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Factorizing magnetic fields triggered by the Dzyaloshinskii–Moriya interaction: Application to magnetic trimers

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ABSTRACT

This work examines the entanglement and correlation state between the assembled spins of a magnetic trimer by using the magnetic specific heat $\mathcal{C}_M$ as a thermal observable and the critical magnetic fields as factorization fingerprints. We show that the entanglement of the $(Cr_7Ni)_2Cu$ trimer as a function of the temperature and external magnetic field becomes a quantifiable quantity when a Dzyaloshinskii–Moriya (DM) interaction is considered. Such entanglement is characterized by critical magnetic fields at which the bipartite density matrices display an abrupt change between factorable and entangled states. In $(Cr_7Ni)_2Cu$ magnets as well as in trimers with local $S\neq\frac{1}{2}$ the DM interaction shifts the critical factorizing fields to quantifiable thermal regimes where they could be detected by magnetic specific heat measurements.

1. Introduction

Molecular nanomagnets conform one of the most promising fields in molecular electronics [1–4]. Their versatility from molecular functionalization viewpoint and exceptional magnetic properties make them one of the systems with the highest potential to future storing and processing information technologies [1–6]. Among the phenomena that have been studied in molecular nanomagnets we have, e.g., the giant-spin-tunneling [1–3], the Berry-phase interference of spin-paths [7,8], the measurable Dzyaloshinskii–Moriya interaction [8–10], and the mesoscopic manifestation of thermal entanglement [11,12]. The single-molecular magnets (SMM) are the most representative systems among the molecular nanomagnets [1–4]. They are characterized by giant-spins and well defined anisotropies which are the key of their outstanding spin-tunneling properties. In this work however, our discussion focus on molecular nanomagnets which differ from the SMM at the effective spin-modeling stage, e.g., they are described by small spins and the anisotropy-barrier separated states are no longer present but the spin dynamics involves transitions between entangled and factorable states. We study a magnetic trimer belonging to the family of the Cr_xNi-based magnets [11–19]. Among the characteristics of this aforementioned family there are two complementary ones that have motivated this work, namely, the assembling of antiferromagnetic rings in this magnets and the rings themselves display thermal observables with interesting tunable properties depending on the intra and inter-rings exchange couplings [19–22]; the second one is that these nanomagnets evidence experimentally quantifiable entanglement features [11]. Understanding the quantities witnessing the molecular entanglement at finite temperature is a step forward in both fundamental and applicative efforts focused on control molecular interactions and the consequences of its functionalization [11,12,23]. According to such an idea it was shown very recently [12] how a functionalization of the exchange interactions between the effective rings of the Cr_7Ni–Cu–Cr_7Ni [13] would give rise to a very particular entanglement which is similar to the one found in the Cr_7Ni-purple [11]. Nevertheless, these studies let several questions open, e.g., the possibility of having another interaction able to trigger such an entanglement, the role of this last interaction in magnetic trimers describing connected molecular rings with different effective spin size, a general underlaying connection between the principal characteristics of the triggered spin-entanglement, the more distinguishable being the presence of factorizing magnetic fields, and the thermal properties witnessing such spin correlations; and the reliability of the thermal observables in predicting spin correlations in more complex systems for which the entanglement of formation has not even been well defined. In this work we address these questions through the study of the factorizing magnetic fields of a magnetic trimer representing both the

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Cr_7Ni–Cu–Cr_7Ni magnet or a hypothetic molecule with larger local spins than the Cr_7Ni–Cu–Cr_7Ni one. A small Dzyaloshinskii–Moriya (DM) interaction is introduced in the effective spin model and the critical magnetic fields appearing in the spin-entanglement and correlation as a consequence of such interaction are witnessed through the magnetic specific heat $\mathcal{C}_M$. Such an entanglement as well as the critical factorizing fields are studied along the next sections as follows. In Section 2, we describe the effective quantum model and calculate the entanglements of formation E_f for the effective magnetic trimer. In Section 3, we demonstrate that E_f is nonzero if we allow the system to experiment a moderate DM interaction. In Section 4, we capture the main characteristics of E_f as a function of the temperature and magnetic field through the magnetic specific heat. In Section 5, we extend the model to $S\neq\frac{1}{2}$-type trimers, and finally in Sections 6 and 7, we discuss the results of the previous sections and present the conclusions, respectively.

2. Cr_7Ni–Cu–Cr_7Ni magnet: entanglement

The Cr_7Ni–Cu–Cr_7Ni molecular magnet is conformed by the coupling of two molecular rings of Cr_7Ni through a Cu^{+2}. The basal states of such a magnet can be well represented by using a reduced spin Hamiltonian with effective parameters fitted from electron paramagnetic resonant measurements [13]. This minimalist Hamiltonian is actually similar for some molecules of the Cr_xNi-based family. For instance, the effective models for the Cr_7Ni-purple and Cr_7Ni–Cu–Cr_7Ni molecules differ by one site in a $\frac{1}{2}$-open-XXZ-chain [12]. Therefore while the first molecule behaves approximately like a spin-dimer the second one behaves as a spin trimer, respectively. In the case of the $(Cr_7Ni)_2Cu^{+2}$ the Cu introduces an effective Landé factor, which is slightly different to the one describing the antiferromagnetic rings. Our initial effective Hamiltonian reads:

$$\mathcal{H}=J_1(S^+_{ef}S^-_{Cu}+S^-_{ef}S^+_{Cu})+J_2S^z_{ef}S^z_{Cu}+\vec{\alpha}.(g_{Cu}\vec{S_{Cu}}+g_{ef}\vec{S_{ef}}), \tag{1}$$

where the first term corresponds to the non-longitudinal exchange couplings and the second one describes the longitudinal interactions along the ring axis. In Eq. (1) $\vec{S}_{ef}=\vec{S}_{R_1}+\vec{S}_{R_2}$ for the Cr_7Ni–Cu^{+2}–Cr_7Ni molecule ($\vec{S}_{ef}=\vec{S}_{Cu}=\vec{S}_R$ for the Cr_7Ni-purple one). We take the external magnetic field such that $\vec{\alpha}=\alpha\hat{z}$ and $\alpha=\mu_BB$. Spin operators are $|\vec{S}_A|=|\vec{S}_B|=|\vec{S}_C|=1/2$, and $g_{ef}=g_R$ and g_C are taken from experimental results [13]. $(J_1(D_1,D_2),J_2(D_1,D_2))$, with (D_1,D_2) being parameters that obey the relations $J_1=(D_1-D_2)/2$ and $J_2=D_1+2D_2$, retrieve the fitted effective Hamiltonians [21]. Fig. 1 shows the "intuitive" scheme of our molecular prototype.

Now, in order to study the behavior of the entanglement in the $(Cr_7Ni)_2Cu$ trimer we use the entanglement of formation [24], which establishes that for a density matrix ρ the bipartite entanglement associated to the partial density matrix ρ_{tr} that appears after projecting out the states different than the bipartite ones is defined as $E_f(\rho_{tr})=\mathcal{E}(C(\rho_{tr}))$, where $\mathcal{E}(C)=\sum_{\sigma=\pm\frac{1}{2}}-(|\sigma|+\sigma\sqrt{1-C^2})\log_2(|\sigma|+\sigma\sqrt{1-C^2})$ and the concurrence $C(\rho_{tr})=\max\{0,\lambda_1-\lambda_2-\lambda_3-\lambda_4\}$ [24]. λ_i are the decreasing square roots of the eigenvalues of $\rho_{tr}(\sigma_y\otimes\sigma_y)\rho^*_{tr}(\sigma_y\otimes\sigma_y)$, the spin-flipped non-Hermitian density matrix with σ_y the Pauli matrix [24,25]. In our case we have three sites in the spin chain, one associated with the Cu ion and two to the heterometallic rings. ρ_{tr} stands then for $trace(\rho)$, ρ being the full thermal density matrix for the solutions of the Hamiltonian equation (1) and where the sum is made over the projections upon the spin states $\uparrow$ or $\downarrow$ associated with either the Cu ion or each one of the heterometallic rings. For sake of simplicity we simplify the notation of the aforementioned states and we name them $|ABC\rangle$. In our model, the forms of the density matrix and concurrence follow the description presented in Ref. [25] so here we explicitly present just these last ones:

$$C(\rho_{tr})=2\max\{0,|Z_{tr}|-\sqrt{W_{tr}}\}, \tag{2}$$

with $tr=B$, Z_B and W_B being

$$Z_B=\sum_{i=\pm\frac{1}{2}}\sum_{j=\pm\frac{1}{2}}\frac{|\mathcal{O}(i,j)|^2P_{i,j}}{2|\mathcal{O}(i,j)|^2+1}-\frac{P_{i,0}}{2},$$

$$W_B=\prod_{i=\pm\frac{1}{2}}P_{3i}+\sum_{j=\pm\frac{1}{2}}\frac{P_{i,j}}{2|\mathcal{O}(i,j)|^2+1},$$

whereas with $tr=A,C$, we have

$$Z_{A,C}=\sum_{i=\pm\frac{1}{2}}\sum_{j=\pm\frac{1}{2}}\frac{|\mathcal{O}(i,j)|P_{i,j}}{2|\mathcal{O}(i,j)|^2+1},$$

$$W_{A,C}=\prod_{i=\pm\frac{1}{2}}P_{3i}+\frac{P_{i,0}}{2}+\sum_{j=\pm\frac{1}{2}}\frac{|\mathcal{O}(i,j)|^2P_{i,j}}{2|\mathcal{O}(i,j)|^2+1},$$

In Eq. (2) we have $\mathcal{O}(i,j)=-J_1/(\mathcal{P}(i)-2j\sqrt{2J_1^2+\mathcal{P}^2(i)})$, with $\mathcal{P}(i)=J_2/4-i\alpha(g_{ef}-g_c)$. $P_{i,0},P_{i,\pm\frac{1}{2}},P_{\pm\frac{3}{2}}$ are the quantum Boltzmann populations for the energy solutions of Eq. (1) namely $E_i^0,E_i^{\pm\frac{1}{2}},E_{\pm\frac{3}{2}}$, respectively, which are calculated from the subspaces given by the Hamiltonian projections upon the state with the same total S_z as it is shown in Ref. [21]. Obtaining the entanglement of formation with $\mathcal{E}(C(\rho_{tr}))$ is straightforward after to find the concurrences. The Cr_7Ni–Cu–Cr_7Ni molecule has the couplings $J_1(K)\approx-0.23$ and $J_2(K)\approx-0.58$, so after using these values in Eq. (2) and calculating the entanglement we find $E_f(\rho_B)=E_f(\rho_{A,C})=0$. Now, if the molecular system could be functionalized in such a way that its exchange couplings J_1J_2 would effectively change to positive values, e.g., $J_1(K)=0.015$ and $J_2(K)=0.038$, therefore the entanglement of formation would be the one pictured in Fig. 2. Here the most important thing to point out about Fig. 2 is that the molecular prototype can be entangled just when the exchange couplings are adequately modified. To get this last bipartite entanglement by manipulating the couplings between Cr_7Ni rings could seem unrealistic at a first look however there is already an evidence that suggests it actually could be done [11]. That is the case of the Cr_7Ni-purple [11] which displays a concurrence that follows a behavior similar to the entanglement shown in Fig. 2. Let us continue with further discussion in the next sections. For now, we need two remarks: (1) The molecular prototype may be not entangled when just exchange interactions are considered, and the exchange anisotropy $\Delta=J_2/J_1$ does not strictly define whether the entangling process occurs or not; (2) the second remark is associated with the fact that when the trimer is entangled the low-temperature main features of $E_f(\rho_{tr})$ are represented by a multivalued behavior with respect to the magnetic field and there is a critical magnetic field at which $E_f(\rho_{tr})$

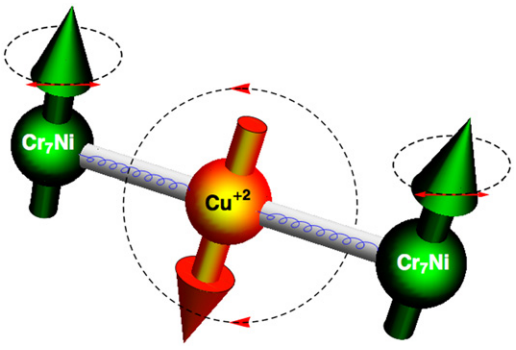

Fig. 1. Effective spin-picture of the Cr_7Ni–Cu–Cr_7Ni.