Dzyaloshinskii-Moriya anisotropy induced by magneto-elastic interactions in antiferromagnetic molecular wheels

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Purpose of my Visit

The purpose of my visit to Prof. F. Mila and his group at the *EPFL* of Lausanne was the investigation of the role of magneto-elastic interactions in inducing Dzyaloshinskii-Moriya (DM) anisotropies in antiferromagnetic molecular wheels. The main motivation comes from the suggestion, in a very recent paper [2], that the spin-lattice coupling can be at the origin of a peculiar torque signal in the ferric wheel $CsFe_8$.

Depending on the orientation of the DM vectors, the DM interaction can induce frustration or not. In fact, it can be shown that the DM interaction generates an effective staggered field, which can compete with the super-exchange coupling and even induce a phase transition [1]. In the model we dealt with, the anisotropic coupling indeed competes with the antiferromagnetic (AFM) exchange and generates a staggered magnetization transverse to the field and opposite to one due to the AFM interaction.

Introduction

In recent years, there has been an intense theoretical activity devoted to the effect of DM interactions[3, 4] in otherwise isotropic magnets. When present, this type of anisotropy gives rise to a number of "anomalous" effects such as the opening of a gap in otherwise gapless regions, and the development of a staggered magnetization perpendicular to the applied magnetic field.[5, 6, 7, 8] The latter may be experimentally manifested by an accompanying characteristic torque signal. It is already invoked to interpret numerous experimental results.[6] For two spins localized at orbitals i and j, it can be effectively written in spin space as $\mathbf{d} \cdot \mathbf{s}_i \times \mathbf{s}_j$.

At the same time, it is well established that the electron-phonon interaction is at the origin of a number of fascinating effects in isotropic Mott insulators. The balance between the corresponding magnetic free energy gain and the elastic energy cost determines whether the lattice distorts or not. All previous investigations focus on the effect of magneto-elastic interactions on the dominant exchange energy. In principle however, since a lattice distortion breaks some or all of the symmetries of the problem a development of an accompanying anisotropy, such as the DM interaction, is also possible. This scenario of a magneto-elastically induced spin anisotropy has been recently implied to be the possible origin of a peculiar torque signal in the context of finite molecular magnets (MM's) and, in particular, in the ferric wheel $CsFe_8$.[2]

An increasing magnetic field $\mathbf{B}$ drives the system through subsequent level crossings (LC's) between the two lowest energy levels, whereby the ground state

changes its total spin from S to $S+1$ to $S+2$ etc. The system may then become susceptible to a small anisotropy around the degenerate LC point provided that this anisotropy admixes the two relevant levels: A magneto-elastic mechanism may induce such an anisotropic interaction. Indeed, the torque measurements of the ferric wheel $CsFe_8$ reveal a severe mixing of the two lowest levels at the lowest two LC fields.

Motivated by these experimental results, we put forward a minimal model which captures the interplay between magneto-elastic and DM interactions. The model assumes a magnetic ring with a sufficient number of symmetries that do not allow any avoided level crossing (ALC). The possibility of an induced DM anisotropy able to provide an ALC is then taken into account by including a set of DM terms proportional to the distortion amplitudes. Consistency is ensured by forcing all DM terms to be compatible with the remaining symmetries after the distortion. More explicitly, the full Hamiltonian of our model reads

$$\mathcal{H} = \mathcal{H}_m + \mathcal{H}_{\rm ph} + \mathcal{V} \ , \tag{1}$$

where $\mathcal{H}_m$, $\mathcal{H}_{ph}$ are the energies of the spin and phonon system respectively, and $\mathcal{V}$ denotes their mutual interaction. The first contains the exchange and Zeeman energies

$$\mathcal{H}_m = J \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j - \mathbf{M} \cdot \mathbf{B} \ , \tag{2}$$

where $\mathbf{M} = -g\mu_B \mathbf{S}$ is the total magnetization, $\mathbf{S} = \sum_i \mathbf{s}_i$ is the total spin, μ_B denotes the Bohr magneton, and g the electronic $g-$factor, hereby assumed isotropic and common for all spins. The magneto-elastic contribution is taken to be

$$\mathcal{V} = \sum_{\langle ij \rangle} \mathbf{d}_{ij} \cdot \mathbf{s}_i \times \mathbf{s}_j \ . \tag{3}$$

The direction of each $\mathbf{d}_{ij}$ is controlled by symmetry, while its amplitude is taken to be linear in the set of relevant distortion amplitudes $\boldsymbol{\delta}_q$. With this choice, we basically keep only the first term of the *analytical* expansion of the magneto-elastic energy around the undistorted $\boldsymbol{\delta}_q = 0$ limit where all $\mathbf{d}_{ij}$ vanish by symmetry. Finally, the associated elastic energy cost reads

$$\mathcal{H}_{\rm ph} = \sum_q \frac{1}{2} \{ K_q \boldsymbol{\delta}_q^2 \ + K_q^{'} \boldsymbol{\delta}_q^4 \}, \tag{4}$$

where we have included both quadratic and quartic contribution with K_q and $K_q^{'}$ denote the corresponding stiffness constants. To keep the model as simple as possible we shall restrict to the case $K_q = K$ and $K_q^{'} = K^{'}$ for all q. This symmetric choice necessitates a minimum energy state with a set of correspondingly symmetric distortions where all $|\boldsymbol{\delta}_q|$ and the induced $|\mathbf{d}_{ij}|$ are the same. These simplifications facilitate considerably the numerical effort of finding the minimum energy configuration.

In order to best illustrate the main physical arguments and results we first treat a much simpler molecular ring, namely the spin s tetramer depicted schematically in Fig. 1. Due to the symmetries of the lattice, when any distortion mode is excluded, the DM vectors have all the same intensity and lie along the axis perpendicular to the plane of the molecule (z-axis) (see Fig. 1A).

The symmetries of this DM interaction prevent the opening of a gap at the LC's. When a buckling type of distortion, (consisting of a equal amplitude

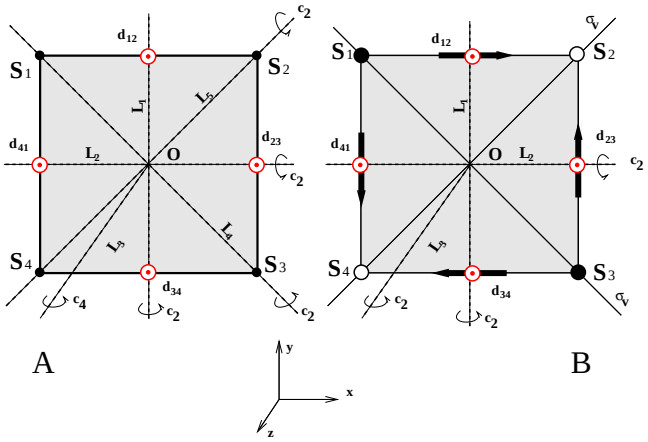

Figure 1: (color online) Pictorial representation of the tetramer with DM interaction without (A) and with (B) spin-lattice coupling. In B the sites are alternatively above (full dots) and below (open dots) the xy-plane. The direction of the DM vectors is set by symmetry arguments. Red dots represent the z-component of DM vector, while black arrows the in-plane component: $d^x_{12} = d^y_{23} = -d^x_{34} = -d^y_{41} = d \sin\alpha$ and $d^z_{i\,i+1} = d\cos\alpha\ \forall i$, where α is a degree of freedom. In A $\alpha = 2\pi$.

shift of the pair of sites $1, 3$ and $2, 4$ above and below the plane of the tetramer, respectively), is considered, a set of DM interactions as in Fig1B is now allowed. This configuration leads to frustration in the sense of [1].

Description of the work carried out during the visit

At first we identified a simple spin model in which the induction of anomalous features by DM interactions is prevented by symmetry arguments (1A). Then we proposed a simple distortion mode which breaks the minimal number of symmetries necessary to induce effective anisotropies (1B). We first investigated the system in the presence of a static distortion. By means of exact diagonalization, we analyzed the dependence of the gap on the spin s, on the intensity of the DM interaction and the orientation of the magnetic field. We also obtained the behavior of the torque as a function of the magnetic field amplitude. Our results are compared with a perturbation theory analysis We then turned to an investigation of the magneto-elastic interactions, which induce the distortion mode mentioned above. By means of full diagonalization, we determined the minimum energy configurations at fixed distortion parameters and then we selected the displacements which minimize the GS energy.

Description of the main results obtained

static distorted lattice

- Numerical results confirmed that the gap at the LC's is a quadratic function of the intensity of the DM interaction, as deduced from symmetry arguments.
- The DM vectors at opposite bonds of the tetramer competes with the

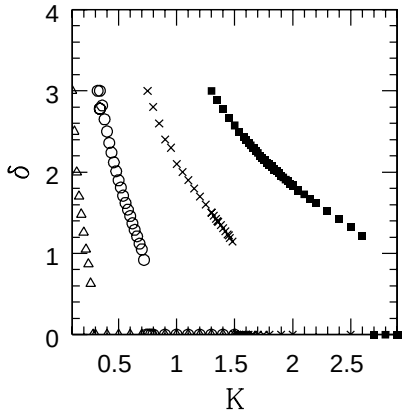

Figure 2: Displacement δ as a function of the coupling constant K at magnetic field $B = 1$ $\theta = 45^0$, $\alpha = 45^0$ and $K^{'} = 0$ for $s = 1/2$ (triangles), 1 (dots), 3/2 (crosses), 2 (square).

AFM exchange between the two bonds and the frustration manifests itself with a peculiar sign reversal of the torque at the LC's.

- The directionality of the torque is consistent with our perturbative analysis.

magneto-elastically distorted lattice

- Since the lowest order correction to the GS energy due to the DM interaction in perturbation theory is quadratic in the intensity of the DM vector and thus of the same order of the dominant term in the elastic energy cost, we foresee the transition from a *rigid* to a distorted lattice to be first order, if present. This is confirmed by numerical results (see Fig. 2).

- The non-zero values of δ selected by the magneto-elastic mechanism imply that the DM contribution is a noticeable fraction of the total energy.

- The magneto-elastic mechanism is effective only close to the LC, where the two lowest energy levels are almost degenerate.

- The main features (direction and amplitude of the torque signal and distortion δ) vary with the spin, the direction of the magnetic field and the intensity of the elastic stiffness. In Fig. 3, a representative case is shown for the $s = 1/2$ tetramer.

Future of the project

In view of interpreting the experimental data on the $CsFe_8$ wheel, we are currently investigating the $s = 5/2$ octagon, in the presence of magneto-elastic interaction. Since the experimental results suggest a strong admixing of the lowest energy levels in a quite large region around the LC's, while our preliminary results indicate a gap opening as the fourth power of the DM intensity and a lattice very stable with respect to magneto-elastic instabilities, we are planning

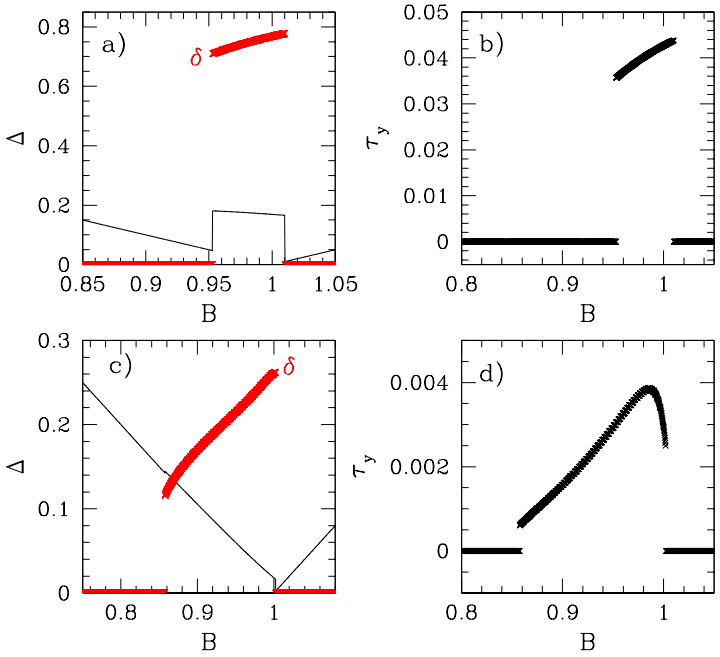

Figure 3: (color online) Upper panels: gap and optimal displacement δ (red) (Fig.a) and non-zero component of the torque versus magnetic field B for $K = 0.25$, $K^{'} = 0$, $\theta = 45^0$ and $\alpha = 45^0$. Lower panels: the same as in the upper ones with $K = 0.2$, $K^{'} = 1.5$, $\theta = 45^0$ and $\alpha = 45^0$

to allow more complex distortion modes, with a larger number of symmetries broken. In fact, some of our numerical simulations have confirmed that, when the DM interaction is less symmetric, the features displayed by our minimal model are closer to that suggested for the real molecule. Namely, a first order correction to the ground state energy, a continuous growth of the displacement δ as the elastic stiffness decreases and a gap at the LC's opening linearly with the intensity of the DM interaction. We are further planning to investigate the role of single ion anisotropies and anisotropic g-tensors in the presence of spin-lattice coupling. A paper on the tetramer case is already in preparation and we are planning to submit it to Phys. Rev. B.

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