Theory of the orthogonal magnetic structure in GdB_4

Geometrical magnetic frustration combined with competing anisotropies leads to a rich and complex behavior of rare-earth tetraborides RB_4. In our study we focus on GdB_4, which exhibits a chiral spin cross antiferromagnetic state in zero magnetic field. We develop a microscopic spin model that explains the origin of this unusual 90° magnetic structure and study the magnetization process in high magnetic fields.

Metallic rare-earth tetraborides exhibit interesting magnetic and electrical properties. Their crystal lattice can be viewed as an array of two-dimensional layers of orthogonal dimers formed by pairs of nearest-negihbor magnetic rare earths, see Fig. 1. This lattice is topologically equivalent to the famous Shastry-Sutherland lattice, which in the case of small spins $S = 1/2$ and antiferromagnetic interactions hosts an exact spin-liquid state [1]. Rare-earth tetraborides provide an experimental realization of the Shastry-Sutherland model with large spins. GdB_4 is a special member of the tetraboride family, which possess a unique 90° spin structure in zero magnetic field shown in Figure 1 [2]. The origin of such a chiral antiferromagnetic structure as well as the behavior in high magnetic fields [3] remain unexplained to date. In our recent study [4] supported by ICC-IMR, Tohoku University, we address these fundamental questions related to GdB_4 and other planar tetraborides like TbB_4.

Gadolinium $4f^7$ ions have the electronic configuration and are described by the spin-only magnetic ºmoments with $S = 7/2$. The minimal spin model for GdB_4 must include at least two exchange constants J_1 and J_2 corresponding to interactions inside and between dimer pairs, see Fig. 1. In the tetragonal crystal lattice of tetraboriodes, the local symmetry on rare-earth sites is only orthorhombic. Accordingly, the lowest-order crystal-field Hamiltonian has a biaxial form:

$$\hat{\mathcal{H}} = \sum_{\langle ij\rangle} J_{ij}\,\mathbf{S}_i\cdot\mathbf{S}_j + \sum_{\langle ij\rangle} \mathbf{D}_{ij}\cdot(\mathbf{S}_i\times\mathbf{S}_j) + \sum_i \left[D S_i^{z2} + E\left(S_i^{x2} - S_i^{y2}\right)\right] \qquad (1)$$

In addition to the exchange and the single-ion terms D the above spin Hamiltonian includes the antisymmetric Dzyaloshinskii-Moriya (DM) interactions, which is responsible for a specific chirality in the spin cross state. The DM interactions are forbidden on the dimer bonds because of the inversion symmetry with respect to the bond center. Such symmetry is not present on the second neighbor bonds. Furthermore, because the ab plane is the mirror plane, the DM vectors on the second-neighbor bonds must be parallel to the z axis. The corresponding sign convention for the antisymmetric couplings is indicated by arrows on the second neighbor bonds (Fig.1).

We have used the spin Hamiltonian (1) to compute the basic properties of GdB_4. The observed 90° spin structure is stabilized by a negative in-plane anisotropy constant $E < 0$. The chiral state is favored by the positive DM constant $D_2 > 0$. Values of the microscopic parameters J_1, J_2 and D are constrained by the Curie-Weiss temperature $\theta = -67$ K in GdB_4 and the measured values of the saturation field $H_s = 52$ T for H // [100] and 54T for H // [001] [3]. This procedure gives $J_1 = 8$K, $J_2 = 0.9$K, $D = 0.45$K, $E = 0.1$K. We further theoretically simulated the magnetization process in GdB_4 by performing energy minimization on finite lattice clusters starting with random spin configurations. The calculated $M(H)$ curves are presented in the middle and the right panels of Figure 1.

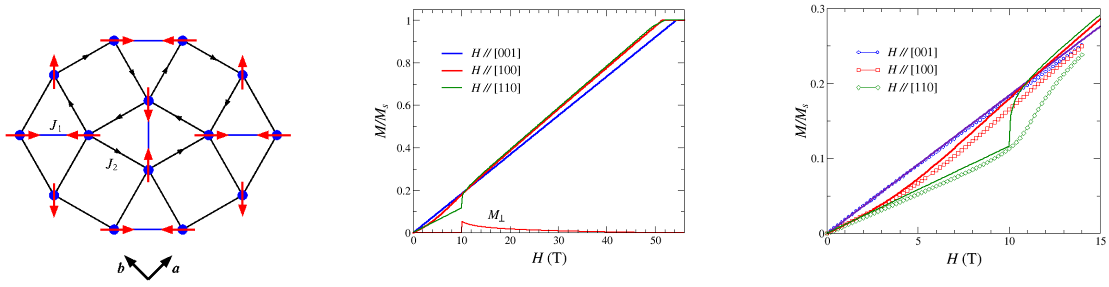

Fig. 1. Left panel: Lattice of gadolinium atoms in GdB_4. Red arrows show the ordered magnetic moments as observed in the neutron diffraction experiments [2]. Middle panel: Magnetization curves $M(H)$ computed for the spin model (1) of GdB_4 with J_1 = 8K, J_2 = 0.9K, D = 0.45K, E = -0.1K for different orientations of an applied field. For H //[100] the magnetization develops a transverse component above the spin-flop transition at 10T. Right panel: Comparison of the theoretical results to the experimental data [3].

Using our spin model we were able to successfully describe the magnetization process in GdB_4 for all three principal orientations of the field. Furthermore, we explain an unusual feature on the curve H // $[110]$ as the spin-flop transition, which affects only a half of Gd spins. The Monte Carlo simulations of the spin model (1) are under way and will further help to validate our set of microscopic parameters by comparing to the observed transition temperature in zero magnetic field.

References

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Keywords: **magnetism, ordering, high magnetic fields**

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