hundreds of atoms. Since then, near thousand papers on molecular magnets have been published (7).

The two types of experiments discussed here refer first to the analysis of the electromagnetic radiation accompanying the fast demagnetization process in molecular magnets, and second to the results of new experiments performed to better understand the spin tunneling transitions in the case of extremely high sweep rate of the bias magnetic field. As a matter of fact, by discussing the results of these two experiments we are in a position to better connect the behaviour of our magnetic units as qubits and the superradiance emission. The case is that the most striking effect for decoherence in magnetic qubits may be the superradiance emission.

The materials used in our experiments were always a set of single crystals to have in all cases more than 10^{18} molecules to cooperate in the coherent decay process. The main idea is the following: the decay process of the spin levels of the molecular magnets is mainly mediated by phonons as the phonon density is much higher than the photon density. In order to have the emission of radiation, the number of molecules cooperating coherently should be at least as high as the ratio between the transition probability via the emission of photons and phonons.

The bias magnetic field producing the magnetization/demagnetization process was changed at a speed such to produce the so-called thermal avalanches in the single crystals. The tunneling process at a certain resonant field is thus accompanied by the emission of phonons which excite molecules to tunnel and produce more phonons when decaying to the ground state. This very non linear effect produces the population inversion in a time of the order of 1 ms. The population inversion is followed by the tunneling of most of the molecules from the same spin level of the metastable well to the same spin level in the stable well. This tunneling process is followed by the staircase process to the ground spin state. Experimentally we detect: 1) how much magnetization is tunneling, 2) the time of the duration of the spin tunneling, 3) the change in the temperature of the sample after the thermal avalanche, 4) the time when the emission of radiation occurs, and 5) the power of the emitted radiation.

The main aim of the experiments performed at ultra high sweep rates of the bias magnetic field was to test the Landau-Zener transition probability in the spin tunnelling case. That is, it is well established that single quantum transitions obey the Landau-Zener law. In our experiments, however, we have seen that this law is no longer obeyed and interpreted it as due to the simultaneous transition of a huge number of spin levels. This result provides strong evidence for the occurrence of the superradiance emission. These experiments are also crucial to better understand the qubit behaviour of the magnetic

clusters when working as logic gates. Let us consider a pair of tunnel-spin levels ε_+ and ε_- versus the bias energy $W = \varepsilon_m - \varepsilon_{m'} = g |m - m'| \mu_B (H(t) - HR)$, where m and m' are the initial and final state, HR denotes the resonance field, and W denotes the energy difference between the two spin levels the tunneling transition is expected to occur in between. It has been theoretically discussed that the individual LZ spin tunneling transitions occurs at the value of the magnetic field $H(t) = HR$. The total magnetization reversal occurs after crossing the resonance $H(t) > HR$. That is, the first stage is the LZ process that leaves the fraction of magnetic molecules P in the excited states (the upper energy branch ε_+). This fraction is given by the Landau-Zener formula: $PLZ = \exp(-\varepsilon)$, where $\varepsilon = \pi\Delta^2/2\hbar v$, being Δ the tunnel splitting and v the energy sweep rate, $W = v t$. During the second stage, these excited states decay due to the Dicke superradiance onto the lower branch ε_- . At high sweep rate r , the majority of the molecules initially cross to the upper branch and then decay simultaneously to the lower branch due to superradiance. If this were the case, the magnetic susceptibility should obey the scaling $rl/2 (dM/dH)$ versus $\delta H/rl/2$, where $\delta H = H(t) - HR$. In our experiments we measured the differential magnetic susceptibility associated to the total reversal of the magnetic moment as a function of the sweep rate for the case of using the Mn12 single crystals. The conclusion of this work is that we have found a new spin relaxation effect which has been tested using both single crystals of Mn12 and Fe8. The magnetization reversal occurs at fields lower than those occurring at very slow sweep rate. The observed dependence of the differential susceptibility on the sweep rate correlates well with the theory of collective electromagnetic transitions.

In conclusion, these two set of experimental results are of very high practical importance besides their novelty in science. The superradiance emission may be the most important decoherence phenomenon when operating with magnetic qubits at large scale.

References:

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