

# Nanoscale magnetic field imaging for 2D materials

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# 1 Technical Review

2  
3 Imaging weak magnetic field patterns on the nanometer-scale and its application to 2D  
4 materials

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## 8 9 Abstract

10 Nanometer-scale imaging of magnetization and current density is the key to deciphering the  
11 mechanisms behind a variety of new and poorly understood condensed matter phenomena. The  
12 recently discovered correlated states hosted in atomically layered materials such as twisted bilayer  
13 graphene or van der Waals heterostructures are noteworthy examples. Manifestations of these states  
14 range from superconductivity, to highly insulating states, to magnetism. Their fragility and  
15 susceptibility to spatial inhomogeneities limits their macroscopic manifestation and complicates  
16 conventional transport or magnetization measurements, which integrate over an entire sample. In  
17 contrast, techniques for imaging weak magnetic field patterns with high spatial resolution overcome  
18 inhomogeneity by measuring the local fields produced by magnetization and current density. Already,  
19 such imaging techniques have shown the vulnerability of correlated states in twisted bilayer graphene  
20 to twist-angle disorder and revealed the complex current flows in quantum Hall edge states. Here, we  
21 review the state-of-the-art techniques most amenable to the investigation of such systems, because  
22 they combine the highest magnetic field sensitivity with the highest spatial resolution and are  
23 minimally invasive: magnetic force microscopy, scanning superconducting quantum interference  
24 device microscopy, and scanning nitrogen-vacancy center microscopy. We compare the capabilities  
25 of these techniques, their required operating conditions, and assess their suitability to different types  
26 of source contrast, in particular magnetization and current density. Finally, we focus on the prospects  
27 for improving each technique and speculate on its potential impact, especially in the rapidly growing  
28 field of two-dimensional materials.

## 29 30 Introduction

31 In the early 1800s, images of the stray magnetic fields around permanent magnets and current-  
32 carrying wires made with tiny iron filings played a crucial role in the development of the theory of  
33 electromagnetism. Today, magnetic imaging techniques continue to provide invaluable insights well  
34 beyond producing pretty pictures. They shed light on magnetization patterns, spin configurations, and  
35 current distributions, which are invisible in optical or topographic images. Unlike bulk measurements  
36 of transport, magnetization, susceptibility, or heat capacity, they provide microscopic information  
37 about length-scales, inhomogeneity, and interactions. This kind of local information is proving crucial

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38 in ongoing efforts to understand and harness an emerging class of two-dimensional (2D) van der Waals  
39 (vdW) materials and their heterostructures.

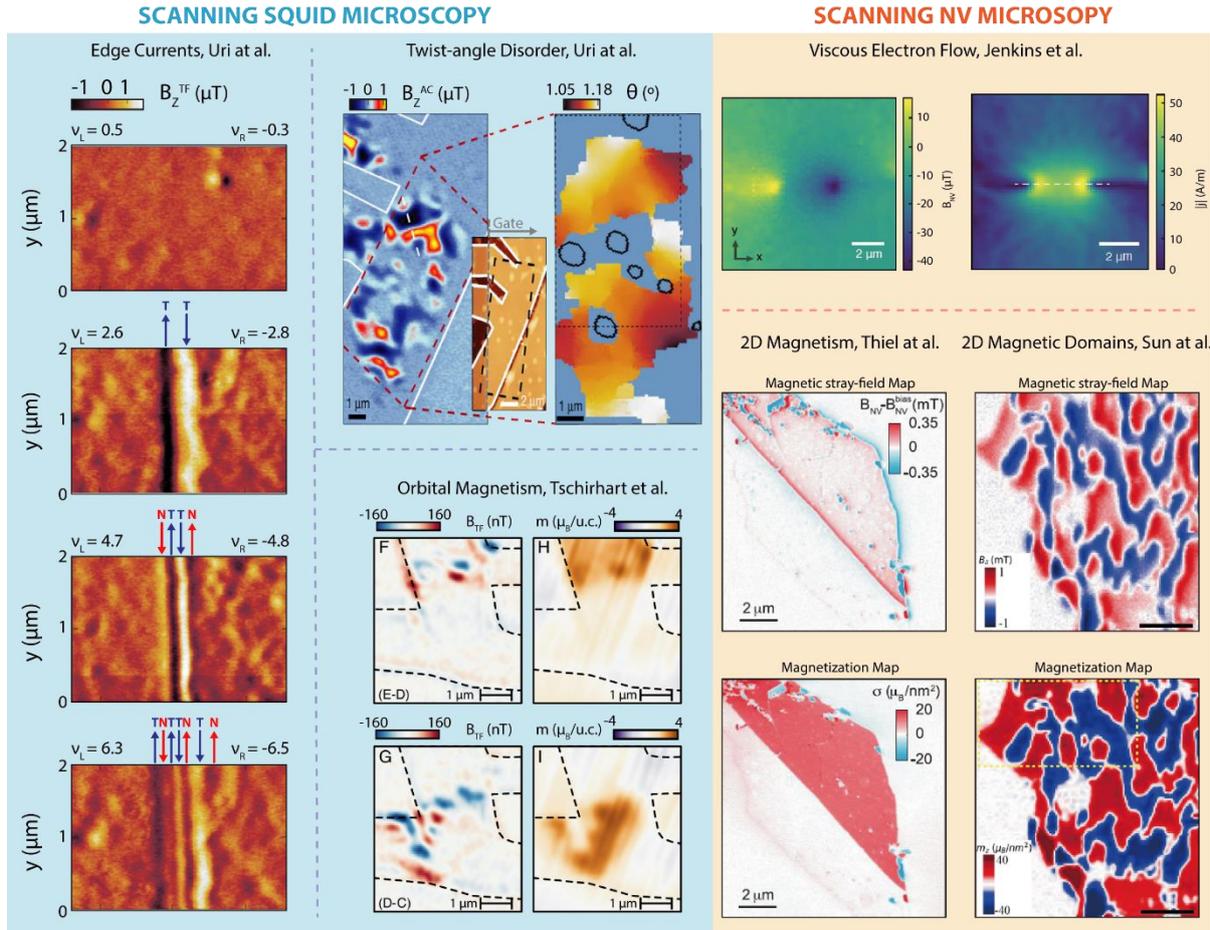
40 The demonstration of the first graphene device in 2004 [1] launched the field of 2D and layered  
41 materials. Graphene itself, however, represents just one of manifold atomically thin vdW materials  
42 with a variety of compositions and crystal structures. Furthermore, heterostructures of these  
43 materials can be engineered, due to the weak vdW interactions that typically dominate their interlayer  
44 coupling: these interactions allow the stacking and twisting of individual atomically thin layers without  
45 lattice mismatch adversely affecting the quality of the structure. This flexibility in both material choice  
46 and structure design has led to the synthesis and fabrication of 2D materials with a range of properties  
47 than span those of insulators, semiconductors, and metals.

48 Recent observations of correlation phenomena such as superconductivity, Mott insulating states, and  
49 magnetically ordered states in such materials are particularly intriguing and are just beginning to be  
50 understood. Such macroscopic manifestations of quantum mechanics are sensitive to the local  
51 environment. In many cases, nanometer-scale spatial resolution is required to investigate and identify  
52 the conditions for their emergence. As a result, there is now an urgent need for sensitive and high-  
53 resolution imaging to zero-in on the nanometer-scale mechanisms behind these phenomena. In  
54 particular, the techniques most adept at tackling this problem are scanning probe microscopies (SPMs)  
55 designed to map subtle magnetic field patterns non-invasively. In 2D systems, such maps can be used  
56 to image magnetization configurations and charge transport, giving crucial local information on  
57 quantum phases, including on the spatial variation of order parameters, the presence of domains, and  
58 the role of defects.

59 In the last few decades, the development of magnetic imaging technologies has been driven by  
60 applications in magnetic storage and information processing. The need to understand magnetostatics  
61 and dynamics on the nanometer-scale and with high temporal resolution has led to powerful optical,  
62 electron, x-ray, and scanning probe microscopies. There are a number of excellent reviews on these  
63 techniques and their myriad applications [2–7]. Most of these techniques, however, are not suitable  
64 for resolving the weak contrast produced by both magnetization and current density in atomically thin  
65 vdW materials.

66 This Technical Review focuses on the subset of state-of-the-art techniques best equipped for this  
67 timely task. Scanning superconducting quantum interference device (SQUID) microscopy has already  
68 demonstrated its ability to map superconducting currents [8] and magnetization [9] in magic-angle  
69 twisted bilayer graphene (MATBG) or quantum Hall edge channels in mono-layer graphene [10,11].  
70 Scanning nitrogen-vacancy (NV) center microscopy has been used to image layer-dependent  
71 magnetization in Cr-based vdW magnets [12–14], as well as hydrodynamic electron flow in  
72 graphene [15,16] and  $WTe_2$  [17]. Sensitive forms of magnetic force microscopy (MFM), including  
73 dissipation microscopy and nanowire (NW) MFM [18,19], are also poised to make an impact.

74 We treat these techniques in the following, briefly explaining how each works and specifying its  
75 magnetic sensitivity and spatial resolution limits. We also touch on the process of reconstructing  
76 spatial maps of measured magnetic field into images of magnetic moment or current. Finally, we  
77 compare the different microscopies and speculate on which is most suitable for which type of contrast  
78 and how each might best be applied in measurements of 2D materials.



79  
 80 Figure 1: Recent SPM measurements of magnetic field on 2D systems. On the left are measurements  
 81 carried out by SSM (clockwise from top left): [8–10]; on the right by SNVM (clockwise from  
 82 top): [12,13,15]. Measurements show magnetic field due both to magnetization and current density.

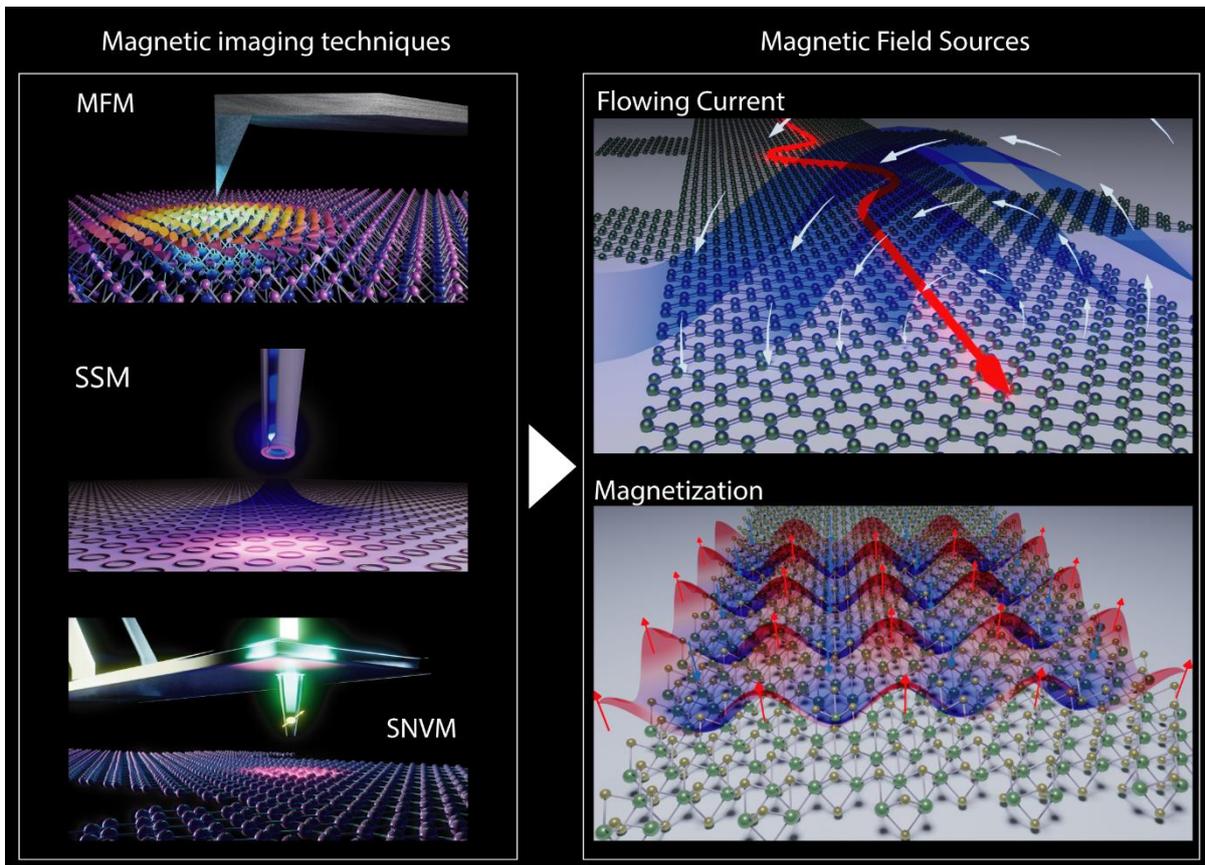
83

84 **Imaging magnetization and current**

85 Mapping magnetization patterns is important for investigations of magnetic domains,  
 86 antiferromagnetism, magnetic skyrmion phases, and the spin-Hall effect. Measurements directly  
 87 sensitive to magnetization include synchrotron-based x-ray techniques, neutron diffraction, and  
 88 electron polarization techniques. For most, the tiny total magnetic moment of atomically-thin  
 89 materials complicates their application to 2D systems. Particularly sensitive techniques such as  
 90 magneto-optic microscopy and spin-polarized scanning tunneling microscopy have been used to  
 91 reveal layer-dependent magnetism in flakes of CrI<sub>3</sub> [20] and films of CrBr<sub>3</sub>, grown by molecular beam  
 92 epitaxy [21], respectively. However, the spatial resolution of magneto-optical techniques is limited to  
 93 the micrometer-scale and interference effects can obscure magnetic signals in thin samples. SP-STM  
 94 requires atomically-clean conducting surfaces, which can often only be obtained by thermal annealing.  
 95 Because many magnetic vdW materials are volatile at high temperatures, this step is sometimes not  
 96 possible. Magnetic imaging via the magnetic circular dichroism of x-ray photoemission electron  
 97 microscopy has not yet been applied to 2D systems. Nevertheless, its sensitivity should be sufficient  
 98 to resolve single layer magnetism [22]. Huang et al. provide a recent survey on the application of  
 99 magnetization-sensitive techniques to 2D materials and especially to 2D magnets [23].

100 Here, we consider techniques capable of mapping magnetic stray field, because they are applicable to  
 101 a wider set of phenomena than direct magnetization imaging. Stray fields are produced not only by  
 102 magnetization patterns, but also by current distributions. Transport imaging can be used to visualize  
 103 local disorder, bulk and edge effects, electron guiding and lensing, topological currents, viscous  
 104 electron flow, microscopic Meissner currents, and the flow and pinning of superconducting vortices.  
 105 Common methods of mapping field include the use of fine magnetic powders as demonstrated by  
 106 Bitter, Lorentz microscopy, electron holography, and a number of SPM techniques. Those most  
 107 applicable to 2D systems, for their combination of high spatial resolution and high magnetic field  
 108 sensitivity, are MFM, scanning SQUID microscopy (SSM), and scanning NV center microscopy (SNVM).

109 Although, in general, a map of magnetic field cannot be reconstructed into a map of the source current  
 110 or magnetization distribution, under certain boundary conditions the source can be uniquely  
 111 determined. In particular, for 2D structures such as 2D materials, patterned circuits, thin films, or  
 112 semiconductor electron and hole gases, a spatial map of a single magnetic field component can be  
 113 used to fully reconstruct the source current or out-of-plane magnetization distribution. Since some of  
 114 the most interesting and elusive effects are observed over length-scales of less than  $1\ \mu\text{m}$  and with  
 115 currents less than  $1\ \mu\text{A}$  or magnetizations of few  $\mu\text{B}/\text{nm}^2$ , techniques are required with both  
 116 nanometer-scale spatial resolution and a sensitivity to fields smaller than a  $\mu\text{T}$ .



117  
 118 Figure 2: Schematic showing the principal magnetic imaging techniques and sources of magnetic field  
 119 discussed in this review.

120

## 121 Imaging weak magnetic field patterns with high spatial resolution

122 In SPM, high spatial resolution is achieved by minimizing both sensor size and its distance from the  
123 sample. High sensitivity is obtained by maximizing signal-to-noise ratio for the magnetic signal of  
124 interest and the fundamental noise of the measurement. In evaluating the sensitivity of different  
125 techniques to magnetic contrast, we follow Kirtley [24] and consider their response to two idealized  
126 sources of magnetic field: a magnetic dipole moment and a line of current. This procedure allows us  
127 to assess and compare the sensitivity of each technique to magnetization and current density in a  
128 sample below.

## 129 Magnetic force microscopy

### 130 *Working principle and conditions*

131 Near surfaces, the most common technique for imaging magnetic fields with high spatial resolution is  
132 MFM, which was introduced in the late 1980s as a natural extension of atomic force  
133 microscopy [25,26]. Contrast results from the magnetostatic interaction between the stray magnetic  
134 fields of a sample and the magnetic tip of a mechanically compliant scanning probe. The vibration  
135 frequency and amplitude of a cantilever probe, whose tip has been coated with a ferromagnetic film,  
136 are recorded as the probe is scanned above a sample. The response typically depends on a gradient  
137 of the stray field. Although some simplifying assumptions can often be made, extracting exact  
138 magnetic field maps from MFM images involves a deconvolution requiring knowledge of the shape  
139 and magnetization configuration of the tip.

140 MFM is possible under a wide variety of conditions, including in air, liquid, vacuum, and over a broad  
141 range of temperatures. As shown in Fig. 3, a MFM system consists of the cantilever, piezoelectric  
142 positioners for moving the sample, and a setup for detecting cantilever motion, usually by optical  
143 deflection or interferometry. Scan areas are typically in the range of a few micrometers on a side and  
144 take several minutes. The cantilever's mechanical frequency, typically a few hundred kHz, sets the  
145 upper limit on the speed of the dynamics that can be measured. In fact, measurement bandwidths are  
146 limited to tens of Hz due to the linewidth of the mechanical resonance or the speed of the phase-  
147 locked loop used for determining the cantilever's frequency.

148 Cantilevers are typically made from Si, SiO<sub>2</sub>, or Si<sub>3</sub>N<sub>4</sub> and their tips are coated with a magnetic film of  
149 Co or Ni. Because cantilevers are optimized to probe surfaces on the atomic-scale, they are designed  
150 to have spring constants around 1 N/m, which is smaller but on the order of spring constant of an  
151 atomic bond at the surface of a solid. As a result, conventional MFM can have extremely high spatial  
152 resolution, down to 10 nm [27,28] at cryogenic temperatures and in vacuum, but more typically from  
153 30 to 100 nm. This large spring constant, however, makes MFM responsive only to strong magnetic  
154 field modulations on the order of tens of T/(m Hz<sup>1/2</sup>) (few μT over 100 nm measured in 1 s). It is,  
155 therefore, well-suited for the measurement of highly magnetized samples, however, ineffective for  
156 detecting the weak stray fields produced by subtle magnetization patterns or Biot-Savart fields of  
157 currents flowing through nanometer-scale devices.

158 The advent of cantilever probes consisting of individual nanowires (NWs) [29,30] or even carbon  
159 nanotubes [31] have given researchers access to much smaller force transducers. This reduction in  
160 size implies both a better force sensitivity and potentially a finer spatial resolution [32]. Sensitivity to  
161 small forces provides the ability to detect weak magnetic fields and therefore to image subtle  
162 magnetic patterns; tiny concentrated magnetic tips have the potential to achieve nanometer-scale  
163 spatial resolution, while also reducing the invasiveness of the tip on the sample under investigation.

164 NWs have been demonstrated to maintain force sensitivities around 1 aN/Hz<sup>1/2</sup> near sample surfaces  
165 (within 100 nm) when operated in high vacuum and at cryogenic temperatures, due to extremely low

166 noncontact friction [33]. In recent proof-of-principle experiments, both magnet-tipped NWs and fully  
 167 magnetic NWs were shown to be sensitive to magnetic field gradients of just a few mT/(m Hz<sup>1/2</sup>) [10]  
 168 and a few nT/Hz<sup>1/2</sup> [19], respectively. These are the gradients and fields produced by tens of  $\mu_B/\text{Hz}^{1/2}$ ,  
 169 where  $\mu_B$  is a Bohr magneton, or several nA/Hz<sup>1/2</sup> of flowing current, each at a distance a hundred or  
 170 so nanometers.

#### 171 *Sensitivity to different types of contrast*

172 Depending on the type of transducer and its tip, MFM maps magnetic field or magnetic field gradients.  
 173 The ultimate noise limiting these measurements is thermal noise acting on the transducer. Such noise  
 174 causes random fluctuations in the measured vibration amplitude and frequency. As shown in Box 1,  
 175 thermal noise sets a minimum measurable magnetic field or field gradient, depending on the  
 176 measurement type and the magnetization configuration of the tip. For example, a frequency shift  
 177 measurement of a conventional MFM transducer [34] has a thermal limit at 4 K to static gradients of  
 178  $\left(\frac{\partial B}{\partial r}\right)_{min} \approx 30 \text{ T}/(\text{m Hz}^{1/2})$ . Recently demonstrated NW MFM has a thermal limit for the same  
 179 measurement that is about 1000 times smaller [19].

#### 180 *Box 1: MFM*

181 Force microscopy contrast is generated by the interaction of a cantilever tip with the sample  
 182 underneath. By monitoring the vibration amplitude, one can measure tip-sample forces at the  
 183 cantilever resonance frequency, while by monitoring the vibration frequency, one can measure static  
 184 tip-sample force gradients. The ultimate noise limiting these measurements is the thermal (Brownian)  
 185 motion of the cantilever. Thermal noise sets a minimum measurable resonant force  $F_{min} = \sqrt{4 k_B T \Gamma}$   
 186 in an amplitude measurement and a minimum measurable static force gradient  $\left(\frac{\partial F}{\partial r}\right)_{min} =$   
 187  $\frac{1}{r_{rms}} \sqrt{4 k_B T \Gamma}$  in a frequency measurement, where  $k_B$  is the Boltzmann constant,  $T$  is the  
 188 temperature,  $\Gamma$  is the mechanical dissipation,  $r_{rms}$  is the cantilever oscillation amplitude, and  $\hat{r}$   
 189 indicates the direction of cantilever oscillation.

190 In MFM, the magnetic tip transduces a magnetic field profile into a force profile. This interaction can  
 191 often be approximated using a point-probe model, in which an effective magnetic multipole –  
 192 including a monopole  $q$  and a dipole  $\mathbf{m}$  – represents the magnetization distribution of the tip. A  
 193 magnetic field profile  $\mathbf{B}$  then produces a magnetic force acting on the cantilever given by  $\mathbf{F}_{MFM} =$   
 194  $q \mathbf{B} \cdot \hat{r} + \nabla(\mathbf{m} \cdot \mathbf{B}) \cdot \hat{r}$ . Note that, in most cases, the contribution of the torque generated by  $\mathbf{B}$  is  
 195 negligible. For conventional MFM, where the tip-sample interaction can be approximated by a pure  
 196 magnetic monopole, this results in a minimum measurable resonant magnetic field  $B_{min} =$   
 197  $\frac{1}{q} \sqrt{4 k_B T \Gamma}$  and a minimum measurable static magnetic field gradient  $\left(\frac{\partial B}{\partial r}\right)_{min} = \frac{1}{q r_{rms}} \sqrt{4 k_B T \Gamma}$ .  
 198 Purely dipolar tips, such as those on the ends of some NWs [18], are sensitive to a further spatial  
 199 derivative of the magnetic field, compared to monopolar tips. Similar expressions can be written  
 200 limiting those measurements.

201 By comparing the thermal noise background to the expected magnetic field or field gradient from a  
 202 single Bohr magneton  $\mu_B$  or a line or current  $I$ , as calculated in Box 2, we can assess the sensitivity of  
 203 MFM. For example, conventional MFM scanning 50 nm above a sample is sensitive to frequency shifts  
 204 equivalent to a magnetic moment of a few thousand  $\mu_B/\text{Hz}^{1/2}$  or currents of a few  $\mu\text{A}/\text{Hz}^{1/2}$  [34]. The  
 205 same type of measurement carried out with newly demonstrated NW MFM probes 100 nm above a  
 206 sample is about 100 times more sensitive to each type of contrast [18,19]. Estimates of sensitivity to  
 207 magnetic moment and current as a function of probe-sample spacing are shown in Figs. 4 a) and b).

208 It should be noted that the thermal limit on frequency measurements is rarely reached in practice.  
 209 Most frequency measurements are limited by other noise sources, such as temperature variations,  
 210 adsorption-desorption noise, or other microscopic mechanisms intrinsic to the resonator, that are  
 211 typically an order of magnitude larger [35]. On the other hand, measurements of resonant oscillation  
 212 amplitude, which are sensitive to modulations at the mechanical frequency of the sensor (typically in  
 213 the 100 kHz regime), are often thermally limited. In such measurements, conventional MFM  
 214 cantilevers can be sensitive to a few hundred  $\mu_B/\text{Hz}^{1/2}$  or a few hundred nA/Hz<sup>1/2</sup>, while NW MFM  
 215 transducers reach down to a few  $\mu_B/\text{Hz}^{1/2}$  or a few nA/Hz<sup>1/2</sup>.

#### 216 *Box 2: Magnetic field sources*

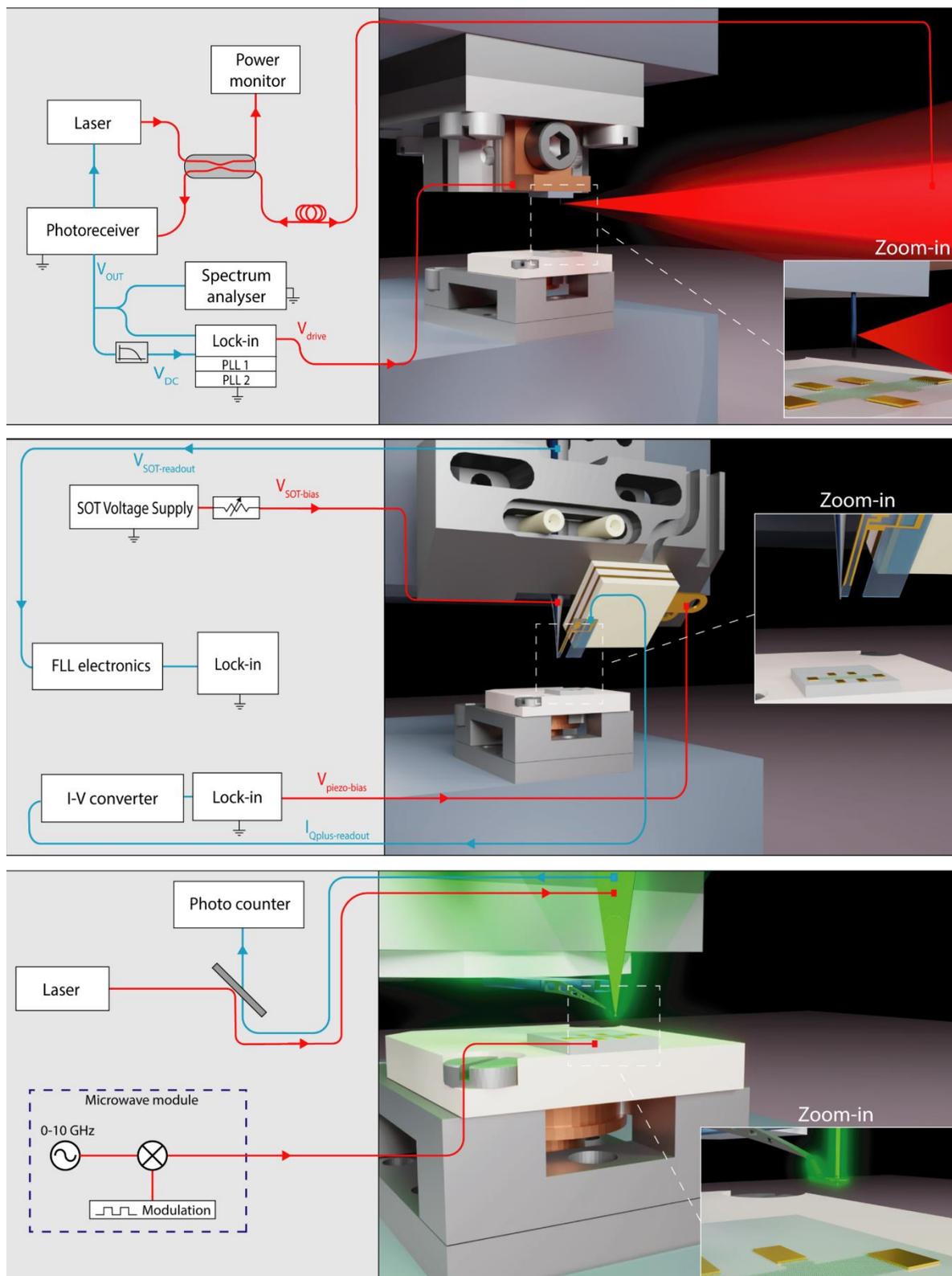
217 The magnetic field of a magnetic moment  $\mathbf{m}$  at distance  $\mathbf{r}$  is given by  $\mathbf{B}_m = \frac{\mu_0}{4\pi r^3} \left( \frac{3(\mathbf{m}\cdot\mathbf{r})\mathbf{r}}{r^2} - \mathbf{m} \right)$  and  
 218 the magnetic field of a line of current  $I$  is given by:  $\mathbf{B}_I = \frac{\mu_0 I \times \mathbf{r}}{2\pi r^2}$ , where  $\mu_0$  is the vacuum permeability.  
 219 Using these two equations, we can express the various quantities measured by our scanning probe  
 220 sensors as a function of tip-sample spacing in terms of  $\mu_B$  of magnetic moment or A of current. For  
 221 example, for SNVM measuring the z-component of the stray magnetic field, the maximum measurable  
 222 signal from a single  $\mu_B$  moment pointing along the z-direction at a tip-sample spacing  $z$  is  $B_{\mu_B,z} =$   
 223  $\frac{\mu_0 \mu_B}{2\pi z^3}$ , while the maximum from a line of current  $I$  flowing in the plane is  $B_{I,z} = \frac{\mu_0 I}{4\pi z}$ . Similar expressions  
 224 can be written for the maximum magnetic flux in the z-direction from the same moment and current  
 225 measured by SSM:  $\Phi_{\mu_B,z} = \frac{\mu_0 \mu_B R^2}{2(z^2 + R^2)^{3/2}}$ , where  $R$  is the SQUID radius and  $\Phi_{I,z} =$   
 226  $\frac{\mu_0 I D}{4\pi} \ln \left( \frac{D^2 + 4z^2 + D\sqrt{D^2 + 4z^2}}{D^2 + 4z^2 - D\sqrt{D^2 + 4z^2}} \right)$ , where  $D$  is the length of one side of a square SQUID loop (to simplify the  
 227 calculation, the current is integrated over a square rather than a circular loop). The corresponding  
 228 maximum static magnetic field gradients measured by standard MFM are:  $\frac{\partial B_{\mu_B,z}}{\partial z} = \frac{3\mu_0 \mu_B}{2\pi z^4}$  and  $\frac{\partial B_{I,z}}{\partial z} =$   
 229  $\frac{3\sqrt{3}\mu_0 I}{16\pi z^2}$ .

#### 230 *Applications to 2D materials*

231 Despite the lack of conventional MFM studies on 2D materials, researchers are starting to employ  
 232 high-sensitivity MFM probes to visualize correlated states in 2D systems via frequency shift maps,  
 233 which can ultimately be reconstructed in current density or magnetization contrast. Such images  
 234 would be particularly useful for measuring the spatial localization of flowing currents, as in edge states,  
 235 and for the determination of length scales such as magnetic domain sizes and coherence lengths.  
 236 Visualizing current flow in MATBG [36] and  $\text{Wt}_2$  [37,38] while they are electrostatically tuned into  
 237 their superconducting states, would help reveal the origin of this superconductivity and whether or  
 238 not it is topological. NW MFM may also help provide direct evidence for magnetism in 2D magnets or  
 239 even in the 2D semiconductor, monolayer  $\text{MoS}_2$  [39,40]. Optical spectroscopy has provided evidence  
 240 of a high-field spin-polarized state in this material, however, confirmation of its presence via a direct  
 241 measurement of magnetic field has not yet been possible. NW MFM's high sensitivity and ability to  
 242 operate in high-field conditions make it promising for such an investigation.

243 MFM can also be used to map dissipation in a sample by measuring the power required to maintain a  
 244 constant oscillation amplitude. This type of contrast maps the energy transfer between the tip and  
 245 the sample and provides excellent contrast for nanometer-scale magnetic structure [41]. Since energy  
 246 dissipation plays a central role in the breakdown of topological protection, it may provide important  
 247 contrast in spatial studies of strongly correlated states in 2D vdW materials. Dissipation contrast has  
 248 been used to observe superconducting [42] and bulk structural phase transitions [43], as well as the  
 249 local density of states. 2D materials engineering allows for the fabrication of devices, in which a variety

250 of different physical phases can be accessed by the application of a gate voltage. Local measurements  
 251 of dissipation via MFM could be an important tool for making spatial maps of the transitions between  
 252 those states.



253 Figure 3: Representative schematic diagrams for the field-sensitive SPMs most applicable to 2D  
 254 systems for their combination of high spatial resolution and high magnetic field sensitivity. From top  
 255 to bottom these are NW MFM, SSM via SOT, and SNVM. In the bottom portion, each diagram shows  
 256

257 a sample mounted on a movable stage, actuated by piezoelectric positioners. For scale, the white  
258 sample-holder in each diagram is 12 x 12 mm in lateral size. Above this sample, is the scanning probe  
259 along with its corresponding readout scheme. Insets show zoomed-in views of each probe, which  
260 more clearly depict the detection schemes. For NW MFM, in red, incident from the right, we see the  
261 focused laser light used for interferometric detection of the NW's flexural motion. For the SSM, we  
262 see the mechanically-coupled tuning fork used for tip-sample distance control. For the SNVM, in  
263 green, incident from above, we see focused laser light for NV excitation.

## 264 Scanning SQUID microscopy

### 265 *Working principle and conditions*

266 Taking advantage of a SQUID's extreme sensitivity to magnetic flux, SSM was first realized in the early  
267 1980s [44]. Contrast results from the magnetic flux threading through a superconducting loop that is  
268 interrupted by at least one JJ. The SQUID's critical current is periodic in this flux – given by the magnetic  
269 field integrated over the area of the loop  $\Phi_z = \int \mathbf{B} \cdot d\mathbf{A}$  – with a period given by the flux quantum  
270  $\Phi_0$ . By applying the appropriate current bias, one can detect voltages across the SQUID which  
271 correspond to changes in magnetic field threading the SQUID loop corresponding to fractions of a  $\Phi_0$ ,  
272 typically down to  $10^{-6} \Phi_0/\text{Hz}^{1/2}$ . For imaging applications, a DC SQUID with two JJs is most often used.  
273 This loop – or a pick-up loop inductively coupled to it – is scanned above a target sample in order to  
274 map the magnetic field profile. The loop's size is minimized in order to optimize spatial resolution.  
275 SQUIDs operate only below a superconducting transition temperature, which is typically below 10 K,  
276 but can be above the temperature of liquid nitrogen (77 K) for some high- $T_c$  superconductors.

277 As shown in Fig. 3, an SSM system consists of the SQUID sensor or pick-up loop and piezoelectric  
278 positioners for moving the sample. In high-sensitivity and high-resolution applications, these elements  
279 are in a cryostat and in vacuum. Precise control of the sensor-sample distance can be achieved, for  
280 example, by coupling it to a micromechanical tuning fork [45,46]. As in MFM, scan areas are in the  
281 micrometer range and take several minutes. The SQUID sets the system's ultimate bandwidth, which  
282 can be in the GHz range, however stray capacitance, cabling, and detection electronics typically limit  
283 the bandwidth to tens of MHz or below.

284 As imaging resolution has improved from the micrometer- down into the nanometer-scale, a number  
285 of strategies have been employed to realize ever-smaller sensors, which simultaneously retain high  
286 magnetic flux sensitivity and can be scanned in close proximity to a sample. One strategy has involved  
287 miniaturizing the pick-up loop of a conventional SQUID and placing it at the extreme corner of the chip  
288 where it can come close to a sample. The most advanced of such devices use a loop with a 200-nm  
289 inner diameter to achieve sub-micrometer imaging resolution and a sensitivity of  $130 \text{ nT}/\text{Hz}^{1/2}$  [47].  
290 Although this design has the advantage of allowing for susceptibility measurements, the size of the  
291 sensor and minimum distance from the sample, which together determine the imaging resolution, are  
292 limited by the complex fabrication process. In the last decade, this limitation has been addressed  
293 through the development of SQUID-on-tip (SOT) sensors, consisting of a SQUID fabricated by shadow  
294 evaporation or directional sputtering of a metallic superconductor directly on the end of a pulled  
295 quartz tip [48,49]. This process has resulted in scanning SQUID sensors with diameters down to 50 nm,  
296 100 nm imaging resolution, and a sensitivity of  $5 \text{ nT}/\text{Hz}^{1/2}$  [50].

### 297 *Sensitivity to different types of contrast*

298 The noise limiting the measurement of magnetic flux in a SQUID arises from several sources including  
299 Johnson noise, shot noise,  $1/f$  noise, and quantum noise [24]. For SQUIDs smaller than  $1 \mu\text{m}$  and at  
300 frequencies high enough to avoid  $1/f$  noise, quantum noise sets the fundamental limit on detectable  
301 flux to be  $\Phi_Q = (\hbar L)^{1/2}$ , where  $\hbar$  is Planck's constant and  $L$  is the loop inductance [24,51,52]. State-

302 of-the-art SOT sensors made from Pb combine the highest flux sensitivity with the smallest sensor  
303 size. In the white-noise limit (measured in the kHz range), sensors with 50 nm diameter reach  $\Phi_{min} =$   
304  $50 \text{ n}\Phi_0/\text{Hz}^{1/2}$ , which is about 4 times larger than  $\Phi_Q$  [50]. Near DC (measured in the Hz range), where  
305 the same sensor is limited by 1/f noise,  $\Phi_{min}$  is about 10 times larger. In these devices,  $L$  is dominated  
306 by kinetic rather than geometric inductance. For this reason, optimizing material parameters for low  
307 kinetic inductance provides the best route for improving  $\Phi_{min}$ .

308 What this sensitivity means in terms of magnetization or current sources requires knowing the tip-  
309 sample spacing. Using the best 50-nm-diameter SOT at a spacing of 50 nm – closer approach than the  
310 characteristic sensor size does not improve spatial resolution – the white noise level is equivalent to  
311 the field of a few  $\mu_B/\text{Hz}^{1/2}$  or a few tens of  $\text{nA}/\text{Hz}^{1/2}$ , while at DC the device is ten times less sensitive.  
312 Again, such estimates are shown as a function of probe-sample spacing in Figs. 4 a) and b).

### 313 *Applications to 2D materials*

314 SSM has already been successfully used to image current density via local measurements of Biot-  
315 Savart fields. In particular, maps of the flow of equilibrium currents in graphene made using SOT  
316 probes revealed the topological and non-topological components of edge currents in the quantum  
317 Hall state [10]. The non-topological currents, which are of opposite polarity to the topological  
318 currents, were predicted theoretically [53], but are not typically considered because they do not affect  
319 conventional transport measurements [54]. In fact, although previous SPM experiments, including  
320 Kelvin probe [55], scanning single-electron transistor [56], and scanning capacitance [57], revealed the  
321 presence of compressible and incompressible regions, non-topological currents were never observed.  
322 This new insight into the microscopic make-up of orbital currents in the quantum Hall systems was  
323 made possible by the SSM's sensitivity to tiny magnetic fields. Similar images of equilibrium currents  
324 in MATBG, revealed the twist-angle disorder in these samples with a resolution and over an extent  
325 not possible by other techniques [8]. In those experiments, SSM also provided a direct correlation  
326 between the degree of disorder and the presence of correlated states, including superconductivity. In  
327 another set of measurements, SSM with the same kind of SOT sensor found evidence for orbital  
328 magnetism in twisted bilayer graphene [9]. Images of the weak orbital magnetization and the  
329 presence of micrometer-scale domains, both of which have not been previously observed, were –  
330 once again – made possible by the technique's sensitivity to magnetic field combined with its spatial  
331 resolution.

332 Given the SOT's exquisite sensitivity to local temperature, such probes can also be applied to measure  
333 local sources of dissipation, as was demonstrated in experiments on graphene [58,59]. Similar  
334 scanning probe measurements of magnetic field and dissipation could be carried out on other moiré  
335 systems, including twisted transition metal dichalcogenides and twisted multi-layer graphene. These  
336 systems are also predicted to host a variety of correlated states, including superconductivity, Mott  
337 insulating states, magnetic states, and Wigner crystal states [60].

### 338 *Scanning NV center microscopy*

#### 339 *Working principle and conditions*

340 Following proposals in 2008 pointing out its potential for high-resolution, high-sensitivity magnetic  
341 field imaging [61,62], the last decade has seen a flurry of activity in the development of SNVM. In this  
342 scheme, NV centers, which are optically addressable electronic defect spins in diamond, are used as  
343 scanning single-spin sensors. Magnetic field measurements are carried out via optically-detected  
344 magnetic resonance (ODMR) spectroscopy, where the EPR spectrum of the NV is recorded by  
345 simultaneous microwave excitation and optical readout of the defect's spin state as the probe is  
346 scanned in close proximity to the sample surface. Thanks to the technique of single-molecule

347 fluorescence, these experiments can be performed on a single spin [63]. The magnetic field sensitivity  
348 results from a Zeeman shift of the spin resonances. In the regime of a weak orthogonal component of  
349 an external magnetic field, the field component parallel to the NV symmetry axis leads to a linear shift  
350 of the  $m_s = \pm 1$  spin states with a proportionality given by the free-electron gyromagnetic ratio  $\gamma =$   
351  $2\pi \times 28 \text{ GHz/T}$  [64]. The ODMR spectrum is measured as a change in optical intensity as a function  
352 of continuous-wave or pulsed microwave excitation [65]. Other forms of contrast include ODMR  
353 quenching in magnetic fields larger than 10 mT due to energy-level mixing by the off-axis field  
354 component [66,67] and spin relaxometry [83]. The latter probes high-frequency fluctuations near the  
355 NV resonance (GHz range) and allows for the investigation of magnetic fluctuations and spin waves in  
356 ferromagnets [68–70]. Further, dynamical decoupling techniques can be used to perform frequency  
357 spectroscopy in the kHz-MHz range [71,72].

358 In scanning probe applications, as shown in Fig. 3, the NV center is hosted within a crystalline diamond  
359 nanopillar and scanned over the sample of interest [61,73]. State-of-the-art diamond probes are  
360 engineered with shallow NV centers, which are implanted at depths around 10 nm [74], in order to  
361 minimize the distance between the NV center and the sample and thus to optimize both sensitivity  
362 and spatial resolution. However, in most SNVM literature, the NV stand-off distance is 50 to 100 nm,  
363 indicating that NV centers may be deeper than expected. As in MFM and SSM, the sample is scanned  
364 below the probe, usually using piezoelectric positioners, while precise distance control is achieved by  
365 coupling to a micromechanical tuning fork. An objective lens above the probe is used to optically excite  
366 the NV center and to detect its fluorescence.

367 Using advanced sensing protocols and sequences of microwave and laser pulses, scanning NV center  
368 microscopes have achieved field sensitivities down to a few  $\mu\text{T/Hz}^{1/2}$  [75] for DC signals and around  
369  $100 \text{ nT/Hz}^{1/2}$  [17] for AC signals. The best resolutions reported for scanning setups are between 15  
370 and 25 nm [76,77], although resolution better than 10 nm should ultimately be possible for optimized  
371 scanning tips with very shallow NV centers. On top of high sensitivity and spatial resolution, scanning  
372 NV microscopy offers additional benefits: a large temperature range – including room temperature –  
373 a quantitative measurement of the magnetic field that is intrinsically calibrated via natural constants,  
374 vector sensitivity, and a number of spin manipulation protocols for performing spectroscopy from DC  
375 to GHz signal frequencies.

376 These advantages notwithstanding, scanning NV microscopy remains challenging at high fields due to  
377 the high microwave frequencies (10s to 100s of GHz) required to actuate the sensor electron spin, and  
378 the spin-level mixing for magnetic fields that are not aligned with the NV symmetry axis [66,67].  
379 Although NV center detection has been reported below 1 K, experiments at cryogenic temperatures  
380 are hampered by reduced photoluminescence contrast and poor charge stability. Furthermore, the  
381 required optical and microwave excitation sometimes poses a limit on the possible samples, since it  
382 can perturb materials such as direct-band-gap semiconductors, nanomagnets, and fragile biological  
383 structures.

#### 384 *Sensitivity to different types of contrast*

385 SNVM is typically limited by photon shot noise from the optical readout, and can be expressed by a  
386 simple signal-to-noise formula typical for optical magnetometry [78]. Specifically, the magnetic  
387 sensitivity of the scanning NV magnetometer is determined by a combination of the spin dephasing  
388 or decoherence time  $T_2$ , the optical contrast  $\epsilon$  and the maximum photon count rate  $I_0$ . A generic  
389 estimate for the minimum detectable magnetic field is given by  $B_{\min} \approx [\gamma\epsilon\sqrt{I_0 t_{\text{acq}} T_2}]^{-1}$ , where  $\gamma$  is  
390 the gyromagnetic ratio and  $t_{\text{acq}}$  is the photon integration time. Using typical values ( $\epsilon = 0.2$ ,  $I_0 =$   
391  $200 \text{ kC/s}$ ,  $t_{\text{acq}} = 300 \text{ ns}$ ,  $T_2 = T_2^* = 1.5 \mu\text{s}$ ), the minimum detectable field is about  $1 \mu\text{T/Hz}^{1/2}$  for

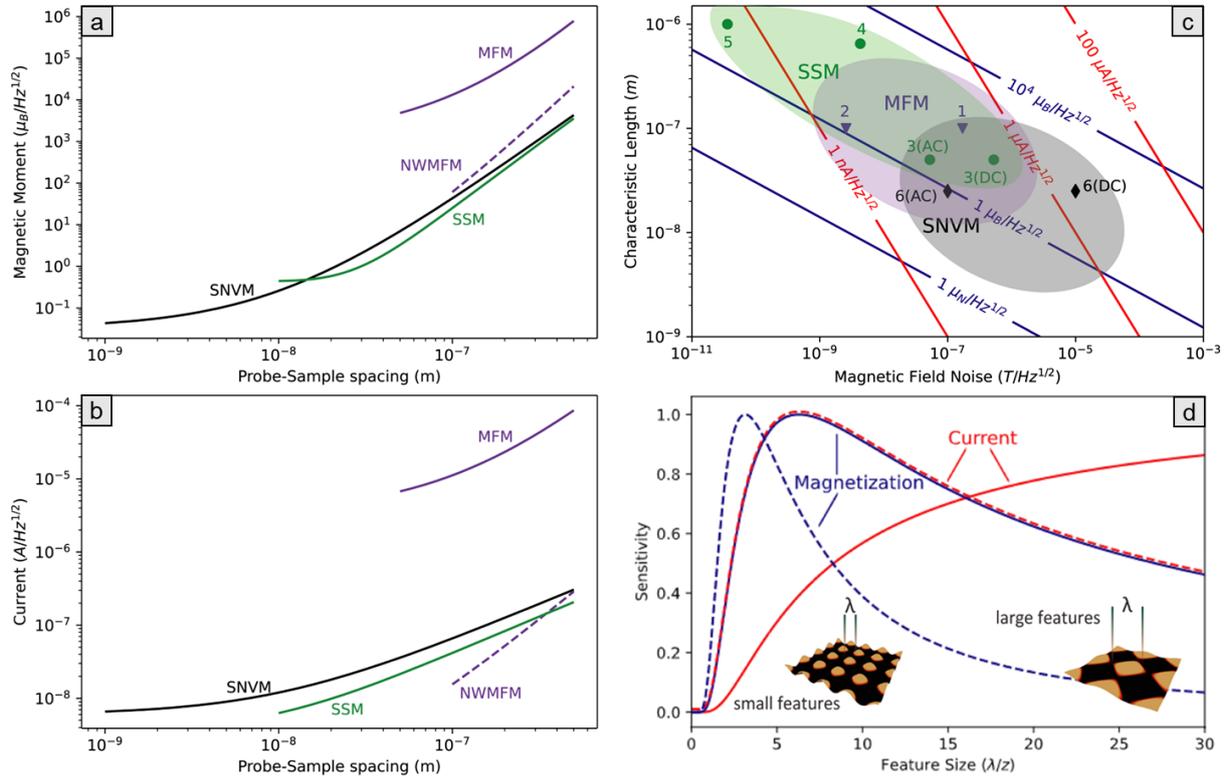
392 pulsed operation and  $10 \mu\text{T}/\text{Hz}^{1/2}$  for continuous-wave operation. Recent SNVM experiments have  
393 shown state-of-the-art pulsed sensitivity of  $100 \text{ nT}/\text{Hz}^{1/2}$  [17]. In the future, the sensitivity can be  
394 improved by extending  $T_2$  using isotopically-purified (free of  $^{13}\text{C}$ ) material [79] and AC magnetometry  
395 techniques [80], improving the contrast through alternative readout schemes [81], and improving the  
396 count rate by photonic shaping [82,83].

397 If we assume the best demonstrated pulsed sensitivity and a 25 nm NV-sample distance, SNVM is  
398 sensitive to one  $\mu_B/\text{Hz}^{1/2}$  or a few tens of  $\text{nA}/\text{Hz}^{1/2}$ . Fig. 2 shows such sensitivity estimates for some of  
399 the best SNVM as a function of probe-sample spacing.

#### 400 *Applications to 2D materials*

401 SNVM has been applied to image magnetization in the 2D ferromagnets [12–14] and current flow in  
402 graphene [15,16,84] and layered semimetals [17]. Given SNVM's particularly high sensitivity to  
403 magnetic moment, the technique is particularly suited for mapping magnetism in vdW magnets to  
404 distinguish domain structure, quantify the strength of the magnetism, and confirm its origin. The  
405 ability to distinguish the magnetism of single atomic layers, as first shown in  $\text{CrI}_3$  [12] and later in  
406  $\text{CrBr}_3$  [13] and  $\text{CrTe}_2$  [14], is crucial for investigating the effect of each layer in vdW heterostructures.  
407 The ability of SNVM to retain high sensitivity at room temperature and under ambient conditions  
408 makes it applicable to magnetic systems with potential practical application in spintronic devices. Sub-  
409 micrometer spatial resolution also distinguishes SNVM from optical techniques such as a Kerr  
410 effect [20,85] and magnetic circular dichroism microscopy [86,87], allowing it to resolve, for example,  
411 domain walls pinned by defects [13]. Moreover, its ability to quantitatively measure stray field allows  
412 the mapping of local 2D magnetization with a precision not possible via optical techniques. High-  
413 frequency sensing with SNVM [88] may also be useful for investigating magnonic excitations in 2D  
414 magnets.

415 Although current mapping at temperatures below 4 K, such as required for studies of  
416 superconductivity in 2D materials, is still challenging, SNVM is ideal for experiments across a broad  
417 and higher temperature range. In fact, researchers have used SNVM to map hydrodynamic flow in  
418 graphene [15] and  $\text{WTe}_2$  [17], which is strongest at intermediate temperatures. The ability to measure  
419 current flow over a wide range of temperatures, allowed, in both of these systems, the observation of  
420 a crossover from diffusive to viscous electron transport. In  $\text{WTe}_2$ , SNVM revealed an unexpected  
421 temperature dependence, indicating that strong electron-electron interactions are likely phonon-  
422 mediated. Similar studies could be carried out in a plethora of other 2D systems, in which viscous  
423 electron transport may dominate under certain conditions.



424

425 Figure 4: Comparing sensitivity and resolution. Plots comparing the sensitivity to magnetic moment  
 426 (a) and current (b) of the 3 magnetic imaging techniques under the most favorable conditions, i.e. in  
 427 vacuum and at liquid helium temperatures. We use parameters from van Schendel et al. for  
 428 conventional MFM [34], Mattiat et al. for NW MFM [19], Vasyukov et al. for SSM [50], and Vool et al.  
 429 for SNVM [17]. MFM and NW MFM sensitivities are based on frequency shift measurements at DC,  
 430 while SNVM and SSM sensitivities are based on AC measurements usually in the tens of kHz range. (c)  
 431 Plot showing the characteristic length and magnetic field noise of state-of-the-art scanning magnetic  
 432 probes under ideal conditions, i.e. in vacuum and at liquid helium temperatures. The characteristic  
 433 length sets the scale of the possible spatial resolution. Diagonal lines show the sensitivity required to  
 434 measure the labelled magnetic moments and currents. Data points correspond to state-of-the-art  
 435 SPMs demonstrated in the corresponding reference: 1, van Schendel et al. [34]; 2, Mattiat et al. [19];  
 436 3, Vasyukov et al. [50]; 4, Kirtley et al. [24]; 5, Jeffery et al. [89]; 6, Vool et al. [17]. (d) Sensitivity as a  
 437 function of feature size, expressed as the ratio between the feature's spatial wavelength  $\lambda$  and the  
 438 probe-sample spacing  $z$ . (Solid lines) Magnetic field imaging is most sensitive to spatially large current  
 439 features (red) and to magnetization features (blue) with a size similar to the probe-sample spacing  $z$ .  
 440 (Dashed lines) Magnetic gradient imaging shifts the maximum sensitivity towards smaller feature size.

441

#### 442 Comparison between techniques

443 Having quantified the sensitivity of MFM, SSM, and SNVM to magnetic moment and electrical current,  
 444 we can now discern which techniques are best suited for mapping which type of contrast. Fig. 4 a) and  
 445 b) show the sensitivity of all techniques to the magnetic field profile produced by a magnetic moment  
 446 and a line of current as a function of probe-sample spacing. In the case of conventional and NW MFM,  
 447 we refer to thermal limit of frequency shift measurements, which applies to DC or low-frequency  
 448 measurements. In the other two cases, we use the minimum flux and field noise achieved in these  
 449 devices in AC measurements in the tens of kHz range.

	MFM (conventional) [27,28,34,90]	MFM (NW) [19]	SSM (susceptometer) [47]	SSM (SOT) [50]	SNVM [17,75– 77]
Sensor size	10-100 nm	100 nm	0.5 $\mu\text{m}$	50 nm	< 1 nm
Sensor stand-off	10-100 nm	50 nm	330 nm	25 nm	50 nm
Spatial resolution	10-100 nm	100 nm	0.5 $\mu\text{m}$	100 nm	15-25 nm
DC sensitivity	10-100 $\mu\text{T}/(\text{Hz})^{1/2}$	3 $\text{nT}/(\text{Hz})^{1/2}$	660 $\text{nT}/(\text{Hz})^{1/2}$	50 $\text{nT}/(\text{Hz})^{1/2}$	4 $\mu\text{T}/(\text{Hz})^{1/2}$
AC sensitivity	170 $\text{nT}/(\text{Hz})^{1/2}$	3 $\text{nT}/(\text{Hz})^{1/2}$	130 $\text{nT}/(\text{Hz})^{1/2}$	5 $\text{nT}/(\text{Hz})^{1/2}$	100 $\text{nT}/(\text{Hz})^{1/2}$
Operating field	< 10 T	< 10 T	< 30 mT	< 1.2 T	< 100s mT
Operating temp.	< 500 K	< 300 K	< 9 K	< 7 K	< 600 K

451 Table 1: Parameters for state-of-the-art magnetic SPM combining the highest-sensitivity with the  
452 highest resolution, based on the devices discussed in the cited references. Values shown in gray  
453 represent estimates based on the properties of the sensors, which have not yet been experimentally  
454 confirmed.

455

456 Together with sensor size, probe-sample spacing sets the spatial resolution of an SPM technique.  
457 Depending on the type of contrast, this spacing also strongly affects sensitivity. SSM sensitivity is not  
458 shown closer than 10 nm, because sensors are difficult to operate closer without a catastrophic crash.  
459 MFM sensitivity is not shown closer than 50 nm and NW MFM is not shown closer than 100 nm,  
460 because the point-probe approximation breaks down at tip-sample spacings smaller than the tip size  
461 and non-contact friction starts to dominate the force noise [91]. Also, at such close spacing, the stray  
462 field produced by the MFM tip at the sample is often invasive. Since SNVM can essentially be operated  
463 in contact with the sample, we plot its sensitivity down to 1 nm of probe-sample spacing.

464 Depending on tip-sample spacing, either SNVM or SSM have the highest sensitivity to magnetic  
465 moment. SSM appears best for tip-sample distances larger than 25 nm, while SNVM is better for closer  
466 approach. Conventional MFM is the least sensitive, while NW MFM is competitive with the other  
467 techniques. While very promising, NW MFM tip size must be reduced from state-of-the-art diameters  
468 of 100 nm in order for the technique to become competitive in high spatial resolution imaging of  
469 magnetic moment.

470 Among proven techniques, SSM is most sensitive to current. While conventional MFM is the least  
471 sensitive, NW MFM appears to surpass all techniques between 500 and 50 nm. Once again, for spatial  
472 resolutions better than 10 nm SNVM appears to be the best choice.

473 Fig. 4 c) provides another way to compare the three techniques, by showing the characteristic length  
474 of each sensor (its size in one dimension) together with its sensitivity to magnetic field. We plot a few

475 state-of-the-art sensors of each type and give an approximate idea of each technique's operating  
476 regime. The characteristic length of a sensor not only sets its ultimate spatial resolution, but also sets  
477 the optimum probe-sample spacing, since closer approach is either impossible or does not improve  
478 sensitivity. Diagonal lines represent the combined probe-sample spacing and field noise required to  
479 achieve a certain sensitivity to magnetic moment or current.

480 Fig. 4 c) makes clear that SNVM has the smallest characteristic length, due to the atomic-scale of the  
481 NV center and the possibility to implant NVs with long coherence times just 10 nm from the surface  
482 of a scanning probe. This makes SNVM the technique of choice for spatial resolution under 25 nm and  
483 for the detection of small magnetizations. Because the magnetic field produced by a magnetic  
484 moment drops off with the inverse cube of the probe-sample distance, a small sensor able to work in  
485 close proximity to the sample is crucial for this type of contrast.

486 Fig. 4 c) also shows that SSM has the highest field sensitivity, but that it comes at the expense of large  
487 sensor size. While conventional MFM appears too insensitive to measure weak magnetization or  
488 current density, the increased force sensitivity of NW MFM makes it competitive with the other two  
489 techniques. In fact, for the measurement of currents, where spatial resolutions better than 100 nm  
490 are not required, SSM and NW MFM are the best techniques. Because Biot-Savart fields fall off only  
491 with the inverse power of the probe-sample spacing, a small sensor is not as important in current  
492 measurements as it is in magnetization measurements.

493 Aside from their sensitivity and resolution, each technique has properties making it more or less  
494 advantageous for certain samples. The strongly magnetic tip of an MFM can produce tens of mT of  
495 magnetic field on a sample 50 nm away. This field can in turn perturb the sample, potentially altering  
496 its state. SNVM requires the excitation of the probe with visible laser light. This optical excitation can  
497 perturb optically active samples below the probe. On the other hand, the stray fields due to the  
498 Meissner effect on an SSM probe are nearly negligible, making these sensors minimally invasive. SSM,  
499 however, is the most limited from the environmental point of view, functioning only at temperatures  
500 below the superconducting transition of the SQUID, typically below 10 K. Both MFM and SNVM  
501 function at a wide range of temperatures and pressures. SSM must also work below its critical field,  
502 which for state-of-the-art SOTs can be as high as a few T. SNVM is also limited in field, in that the  
503 frequency of the microwaves used to address the NV center scale linearly with field and become  
504 impractically high above 1 T.

505

## 506 [Reconstruction of magnetization or current from field images](#)

507 Since magnetic field microscopy techniques do not directly image the current or magnetization  
508 pattern, but rather their stray field, the question arises whether and how the former may be  
509 reconstructed from a stray field map. The relation between stray field and current density is governed  
510 by the Biot-Savart law that, via the concept of bound currents, can also be applied to magnetization.

511 Work in the late eighties by Roth [92] and Beardsley [93] established a framework to compute the  
512 stray fields of two-dimensional current density  $\mathbf{J}(x,y)$  and two-dimensional magnetization patterns  
513  $\mathbf{M}(x,y)$ , respectively. The same work also specified the conditions, in which a reconstruction of  $\mathbf{J}$  and  
514  $\mathbf{M}$  is possible. In particular, they showed that three-dimensional current densities and magnetization  
515 patterns do not produce a unique magnetic stray field pattern, and can therefore not be determined  
516 by stray field imaging. Further, even an arbitrary two-dimensional magnetization pattern does not  
517 possess a unique stray field because the divergence-free part of  $\mathbf{M}$  does not generate an external  
518 stray field and is left arbitrary [93]. A rigorous solution, on the other hand, exists for two-dimensional

519 current densities  $\mathbf{J} = (J_x, J_y, 0)$  and out-of-plane magnetized films  $\mathbf{M} = (0, 0, M_z)$ . It has further been  
 520 shown that this solution can be extended to thick films if the magnetization, or current density, is  
 521 uniform through the thickness [76]. As a consequence, magnetic field imaging is especially useful for  
 522 analyzing 2D systems and thin-film devices.

523 Magnetic field maps do not reproduce all current or magnetization features with the same sensitivity.  
 524 Looking at the mechanics of the reconstruction, shown in Box 3, it becomes clear that features smaller  
 525 than the probe-sample spacing  $z$  produce negligible magnetic field at the sensor location, because  
 526 stray fields decay exponentially with distance from the surface. The decay length is given by  $\lambda/2\pi$ ,  
 527 where  $\lambda$  is the spatial wavelength of the current or magnetization feature, as shown in Fig. 4 d).  
 528 Interestingly, large features compared to the probe-sample spacing, i.e. large  $\lambda/z$ , produce a strong  
 529 signal for currents, but not for magnetization.

530 Imaging magnetic field gradients rather than magnetic fields, allows one to push the maximum  
 531 sensitivity towards smaller feature size. Magnetic gradient detection is the standard mode for MFM,  
 532 but can also be implemented for SSM and SNVM by a mechanical oscillation of the sensor [10,61].  
 533 Using lock-in techniques to demodulate the resulting signal can also significantly reduce noise through  
 534 spectral filtering. Gradient detection is especially attractive for imaging currents, because the  
 535 magnetic gradient image closely resembles the current density image, so that no reconstruction is  
 536 needed [10]. For SNVM, gradient imaging is attractive because it upconverts DC signals to AC where  
 537 much more sensitive magnetometry protocols are available [61,80].

### Box 3: Reconstruction of current density and magnetization from a magnetic field image

539 The current density  $\mathbf{J} = (J_x, J_y)$  and in-plane magnetization  $M_z$  of a two-dimensional sample can be  
 540 conveniently reconstructed from a magnetic field image by expressing the Biot-Savart law in  $k$ -space.  
 541 Assume that we image in a plane at distance  $z$  above the sample, the magnetic stray field, in  $k$ -space  
 542 is given by:  $B_z(k_x, k_y, z) = ig(k, z)[\frac{k_y}{k}J_x(k_x, k_y) - \frac{k_x}{k}J_y(k_x, k_y)]$ , where  $g(k, z) = \frac{1}{2}\mu_0de^{-kz}$  is a  
 543 transfer function with  $d \ll z$  being the film thickness,  $k_x$  and  $k_y$  are the  $k$ -vectors, and  $k =$   
 544  $(k_x^2 + k_y^2)^{1/2}$ . Similar expressions can be derived for  $B_x$  and  $B_y$  as well as for  $d \geq z$  [76,92]. To  
 545 reconstruct the current density from a magnetic field map, the relation is inverted:  $J_x(k_x, k_y) =$   
 546  $-\frac{ik_yWB_z(k_x, k_y, z)}{kg(k, z)}$  and  $J_y(k_x, k_y) = -\frac{ik_xWB_z(k_x, k_y, z)}{kg(k, z)}$ , where  $W$  is a window function, whose cut-off  
 547 wavelength is adjusted to suppress high-frequency noise. Different choices for the window function  
 548 have been reported in the literature, including Hann and rectangular and Tikhonov-based windows.  
 549 The cut-off wavelength typically is of order  $z$ . An expression for reconstructing  $\mathbf{J}$  from an arbitrary  $B$ -  
 550 field component is given in [76].

551 Similar expressions can be derived for reconstructing an out-of-plane magnetization  $M_z$  or to  
 552 reconstruct magnetic gradient images. To reconstruct  $M_z$ , note that  $\mathbf{J} = \nabla \times \mathbf{M}$ , and therefore:  
 553  $B_z(k_x, k_y) = kg(k, z)M_z(k_x, k_y)$  for the forward problem as well as  $M_z(k_x, k_y) = \frac{WB_z(k_x, k_y, z)}{kg(z, k)}$  for the  
 554 reverse problem. To reconstruct a magnetic gradient image, the transfer function incurs an additional  
 555 factor of  $k$  due to the derivative.

556

### 557 Prospects for improvement

558 Improving MFM sensitivity requires stronger magnetic tips or transducers with better force sensitivity.  
 559 Up to an order of magnitude in force sensitivity could be gained by using optimized NW transducers.  
 560 MFM cantilevers have recently been realized with spring constants in the hundreds of mN/m and

561 mechanical quality factors above  $10^6$ , resulting in nearly 100 times more sensitivity than conventional  
562 transducers. In general, however, improving the sensitivity of a mechanical transducer is achieved by  
563 reducing its size [94], as in recent work on NW MFM. Another route to improve magnetic field  
564 sensitivity is to increase the magnetic moment and size of MFM tips. This gain, however, comes at the  
565 cost of reducing spatial resolution and increasing the perturbative effect of the probes, which now  
566 produce larger stray fields at the sample.

567 The spatial resolution of the MFM could be improved by utilizing the sharpest possible magnetic tips.  
568 Extensive work has been done in this area in the context of conventional MFM, achieving spatial  
569 resolutions down to 10 nm [95–98]. Such work could be extended to high-force-sensitivity NW MFM.  
570 Smaller tips, however, have reduced magnetic moment and, consequently, a worse sensitivity to  
571 magnetic field profiles. In order to maintain high sensitivity, in general, the reduction in tip size should  
572 be accompanied with a reduction in transducer size.

573 Improvements in SSM field sensitivity could come from a reduction in the SQUID inductance. Given  
574 that this quantity is dominated by kinetic inductance in state-of-the-art devices, optimizing the  
575 superconducting material from which the device is made could be a fruitful pursuit. Further reduction  
576 of the characteristic size of SSM probes is difficult to imagine. SOT probes have been fabricated with  
577 diameters just under 50 nm. Reducing this size further would make the device size similar to the  
578 thickness of the deposited superconducting film, complicating much of the process, on which the  
579 fabrication is based. SQUIDs with feature sizes of only a few nanometers have been fabricated in YBCO  
580 using a focused ion beam of He [99], raising the possibility of devices that are an order of magnitude  
581 smaller and potentially work at liquid nitrogen temperature. Nevertheless, significant work remains  
582 to be done before such devices can be integrated onto scanning probes.

583 In order to reduce the characteristic length scale of SNVM, a number of researchers have focused on  
584 simultaneously reducing the implantation depth of NV centers and maintaining their coherence  
585 properties. Implantation depths of less than 3 nm have been reported combined with greater than 10  
586  $\mu\text{s}$  coherence times [100], giving a perspective of better than 10 nm imaging resolution combined with  
587 sub-10 nT/Hz<sup>1/2</sup> sensitivity. So far, however, most reported stand-off distances remain between 50  
588 and 100 nm and the best magnetic field sensitivities at 100 nT/Hz<sup>1/2</sup> and significant work may be  
589 needed to reduce either figure of merit.

590

## 591 Conclusion

592 The confluence of substantial improvements in nanometer-scale magnetic imaging with the advent of  
593 engineered 2D materials creates the perfect opportunity to gain new insight into the physics of  
594 correlated states in condensed matter. The unprecedented control provided by layer-by-layer  
595 material engineering gives physicists a vast playground on which to test theories on superconductivity,  
596 magnetism, and other correlated phenomena. With this control, however, comes sensitivity to  
597 disorder and inhomogeneity. In such a fragile environment, local measurements – with sensors whose  
598 characteristic size is smaller than the length scale of the disorder – are essential for making sense of  
599 the system. For this reason, SPM techniques will become ever more important tools in this growing  
600 field, perhaps only losing traction, once fabrication techniques have been honed and substantially  
601 improved.

602 There are a number of SPM techniques, which have emerged as important tools for the investigation  
603 of 2D systems. Conventional atomic force microscopy has been used extensively for topographic  
604 characterization of 2D materials. In graphene, scanning single electron transistors have been used to

605 map the local density of states [101] and for imaging hydrodynamic flow [102]. Scanning gate  
606 microscopy has been used to image localized states [103] and scanning microwave impedance  
607 microscopy for visualizing the structural details of moiré lattices [104]. Electronic properties of 2D  
608 transition metal dichalcogenides have also been studied by scanning tunnelling microscopy [105].  
609 Scanning near-field optical microscopy has even been used to measure polaritonic response in  
610 graphene-hexagonal boron nitride heterostructures [106].

611 As discussed in this review, among these SPM techniques, those involving non-invasive magnetic field  
612 imaging are particularly suited to investigating the correlated states present in 2D systems, because  
613 of their ability to map both current and out-of-plane magnetization. Given the high sensitivity and  
614 spatial resolution required to investigate correlated states in 2D materials, it is important to choose  
615 the appropriate magnetic SPM for the physical system under investigation. The different scaling of  
616 magnetization and current contrast with probe-sample spacing and the different physical quantities  
617 that are measured by various magnetic SPM make certain techniques more amenable to certain  
618 systems. We hope to have provided some insight in this regard, both to experimentalists wanting to  
619 apply magnetic SPM to 2D systems and to physicists working on the next generation of magnetic  
620 imaging techniques.

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