

Opportunities for nitrogen-vacancy-assisted magnetometry to study magnetism in 2D van der Waals magnets

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ABSTRACT

Exploring and understanding magnetism in two-dimensional (2D) van der Waals (vdW) magnetic materials present a promising route for developing high-speed and low-power spintronics devices. Studying their magnetic properties at the nanoscale is challenging due to their low magnetic moment compared to bulk materials and the requirements of highly sensitive magnetic microscopy tools that work over a wide range of experimental conditions (e.g., temperature, magnetic field, and sample geometry). This Perspective reviews the applications of nitrogen-vacancy center (NV) based magnetometry to study magnetism in 2D vdW magnets. The topics discussed include the basics, advantages, challenges, and the usage of NV magnetometry.

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I. OVERVIEW

After the demonstration of the intrinsic magnetic behavior in 2D $\text{Cr}_2\text{Ge}_2\text{Te}_6$ and CrI_3 materials in 2017,^{1,2} a wide range of other vdW materials have been investigated.³ The advantages of 2D magnets in comparison with bulk crystals, such as easy and low-cost fabrication and a wide variety of control mechanisms, make them and their heterostructures promising candidates for the next generation of spintronic devices.^{4–7} For example, the extensively studied materials CrI_3 (a semiconductor) and Fe_2GeTe_3 (a metal) are soft ferromagnets in the bulk crystal^{8,9} and become hard ferromagnets when exfoliated to a few atomic layers.^{10–16} Another advantage of 2D materials is that the atomically thin nature of the material makes it susceptible to atom engineering and proximity effects.^{17,18} By placing graphene on top of a ferromagnetic insulator, anomalous Hall measurements showed that the graphene becomes ferromagnetic.^{19,20} Additionally, layer-by-layer engineering and twisting and van der Waals bonding in these materials provide a myriad of magnetic phenomena at the nanoscale, including edge-topological magnetic states,²⁰ unconventional superconductivity,²¹ and topological spin textures such as skyrmions³² and moiré magnetism.²³ Dopant and defect-induced ferromagnetism has been predicted theoretically^{24,25} and demonstrated experimentally in 2D transition metal dichalcogenides (TMD) such as MoSe_2 , WS_2 , MoS_2 , VSe_2 , and

MnSe_2 .^{26–30} This progress has led to the proposal of the usage of 2D magnets in spintronics.^{6,7} Examples include the tunneling magnetoresistance devices,^{31,32} anomalous Hall effect,¹⁴ spin transistors,³³ spin valves,³⁴ spin filters and magnetoelectric switches,¹⁶ and magnetic memories based on skyrmions.²² However, despite this progress, little is known about the mechanisms governing the nanoscale fundamental magnetic processes in 2D vdW magnets.

A wide range of magnetic probing techniques have been used to study their magnetic properties. Standard techniques include magneto-transport measurements,^{31,35–39} magneto-optical Kerr effect microscopy (MOKE),^{1,2} and magnetic force microscopy (MFM).⁴⁰ MOKE is sensitive enough to detect ferromagnetism in few-layer magnets, such as $\text{Cr}_2\text{Ge}_2\text{Te}_6$,^{1,2} but it lacks spatial resolution (≥ 300 nm).^{41,42} MFM uses magnetic tips to image the stray magnetic fields generated at surfaces of magnetic materials and has a good spatial resolution ≥ 10 nm, limited by the magnetic tip size and the local magnetic gradient.⁴³ Apparent contrasts in MFM due to magnetic forces are often contaminated by other long-range forces associated, for instance, with surface charges, that make it hard to interpret the measured magnetic signals quantitatively.⁴⁴ MFM also lacks the magnetic sensitivity to image weakly magnetized materials.⁴⁴ Advanced microscopy techniques, such as ferromagnetic resonance force microscopy (MRFM),⁴⁵ suffer from limited knowledge

about the magnetic tip, and magnetic tip influences the local properties. Spin-polarized current-based tunneling microscopy (SP-STM) provides a very good magnetic sensitivity with a spatial resolution of few nanometers,^{46,47} but it requires electrically conductive samples. Scanning superconducting quantum interference devices (SQUIDS) integrated to SPM have been used to probe weakly stray-field produced materials, i.e., to measure quantum Hall edge currents in graphene⁴⁸ and image orbital ferromagnetism in twisted bilayer graphene.⁴⁹ It offers a magnetic sensitivity down to 5 nT/Hz^{1/2} with a spatial resolution below 100 nm, but it needs lower temperatures and complicated SQUID probe designs.^{50–52}

Recently, a technique has emerged for measuring magnetic fields at the nanoscale based on optical detection of the electron spin resonances of nitrogen-vacancy (NV) centers in diamond,⁵³ opening up frontiers to study condensed matter phenomena.⁵⁴ NV magnetometry can detect weak static and dynamic magnetic stray fields with frequencies from DC to > few GHz and works at a wide range of experimental conditions, i.e., temperatures from 0.35 to 600 K,^{55–58} and applied magnetic fields up to 4 T.⁵⁹ It is until very recently where NV magnetometry has been used to study vdW 2D magnets, including CrI₃,^{13,23} VI₃,⁶⁰ CrTe₂,⁶¹ and CrBr₃.⁶² There are very excellent Review/Perspective papers on studying the magnetic properties of 2D magnets by using various techniques,^{4,5,40,63,64} and only a few of them include short discussions of using NV magnetometry.⁴⁰ In this Perspective paper, we mainly discuss the advantages and challenges of using NV microscopy to study 2D vdW magnets by including the basics of NV magnetometry sensing/imaging schemes and recent NV-magnetic measurements on 2D magnets, as well as the outlooks of the field.

II. INTRODUCTION TO NV-BASED MAGNETOMETRY SCHEMES

The NV center is a lattice defect in diamond^{54,65–67} with remarkable properties, including sensing magnetic fields,^{68–75} electrical

fields,^{76–79} and temperature,^{80–82} even at extremes pressure conditions.^{83,84} The negatively charged NV center, a substitutional nitrogen adjacent to a vacancy site [Fig. 1(a)], is an $S = 1$ electron spin state that can be initialized by optical excitation (500–550 nm) and detected through spin-dependent photoluminescence (650–800 nm) known as optically detected magnetic resonance (ODMR)⁵³ [Figs. 1(b) and 1(c)]. Intersystem crossing to metastable singlet states takes place preferentially for NV centers in the $m_s = \pm 1$ states, allowing optical readout of the spin state via spin-dependent fluorescence.⁵³ The application of a magnetic field breaks the degeneracy of the $m_s = \pm 1$ state via the Zeeman effect and leads to a pair of transitions whose frequencies depend on the magnetic field component along the NV symmetry axis.⁵³ The NV electron spin has millisecond spin coherence time at room temperature^{85–87} and is among the best quantum sensors found in nature.⁸⁸ Magnetic field sensing via NV center can be divided into two broad categories based on the spectral characteristics of the magnetic fields to be detected. For example, DC sensing schemes are sensitive to static, slowly varying, and broadband near static magnetic fields, whereas AC sensing schemes typically detect narrowband time-varying magnetic fields of frequency up to few GHz.^{54,89,90} NV magnetometry is now widely used to detect static and dynamic magnetic stray fields, temperature, electric field, and strain in solid-state materials, opening up new frontiers in condensed matter research.⁵⁴

The Zeeman shifts of NV's electron spin states are used for DC sensing by measuring the peak positions of NV's spin resonance frequencies.⁵³ The optimized DC magnetic sensitivity is limited by photon-shot-noise given by Refs. 91–93 as $\eta_{DC} \cong (\gamma_{NV} R)^{-1} 1/(I_0 t)^{1/2} [1/T_2^*]^{1/2}$, where $\gamma_{NV} = 28$ GHz/T is the NV gyromagnetic ratio, R is the ODMR contrast, I_0 is the NV fluorescence rate, T_2^* is the decoherence time of NV, and t is the measurement time. For standard surface NV measurements $T_2^* =$ hundreds of ns to 1.5 μ s, giving a DC sensitivity of 4–10 μ T/Hz^{1/2} for single NVs and < 0.1–1 μ T/Hz^{1/2} for NV

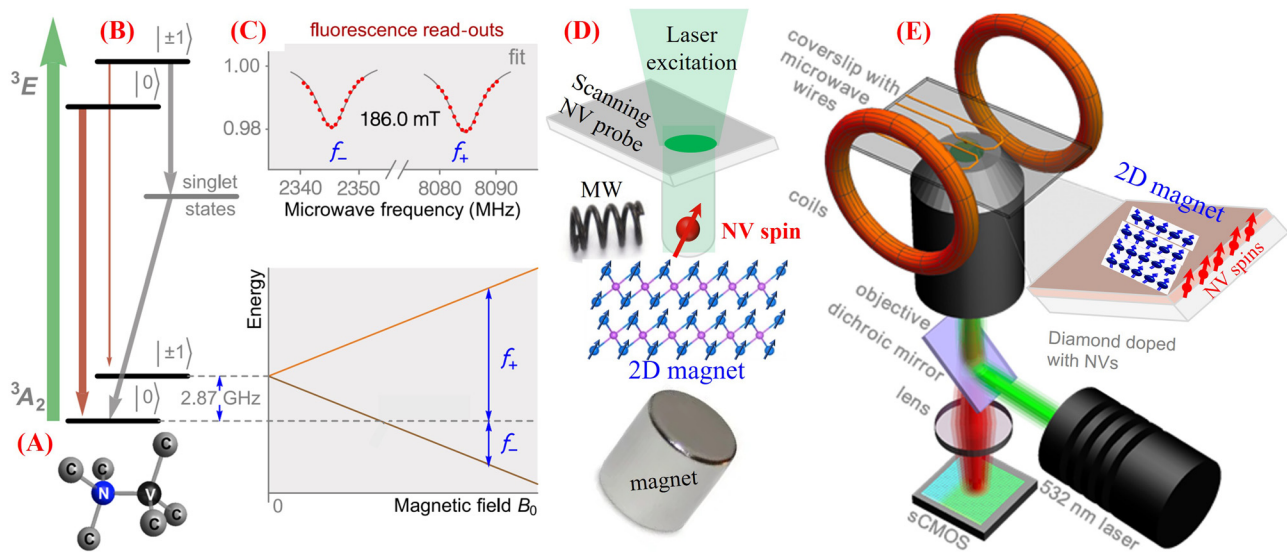


FIG. 1. NV-magnetometry. (a) NV center in the diamond lattice. (b) Energy levels of the NV center. The NV spin is pumped into the $|0\rangle$ state by off resonance optical excitation, and the ground-state spin can be manipulated by microwave excitation. NV ODMR spectra [above (c)] and magnetic field dependence of NV resonances [below (c)]. (d) Schematic of NV-SPM for studying magnetic properties of 2D magnets. (e) Schematic of NV-WFM for studying magnetic properties of 2D magnets. (b) and (c) are reproduced with permission from Fescenko et al., "Diamond magnetic microscopy of malarial hemozoin nanocrystals," *Phys. Rev. Appl.* **11**, 034029 (2019). Copyright 2021 American Physical Society.⁹³

ensembles (densities \sim tens of ppb to few ppm).⁹¹ The variation of the magnetic sensitivities is related to the number of NVs and fluorescence collection efficiency⁹¹ (discussed below). The DC magnetometry scheme is a quick and simple approach to extracting static unknown stray magnetic field B_{NV} (variable magnitude and direction) from magnetic surfaces. Measuring along one of the diamond orientations allows retrieving vector stray field components (B_x , B_y , and B_z) and indirectly retrieving the vector magnetization.^{13,54,94}

An AC sensing scheme is preferred to study dynamic magnetic phenomena in 2D magnets,⁹⁵ and it offers better magnetic sensitivity and frequency selection as compared to DC sensing protocols.^{54,69,96} The NV-magnetometry AC sensing scheme is based on using well-established electron spin resonance pulse sequences such as Hahn echo to extend T_2^* to T_2 (usually one order of magnitude longer).⁹¹ The typical AC magnetic sensitivity for such protocol is about 50–100 nT/Hz^{1/2} for single NVs and can be boosted to few nT/Hz^{1/2} by using pulse dynamical decoupling (DD) protocols and photonics diamond engineering.⁹¹ For example, single NV centers can detect random AC (up to 20 kHz) magnetic fields with a sensitivity of \sim 50–100 nT/Hz^{1/2} [100]. This AC sensing pulse protocol can be modified to perform double spin resonance experiments and detect high frequency (few GHz) spins.^{54,69,70} By using a spin-correlation pulse protocol,⁹⁸ the NV AC sensitivity can be pushed down to few nT/Hz^{1/2}, limited by the NV spin-lattice relaxation T_1 (few milliseconds at ambient conditions).^{91,92,98,99} The spin-correlation pulse protocol is later integrated with DD pulse protocols to probe 30 nuclear ¹¹B spins in atomically thin hBN¹⁰⁰ and map the distribution and direction of the photocurrent flow in the 2D MoS₂ material⁹⁰ with sensitivities to current densities as low as 20 nA/ μ m, comparable to state-of-the-art SQUID.¹⁰¹ By using NV ensembles in combination with a narrowband synchronized readout protocol,^{102,103} AC magnetic sensitivities down to tens of pT/Hz^{1/2} are obtained.^{104,105}

III. SCOPE TO DEVELOP FAST, HIGH SPATIAL RESOLUTION, AND HIGHLY SENSITIVE NV MAGNETOMETRY-BASED IMAGING METHODS TO STUDY 2D VDW MAGNETS

There are two approaches for using NV magnetometry for magnetic imaging: single NV scanning probe microscopy [NV-SPM, Fig. 1(d)] and NV ensemble wide-field microscopy [NV-WFM, Fig. 1(e)]. We discuss below in detail the basics of both approaches, advantages, and challenges in comparison with existing magnetic imaging methods.

NV-SPM is proposed first by Degen;¹⁰⁶ NV-SPM is based on scanning a diamond probe with a single NV center across a magnetic sample and measures the stray fields generated from its surface [Fig. 1(d)]. Magnetic imaging with NV-SPM was first realized from DC magnetic stray fields generated by Ni nanoelements⁶⁸ but has been extended to imaging skyrmions in ferromagnetic multilayers/films,^{94,107,108} spin textures in antiferromagnets,^{107,109–112} magnetic vortices in high T_c superconductors,^{54,113} and moiré superlattices of twisted 2D magnet CrI₃²³ and to probe magnetic dynamic excitations in ferromagnetic materials.^{114,115}

NV-SPM has some advantages to probe magnetism at the nano-scale in comparison with existing techniques because (1) it provides high spatial resolution (\sim 50 nm), limited by the distance of NV center in the diamond probe from the target surface and by the ambient large tip oscillation amplitude and low Q-factor.^{13,58,109,116–118} By using ion beam

implantation and slow oxidative etching of implanted diamond, NVs can be created as shallow as 2 nm from the diamond surface.¹¹⁸ Integrating NV-SPM with an ultra-high vacuum system enhances the diamond tip Q-factor by orders of magnitude, which allow for a lower tip oscillation amplitude.¹¹⁹ By using these two approaches, the NV-SPM spatial resolution can be pushed down to below 20 nm; (2) commercial diamond tips are available with a single NV center at all diamond orientations that allows measuring samples with different magnetization configurations (e.g., out-of-plane and in-plane); (3) measurements work for both conductive and non-conductive samples, i.e., no surface treatment is required; (4) NV center weakly interacts with the sample, thus, less perturbation, in contrast to MFM; and (5) high sensitivity to static and dynamic magnetic properties⁹¹ (discussed above).

NV-SPM is a still evolving magnetic imaging technique that can be generalized as a standard tool if the challenges discussed below are taken care of. First, it is a slow imaging technique. A typical static magnetic image of 100×100 pixels² (a dwell time of \sim few seconds per pixel) takes several hours to complete. This is mainly limited by three factors: A long averaging time (tens to hundreds of ms) is required to get a good signal-to-noise ratio, a high number (>20) of MW frequency sweeping points across the ODMR spectrum (linewidth \sim 5–10 MHz) is needed to resolve weak (<10 μ T) stray magnetic fields, and data fitting is used to determine the local magnetic field information per pixel.^{13,40,54} There are recent efforts to speed up NV-SPM. Recently, Ambal and McMichael demonstrated a lock-in amplifier detection scheme by using a proportional–integral–derivative (PID) feedback controller to track the NV's ODMR peak with a dwell time of 0.1–1 s.^{120,121} However, this method fails to track sudden changes in the local magnetic field.¹²⁰ To circumvent this issue, Dushenko *et al.*¹²² used an optimized Bayesian algorithm and demonstrated 45 faster acquisition time in comparison with the traditional frequency sweep technique. In this method, the Bayesian algorithm predicts the next best frequency to measure based on previous measurements by learning from each measurement¹²³ as opposed to the sequentially (step-by-step) frequency sweep. Very recently, Welter *et al.* used a spectrum demodulation method and imaged stray fields above antiferromagnet α -Fe₂O₃ at pixel rates of up to 100 Hz (dwell time = 0.01 s) with an image resolution exceeding one megapixel.¹²⁴ This technique is specifically useful for magnetic samples with a large signal dynamic range (\sim few mT).¹²⁴

Another approach used to speed up NV-SPM imaging is to optimize the NV's fluorescence collection efficiency in the diamond probe. Placement of the NV center in the diamond probe based on nanopillar geometry boosts the NV fluorescence in comparison with bulk substrates.¹²⁵ Hedrich *et al.* fabricated diamond probes with a truncated parabolic profile and optimized the fluorescence signal from single and near-surface NV centers to \sim 2.1 Mc/s,¹²⁶ one order of magnitude better than conventional nanopillar tips (\sim 200 Kc/s).¹¹⁷ Therefore, combining the parabolic profile of the diamond tip with PID-based tracking or mathematical analysis (e.g., Bayesian algorithm) and spectrum demodulation methods could cut down the NV-SPM imaging time from few tens of seconds to few tens of minutes.

Other challenges of NV-SPM include (i) the requirement of optical and microwave excitation that sometimes sets up a limit on the possible materials to be studied. Laser can excite direct-bandgap 2D magnets and may induce fluorescence in the NV wavelength detection bandwidth.^{24,127} Microwave can excite thermal magnon modes in the

material and may lead to NV fluorescence quenching and/or ODMR broadening.^{128,129} It is possible to reduce these effects by using pulsed AC sensing detection schemes,^{54,91} (ii) the requirement to align the applied magnetic field along the NV axis to prevent spin level mixing,⁵³ which leads to a decrease in the NV ODMR contrast and, therefore, deteriorates the NV-SPM sensitivity; and (iii) cryogenic measurements are affected by the reduced photoluminescence contrast, explained by the strain-dependent variations of the NV's orbital g factor.⁵⁶ Very recently, NV-SPM imaging has been performed at a temperature of 350 mK,¹³⁰ providing opportunities to study high correlated magnetic phenomena in 2D vdW magnets.

NV-WFM: NV-SPM imaging is preferred to map high spatial resolution and small sample regions ($\leq 100 \mu\text{m}^2$) with magnetic structures below 100 nm. With NV-ODMR peak tracking techniques and diamond probe engineering (discussed above) could speed up NV-SPM imaging. Though it still may take extra hours to acquire megapixel images or scan bigger regions (area $> 400 \mu\text{m}^2$).¹³ NV-WFM has emerged as an alternative technique to spatially map solid-state materials and biomolecules.⁵⁷ The imaging modality is based on using a diamond chip implanted with a dense layer of NV centers (few nanometers to tens of micrometers) near its surface, interrogated in a wide-field optical microscope with an sCMOS camera, to map the magnetic stray field, lattice strain, temperature, and current density of samples or devices placed in proximity⁵⁷ [Fig. 1(e)]. The high density of NVs (a few ppm) allows a DC magnetic sensitivity $< 0.1 \mu\text{T}/\text{Hz}^{1/2}$ and AC magnetic sensitivity down to tens of $\text{pT}/\text{Hz}^{1/2}$, depending on the detection volume.^{57,91,97} It is used initially to map biomaterials, such as live cells labeled with magnetic nanoparticles¹³¹ and malarial biocrystals⁹³ and later extended to study magnetism of rocks,¹³² and to map local strain in diamonds.^{133,134} NV-WFM imaging modality is well suited for rapid analysis (few tens of minutes) of multiple micrometer-sized samples (scanning area $> 400 \mu\text{m}^2$) and allows to study of condensed matter (strain, temperature, and magnetism) phenomena in vdW magnets with sub-300 nm spatial resolution.⁵⁷ Measuring the ODMR peaks from the four NV orientations enables vector magnetometry,¹³⁵ which is useful for reconstructing the vector magnetization/magnetic moment of 2D magnets. Recently, this modality has been used to study 2D vdW materials, for example, to map current density in a graphene ribbon¹³⁶ and stray magnetic field map of VI_3 crystals⁶⁰ (further discussion below). Compared with other magneto-optical imaging techniques, such as MOKE, NV-WFM imaging features a similar spatial resolution ($\geq 300 \text{ nm}$, limited by optical diffraction) but presents the advantage of being quantitative, enabling the absolute magnetization of individual flakes to be determined.⁵⁷ We discuss below few examples of recent contributions of using NV-SPM and NV-WFM magnetometry to study magnetism in 2D vdW magnets.

IV. RECENT CONTRIBUTION OF NV MAGNETOMETRY TO STUDY 2D MAGNETS

Recently, Thiel *et al.* used NV-SPM to quantitatively study the magnetic properties of CrI_3 monolayers and directly image magnetic domains with a spatial resolution of $\sim 50\text{-nm}$.¹³ Figure 2(a) shows NV measured stray-magnetic field B_{NV} image on an area containing bilayer and trilayer CrI_3 in an applied magnetic field of 172.5 mT. The imaging is performed by measuring the NV full ODMR peaks at each pixel (dwell time of 2 s). The stray magnetic fields come mostly from the edges of the trilayer flake, as expected for a largely uniform

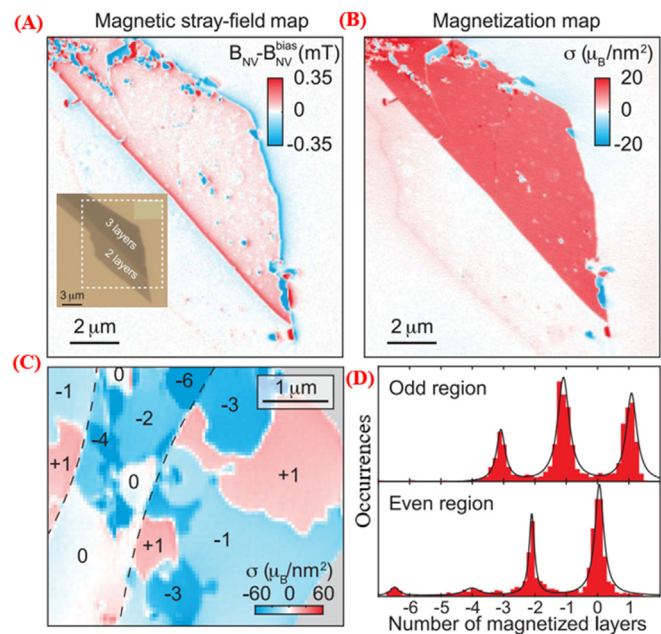


FIG. 2. NV-SPM imaging of CrI_3 layers. (a) NV magnetic stray field map measured at a magnetic field 172.5 mT, optical image of CrI_3 flake is shown in the inset. (b) Map of the magnetization distribution of the same region, determined by reverse propagation of the NV stray field map in (a). (c) Spontaneously occurring magnetic domains observed in the nine-layer flake of another CrI_3 sample. (d) Histograms of magnetization pixel values obtained in the odd- and even-numbered regions of the data in (c). The figure reproduced with permission from Thiel *et al.*, "Probing magnetism in 2D materials at the nanoscale with single-spin microscopy," *Science* **364**(6444), 973–976 (2019). Copyright 2019 American Association for the Advancement of Science.¹³

magnetization of few-layer CrI_3 .¹³⁷ The 2D out-of-plane magnetization map of CrI_3 in Fig. 2(b), retrieved by using a reverse-propagation protocol,¹³⁸ shows a uniform magnetization of the trilayer CrI_3 with an average magnetization of $\sim 13 \mu_B/\text{nm}^2$, where μ_B is the Bohr magneton. Additional measurements on different CrI_3 samples with even and odd numbers of layers confirmed the zero net and homogenous magnetization, respectively, consistent with antiferromagnetic exchange coupling.^{31,139} The change in the magnetic order from ferromagnet in bulk CrI_3 crystal¹¹ to antiferromagnetic in CrI_3 layers^{10,11,13,139} is explained by the interplay between the structure (stacking order) and exchange coupling.¹³ To further explain this effect, NV-SPM measurements were performed on nine-layer flake CrI_3 [Fig. 2(c)]. The spatial variations of the exchange coupling due to the locale change of the stacking order resulted in regions with varying numbers of anti and ferromagnetically coupled layers.¹³ The magnetization was found to be discretized in integer multiples of the monolayer magnetization. This domain formation mechanism conserves the parity of well-separated regions on the sample [histogram in Fig. 2(d)] and can be explained by adding or removing a CrI_3 monolayer during sample preparation.¹³

Sun *et al.*⁶² employed similar cryogenic NV-SPM to study the existence of magnetic domains in atomically thin CrBr_3 . CrBr_3 is a ferromagnetic insulator with a unique spin system to study ferromagnetism in the 2D limit.¹⁴⁰ The authors studied magnetic domains in few-layer samples of CrBr_3 by quantitatively mapping the stray

magnetic field.⁶² They showed that domain wall pinning is the dominant coercivity mechanism by observing the evolution of both the individual magnetic domains and the average magnetization under variable applied magnetic fields.⁶² Song *et al.* used NV-SPM to study the magnetism of twisted layers of the 2D magnet CrI₃ and imaged the magnetic domains in both twisted monolayer and twisted trilayer structures.²³ For twisted trilayers, a periodic moiré pattern of ferromagnetic and antiferromagnetic domains was shown.¹⁴¹ In a recent NV-SPM study, Fabre *et al.* observed room-temperature ferromagnetism with in-plane magnetic anisotropy in micro-sized CrTe₂ flakes (thickness ~ 20 nm).⁶¹ The analysis of NV-SPM measurements indicated that the orientation of the magnetization of these flakes is not determined only by shape anisotropy, which suggests the presence of magnetocrystalline anisotropy.⁶¹

NV-WFM was employed recently by Broadway *et al.* to directly study the magnetic processes in few-layer flakes of VI₃.⁶⁰ VI₃ is a magnetic semiconductor that has a hard ferromagnetic behavior in bulk substrates with an out-of-plane anisotropy and a high coercive field H_c of 1 T at low temperatures.^{142,143} An optical image of a VI₃ sample on a Si substrate [Fig. 3(a)], prior to transfer to diamond, shows flakes of different thicknesses from three atomic layers up to 10 nm. The NV magnetic field (B_{NV}) image of the same sample, after transfer to diamond [Fig. 3(b)], revealed magnetic signals of up to 50 μ T at an applied magnetic field of 5 mT and at a temperature of 5 K. Using a similar approach in reference,¹³ the map of the out-of-plane magnetization (M_z) is reconstructed in Fig. 3(c) and found to be in the range of ~ 50 μ_B/nm^2 . A complete and abrupt switching of most flakes is observed at coercive field $H_c \approx 0.5$ –1 T, independent of VI₃ flake thickness, as shown in Fig. 3(d). H_c decreases as the temperature approaches the Curie temperature T_c (≈ 50 K). To understand the

switching processes in the VI₃ flakes, 1 s pulses of magnetic field are applied in the $-z$ -direction of the samples (initially magnetized in the $+z$ -direction) and the NV-ODMR imaging is performed by increasing the magnetic field pulse amplitude up to 1.3 T and temperature up to 50 K. The magnetization was found to reverse abruptly, and H_c decreases with the increase in temperature [Fig. 3(e)]. This observation has a similar signature of magnetic domain nucleation-type as in ferromagnetic systems.¹⁴⁴ Further NV-WFM measurements from the zero-field cooled state to high magnetic fields up to 0.4 T and at 5 K are performed on the same flake. The domain wall depinning field in the range of 0.1 T to 0.4 T increases with the decrease in the flake thickness.⁶⁰ These values, expected from thin flakes, are way less in the case of bulk crystals (\sim few mT) measured by MOKE.¹⁴³

These demonstrations of using NV magnetometry in both NV-SPM and NV-WFM are promising, paving the way toward the widespread use of this technique as a powerful microscopy tool to study magnetism in 2D vdW magnets.

V SUMMARY AND FUTURE PERSPECTIVE

NV magnetometry is still an emerging technique with impressive progress over the last few years. We highlighted some of the progress toward a wide usage of such techniques: The commercialization of single NV diamond probes with different orientations that allow measuring 2D magnets with any magnetization orientation; the commercialization of turn-key RT and cryogenic NV-SPM microscopes may expand the usage to non-NV experts, the demonstration of 350 mK NV-SPM operation may provide opportunities to study high correlated magnetic phenomena in 2D magnets;⁵⁵ the integration of lock-in detection, Bayesian approach, or demodulation methods can speed up the acquisition time of NV-SPM.^{122–124} The usage of NV-WFM can help in rapid analysis of multiple micrometer-sized samples and allow for studying condensed matter (strain, transport, temperature, and magnetism) phenomena in vdW magnets.⁵⁷

In comparison with standard magnetic imaging techniques, NV magnetometry has several advantages summarized here: its high sensitivity to surface magnetic samples,^{13,40,58,109} detection of dynamic magnetic excitation,^{37,69,145} and sensitivity to other parameters, such as electric field, local strain, and temperature, that make it an ideal hybrid quantum sensing platform for 2D magnets. However, the requirements of magnetic field alignment, laser, and high-frequency MW excitation, and short (few nanometers) NV-to-target sample distance^{58,128} add few limitations to NV magnetometry. By complementing NV magnetometry with magneto-transport measurements and with other magnetic probing techniques, it can be an ideal tool to study spin textures, spin dynamics and relaxation, and the origin of magnetism in 2D vdW magnets. For example, there are many recent studies where NV magnetometry (both NV-SPM and NV-WFM) is integrated with magneto-transport measurements to study current-induced nanoscale fragmentation of non-uniform antiferromagnetic domain patterns in CuMnAs devices¹⁴⁶ and study spin-orbit torque induced deterministic magnetic switching and chiral spin rotation in non-collinear antiferromagnet Mn₃Sn,¹⁴⁷ opening a new door to study antiferromagnetism in 2D magnets.

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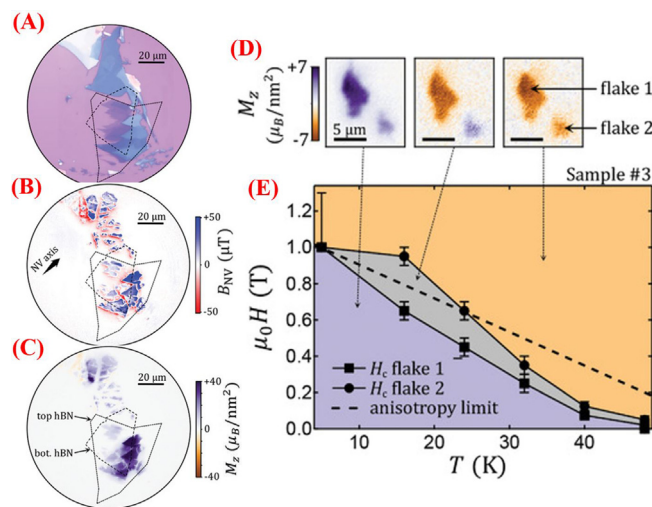


FIG. 3. NV-WFM imaging of few-layer VI₃ flakes. (a) Optical image of VI₃ flake with varying thickness prior to transfer to the diamond. (b) The corresponding NV B_{NV} map at 5 K and bias magnetic field of 5 mT. (c) Map of z magnetization M_z determined from (b). (d) M_z maps of each magnetic state in (e), recorded at 5 K. (e) H - T phase diagram of the magnetic state of two flakes. The figure is reproduced with permission from Broadway *et al.*, "Imaging domain reversal in an ultrathin van der Waals ferromagnet," *Adv. Mater.* **32**, 2003314 (2020). Copyright 1999–2021 John Wiley & Sons, Inc.⁶⁰

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Abdelghani Laraoui: Conceptualization (equal); Writing – original draft (equal); Writing – review and editing (equal). **Kapildeb Ambal:** Conceptualization (equal); Writing – original draft (equal); Writing – review and editing (equal).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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