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Control of luminescence in resonant nanodiamonds with NV-centers

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Abstract – **Resonant high-index nanostructures possess unique properties allowing development of nanophotonic devices: surprising ways of emission manipulation at subwavelength scale, efficient control of radiation pattern, and low dissipative losses. Here, the resonant properties of nanodiamonds with NV-centers in visible region were studied and the influence of resonance nature of nanodiamonds on the luminescence lifetime at zero-phonon line was demonstrated.**

I. Introduction

Resonant high-refractive index nanostructures provide new physical effects as well as effective light manipulation at nanoscale and pave the way for creation of novel photonic devices. One of the most interesting features of such structure is enhancement of the spontaneous emission rate [1, 2].

Nanodiamond with nitrogen-vacancy (NV) centers are of special interest. Indeed, diamond is characterized by relatively high refractive index (n $\approx$ 2.4) and almost zero absorption in visible and NIR regions. This makes nanodiamonds with NV-centers good candidates for the observation of morphology- and size-dependent resonances. It should be mentioned, that the optical properties of nanodiamonds containing colour centers are of interest for development of single photon quantum emitters integrated into photonic circuits [3]. Colour centers in nanodiamons such as nitrogen-vacancy, silicon-related and nickel-related colour centers are artificially created defects [4, 5]. NV-center is the only diamond colour center that has demonstrated coherent optical effects [6, 7]. Such defect along with the high-index of diamond have performed the unique properties for photonic devices, like stability of diamond structure and the existing of zero phonon line (ZPL) at 637 nm.

In this work we present experimental observation of Mie-type resonances in nanodiamonds with NV-centers. We demonstrate that spectral position of the resonances has a direct influence on the lifetime at zero-phonon line providing lifetime reduction in the resonant case up to 1.5 times compared to non-resonant one.

II. Results and Discussions

The samples of nanodiamonds were fabricated by plasma-enhanced chemically vapor deposition (PECVD) method and represent pieces of complicated shape. The example of SEM (scanning electron microscope) image of studied nanodiamonds is shown in the Figure 1a. All the nanodiamonds can be confined by spheres with radii in the range from 300 to 1000 nm. The study of luminescent properties demonstrates, that all the studied nanodiamonds possess pronounced peak at 637 nm in photoluminescence spectra (Figure 1c). Photoluminescence properties were studied with an objective (10, NA=0.28, Mitutoyo Plan Apo VIS) and supercontinuum laser system (Fianium). The laser system excited nanoparticles at the wavelength of 530 nm and bandwidth of 40 nm. The measurements of lifetime were carried out by the Time-Correlated Single Photon Counting method. Analysis of this signal was conducted with confocal system with a Raman spectrometer (HORIBA LabRam HR) and cooled CCD Camera (Andor DU 420A-OE 325) equipped with a 150 g/mm diffraction grating. Additional CCD camera (Cannon 400 D) allowed to control the location of nanoparticles.

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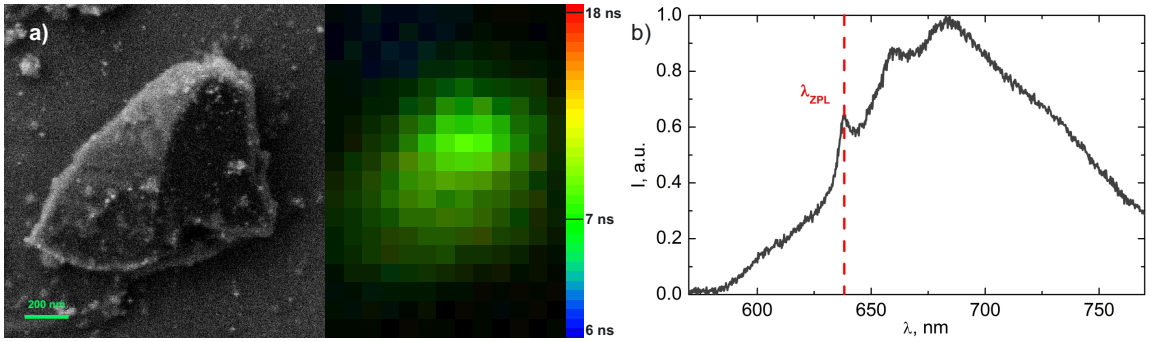

Fig. 1: a) Studied nanodiamond containing NV-center: left - the SEM image, right - the map of lifetime measurement; b) The photoluminescence spectrum

The investigation of scattering properties demonstrates Mie-type resonances. For studying of scattering properties of nanodiamonds we used a dark-field setup with an infinity corrected objective 10, NA=0.26, Mitutoyo Plan Apo NIR placed under the 67 degrees angle to the surface normal and an objective 50 NA=0.42 Mitutoyo Plan Apo NIR - perpendicularly to the sample surface. The first objective was employed for illumination with a white light source (HL-2000 halogen lamp) and the second one was applied for scattering signal collection. According to Mie theory the spectral position of resonances depends on shape and size of the nanoparticles, and in a sphere made of high-index material the magnetic dipole mode provides the main contribution to the scattering when the wavelength of light inside the nanoparticle equals to its diameter $2R \approx \lambda_0/n$ [10, 11, 12]. In contrast to plasmonic nanoparticles high-index dielectric ones are characterized by low dissipation losses and exhibit both electric and magnetic dipole resonances [9]. In our experiments we have observed only higher-order multipole Mie resonances due to the large sizes of the samples.

Resonances at 637 nm were studied detailed for the attempts to manipulate by ZPL in nanodiamond spectra. The series of measurements of scattering spectra have demonstrated that spectral position of resonances relatively to ZPL in the photoluminescence spectra has a direct influence on the lifetime. To demonstrate this effect, two samples with different spectral positions of resonances at the wavelength of ZPL were chosen (see Figure 2a,b.).

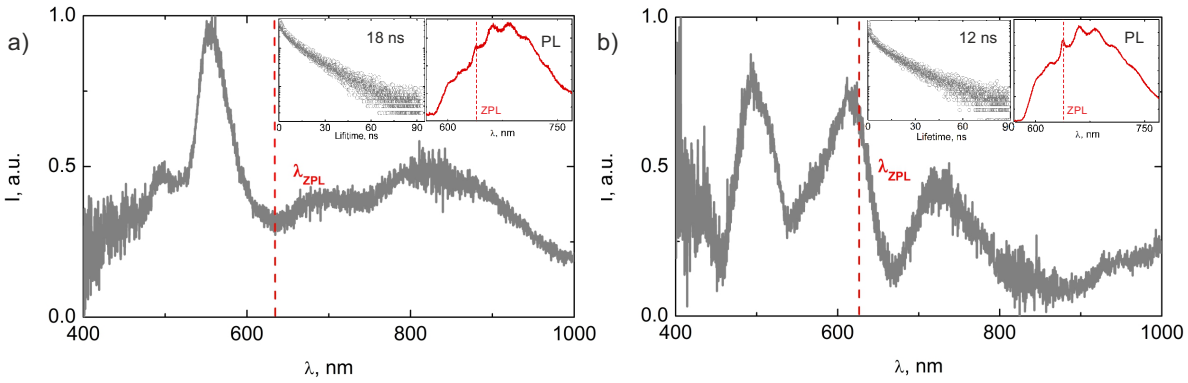

Fig. 2: The dark-field scattering spectra of nanodiamonds a) off-resonance case with the measured lifetime of 18 ns and b) resonance case with the measured lifetime of 12 ns. Insets: left - lifetime decay and right - photoluminescence for the off-resonance and resonance cases

The scattering spectra of samples with nanodiamonds are demonstrated in Figure 2 for resonance and off-resonance cases. The off-resonance case (Figure 2a) we obtain the measured lifetime value of 18 ns. The resonance excitation case (Figure 2b) leads to decreasing of lifetime by a factor of 1.5. In this way spectral position of resonance directly affects the ZPL emission and lead to lifetime decreasing. Due to the fact that lifetime is linked directly with emission of colour centers in nanodiamonds [8] the manipulation of resonance position allows to

control ZPL emission.

III. Conclusion

In this work we have experimentally demonstrated control of ZPL emission at room temperature via Mie-type resonances in nanodiamonds. In this way the emission from nanodiamonds with NV-centers can be controlled by changing the spectral position of resonance. We have observed that the decrease of luminescence lifetime is 1,5 time for resonant case compared to non-resonant one. Such effect makes resonant nanodiamonds with NV-centers a promising platform for low-loss nanophotonic devices and allow to control emission properties at ZPL. We believe, the achieved results can be applied for creation of new-generation photonic devices based on resonant luminescent nanodiamonds.

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