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Low-temperature phonoemissive tunneling rates in single molecule magnets

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ABSTRACT

Tunneling between the two lowest energy levels of single molecule magnets with Ising type anisotropy, accompanied by the emission or absorption of phonons, is considered. Quantitatively accurate calculations of the rates for such tunneling are performed for a model Hamiltonian especially relevant to the best studied example, Fe_8. Two different methods are used: high-order perturbation theory in the spin–phonon interaction and the non-Ising-symmetric parts of the spin Hamiltonian, and a novel semiclassical approach based on spin-coherent-state-path-integral instantons. The methods are found to be in good quantitative agreement with other, and consistent with previous approaches to the problem. The implications of these results for magnetization of molecular solids of these molecules are discussed briefly.

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1. Introduction and background

Single molecule magnets, also known as molecular nanomagnets, are a twenty-year old class of magnetic materials consisting of organomagnetic molecules that form molecular solids. Their general characteristics are a total spin of about 10 per molecule in the ground state, an absence of exchange interaction between different molecules, and magnetic anisotropy with an energy difference per

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molecule of tens of Kelvin between easy and hard directions [1,2]. The most studied systems, Fe_8 and Mn_{12}, both have spin magnitude j equal to 10, and uniaxial Ising-type anisotropy. At low enough temperatures, only the $m = \pm 10$ Zeeman levels are occupied. On theoretical grounds, transitions between these two levels can only take place via quantum tunneling, and for Fe_8 there is clear evidence that such tunneling really occurs [3], even though the tunneling matrix element is only 10^{-8} K or 1 peV in energy units. However, quantum tunneling with significant probability transfer between the states involved can only happen if the states are nearly degenerate in energy. If they are not, the tunneling degree of freedom (in this case the spin) must be coupled to an environment which can supply or absorb the energy necessary to maintain energy conservation. This fact, coupled with the general paucity of excitations with large energy, greatly restricts the dynamics of the *total* magnetization of the solid. Several experiments [4–11] find that magnetization relaxation is slow, with non-exponential behavior in time.

The current theoretical understanding of this slow relaxation [12,13] is that interaction of the molecular spins with the nuclear spins renders the quantum tunneling of the former incoherent, but because the nuclear spins that couple to a given molecular spin can exchange only a rather limited amount of energy, the requirement of near-degeneracy of the Zeeman levels of the molecular spin is weakened only moderately, and the two levels must lie within a narrow window of each other in order for transitions to occur. Further relaxation can only take place due to the intermolecular dipole field, which can be quite inhomogeneous. If this field happens to be such at a given molecular spin site as to bring that spin into near degeneracy, it will be able to flip. This flip will change the field at other sites, potentially allowing those spins to relax. Monte Carlo and kinetic equation studies based on this model have been done by several authors [14–19], all of whom obtain slow relaxation, and in some cases, an initial square-root time dependence as seen experimentally.

A central feature of the above model is that the transition rate between the $m = \pm j$ levels is insensitive to which one is lower in energy. It thus allows the magnetization of a bulk sample to relax without relaxation of the energy, and the relaxation is always toward the state of zero magnetization. As a result, this model cannot explain *magnetization* experiments in which a magnetic field is applied to an initially demagnetized sample. In this case, it is essential to understand the relaxation of energy as that is what drives the change in magnetization from zero to a nonzero value.

The obvious environment to which energy can be transferred is the phonons. The immediate puzzle is that the spin–phonon interaction typically involves processes with $\Delta m = 1$ or $\Delta m = 2$, while in the cases of Fe_8 and Mn_{12}, we require $\Delta m = 20$. Thus the relaxation must take place via a combination of spin tunneling and phonon emission. If we accept this hypothesis, the program of understanding the magnetization experiments in molecular magnetic solids divides into two parts. The first part is to understand the relaxation mechanism in a single molecule and calculate the relevant rate. The second part is to insert this rate into whatever theory (for example, the kinetic equations) governs the dynamics of dipole-coupled molecules, and thus understand the behavior of the bulk solid. These two parts are logically separate and entail rather different ideas. The first part is entirely quantum mechanical, while the second is entirely classical.

The purpose of the present paper is to address the first part. There are two prior calculations of phonoemissive tunneling [20,21], but both are rather approximate, almost qualitative. Further, the first one is done for a tetragonal spin anisotropy in the plane perpendicular to the primary (Ising) anisotropy axis, while the second one is done for biaxial anisotropy. Further still, it is not easy to estimate the error involved in these calculations. With this motivation, we attempt in this paper to carry out a more accurate treatment, making fewer approximations, and also trying to develop more than one method of calculation.

To explain more fully what we intend to do, let us consider the case of the Fe_8 molecule. Neglecting interaction with all other degrees of freedom, the dynamics of the total spin of the molecule, $\boldsymbol{\mathcal{J}}$, are governed by the effective Hamiltonian

$$\mathcal{H}^s = -D\mathcal{J}_z^2 + E(\mathcal{J}_x^2 - \mathcal{J}_y^2) + C(\mathcal{J}_+^4 + \mathcal{J}_-^4) - g\mu_B \boldsymbol{H} \cdot \boldsymbol{\mathcal{J}}. \tag{1.1}$$

Here, $\boldsymbol{\mathcal{J}}$ is the dimensionless spin angular momentum operator, $\boldsymbol{H}$ is the external magnetic field, and C, D, and E are anisotropy energies with $D > E \gg |C| > 0$. The g-factor, g, is very close to 2, indicating an all-spin magnetic moment, and the magnitude of the total spin, j, is 10 as already mentioned. The

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