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CONTROL OF QUANTUM CORRELATIONS IN TAILORED MATTER
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Kolloquium

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Phase-transitions in crystals of molecular nanomagnets

Crystals made of large molecular clusters are ideal candidates to investigate the long-range magnetic ordering induced by dipole-dipole interactions alone, of which only a few examples exists in Nature. Observing experimentally such transitions is however not easy, since these molecular clusters exhibit strong uniaxial anisotropy that leads to hysteresis phenomena at low temperatures. Consequently, the spin reversal can only take place via quantum tunneling processes, which gives rise to a very slow relaxation towards the thermal equilibrium spin configuration. In recent years, we have tried to tackle this problem following three different strategies that will be described in the talk [1-3]. The simplest option is to study isotropic clusters having very small magnetic anisotropy. One of these, Mn6, remains in equilibrium down to very low temperatures and shows signatures of a phase transition to long-range ferromagnetic order at T_c = 0.16 K [1]. Relaxation to equilibrium via quantum tunneling is also able to bring the spins to thermal equilibrium provided the tunnel splittings are sufficiently large. This was achieved in clusters of Mn4 by chemical means, lowering the symmetry of the molecules and therefore enhancing those terms in the spin Hamiltonian that induce tunneling [2]. In crystals of well oriented molecules, the tunneling rates can be enhanced at will by the application of an external magnetic field perpendicular to the anisotropy axes. Using neutron diffraction, we have observed the onset of a spontaneous magnetization in a single crystal of Mn12 acetate that sets in below a critical temperature $T_c \approx 0.8$ K. Curiously enough, if the field-induced zero-point fluctuations become sufficiently strong with respect to dipole-dipole interactions, this spontaneus magnetization is suppressed. This points to the existence of a quantum critical point in crystals of mesoscopic-size clusters [3].

References
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