Characterization of room-temperature in-plane magnetization in thin flakes of CrTe$_2$ with a single spin magnetometer

F. Fabre,$^1$ A. Finco,$^1$ A. Purбавати,$^2$ A. Hadj-Azzem,$^2$ N. Rougemaille,$^2$ J. Coraux,$^2$ I. Philip,$^1$ and V. Jacques$^1$

$^1$Laboratoire Charles Coulomb, Université de Montpellier and CNRS, 34095 Montpellier, France
$^2$Univ. Grenoble Alpes, CNRS, Grenoble INP, Institut NEEL, 38000 Grenoble, France

(Dated: November 12, 2020)

We demonstrate room-temperature ferromagnetism with in-plane magnetic anisotropy in thin flakes of the CrTe$_2$ van der Waals ferromagnet. Using quantitative magnetic imaging with a single spin magnetometer based on a nitrogen-vacancy defect in diamond, we infer a room-temperature in-plane magnetization in the range of $M \sim 25$ kA/m for flakes with thicknesses down to 20 nm. These results make CrTe$_2$ a unique system in the growing family of van der Waals ferromagnets, because it is the only material platform known to date which offers an intrinsic in-plane magnetization and a Curie temperature above 300 K in thin flakes.

Ferromagnetic van der Waals (vdW) crystals offer numerous opportunities both for the study of exotic magnetic phase transitions in low-dimensional systems [1] and for the design of innovative, atomically-thin spintronic devices [2, 3]. Since the discovery of a two-dimensional (2D) magnetic order in monolayers of CrI$_3$ [4] and Cr$_3$Ge$_2$Te$_6$ [5] crystals, the family of vdW ferromagnets has expanded very rapidly [6–8]. However, most of these compounds have a Curie temperature ($T_c$) well below 300 K, which appears as an important drawback for future technological applications. An intense research effort is therefore currently devoted to the identification of high-$T_c$ 2D magnets [9].

In this context, the vdW crystal Fe$_3$GeTe$_2$ appears as a serious candidate because it can be grown in wafer-scale through molecular beam epitaxy and it exhibits a strong perpendicular magnetic anisotropy [9, 10]. Although its intrinsic $T_c$ drops to 130 K in the monolayer limit [11], it can be raised above room-temperature either by ionic gating [12], micro-patterning [13], interfacial engineering [14] or by varying the composition of the iron-based Fe$_3$GeTe$_2$ alloy [15, 16]. Another promising strategy consists in incorporating magnetic dopants into 2D materials to form diluted magnetic semiconductors [17]. This approach was recently employed to induce room-temperature ferromagnetism in WSe$_2$ monolayers doped with vanadium [18, 19]. Finally, an intrinsic ferromagnetic order was reported in epitaxial layers of VSe$_2$ [20], MnSe$_2$ [21] and VTe$_2$ [22] under ambient conditions, although the interpretation of these experiments still remains debated [23, 24].

In this work, we follow an alternative research direction by studying the room-temperature magnetic properties of micron-sized flakes exfoliated from a CrTe$_2$ crystal with $IT$ structure. In its bulk form, this layered transition metal dichalcogenide is a ferromagnet with in-plane magnetization, i.e. pointing perpendicular to the $c$ axis, and a $T_c$ around 320 K [25]. This combination of properties is unique in the growing family of vdW ferromagnets. Recent studies have reported that the magnetic order is preserved at room temperature in exfoliated CrTe$_2$ flakes with thicknesses in the range of a few tens of nanometers [26, 27]. However, obtaining quantitative estimates of the magnetization in such micron-sized flakes remains a difficult task, which requires the use of non-invasive magnetic microscopy techniques combining high sensitivity with high spatial resolution. These performances are offered by magnetometers employing a single nitrogen-vacancy (NV) defect in diamond as an atomic-size quantum sensor [28–30]. In recent years, this microscopy technique has found many applications in condensed matter physics [31], including the study of chiral spin textures in ultrathin magnetic materials [32–34], current flow imaging in graphene [35] and the analysis of the magnetic order in vdW magnets down to the monolayer limit [36–38].

Here we use scanning-NV magnetometry to infer quantitatively the in-plane magnetization in exfoliated CrTe$_2$ flakes under ambient conditions. Our measurements confirm that the ferromagnetic order is preserved in few tens of nanometers thick flakes, although with a low room-temperature magnetization $M \sim 25$ kA/m. This value

![FIG. 1. Temperature dependence of the magnetization $M$ in a bulk crystal of CrTe$_2$ with $IT$ polytype. The measurement is performed by vibrating sample magnetometry under a magnetic field of 500 mT. At room temperature, the magnetization is around 120 kA/m (black dashed lines). The inset shows the layered crystal structure of $1T$-CrTe$_2$.](image-url)
is five times smaller that the one measured in a bulk CrTe$_2$ crystal. Such a reduction of the magnetization is attributed to a decreased Curie temperature in exfoliated flakes.

A bulk 1T-CrTe$_2$ crystal was synthesized following the procedure described in Ref. [25]. The in-plane magnetization of this layered ferromagnet was first characterized as a function of temperature through vibrating sample magnetometry under a magnetic field of 500 mT. The results shown in Fig. 1 indicate a Curie temperature around 320 K and a magnetization reaching $M \sim 120$ kA/m under ambient conditions. CrTe$_2$ flakes with thicknesses ranging from a few tens to a hundred of nanometers were then obtained by mechanical exfoliation and transferred on a SiO$_2$/Si substrate. We note that the probability to obtain thin CrTe$_2$ flakes through mechanical exfoliation is still very low compared to other layered transition metal dichalcogenides, such as MoS$_2$ or WSe$_2$ [27]. The thinnest flake studied in this work has a thickness of $\sim 20$ nm. Like all van der Waals ferromagnets known to date, CrTe$_2$ flakes are unstable under oxygen atmosphere. However, a recent study combining X-ray and Raman spectroscopy has shown that oxidation of CrTe$_2$ flakes occurs typically within a day scale under ambient conditions and is limited to the very first outer layers [27]. In this work, CrTe$_2$ flakes were not encapsulated and all the measurements were done within a day after exfoliation to mitigate oxidation.

Magnetic imaging was performed with a scanning-NV magnetometer operating under ambient conditions [30]. As sketched in Fig. 2(a), a single NV defect integrated into the tip of an atomic force microscope (AFM) was scanned above CrTe$_2$ flakes to probe their stray magnetic fields. At each point of the scan, a confocal optical microscope placed above the tip was used to monitor the magnetic-field-dependent photoluminescence (PL) properties of the NV defect under green laser illumination. In this work, we employed a commercial diamond tip (Qnami, Quantilever MX) with a characteristic NV-to-sample distance $d_{NV} = 80 \pm 10$ nm, as measured through an independent calibration procedure [39]. Two different magnetic imaging modes were used. In the limit of weak stray fields ($< 5$ mT), quantitative magnetic field mapping was obtained by recording the Zeeman shift of the NV defect electron spin sublevels through optical detection of the electron spin resonance (ESR). This method relies on microwave driving of the NV spin transition combined with the detection of the spin-dependent PL intensity of the NV defect [40]. For stronger magnetic fields ($> 5$ mT), the scanning-NV magnetometer was rather used in all-optical, PL quenching mode [41, 42]. In this case, localized regions of the sample producing large stray fields are simply revealed by an overall decrease of the PL signal induced by a mixing of the NV defect spin sublevels [43]. We note that the diameter of the scanning diamond tip is around 200 nm in order to act as an efficient waveguide for the PL emission of the NV defect [44, 45]. Consequently, such tips cannot provide precise topographic information of the sample. For the thinnest flakes, the topography was thus imaged by using conventional, sharp AFM tips.

In a first experiment, we imaged a CrTe$_2$ flake with a large thickness $t \sim 150$ nm [Fig. 2(b)]. For a uniformly magnetized flake, stray magnetic fields are produced at the edges of the flake, as sketched in Fig. 2(a). Considering the saturation magnetization of the bulk CrTe$_2$ crystal, magnetic simulations predict a stray field amplitude larger than 10 mT at a distance $d_{NV} \sim 80$ nm above the edges of the 150-nm-thick flake. The scanning-NV magnetometer was thus operated in the PL quenching mode for such a thick flake. Fig. 2(c) displays the resulting PL
map, which is characterized by a strong quenching at the edges of the flake, as expected for a single ferromagnetic domain. Although qualitative, this preliminary experiment confirms that mechanical exfoliation preserves the ferromagnetic order in CrTe₂ under ambient conditions.

Thinner CrTe₂ flakes, i.e., producing less stray field, were then studied through quantitative magnetic field imaging. To this end, a microwave excitation was applied through a copper microwire directly spanned on the sample surface and the Zeeman shift of the NV defect’s ESR frequency was recorded at each point of the scan. In the weak field regime, the ESR frequency is shifted linearly with the projection \( B_{NV} \) of the stray magnetic field along the NV defect quantization axis [30]. This axis was precisely measured by applying a calibrated magnetic field [46], leading to the spherical angles \( \theta_{NV} = 58° \) and \( \phi_{NV} = 103° \) in the laboratory frame \((x, y, z)\), as illustrated in Fig. 2(a).

A map of \( B_{NV} \) recorded above a 40-nm thick CrTe₂ flake is shown in Fig. 3(b). A stray magnetic field around \( \pm 1.5 \) mT was detected at two opposite edges of the flake. In principle, the underlying sample magnetization can be retrieved from such a quantitative magnetic field map by using reverse propagation methods with well-adjusted filters in Fourier-space [31]. Under several assumptions, this method can be quite robust for magnetic materials with out-of-plane magnetization [36–38]. For in plane magnets, however, the reconstruction procedure amplifies noise and is thus much less efficient [47]. As a result, the recorded magnetic field distribution was rather compared to magnetic calculations in order to extract quantitative information on the sample magnetization.

The geometry of the flake used for the calculation was inferred from the AFM image simultaneously recorded with the NV microscope [Fig. 3(a)]. Assuming a uniform in-plane magnetization \( \mathbf{M} \) with an azimuthal angle \( \phi_M = -135° \) in the \((x, y)\) plane and a norm \( M = 23.7 \) kA/m. The calculation is done at a distance \( d_{NV} = 80 \) nm above the flake, whose shape is extracted from the topography image [black dashed line in (a)]. (d) Fitting of an experimental line profile (black markers) with the magnetic calculation (blue solid line). The position of the linecuts are indicated by the white dashed lines in (b) and (c). A magnetization \( M = 23.7 \pm 2.8 \) kA/m is obtained, the uncertainty being illustrated by the blue shaded area. The red and green solid lines indicate the result of the calculation for \( M = 35 \) kA/m and \( M = 15 \) kA/m, respectively.
could be explained by a degradation of the sample surface through oxidation processes, leading to a thinner effective magnetic thickness. However, recent experiments relying on X-ray magnetic circular dichroism coupled to photoemission electron microscopy (XMCD-PEEM) have shown that oxidation is limited to the first outer layers of the CrTe₂ flake [27]. Considering a 40-nm thick CrTe₂ flake, surface oxidation can thus be safely neglected, and cannot explain the observed reduction of the magnetization. This effect rather results from a decrease of the Curie temperature in exfoliated flakes, a phenomenon commonly observed in thin vdW magnets [26]. The decreased magnetization observed in CrTe₂ flakes would correspond to a reduction of $T_c$ by about 20 K.

To support this finding, a similar analysis was performed for a 20-nm thick flake [Fig. 4(a)]. Here, the topography of the flake was recorded by using a conventional AFM tip in order to obtain more precise measurements. The corresponding magnetic field distribution is shown in Fig. 4(b). A stray magnetic field is mainly detected along the bottom edge of the flake. On the opposite edge, the stray field distribution is quite inhomogeneous. This observation is attributed to damages of the flake that can be observed in the AFM image. A simulation of the stray field distribution for a magnetization with an azimuthal angle $\phi_M = -100 \pm 10^\circ$ reproduces fairly well the experimental results [Fig. 4(c)] and a quantitative analysis of line profiles across the bottom edge of the flake leads to $M = 25.8 \pm 4.0$ kA/m [Fig. 4(d)], a similar value to that obtained in the 40-nm thick flake. This observation indicates that the magnetization is not significantly modified for thicknesses lying in the few tens of nanometers range.

To conclude, we have used quantitative magnetic imaging with a scanning-NV magnetometer to demonstrate that exfoliated CrTe₂ flakes with thicknesses down to 20 nm exhibit an in-plane ferromagnetic order at room temperature with a typical magnetization in the range of $M \sim 25$ kA/m. These results make CrTe₂ a unique system in the growing family of vdW ferromagnets, because it is the only material platform known to date which offers an intrinsic in-plane magnetization and a $T_c$ above room temperature in thin flakes. These properties might offer several opportunities for studying magnetic phase transition in 2D-XY systems [49] and to design spintronic devices based on vdW magnets. The next challenge will be to assess if the ferromagnetic order is preserved at room temperature in the few layers limit.

**Acknowledgements:** The authors warmly thank J. Vogel and M. Núñez-Regueiro for fruitful discussions. This research has received funding from the European Union H2020 Program under Grant Agreement No. 820394 (ASTERIQs), the DARPA TEE program, and the Flag-ERA JTC 2017 project MORE-MXenes. A.F. acknowledges financial support from the EU Horizon 2020 Research and Innovation program under the Marie Skłodowska-Curie Grant Agreement No. 846597 (DIMAFT).

**References**

[1] J. M. Kosterlitz and D. J. Thouless, J. Phys. C: Solid State Phys. 6, 1181 (1973).
[2] H. Li, S. Ruan, and Y.-J. Zeng, Adv. Mater. 31, 1900065 (2019).
[3] M.-C. Wang, C.-C. Huang, C.-H. Cheung, C.-Y. Chen, S. G. Tan, T.-W. Huang, Y. Zhao, Y. Zhao, G. Wu, Y.-P. Feng, H.-C. Wu, and C.-R. Chang, Annalen der Physik 532, 1900452 (2020).
[4] B. Huang, G. Clark, E. Navarro-Moratealla, D. R. Klein, R. Cheng, K. L. Seyler, D. Zhong, E. Schmidgall, M. A. McGuire, D. H. Cobden, W. Yao, D. Xiao, P. Jarillo-Herrero, and X. Xu, Nature 546, 270 (2017).
[5] C. Gong, L. Li, Z. Li, H. Ji, A. Stern, Y. Xia, T. Cao, W. Bao, C. Wang, Y. Wang, Z. Q. Qiu, R. J. Cava, S. G.
J. Förster, A. Brunner, T. Taniguchi, K. Watanabe, J. Gräfe, R. Stöhr, X. Xu, and J. Wrachtrup, arXiv:2009.13440 (2020).

[39] T. Hingant, J.-P. Tetienne, L. J. Martínez, K. Garcia, D. Ravelosona, J.-F. Roch, and V. Jacques, Physical Review Applied 4, 014003 (2015).

[40] A. Gruber, A. Dräbenstedt, C. Tietz, L. Fleury, J. Wrachtrup, and C. V. Borczyskowski, Science 276, 2012 (1997).

[41] I. Gross, W. Akhtar, A. Hrabec, J. Sampaio, L. J. Martínez, S. Chouaieb, B. J. Shields, P. Maletinsky, A. Thiaville, S. Rohart, and V. Jacques, Phys. Rev. Materials 2, 024406 (2018).

[42] W. Akhtar, A. Hrabec, S. Chouaieb, A. Haykal, I. Gross, M. Belmeguenai, M. Gabor, B. Shields, P. Maletinsky, A. Thiaville, S. Rohart, and V. Jacques, Phys. Rev. Applied 11, 034066 (2019).

[43] J.-P. Tetienne, L. Rondin, P. Spinicelli, M. Chipaux, T. Debuisschert, J.-F. Roch, and V. Jacques, New Journal of Physics 14, 103033 (2012).

[44] P. Maletinsky, S. Hong, M. S. Grinolds, B. Hausmann, M. D. Lukin, R. L. Walsworth, M. Loncar, and A. Yacoby, Nature Nanotechnology 7, 320 (2012).

[45] P. Appel, E. Neu, M. Ganzhorn, A. Barfuss, M. Batzer, M. Gratz, A. Tschöpe, and P. Maletinsky, Review of Scientific Instruments 87, 063703 (2016).

[46] L. Rondin, J.-P. Tetienne, S. Rohart, A. Thiaville, T. Hingant, P. Spinicelli, J.-F. Roch, and V. Jacques, Nat. Commun. 4, 2279 (2013).

[47] D. A. Broadway, S. E. Lillie, S. C. Scholten, D. Rohner, N. Dentschuk, P. Maletinsky, J.-P. Tetienne, and L. C. L. Hollenberg, Phys. Rev. Applied 14, 024076 (2020).

[48] I. Gross, W. Akhtar, V. Garcia, L. J. Martínez, S. Chouaieb, K. Garcia, C. Carrétéro, A. Barthélémy, P. Appel, P. Maletinsky, J.-V. Kim, J. Y. Chauleau, N. Jaouen, M. Viret, M. Bibes, S. Fusil, and V. Jacques, Nature 549, 252 (2017).

[49] A. Bedoya-Pinto, J.-R. Ji, A. Pandeya, P. Gargiani, M. Valvidares, P. Sessi, F. Radu, K. Chang, and S. Parkin, arXiv:2006.07605 (2020).