From the atom to magnetic nanoparticles

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1 Single-molecule magnets

Model Hamiltonian

Generally modeled as fixed-$\mathbf{S}^2$ spin with

$$H = -DS_z^2 + E(S_x^2 - S_y^2) + g\mu_B \mathbf{S} \cdot \mathbf{H}$$

where $\mathbf{S}$ is the spin, $\mathbf{H}$ the applied field, D and E anisotropy constants, with $D \gg E$.

Zeeman diagram

If $E = 0$ and the field is applied along z, the Zeeman diagram is very simple, with level crossings at equally spaced intervals of field. $E \neq 0$ or a tilt in the field results in avoided level crossings.

Landau-Zener tunneling

If the field is swept through an avoided level crossing with splitting Δ, the transition probability is

$$p = 1 - \exp\left(-\frac{\pi\Delta^2}{2\hbar g\mu_B |m - m'| \mu_0 \mathrm{d}H/\mathrm{d}t}\right)$$

where m abd m' are the S_z quantum numbers of the crossing levels.

Spin-parity and Berry phase

The transition is forbidden when $|m - m'|$ is odd, but only inthe absence of transverse field. The transition probability oscillates as a function of a transverse field in the hard direction with period

$$\Delta H = \frac{2}{g\mu_B\mu_0}\sqrt{2E(E+D)}.$$

It increases monotonically as a function of a transverse field in the intermediate (not hard, nor easy) direction. This can be understood quasi-classically as the result of interference between two possible reversal paths.

Intermolecular couplings

For two molecules coupled together in a dimer, the model Hamiltonian is

$$H = H_1 + H_2 + J\mathbf{S_1} \cdot \mathbf{S_2}$$

where H_i is the one-molecule Hamiltonian of molecule i and J an exchange coupling constant. Qualitatively, each molecule is "exchange biased" by the coupling to the neighbor.

External couplings

For example:

dipolar couplings: broaden the transitions

photon excitations: allow control on the magnetization reversal.

2 Magnetic nanoparticles

The uniform rotation limit

Competition between energies (exchange, demagnetizing and anisotropy) decides whether the single domain state is stable at zero field. The Zeeman term is involved to decide whether reversal occurs via uniform rotation.

The Stoner-Wohlfarth model

We assume the energy

$$E = K \sin^2\theta - \mu_0 M_s H \cos(\theta - \phi),$$

where K is the total anisotropy and θ and ϕ the directions of $\mathbf{M}$ and $\mathbf{H}$. The switching field is then

$$H_{\mathrm{sw}} = \left(\sin^{2/3}\phi + \cos^{2/3}\phi\right)^{-3/2}.$$

This has been generalized to an arbitrary anisotropy function in 2D and 3D.

Thermal dynamics

Superparamagntism happens when $K < 25k_BT$: spontaneous switching of the magnetization between the two stable orientations. At $T \ll K/k_B$, temperature reduces the switching field, which can be evidenced by:

- waiting-time measurements
- switching-field measurements

Fast-field dynamics

Intrinsic dynamics of the magnetization is described by the Landau-Lifschitz-Gilbert equation:

$$\frac{\mathrm{d}\mathbf{M}}{\mathrm{d}t} = -\gamma\mathbf{M} \times \mathbf{H}_{\text{eff}} + \frac{\alpha}{M_s}\mathbf{M} \times \frac{\mathrm{d}\mathbf{M}}{\mathrm{d}t}$$

where γ is the gyromagnetic ratio, α a phenomenological damping constant and

$$\mathbf{H}_{\text{eff}} = -\frac{1}{\mu_0 V}\frac{\partial E}{\partial \mathbf{M}}.$$

This can be probed by fast-varying fields, e.g. the magnetization reversal can be assisted by a field pulse or an RF radiation.