

Supporting Information

Single Silicon Vacancy Centers in 10-Nanometer Diamonds for Quantum Information Applications

Stepan V. Bolshedvorskii^{§,†,#}, Anton I. Zeleneev^{†,‡}, Vadim V. Vorobyov^{§,¶}, Vladimir V. Soshenko^{§,#}, Olga R. Rubinas^{§,†,#}, Leonid A. Zhulikov^{†,‡}, Pavel A. Pivovarov^{}, Vadim N. Sorokin^{§,‡}, Andrey N. Smolyaninov[#], Liudmila F. Kulikova[⊥], Anastasiia S. Garanina^{||}, Sergey G. Lyapin[⊥], Viatcheslav N. Agafonov^{||}, Rustem E. Uzbekov[†], Valery A. Davydov[⊥] and Alexey V. Akimov^{%,§,‡,*}*

[§]P.N. Lebedev Physical Institute, 53 Leninskij Prospekt, Moscow, 119991 Russia

[†]Moscow Institute of Physics and Technology, 9 Institutskiy per., Dolgoprudny, Moscow Region, 141700, Russia

[#]Photonic Nano-Meta Technologies, Moscow, The territory of Skolkovo Innovation Center, Str. Nobel b.7, 143026 Russia

[‡]Russian Quantum Center, 100 Novaya St., Skolkovo, Moscow, 143025, Russia

[¶]Institute of Physics, University of Stuttgart and Institute for Quantum Science and Technology IQST, 70569, Pfaffenwaldring 57, Stuttgart, Germany

^{*}Prokhorov General Physics Institute of Russian Academy of Sciences, 38 Vavilov str., Moscow, 119991, Russia

[⊥]L.F. Vereshchagin Institute for High Pressure Physics, Russian Academy of Sciences, Troitsk, Moscow, 108840, Russia

^{||}GREMAN, UMR CNRS CEA 6157, F. Rabelais University, 37200 Tours, France

[†]Faculty of Medicine, F. Rabelais University, 37032 Tours, France

[%]Texas A & M University, College Station, TX 77843, USA

*aa@rqc.ru

Calculation of probability find SiV center in single nanodiamond

To determine the probability of finding SiV center in single nanodiamond crystal we analyze 3 AFM maps. We find 24 nanodiamonds containing SiV center including 7 nanodiamonds with

single SiV center. Besides, there are 1625 aggregates of nanodiamonds and single nanodiamonds in these 3 AFM maps. Then, we compile histogram of height for these particles. Thus, we can measure the number of single nanodiamonds in all 3 maps. From TEM images we can say that median size of single nanodiamonds is 7.5 nm. Therefore, the number of single nanodiamonds can be calculated from this following formula: $N_{sum} = \sum_i \left(\frac{H_i}{H_{median}} \right)^3 \cdot N_i \approx 4480$, where H_i is a height in bin of the histogram, N_i is a number of particles in this bin in the histogram, $H_{median} = 7.5$ is the median size of single nanodiamond. Using this number, we can calculate probability to find at least one SiV center in single nanodiamond as $24 / 4480 \approx 0.5\%$, whereas the probability to find single SiV center in single nanodiamonds is $7 / 4480 \approx 0.15\%$. Thereby, probability to find nanodiamond with single SiV is approximately 30% among nanodiamonds with SiV color centers.

Size measurements from AFM maps

To make the statistical data more sufficient we analyze the additional 3 AFM maps (see Fig S1a) containing 1019 nanodiamonds or small aggregates of nanodiamonds. The histogram of size distribution depicted on the Figure S1b.

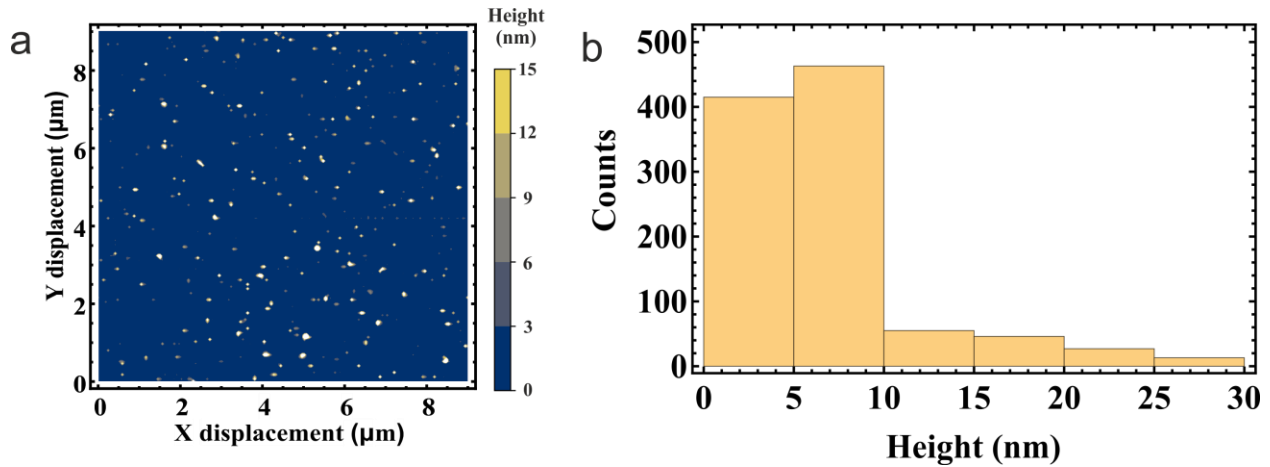


Figure S1.(a) Typical AFM map an the sample (b)The size distribution histogram obtained from the AFM particle analysis.

The median size is 5.5 nm with the standard deviation 5.5 nm. Including error of estimation, the data are in a good agreement with the TEM measurements (see Fig 3b in the paper).

Raman measurements and preliminary characterization of grown diamond material

For preliminary characterization of full sample diamond material obtained during growing process, we use Raman spectra measurements. Raman spectra were measured on a Princeton Instruments TriVista 555 triple spectrometer with the use of the 50x objective of an Olympus BX51 microscope and the 488 nm Ar⁺ laser line as the excitation source. The laser power incident on the sample was 30 mW.

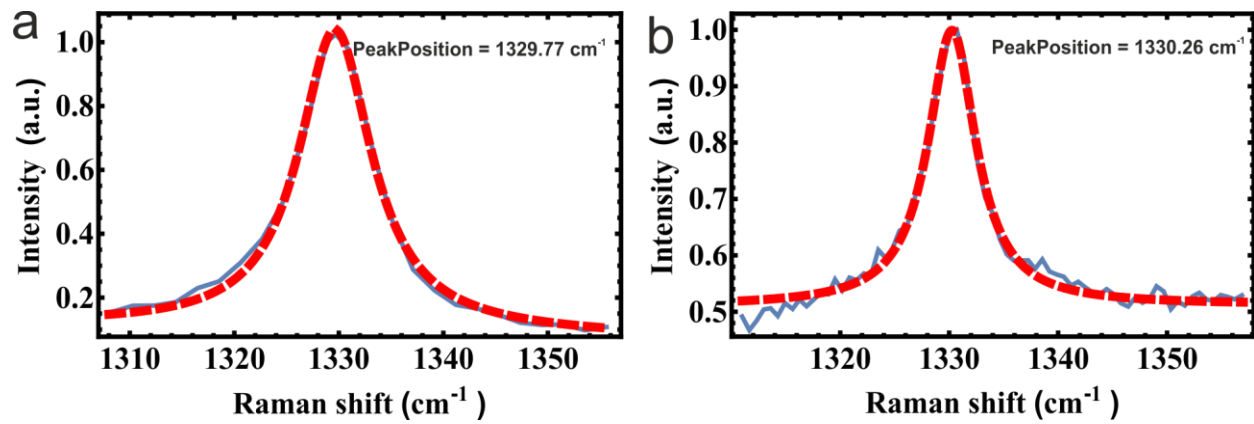


Figure S2. (a) Diamond Raman peak for nanodiamonds right after growing with Lorentzian fit. (b) Diamond Raman peak for nanodiamonds after cleaning and centrifugation with Lorentzian fit.

Figure S2a shows Raman spectra for a central part of pallet with full sample diamond material right after growing. We use the Lorentzian fit to find the central position of the diamond Raman line and observed it at 1330.26 cm⁻¹.

Using the relation between the pressure corresponding for compression-expansion, P , and the change in diamond Raman frequency ($\Delta\nu = \nu - 1331.8$ cm⁻¹, where ν - peak position for a diamond Raman line)¹ we can calculate the strain level following way²:

$$P = 0.34 \frac{\text{GPa}}{\text{cm}^{-1}} \Delta\nu = 0.34 \frac{\text{GPa}}{\text{cm}^{-1}} \times (1330.26 - 1331.8) \text{ cm}^{-1} = -0.524 \text{ GPa}. \quad (1)$$

We performed the Raman measurements also for a nanodiamonds after cleaning procedure and centrifugation (see Figure S2b). Lorentzian fit of the diamond peak position for ultra-small nanodiamonds is 1329.77 cm^{-1} .

Using the similar to (3) relation of the diamond peak position we can calculate the strain level for ultra-small nanodiamonds:

$$P = 0.34 \frac{\text{GPa}}{\text{cm}^{-1}} \Delta \nu = 0.34 \frac{\text{GPa}}{\text{cm}^{-1}} \times (1329.77 - 1331.8) \text{ cm}^{-1} = -0.69 \text{ GPa}. \quad (2)$$

The sign of the pressure stands for expansion nature of the stress, which quite standard for nanodiamonds. Besides, the small downshift of the Raman frequency may be caused by an asymmetric broadening due to the phonon confinement effect³ thus pressure estimate (1) and (2) is upper limit on possible strain.

The Raman measurements shows the mostly sp^3 hybridization bonding for carbon atoms in the grown diamond material and small strain level lower than 0.7 GPa even for ultra-small nanodiamonds fraction.

Strain assessment by the ZPL position analysis

To make an assessment on the strain level in nanodiamonds containing optical active SiV color centers we took into accounts the ZPL position statistics obtained during the investigation. The standard deviation of the ZPL position in our measurements (see Figure 6a) is $\sigma_{ZPL} = 0.27 \text{ nm}$.

To analyze the strain level we use the ZPL position dependence on strain⁴:

$$E(P) = E_0 + \alpha P + \beta P^2. \quad (3)$$

We took into account only the linear coefficient: $\alpha = 0.48 \frac{\text{nm}}{\text{GPa}}$ due to the fact that $\alpha \gg \beta$, than

we can calculate the strain level in nanodiamonds as following:

$$Strain = \frac{\sigma_{ZPL}}{\alpha} = \frac{0.27 \text{ nm}}{0.48 \frac{\text{nm}}{\text{GPa}}} = 0.562 \text{ GPa}. \quad (4)$$

Because the standard deviation cannot take negative values, we cannot make any statements about the sign of the stress from these measurements. Nevertheless, the module of the strain level from the diamond Raman frequency position line analyses and the SiV ZPL position analyses deviates no more than 25%. Thus we can conclude that the strain level in produced nanodiamonds less than 0.7 GPa.

APD jitter measurements and deconvolution procedure

For the deconvolution process, we first measured the jitter of our setup by focusing on the pulsed laser (5 MHz repetition rate, pulse duration is 70 ps) on the golden mask and measured $g_2(\tau)$ (Figure 5a inset) of reflected green light. We then used Gaussian response model⁵ to fit the data:

$$f(t) = offset + amp \times \frac{e^{-\frac{(t-t_1)^2}{2t_j^2}}}{\sqrt{2\pi t_j^2}}, \quad (5)$$

where t_j – jitter of the our HBT interferometer. From the fit, we determined that the jitter of our setup was 672 ps. For the deconvolution process, we used the convolution of jitter function and $g_2(\tau)$ function with the following formula:

$$f(t) = (offset - amp \times e^{-\gamma(t-t_1)}) \times \frac{e^{-\frac{(t-t_1)^2}{2t_j^2}}}{\sqrt{2\pi t_j^2}} \quad (6)$$

After fitting the raw data, we can estimate that what is not affected is the detector's jitter $g_2(0)$ depth.

REFERENCES

- (1) Liscia, E. J. Di; Álvarez, F.; Burgos, E.; Halac, E. B.; Huck, H.; Reinoso, M. Stress Analysis on Single-Crystal Diamonds by Raman Spectroscopy 3D Mapping. *Mater. Sci. Appl.* **2013**, *4*, 191–197. <https://doi.org/10.4236/msa.2013.43023>.
- (2) Stehlik, S.; Varga, M.; Ledinsky, M.; Jirasek, V.; Artemenko, A.; Kozak, H.; Ondic, L.; Skakalova, V.; Argentero, G.; Pennycook, T.; Meyer, J. C.; Fejfar, A.; Kromka, A.; Rezek,

- B. Size and Purity Control of HPHT Nanodiamonds down to 1 Nm. *J. Phys. Chem. C* **2015**, *119* (49), 27708–27720. <https://doi.org/10.1021/acs.jpcc.5b05259>.
- (3) Yoshikawa, M.; Mori, Y.; Maegawa, M.; Katagiri, G.; Ishida, H.; Ishitani, A. Raman Scattering from Diamond Particles. *Appl. Phys. Lett.* **1993**, *62* (24), 3144–3116. <https://doi.org/10.1063/1.109154>.
- (4) Lyapin, S. G.; Ilichev, I. D.; Novikov, A. P.; Davydov, V. A.; Agafonov, V. N. Study of Optical Properties of the NV and SiV Centres in Diamond at High Pressures. *Nanosyst. Physics, Chem. Math.* **2018**, *9* (1), 55–57. <https://doi.org/10.17586/2220-8054-2018-9-1-55-57>.
- (5) Vlasov, I. I.; Shiryaev, A. A.; Rendler, T.; Steinert, S.; Lee, S.-Y.; Antonov, D.; Vörös, M.; Jelezko, F.; Fisenko, A. V.; Semjonova, L. F.; Biskupek, J.; Kaiser, U.; Lebedev, O. I.; Sildos, I.; Hemmer, P. R.; Konov, V. I.; Gali, A.; Wrachtrup, J. Molecular-Sized Fluorescent Nanodiamonds. *Nat. Nanotechnol.* **2014**, *9* (1), 54–58. <https://doi.org/10.1038/nnano.2013.255>.