Unitary version of the classical hamiltonians of the Fe_4 and Fe_8 SMMs

This text is part of sections 2 and 4 of Ref. [1].

The main terms in the hamiltonian of the Fe_4 and Fe_8 single-molecule magnets (SMMs) are of the same type. Their hamiltonian in the presence of an external constant magnetic field with arbitrary direction is [2, 3]

$$\mathbf{H}_S = \sum_{j=1}^{N} \mathbf{H}_j^{(S)}, \qquad j = 1, 2, \cdots, N, \tag{1a}$$

where

$$\mathbf{H}_j^{(S)} = -D_S (S_j^z)^2 + E_S[(S_j^x)^2 - (S_j^y)^2] - h_x S_j^x - h_y S_j^y - h_z S_j^z. \tag{1b}$$

The hamiltonian (1a) describes a Fe_4 or Fe_8 SMM with N sites, in which h_x, h_y and h_z are the components of the constant external magnetic field. For the Fe_4 SMM, (S_j^x, S_j^y, S_j^z) corresponds to spin-5 operators at the jth-site, whereas for the Fe_8 SMM they are the spin-10 operators.

The classical versions of these two SMMs are obtained by replacing, in (1b), the spin-S operators by the classical functions

$$S_x^{class} = \sqrt{S(S+1)}\ \sin\theta\ \cos\phi \tag{2a}$$
$$S_y^{class} = \sqrt{S(S+1)}\ \sin\theta\ \sin\phi \tag{2b}$$
$$S_x^{class} = \sqrt{S(S+1)}\ \cos\theta\ , \tag{2c}$$

with $\theta \in [0, \pi]$ and $\phi \in [0, 2\pi]$.

An interesting comparison of some thermodynamical functions of the Fe_4 and Fe_8 SMMs can be made when we study the thermodynamics of the unitary (normalized) version of the hamiltonian (1b) for the spins $S = 5$ and $S = 10$. For each value os spin in this hamiltonian, the unitary version of $\mathbf{H}_j^{(S)}$ is obtained by replacing the spin matrices by the unitary spin operator $\vec{s}$, defined as

$$\vec{s}_j \equiv \frac{\vec{S}_j}{\sqrt{S(S+1)}}, \qquad S \in \{5, 10\}, \tag{3}$$

where $\vec{S}_j$ is the spin operator at the jth- site, with norm $||\vec{S}_j|| = \sqrt{S(S+1)}$, for $S \in \{5, 10\}$.

The unitary version of the hamiltonian (1b) is

$$\mathcal{H}_j^{(S)} = -D_S(s_j^z)^2 + E_S[(s_j^x)^2 - (s_j^y)^2] - h_x s_j^x - h_y s_j^y - h_z s_j^z. \tag{4}$$

We denote by $\omega_S(D_S, E_S, h_x, h_y, h_z; \beta)$ the Helmholtz free energy (HFE) derived from (4) for $S = 5$ and $S = 10$.

It is simple to show that the HFEs derived from the hamiltonians (1b) (we call it $\mathcal{W}_S(D_S, E_S, h_x, h_y, h_z; \beta)$) and (4) are related by

$$\mathcal{W}_S(D_S, E_S, h_x, h_y, h_z; \beta) = S(S+1)\, \omega_S(D_S, E_S, \tilde{h}_x, \tilde{h}_y, \tilde{h}_z; \tilde{\beta}). \tag{5}$$

where

$$\tilde{h}_i \equiv \frac{h_i}{\sqrt{S(S+1)}}, \qquad \tilde{\beta} \equiv S(S+1)\beta \tag{6}$$

and $i \in \{x, y, z\}$.

References

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