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Single photon emission of “silicon-vacancy” centers in nanodiamonds placed in cylindrical pits on a gold film

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Abstract. In this paper, we study photon emission of single “silicon-vacancy” centers in HPHT-nanodiamonds on 1) a gold surface and 2) inside nanophotonic cavity which is cylindrical nanopit on a gold film. It was found that the saturation intensity of single SiV centers in the nanocavity increases by up to 3 times compared to similar diamonds on the gold surface, along with a twofold decrease in saturation power and a 15% reduction in lifetime. The obtained results are explained by the interaction of SiV centers with the surface plasmons in cylindrical nanopit in gold film, as well as by the narrow directivity of the emitter related to geometry factor of the plasmonic nanocavity.

Keywords: nanodiamonds, color centers, microcavities, single photon emitters

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Материалы конференции
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Флуоресценция одиночных центров окраски «кремний-вакансия» в наноалмазах в цилиндрических углублениях на золотой пленке

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Аннотация. В настоящей работе изучалась однофотонная эмиссия центров окраски «кремний-вакансия» в HPHT-наноалмазах 1) на поверхности золота и 2) в цилиндрическом углублении (ЦУ) на золотой пленке. Было обнаружено, что интенсивность насыщения одиночных центров SiV в ЦУ увеличивается до 3 раз по сравнению с аналогичными алмазами на поверхности кремния, наряду с двукратным снижением мощности насыщения и сокращением времени жизни на 15%. Полученные результаты объясняются взаимодействием SiV-центров с поверхностными плазмонами в микрорезонаторе, а также узкой направленностью излучения в виду геометрического фактора плазмонного ЦУ.

Ключевые слова: наноалмазы, центры окраски, микрорезонаторы, однофотонные эмиттеры

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Introduction

The single photon source (SPS) is cornerstone in quantum information transmission and processing protocols [1–3]. Such a source must possess high photostability, brightness and emit photons in a narrow spectral range and solid angle [4]. The “silicon-vacancy” color centers (SiV) in nanodiamonds (NDs), having a bright and narrow (~ 6 nm) zero-phonon line (ZPL) in the near IR region (738 nm), are promising candidates for this role [5, 6]. The emission properties of SiV centers benefit significantly from placing NDs in microresonators [7–10] due to enhancing the rate of spontaneous transitions and increasing the radiation directivity. Moreover, the rate of quantum emitter photoexcitation can be significantly enhanced, which is most clearly manifested in the case of plasmonic nanoantennas [10].

In this work, the fluorescence properties of single SiV centers in HPHT nanodiamonds placed 1) on a gold film and 2) in cylindrical nanopits on a gold film were studied. At the first stage, saturation curves, spectra and decay times of fluorescence were measured for the particles on the gold film. Next, the “pick-and-place” method we developed was used to move the selected nanodiamonds to the cylindrical nanopits on a gold surface, where similar measurements were carried out. Finally, we provide a comparison of optical properties before/after ND moving and discuss the mechanisms contributing to the fluorescent enhancement.

Materials and Methods

The NDs under study were synthesized by the High-Pressure High-Temperature (HPHT) method from a mixture of adamantane ($C_{10}H_{16}$) and detonation nanodiamonds at a pressure ~ 7.5 GPa, temperatures 1500–1600 °C and time exposition 20 s. To form SiV-centers in diamond matrix, a small amount of tetrakis(trimethylsilyl)silane ($C_{12}H_{36}Si_5$) was added to the initial growth mixture with the ratio Si/(Si + C) = 0.01%. The 0.1 mg of output ND-powder was diluted in 2 ml of ethanol and then sonicated in the ultrasonic bath for 30 min to form a homogeneous suspension of NDs. Eventually, 2 μl of the resulting suspension was applied to the gold film with cylindrical pits. A pre-characterization in Scanning Electron Microscope (SEM) revealed the ND size distribution between 150 nm and 500 nm.

The process of cylindrical pit fabrication on a gold film was described in detail in our previous work [10]. The diameter and height of such nanopits are 500 nm and 300 nm, respectively. To transfer NDs to the surface of the gold film and then inside the cylindrical nanopit, we used a "pick-and-place" technique we developed earlier, described in [10].

Optical properties of NDs were studied in home-built confocal microscope equipped with two sources: a continuous laser at 660 nm and pulsed one at 630 nm ($\tau_{pulse} \approx 80$ ps). To confirm the single photon emission from selected ND, we used a Hanbury–Brown–Twiss (HBT) interferometer with two avalanche photodiodes (APDs, Excelitas SPCMA–QRH–14–FC). The fluorescence spectra were recorded with Ocean Insight QEPro spectrometer, and for saturation measurements, the APDs with band pass filter (728–749 nm) were employed. A long–distance objective Mitutoyo ×100, NA = 0.7 was used in all these measurements. The fluorescence decay rates were analyzed with Time-Correlated Single Photon Counting (TC SPC) module Picoquant MicroTime 200 and objective Olympus (×100, NA = 0.95).

Results and Discussion

We started by choosing three random NDs on the gold surface whose emission satisfies to criteria of single photon statistics. In order to reveal such sources, we performed precise confocal mapping and measured the second-order autocorrelation function $g^{(2)}(\tau)$ of SiV fluorescence. Fig. 1, *a* demonstrates an example of $g^{(2)}(\tau)$ functions for NDs on the gold surface (NG–G). The dip at zero time–delay ($\tau = 0$) is less than 0.5 indicating a subpoissonian statistics of light and consequently its single-photon nature. Fluorescence spectra, saturation curves and decay times were measured for selected NDs. After characterization of the ND emitters on the gold surface, we moved the NDs to pre-defined cylindrical pits by the "pick-and-place" method and performed similar measurements. A SEM image for ND1–GNP (Fig. 1, *c*) reveals the geometry of the nanophotonic interface and the size of the diamond particle (~ 350 nm).

Fig. 1, *b* illustrates the background-corrected fluorescence spectra recorded for ND1–G and ND1 inside the microcavity (ND1–GNP) at 30 mW excitation. A bright zero-phonon line (ZPL) at 738 nm and a sharp diamond Raman peak are observed in both spectra. For ND1–GNP the peak intensity of ZPL slightly increases by 30% relative to diamond Raman. The spectral position of ZPL is shifted to the IR region by 0.2 nm, while the full width at half maximum (FWHM) is reduced by ~ 10% in comparison to ND1–G. This behavior could be a product of enhanced interaction between the emitter and plasmon in microcavity.

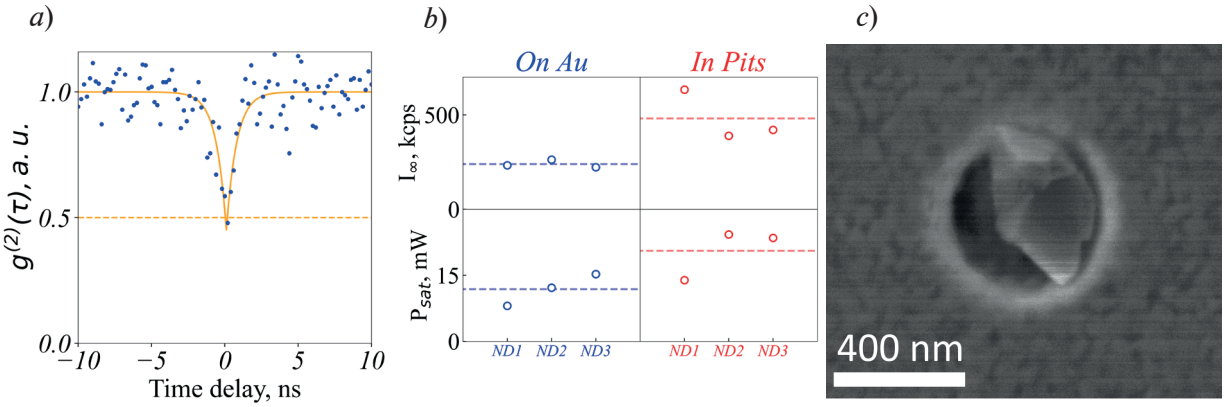

Fig. 1. Second-order autocorrelation function $g^{(2)}(\tau)$ for ND on the surface of the gold film (*a*); Fluorescence spectra of single SiV–centers in NDs on gold film (blue) and in cylindrical pits (red) (*b*); SEM–image of ND1–GNP (*c*)

For quantitative analysis of the fluorescence enhancement factor, the saturation measurements were carried out. Fig. 2, *a* shows the fluorescence intensity of a single SiV as a function of excitation power for ND1–G (blue) and ND1–GNP (red). The data was approximated by the curve

$$I(P) = I_\infty \frac{P}{P_{sat} + P} + c_{bg} \cdot P,$$

where I is the fluorescence intensity, P is the excitation power, I_∞ is the count rate in the limit of large P, P_{sat} is the saturation power, c_{bg} is the coefficient of linear background. The values I_∞ and P_{sat} extracted for three NDs in different photonic environments are presented in Fig. 2, *b*. One can see that emission rates of NDs in nanopits on average exceeds the similar parameter for NDs–G by an enhancement factor $EF = 2$ with the maximum value for ND1 $EF_{\max} = 3$ corresponding to $I^{\infty}_{ND1\text{-}GNP} = 634$ kHz. The saturation power on average also increases from $P^{sat}_{NDs-G} = 11.8$ mW to $P^{sat}_{NDs-GNP} = 20.5$ mW which probably takes explanation from additional absorption by void plasmons [11].

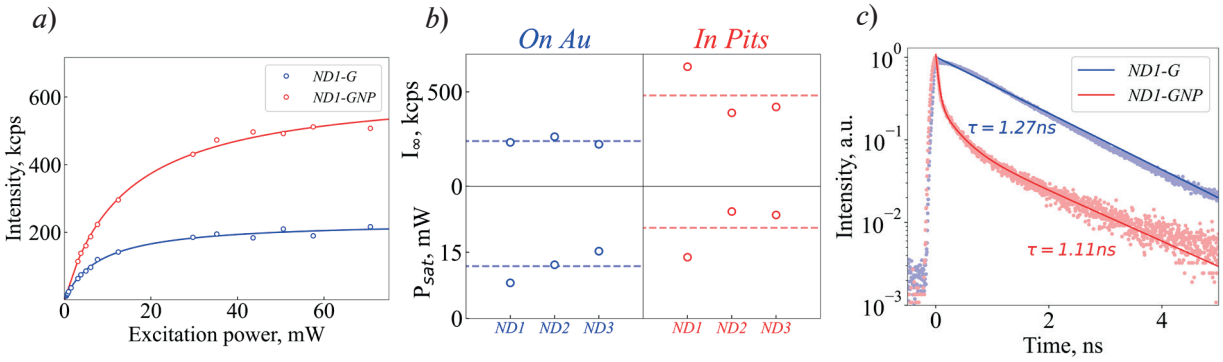

Fig. 2. Saturation curves measured for ND1 on gold surface (blue) and in nanopit (red) (*a*). Extracted I_∞ (top) and P_{sat} (bottom) for three NDs SiV luminescence on gold surface (blue) and in nanopits (red) (*b*). Fluorescent decays for ND on surface (blue) and in nanocavity (red) (*c*)

Finally, we performed time-resolved fluorescence intensity measurements for the NDs both on a gold surface and in a nanophotonic cavity. The SiV-fluorescence decay times extracted from the decay curves were averaged over three NDs and compared between each other. The relaxation time reduces by ~ 15% on average for emitters inside a nanophotonic cavity which can be a sign of effective coupling with void plasmons in a near–surface gold layer. The fluorescence decays for ND1–G demonstrate mono–exponential behavior (Fig. 2, *c*)). In contrast, for ND1–GNP deeply sub-ns component was revealed which we believe is not related to SiV-fluorescence, but rather to an enhanced background fluorescence originating from the gold nanopit [12].

An observed enhancement of the single photon emission can be explained by analyzing contributions from major mechanisms. First, the emitter–plasmon interaction increases in nanophotonic cavity which is confirmed by a slight reduction of the fluorescent lifetime. Second, ND-in-pit interface changes the geometrical factor of the fluorescence, resulting in enhanced directivity of the SiV-center and its extracted emission rate. Third, the small heterogeneity in enhancement factor from particle to particle can be related to inaccuracies in nanomanipulations with diamonds. During the “pick-and-place” operation ND accidently change its spatial position losing the initial orientation of the dipole moment of the emitter relative to the normal of the gold surface. Nevertheless, we believe changes in dipole orientation play a non-primary role in observed enhancement since for all accidentally transferred diamond particles the increase of the emission rate was detected which excludes the factor of accident orientation.

Conclusion

In summary, we investigated single photon emission of SiV-centers in nanodiamonds placed 1) on a gold surface and 2) in plasmonic nanocavity and provided a comparative study of optical properties for these two cases. A nanophotonic structure nanodiamond-in-pit exhibits a 3-fold enhancement of fluorescence intensity with reduced by 15% fluorescent lifetime and narrowed by 10% spectral range of the emission. Such a structure demonstrates promising potential for state-of-the-art quantum applications giving rise to scalable photonic interfaces.

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