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Direct Observation of 2D Magnons in Atomically Thin CrI_3

INTRODUCTION:

A magnon is the bosonic quasiparticle associated with spin wave excitations in magnetically ordered materials. The field of magnonics seeks to create logic circuits and other devices using magnons to transport spin information. Magnonic circuits have several potential advantages over electronic circuits: since magnons propagate with no dissipation losses, are easily tunable using applied magnetic fields, perform with low noise in low-power applications, and have wavelengths several orders of magnitude shorter than those of electromagnetic waves in practical scenarios, they are promising candidates for use in the development of miniaturized, low-power devices[1]. Consequently, the study of magnons has attracted intense interest of late. Recent studies of these systems have focused primarily on magnons in the gigahertz regime, which have limited applicability in the design of ultrafast devices. Antiferromagnetic magnons, however, can reach the terahertz regime and provide a chance to study ultrafast magnetization dynamics[2].

Chromium triiodide (CrI_3) is an antiferromagnet with several properties that make it a promising material for the study of magnons. Its unique layer-dependent magnetic phases—ferromagnetic (FM) in the monolayer below the Curie temperature and antiferromagnetic (AFM) in the bilayer—enable the study of magnon dynamics in both FM and AFM states. In addition, the crystal's high anisotropy could allow for high frequency magnons. Furthermore, CrI_3 can be incorporated into Van der Waals heterostructures, allowing for the study of magnons in novel situations (e.g., electrical gating, proximity effects).

Results from recent neutron scattering experiments indicate that magnon modes are expected in CrI_3 at approximately 1 meV (the 'acoustic' mode) and approximately 19 meV (the 'optical' mode)[3]. The acoustic mode corresponds to in-phase oscillations between two sublattice Cr spins, while the optical mode corresponds to out-of-phase oscillations. However, true 2D magnons (i.e., in monolayer or bilayer) have not yet been directly studied through inelastic light scattering.

METHODS:

To study spin waves in atomically thin CrI_3, we encapsulated monolayer and bilayer CrI_3 between 20-30 nm flakes of hexagonal boron nitride (hBN). Using mechanical exfoliation techniques, I produced the hBN flakes used for encapsulation, and assisted in the fabrication of the heterostructure. The resulting heterostructure (Fig. 1a) was then mounted on a cold finger cryostat, which can cool the sample to temperatures as low as 15K. I then used polar reflective

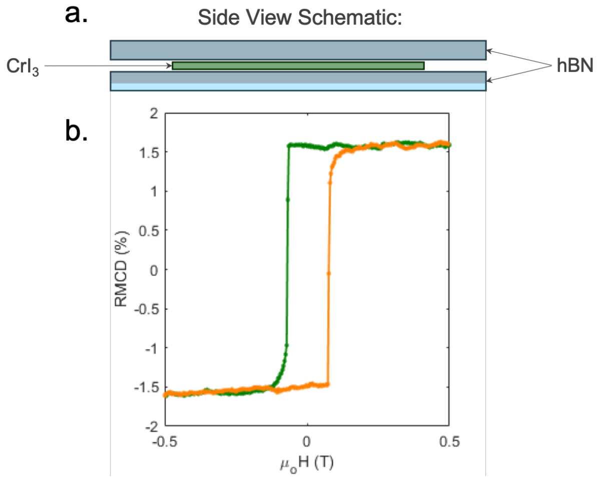

Fig. 1. a. Side-view schematic of double hBN encapsulated CrI_3. **b.** RMCD sweep of CrI_3 monolayer showing clear hysteresis loop—this is indicative of magnetization.

magnetic circular dichroism (RMCD) measurements to confirm the magnetic behavior of the samples (Fig. 1b), before rearranging the optical setup to perform magneto-Raman measurements on the samples. In these measurements, I focused 632.8 nm light (emitted by a HeNe laser) down to a 3 μm beamspot, using 400 μW of laser power to study the monolayer and bilayer samples and 1.5 mW to study the thin bulk. I used MATLAB to control the magnet and spectrometer, and to analyze and plot the data I collected.

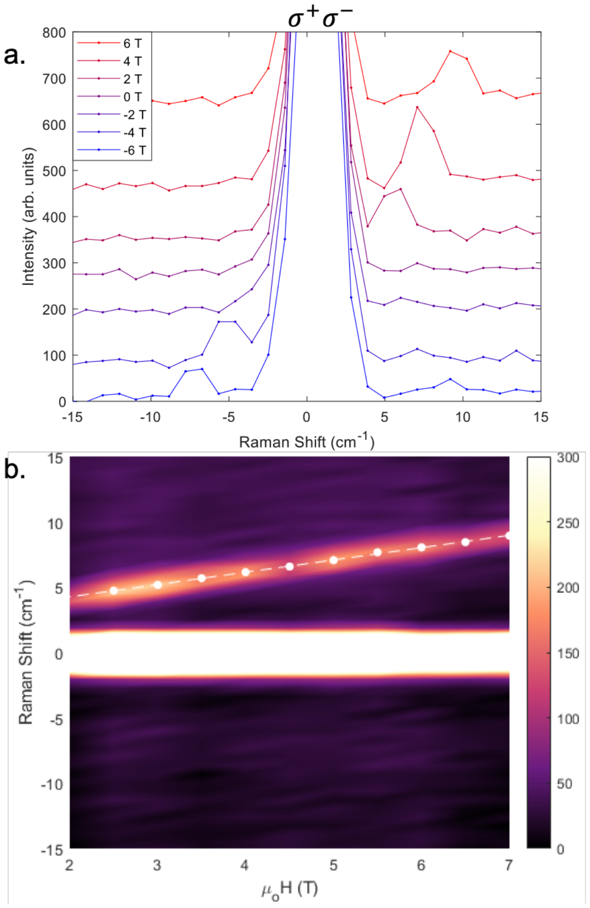

Fig. 2. Monolayer CrI$_3$. **a.** Stack plot showing intensity of magnon signal vs Raman shift for applied magnetic fields from -6T to 6T (2T increments). Note that the small peak (magnon) shifts with the applied field—this is characteristic of a magnon. **b.** Color plot showing magnon (sloped, dimmer line) with a fit for the g factor overlaid (dashed white line).

RESULTS:

In monolayer CrI$_3$, we see a sharp one-magnon feature at a zero-field energy of ~.3 meV, which blue-shifts with a g factor of 2 in accordance with theoretical predictions (Fig. 2). The same magnon feature is observed in the bilayer (Fig. 3a), but with a discontinuous red-shift occurring at the metamagnetic transition (Fig. 3b), confirming the dominance of the magnetic anisotropy over the interlayer exchange in CrI$_3$. Furthermore, we also observe a high-frequency magnon feature at ~19 meV (~4.6 THz) in samples with 2 or more layers (Fig. 4). As this is at a far higher

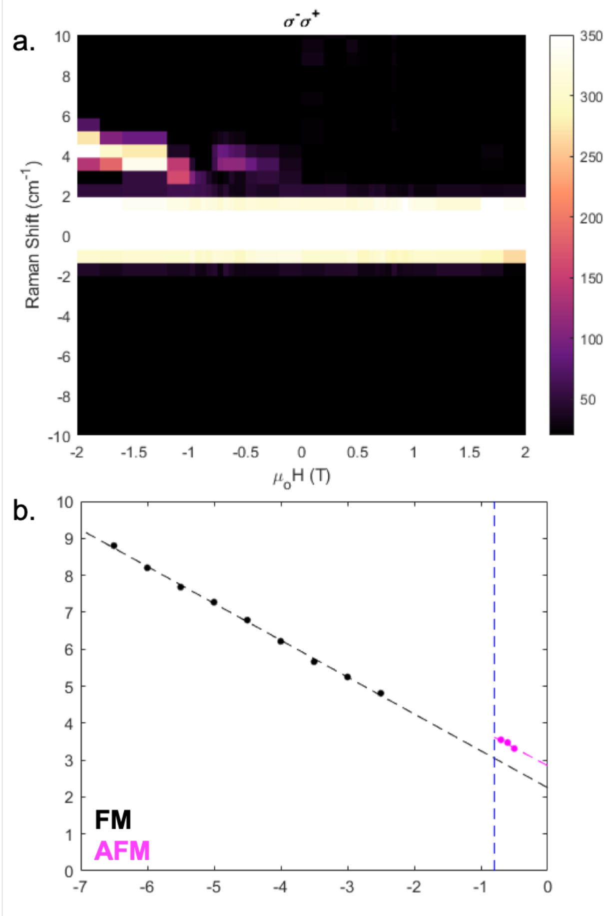

Fig. 3. Bilayer CrI$_3$. **a.** Low-field color plot of bilayer magnon (slanted lines) from -2T to 2T. The discontinuity occurs at the metamagnetic transition. **b.** Fitted values for FM and AFM states, also showing the discontinuous red-shift at transition.

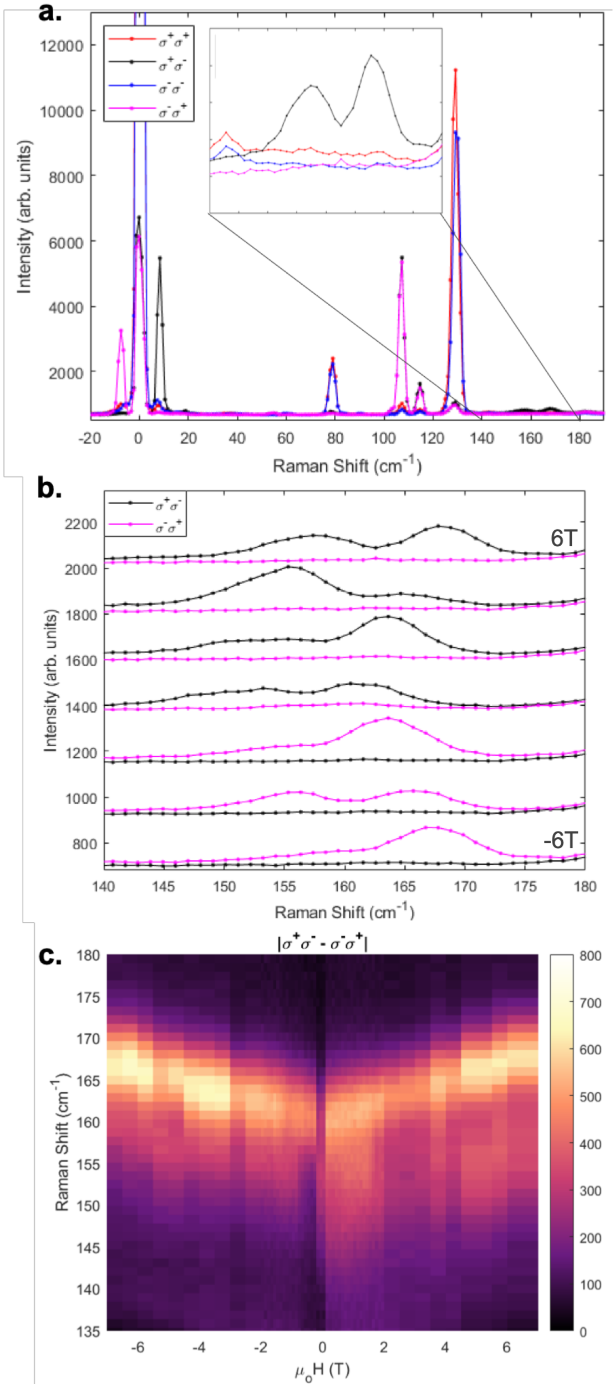

Fig. 4. High-frequency magnon (thin bulk). **a.** Thin bulk spectra for CrI$_3$. Inset magnifies high-freq. magnon region. **b.** Stack plot for high-frequency magnon, -6T to 6T (2T increments). Magnon shifts with field, as expected. **c.** Color plot, -7T to 7T.

frequency than Γ point magnons in most ferromagnets, our results indicate that CrI$_3$ has high potential for application in THz magnonic devices.

CONCLUSION:

This work marks the first direct observation of magnons in a truly 2D system using Raman spectroscopy. The monolayer data I obtained agrees with the theoretically predicted Zeeman energy shift quite well, and can help inform future theoretical studies by providing a direct measurement of the monolayer spin wave gap at the Γ point. Additionally, the high frequency magnon I observed is several orders of magnitude higher in frequency than typical FM magnons, which points to CrI$_3$ for use in THz magnonic devices.

Future investigations could study the effects of electric fields on magnons using a gated device. Application of pressure or tension to the crystal lattice could reveal strain effects—another interesting direction of future inquiry. Alternatively, since CrI$_3$ is well suited to for use in heterostructure devices, further studies could explore possible proximity effects and other unique phenomena. The research I conducted on magnons in CrI$_3$ helps to elucidate magnetism in this material, revealing the abundance of interesting and potentially useful physics that can be uncovered by studying magnons.

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REFERENCES:

[1] V. V. Kruglyak, S. O. Demokritov, and D. Grundler, Journal of Physics D: Applied Physics **43**, (2010).

[2] M. B. Jungfleisch, W. Zhang, and A. Hoffmann, Physics Letters A **382**, 865 (2018).

[3] L. Chen, J.-H. Chung, B. Gao, T. Chen, M. B. Stone, A. I. Kolesnikov, Q. Huang, and P. Dai, Physical Review X **8**, (2018).