CMOS compatible nanoantenna-nanodiamond integration

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Abstract. Here we demonstrate CMOS compatible method to deterministically produce nanodiamonds with nanodiamonds systems on example of bull-eye antenna on top of on hyperbolic metamaterials. We study the statistics of the placement of nanodiamonds and measure the fluorescence lifetime and the second-order correlation function of NV-centers inside nanodiamonds.

1 Introduction
Quantum technology is a relatively young field that manipulates at the atomic or microscopic level, based on the laws of quantum mechanics. For the practical implementation of quantum technology, photon sources are of interest, since photons are ideal information carriers. The ideal source of photons is a device that emits one photon on demand. A promising candidate for single-photon source is a nanodiamond with a nitrogen-vacancy (NV) color center. Nitrogen-vacancy-center nanodiamonds will emit single photon after excitation. Previously nitrogen vacancies (NVs) shown to be one of the most promising quantum system, but their optical characteristics (the generation rate and directivity) can be significantly improved. One of the way to enhance optical properties of NV is its positioning on hyperbolic metamaterials (HMM), but in this case HMM mostly increase radiation into high k-modes which are absorbed inside the material. One of the way to change it is to use nanooantennas [2]. We were pursuing this way in order to outcouple high k-modes into the free-space.

In this paper we continue our previous study of an controlled on-chip NV placements [1] and demonstrate CMOS compatible method to deterministically produce nanodiamonds and a bull-eye nanoantenna.

2 Device design and fabrication
At the beginning of the integration process nanoantennas and nanodiamonds a TiN layer of 60 nm thick on a HMM using reactive magnetron sputtering was deposited. We used a spin-coated negative e-beam resist (ma-N 2403) spin coated for 30 seconds with a speed of 3000 rpm. The resulting layer was dried on a hot plate at a temperature of 90 °C for 1 minute, providing resist thickness approximately 270 nm. After e-beam lithography drawing bull-eye antennas, we developed the resist in ma-D 525 and stopped reaction in water during two minutes. The etching of TiN was carried out with reactive ion etching (RIE) procedure in SF₆ atmosphere with a plasma power of 50 Watts. Images of four nanoantennas obtained with an atomic force microscope (AFM) can be seen in Fig. 1(a). To determine the location for the subsequent positioning of nanodiamonds, we used spin-coated 100 nm thick polymethyl methacrylate (PMMA) positive resist and baked at 150 °C for 2 minutes on a hot plate. Using e-beam lithography, we created an array of 33 by 33 holes with a diameter of 170 nm. Each hole we combined with the center of the corresponding nano-antennas. The accuracy of the resulting alignment ranges from 5 to 50 nm in different arrays. Then we developed the chip in a solution of isopropanol and water (8:1) for 20 seconds (Fig. 1 (b)). The next step, that is illustrated in
Fig. 1 (c), we put a drop of nanodiamonds solution and left it to incubate for 30 minutes. To avoid unnecessary particles, we first removed a drop of the solution with a pipette and sent a stream of isopropanol on the sample. Finally, for PMMA removing, we washed chip by an acetone stream heated to 55 °C, followed by washing in isopropanol and drying in stream N₂.

One of the fabricated devices is shown in Fig. 1 (d). Concentric circles represent a nanoantenna, in the center of which there is a nanodiamond with an NV center. The average size of nanodiamonds is 50 nm. Also, only every eighth nanodiamond in solution has a NV-center, which helps to create a single-photon source, even if there are a lot of nanodiamonds left after removing PMMA.

3 Experimental study and results
The first stage of research is devoted to the study of the effectiveness of the placement of nanodiamonds inside the nanoantennas. To count placement efficiency we used a Scanning Electron Microscope (SEM) images (one of them is shown in Fig.2 (a)). Totally, we investigated 700 on-chip nanoantennas, which were classified into 3 categories:

1) single diamond in the center of the nanoantenna (42.8%);
2) single diamond in the center of the nanoantenna is absent (33.5%);
3) diamonds are not located in the center of the antenna or located as agglomerate of nanodiamonds (23.7%).

Figure 2 (a-b). (a) AFM image four nanoantennas; (b) Scanning electron microscope (SEM) image of device after development of e-beam PMMA resist.

Diagram of placement efficiency is shown in Fig. 2 (b). It should be noted that we cannot reliably distinguish nanodiamond from dust particles of a similar size without confocal technique.

The second phase of the study is devoted to the study of nanodiamonds using confocal microscopy.

To analyze optical properties of NV-centers on the sample, we used home built confocal microscope employing high numerical aperture immersion oil objective (NIKON Apo TIRF 100X NA 1.49) with working distance of 120 µm (Fig. 3(a)). A continuous wave laser (Coherent Compass 300, wavelength 532 nm) served as the source of NV excitation and galvo mirrors (Cambridge Technologies) were used as a scanning element, enabling a 100 × 100 µm field of view for our microscope design (Fig. 3(b)). The collected fluorescence was coupled into a Hunbury-Brown-Twiss (HBT) interferometer that consisted of two avalanche photodiodes (PerkinElmer SPCM-AQRH-14-FC) and a 45:55 beam splitter. We used the combination of an optical notch filter with a stop-band centered at 532 nm and longpass optical filter with cut-off at 600 nm to remove the residual green excitation light and Raman signal from the collected emission. A time-correlated single photon counting module (Picoquant Picoharp 300) was used to obtain second-order photon correlation histograms from NV-centers (Fig. 4(b)).

For lifetime measurements, a picosecond diode laser (Picoquant LDH-P-FA-530 XL) was used with the same time-correlation single photon counting module. Due to the fact that lifetime of NV-center excited state is much longer than the length of the laser pulse, the fluorescence signal recorded after the end of the applied laser pulse should exhibit exponential decay (Fig. 4(a)). In Fig. 4(a), the actual signal observed consist of two exponents – fast and slow one. The fast exponential component corresponds to fluorescence of the sample and other similar background fluorescence signatures that do not have a slowly decaying exponential component. This was verified by focusing the microscope on a region of the sample that contained no diamonds and measuring the fluorescence decay time collected from this region. We therefore associate long-lived exponential tail in Fig. 4(a) with the NV-center luminescence.

Confocal microscope allow distinguish nanoantenna with NV-centers nanodimonds in the antennas array (Fig. 3(b)) and investigate the NV-centers optical properties. The orange circle shows the single NV-center nanodiamond (Fig 4(b)) inside the bull eye antenna. Figure 4 (b) demonstrates
second-order autocorrelation function measurements deep $g^2(0) \approx 0.3$ which associated with single NV-center. Figure 4(a) demonstrates lifetime measurements of 8.3 ns for single NV-center inside bull eye antenna.

![Diagram](image1)

**Figure 3 (a-b).** (a) Scheme of the confocal microscope; (b) CLSM image of array of nanodiamonds inside nanoantennas.

Totally, we investigate 11 single NV-centers on glass and 5 single nanodiamonds on HMM nanostructures. The average lifetime for NV on nanostructures was 8.5 ns, while on glass it is 22.5 ns.

![Diagram](image2)

**Figure 4 (a-b).** (a) Lifetime measurements for single NV-center inside nanoantenna; (b) second-order autocorrelation function $g^2(\tau)$ for single NV-center.

Thus, the placement technology gives satisfactory results. At the same time, the enhancement of the radiation of nano-amps with antennas in the metamaterial is ambiguous, since only 5 cases were investigated there. The investigation of the antennas efficiency requires further research.

### 4 Conclusion

We demonstrated nanodiamond with color centers integrated into the CMOS process to create integrated optical circuits. The placement technology gives satisfactory results when 42.8% of nanodiamonds were placed as desired. Further research will be directed to study optical and spin properties of NV [2-4] inside nanoantennas. Moreover, nanodiamonds with single NV-centers were found and their optical were characterized.
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