Wide field imaging of van der Waals ferromagnet Fe$_3$GeTe$_2$ by spin defects in hexagonal boron nitride

Emergent color centers with accessible spins hosted by van der Waals materials have attracted substantial interest in recent years due to their significant potential for implementing transformative quantum sensing technologies. Hexagonal boron nitride (hBN) is naturally relevant in this context due to its remarkable ease of integration into devices consisting of low-dimensional materials. Taking advantage of boron vacancy spin defects in hBN, we report nanoscale quantum imaging of low-dimensional ferromagnetism sustained in Fe$_3$GeTe$_2$/hBN van der Waals heterostructures. Exploiting spin relaxometry methods, we have further observed spatially varying magnetic fluctuations in the exfoliated Fe$_3$GeTe$_2$ flake, whose magnitude reaches a peak value around the Curie temperature. Our results demonstrate the capability of spin defects in hBN of investigating local magnetic properties of layered materials in an accessible and precise way, which can be extended readily to a broad range of miniaturized van der Waals heterostructure systems.

Optically active spin defects in wide band-gap semiconductors promise to enable a broad range of emerging applications in quantum information sciences and technologies. To date, nitrogen-vacancy centers in diamond, as well as divacancy and silicon-vacancy centers in silicon carbide, have been among the most prominent candidates, and have been successfully applied to quantum sensing, computing, and network research, enabling unprecedented field sensitivity, spatial resolution, and state-of-the-art spin-qubit operations. Many of these advantages derive from the quantum-mechanical nature of these spin defects, which are endowed with excellent quantum coherence, single-spin addressability, and remarkable functionality over a broad temperature range. More recently, the flourishing catalog of van der Waals materials has provided a diverse new playground to enrich this field. There is ongoing and intense activity to explore emergent spin defects and color centers in atomic layers of van der Waals crystals, e.g. transition metal dichalcogenides MoS$_2$, WSe$_2$, and hexagonal boron nitride (hBN). In comparison with their conventional counterparts imbedded in three-dimensional solid-state-media, spin defects hosted by two-dimensional (2D) materials exhibit improved versatility for implementing ultrasensitive quantum sensing of proximate objects and remarkable compatibility to device integration. For instance, hexagonal boron nitride (hBN), one of the most intensively studied candidates, has been widely employed as an encapsulation layer and gate dielectric material in fabricating functional 2D devices. Thus, nanoscale proximity between spin defects in a hBN thin sheet and a layered 2D material can be readily established in van der Waals heterostructures, offering a previously unexploited quantum sensing platform to explore the local physical quantities of interest in an accessible and precise way.

Despite these potential benefits and many pioneering studies, to date, experimental demonstration of quantum microscopy using spin...
defects in a real van der Waals heterostructure remains a formidable challenge. In this work, we report nanoscale quantum sensing and imaging of exfoliated 2D ferromagnet Fe₃GeTe₂ (FGT) flakes by boron vacancy VₓBₐ spin defects in an adjacent hBN capping layer. Exploiting a wide-field magnetometry method, we directly image the local magnetic texture of the FGT flake and its characteristic temperature and field dependent magnetization evolution behavior. Taking advantage of spin relaxometry techniques, we have observed spatially varying spin fluctuations in the FGT flake, whose magnitude reaches a peak value around the Curie temperature, consistent with the expected ferromagnetic phase transition. We highlight the local magnetic texture of the FGT flake, bringing new opportunities for investigating the local spin, charge, and thermal properties of emergent quantum materials and devices.

Results

Before discussing the details of our experimental results, we first review our measurement platform and device structure, as illustrated in Fig. 1a. We exfoliated an FGT flake and mechanically transferred it onto a patterned Au microwave transmission line. Following this, we encapsulated the sample with an hBN layer. The device preparation process was performed in a glove box with argon environment to minimize environmental effects (see Methods for details). An optical microscope image shown in Fig. 1b provides an overview of a prepared device, where the thickness of the FGT and hBN flakes were characterized by atomic force microscopy (see Supplementary Information Note 1 for details). Boron vacancy VₓBₐ spin defects in the hBN flake were created by Helium ion implantation with an energy of 5 keV and a dose of 5 × 10ⁱ⁹ cm⁻². Figure 1c illustrates the structure of VₓBₐ in a hexagonal crystalline structure with alternating boron (red) and nitrogen (green) atoms, where three nitrogen atoms are adjacent to each boron atom vacancy VₓBₐ. The negatively charged VₓBₐ spin defect has an S = 1 electron spin and serves as a three-level quantum system, as shown in Fig. 1d. In the present study, we used microwave currents flowing in the Au microwave transmission line to control the quantum spin state of VₓBₐ spin defects, which can be optically accessed via spin-dependent photoluminescence (PL). The Au underlayer also enhances the PL and optical contrast of VₓBₐ spin defects, aiding the quantum microscopy measurements discussed below. The distance between the VₓBₐ defect centers and the top surface of the FGT sample is estimated to be ~50 nm based on Stopping and Range of Ions in Matter (SRIM) simulations. Exfoliated FGT flakes show spontaneous perpendicular magnetization due to reduced crystal symmetry of the layered structure. We have prepared separate devices to systematically and reproducibly characterize the magneto-transport properties of exfoliated FGT flakes, whose Curie temperature is measured to be ~200 K, in qualitative agreement with previous studies (see Supplementary Information Note 2 for details).

We now utilize wide-field microscopy to demonstrate optically detected magnetic resonance (ODMR) and spin relaxation of VₓBₐ defect centers (see Methods for details). The top panel of Fig. 1e shows the optical and microwave measurement sequence. We utilize 1-μs-long green laser pulses for spin initialization and readout, and ~100-ns-long microwave π pulses to induce spin transitions of VₓBₐ spin defects. We sweep the frequency f of the microwave π pulses and measure the fluorescence across the field of view projected on a CMOS camera. When f matches the electron spin resonance (ESR) frequencies, VₓBₐ spin defects are excited to the mₓ = ± 1 states, which are more likely to relax through a non-radiative pathway back to the mₓ = 0 ground state and emit reduced PL. The bottom panel of Fig. 1e shows a series of
ODMR spectra of $V_B$ defect centers measured at room temperature with different external magnetic fields $B_{ext}$ applied along the out-of-plane direction. For $B_{ext}=0$, the energy level of the $m_s = \pm 1$ states of $V_B$ spin defects exhibit a small separation of -100 MHz due to the off-axis zero field splitting effect\cite{42,46,53} and the average ESR frequency equals -3.47 GHz at room temperature\cite{42,46,53}. For $B_{ ext}>0$, the Zeeman coupling separates the $m_s = -1$ and $m_s = +1$ spin states by an energy gap of magnitude $2\gamma B_{ext}$ (see Supplementary Information Note 3 for details), where $\gamma$ denotes the gyromagnetic ratio of $V_B$ defect centers. To characterize the quantum coherence of $V_B$ spin defects, we perform spin relaxometry measurements with a measurement protocol shown in the top panel of Fig. 1. A microsecond scale green laser pulse is first applied to initialize the $V_B$ spin defects to the $m_s = 0$ state. During the time delay, fluctuating magnetic fields at the ESR frequencies will accelerate spin relaxation of $V_B$ spin defects. After a delay time $t$, we measure the occupation probabilities of the $V_B$ spin defects at the $m_s = 0$ and $\pm 1$ states by applying a microwave $\pi$ pulse on the corresponding ESR frequencies and measuring the spin-dependent PL by a green-laser readout pulse. The bottom panel of Fig. 1f shows the integrated PL intensity of the $m_s = 0$ and $\pm 1$ states measured as a function of the delay time $t$. By fitting the data with a three-level model\cite{51,56}, the spin relaxation rate $T_1$ of $V_B$ spin defects is obtained to be 39 KHz (38 KHz) for $m_s = 0 \rightarrow -1 (+1)$ transition at 295 K, consistent with previous studies\cite{56,53,53}.

After demonstrating the ODMR and spin relaxometry measurement capabilities, next, we use $V_B$ spin defects in hBN to directly image magnetic textures of an exfoliated FGT flake. Wide-field magnetometry exploits the Zeeman splitting effect of the ensembles of $V_B$ defect centers to measure the local magnetic stray fields generated from the proximate FGT flake, as illustrated in Fig. 2a. It is worth noting that the $V_B$ spins are naturally orientated along the out-of-plane direction\cite{14}, serving as an ideal local sensor to investigate the magnetic dynamics and phase transition of FGT with spontaneous perpendicular anisotropy. The magnitude of the local static magnetic field $B_{ ext}$ can be extracted as follows: $B_{ ext} = \pi \Delta f_{ ESR} / \gamma$, where $\Delta f_{ ESR}$ characterizes the Zeeman splitting of the $V_B$ spin defects. Subtracting the contribution from the external magnetic field $B_{ ext}$, the magnetic stray field $B_{ tot}$ generated from the FGT flake can be quantitatively measured. By performing spatially dependent ODMR measurements over the $V_B$ spin ensembles, we are able to obtain a 2D stray field map as shown in Fig. 2b, which is measured at a temperature $T = 6K$ and an external magnetic field $B_{ ext} = 142 G$. Through well-established reverse-propagation protocols\cite{30,32,38} (see Supplementary Information Note 3 for details), we can reconstruct the corresponding magnetization $4\pi M$ map of the FGT flake, as shown in Fig. 2c. The spatially averaged magnetization of the FGT flake is 1.3 kG at 6 K, in qualitative agreement with the bulk value\cite{31}. The variation of the local magnetization could result from inhomogeneities, magnetic domains, or localized defects\cite{39,52}.

We now present systematic wide-field magnetometry results to directly image the magnetic phase transition of the FGT flake across the Curie temperature. Figure 3a–g show the reconstructed magnetization maps of the FGT flake measured with temperatures varying from 6 to 225 K and an external perpendicular magnetic field $B_{ ext}$ of 142 G. In the low temperature regime ($T < 100 K$), the exfoliated FGT flake exhibits robust magnetization, as shown in Fig. 3a, b, indicating a

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**Fig. 2** | Wide-field imaging of magnetization of an exfoliated FGT flake by $V_B$ spin defects in hBN. a Schematic illustration of quantum sensing of local stray fields $B_t$ generated from FGT by proximate $V_B$ spin defects. b, c Two-dimensional maps of static stray field $B_t$ (b) and reconstructed magnetization $4\pi M$ (c) of an exfoliated FGT flake measured at 6 K with an external perpendicular magnetic field $B_{ ext}$ of 142 G. The black and purple dashed lines in Fig. 2b outline the boundary of the FGT and hBN flake, respectively, and the scale bar is 5 $\mu m$.

**Fig. 3** | Quantum imaging of temperature dependence of FGT magnetization. Reconstructed magnetization $4\pi M$ maps of the FGT flake at $B_{ ext} = 142$ G and temperatures of 6 K (a), 60 K (b), 115 K (c), 162 K (d), 169 K (e), 176 K (f), and 225 K (g), respectively. The white dashed lines outline the boundary of the exfoliated FGT flake, and the scale bar is 5 $\mu m$. h Temperature dependence of spatially averaged magnetization of the FGT flake (blue points), in agreement with the variation behavior of the normalized Hall resistance presented in arbitrary units (a. u.) (red curve). The magneto-transport results were measured in a separate FGT flake with similar thickness.
Fig. 4 | Quantum imaging of field dependence of FGT magnetization. Reconstructed magnetization (4m) maps of the FGT flake measured at 178 K with an external perpendicular magnetic field \( B_{\text{ext}} \) of 142 G (a), 180 G (b), 199 G (c), 266 G (d), 326 G (e), 399 G (f), and 698 G (g), respectively. The white dashed lines outline the boundary of the FGT flake, and the scale bar is 5 μm. h Field dependence of spatially averaged magnetization of the FGT flake (blue points), consistent with the variation behavior of the normalized Hall resistance presented in arbitrary units (a. u.) (red curve). The magneto-transport results were measured in a separate FGT flake with similar thickness.

In summary, we have demonstrated \( V_\text{B} \) spin defects in hBN as a local probe to image magnetic phase transitions and spin fluctuations in the archetypical van der Waals ferromagnet FGT at the nanoscale. The spatially resolved wide-field magnetometry results reveal the characteristic evolution behavior of magnetic domains during the phase transition of FGT. By using \( V_\text{B} \) spin relaxometry techniques, we are also able to access the spin fluctuations in the FGT flake, whose magnitude reaches a maximum value around the Curie temperature. Our results illustrate the appreciable capability of \( V_\text{B} \) spin defects hosted by hBN.
of investigating local magnetic properties of layered materials in van der Waals heterostructure formats. The presented measurement platform also shows optimal field sensitivity with nanoscale sensor-to-sample distance\(^\text{164,45}\), offering new opportunities for advancing the state of the art of existing quantum sensing technologies. While the current study is conducted using wide-field magnetometry with a spatial resolution set by the optical diffraction limit, we anticipate that the spatial sensitivity of \(V_{\text{B}}\) spin defects could potentially reach the tens of nanometers regime by utilizing single-spin defects and developing scanning microscopy measurement schemes\(^\text{24,44,50,57,65,66}\), opening the possibility of uncovering detailed microscopic features in a broad range of 2D material systems.

**Methods**

**Materials and device fabrication**

The hBN and FGT crystals used in this study were commercially available from 2D Semiconductors. Thin samples were mechanically exfoliated onto Si/SiO\(_2\) (285 nm) substrates. hBN flakes were subsequently irradiated by Helium ions with an energy of 5 keV and a dose of \(5 \times 10^{13}\) cm\(^{-2}\). Irradiated hBN flakes with desirable lateral shape and dimensions are selected for fabricating FGT/hBN bilayer devices. FGT/hBN stack was prepared using the standard polydimethylsiloxane stamp process\(^\text{67}\). Selected flakes were picked up one-by-one by a stamp consisting of a thin layer of polycarbonate on polydimethylsiloxane, then released onto an Au transmission line pre-patterned on a separate Si/SiO\(_2\) (285 nm) substrate. The residual polycarbonate on the device was dissolved in chloroform before measurements. All the device fabrication processes involving handling FGT flakes were performed inside a glovebox filled with argon to minimize environmental degradation. Multiple control samples have been prepared and tested to ensure the reproducibility of the presented results (see Supplementary Information Note 5 for details).

**Pulsed ODMR and spin relaxometry measurements**

Pulsed ODMR and spin relaxometry measurements were performed by a wide-field microscope. The prepared FGT/hBN heterostructure was positioned in a closed-cycle optical cryostat allowing for measurements from 4.5 K to 350 K. Microsecond-long green laser pulses used for spin initialization and readout were generated by an electrically driven 515-nm laser. The laser beam spot width after passing the objective was about 25 \(\mu\)m \(\times\) 25 \(\mu\)m, and was subsequently focused on the hBN layer. Fluorescence of \(V_{\text{B}}\) spin defects was imaged using a CMOS camera. Pulses to drive the green laser and to trigger the camera exposure were generated by a programmable pulse generator. Continuous microwave currents were generated using Rohde & Schwarz SGS100a and/or Rohde & Schwarz SMB100a signal generators. 100-nanosecond-long microwave current pulses were generated by sending the continuous microwave currents to a microwave switch (Mini-Circuits ZB3PD-63-S+) and amplified by +50 dB (Mini-Circuits ZHL-25W-63+) before being delivered to the on-chip Au stripline. The external magnetic field was generated by a cylindrical NdFeB permanent magnet attached to a scanning stage. The field sensitivity and other merits of the presented ODMR measurement platform are discussed in Supplementary Information Note 6 in detail.

**Data availability**

All data supporting the findings of this study are available from the corresponding author on reasonable request.

**Code availability**

All code not included in the paper are available upon reasonable request from the corresponding authors.

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Author contributions
M.H. performed the optical measurements and analyzed the data with H.L., N.M., S.L., and H.W.J.Z. and D.D. prepared the devices with assistance from M.A. and J.S.D.C. performed ion implantations. C.R.D. supervised this project.

Competing interests
The authors declare no competing interests.

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