Hamiltonian of the Mn_{12}-ac SMM

This text is part of subsection 3.1 and Appendix B of Ref. [1].

The one-site hamiltonian that fits the experimental data of the Mn_{12}-ac molecular clusters, in the presence of a skew magnetic field, at the i-th site is[2]

$$\mathbf{H}_i = -D(S_z^i)^2 - B(S_z^i)^4 - C[(S_+^i)^4 + (S_-^i)^4] + E[(S_x^i)^2 - (S_y^i)^2] - h_x S_x^i - h_z S_z^i, \qquad (1)$$

where S_x^i, S_y^i and S_z^i are the spin operators for $S = 10$, at the i-th site; for the creation and destruction operators we have[1] $S_\pm \equiv \frac{1}{\sqrt{2}}(S_x \pm iS_y)$. The total hamiltonian is $H = \sum_i^N \mathbf{H}_i$, where N is the number of Mn_{12} molecules in the medium (we take $N \to \infty$ in the thermodynamical limit). The constant external magnetic field has components h_x and h_z, the direction z being the easy-axis. We introduce a transverse component of the magnetic field in the x-direction in order to mimic the transverse magnetic field due to dislocations in Mn_{12}-ac crystals. The terms proportional to the parameters C and E in (1) do not commute with S_z and are responsible for transitions among its eigenstates. The term proportional to E appears due to local rotations of the easy-axis[3, 4]. We can write the components of the external magnetic field, constrained to the xz-plane without loss of generality, as functions of the angle θ between the constant external magnetic field and the easy-axis (z-axis),

$$h_x = h\sin(\theta) \qquad \text{and} \qquad h_z = h\cos(\theta), \qquad (2)$$

where $\theta \in [0, 2\pi]$. The magnetic field has norm h ($h = |\vec{h}|$).

In the classical version of the $S = 10$ Mn_{12}-ac molecule we have a classical spin with norm $||\mathbf{S}|| = \sqrt{S(S+1)} = \sqrt{110}$ with components

$$S_x = \sqrt{110}\ \sin(\theta)\cos(\phi), \quad S_y = \sqrt{110}\ \sin(\theta)\sin(\phi) \quad \text{and} \quad S_z = \sqrt{110}\ \cos(\theta), \qquad (3)$$

with $\theta \in [0, \pi]$ and $\phi \in [0, 2\pi]$. The relations (3) are replaced in hamiltonian (1) in order to derive its classical version $H^{class}(\theta, \phi)$.

The values of Ref.[2] for the parameters in hamiltonian (1) for the Mn_{12}-ac SMM is,

$$\frac{D}{k} = +0.548(3)K; \quad \frac{B}{k} = 1.173(4)\,10^{-3}K \quad \text{and} \quad \frac{C}{k} = \mp 1.16\,10^{-4}K, \qquad (4)$$

k being the Boltzmann constant.

More recently, Su, Shen and Tao[5] proposed a gaussian distribution

[1] The definition of these operators differs from that used in previous articles on the subject. In hamiltonian (1) we have: $C = -4\,C_{\text{Mertes}}$.

$$P(E) = \frac{1}{\sqrt{2\pi}} \frac{1}{\sigma} e^{\frac{-(E-E_0)^2}{2\sigma^2}}, \tag{5}$$

for the parameter $E \in (-\infty, \infty)$.

The numerical analysis performed in Ref.[5] for the experimental values (4) of parameters gives

$$\frac{E_0}{k} = +0.018K \quad \text{and} \quad \frac{\sigma}{k} = 0.006K. \tag{6}$$

The average specific heat (with respect to the orientation of the chain to the external constant magnetic field) of the powder sample of the Mn_{12}-ac is given by

$$\bar{\mathcal{C}}(\beta) = \int_0^{\pi/2} \mathcal{C}(h, \theta; \beta)\ P(\theta)\, d\theta, \tag{7}$$

where $P(\theta)\ d\theta$ is the probability of finding an angle between θ and $\theta + d\theta$ between the easy-axis of the SMM and the external magnetic field. This distribution of probability depends on the experimental arrangement. Since our expansions of the quantum and classical specific heat are continuous functions of θ, they can be used in the calculation of $\bar{\mathcal{C}}(\beta)$ for any particular distribution $P(\theta)$.

Assuming a homogeneous probability distribution of chain orientations $P(\theta) = \frac{2}{\pi}$, for $\theta \in [0, \pi/2]$ we calculate $\bar{\mathcal{C}}(\beta)$.

References

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