Controlling the Topological Sector of Magnetic Solitons in Exfoliated Cr$_{1/3}$NbS$_2$ Crystals


1Department of Quantum Matter Physics and Group of Applied Physics, University of Geneva, 24 quai Ernest-Ansermet, CH-1211 Geneva, Switzerland
2Key Laboratory of Flexible Electronics (KLOFE) & Institute of Advanced Materials (IAM), Jiangsu National Synergetic Innovation Center for Advanced Materials (SICAM), Nanjing Tech University (NanjingTech), 30 South Puzhu Road, Nanjing 211816, China
3Institute of Physics, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland
4Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee 37996, USA
5Theoretical Division, T-4 and CNLS, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
6Department of Applied Physics, University of Tokyo, Hongo, 7-3-1, Bunkyo, Tokyo 113-8566, Japan
7Nanofabrication Research Laboratory, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6493, USA
8Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA
9Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
10Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996, USA

We investigate manifestations of topological order in monoaxial helimagnet Cr$_{1/3}$NbS$_2$ by performing transport measurements on ultrathin crystals. Upon sweeping the magnetic field perpendicularly to the helical axis, crystals thicker than one helix pitch (48 nm) but much thinner than the magnetic domain size (~1 μm) are found to exhibit sharp and hysteretic resistance jumps. We show that these phenomena originate from transitions between topological sectors with a different number of magnetic solitons. This is confirmed by measurements on crystals thinner than 48 nm—in which the topological sector cannot change—that do not exhibit any jump or hysteresis. Our results show the ability to deterministically control the topological sector of finite-size Cr$_{1/3}$NbS$_2$ and to detect intersector transitions by transport measurements.

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The properties of many electronic systems are characterized by topological indices that allow all possible states to be grouped into distinct topological sectors [1–5]. Since topological indices assume discrete values, changes in the topological sector can only occur through abrupt transitions that can be detected experimentally. Investigating these transitions under controlled conditions and probing properties associated with “topological order,” however, is not simple, as it requires the ability to tune the state of the system in a predefined topological sector, i.e., to deterministically set the topological indices of the system by acting on experimental parameters. Here we show that this level of control can be achieved by means of magnetoresistance measurements on mechanically exfoliated crystals of the monoaxial chiral helimagnet Cr$_{1/3}$NbS$_2$.

Our experiments build on earlier work that has established the key properties of Cr$_{1/3}$NbS$_2$, a layered material consisting of alternating NbS$_2$ and Cr planes [see Fig. 1(a)] [6–17]. At low temperature, the $S = 3/2$ spins on the Cr atoms order ferromagnetically in each plane, forming a helix that winds around the direction perpendicular to the planes [Fig. 1(b)], the zero-field helix pitch $L_0$ is 48 nm [9,11–14]. Lorentz microscopy has shown that the in-plane magnetic field $B$ causes the helix to deform, resulting in a so-called chiral soliton lattice [12]. The lattice [Fig. 1(b)] consists of narrow regions in which the spins make a complete $2\pi$ revolution (the solitons), separated by stretches of ferromagnetically aligned spins, whose extension—which determines the lattice period $L_B$—increases upon increasing $B$ [Fig. 1(c)] [12]. The observed microscopic evolution of the helix, as well as the detailed magnetic response of bulk crystals, conform quantitatively to a theoretical (one-dimensional) model proposed by Dzyaloshinskii over 50 years ago, comprising Heisenberg and Dzyaloshinskii-Moriya interactions between nearest neighboring spins (besides the Zeeman term in the presence of a magnetic field and a magnetic anisotropy term that forces the spins to point perpendicularly to the helix direction) [18,19]. Whenever it is spatially confined, the chiral soliton lattice in Cr$_{1/3}$NbS$_2$ is predicted to exhibit interesting phenomena [20–23]. Some of these phenomena have been observed recently in a small specimen (10 μm in linear dimensions) cut from bulk crystals, in which confinement originates from the presence of magnetic domains extending for approximately 1 μm in the helix direction [22]. Upon increasing $B$, the separation between solitons increases, so that their total number in each finite-size domain decreases. Since the number of solitons corresponds to the total spin winding number that is a topological index, the soliton number can only change through discrete transitions. These transitions
FIG. 1. (a) Structure of layered Cr$_{1/3}$NbS$_2$. The $c$-axis lattice constant is 1.21 nm. (b) Schematic illustration of the spin configurations along the $c$-axis in bulk Cr$_{1/3}$NbS$_2$ (the arrows represent the magnetization in the Cr planes). Upon increasing magnetic field ($B$) in the $ab$ plane, the $B = 0$ magnetic helix (top) is gradually transformed into a chiral soliton lattice with period $L_B$ (middle), finally becoming ferromagnetic above the critical magnetic field ($B_c$; bottom). (c) In the process, the period $L_B$ increases and diverges close to $B_c$ (plot obtained from theoretical calculations based on the model discussed in the main text). (d) Magnetoresistance, $\Delta R/R(0) \equiv (R(B) - R(0))/R(0)$, of bulk Cr$_{1/3}$NbS$_2$ measured at 250 mK. The blue or red curve corresponds to measurements performed upon sweeping the in-plane field $B$ in the direction indicated by the blue or red arrow. An abrupt resistance drop at $B_c \sim 0.17$ T with no hysteresis is observed.

have been detected by analyzing Lorentz microscope images [22]. It was argued that hysteresis and a sequence of jumps present in the magnetoresistance of small specimen, but absent in bulk crystals [see Fig. 1(d)], are a transport manifestation of the changes in soliton number [22,23]. This conclusion is very interesting, as it implies the ability to probe topological aspects of the magnetic state of Cr$_{1/3}$NbS$_2$ by monitoring transport properties. Its validity is, however, unclear because the magnetoresistance jumps could not be directly linked to specific changes in the soliton configuration, and because it was not ruled out that the jumps may originate from domain switching.

To avoid these problems, we investigate these same phenomena by working with Cr$_{1/3}$NbS$_2$ crystals much thinner than the magnetic domain size. Figure 2(a) shows optical microscope images of a selection of crystals having thickness ($t$) between 28 and 300 nm, whose surfaces are parallel to the NbS$_2$ and the Cr planes. The magnetic helix is oriented perpendicular to the substrate, so that the crystal thickness determines the number of solitons present at $B = 0$. We produced these crystals, up to 500 times thinner than the specimen studied in Ref. [22], by means of mechanical exfoliation, following the same procedure used to extract graphene from graphite. Exfoliation is more difficult for Cr$_{1/3}$NbS$_2$ because of strong chemical bonds between the Cr and S atoms; nevertheless, atomic force microscope images [see Figs. 2(b) and 2(c)] show that the crystal surface is flat and uniform.

Transport through Cr$_{1/3}$NbS$_2$ crystals as thin as the ones discussed here had not been investigated earlier and it is important to identify which properties depend on thickness and which do not. Figure 2(d) shows that all exfoliated crystals exhibit the same temperature dependence of the resistance (identical to that of the bulk). A pronounced decrease in resistance starts around $T = 130$ K, corresponding to the transition temperature $T_c$ to the helimagnetic state. Since $T_c$ is determined by the strength of the microscopic interactions between nearest neighboring spins (and—in the range investigated here—not by the thickness) this behavior is not surprising [24]. Nevertheless, the excellent reproducibility upon varying the thickness is worth commenting on, as it indicates the absence of any significant material degradation (not warranted a priori for thin crystals exposed to air during exfoliation and device fabrication).

The relative change in resistance upon the application of an in-plane magnetic field, $\Delta R/R(0) \equiv (R(B) - R(0))/R(0)$, is shown in Figs. 3(a)–3(d) for several crystals containing one [(a),(b); $t = 57$ nm and 79 nm, respectively], two [(c); $t = 113$ nm], and five [(d); $t = 280$ nm] complete helix periods at $B = 0$. The behavior is representative of what we observed in more than 10 devices realized with crystal having a thickness between approximately 50 and 300 nm, exhibiting common features and systematic trends. For these crystals, hysteresis in the magnetoresistance upon reversing the sweeping direction of the applied field is always present, and is accompanied by sharp jumps. The phenomena cannot originate from

FIG. 2. (a) Optical microscope images of exfoliated Cr$_{1/3}$NbS$_2$ crystals with thicknesses between 28 nm and 280 nm. (b) Atomic force microscope image of the 57-nm-thick crystal, showing a flat and uniform surface, as visible in the thickness profile (c) taken along the red line. (d) Temperature dependence of the resistance $R(T)/R(280K)$ for crystals of different thicknesses (see legend). All curves merge together and exhibit a resistance drop starting around 130 K, the bulk transition temperature of the helimagnetic state. The inset in (d) shows an optical image of a device, with nanofabricated Ti/Au contacts. In all images, the scale bar is 3 μm.
magnetic domains, since all crystals are significantly thinner than the domain size [22]. We find that the number of jumps tends to increase with increasing the crystal thickness, seemingly correlating with the number of complete periods present in the magnetic helix at $B = 0$ (determined by $t/L_0$ where $L_0$ is the helix pitch). For instance, the crystals in Figs. 3(a) and 3(b) contain one full period and exhibit one jump, the crystal with $t=113$ nm [Fig. 3(c)] contains two full periods and exhibits two jumps. The $t=280$ nm crystal contains five full periods at $B = 0$, and four jumps are observed, but the jump at $B \sim 0.17$ T appears to be smeared, suggesting that it may originate from two jumps that are not resolved individually. The magnetic field at which the last jump occurs systematically increases with increasing crystal thickness, as shown in Fig. 3(e). Finally, the total change in the relative magnetoresistance measured after the last jump decreases with increasing thickness [see Fig. 3(f); this same quantity vanishes in bulk crystals].

All the observed trends can be understood in terms of a theoretical model known to properly describe the magnetic state of Cr$_{1/3}$NbS$_2$ (the model is discussed in several papers and here we only recall the key conceptual aspects; see also the Supplemental Material [25]) [12,14–16,21,26,27]. The model enables the spin configuration to be determined through the minimization of the system energy expressed as a functional of the local magnetization. Its validity for Cr$_{1/3}$NbS$_2$ has been established by direct comparison with Lorentz microscopy experiments and magnetization measurements, which enable the model parameters to be extracted quantitatively [12,22]. The model has also been applied to strained MnSi thin films [28,29] [30] to interpret magnetotransport data closely related to the ones discussed here. In this context, it was assumed that all changes causing a better aligned spin configuration result in smaller measured resistance [25].

In this same spirit, we reproduce some of the known results by solving the model numerically as a function of $B$ for crystals of different thicknesses, and we use the numerical solutions to illustrate the aspects of the behavior that account for the experimentally observed trends [25]. Figures 4(a)–4(c) show the lowest energy spin configuration—represented by the $x$ component of the spin on the chromium atoms planes—calculated at $B = 0$ for crystals having $t=1.5$ $L_0$, 2.5 $L_0$, 5.5 $L_0$.
domain size, so that domain switching can be excluded as a possible origin of the phenomena. This and the agreement between observations and theoretically expected trends allow us to conclude that the observed jumps and the hysteresis do originate from transitions between states with a different soliton number. The use of exfoliated crystals enables us to obtain further evidence supporting this conclusion, as it allows us to investigate crystals thinner than the helix pitch, a regime that has not been explored so far. No jumps and hysteresis in the magnetoresistance should be expected, because in this regime the soliton number vanishes for all values of $B$, and no topological transition can occur. Figure 5 shows that this is indeed the case: in all Cr$_{1/3}$NbS$_2$ crystals thinner than the helix pitch at $B = 0$ only a negative magnetoresistance is observed due to the gradual spin alignment upon increasing $B$. No jumps or hysteresis are present. This drastic qualitative difference in behavior observed upon changing the crystal thickness by only a few nanometers is striking. It provides conclusive evidence that in Cr$_{1/3}$NbS$_2$ the topological sector of the system can be controlled and probed by transport measurements.

In summary, we have performed experiments on very thin exfoliated crystals showing that the number of solitons in Cr$_{1/3}$NbS$_2$ can be controlled by selecting the appropriate crystal thickness and by acting on the applied field. The experiments further show that changes in the number of solitons manifest themselves in hysteretic magnetoresistance jumps. These results imply that the topological sector of the system can be deterministically controlled and probed by transport measurements.

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[30] Bulk MnSi is not a chiral helical magnet, but MnSi thin films are in a narrow range of thicknesses, due to strain from the substrate [28,29].

[31] The B values at which the transitions occur depend on thickness that—for exfoliated crystals—cannot be deterministically controlled. This makes it difficult to compare the precise value of magnetic field for which a generic resistance jump is expected to occur with theory. Since devices of all thicknesses $t > t_0$ exhibit the jump corresponding to the transition to the ferromagnetic state, for this transition it is nevertheless possible to obtain enough data to make a statistically significant comparison.