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Imaging crystal stress in diamond using ensembles of nitrogen-vacancy centers

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We present a micrometer-resolution millimeter-field-of-view stress imaging method for diamonds containing a thin surface layer of nitrogen vacancy (NV) centers. In this method, we reconstruct stress tensor elements over a two-dimensional field of view from NV optically-detected magnetic resonance (ODMR) spectra. We use this technique to study how stress inhomogeneity affects NV magnetometry performance, and show how NV $M_{z,\kappa}$ imaging is a useful and direct way to assess these effects. This new tool for mapping stress in diamond will aid optimization of NV-diamond sensing, with wide-ranging applications in the physical and life sciences.

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Nitrogen-vacancy (NV) color centers in diamond are 18 an increasingly popular tool for sensing and imag-19 ing electromagnetic fields and temperature, with wide-20 ranging applications. In particular, widefield 2D mag-21 netic imaging using ensembles of NV centers can pro-22 vide micrometer spatial resolution and millimeter field-23 of-view in ambient conditions, enabling investigations 24 of condensed-matter physics, paleomagnetism, and bio-25 magnetism problems [1-5]. However, one limitation 26 to an NV magnetic imager's sensitivity is intrinsic di-27 amond stress variation, which inhomogeneously shifts 28 the NV ground-state resonance frequencies and spoils 29 the NV spin dephasing time [6]. Diamond crystal stress 30 and strain are therefore important to understand and 31 minimize when optimizing NV magnetometry [7, 8] and 32 magnetic microscopy. 33

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In this work we use an ensemble NV surface laver 34 to image diamond stress across a millimeter-scale field 35 of view, and explore how stress inhomogeneity impedes 36 NV magnetic microscopy. A 532 nm laser illuminates 37 the micrometer-scale NV layer at the top surface of a di-38 amond chip $(4 \text{ mm} \times 4 \text{ mm} \text{ wide and } 0.5 \text{ mm} \text{ thick})$ and 39 an optical microscope images the spin-state-dependent 40 NV fluorescence onto a camera (Fig. 1). Probing the 41 transition frequencies between NV ground-state sub-42 levels by sweeping the frequency of an applied mi-43 crowave field yields an optically-detected magnetic res-44 onance (ODMR) spectrum in each pixel. From the re-45 sulting 2D map of NV resonance frequencies, we extract 46 magnetic field components and crystal stress tensor el-47 ements (which have units of pressure). As crystal stress 48 and strain are related through the compliance tensor 49 (with 1050-1210 GPa Young's modulus for diamond, 50 depending on orientation [9]), we refer to the crystal de-51 fects that induce stress within the diamond (shifting the 52 NV ground-state sublevels and causing birefringence) 53 as strain defects. We first demonstrate the NV stress 54 imaging technique with diamond Sample A, which con-55

tains a nitrogen-rich layer (25 ppm, 13 μ m thick) grown on an electronic-grade single-crystal substrate with ppb nitrogen density. This sample was electron-irradiated and annealed to increase the NV density. We also apply the NV stress imaging technique to several other diamonds (Samples B through J) [10, 11], which also exhibit a variety of strain defects (see Supplementary Material [12]).



FIG. 1. Schematic of the combined NV stress and birefringence imager. The NV stress imager (blue labels) uses a 532 nm laser to illuminate the diamond, an applied microwave field to drive transitions between NV ground-state sublevels, and a bias magnetic field. The birefringence imager (maroon labels) uses an LED illuminator, two linear polarizers, and a quarter-wave plate. Both imagers use the same microscope and CCD camera (black labels) to collect and image the transmitted light. The photograph on the right shows the diamond Sample A studied in this work.

Previous diamond strain imaging studies used X-64 ray topography, Raman spectroscopy, cathodolumines-65 cence, and birefringence to characterize diamond strain 66 and how it affects diamond applications [13–15]. By 67 comparison, NV stress imaging gives a more direct char-68 acterization of how diamond stress inhomogeneity af-69 fects NV magnetic imaging, as both techniques probe 70 the NV ODMR frequencies. In addition, NV stress 71 imaging yields quantitative maps of the diamond stress 72 tensor components localized in the NV layer with mi-73 crometer resolution [16]. The stress tensor reconstruc-74 tion can help identify how strain features formed during 75 diamond sample preparation and thereby inform future 76 sample fabrication. Finally, high-resolution NV stress 77 imaging is essential in ongoing efforts to identify dam-78 age tracks from recoiling carbon nuclei to search for 79 high-energy particle collisions in diamond [17]. 80

In the following sections, we describe NV stress imaging and compare NV-based and birefringence images acquired with the same optical microscope. We next consider how stress inhomogeneity compromises NV magnetometer sensitivity, and then present a survey of common strain defects found in fabricated diamond and their impacts on magnetic microscopy.

88 WIDEFIELD NV STRESS IMAGING

The NV center in diamond consists of a substitu-89 tional nitrogen atom in the carbon lattice adjacent 90 to a vacancy (Fig. 2a). It has an electronic spin-91 triplet ground state (S = 1) with magnetic sublevels 92 $m_s = \{-1, 0, +1\}$. The sublevel energies shift in re-93 sponse to local magnetic fields, crystal stress, tempera-94 ture changes, and electric fields. We measure these en-95 ergy (i.e., frequency) shifts using ODMR spectroscopy, 96 where a resonant microwave field induces transitions be-97 tween the $m_s = 0$ and ± 1 sublevels and causes reduced 98 NV fluorescence under continuous illumination by 532 99 nm laser light (Fig. 2b). Each NV is oriented along one 100 of four crystallographic directions (labeled with the in-101 dex $\kappa = \{1, 2, 3, 4\}$). An NV ensemble usually contains 102 an equal number of NVs for each κ . The ODMR spec-103 tra from all NV orientations yields the information to 104 reconstruct stress tensor elements and vector magnetic 105 field components [3]. 106

We now describe how to extract the local magnetic field and crystal stress from the measured NV resonance frequencies. The NV ground-state Hamiltonian in the presence of stress and a static magnetic field is [18–20]

$$H_{\kappa} = (D + M_{z,\kappa}) S_{z,\kappa}^{2} + \gamma \vec{B} \cdot \vec{S}_{\kappa} + M_{x,\kappa} \left(S_{y,\kappa}^{2} - S_{x,\kappa}^{2} \right)$$

$$+ M_{y,\kappa} \left(S_{x,\kappa} S_{y,\kappa} + S_{y,\kappa} S_{x,\kappa} \right)$$

$$+ N_{x,\kappa} \left(S_{x,\kappa} S_{z,\kappa} + S_{z,\kappa} S_{x,\kappa} \right)$$

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$$+ N_{y,\kappa} \left(S_{y,\kappa} S_{z,\kappa} + S_{z,\kappa} S_{y,\kappa} \right).$$



FIG. 2. (a) NV centers in the diamond lattice, with the four N-V axes shown in green [18]. Carbon atoms are black, nitrogen atoms are red, and vacancies are gray. (b) Example ODMR spectrum with $\vec{B} = \{220, 593, 1520\} \ \mu\text{T}$ in the diamond chip coordinate system (fit function plotted in red). The labels indicate the resonances from the different NV orientations. Each NV resonance is split into three lines due to hyperfine interactions with the spin-1 ¹⁴N nucleus.

Here, $D \approx 2870$ MHz is the zero-field splitting, $S_{i,\kappa}$ are the dimensionless spin-1 projection operators, $\gamma = 2.803 \times 10^4$ MHz/T is the NV electronic gyromagnetic ratio, \vec{B} is the magnetic field in the NV coordinate system, and $M_{i,\kappa}$ and $N_{i,\kappa}$ are terms related to the crystal stress and temperature. The indices $i = \{x, y, z\}$ represent the coordinate system for the particular NV orientation. We neglect the electric-field contributions to Eq. 1, as explained in the Supplementary Material [12]. In addition, if $|\vec{B}| > 1$ mT, as is the case in this work, the contributions from the $\{M_{x,\kappa}, M_{y,\kappa}, N_{x,\kappa}, N_{y,\kappa}\}$ terms are negligible, and Eq. 1 simplifies to

$$H_{\kappa} = (D + M_{z,\kappa})S_{z,\kappa}^2 + \gamma \vec{B} \cdot \vec{S}_{\kappa}.$$
 (2)

When \vec{B} is aligned along the z-axis for one NV orientation, the Hamiltonian for the selected orientation reduces to

$$H_{\kappa} = (D + M_{z,\kappa})S_{z,\kappa}^2 + \gamma B_z S_{z,\kappa}, \qquad (3)$$

and the resonance frequencies are

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$$f_{\pm} = (D + M_{z,\kappa}) \pm \gamma B_z. \tag{4}$$

Measuring f_{\pm} yields the magnetic field projection B_z and the $M_{z,\kappa}$ for that NV orientation. This measurement forms the basis of a sensing modality called Projection Magnetic Microscopy (PMM) [3], where we align the bias magnetic field along the z-axis of each

NV orientation and record the associated resonance fre-164 133 quencies individually. An alternative sensing modality, 134 called Vector Magnetic Microscopy (VMM) [3], allows 166 135 us to determine \vec{B} and all four $M_{z,\kappa}$ terms from a sin-136 gle measurement (Fig. 2b). In VMM, the selected bias 137 magnetic field generates unique Zeeman splittings and 138 non-overlapping ODMR spectra for each NV orienta-139 tion. We extract the magnetic field components and 140 $M_{z,\kappa}$ values by fitting Eq. 2 for all four NV orientations. 141 Both VMM and PMM yield the same $M_{z,\kappa}$ results; we 173 142 detail advantages of each method in the Supplemental 174 143 Material [12, 21, 22]. We used VMM in this work to 175 144 measure the four necessary $M_{z,\kappa}$ maps (which we refer 145 to as "NV $M_{z,\kappa}$ imaging") needed to reconstruct stress 146 tensor elements for each pixel, as described below. 147

Stress tensor reconstruction

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In each pixel, the stress tensor components can be determined from the four $M_{z,\kappa}$ parameters, allowing us to generate a quantitative image of the local stress across the diamond. Following the derivations in Refs. [18–20], we obtain

$$M_{z,1} = a_1 \sigma_{\text{diag}} + 2a_2 \left[\sigma_{XY} + \sigma_{XZ} + \sigma_{YZ} \right], \qquad (5)$$

$$M_{z,2} = a_1 \sigma_{\text{diag}} + 2a_2 \left[\sigma_{XY} - \sigma_{XZ} - \sigma_{YZ} \right], \qquad (6)$$

$$M_{z,3} = a_1 \sigma_{\text{diag}} + 2a_2 \left[-\sigma_{XY} + \sigma_{XZ} - \sigma_{YZ} \right], \quad (7)$$

$$M_{z,4} = a_1 \sigma_{\text{diag}} + 2a_2 \left[-\sigma_{XY} - \sigma_{XZ} + \sigma_{YZ} \right]. \tag{8}$$

Here, $\{a_1, a_2\} = \{4.86, -3.7\}$ MHz/GPa are spin-stress 149 coupling constants [19], σ_{ij} are elements of the 3×3 150 stress tensor in GPa in the coordinate system shown 151 in Fig. 2a, and $\sigma_{\text{diag}} \equiv \sigma_{XX} + \sigma_{YY} + \sigma_{ZZ}$. The σ_{ii} 152 are normal stress terms, while σ_{XY} , σ_{XZ} , and σ_{YZ} are 153 shear stress terms. The σ_{ij} are written in the diamond 154 unit cell coordinate system $\{X, Y, Z\}$ (rather than the 155 NV coordinate system $\{x, y, z\}$ for a given κ), and are 156 felt by all four NV orientations. Each NV orientation 157 exhibits the same $a_1 \sigma_{\text{diag}}$ contribution to $M_{z,\kappa}$. The a_2 158 contributions change as we transform the stress tensor 159 for each of the four NV orientations. 160

Solving Eqs. 5-8 to extract $\sigma_{\text{diag}}, \sigma_{XY}, \sigma_{XZ}$, and σ_{YZ} in each pixel yields

$$\sigma_{\text{diag}} = \frac{1}{4a_1} \left[M_{z,1} + M_{z,2} + M_{z,3} + M_{z,4} \right], \qquad (9)$$

$$\sigma_{XY} = \frac{1}{8a_2} \left[M_{z,1} + M_{z,2} - M_{z,3} - M_{z,4} \right], \qquad (10) \quad {}^{208}_{209}$$

$$\sigma_{XZ} = \frac{1}{8a_2} \left[M_{z,1} - M_{z,2} + M_{z,3} - M_{z,4} \right], \qquad (11) \quad \stackrel{^{210}}{\underset{^{212}}{}}$$

$$\sigma_{YZ} = \frac{1}{8a_2} \left[M_{z,1} - M_{z,2} - M_{z,3} + M_{z,4} \right].$$
(12)

For the σ_{ii} normal stress terms, the measurements pre-161

sented here are only sensitive to the total normal stress ²¹³ 162 σ_{diag} rather than the individual σ_{ii} contributions [19]. 214 163

A more sophisticated algorithm could use VMM spectra measured at several magnetic fields and keep all of the terms in Eq. 1 to obtain each σ_{ii} separately.

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Since the $D + M_{z,\kappa}$ terms change with temperature, $M_{z,\kappa}$ and σ_{diag} can only be evaluated up to an overall constant [23, 24]. However, the shear stress terms should be unaffected by temperature changes, and thus shear stress images are absolute. For measurements acquired with 10 mK temperature stability, an NV $M_{z,\kappa}$ imager can determine $M_{z,\kappa}$ to about 1 kHz, or ~0.1 MPa. As a further example, a 1 $\mu T/\sqrt{Hz}$ magnetic sensitivity per pixel (28 kHz/ $\sqrt{\text{Hz}}$ frequency sensitivity) corresponds to approximately 10 MPa/ $\sqrt{\text{Hz}}$ stress sensitivity.

Figure 3 shows the measured $M_{z,\kappa}$ maps and the resulting $\{\sigma_{\text{diag}}, \sigma_{XY}, \sigma_{XZ}, \sigma_{YZ}\}$ maps for Sample A, illustrating a practical example of NV $M_{z,\kappa}$ and stress imaging. This diamond has a variety of strain features (their origins are described below), in addition to more homogeneous regions. For Sample A and most of the other diamond samples we investigated in this work, we found the shear stress inhomogeneity was greater in σ_{XY} than in σ_{XZ} or σ_{YZ} [12]. The $M_{z,\kappa}$ variations were usually due to σ_{diag} and σ_{XY} inhomogeneity in roughly equal amounts.

COMPARISON WITH BIREFRINGENCE IMAGING

Here we compare NV stress imaging to birefringence imaging, which is a prominent characterization tool in the diamond community [13, 25]. In this work, both methods were implemented within the same optical microscope for a straightforward comparison (Fig. 1). Both the NV $M_{z,\kappa}$ terms and the diamond refractive index depend on crystal stress, but NV stress imaging more directly captures relevant information about stress inhomogeneity in the NV layer and its effects on NV sensing. This makes NV stress imaging the more appropriate tool for optimizing NV diamond samples for magnetic microscopy.

In a birefringent material, light with orthogonal polarizations transmitted through a sample of thickness L accumulates a relative optical retardance phase $\delta =$ $\frac{2\pi}{\lambda}\Delta nL$, where λ is the wavelength and Δn is the difference in refractive indices for orthogonal polarizations. We used a rotating-linear-polarizer method, also known as Metripol, to extract $|\sin \delta|$ by probing the sample with light of varying polarization angles [12, 26-28]. The measured transmission intensity I_i for a given polarizer rotation angle α_i is

$$I_{i} = \frac{1}{2} I_{0} [1 + \sin 2(\alpha_{i} - \phi) \sin \delta].$$
(13)

Here I_0 is the transmittance of a given pixel and ϕ is the retardance orientation angle. Sweeping α_i across



FIG. 3. Example NV $M_{z,\kappa}$ and $\{\sigma_{\text{diag}}, \sigma_{XY}, \sigma_{XZ}, \sigma_{YZ}\}$ maps for Sample A. After measuring the $M_{z,\kappa}$ maps in the top row from the NV resonance frequencies, we calculate the stress tensor element maps in the bottom row using Eqs. 9-12. The diamond chip has high-stress and low-stress regions, and most of the $M_{z,\kappa}$ inhomogeneity comes from σ_{diag} and σ_{XY} stress terms.



FIG. 4. Birefringence $\sin^{-1} | \sin \delta |$ and NV stress maps for the lower-left corner region of Sample A. Both techniques show similar phenomena, though the NV stress imaging maps are immune to the $\delta > \pi/2$ phase ambiguity, can resolve the petalshaped defects localized in the NV layer, separate out strain phenomena into different stress tensor contributions, and predict how strain features affect the NV magnetic microscopy performance.

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180° of polarization rotation allows us to determine I_0 , 229 215 $|\sin \delta|$, and ϕ [12]. 216

Figure 4 shows a comparison between $\sin^{-1} |\sin \delta|$, 217 σ_{diag} , and σ_{XY} maps collected using birefringence and 218 NV stress imaging with the same diamond field of view. 219 Despite the general similarity in results between the two 220 methods, there are some stark differences. The σ_{XY} 221 map shows petal-shaped strain features in the NV layer, 222 whereas the birefringence map (which integrates phase 223 retardance through the entire thickness) does not cap-224 ture these fine details. Furthermore, the NV stress maps 225 can distinguish that the diagonal stripe causing $M_{z,\kappa}$ 226 inhomogeneity arises from $\sigma_{
m diag}$ stress, while the petal- $_{241}$ 227 shaped strain features are caused by σ_{XY} stress. We 242 228

can exploit such component-separated NV stress maps to investigate the sources and phenomenology of observed strain features.

Crystal strain and δ are linearly related through the diamond photo-elastic parameters [29–32]. However, this relationship typically assumes uniform stress over the optical path, meaning that the δ we measure is integrated over L even though the strain may be localized to one layer. By comparison, the NV $M_{z,\kappa}$ technique provides stress information localized to the NV layer, and converting from $M_{z,\kappa}$ to stress tensor elements is more straightforward.

Figure 4 illustrates an additional limitation for birefringence imaging. For high-strain regions, the inte-

grated δ through the sample thickness may be greater 243 than $\pi/2$, leading to ambiguity when calculating stress 244 from $|\sin \delta|$ since multiple δ values can yield the same 245 $|\sin \delta|$. This occurs in the middle of the stripe feature 246 in Fig. 4, where the reconstructed δ reaches its maxi-247 mum value of $\pi/2$ before decreasing. NV stress imaging 248 is not susceptible to this ambiguity. The NV σ_{diag} map 249 instead shows that the stress amplitude increases to the 250 middle of the stripe. Accounting for the extra $\sim \pi/4$ 251 of phase accumulation in the birefringence map yields a 252 maximum stress amplitude of ~ 130 MPa, which is con-253 sistent with the 140 MPa maximum stress amplitude in 254 the σ_{diag} map [12]. Despite the $|\sin \delta|$ ambiguity, the 255 NV and birefringence methods yield consistent stress 256 measurements. 257

STRESS AND NV MAGNETOMETRY 258

NV $M_{z,\kappa}$ inhomogeneity causes each NV in an en-259 semble to have different resonance frequencies, which 260 reduces the magnetic sensitivity and degrades NV mag-261 netometer performance [6]. A useful NV-magnetometer 262 figure of merit is the slope of the ODMR lineshape 263 |F'(f)|, where f is the probe-microwave frequency and 264 F'(f) is the derivative of the NV fluorescence intensity 265 at frequency f (Fig. 1b). The maximum |F'(f)| slope 266 is proportional to the quantity C/Γ , where C is the 267 fluorescence contrast and Γ is the resonance linewidth 268 [33]. $M_{z,\kappa}$ inhomogeneity reduces magnetic sensitivity 269 by making the resonance lineshape broader, the contrast 270 weaker, and thus the maximum slope $|F'(f)| \propto C/\Gamma$ 271 smaller [6]. 272

For NV-diamond magnetometers that use fewer probe 273 microwave frequencies for improved magnetic sensi-274 tivity, $M_{z,\kappa}$ inhomogeneity is even more detrimen-275 tal. High-sensitivity magnetometers typically measure 276 at two microwave frequencies (called the "two-point 277 method") instead of probing the full width of the 278 ODMR lineshape (the "full-sweep method") [34]. The 279 two microwave frequencies are typically chosen to max-280 imize the two-point responsivity (defined as the change 281 in fluorescence per unit frequency shift of the NV res-282 onance). If $M_{z,\kappa}$ varies substantially over the field of 283 view, no pair of frequencies can be optimal for all NVs, 284 resulting in decreased sensitivity for many pixels in the 285 magnetic image. A larger variation in $M_{z,\kappa}$ across the 286 ensemble also implies a narrower magnetic-field range 287 before the NVs in some pixels fall out of resonance. As 297 288 such, $M_{z,\kappa}$ inhomogeneity limits the field of view and 289 dynamic range of high-sensitivity NV magnetic imagers. 299 290 Figure 5 shows a zoomed-in $M_{z,\kappa}$ map together with 291 single-pixel ODMR spectra corresponding to regions of 301 292 Sample A with different local strain properties. For ex-293 ample, one pixel shows a region with a low $M_{z,\kappa}$ gradi-294 ent and offset from the mean (i); a second pixel shows a 295 region with a low $M_{z,\kappa}$ gradient and a high $M_{z,\kappa}$ offset 305 296



FIG. 5. (a) Zoomed-in $M_{z,\kappa}$ map (lower-left corner of Fig. 3), showing the locations of the example pixels. (b)Fitted ODMR spectra for example pixels (i), (ii), and (iii) (green, red, and blue, respectively). Each has varying $M_{z,\kappa}$ gradients and offsets. (c) Derivatives F'(f) for the ODMR lineshapes plotted in (b). Pixel (i) has the best C/Γ slope and two-point responsivity, while pixel (ii) has a good C/Γ slope but a poor two-point responsivity since the $M_{z,\kappa}$ offset means we probe this pixel at a suboptimal microwave frequencies compared to the others. Pixel (iii) has poor C/Γ slope and two-point responsivity due to the high $M_{z,\kappa}$ inhomogeneity in this pixel.

(ii); and a third pixel shows a region with a high $M_{z,\kappa}$ gradient and a low $M_{z,\kappa}$ offset (iii). These local strain conditions are caused by a ~ 0.3 MHz $M_{z,\kappa}$ offset in the diagonal stripe and high $M_{z,\kappa}$ variation in the $\sim 30 \ \mu m$ petal defects. Pixels (i) and (ii) have a comparable C/Γ slope and therefore a comparable NV magnetic sensitivity when using the full-sweep method. However, when using the two-point method optimized for pixel (i), pixel (ii) will have a poor responsivity due to its large $M_{z,\kappa}$

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offset. By comparison, pixel (iii) will exhibit poor per-306 formance with both methods. As these example pixels 307 demonstrate, $M_{z,\kappa}$ inhomogeneity reduces the magni-308 tude and uniformity of the magnetic sensitivity across 309 an image. 310

STRAIN FEATURE SURVEY AND EFFECTS 311 ON NV MAGNETOMETRY 312

We used NV $M_{z,\kappa}$ imaging to study and categorize 313 different types of strain features in diamond samples. 314 As shown in the regions highlighted in Fig. 6, different 315 types of strain features have a variety of typical dimen-316 sions, $M_{z,\kappa}$ amplitudes and gradients, and stress ten-317 sor contributions. From our $M_{z,\kappa}$ maps, we categorized 318 strain features into general types. We identified how 319 each type impacts the C/Γ slope and two-point respon-320 sivity. Here we concentrate on strain features observed 321 in Sample A. Surveys of additional diamonds exhibit-322 ing similar phenomena are included in the Supplemental 323 Material [12]. 324

Figure 6a shows the same field of view as in Fig. 5. 325 The broad-scale plastic deformation in the diagonal 326 stripe is perhaps associated with the lower-left corner of 327 the diamond sample, as high stress is common at sharp 328 corners, edges, and fractures. The stress from the di-329 agonal stripe is largely σ_{diag} stress, causing millimeter-330 scale $M_{z,\kappa}$ gradients, resulting in a wide span in NV 331 resonance frequencies (~ 1 MHz). As anticipated, the 332 diagonal stripe spoils the two-point responsivity while 333 the full-sweep C/Γ slope is largely unaffected. In this 334 example the $M_{z,\kappa}$ span is large enough to cause a nega-335 tive responsivity in the diagonal stripe, as the resonance 336 frequency is offset far enough that one of the probe fre-337 quencies is on the opposite side of its resonance peak. 338

The 20-30 μ m petal-shaped strain defects in Fig. 6a 339 and Fig. 4 are caused by lattice dislocations that can 340 form on top of the seed crystal during homoepitaxial 341 growth, as studied in previous work [13, 25, 35–37]. 342 The three types of lattice dislocations (edge, screw, and 343 mixed dislocations) contribute to different crystal stress 344 terms [38]. The petal features appear most strongly in 345 the σ_{diag} and σ_{XY} maps (and to a lesser degree in the 400 346 σ_{XZ} and σ_{YZ} maps), which suggests that the petal-347 shaped strain features we observed are predominantly 348 caused by edge and mixed dislocations. 349

Figure 6b shows a $\sim 200 \ \mu m$ strain feature (likely 350 caused by a dislocation bundle), surrounded by smaller 351 petal-shaped defects. From birefringence imaging, we 352 know that such strain features are typically edge dislo-353 cations (with σ_{diag} and σ_{XY} stress). They often have 354 four quadrants with lines emanating from the center 409 355 along the [001] and [010] directions, and are a few hun-356 dred micrometers across [39, 40]. As shown in the Sup-357 plemental Material [12], the birefringence map displays 358 lobes associated with the strain feature in Fig. 6b, with 359

the expected orientation. The lobes appearing in the σ_{diag} and σ_{XY} NV stress maps are rotated by 45°. These characteristics lead us to conclude that this strain feature is a dislocation bundle. For this particular strain feature, the range of $M_{z,\kappa}$ values is narrow enough that it has only a minor effect on NV magnetometry performance for both the full-sweep and two-point methods.

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Figure 6c shows a prominent $\sim 30 \ \mu m$ dislocation strain feature. Here the single-pixel $M_{z,\kappa}$ gradients are substantial enough to spoil the NV magnetic sensitivity of both methods. Severe $M_{z,\kappa}$ gradients also interfere when fitting the ODMR spectra to a Lorentzian lineshape model, introducing systematic errors in the extracted resonance frequencies. Such errors can produce false features in NV magnetic images [12].

Figure 6d shows a ~ 0.8 mm X-shaped strain feature. Though visually most similar to the petal-shaped strain features discussed above, X-shaped strain features are larger, display sharp edges pointing along the diamond [100] and [010] directions, and have no lobe structures. The X-shaped strain features also exhibit mainly σ_{diag} and σ_{XY} stress (like an edge dislocation), whereas the σ_{XZ} and σ_{YZ} values are nearly zero. Despite the similarities to the previously-discussed strain features, the origin of the X-shaped strain features remains under investigation. They mainly affect the two-point responsivity for NV magnetometry, whereas the full-sweep C/Γ slope is mostly immune.

SUMMARY AND OUTLOOK

We presented a method for quantitative stress imaging in diamond with micrometer spatial resolution and millimeter field of view using a layer of NV centers. We compared NV stress imaging to the more traditional birefringence imaging method, implemented in the same experimental setup, and found quantitative and qualitative consistency. NV $M_{z,\kappa}$ imaging offers a straightforward way to reconstruct stress tensor elements within a diamond sample and provides a more direct measure of how the strain features affect NV magnetic imaging. NV $M_{z,\kappa}$ imaging is therefore a useful tool to support NV magnetic microscopy and other diamond applications that rely on crystal homogeneity for optimal performance.

To further improve the NV $M_{z,\kappa}$ imaging method, one can implement NV sensitivity and resolution enhancements. For example, one can boost the sensitivity by implementing a double-quantum Ramsey spectroscopy protocol, creating a superposition of the NV $m_s = \pm 1$ magnetic sublevels. This doubles the $M_{z.\kappa}$ part of Eq. 2 and cancels the magnetic contribution [6, 41]. NV $M_{z,\kappa}$ imaging with double-quantum Ramsey spectroscopy should be beneficial for NV layers where the magnetic field inhomogeneity dominates the $M_{z,\kappa}$ inhomogeneity. Furthermore, for specific applications,



FIG. 6. Comparisons between $M_{z,\kappa}$, C/Γ slope, and two-point responsivity for common strain feature types found in Sample A. The C/Γ and responsivity maps are related to the magnetic microscopy performance when using the full-sweep method and the two-point method, respectively. Note that the two-point responsivity is more susceptible to $M_{z,\kappa}$ inhomogeneity, while the full-sweep method can tolerate some range of $M_{z,\kappa}$ offsets.

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one can perform additional measurements to disentan- 439 414

- gle the σ_{ii} normal stress terms. Finally, one can employ 415
- NV super-resolution techniques to map the stress ten-416
- sor components with a resolution beyond the optical 417
- diffraction limit [42, 43]. 418

Looking to future diamond applications for particle 419 physics, diamond stress characterization is important 445 420 for the recently-proposed diamond directional weakly-421 interacting massive particle (WIMP) detector [17]. This 447 422 approach aims to use NV centers to image the stress cre- 448 423 ated by ~ 100 nm tracks from recoiling carbon nuclei. 449 424 Mapping the intrinsic $M_{z,\kappa}$ and stress inhomogeneity 425 is a first step to exploring the feasibility of directional 451 426 WIMP detection with NVs. In particular, since σ_{XZ} 452 427 and σ_{YZ} stress are typically smaller than $\sigma_{\rm diag}$ and 453 428 σ_{XY} stress, detecting deviations in σ_{XZ} or σ_{YZ} may 454 429 exhibit a larger signal-to-background ratio. Anticipated 455 430 next steps include NV $M_{z,\kappa}$ imaging with higher spa-431 tial resolution ($<1 \mu m$) and variable depth; cataloging 457 432 the $M_{z,\kappa}$ distribution from many individual NV cen- 458 433 ters in a low-density bulk sample (ppb NV density); in- 459 434 vestigating hybrid-sensor schemes (such as a combined 460 435 cathodoluminescence/ $M_{z,\kappa}$ method) to rapidly survey 461 436 diamond chips for damaged voxels; and imaging the re- 462 437 coil tracks from implanted ¹²C nuclei. 438

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