Magnetic Correlations in the Quasi-2D Semiconducting Ferromagnet CrSiTe₃

T.J. Williams, A.A. Aczel, M.D. Lumsden, S.E. Nagler, and M.B. Stone
Quantum Condensed Matter Division, Neutron Sciences Directorate,
Oak Ridge National Lab, Oak Ridge, TN, 37831, USA

J.-Q. Yan and D. Mandrus
Materials Science & Technology Division, Physical Sciences Directorate,
Oak Ridge National Lab, Oak Ridge, TN, 37831, USA and
Department of Materials Science & Engineering,
University of Tennessee, Knoxville, TN, 37996, USA
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Intrinsic, two-dimensional ferromagnetic semiconductors are an important class of materials for overcoming the limitations of dilute magnetic semiconductors for spintronics applications. CrSiTe₃ is a particularly interesting member of this class, since it can likely be exfoliated down to single layers, where Tₓ is predicted to increase dramatically. Establishing the nature of the magnetism in the bulk is a necessary precursor to understanding the magnetic behavior in thin film samples and the possible applications of this material. In this work, we use elastic and inelastic neutron scattering to measure the magnetic properties of single crystalline CrSiTe₃. We find that there is a very small single ion anisotropy favoring magnetic ordering along the c-axis and that the measured spin waves fit well to a model where the moments are only weakly coupled along that direction. Finally, we find that both static and dynamic correlations persist within the ab-plane up to at least 300 K, strong evidence of this material’s two-dimensional characteristics that are relevant for future studies on thin film and monolayer samples.

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I. INTRODUCTION

Spintronics requires semiconductors that possess ferromagnetic properties, particularly those that can be manipulated into one- or two-dimensional forms. Much work has focused on dilute magnetic semiconductors, which offer a large range of magnetic properties. These compounds have been the subject of much debate since their properties are highly dependent on epitaxial growth conditions, dopant distributions and even how the dopants are incorporated into the overall band structure[1]. In an effort to overcome these limitations, there has been an emergence of research into the much less common spintronic candidate: intrinsically ferromagnetic semiconductors[2]. The compound CrSiTe₃ belongs to this class, having an electronic band gap of 0.38 meV and a ferromagnetic transition at Tₓ = 33 K.

Much of the recent work on CrSiTe₃ has been in the form of theoretical calculations focused on the band structure and semiconducting properties, particularly calculations of the monolayer properties. Generalized gradient approximation (GGA) calculations on bulk CrSiTe₃ have calculated a bulk band gap of 0.6 meV, close to the experimental value, with the gap formed by the splitting of the Cr and Te levels[3]. These calculations correctly predict a ferromagnetic ground state that induces a slight spin polarization in the Si and Te atoms[4]. When the spins were assumed to be Ising-like and aligned along the c-axis, local density approximation (LDA) calculations predicted a nearest-neighbor exchange J = -0.58 meV and transition temperature Tₓ = 23 K[5]. Experimentally, the structure has been determined in earlier work[6]. Hexagonal planes of Cr³⁺ (S = 3/2) atoms are stacked along the c-axis, with each atom octahedrally-coordinated by Te. The Te-Te bond lengths are ≈ 3.15 Å in the ab-plane and ≈ 3.48 Å out of the plane, suggesting a very small octahedral distortion[7,8]. Previous neutron measurements also found a magnetic transition at 32.1 K corresponding to the Cr³⁺ moments ordering ferromagnetically along the c-axis[9]. Magnetic fluctuations were observed above Tₓ, and inelastic measurements below Tₓ showed modes with no dispersion along L, both indicative of two-dimensional behavior[10,11]. These measurements found a spin gap of nearly 6 meV, which was taken as evidence for Ising behavior. However, these measurements were not able to provide a conclusive origin for the excitations and the exchange parameters were not able to be determined. Furthermore, the peaks seen in this work were quite weak, casting doubt on the accuracy of the determined magnetic dynamics.

The resurgent interest in studying CrSiTe₃ has been driven by its applicability to spintronics. This has been prompted by the speculation that it may be possible to exfoliate the material down to single layers[12], which has been predicted to have the desirable effect of increasing the band gap to ≈ 0.59 eV[13] and the Curie temperature to ≈ 92 K[14]. These features have quickly created the need to understand the electronic and magnetic properties of CrSiTe₃.
FIG. 1. (color online) (a) The crystal structure of CrSiTe$_3$. The Cr$^{3+}$ ions form hexagonal arrangements in the $ab$-plane, which are then stacked along the $c$-axis in an $ABC$-type stacking. This creates two magnetically-inequivalent Cr sites, shown in red and blue. The Cr-I site (blue) has a Cr-II atom (red) above and a Si$_2$ pair (green) below, while it is reversed for the Cr-II site. Below $T_C = 33.2$ K, the Cr$^{3+}$ spins align ferromagnetically along the $c$-axis, due to a single-ion anisotropy, $D_z$. The nearest-neighbour exchanges out to 8 Å are shown in the figure, which includes 3 in-plane exchanges, $J_{ab1}$, $J_{ab2}$ and $J_{ab3}$, and one out-of-plane exchange, $J_c$. (b) The structure of a single layer is shown, highlighting the different exchange interactions in this plane.

II. EXPERIMENTAL DETAILS

CrSiTe$_3$ single crystals were grown using a self-flux technique, as previously reported$^{10}$. CrSiTe$_3$ is rhombohedral, crystallizing in the space group $R-3$, which was confirmed with x-ray diffraction measurements on the samples used in this study. The crystal structure is shown in Fig. 1. Susceptibility measurements showed a ferromagnetic transition at $T_C = 33(1)$ K$^{10}$. The large $c/a$ ratio and the reasonably accessible magnetic transition make this a good candidate to study for its application as a two-dimensional spintronic material. In order to fully characterize the nature of the magnetic correlations, including their lower dimensional properties, neutron scattering measurements were performed at the HB-3 and CG-4C (CTAX) triple axis spectrometers of the High-Flux Isotope Reactor, as well as the SEQUOIA time-of-flight spectrometer at the Spallation Neutron Source of the Oak Ridge National Laboratory.

Five single crystals of total mass 4.1g and a mosaic of 2.25° were coaligned in the [H 0 L] scattering plane for use in the SEQUOIA and cold neutron triple axis (CTAX) experiments, while the largest single crystal (mass ≈ 1.2g) was used for the HB-3 experiment. The HB-3 measurements were performed in a closed-cycle refrigerator with a base temperature of 4.0 K using a fixed final energy of 14.7 meV. PG (002) monochromator and analyzer crystals were used, with PG filters and the collimation was 48'-40'-40'-120'. The SEQUOIA measurements were also performed in a closed-cycle refrigerator using fixed incident energies of 30 and 65 meV. The CTAX measurements were performed in a He-4 cryostat with a base temperature of 1.5 K using fixed final energies of 3 and 5 meV. This experiment used a PG002 monochromator, a Be filter and collimation settings of guide-open-80'-open.

III. ELASTIC NEUTRON SCATTERING

Fig. 2(a) shows the temperature dependence of the (1 0 1) Bragg peak, which is both a nuclear and a magnetic peak, but the magnetic contribution is approximately 10 times larger. The red line is a fit to a critical exponent, which yields a transition temperature of 33.2(1) K and a critical exponent, $\beta = 0.151(12)$. This is close to the value expected for a two dimensional transition ($\beta_{2D,\text{Ising}} = 0.125$), and well below the values expected for a three dimensional transition ($\beta_{3D,\text{Ising}} = 0.326$ and $\beta_{3D,\text{Heisenberg}} = 0.367$). The low value of critical exponent is likely a consequence of strong two-dimensional correlations in this material. The long-range order below 33.2 K is, however, three-dimensional in nature, with the spins aligned ferromagnetically along the $c$-axis direction. This was confirmed by measuring 39 magnetic peaks in the [H 0 L] scattering plane, whose intensities below $T_C$ were consistent with $c$-axis ordering. This is in agreement with previous work$^{11}$.

In addition to the magnetic diffraction peaks that emerge below $T_C$, diffuse scattering develops around the Bragg peaks, with an intensity that has a maximum at $T_C$, shown in the inset to Fig. 2(a). This diffuse scattering arises due to the in-plane correlations that are present, particularly above the ordering temperature. In order to study the two-dimensional static correlations within the $ab$-plane in more detail, a series of two-axis measurements were performed on the HB-3 instrument. This was done by measuring along $H$ about the wavevector (1 0 0.545), chosen such that the $c$-axis was parallel to

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FIG. 2. (color online) (a) The temperature dependence of the measured intensity at the (1 0 1) Bragg peak. This peak is both nuclear and magnetic, but the magnetic intensity is larger by approximately a factor of 10. A fit to this curve gives a transition temperature of $T_C = 33.2(1)$ K and a critical exponent $\beta = 0.151(12)$. (inset) Scans through the (1 0 1) Bragg peak along $H = 10$ K (black squares), 33 K (red circles) and 50 K (blue triangles), plotted on a logarithmic scale. This shows the sharp increase in the intensity at (1 0 1), corresponding to the 3D order, while the diffuse scattering, corresponding to the 2D correlations, is most intense at the transition. (b) The temperature dependence of the in-plane correlation length, determined from the 2-axis scans described in the text. The line is a guide to the eye. We note that the in-plane correlation length is greater than the nearest-neighbor Cr-Cr distance at all temperatures measured, suggesting that the in-plane magnetic correlations are important well above the bulk ordering temperature.

The two-axis measurements are peaked at $H = 1$ for all temperatures measured and we find that the intensity of the peak is largest at $T_C$. The width of the peak can be used to calculate the in-plane correlation length, which is plotted in Fig. 2b. As expected, the correlation length peaks at $T_C$, but decays as the temperature increases. The correlation length remains larger than the nearest-neighbor distance at all temperatures measured, up to 250 K. This suggests that short-range correlations between the moments in the $ab$-plane exist well above the ordering transition.

IV. SPIN WAVE MEASUREMENTS

Fig. 3a) and (c) show the spin wave dispersions measured on the SEQUOIA spectrometer, with an incident energy $E_i = 30$ meV. Panel (c) shows the dispersion along ($0 0 L$) with integration ranges of 0.4 r.l.u along $H$ and $K$. This shows that the dispersion is very weak along the $c$-axis, and that the minimum in the spin wave dispersion is below 1 meV at the zone center, (0 0 -3). Since the dispersion along $L$ is relatively weak, the spectrum shown in Fig. 3a) was constructed by integrating over the full $L$-range, $-12 \leq L \leq 12$, while using an integration range of 0.2 r.l.u. along $K$. This shows two spin wave bands, one peaking at $\approx 8$ meV and the higher band peaking at $\approx 15$ meV, which meet at the zone boundaries. The data along ($H 0 0$) using a smaller integration range for $L$ showed identical dispersions, supporting the conclusion that the spin waves are close to two-dimensional. The spin waves were modeled with a Hamiltonian given by:

$$H = J_{ab1} \sum_i \vec{S}_i \cdot \vec{S}_{i+1} + J_{ab2} \sum_i \vec{S}_i \cdot \vec{S}_{i+2} + J_c \sum_i \vec{S}_i \cdot \vec{S}_{i+3} + J_{ab3} \sum_i \vec{S}_i \cdot \vec{S}_{i+4} - D_z \sum_i \vec{S}_i^2$$

where the $J$’s are exchange constants between neighboring spins, $\vec{S}_i$, $D_z$ represents a single-ion anisotropy and the spins are assumed to be localized Cr$^{3+}$, $S = 3/2$ moments, as shown in Fig. 3. The spin gap is below the resolution of the spectrometer and the $c$-axis dispersion is too weak to be accurately determined from this data. The magnetic anisotropy necessitates a non-zero spin gap, but the SEQUOIA measurements have shown that it is less than 1 meV. This is consistent with magnetization data taken to characterize the sample, which observes a coercive field of 1.5 T and very little magnetic hysteresis. Additionally, DFT calculations based on purely van der Waals interactions have found an entropy difference of $\approx 20 \mu$eV, favoring $c$-axis ordering. This is very close to the value obtained for the single-ion anisotropy, while both quantities are close to the resolution limits of the respective technique. The fits described below produce the simulations shown in Fig. 3b) and (d). These show very good agreement with the experimental data, but we do observe a discrepancy at the integer values of $H$. At these points in the experimental data,
FIG. 3. (color online) The panels on the left show the spin waves of CrSiTe$_3$ measured on the SEQUOIA spectrometer at $T = 10$ K, while the panels on the right show the calculated spin wave dispersions, along the $(H\ 0\ 0)$ direction (panels (a) and (b)) and the $(0\ 0\ L)$ direction (panels (c) and (d)). The calculated patterns use the model and exchange parameters described in the text, producing very good agreement with the experimental data.

we observe broadening of the optical mode and intensity shifts to the acoustic mode. These features are not seen in the simulation, though the origin of this difference is unclear.

In order to measure the magnitude of the spin gap and the dispersion out of the plane, measurements were performed on the cold neutron triple axis (CTAX) instrument at Oak Ridge National Laboratory. The resolution of a constant-$Q$ scan was insufficient to separate the gapped mode from the elastic scattering. To extract the spin gap value, a series of constant-$E$ scans was performed, shown in Fig. 4(a). By assuming that the energy of the spin waves, $E \propto |q|^2$ in the long wavelength limit, we can extract an approximate measure of the gap. The data was fit in multiple zones, as shown in Fig 4(b). This gave a value of the spin gap, $\Delta = 0.075(24)$ meV, allowing a measurement of the single-ion anisotropy, $D_z = \Delta/2S = 0.0252(80)$ meV. The single-ion anisotropy arises from the small $c$-axis distortion in the Te octahedra surrounding the Cr$^{3+}$ ions. This nearly perfect octahedral environment makes spin-orbit effects very weak and the Cr-Cr bond is a point of local centrosymmetry, ruling out Dyaloshinskii-Moriya interactions as an origin for the energy gap. This value of $D_z$ was then used with constant-$Q$ scans at the zone boundary to determine the out-of-plane coupling, shown in Fig. 4(c). This gave a value for the $c$-axis coupling, $J_c = -0.730(96)$ meV.

These values were then fixed for the purposes of fitting the SEQUOIA data to obtain the in-plane exchange constants. The fitting was performed using the fitting routines built into the Horace software package which used
the dispersions calculated by SpinW as its model, comparing the intensities of the data and calculation over the entire range in the $Q$-$E$ slice. This allowed the in-plane exchange constants to be fit simultaneously, utilizing the two-dimensional dataset shown in Fig. 3(a). The values of these three exchange constants obtained from fitting the data shown in Fig. 3(a) to Eq. [4] as well as those determined from the CTAX measurements, are given in Table I.

| Exchange  | Description                     | Distance (Å) | Value (meV)        |
|-----------|---------------------------------|--------------|--------------------|
| $J_{ab1}$ | $1^{st}$ NN in-plane            | 3.907        | -1.27(23)          |
| $J_{ab2}$ | $2^{nd}$ NN in-plane            | 6.768        | -0.10(50)          |
| $J_{ab3}$ | $3^{rd}$ NN in-plane            | 7.814        | -0.285(73)         |
| $J_c$     | $1^{st}$ NN out-of-plane        | 6.852        | -0.730(96)         |
| $D_z$     | Single Ion Anisotropy           |              | 0.0252(80)         |

TABLE I. The exchange constants obtained from fitting the inelastic measurements using Eq. [4]. To properly describe the spin waves, it was necessary to use couplings up to 8 Å, which requires 3 in-plane interactions and one out-of-plane. Consistent with the quasi-2D nature of the material, the value of $J_c$ gives a very small dispersion along the c-axis and, as expected from the very small octahedral distortion, the value of the single-ion anisotropy, $D_z$, is very close to zero.

As expected for a quasi-two-dimensional system, the nearest neighbor in-plane coupling, $J_{ab1}$, is the dominant interaction and is ferromagnetic, as are the other in-plane exchange constants. The ferromagnetic origin of the in-plane terms is likely due to superexchange mediated by the Te ions, shown in Fig. 1(b). The Cr$^{3+}$ ion direct exchange is antiferromagnetic, though the large nearest-neighbor Cr-Cr distance of 3.987 Å should make this a weak effect. Finally, the Cr octahedra are edge-sharing and the Cr-Te-Cr angle is 88.8°, very close to perfectly orthogonal. This arrangement of the ions would suggest that superexchange of the type described by the Goodenough-Kanamori rules would be ferromagnetic, consistent with the observations of ferromagnetism in other layered Cr$^{3+}$ compounds with similar superexchange pathways. The other exchange constants are also ferromagnetic, including the out-of-plane coupling, $J_c = -0.730(96)$ meV, which is weaker than the in-plane coupling. First principles DFT calculations on monolayer CrSiTe$_3$ suggest that the $2^{nd}$ and $3^{rd}$ nearest-neighbor in-plane interactions are Cr-Te-Cr double-superexchange interactions, leading to the small values we observe for $J_{ab2}$ and $J_{ab3}$. Furthermore, the observation that $|J_{ab2}| < |J_{ab3}|$ was also predicted by the DFT calculations. There are two double-superexchange pathways that contribute to $J_{ab2}$, one of which is ferromagnetic and the other antiferromagnetic, making it a very weak exchange.

The neutron scattering measurements are also consistent with the theoretical predictions of an enhanced transition temperature in CrGeTe$_3$ and CrSnTe$_3$, as well as monolayer CrSiTe$_3$. Despite the spins being only weakly Ising-like, the easy-axis spin anisotropy created by the imperfect Te octahedra still allows for ordering in two-dimensions, such as when the materials are reduced to monolayers. The loss of the out-of-plane coupling is compensated by an increased in-plane ferromagnetic contribution, as predicted by several theoretical studies that compare bulk CrSiTe$_3$ to its monolayer version and the chemically-substituted CrGeTe$_3$ and CrSnTe$_3$.

The increased ferromagnetic contribution within the $ab$-plane arises due to the increase of the nearest-neighbor Cr-Cr distance in all of these cases. This serves to re-
duce the Cr-Cr direct exchange, which is antiferromagnetic, while pushing the Cr-Te-Cr bond angle closer to 90°. This increases the effect of the ferromagnetic superexchange, resulting in the increased values of T\textsubscript{C}[13].

Inelastic neutron scattering measurements performed above the ferromagnetic transition are shown in Fig. 5. Measurements at 40 K along the (H 0 0) direction are presented in Fig. 5(a). This data indicates that the spin waves are still present within the planes, but have been significantly broadened. The minima in the spin waves seen at (1 0 0) and (2 0 0) in Fig. 3(a) have disappeared, as they arose from the weak dispersion along the L-direction. This indicates that there is no dispersion along L above T\textsubscript{C}, and the dynamic correlations are only present within the ab-planes at these temperatures. To track the temperature evolution of the spin waves, constant-E measurements were performed along (H 0 0) at E = 1.75 meV and (0 0 L) at E = 1.5 meV as a function of temperature. These are shown in Fig. 5(b) and (c), respectively. We see that the spin waves along L disappear abruptly above T\textsubscript{C}, but that the broadened spin waves exist at the same \( \vec{Q} \) along (H 0 0) well above the ordering transition. As in the case of the static correlations, these dynamic correlations within the plane persist up to at least 300 K.

V. CONCLUSIONS

We have measured CrSiTe\textsubscript{3} using elastic and inelastic neutron scattering. These measurements observe bulk ferromagnetism below T\textsubscript{C} = 33.2(1) K and the critical exponent of the neutron scattering intensity was found to be \( \beta = 0.151(2) \), close to the value expected for a two-dimensional system. The magnetic Bragg peaks also exhibited a diffuse component, the intensity of which peaks at T\textsubscript{C}, indicating two-dimensional ferromagnetic correlations are present in the ab-plane above the ferromagnetic transition. To characterize these correlations, two-axis measurements were performed, which provided an effective quantitative measure of the in-plane correlation length. As expected, the correlation length diverged at T\textsubscript{C} but was still larger than the nearest-neighbor Cr-Cr distance at all temperatures measured, up to 250 K. These measurements suggest that while CrSiTe\textsubscript{3} only orders in three dimensions below 33 K, there are strong two-dimensional static correlations that persist up to at least room temperature.

In contrast to the previous assumption of Ising spins, the spin wave measurements suggest that the spins are very Heisenberg-like, with a very small spin gap of 0.075(24) meV due to a small single-ion anisotropy. X-ray diffraction measurements[14] and DFT calculations[15] find evidence of van der Waals interactions creating a very small octahedral distortion along the c-axis, creating the anisotropy. We find that the dominant interaction, \( J_{ab1} = -1.27(23) \) meV, is likely due to the superexchange mediated by the Te ions. The 2nd and 3rd nearest-neighbor in-plane interactions are much weaker, likely a result of double-superexchange interactions.

Above T\textsubscript{C}, no spin waves are present along the L direction, but they persist in a broadened form along the H direction. Measurements at 40 K along (H 0 0) showed broadened spin waves consistent with dynamics confined to the ab-planes, a clear indication of two-dimensional behavior, while temperature-dependent measurements show an inelastic signal that is visible up to 300 K. The latter finding suggests that there are dynamic in-plane correlations that persist up to at least room temperature as well. Considering these results in conjunction with the neutron diffraction measurements, we find that both static and dynamic in-plane magnetic correlations
exist up to at least 300 K, ten times $T_c$. This illustrates the importance of two-dimensional correlations in these materials and may suggest that similar physics is relevant in the isostructural compounds CrGeTe$_3$ and CrSnTe$_3$, as well as for monolayer CrSiTe$_3$, where it has been predicted that separation and decoupling of the hexagonal Cr layers puts an increased emphasis on the two-dimensional correlations. Taken together with the dramatic increase in the magnetic transition temperature driven by a separation of the intralayer Cr atoms in these materials, this suggests that spintronic devices that make use of these semiconductors may be capable of supporting intrinsic ferromagnetism at temperatures approaching, or even surpassing, room temperature.

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* williamstj@ornl.gov

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