Investigation of a single-photon hybrid emitting system based on NV-centers in nanodiamonds integrated with GaP NWs

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Abstract: NV-centers can be used for quantum informatics, quantum communication and quantum sensing. The calculation of optical modes formed in a GaP cylindrical nanocavity covered by nanodiamonds has been performed. GaP nanowires have been synthesized with molecular beam epitaxy and played the role of optical resonators for light-emitting centers on the base of nanodiamonds with NV-centers. The optical characteristics of the GaP-based nanocavity were analyzed. The increase in the rate of spontaneous emission of NV-centers optically coupled to the nanocavity was estimated by the time correlated single photon counting method.

1. Introduction
Nanoscale quantum optical emitters are main elements of quantum optics and there are widely used for sensing applications. Nanodiamonds (NDs) with optically active point defects in the crystal lattice can be used as such sources. One of the most studied crystal lattice defects is a nitrogen-substituted vacancy or NV-center. The spins of electrons in NV-centers can be adjusted at room temperature using a magnetic field, electric field or optical radiation [1]. The main problem that limits application of NV-centers in quantum nanophotonic systems is associated with the low brightness of zero-phonon line (ZPL) at room temperature due to broad phonon side-band [2]. A conventional approach to solve the problem of increasing the NV-center ZPL emission rate as well as efficient collection of emitted photons is based on the integration of the radiative nanodiamonds with optical cavities or waveguides [3]. Gallium phosphide (GaP) nanowires (NWs) which have large length-to-diameter ratio are the most promising candidates to be used as optical resonance systems since GaP NWs have an indirect band gap with relatively large energy that makes its transparent in the visible range including ZPL line of NV-centers. Another advantage is the optical refractive index, which is about 3.2 within the visible range which is higher than most optical materials. For example, the refractive index of diamond is 2.4.

2. Numerical calculation
The primary calculation aim is to determine the conditions providing efficient coupling between a nanocavity based on a single GaP NW and nanodiamond with an emitting NV-center. Another main task is related to assess the system resistance to coupling errors. To solve this problem we used...
numerical simulation of a coupled optical system in the wave optics approximation with Comsol Multiphysics software package. The analysis of the system was led into two steps. At the first step we looked for NW diameters having optical resonance near the ZPL frequency (470 THz). Also we studied the near-field distribution in the cavity and the corresponding far-field radiation pattern. At the second step the nanodiamond was placed to NW sidewall and the Purcell factor and the radiation pattern as a function of the NW diameter and the polarization of the NV-center were determined for ZPL frequency.

In the numerical simulation GaP NW was represented as a 3D elongated object with a hexagon cross-section and 5 μm length. The nanodiamond was represented by a cube with a size of 60 nm. The refractive indexes and absorption coefficients for GaP and diamond were taken from [4, 5]. Analysis of the obtained data was carried out for NW diameters which provides eigen frequency of optical resonance equaled to ZPL.

According to the results of numerical simulation we plotted the Q-factor of optical nanocavities based on GaP NWs at ZPL frequency (470 THz), see Figure 1. Figure 2 shows the maps of the electric field distribution inside the resonator and the corresponding radiation patterns for three typical NW diameters. One can see that the increase in the NW diameter leads to the increase in the Q-factor of the resonator. It can be associated with the size-dependence of the energy localization inside the NW, Figure 1.

![Figure 1. Plot of Q factor of optical modes of GaP NW.](image)

Analysis of the distribution of electromagnetic fields shows that with a diameter lower than 160 nm the resonator modes are mainly represented by the fundamental TEM\(_{00}\) mode, Figure 2 (137 nm), which is optimal for obtaining directional (along the NW axis) radiation. A further increase in diameter leads to the appearance of higher order transverse modes such as TEM\(_{01}\) and TEM\(_{11}\), which lead to the increase in the contribution of side share to the radiation pattern, Figure 2 (160 nm), (188 nm).

The calculation of composite GaP-NW / NV-center system was carried out for the obtained at previous stage NW diameters. Nanodiamond was placed onto NW sidewall and play a role of the optical dipole source. The calculation was performed for three polarizations of the dipole source: the one is along the NW axis (Z) as well as two transverse polarizations – along (X) and across (Y) the sidewall. The distribution of the electromagnetic field of a nanocavity with a diameter of 137 nm and the radiation pattern for three orientations of the dipole are presented in Figure 3. As can be seen from the maps of the near field the maximum amplitude of the electromagnetic wave induced in the NW is...
achieved for the Y polarization which is associated with the penetration of the near field of the NV-center dipole into the core of the NW. The minimum coupling, in turn, is observed for the X orientation of the polarization. Far-field analysis shows that for a NW diameter of 137 nm, Y and Z polarizations of the NV-center provide the light propagation along the NW axis, which can be successfully used to suppress the directivity pattern of the NV-center.

![Image of far and near fields](Image)

**Figure 2.** Far and near fields’ for three characteristic diameters of GaP NW: b) 137 nm, c) 160 nm, d) 188 nm.

![Image of far and near fields](Image)

**Figure 3.** Far and near fields’ distributions for GaP NW (137 nm in diameter) for three different polarization of NV-centers.

According to the results of numerical calculation, the dependence of the Purcell factor on the nanocavity diameter was calculated for three different polarizations of the NV-center. Figure 4 shows a graph of the obtained dependencies.

As noted above, for the X polarization (when the dipole weakly interacts with the cavity and the near field strongly goes beyond its limits), we observed the smallest Purcell factor (gray curve). It confirms the weak optical coupling of the NV-center with GaP NWs. For NW diameter about 160 nm, we observed the highest value of the Purcell factor. Presumably, it is associated with the complication of the mode composition of NWs and the enhancement of the optical coupling between NWs and nanodiamonds. Y (red curve) and Z (blue curve) polarizations are characterized by enhanced values of the Purcell factor, since they have a larger optical coupling with the resonator. The dependence of the slope of the curve for Y polarization is related to the fact that with an increase in the diameter the fraction of the energy contained in the evanescent field decreases, while the fraction of the energy inside the resonator increases. For Z orientation, there is also nonlinear dependence and the optimal
value is determined by the coincidence of the field lines generated by the dipole with the localization maxima of the modes in the nanocavity. This is due to the fact that with an increase in the diameter of GaP NWs, the effective refractive index of the system increases and the field maxima begin to approach each other. Thus, we obtain the optimal configuration of the field maxima near NW diameter of 150 nm, which leads to the best optical coupling between the NV-center and GaP NWs. Oscillations on the graph are associated with the dipole “hitting” an even or odd localized mode in the near field, Figure 4.

![Figure 4](image-url)

**Figure 4.** Plot of the Purcell factor on NW diameter for different NV-center orientations. Gray curve – polarization along the lateral face of the NW, Red curve – polarization across the lateral face of the NWs, Blue curve – polarization is directed along the NWs. Solid lines – fitting of the acquired data.

Based on the obtained model, we determined the optimal geometric parameters of a nanocavity based on nanodiamond combined with GaP NWs for three different polarization. The NW diameter near ~ 150-160 nm provides the best characteristics for increasing the luminosity of NV-centers due to the high Purcell factor.

3. Experiment

Arrays of GaP NWs were synthesized on a Si (111) substrate using a molecular beam epitaxy Veeco GEN III machine. The morphology of the grown GaP NWs was studied using a scanning electron microscope (SEM) (Zeiss SUPRA 25-30-63). The obtained NWs had the length of 12 ± 2 µm and diameter 155 ± 15 nm. Nanodiamonds were deposited to NWs array from a liquid suspension with microdispenser, Figure 5 (a).

Microspectroscopic studies of the photoluminescence (PL) response and Raman scattering of light (RS) of individual NWs covered with the ensemble of nanodiamonds were performed at a room temperature (300 K) (see Figure 5). Figure 5 (b) demonstrates two typical peaks corresponding to two charge states of the defect, namely: NV\(^{0}\) (575 nm) and NV\(^{-}\) (637 nm). As shown in Figure 5 (d) the PL spectrum for a single NW with deposited nanodiamonds demonstrates intensity modulation associated with the eigenmodes of the cavity which also confirms the optical coupling between emitting NV-centers and NWs.

The photoluminescence decay time of NV-centers was investigated using the time-correlated single photon counting (TCSPC) method on the sample with single NWs as well as reference glass covered by NDs. We used a 532 nm femtosecond laser with a Mitutoyo M Plan 100X NA 0.7 objective. Figure 6 shows the intensity and lifetime maps of NW-NV-center system.
Figure 5. a) Scheme of the synthesized GaP nanowire array covered by nanodiamonds b) PL spectrum of NW array with NDs, c) Scheme of a single NW with NDs transferred on an arbitrary glass substrate, d) PL spectrum of a single NW with ND; the inset shows an optical image of NWs and demonstrates the light trapping effect by the NW.

Figure 6. Intensity map and map of lifetime for: a), b) - a reference sample, c), d) - a single NW with a deposited ensemble of nanodiamonds, respectively.

The average lifetime of NV-centers on the reference sample was ~ 18.2 ns, while on the sample with a single GaP NWs, ~ 9.2 ns. The lifetime ratio for these samples gives the Purcell factor corresponding to ~ 2 [6]. This value lower compared to the results of numerical calculation due to many reasons. Firstly, the lifetime value of the reference sample is underestimated due to presence of greater than unity inherent Purcell factor of reference sample. Thus, this leads to decreasing the lifetime ratio. Secondly, in our samples there are many NV-centers located at different points inside the diamond particle thus we observe the PL decay rate averaged over the ensemble of emitting NV-centers. It is known that such averaging usually leads to lower values of the Purcell factor [7]. Thirdly, in our optical measurement geometry collection and pumping occur perpendicular to the NW axis and
therefore the optical signal is mainly contributed by two dipole components: X and Z which have lower Purcell factor compared to Y part (on average ~ 2-5, see Figure 4). The last conclusion is supported by numerical calculation.

4. Conclusions
In this work, we performed a numerical calculation of the Q-factor of the optical modes of a nanocavity based on GaP NW at the ZPL frequency (470 THz), and obtained diagrams of the electric field distribution inside a cavity based on GaP NW combined with an NV-center. The Purcell factor was analyzed as a function of the NW diameter for three different dipole polarization of the NV-center in nanodiamonds. A method of nanodiamond integration with NV-centers into an array of GaP NWs has been developed. The analysis of the optical properties of the fabricated structure was carried out and the PL and Raman spectra were obtained, which confirmed the waveguide properties of NWs. Increasing the spontaneous emission rate of an NV-center in a nanocavity system in comparison with the emission of reference samples has been experimentally demonstrated.

Acknowledgments
Modeling and development of a nanocavity based on GaP NWs combined with a diamond nanoparticle containing an NV-center was supported by the Russian Science Foundation (grant 19-19-00693).

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