A robust diamond sensor for nanoscale sensing and imaging with single nitrogen vacancy centers

P. Maletinsky*, S. Hong*, M.S. Grinolds*, B. Hausmann, M.D. Lukin, R.L. Walsworth, M. Loncar, and A. Yacoby

* These authors contributed equally to this work
The following supplementary material is divided into five sections. Each section provides background information related to specific topics of the main text. The sections are not built upon each other and can be read independently. Section S1 provides details for the model-calculation used to simulate the NV magnetic image in Fig.3d. In section S2, we discuss limitations to NV magnetic imaging if the NV sensor is in close proximity to a strongly magnetized sample. Experimental limitations to the achievable NV-sample distance are discussed in section S3. The fabrication of the sharp metallic tips employed in the measurement for Fig.4a in the main text is detailed in section S4. Finally, section S5 contains a description and simple model for the topographic features observed in Fig.4a in the main text.

S1. SIMULATION OF MAGNETIC IMAGES

In order to reproduce the magnetic images obtained with the scanning NV sensor, we performed a model-calculation of the local magnetic fields in proximity to the hard-disc sample we imaged in our experiment. The magnetic domains were approximated by an array of current-loops in the sample-plane as illustrated in Fig. S1a. We chose the sizes of the loops to match the nominal size of the magnetic bits on the sample (bit-with 200 nm and bit-length 125 nm and 50 nm for the tracks in the figure) and set the current to 1 mA (corresponding to a density of \( \approx 1 \) Bohr magneton per \((0.1 \text{ nm})^2\)), which we found to yield the best qualitative match to the magnetic field strengths observed in the experiment. We then applied Biot-Savart’s law to this current-distribution to obtain the magnetic field distribution in the half-plane above the sample.

Fig. S1b shows the resulting magnetic field projection onto the NV center at a scan height of 50 nm above the current loops. The NV direction was experimentally determined to be along the ([0\( \bar{1} \bar{1} \bar{1} \]) crystalline direction of the diamond nanopillar (in a coordinate-system where \( x- \), \( y- \), and \( z- \) correspond to the horizontal-, vertical and out-of plane directions in Fig. S1b), by monitoring the NV-ESR response to an externally applied magnetic field (using 3-axis Helmholtz-coils). We then allowed for slight variations of the NV orientation due to alignment errors between the diamond crystallographic axes and the scan directions to find the NV orientation that reproduced our experimental data best. With this procedure, we found an NV orientation \( (\sqrt{2}\sin(\phi), \sqrt{2}\cos(\phi), 1)/\sqrt{5}, \) with \( \phi = \pi 162/180. \)
FIG. S1: Simulation of NV response to bits of a magnetic memory. (a) Current distribution used to simulate the magnetic bits imaged in this work. Red (blue) loops indicate currents of 1 mA in the (counter-)clockwise direction. (b) Magnetic field generated by the current-distribution in (a), projected on the NV axis at a height of 50 nm above the current loops. The NV axis was tilted by 37° out of the scan-plane ([111] crystalline direction) with an in-plane component as illustrated by the blue arrow. (c) NV magnetometry response obtained from the magnetic field distribution in (b), assuming a Lorentzian NV-ESR response and RF detunings as in the original experiment (see text). Note that this experimental situation only resolves magnetic field lines corresponding to the microwave detuning (here: $B_{NV} = \pm 3$ G).

Finally, we used this magnetic-field distribution to calculate the response of the NV center to a magnetometry scan as described in the main text. For this, we assumed a Lorentzian ESR response with a full-width at half maximum of 9.7 MHz, a visibility of 20 % and two external RF sources with detunings $\pm 10$ MHz from the bare ESR frequency, all in accordance with our original experimental parameters.

S2. NV MAGNETOMETRY IN CLOSE PROXIMITY TO A STRONGLY MAGNETIZED SAMPLE

The presence of a strong magnetic field $B_\perp$, transverse to the NV axis leads to a reduction of contrast in optically detected ESR and moreover reduces the overall fluorescence intensity of the NV center. These effects result from a mixing of the NV spin-levels in the optical ground and excited states of the NV center in the presence of $B_\perp$. Such mixing on one hand allows for spin non-conserving optical transitions and on the other hand suppresses...
FIG. S2: Quenching of NV fluorescence and ESR contrast in hard-disc imaging. (a) Total NV fluorescence $I_{\text{norm}}$ as a function of sample position for an NV in close proximity to the hard-disc sample. $I$ was normalized to the average fluorescence intensity of $I_0 \approx 15000$ cps in the scan. Dark regions in the scan correspond to individual magnetic domains and are caused by strong magnetic fields transverse to the NV axis which occur in close proximity to the domains. (b) NV magnetic image recorded simultaneously with (a). Data acquisition and integration time per pixel was analogous to the magnetic imaging described in the main text. However here, due to strong transverse magnetic fields, NV ESR contrast almost completely disappeared and prevented NV magnetic imaging using optically detected ESR. The color-bar applies to (a) and (b). (c) Line-cut along the white line in (a), averaged over 7 adjacent pixels. $I_{\text{norm}}$ shows a periodicity of $\approx 64$ nm, indicating a bit-width of 32 nm. (d) Fluorescence approach curve on the magnetic memory medium. NV fluorescence $I$ was normalized to the fluorescence rate $I_\infty = 27'000$ cps when the NV center was far from the sample. In contact with the magnetic sample (last data-point to the right), NV fluorescence was reduced by almost a factor of two compared to the NV counts far from the sample. (e) Magnetic imaging with the same NV sensor: Even in close contact to the sample, NV magnetic imaging using ESR is still possible, albeit with a strongly reduced ESR contrast and signal to noise ratio compared to the data shown in the main text. Data in (e) was acquired over 180 minutes, for the smallest resolvable magnetic domains (top third of image) we measure a mean width of 16.5 nm. The laser power was set to 100 $\mu$W in a-d and 60 $\mu$W in e.

...the spin-dependance in shelving from the NV excited state (triplet) to the metastable NV singlet state. Both, spin-conservation under optical excitation and spin-dependant shelving are responsible for the non-zero contrast in optically detected ESR of NV centers.
consequently, their suppression with transverse magnetic fields explains the disappearance of NV magnetometry features when closely approaching a strongly magnetized sample.

Fig. S2a shows the raw NV fluorescence counts observed when scanning an NV in a diamond nanopillar in close proximity (estimated $10 - 20$ nm distance between NV and sample surface) to the sample. Dark features appear when the NV is scanned over magnetic bits that enhance $B_\perp$, while the inverse happens when $B_\perp$ is reduced (or the longitudinal field $B_{NV}$ enhanced) by local fields. This mode of bit-imaging allows for spatial resolutions $\approx 20 - 30$ nm (Fig. S2c). At the same time, a magnetic image recorded with the technique described in the main text shows no appreciable imaging contrast (Fig. S2b). Only exceedingly long integration times on the order of hours allowed us to reveal weak magnetic features with dimensions on the order of $20$ nm (Fig. S2d).

The rates of the two effects which lead to a disappearance of ESR contrast, i.e. spin-flip optical transitions and shelving of $m_s = 0$ electronic states into the metastable singlet, scale approximately as $(B_\perp D_{GS} - D_{ES})^2$ and $(B_\perp D_{ES})^2$, respectively, with $D_{GS(ES)}$ the ground- (excited-) state zero-field spin-splitting of $2.87$ GHz and $1.425$ GHz, respectively. Given that $D_{GS} \approx 2D_{ES}$, the scaling of the two mechanisms with $B_\perp$ will be very similar. The characteristic scale of $D_{ES} (D_{GS}/2)$ for the disappearance of ESR contrast thus allows us to estimate $B_\perp$ close to the sample to be $B_\perp \approx D_{ES}/\gamma_{NV} \approx 514$ Gauss. We note however that this simple argument likely gives an over-estimation of $B_\perp$ as smaller values can already significantly affect ESR contrast and NV fluorescence intensity due to the complex dynamics of NV spin pumping. Indeed, strong reductions of NV fluorescence rates for $B_\perp$ less than $100$ G have been observed in the past. Transverse magnetic fields on this order were consistent with the largest on-axis magnetic fields observed on our experiments as well as with the calculations of magnetic field profiles presented in Sect. S1 (for the parameters used in Fig. S1, we obtain maximal values of $B_\perp \approx 200$ Gauss for an NV-to-sample distance of $20$ nm).

S3. LIMITATIONS TO NV-SAMPLE DISTANCE

As mentioned in the main text, NV-sample distance is an essential parameter for the performance of our microscope as it determines the overall resolving power with which weak magnetic targets can be imaged. We identified three critical parameters that can affect NV-sample distance:
FIG. S3: **Contamination of diamond tips.** (a) AFM image of the end of a scanning diamond nanopillar after contamination during scanning. The image was acquired by scanning the diamond nanopillar over a sharp diamond tip as shown in Fig. S4. (b) AFM Image of the same nanopillar as in (a) after cleaning of the pillar’s end-face by repeated “scratching” over the sharp diamond tip.

- **Implantation-depth of NV centers in the diamond nanopillars**
  The depth of the NV centers created using ion implantation can be controlled by the energy of the ions used for NV creation. However, the stopping of ions in matter is a random process and the depth of the created NV centers therefore not perfectly well-defined. This straggle in ion implantation poses an intrinsic uncertainty to the distance between the scanning NV and the end of the diamond nanopillar. For implantation energies of 6 keV (with nominal implantation-depths of 10 nm) as used in this work, NV straggle has recently been shown to be as large as $10 - 20$ nm. We note that since straggle in NV implantation is hard to circumvent it is essential for the future to develop techniques to precisely pre-determine the depth of a given sensing NV in a diamond nanopillar. This could be performed using recently developed nanoscale imaging methods for NV centers, or by scanning the NV sensor over a well-defined magnetic field source.

- **Contamination of scanning diamond nanopillars**
  During scanning-operation, the scanning diamond nanopillar can gather contamination from the sample or environment. An example for such a contaminated diamond-tip is shown in the AFM image shown in Fig. S3a (which was acquired with the scanning
protocol employed in Fig.4, using the a sharp diamond tip as shown in Fig. S4). Such contamination can artificially increase the distance of the scanning NV center to the sample by several 10’s of nm (see Fig. S3a). To undo contamination of the diamond-tip after excessive scanning over dirty samples, we developed a “tip-cleaning technique” that allowed us to revert a contaminated tip to its initial, clean state (as illustrated by the transition from Fig. S3a to b). Tip cleaning is performed by repeated scanning of the diamond nanopillar over a sharp diamond tip (Fig. S3a) in the absence of AFM feedback. Such feedback-free scanning can partly remove contamination from the diamond pillar, which after repeated operation leads to a clean device as the one shown in Fig. S3b.

We note that with proper sample-cleaning, control over environmental conditions and occasional “tip-cleaning” runs, adverse effects of tip-contamination can be essentially eliminated. This, together with the excellent photo-stability of NV centers, then allows for long-term operation of the scanning NV sensor.

- **AFM control**

Proper AFM control is necessary to assure close proximity of the NV center to the sample surface. It has been shown in the past that bad mounting or improper AFM feedback control can lead to AFM tip-sample distances in excess of 20 nm. Careful mounting of AFM tips and proper setup and tuning of AFM feedback (here provided by an Attocube ASC500 controller) was therefore essential to observe, for instance, the fluorescence quenching features discussed in Fig.4 of the main text.

## S4. FABRICATION OF SHARP DIAMOND TIPS

For the experiment presented in Fig.4a of the main text, we fabricated sharp diamond tips which were metal coated for in order to localize the NV in the scanning nanopillar through fluorescence quenching. Diamond tip fabrication was based on the nanofabrication techniques that we already employed for the production of the scanning diamond nanopillars presented in Fig.1. A type Ib diamond (Element six) was patterned with circular etch-masks (flowable oxide, FOx XR, Dow Corning) of 100 nm diameter. Here, in order to obtain sharp diamond tips instead of cylindrical diamond nanopillars, we modified the RIE etching recipe.
FIG. S4: Sharp diamond tip used for NV fluorescence quenching. Scanning electron microscopy image of a sharp diamond tip similar to the one used for the experiments presented in Fig.4a of the main text. Typical tip-radii are on the order of 10nm.

we had previously used: While we kept the (oxygen) etching chemistry identical to pillar fabrication, we significantly increased the etching time, such as to completely erode the etch mask on the diamond substrate. As a result, the etched diamond structures acquired the form of sharp tips as shown in the representative SEM image in Fig. S4. Typical tip-radii were in the range of 10 nm and tip lengths were on the order of 200 nm.

For the experiments described in the main text, we then coated the sharp diamond tips with a thin metallic layer using thermal metal evaporation. To avoid oxidation of the metal, we chose gold as the quenching metal and used a chrome adhesion layer between the gold and the diamond. For the tips employed in this work, we deposited 5 nm of gold and 5 nm of chrome.

S5. EXPLANATION OF TOPOGRAPHIC FEATURES IN FIG.4

The features observed in Fig.4a of the main text were governed by direct fluorescence quenching through metallic objects (as highlighted by the red square in the figure) and by a confluence of the distance-dependance of the NV fluorescence with topographic features on the sample (bright, ring-shaped feature in the figure). When approaching the NV to our metallic sample-surface, the total NV fluorescence collected in the far-field through the pillar changed as shown in the measurement in Fig. S5b. This well-known variation of NV fluorescence is a result of the variable electromagnetic density of states in the vicinity of metallic or dielectric interfaces, which influences the NV radiative lifetime as well as the
FIG. S5: **Explanation of topographic features in fluorescence quenching.** (a) AFM topography recorded during the experiment presented in Fig. 4 (same data as shown in Fig. 4a). (b) “Approach-curve” of the far-field NV fluorescence rate as the nanopillar with the NV center was approached to the sample. “z=0” was defined as the point of AFM contact (leftmost data-point) (c) Total fluorescence image reconstructed from the datasets in (a) and (b): Looking up the NV fluorescence intensity in (b) for every tip-sample displacement measured in (a) yields the reconstructed topographic features shown in the panel. We note that here, fluorescence quenching is solely induced by the sample surface, while the tip merely acts as a “spacer” between NV center and sample. Fluorescence-quenching that is directly induced by the tip (additional dark spot in d) is not reproduced here. (d) Original data (same data as Fig. 4a). The features common to (c) and (d) are attributed to effects of sample topography. The additional, dark feature in the center of (d) (red square in Fig. 4a) has no correspondence in topography and stems from direct fluorescence quenching of the NV center on the sharp metallic tip.

total effective laser excitation intensity impinging on the NV center. During our scanning experiments, the topography causes the mean distance between the scanning NV center in the nanopillar and the metallic substrate to vary, which in turn causes variations in the collected NV fluorescence rate. Assuming to first order that the metallic tip does not itself affect NV fluorescence (so long as it is not placed in direct contact to the NV center as in the “red-square region”), one can understand most features observed in Fig. 4a as a pure effect of topography. Based on this principle, in Fig. S5 we reconstruct the data in Fig. 4a from a measurement of sample topography (a) and an independently acquired fluorescence “approach-curve” (b), characteristic for fluorescence quenching of an emitter approaching a metallic surface\(^{41}\). The reconstructed image (Fig. S5c) was obtained by taking
the value of the AFM z-displacement for each point in the scan in Fig. S5a and looking up the corresponding fluorescence-rate obtained in the approach-curve. The resulting image shows striking similarity with the actually measured data (Fig. S5e; same data as Fig. 4a) and supports the validity of our explanation.

In stark contrast to these topography-induced changes in fluorescence, the sharp spot of reduced fluorescence at the location of the NV center (Fig. 4b in the main text), occurs in the absence of any corresponding features in the AFM image (see AFM image in Fig. S5a which was recorded simultaneously with the data in Fig. S5d). Besides this local feature, all measured changes of NV fluorescence in Fig. S5d could qualitatively be reproduced with our simple model shown in Fig. S5b. We therefore conclude, that the strong local reduction of NV fluorescence cannot be attributed to a topographic artifact, but instead is a result of the metallic tip being scanned in close vicinity of the NV center. We note that with the data at hand, an unambiguous differentiation between local fluorescence quenching (i.e. a reduction of radiative lifetime of the NV center induced by the metallic tip) and a mere “antenna-effect”, i.e. a modification of the excitation light intensity and NV emission pattern due to the presence of the metallic tip is not possible. However, within the scope of this paper such a differentiation is of limited importance as both effects would lead to the same conclusions with respect to localization of the NV center within the nano pillar.


