Spin Structure of the Anisotropic Helimagnet Cr$_{1/3}$NbS$_2$ in a Magnetic Field

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In this letter we describe the ground-state magnetic structure of the highly anisotropic helimagnet Cr$_{1/3}$NbS$_2$ in a magnetic field. A Heisenberg spin model with Dzyaloshinskii-Moriya interactions and magnetocrystalline anisotropy allows the ground state spin structure to be calculated for magnetic fields of arbitrary strength and direction. Comparison with magnetization measurements shows excellent agreement with the predicted spin structure.

Controlling the electrical properties of materials by manipulating their magnetic structure has been one of the primary themes in the field of magnetism research and its applications. Major technological innovations have been based on these efforts, such as giant magneto-resistance in magnetic multilayers systems 2 and magnetic tunneling effects 3, 6–10. These objects are especially interesting because of the stability granted by their topology. A detailed understanding of such spin structures, in relation with their effect on electrical properties, is expected to shed light on developing spin-texture based applications 10.

With its layered noncentrosymmetric crystal structure, the helimagnet Cr$_{1/3}$NbS$_2$ is well-suited for investigations of spin structure, especially toward controlling electrical properties 11, 12. In Cr$_{1/3}$NbS$_2$, Cr$^{3+}$ ions are intercalated between the hexagonal 2$H$-NbS$_2$ layers and magnetically order at $T_C = 133$ K 11, 12. The crystal structure’s lack of inversion symmetry, caused by Cr intercalation, results in a helical magnetic ground state oriented along the crystalline c-axis, with spins aligned ferromagnetically within the ab planes. Unlike other well known helimagnets within the B20 crystal structure 4, 5, Cr$_{1/3}$NbS$_2$ only breaks inversion symmetry along the c-axis, making it ideal for studying spin-textures in magnetic thin films 13, which also break inversion symmetry only along the single axis normal to the plane of film.

The quasi 2-dimensional (2D) nature of these layered ferromagnetic planes, paired with strong magnetocrystalline anisotropy, allows a clear distinction between the magnetically hard axis (i.e. c-axis of the crystal) and the easy plane (ab-plane). The above qualities of Cr$_{1/3}$NbS$_2$ greatly resemble those of planar magnetic devices fabricated on a substrate, making this material especially attractive as a model system.

Furthermore, Cr$_{1/3}$NbS$_2$ hosts a chiral soliton lattice phase in the presence of a magnetic field applied within the $ab$-plane 4. In this context, solitons are essentially 360° domain walls in the spin structure, as illustrated in Fig. 1 (e). As the field strength increases, the space between adjacent solitons grows 3, 14 until a phase transition to a spin-polarized state occurs at $B_c = 0.175$ T 12, 15. Alternatively, if a magnetic field is applied along the c-axis (also the helical axis), the transition to a polarized state occurs at a much larger field of 2.45 T 11, 16, through a conical state. A complete description of the spin structure in Cr$_{1/3}$NbS$_2$ must unite these disparate magnetic field scales. It must also describe how the local magnetic structure changes with magnetic fields of varying strength and direction.

In this letter we show that the spin structure of Cr$_{1/3}$NbS$_2$ in a magnetic field $B$ is well-described by a 1-dimensional (1D) Heisenberg spin model with Dzyaloshinskii-Moriya (DM) interactions 17, 18, a Zeeman interaction, and strong magnetocrystalline anisotropy. After solving for the ground state of the model and its magnetization, we compare with magnetization measurements of Cr$_{1/3}$NbS$_2$. The predictions follow the data closely, and capture the soliton lattice transition at low magnetic fields applied in the ab-plane, the conical transition at high fields applied along the c-axis, as well as the behavior at intermediate fields applied at oblique angles.

The inset of Fig. 1 (a) shows a schematic of these measurements, where a magnetic field is applied at an angle $\theta_B$ to the c-axis of a Cr$_{1/3}$NbS$_2$ sample and the component of the magnetization parallel to that field $M_{||}$ is detected. In all the measurements, the temperature was held fixed at $T = 4$ K $\ll T_C$, justifying the zero-temperature approximation used in calculations. Typical data from these measurements (blue circles) are shown in Fig. 1 (a-d) for different fixed values of $\theta_B$, with the model’s predictions plotted as the solid red lines. The

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agreement is remarkable.

We now describe the details of the spin model. Within a single $ab$-plane, the only spin-spin interaction is exchange, which results in a uniformly polarized state with all spins in the plane aligned. This allows the low-temperature magnetic structure of Cr$_{1/3}$NbS$_2$ to be studied on a 1D lattice, where sites correspond to local moments of Cr$^{3+}$ ions at different position along the $c$-axis. The magnetic moment of the spin on the $i$th site is represented classically as a 3 dimensional vector of magnitude $s_i = s n_i$, $n_i$ a unit vector. The Hamiltonian for the system is

$$H = \sum_i \left[ -J s_i \cdot s_{i+1} - D \cdot (s_i \times s_{i+1}) - \mu_B B \cdot s_i + A (\hat{z} \cdot s_i)^2 \right],$$

with $J$ and $A$ both positive and $\hat{z}$ along the $c$-axis. The four terms in the above expression represent the exchange interaction, the DM interaction, a Zeeman interaction, and magnetocrystalline anisotropy, respectively. $\mu_B$ is the Bohr magneton. In Cr$_{1/3}$NbS$_2$, the crystalline lattice has a 3-fold rotation symmetry about the $c$-axis$^{11}$, so for the DM term to remain rotationally invariant, $D$ has a non-zero component only along $c$-axis. We thus put $D = D\hat{z}$.

The symmetry of this Hamiltonian allows us to confine our study to magnetic field directions in the $ac$ plane. That is, we fix the azimuthal angle of the magnetic field at $\phi_B = 0$ and examine the ground state at varying field strengths, for different polar angles $\theta_B$ in the interval $[0, \pi/2]$. To make direct comparisons between the spin-model and measurements of the magnetization, we choose model parameters appropriate for Cr$_{1/3}$NbS$_2$. Cr$^{3+}$ ions have localized spins $S = 3/2$ with magnetic moments of magnitude $\mu_{Cr} \equiv g\mu_B S = 3\mu_B$, consistent with the observed value. We thus take $s = 3$ in our calculations. To fix the remaining parameters $J$, $A$, and $D$ we examine predictions of the model. At zero field the ground state is helical with pitch $\lambda = 2\pi a/\delta$, where $a$ is the $c$-axis lattice constant and $\delta \equiv \arctan(D/J)$. The handedness of the helix is determined by the sign of $D$. In Cr$_{1/3}$NbS$_2$, $\lambda \approx 40a^{1,16}$ and the helices are left-handed$^3$, making $\delta \approx D/J = -0.16$ a small parameter. In magnetic fields of sufficient strength all moments polarize along $B$. When $B$ is in the $ab$-plane ($\theta_B = 90^\circ$), that critical field is $\mu_B B_c = [(\pi/2)^2 + O(\delta^3)]J a$, while a field along the $c$-axis ($\theta_B = 0$) requires a stronger field of $[(\delta^2 + 2\alpha) + O(\delta^3)]J a$. Here $\alpha \equiv A/J$. $J$ and $A$ may then be determined by comparison with experiments which determine the saturating magnetic fields in these two different field orientations. This fixes $\alpha \equiv A/J = 0.10$ and $J = 0.2$ meV, the only free parameters in the model.

To efficiently identify the ground state of Eq. (1) for arbitrary $B$, we make the assumption that all spins have the same polar angle ($\theta$) or $z$-component. This is a valid assumption for most magnetic fields, and breaks down only when $0 < \theta_B < \pi/2$, as has been confirmed by direct numerical minimizations of Eq. (1) where $\theta_i$ varies with $i$. Further discussion on the validity of this assumption is momentarily deferred.

In that case, Eq. (1) may be converted to spherical coordinates. In a continuum limit, the energy per unit length $\mathcal{E}$ is

$$\mathcal{E} = -\frac{J s^2}{a^2} \int_{-L/2}^{L/2} dz \left[ 1 + \beta_z \cos \theta - \alpha \cos^2 \theta + \sin^2 \theta \left( -\frac{1}{2} a d\phi \left( \frac{dz}{a} \right)^2 + \delta_\phi d\phi + \frac{\beta_x}{\sin \theta} \cos \phi \right) \right].$$

Here $\phi(z)$ is the azimuthal angle of spin moments, and the dimensionless field $\beta_x \equiv \mu_B B_x/J a$. A stationarity condition on $\mathcal{E}$ for $\phi(z)$ yields a sine-Gordon equation with solution$^{19}$,

$$\phi(z) = \phi_0 - 2\arctan \left( \frac{\beta_x}{\sin \theta} \frac{z}{ka} \right),$$

where $\phi_0$ is the initial angle, $am$ is the Jacobi amplitude function, and $k$ is an elliptic modulus, chosen to minimize

FIG. 1. (a-d) Comparison of calculated $M_{||}$ (red lines) with experiments (blue circles) as a function of applied field $H$. A schematic of the measurement is shown in the inset of (a). $\theta_B$ is fixed at (a) $90^\circ$, (b) $27^\circ$, (c) $12^\circ$, and (d) $0^\circ$. (e) The left-handed spin structure of Cr$_{1/3}$NbS$_2$ calculated from Eq. (3), when $\theta_B = 90^\circ$ and $B \rightarrow B_c$ from below. At this field, the magnetic unit cell has grown to more than double its zero-field length $\lambda$. Two solitons are visible.
Substituting this result into Eq. (2) yields

$$\mathcal{E} = -\frac{J s^2}{a} \left[ 1 - \alpha \cos^2 \theta + \beta_z \cos \theta 
+ \beta_x \sin \theta \left( \frac{2}{k^2} + \frac{\pi \delta}{k K(k)} \sqrt{\frac{\sin \theta}{\beta_x}} \frac{4E(k)}{k^2 K(k)} - 1 \right) \right]$$

(4)

where $K$ and $E$ are the complete elliptic integrals of the first and second kind. In this form, $\mathcal{E}$ can be numerically minimized to determine $\theta$ and $k$. For that we used an interior-point algorithm. This fixes $\phi(z)$ as well as the components of the magnetization, $M_x = \mu_C \sin \theta \left( \frac{2}{k^2} - 1 - \frac{2E(k)}{k^2 K(k)} \right)$, and $M_z = \mu_C \cos \theta$.

The red lines shown in Fig. 1 (a-d) are made by combining these results to find $M_\theta$. Fig. 2 shows their variation over the entire parameter space of interest, with $M_x$ in (a), $M_z$ in (b), and $M_\theta$ in (c).

The field for which the length of the magnetic unit cell diverges corresponds to the soliton lattice transition. Its magnitude depends on $\theta_B$, which we write as $B_c(\theta_B)$. In Fig. 2 (a), $B_c(\theta_B)$ is clearly evident in the sudden increase in $M_x$. Throughout this letter, we denote $B_c(\theta_B = 90^\circ)$ by $B_c$. When $\cos \theta_B \ll 1$, the model predicts a critical field $B_c(\theta_B) \approx B_c/\sin \theta_B$, shown as the dashed black line in Fig. 2 (a). Only as $\theta_B \to 0$ does the calculated $B_c(\theta_B)$ begin to depart from this estimate.

Note that when $\theta_B < 90^\circ$ the magnetization in the $x$ direction grows rapidly through the soliton lattice transition at $B_c(\theta_B)$, but then decreases slowly as $B$, the magnitude of $B$, increases. This corresponds to a polarization of all moments in (or close to) the $ab$-plane, followed by a slow tilting of the spins out of the plane and along $B$. This effect is most noticeable when $B$ is near to but distinct from the $c$-axis. In this regime, $M_x/\mu_C$ can approach unity at $B = B_c(\theta_B)$, but when $B \gg B_c(\theta_B)$, $M_x(B)/\mu_C \to \sin \theta_B \approx \theta_B \ll 1$.

We return now to the discussion of the constant $\theta$ approximation used with Eq. (1). This is expected to be valid when $B_c(\theta_B) \ll B_c(\theta_B = 0)$, as this means the helix will polarize in the $ab$-plane via a soliton lattice transition before spins develop appreciable components out of the plane. In Cr$_{1/3}$NbS$_2$ this criteria is fairly unrestrictive, as the two fields of $B_c(\theta_B)$ and $B_c/\sin \theta_B$ are equal only when $\theta_B \approx \arcsin(\frac{2}{B_c(\theta_B = 0)}) \approx 4^\circ$. When the assumption is relaxed and $\theta(z)$ is allowed to vary with $z$, the effect is minimal: as $B$ approaches $B_c$ at a given $\theta_B$, moments whose $x$ components align with $B_x$ tilt slightly downward on average, inclining toward the $ab$-plane, while moments with $x$ components opposite $B_x$ tilt slightly upward. When $\theta_B = 0$, however, the model is again accurate as $\theta(z)$ is constant through the conical transition.

The agreement in $B_c(\theta_B)$ between the model and these measurements is further evidence of electrical transport’s sensitivity to the magnetic structure in this material. The markers in Fig. 2 (c) are measurements of $B_c$ as a function of $\theta_B$ deduced from magnetization (black circles) and $ab$-plane magnetoresistance (white squares) reported previously. Interestingly, $M_\parallel/\mu_C$ is less than one for a wide range of angles $\theta_B$, even when the magnetic field is of the order of several tesla. This is visible in the broad and shallow dip seen in the center of Fig. 2 (c), and could be relevant to experiments operating in this regime.

In summary we presented a spin model to describe the ground state magnetic structure of Cr$_{1/3}$NbS$_2$ in a magnetic field applied in arbitrary direction. We also detailed an efficient method to calculate that ground state, along with its magnetization. Comparison with experiments
reveals that the model captures the complex spin structure in this material. This work supports future efforts to engineer technologies that rely on the manipulation of spin structures in the presence of high magnetoanisotropy, like that of Cr_{1/3}NbS_2.

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