Collective Relaxational Dynamics in Molecular Nanomagnets

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Although molecular solids based on nanomagnets such as Fe_8 and Mn_{12} display many quantum phenomena that can be understood at the level of one molecule, and are often called single-molecule-magnets for that reason, they also display other dynamical behaviours that require going beyond the single-molecule description. Examples include relaxation in zero field from a magnetized state, magnetization reversal in a swept field (the Landau-Zener-Stuckelberg protocol), and magnetization in nonzero field starting from a nonmagnetized state. Here one sees nonexponential behaviour in time, characteristic of glassy dynamics. It is found essential to consider decoherence in understanding the dynamics of one molecule, and the dipole-dipole interaction between them in understanding the dynamics of the solid as a whole. We have performed a detailed study of the decohering effects of nuclear spins and of the dipole-dipole interaction, showing how quantum mechanical tunneling between deep levels of one molecule is transformed into incoherent relaxation, and providing good justification for a classical model of relaxation for the solid as a whole [1]. In this model the relaxation probability of a given molecular spin is strongly dependent on the local field seen by that molecule, and because of the dipole-dipole interaction's long range and enormous value compared to the tunnel splitting and nuclear spin magnetic field, the relaxation of any one molecule alters the field seen by many other molecules. We have performed Monte Carlo simulations of this model, and also developed and solved rate equations and kinetic equations to compare with the simulations and the experiments [2, 3]. The agreement between simulations, kinetic equation, and experiments is very good in most respects, but not so good for ultra long times and ultra-slow phenomena. The problem of magnetization is particularly interesting, as at first sight it entails the relaxation of energy in addition to spin, whereas the as originally conceived model has no provision for energy relaxation per se. However, a modification of this model including a secondary relaxation mechanism due to phonoemissive tunneling [4] does not appear to be satisfactory. Thus, the magnetization problem remains open. Work is currently underway to see if it is possible to achieve energy relaxation from the original model without building it in at the single molecule level.

References:

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