

Preparation and characterization of AFM tips with nitrogen-vacancy and nitrogen-vacancy-nitrogen color centers

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Abstract—We demonstrate a simple dip-coating method of covering standard AFM tips with nanodiamonds containing color centers. Such coating enables convenient visualization of AFM tips above transparent samples as well as tip using for performing spatially resolved magnetometry.

Atomic force microscopy (AFM) is a well-established field of research that offers multiple measurements including surface topology mapping, magnetic force measurements and electric conductance imaging with high-spatial resolution in various environments, like air, vacuum and/or liquids modalities [1–2]. Studies of nanomechanical properties of biological molecules, organella and living cells may be performed through local indentation by the AFM probe [3–4]. Measurements performed on optically transparent objects often require fluorescence imaging and the acquired AFM signal needs to be spatially correlated with the optical image [5].

In this work, we demonstrate a simple way of coating AFM tips and cantilevers with fluorescent nanodiamonds (NDs), which enables their localization using fluorescence microscopy techniques. Standard silicon AFM probes were dip-coated in a suspension of NDs in isopropyl alcohol and the NDs used contained two types of color centers: nitrogen-vacancy (NV) and nitrogen-vacancy-nitrogen (NVN, also designated as H3). They can be easily imaged under blue (for NVN centers) or green (for NV centers) light excitation with their fluorescence detected in the green and red wavelength bands, respectively. In the case of NV diamonds, additional measurement modalities are possible, e.g., magnetic field sensing [6]. While there have been many examples of scanning probe microscopy with diamond probes, including the use of single NV centers for nanoscale magnetometry [7–15], we focus here on the customization of more standard, silicon-based probes.

NV color centers are atom-like systems finding many practical applications in science and nanotechnology owing to their bright and stable fluorescence and spin properties, particularly when they are in their negative electronic charge state, NV⁻ [16–18]. The NV⁻ defect possesses a spin-triplet ground state which enables magnetic and electric field sensing using so-called, optically detected magnetic resonance (ODMR) [19], [20]. Green laser light is used for the spin polarization and

readout while a microwave (MW) field around 2.87 GHz is used for the spin manipulation. Small (~1 – 10 %) fluorescence dips are observed when the MW field is in resonance with the NV spin transition. External fields shift the resonance frequency which is monitored by the fluorescence change. The ODMR spectrum can be used to determine the magnitude and, in some arrangements, even the vector orientation of the perturbing field.

The spatial resolution of a magnetometer based on ND with an ensemble of NV centers, is a function of the size of the crystal used and its distance from a field source. Thus, to achieve good spatial resolution, placing NDs close to the tip apex is desired.

Our experiments were performed with commercially available silicon AFM probes (type PPP-CONTR, Nanosensors). The overall chip size is $3.4 \times 1.4 \times 0.3$ mm with the cantilever thickness, length, and width being 2.0 ± 0.1 μ m, 450 ± 10 μ m, and 50 ± 7.5 μ m, respectively. The tip height is within 10 – 15 μ m range. For dip-coating, the chip tip was carefully immersed for a few seconds into a suspension of nano-diamonds (MDNVN1 μ m powder with a 750 nm mean particle size, Adamas Nanotechnologies Inc.) in isopropyl alcohol and then allowed to dry in air. This type of ND powder contains on average ~2 ppm of NVN centers and estimated ~0.1 ppm of NV centers. Importantly, observations under the microscope indicated fluorescence of individual crystals being dominated by either the NV or NVN centers. Therefore, to achieve a satisfactory coating with less abundant NV diamonds, the dipping process was repeated, and two repetitions were found to be typically sufficient.

Fluorescence imaging of coated cantilevers and ODMR measurements have been performed with a wide-field microscope setup schematically presented in Fig. 1. Excitation of the NV color centers is ensured by a green (532 nm), PM-fiber-coupled laser (532-L-OE-115-1 W, Integrated Optics) with an output power of 60 mW. The laser beam after the fiber was expanded to ~9 mm diameter and collimated, and then focused at the back focal plane of the microscope objective (LUCPLFLN 40x, NA = 0.6, Olympus) with a help of a dichroic mirror (DMLP567, Thorlabs). Fluorescence light collected by the objective was filtered by a 600-nm long-pass filter (FEL0600, Thorlabs) and projected onto a CMOS camera

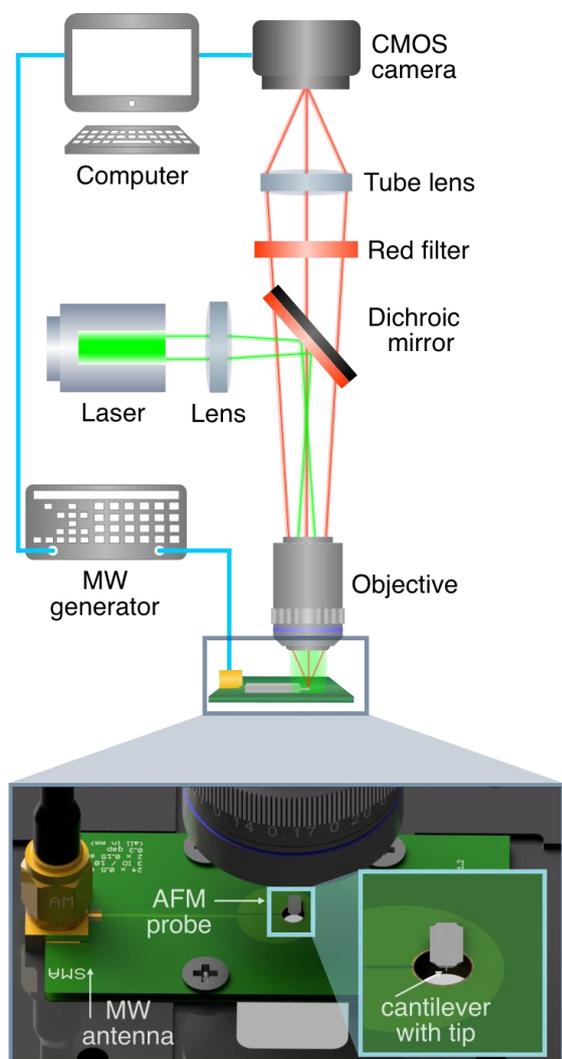


Fig. 1. Experimental apparatus for wide-field fluorescence microscopy and ODMR measurements. Inset shows the position of the AFM chip on the MW antenna.

sensor (IDS, UI-3240CP Rev. 2) using a 100-mm focal distance lens, which resulted in a field of view of $305 \times 244 \mu\text{m}$. For the ODMR measurements, the MW field was generated using a signal generator (SG396, Stanford Research Systems), amplified by a high-power amplifier (ZHL-16W-43+, Mini-Circuits) and delivered to a loop-gap type microstrip patch antenna [21] which has a 2-mm-diameter central hole size and ensures spatially uniform magnetic field over the whole cantilever area (see the inset in Fig. 1). An additional coil (not shown in Fig. 1) was used for applying a static magnetic field of up to 1 mT induction.

The fluorescence image of the ND-coated AFM cantilever is shown in Fig. 2. Under green light illumination, NDs containing NV centers are well visible as bright spots located randomly on the surface of the cantilever, which enables a convenient way of localizing the structure with a fluorescence microscope. Additionally,

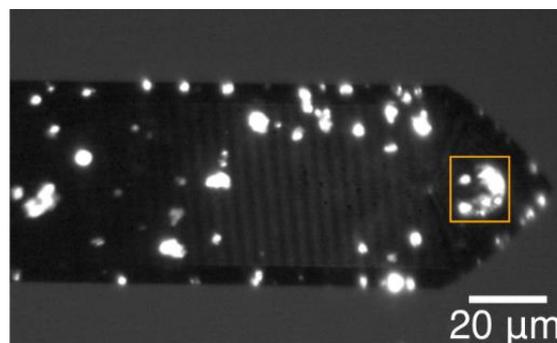


Fig. 2. Fluorescence image of the AFM cantilever coated with 750-nm-size nanodiamonds. The orange square marks the area from where the ODMR signal was collected.

there is a number of NDs visible around the tip area marked with the orange square in Fig. 2. The tip extends there from the cantilever plane towards the microscope objective and, due to the limited field depth of the optical setup, some NDs do not form a sharp image. However, this doesn't preclude ODMR measurements as long as fluorescence is integrated over the area containing a blurred image of all NDs. If the integration area is set too small, any drifts occurring during the MW frequency scans may cause fluorescence signal variations which can easily hinder the visibility of the ODMR signal.

Figure 3 shows the ODMR signals acquired for several magnetic field values with the fluorescence signal integrated over the region marked in Fig. 2. While in a zero magnetic field the measured nanodiamond ODMR spectrum resembles closely the single-crystalline one [16], the spectra recorded in non-zero fields are significantly different. Rather than exhibiting discrete pairs of resonances corresponding to four possible NV orientations along the [111] crystallographic direction in a bulk diamond, they are inhomogeneously broadened because of the random orientation of fluorescing NDs with respect to the direction of the magnetic field. Because of adding fluorescence from multiple randomly oriented NDs, the orientation of the magnetic field vector cannot be retrieved. However, the information on the field magnitude still can be extracted from the overall width of the spectrum [22], [23]. For the accurate readout of the field strength, this requires multiple, truly randomly oriented NDs contributing to the signal, but a less rigorous or qualitative magnetic field mapping is feasible with any number of NDs.

In this work, we have demonstrated a simple and efficient method of making the AFM chips visible under a fluorescence microscope by dip-coating them with fluorescent NDs. The method can be useful for the localization of the AFM tip position with respect to optically transparent but fluorescing samples. Moreover, we have successfully recorded ODMR spectra from a group of NDs located around the pyramidal tip of the

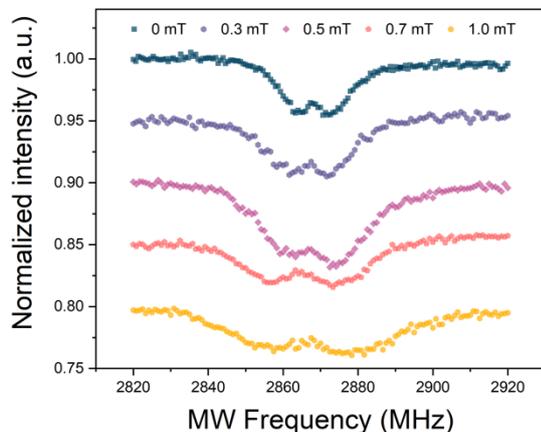


Fig. 3. ODMR signal for different value of magnetic fields. The visible broadening of a signal is caused by an increase in the applied magnetic field.

cantilever. This enables performing local magnetic field and temperature measurements with the same chip used for AFM measurements. The spatial resolution in the case of ND magnetometry is strongly dependent on the size of NDs and their positioning around the tip and may be improved by applying smaller diamond particles and limiting the fluorescence collection area to the tip apex. Since the NDs are addressed optically, the ultimate limit on spatial resolution is coming from the optical diffraction limit and is on the order of hundreds of nanometers. Despite offering significantly worse spatial resolution than the AFM, this modality may find practical applications for discerning nanostructures characterized by similar topography but different magnetic properties.

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