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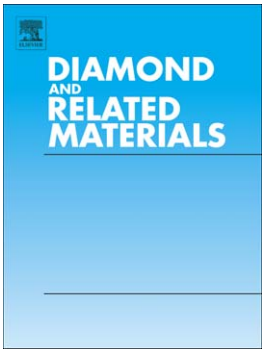

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PII: S0925-9635(16)30011-5
DOI: doi: 10.1016/j.diamond.2016.01.011
Reference: DIAMAT 6553

To appear in: *Diamond & Related Materials*

Received date: 4 December 2015
Revised date: 15 January 2016
Accepted date: 15 January 2016

Please cite this article as: Nina Felgen, Boris Naydenov, Stuart Turner, Fedor Jelezko, Johann Peter Reithmaier, Cyril Popov, Incorporation and study of SiV centers in diamond nanopillars, *Diamond & Related Materials* (2016), doi: 10.1016/j.diamond.2016.01.011

Incorporation and study of SiV centers in diamond nanopillars

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Abstract

We report on the incorporation of SiV centers during hot filament chemical vapor deposition of diamond on top of diamond nanopillars with diameters down to 100 nm. The nanopillars themselves were prepared from nanocrystalline diamond films by applying electron beam lithography and inductively coupled plasma reactive ion etching. The optical investigations revealed the presence of ensembles of SiV color centers incorporated during the overgrowth step.

Keywords: Nanocrystalline diamond; Nanopillars; SiV color centers

1. Introduction

Recently, there has been growing interest in diamond as a platform for quantum information technology (QIT) components and devices like, quantum memories, quantum repeaters [1] or single photon sources [2,3] which is due to the outstanding properties of different color centers in its crystal lattice. Some of these color centers have been identified as bright and stable single photon emitters, e.g. the nitrogen-vacancy centers (NV centers) [4,5], nickel-related NE8 complexes [6], chromium-related centers [7] and silicon-vacancy centers (SiV centers) [8,9]. Some of these solid-state defects are already applied in different fields, like, quantum optics [10] or biological sensing [11,12], but the intensive research on their basic and application relevant properties is still ongoing. Among these defects the NV color center is well studied in synthetic diamond because of its unique spin properties. It possesses broad emission spectra from 600 to 800 nm and is a bright and stable single photon emitter [13]. The disadvantages of these centers are the phonon-broadened emission spectra, where only 4 % are emitted into the zero phonon line (ZPL) [14] and its long lifetime up to 12 ns in bulk diamond [13] which limits the excitation and emission rates [15].

Due to their advantageous properties in comparison to the NV centers, SiV centers have gained a lot of attention recently. In a SiV center a silicon atom sits in the center of a divacancy (Fig. 1.). It does not couple strongly to the phonons of the diamond lattice because of the same symmetry of ground and excited state [16,17]. This results in a linearly polarized emission spectra [18] with a sharp ZPL around 739 nm with a FWHM of 5 nm [19] and a weak phonon side band [20]. At room temperature 70% of the emission is in the ZPL [18], subsequently a photon emission rate of about 6 Mcounts/s for SiV can be reached [18] The SiV complex is stable up to 1350°C [21], but there are some reports of bleaching if high laser power is used for excitation [22]. Additionally to that it has been shown that SiV centers can be observed and are stable in small isolated diamonds down to a size of 2 nm [23].

SiV centers can be created in different ways: during the diamond growth by hydrogen etching (with H atoms stemming from the excited by a microwave plasma or hot filaments precursor gases) of the silicon substrate until the diamond grains coalescence in a closed film or of a piece Si, SiO_2 or SiC placed near the substrate [24,25], by introduction of Si-containing gases into the reaction chamber [26,27] or by implantation of Si (focused ion beam or ion implantation) in diamond films or afterwards in the fabricated structures [18,28]. Coupling of these color centers to optical resonators or cavities, for example, microdiscs [29], pillars or photonic crystals [14,30] enables the control of the emission properties, like, the photon emission yield or the possibility to guide photons through a photonic network. Such cavities and nanostructures can be created not only using top-down approaches as in the present work, but also by bottom-up techniques, like selective diamond seeding on the substrate [31], growth through a patterned SiO_2 mask [32], using an alumina template [33] or scanning probe “dip-pen” lithography [25].

The aim of the present work is the incorporation of SiV centers in diamond nanopillars of different diameters and their optical characterization. The approach here is based on the top-down technique for fabrication of nanopillars described in our previous work [34]

followed by a CVD overgrowth step to incorporate silicon atoms etched from the Si substrate into the existing nanocrystalline (NCD) pillars. Due to the fact that the pillars are freestanding on the substrate, the etching of the latter by hydrogen from the precursor mixture is possible [35]. The SiV color centers will be embedded only in the additionally grown diamond, i.e. very close to the surface. Their optical properties are investigated with confocal microscopy, fluorescence microscopy and fluorescence auto-correlation measurements.

2. Experimental

2.1. Fabrication of NCD pillars with incorporated SiV centers

Initially, nanocrystalline diamond films were grown by hot filament chemical vapor deposition (HFCVD) in a self-designed set-up as described in detail in previous publications [36]. 3-inch (100) silicon wafers were used as substrates after ultrasonic pretreatment in a suspension of diamond powders with different crystallite sizes in *n*-pentane to achieve a nucleation density of about 1×10^{10} cm^{-2} [37]. The precursor gas mixture included 1% CH_4 in H_2 with a total gas flow of 505 sccm. The working pressure of 25 mbar and the substrate temperature of 880°C were kept constant during the deposition time of 180 min.

The obtained NCD films with thicknesses of about 380 nm were used for the fabrication of nanopillars. Like in our previous publications [34,38], also in this work arrays of pillars with diameters of 1 µm down to 100 nm were prepared and investigated. The first step was to spin-coat a positive resist (AR-P 617.06) on the top of NCD films, followed by the definition of the nanopillars using electron beam lithography (EBL, Raith eLine). Arrays of round structures with varying diameters and distances of 5 µm were written in the resist which was subsequently removed from the exposed areas with methyl isobutyl ketone (MIBK)/ isopropanol (IPA) = 1 /3 for 165 s, followed by cleaning in IPA for 30 s. As a hard etch mask 5 nm Ti (as adhesion layer) and 200 nm Au were deposited by electron beam evaporation (Balzers BAK 600). A lift-off process was performed by removing the remaining

resist together with the gold film on top of it immersing the samples in 1-methyl-2-pyrrolidone (NMP) at 80°C, leaving the hard mask for NCD nanostructuring. An inductively coupled plasma reactive ion etching system (ICP-RIE, Oxford PlasmaLab 100) was used to transfer the mask pattern into the diamond films by a RIE process with oxygen as a reactive gas. The ICP-RIE etch recipe included 1000 W ICP power, 200 W RF power, O_2 flow of 10 sccm and a pressure of 5 mTorr (0.7 Pa) [38]. The substrate temperature was kept at 30°C using a temperature controlled table and He-backing. The endpoint was determined by a laser interferometer (675 nm wavelength). After the etching process the mask was removed with aqua regia ($HCl:HNO_3$ 3:1) and NH_4F/HF treatments each for 2 min.

The sample with the structured diamond pillars on silicon was loaded into the HFCVD set-up again and overgrown for 30 min using the same process parameters as for the film growth. No additional pretreatment was applied, the diamond was deposited only on the nanopillars but not on the bare silicon substrate. This allowed etching of the substrate by atomic hydrogen from the gas phase [39,40] and incorporation of Si atoms into the growing diamond.

The process of fabrication of NCD pillars with integrated SiV centers is schematically shown in Fig. 2.

The morphology of the NCD films, as well as the quality and size of the structured nanopillars were investigated with scanning electron microscopy (SEM, Hitachi S-4100). Most of the images were taken with a tilting angle of 45°, however, to determine the real dimensions the samples were also tilted to different angles.

The microstructure of the NCD nanopillars prior the overgrowth was studied by annual dark-field scanning transmission electron microscopy (ADF-STEM) in combination with electron energy loss spectroscopy (EELS). To do so, a single nanopillar was cut out in cross-section using focused ion beam (FIB) lamella cutting in a FIB Helios FIB-SEM instrument. The electron microscopy experiments were performed in a Titan "cubed" 80-300

aberration corrected microscope, operated at 300 kV and equipped with a GIF Quantum spectrometer. The convergence semi-angle α used for imaging was 22 mrad, the ADF inner acceptance angle β was 25 mrad. The EELS acceptance angle used was 28 mrad.

2.2. Optical characterization of NCD pillars with incorporated SiV centers

The fluorescence of the pillars was characterized with a home-built confocal microscope using an air objective (Olympus UPLAN FL 100x-0.95). For excitation we used a frequency-doubled Nd:YAG laser (Laser Quantum GEM) with a power of 400 μW. The set up possesses two single photon counters (avalanche photo-diodes Excelitas SPCM-AQRH-14) allowing to perform second order auto-correlation measurements. The fluorescence spectra were recorded using a spectrometer (Princeton Instruments SP300i).

3. Results and discussion

3.1. Morphology and crystallinity of the NCD pillars

Arrays of NCD pillars with nominal diameters of 1000 nm, 500 nm, 200 nm, and 100 nm were produced as shown in Fig. 3. The silicon substrate between the pillars was very clean, without any diamond remnants. The nanopillars themselves were well shaped and tapered with a top diameter equal to the nominal one. This effect increased with decreasing the nominal diameter of the pillars and was most pronounced by the pillars with diameters of 100 and 200 nm (Fig. 4).

The ADF-STEM image of a single NCD pillar presented in Figure 4 shows clearly that the tapered single pillar is made up of densely-packed NCD crystals. High resolution data presented in Figure 5 reveals the excellent quality of the diamond film grains. Furthermore, an EEL spectrum taken from a single diamond grain shows the typical C-K edge features that correspond to sp^3 bonded carbon (σ*-contribution), together with a small π* contribution at 285 eV, related to sp^2 bonded carbon. This signal most likely has its origin in amorphous

carbon at grain boundaries in the material, and amorphous carbon at the sample surface as a result of sample preparation.

The NCD pillars were overgrown for 30 min, corresponding to ca. 50 nm, and examples from the resulting structures are shown in Fig. 6. It can be seen that no diamond deposition occurred on the Si surface between pillars but only on the pillars. The tapering of the pillars was decreased due to the additional coating mainly on the top. Furthermore, larger crystallites can be observed in the SEM images

3.2. Optical characterization of the SiV centers incorporated in the nanopillars

Typical confocal images of nanopillars with two different diameters are shown in Figure 7. The bright spots correspond to the individual nanopillars in the arrays. The observed fluorescence varied from 3 Mcounts/s down to about 1 Mcounts/s depending on the nanopillar diameter. The fluorescence was collected after a 640 nm long pass filter in order to remove the laser light.

Room temperature photoluminescence (PL) spectra were taken from selected pillars before the overgrowth (Fig. 8 (a)). The broad spectral line observed can be attributed to the nanocrystalline diamond material showing no distinct peak of SiV centers. The presence of SiV centers incorporated in the NCD during the initial growth step can not be completely excluded, however, their emission is masked by the tail of the background signal. After the overgrowth step the selected pillars with different diameters were measured under the same conditions and the PL spectra exhibited strong peaks at ca. 739 nm which we assigned to the ZPL of SiV centers (Fig. 8 (b)). These peaks overlapped with the broadband luminescence typical for CVD diamond and attributed to in-gap state distribution due to the presence of disordered carbon phase in the grain boundary material [41]**.** The SiV ZPL peaks were studied in more detail after the subtraction of the broad band of NCD. Their full widths at half maximum (FWHM) varied between 6 and 9 nm, matching well with literature data [29,42,43].

In some of the spectra a weak vibronic band on the long wavelength side of the ZPL could be observed, which corresponds to its phonon replica [44].

Finally, we applied a small band pass filter (Chroma 740/10) in order to observe only the fluorescence coming out from the SiV centers. Although we could still view confocal spots from the samples, the auto-correlation measurements revealed no anti-bunching. This fact suggested, that either the number of SiV centers in the nanopillars was quite high (> 10) or we still had a rather high background fluorescence disturbing the measurements. In order to reduce the number of incorporated color centers further experiments with reduced overgrowth time are in progress, which should also have impact on the quality of the top facets of the nanopillars. The smaller overgrown diamond could be also favorable for the enhancement of the SiV emission due to lower content of defects, quenching the emission, and especially in the case of additionally incorporated nitrogen atoms as electron donors [45].

4. Conclusions

We have demonstrated the incorporation of SiV color centers in diamond nanopillars during their overgrowth. The pillars with nominal diameters between 100 nm and 1 μm were structured from NCD films by EBL and RIE with oxygen plasma. ADF-STEM and EELS studies revealed the excellent quality of the NCD on which diamond with embedded SiV centers was grown. During the latter step Si atoms were etched from the bare substrate between the nanopillars and incorporated in the diamond lattice. The optical measurements of the nanopillars revealed strong signals of SiV indicating the creation of ensembles of these color centers during the overgrowth process.

Acknowledgements

The authors would like to acknowledge the financial support of the German Federal Ministry of Education and Research (BMBF) under the Program "Quantum Communication" (Project Q.com). ST gratefully acknowledges the FWO for a post-doctoral scholarship.

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Figure captions:

Fig. 1. Schema of a SiV center in the diamond crystal lattice

Fig. 2. Schematic diagram of the fabrication process of diamond nanopillars with incorporated SiV centers

Fig. 3. SEM image of an array of NCD pillars (in this case with a diameter of 500 nm)

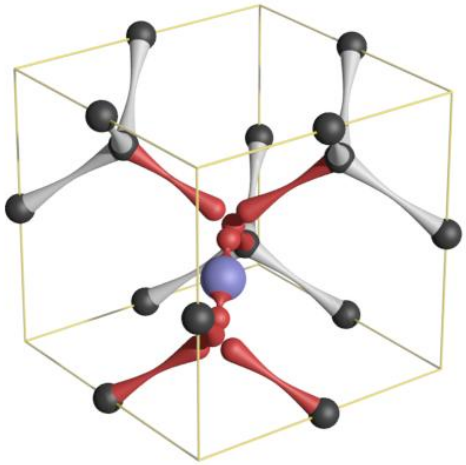
Fig. 4. ADF-STEM image of a NCD pillar with a nominal diameter of 200 nm

Fig. 5. (a) High resolution ADF-STEM image of the boundary between two grains (G1 and G2) in the NCD pillar. (b) Fourier transform pattern of the bottom grain in (a), evidencing the [110] zone axis orientation. (c) Enlarged view of the grain boundary. (d) EELS spectrum taken from a whole diamond grain.

Fig. 6. SEM images of NCD nanopillars with different nominal diameters after 30 min overgrowth: (a) 1 μm, (b) 500 nm, (c) 200 nm and (d) 100 nm

Fig. 7. Confocal images showing the fluorescence of arrays of diamond nanopillars with sizes of 1 μm (a) and 100 nm (b)

Fig. 8 Room temperature PL spectra of diamond nanopillars (in this case with a diameter of 100 nm) before (a) and after the overgrowth step (b)



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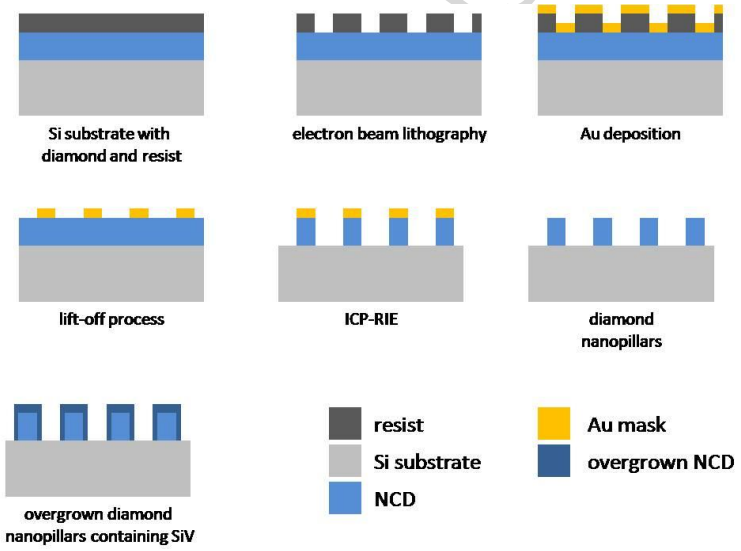

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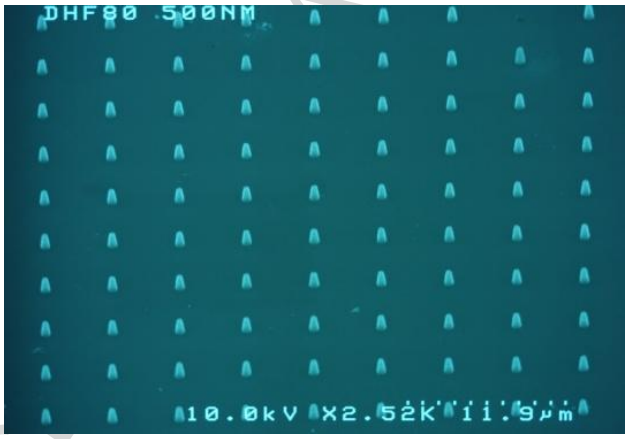

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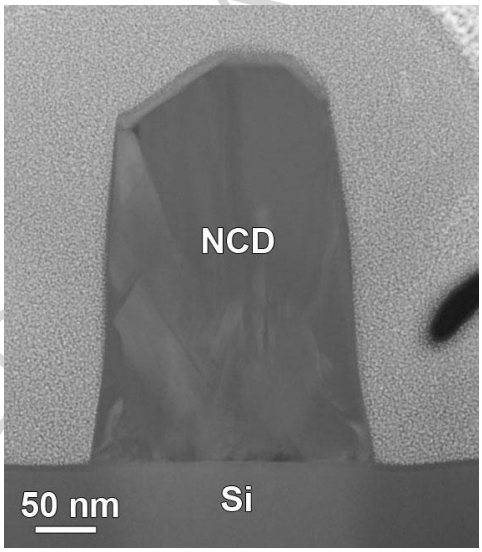


Fig. 4 N. Felgen et al.

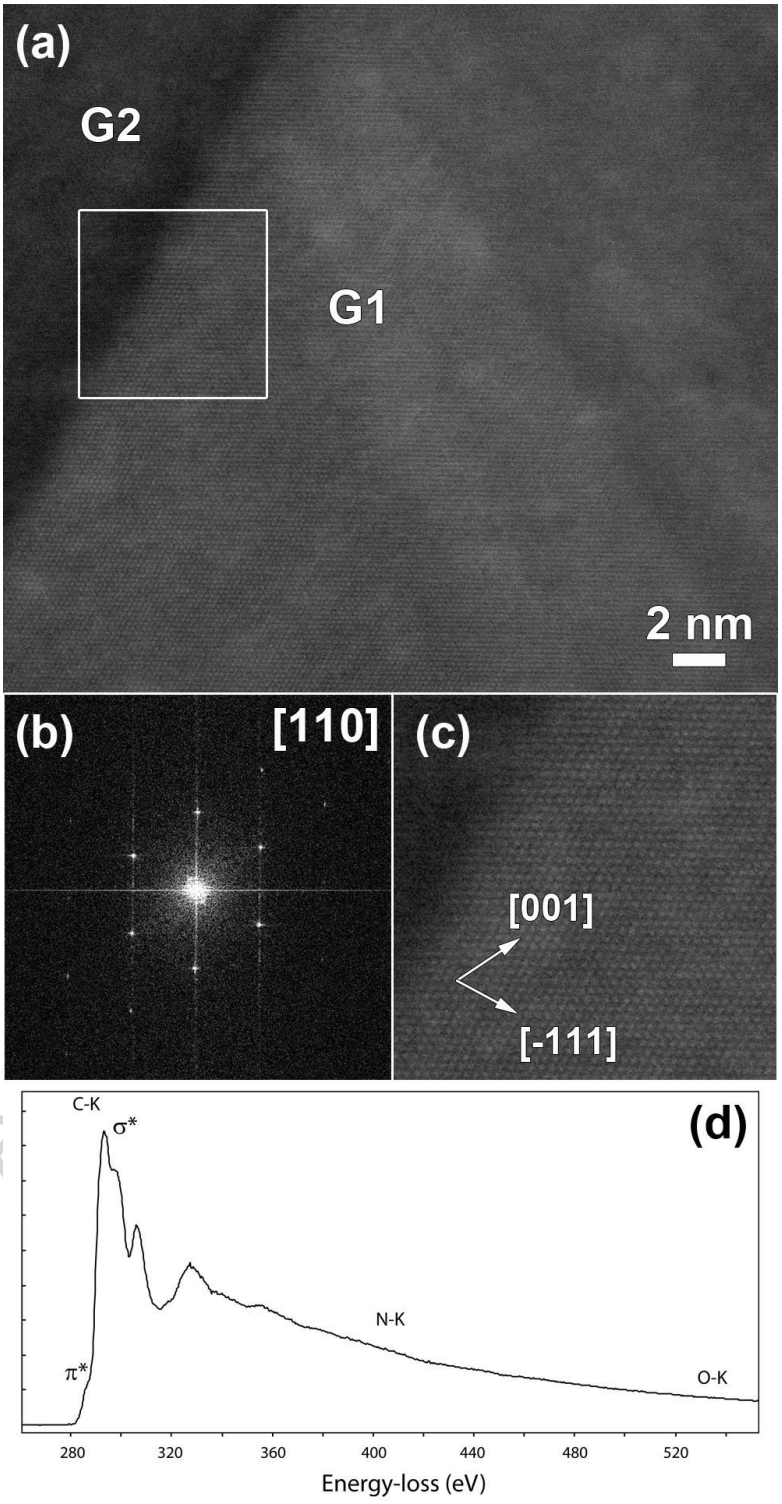


Fig. 5 N. Felgen et al.

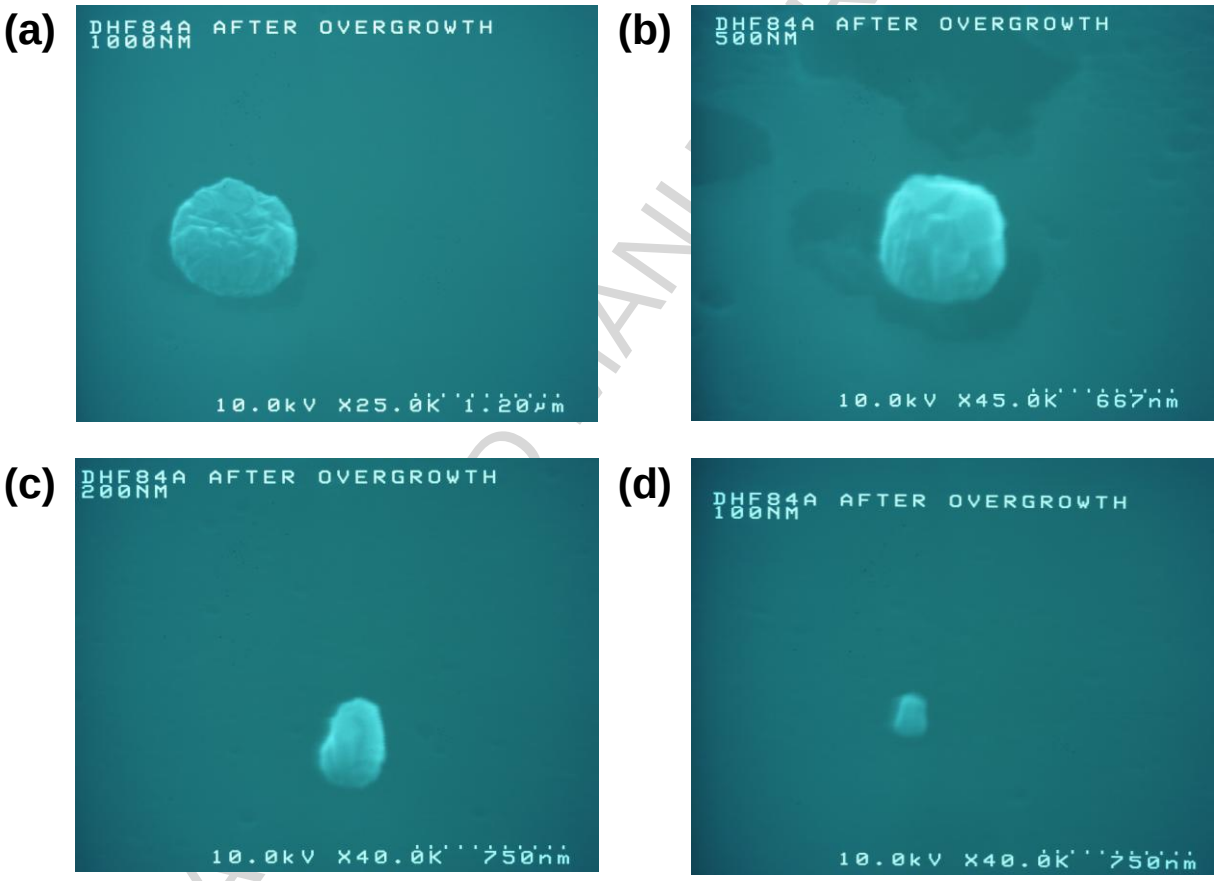


Fig. 6 N. Felgen et al.

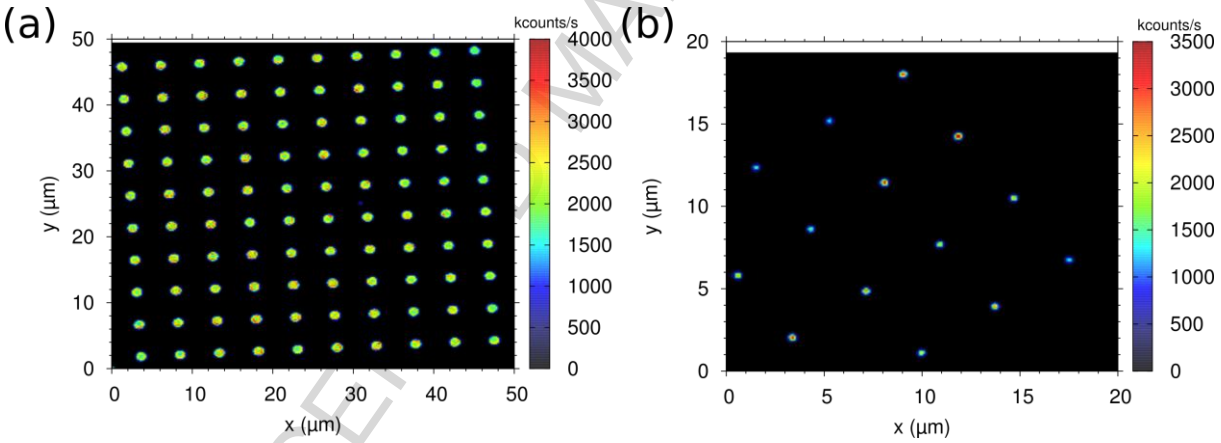


Fig. 7 N. Felgen et al.

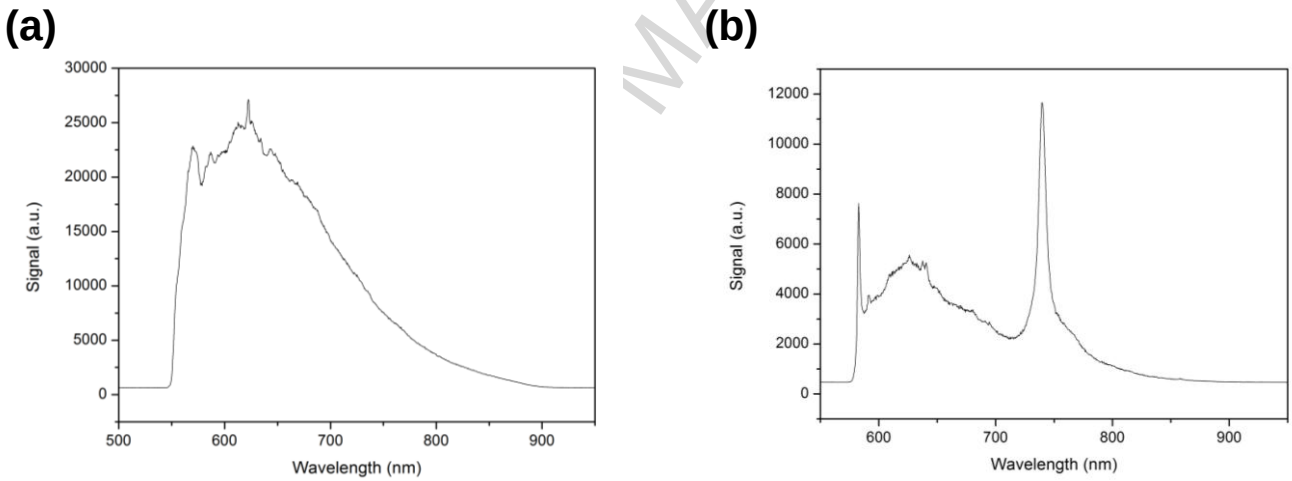
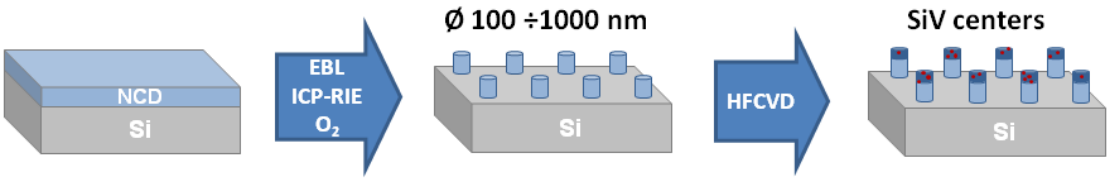


Fig. 8 N. Felgen et al.



Graphical abstract

Highlights

- Diamond nanopillars with diameters down to 100 nm were structured by EBL and RIE with oxygen plasma.
- SiV color centers were incorporated in the nanopillars during an overgrowth step.
- Optical measurements of the nanopillars revealed strong signals of SiV indicating the creation of ensembles.