

Nanopositioning of a diamond nanocrystal containing a single nitrogen-vacancy defect center

T. van der Sar,¹ E. C. Heeres,² G. M. Dmochowski,¹ G. de Lange,¹ L. Robledo,¹
T. H. Oosterkamp,² and R. Hanson^{1,a)}

¹*Kavli Institute of Nanoscience, Delft University of Technology, P.O. Box 5046, 2600 GA Delft, The Netherlands*

²*Leiden Institute of Physics, Leiden University, Niels Bohrweg 2, 2333 CA Leiden, The Netherlands*

(Received 5 March 2009; accepted 28 March 2009; published online 28 April 2009)

Precise control over the position of a single quantum object is important for many experiments in quantum science and nanotechnology. We report on a technique for high-accuracy positioning of individual diamond nanocrystals. The positioning is done with a home-built nanomanipulator under real-time scanning electron imaging, yielding an accuracy of a few nanometers. This technique is applied to pick up, move, and position a single nitrogen-vacancy (NV) defect center contained in a diamond nanocrystal. We verify that the unique optical and spin properties of the NV center are conserved by the positioning process. © 2009 American Institute of Physics.

[DOI: [10.1063/1.3120558](https://doi.org/10.1063/1.3120558)]

The coupling of a single quantum object to degrees of freedom in its environment is a central theme in quantum science and engineering. Examples are the coupling of a single-photon source to an optical resonator^{1,2} or to a plasmonic waveguide,³ and the coupling of a single spin to surrounding spins.^{4,5} Studying and engineering such couplings is not only of fundamental interest, but may also lead to dramatic improvements in fluorescence detection efficiency,⁶ ultrasensitive magnetometry,^{7–11} and applications in quantum information processing.^{2,12,13} Controlled and precise positioning of the quantum object under study is essential for many of these experiments.

Examples of well-studied single-photon emitters in a solid-state environment are quantum dots, fluorescing dye molecules, and nitrogen-vacancy (NV) centers in diamond. In particular NV centers, which consist of a substitutional nitrogen atom next to an adjacent vacancy in the diamond lattice, are extremely stable sources of single photons.^{14,15} In addition, they have the unique property of a paramagnetic spin whose quantum state can be read out optically using fluorescence microscopy¹⁶ and be coherently controlled using magnetic resonance.¹⁷ Crucially, all these properties are retained under ambient conditions. Since NV centers can form in nanocrystals as small as 10 nm,¹⁸ their position can in principle be controlled with an accuracy of a few nanometers.

The potential of NV centers for quantum optical experiments is underlined by recent reports demonstrating coupling to confined optical modes of a microsphere^{19,20} and a micro-disk cavity.²¹ In these experiments, however, the positioning accuracy of the diamond nanocrystals was determined by the resolution of the optical setup (~500 nm), whereas sub-wavelength resolution is desired.

Here, we demonstrate a versatile technique to position a single NV center contained in a diamond nanocrystal to an arbitrary location. We locate and characterize diamond nanocrystals with single NV centers using a scanning confocal microscope. Subsequently, we use a home-built nanomanipu-

lator, consisting of a sharp probe mounted on a piezoelectrically controlled system inside a scanning electron microscope (SEM), for picking up and positioning the nanocrystal with nanometer precision. This technique is directly applicable to studies of the coupling of a single-photon emitter in a nanocrystal to various photonic and plasmonic structures and can facilitate experiments in magnetometry and quantum information processing.

First, we demonstrate our ability to position individual nanocrystals on an empty flat substrate with high spatial accuracy. Then we describe how we locate and identify nanocrystals containing a single NV center. Finally, we demonstrate explicitly through optical characterization and coherent spin manipulation that the unique properties of the NV center are preserved by the positioning process.

Positioning of nanocrystals is performed inside a FEI Nova NanoSEM using a home-built nanomanipulator located inside the SEM main chamber. The nanomanipulator consists of a coarse stage onto which a sample is mounted, and a fine stage holding a sharp tip that can be used to pick up nanometer-size objects [Fig. 1(a)].²² In contrast to manipulation of nano-objects with an atomic force microscope,^{23,24} this setup allows real-time imaging of the probe with nanometer resolution during the manipulation process.

In Figs. 1(b)–1(d) we demonstrate our ability to pick up, move, and accurately position individual diamond nanocrystals. The crystals (mean size ~100 nm) are dispersed onto an oxidized silicon substrate. To pick up a nanocrystal, it is first brought within 15 μm of the tip by moving the coarse stage. Subsequently, control is switched to the fine stage and the tip is translated and lowered until it touches the nanocrystal [Fig. 1(b)]. The crystal is lifted from the substrate by pushing against it with the tip until it sticks (presumably by van der Waals forces), and then retracting the tip [Fig. 1(c)]. At this point, the coarse stage can be moved to position the crystal in a new location on the same substrate or on another substrate within the 3 mm range of the stage. Alternatively, substrates can be exchanged while the crystal remains attached to the tip.

^{a)}Electronic mail: r.hanson@tudelft.nl.

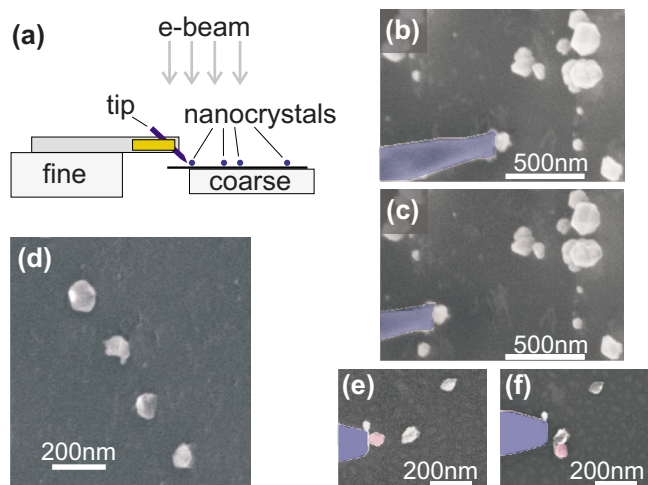


FIG. 1. (Color online) (a) Schematic of the nanomanipulator. The coarse stage can be moved in three dimensions by Attocube piezo steppers (range of 3 mm, smallest step size of 25 nm). A sharp, chemically etched tungsten tip is mounted on the piezo fine stage (range of 15 μm , smallest step size of subnanometer) which can also be moved in three dimensions. Both stages are controlled by an analog joystick, allowing for accurate adjustment of their positions. [(b) and (c)] SEM images showing a nanocrystal being attached to the tip (shaded blue) (b) and subsequently being lifted from the substrate by retraction of the tip (c). (d) SEM image of four diamond nanocrystals that were subsequently picked up and positioned to form a line. [(e) and (f)] SEM images showing a nanocrystal (shaded pink) first attached to the tip (e), then being positioned onto another nanocrystal (f).

The nanocrystal is positioned by following the reverse process. The tip is lowered until the crystal touches the substrate. Then, the tip is moved along the surface, whereby the crystal is reoriented and its contact areas with the substrate and tip are modified until it sticks to the substrate. While the initial placement has a precision of ~ 20 nm [Fig. 1(d)], the positioning can be optimized further by pushing the crystal with the tip, which is only limited by the SEM imaging resolution (a few nanometer). Alternatively, we can position a crystal such that it is touching an on-chip structure such as a nanowire, or another diamond nanocrystal, as is shown in Figs. 1(e) and 1(f).

Next, we demonstrate the positioning of a single NV center. Diamond nanocrystals (~ 10 – 100 nm in size) are dispersed onto a glass cover slip that has been coated with a thin (~ 3 nm) layer of Cr (to allow for SEM imaging, while retaining high optical transparency) and which contains gold markers (defined by e-beam lithography) for position reference.

NV centers are imaged by fluorescence microscopy using a home-built scanning confocal microscope.¹⁶ All data is taken at room temperature. In Fig. 2(a), a photoluminescence (PL) image of a $30 \times 45 \mu\text{m}^2$ area of the sample is shown. The fluorescing spot in the lower-left corner is verified to be a single emitter by time correlation measurements on single-photon detection events,¹⁴ see Fig. 2(b). The subsequent observation of the characteristic zero-phonon line (ZPL) around 637 nm [Fig. 2(c)] shows that this emitter is an NV center.¹⁶

In the SEM, the nanocrystal containing the single NV center is identified using the reference markers [Fig. 2(d)] and subsequently picked up and positioned on the other side of the marker. Although this area contains other nanocrystals, we know from the optical characterization [inset of Fig. 2(a)] that they do not contain fluorescing defects. Also, the energy

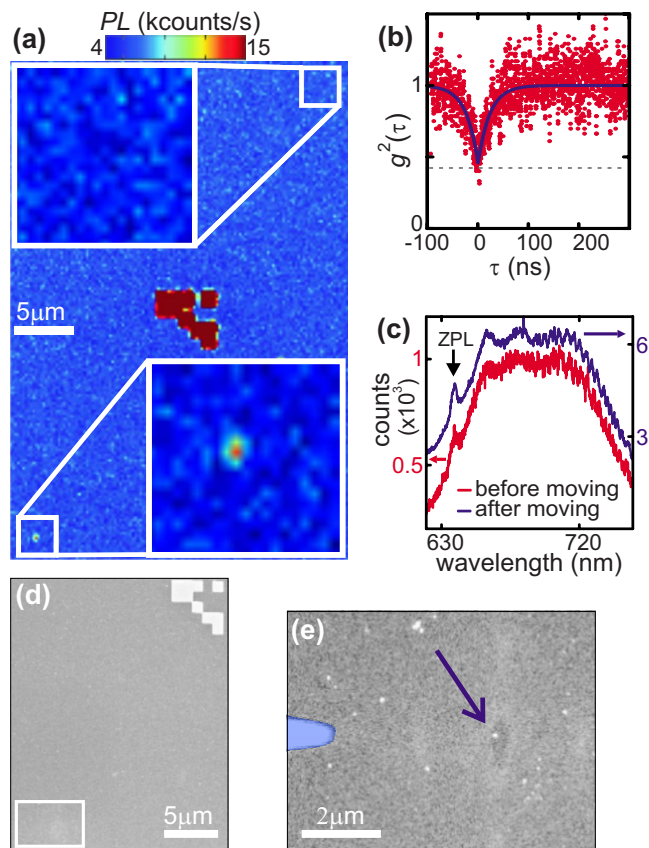


FIG. 2. (Color online) Identifying and positioning a single NV center. (a) PL image of part of the sample around a gold marker. The bright spot in the lower left corner is verified to be a single NV center in (b) and (c). (b) Autocorrelation function $g^{(2)}(\tau)$ of photon detection events, showing an antibunching dip below 0.5 (Ref. 14). The solid line is a fit based on a three-level model (Ref. 14). The dotted line indicates the independently measured contribution of background fluorescence. (c) PL spectrum of the NV center showing the characteristic ZPL around 637 nm (Ref. 16). Red (blue) data is taken before (after) positioning (Ref. 25). (d) SEM image of the same area as in (a). (e) Zoom in on the white box in (d), with the arrow pointing to the nanocrystal containing the NV center. The faint white spots next to it are markings made by focusing the laser onto a spot for 2 min at a power of 10 mW to facilitate identification of the NV-containing nanocrystal. The contrast difference is presumably due to local oxidation or evaporation of the Cr film. The tip is visible on the left.

of the imaging electrons (10 keV) is far too low to displace carbon atoms²⁶ and thus no additional NV centers are formed.

After the nanopositioning, a spatial PL map of the same area as in Fig. 2(a) is made [Fig. 3(a)]. Initially observed fluorescence in the SEM-imaged areas (likely from SEM-deposited carbon) rapidly bleached under laser illumination (500 μW) to a low background value on the time scale of a minute. To ensure that the NV center was not damaged by the positioning, we repeat the optical characterization. The observation of photon antibunching [Fig. 3(b)] and the ZPL in the spectrum [Fig. 2(c)] confirm that the nanocrystal still contains one NV center.

To explicitly show that we have positioned a single spin, we demonstrate coherent control of the quantum state of the positioned spin. Optically detected magnetic resonance (ODMR) reveals the characteristic zero-field splitting of 2.87 GHz of the NV center¹⁶ [Fig. 3(c)]. By using short microwave pulses that are resonant with the spin transition, we observe coherent spin rotations (Rabi nutations) of this single NV center^{12,17} [Fig. 3(d)].

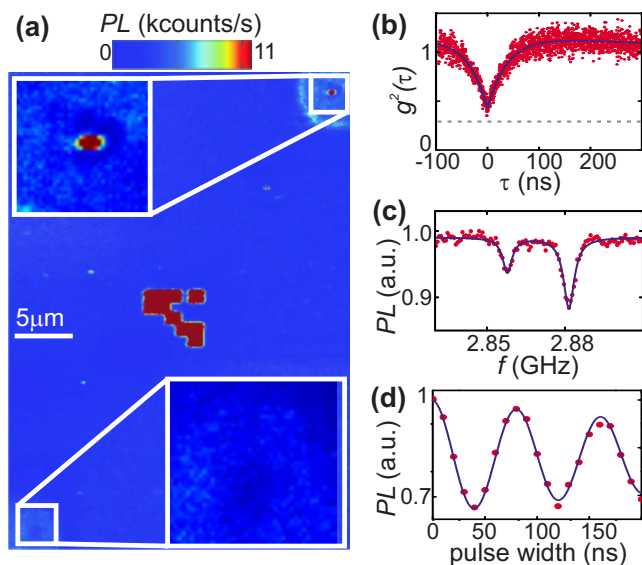


FIG. 3. (Color online) Optical and spin characterization of a single NV center after positioning. (a) PL image of the same area as in Fig. 2(a) (Ref. 25). The NV-containing crystal has been moved from the lower left to the upper right corner. (b) Measurement of $g^2(\tau)$ as in Fig. 2(b), showing an antibunching dip below 0.5. (c) ODMR and (d) coherent oscillations of the NV center electron spin. The ODMR splitting is due to a stray magnetic field (~ 5 G). Microwaves are applied through a $30 \mu\text{m}$ diameter wire that is located in close proximity to the NV center.

In summary, we have demonstrated a technique to pick up, move, and position individual diamond nanocrystals with few-nanometer precision, all under real-time SEM imaging. This technique has been used to position a single NV center contained in a diamond nanocrystal. We have demonstrated that the unique spin and optical properties of the NV center electron spin are preserved in the positioning process. This technique is directly applicable to controlled studies of the coupling of a single NV center to photonic and plasmonic structures.

We thank C. Degen, G. Fuchs, M. Hesselberth, F. Jelezko, T. Schenkel, and D. van der Zalm for discussions and experimental help. This work is supported by the “Stichting voor Fundamenteel Onderzoek der Materie (FOM)” and the “Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO),” and the DARPA QuEST programme. T.H.O.

acknowledges support from Technologiestichting STW and from an ERC Starting Grant.

- ¹K. J. Vahala, *Nature (London)* **424**, 839 (2003).
- ²H. J. Kimble, *Nature (London)* **453**, 1023 (2008).
- ³A. V. Akimov, A. Mukherjee, C. L. Yu, D. E. Chang, A. S. Zibrov, P. R. Hemmer, H. Park, and M. D. Lukin, *Nature (London)* **450**, 402 (2007).
- ⁴L. Childress, M. V. Gurudev Dutt, J. M. Taylor, A. S. Zibrov, F. Jelezko, J. Wrachtrup, P. R. Hemmer, and M. D. Lukin, *Science* **314**, 281 (2006).
- ⁵R. Hanson, V. V. Dobrovitski, A. E. Feiguin, O. Gywat, and D. D. Awschalom, *Science* **320**, 352 (2008).
- ⁶D. E. Chang, A. S. Sørensen, P. R. Hemmer, and M. D. Lukin, *Phys. Rev. B* **76**, 035420 (2007).
- ⁷B. M. Chernobrod and G. P. Berman, *J. Appl. Phys.* **97**, 014903 (2005).
- ⁸C. L. Degen, *Appl. Phys. Lett.* **92**, 243111 (2008).
- ⁹J. M. Taylor, P. Cappellaro, L. Childress, L. Jiang, D. Budker, P. R. Hemmer, A. Yacoby, R. Walsworth, and M. D. Lukin, *Nat. Phys.* **4**, 810 (2008).
- ¹⁰J. R. Maze, P. L. Stanwix, J. S. Hodges, S. Hong, J. M. Taylor, P. Cappellaro, L. Jiang, M. V. Gurudev Dutt, E. Togan, A. S. Zibrov, A. Yacoby, R. L. Walsworth, and M. D. Lukin, *Nature (London)* **455**, 644 (2008).
- ¹¹G. Balasubramanian, I. Y. Chan, R. Kolesov, M. Al-Hmoud, J. Tisler, C. Shin, C. Kim, A. Wojcik, P. R. Hemmer, A. Krueger, T. Hanke, A. Leitenstorfer, R. Bratschitsch, F. Jelezko, and J. Wrachtrup, *Nature (London)* **455**, 648 (2008).
- ¹²R. Hanson and D. D. Awschalom, *Nature (London)* **453**, 1043 (2008).
- ¹³P. Neumann, N. Mizuochi, F. Rempp, P. Hemmer, H. Watanabe, S. Yamasaki, V. Jacques, T. Gaebel, F. Jelezko, and J. Wrachtrup, *Science* **320**, 1326 (2008).
- ¹⁴C. Kurtsiefer, S. Mayer, P. Zarda, and H. Weinfurter, *Phys. Rev. Lett.* **85**, 290 (2000).
- ¹⁵A. Beveratos, R. Brouri, T. Gacoin, J.-P. Poizat, and P. Grangier, *Phys. Rev. A* **64**, 061802 (2001).
- ¹⁶A. Gruber, A. Drabenstedt, C. Tietz, L. Fleury, J. Wrachtrup, and C. von Borzyskowski, *Science* **276**, 1212 (1997).
- ¹⁷F. Jelezko, T. Gaebel, I. Popa, A. Gruber, and J. Wrachtrup, *Phys. Rev. Lett.* **92**, 076401 (2004).
- ¹⁸M. Boersch, R. Reuter, G. Balasubramanian, R. Erdmann, F. Jelezko, and J. Wrachtrup, arXiv:0902.0389v1.
- ¹⁹Y.-S. Park, A. K. Cook, and H. Wang, *Nano Lett.* **6**, 2075 (2006).
- ²⁰S. Schietinger, T. Schroder, and O. Benson, *Nano Lett.* **8**, 3911 (2008).
- ²¹P. E. Barclay, O. Painter, T. J. Watson, Sr., C. Santori, K.-M. Fu, and R. G. Beausoleil, arXiv:0812.4505v2.
- ²²A. J. Katan and T. H. Oosterkamp, *J. Phys. Chem. C* **112**, 9769 (2008).
- ²³Y. Wang, Y. Zhang, B. Li, J. Lu, and J. Hu, *Appl. Phys. Lett.* **90**, 133102 (2007).
- ²⁴M. Barth, J. Sting, J. Kouba, N. Nüsse, B. Lochel, and O. Benson, *Phys. Status Solidi B* **246**, 298 (2009).
- ²⁵Note that optical measurements before and after positioning were done with a different microscope objective and therefore do not allow direct quantitative comparison.
- ²⁶J. Koike, D. M. Parkin, and T. E. Mitchell, *Appl. Phys. Lett.* **60**, 1450 (1992).