Annals of Physics 356 (2015) 37–56

Contents lists available at ScienceDirect

Annals of Physics

journal homepage: www.elsevier.com/locate/aop

Low-temperature magnetization dynamics of magnetic molecular solids in a swept field

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ARTICLE INFO

Article history:
Received 14 November 2014
Accepted 8 February 2015
Available online 17 February 2015

Keywords:
Molecular magnet
Magnetic relaxation
Swept field
Monte Carlo method
Kinetic equation

ABSTRACT

The swept-field experiments on magnetic molecular solids such as Fe_8 are studied using Monte Carlo simulations, and a kinetic equation developed to understand collective magnetization phenomena in such solids, where the collective aspects arise from dipole–dipole interactions between different molecules. Because of these interactions, the classic Landau–Zener–Stückelberg theory proves inadequate, as does another widely used model constructed by Kayanuma. It is found that the simulations provide a quantitatively accurate account of the experiments. The kinetic equation provides a similarly accurate account except at very low sweep velocities, where it fails modestly. This failure is attributed to the neglect of short-range correlations between the dipolar magnetic fields seen by the molecular spins. The simulations and the kinetic equation both provide a good understanding of the distribution of these dipolar fields, although analytic expressions for the final magnetization remain elusive.

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1. Introduction

As a prototype of magnetic molecular solids, the one generally known as Fe_8 has drawn great interest. This material consists of molecules of $[Fe_8O_2(OH)_{12}(tacn)_6]Br_8(H_2O)_9$, in which the Fe(III) ions of one molecule are well separated from those of a neighboring molecule. At low temperatures, each molecule has a spin $S = 10$, a corresponding all-spin magnetic moment of magnitude $g\mu_B S$ with $g \simeq 2$, and an Ising-like anisotropy which translates into an energy barrier of 22 K [1,2]. Among

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http://dx.doi.org/10.1016/j.aop.2015.02.006
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the many experimental investigations of this molecule, the swept-field experiment of Wernsdorfer and Sessoli [3] is one of the most revealing. A partial and simplified description of this experiment is as follows. At low temperatures ($T \lesssim 100$ mK) they first saturate the magnetization of the sample by applying an external magnetic field H_z along the Ising or z axis, and also apply a magnetic field H_x along the hard magnetic axis transverse to the easy axis. They then sweep H_z so as to reverse the magnetization, and measure the rate at which the spins reverse. This rate turns out to be an oscillatory function of H_x, even though the energy barrier and the angle between the two energy minimizing orientations of the spin are both monotonically decreasing functions of H_x. Although surprising at first, the oscillations are now well understood. The simplest explanation for the magnetization reversal is that the sweeping of H_z induces Landau–Zener–Stückelberg (LZS) transitions [4,5] between the two lowest states on opposite sides of the energy barrier at a rate proportional to Δ^2, where Δ is the tunnel splitting between these states. The rate oscillates because Δ does so, and Δ in turn oscillates because there are two tunneling spin trajectories that interfere with a phase that varies with H_x [6–8].

Fe_8 is just one of $\sim 10^3$ magnetic molecular solids that are now known, and which have been the object of much study over the last two decades. Their main characteristics are that the spin of one molecule is large at low temperatures, and to a good first approximation, one may treat the spins on different molecules as non-interacting. (A comprehensive and authoritative review of the entire field is contained in Ref. [2]. Shorter reviews may be found in Refs. [9–11].) These solids are often known as single-molecule-magnets (SMM's), because many phenomena may be understood, at least qualitatively, in terms of the properties of the total ground state spin of a single molecule in a suitable crystal field, via an effective spin Hamiltonian that contains anisotropy terms reflecting the overall symmetry of the molecule and its local environment. The oscillatory tunnel splitting mentioned above is one such phenomenon. Another is that the hysteresis loops are sharply stepped, where the steps coincide with crossings of energy levels on opposite sides of the energy barrier [12,13]. This single molecule behavior has, unsurprisingly, led to suggestions and proposals for using these materials in devices [14], but they are a new class of magnetic materials and worthy of study in their own right for the novel phenomena they display.

A large variety of experimental tools has been employed to study the low temperature magnetization dynamics of such solids. Of these, the swept-field or Landau–Zener–Stückelberg (LZS) protocol has proven to be one of the most fruitful. When the sweep is sufficiently rapid, the accompanying change in the magnetization can be interpreted in terms of LZS transitions as already mentioned, and thereby provides a measurement of the tunneling amplitude between energy levels on opposite sides of a barrier. Tunnel splittings measured by this technique are as low as 10^{-8} K in temperature units. Such low splittings are beyond the reach of any other method. When the sweep rate is slow, on the other hand, the interpretation is not clear-cut. It is essential to consider the dipole–dipole interactions between different molecules, and the transition is influenced by the collective dynamics of all the spins. Indeed, in this case, the SMM designation falls short, and a more intricate analysis is called for.

Collective, dipole-coupled dynamics of the spins are also seen in several magnetization-relaxation type experiments [15–21]. Theoretical discussions and Monte-Carlo simulations of these experiments have been provided by [2,22–25], and many aspects of the experiments are understood. The same is not true of the swept-field experiments, and we are aware of only a few previous investigations that pertain to the collective dynamical aspects [26–28]. We discuss these below. Theories that include the dipole–dipole interactions in a purely static way [29], or via a dynamic mean-field [30] cannot explain all the experimental behavior, especially that at low sweep rates. It is important to have a more complete theory since experimentalists often interpret data in terms of the original LZS analysis [31–34]. In light of the fact that the spins in molecular magnets are subject to strong environmental influences, especially the dipolar couplings to other molecular spins, and that the LZS theory is based on fully quantum mechanically coherent time evolution, it is not at all clear that the LZS description is even applicable a priori. Indeed, as explained in Ref. [30], it is a fortunate fact that it can be used even at high sweep velocities. Wernsdorfer has argued [32], correctly in our view, that the LZS formula should not be used without first verifying that one is in the fast sweep limit, and that it can in general only yield a lower bound on the tunnel splitting. While the use of this formula may provide qualitative proof of the presence of a geometrical phase in the tunneling spectrum [8], one is on shaky ground if one uses the extracted tunnel splittings to build detailed models of intramolecular magnetic interactions. It seems

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