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Investigations on Diamond-NV-Centers as Alternative Labels in STED Microscopy

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Abstract. We investigate diamond nitrogen-vacancy (NV) centers as alternative labels in stimulated emission depletion (STED) microscopy. To this end, artificial diamond is used as a substrate and Raman spectroscopy in photoluminescence (PL) mode is performed for clearly identifying the emission wavelengths of the NV centers. With the aid of a STED microscopy system, we imaged a random feature on substrate surface with the NV-centers in STED and confocal mode. Our first measurements indicate that the combination of NV centers and STED is very promising for resolving the structures and can be further extended to nanoscale structures applied on the diamond substrates.

1 Introduction

Today, developing and fabricating micro- and nano-scaled electronic and optical devices requires combining complex structures of different materials. Finding reliable and precise measurements for analyzing these structures is a challenge for various fields of science and industry. Light microscopy is an optical metrology technique that provides physical and chemical information about a sample. Although light microscopy is a non-contact, non-destructive, and high-speed method, it suffers from a resolution limit due to the wave nature of light [1]. In recent decades, super-resolution microscopy techniques containing a resolution beyond diffraction limit were developed [2]. Although STED microscopy is a promising and precise optical far-field technique for nanoscopy, its use is limited to systems based on fluorescence markers [3-5]. NV-centers in diamond substrates have great potential as alternative labels in super-resolution microscopy if they have a high density and homogeneous distribution throughout the sample surface providing a homogeneous fluorescence [6-7].

To establish NV-centers as an alternative label for superresolution microscopy, in this contribution, first, we investigate the distribution of NV-centers in a diamond substrate using Raman spectroscopy in photoluminescence (PL) mode for the existence of NV centers in the substrate and for comparing the spectral resolution as proposed by Balasubramanian *et al.* [8]. Then, we imaged a random feature on the substrate surface with the NV-centers in STED and confocal mode with the aid of the STED microscopy system. As a result of preliminary tests, STED mode already provided higher resolution images of the structures on the substrate surface than the confocal mode.

2 Measurement methodology

The investigated sample is an artificial diamond substrate with NV-centers. The dimensions of the sample are $3 \times 3 \times 0.5$ mm^3. To evaluate the PL spectra of the sample, we used Raman spectroscopy in PL mode for different positions as well as bottom side of the sample. We used an Abberior Instrument STED microscopy system to obtain a high-resolution NV-center image. The STED microscope is equipped with a pulse laser line with wavelengths of 561 nm and 775 nm for excitation and saturating depletion of the excitation state, respectively. A vortex phase plate is inserted into the optical path of the 775 nm laser to generate a donut shaped beam. Both laser beams are overlapped and focused onto the sample using an oil-immersion objective with NA=1.4. Finally, the emission signal of the NV-center is recorded under a reflection confocal detection.

3 Measurement results

For different positions on the sample surface as well as from the bottom of the sample, PL spectra were measured and depicted in Figure 1. The photoluminescence measurement shows that there are two peaks at λ=575 nm and λ=637 nm for the neutral charge state of NV-center (NV^0) and the negative charge state of NV-center (NV^-), respectively. These peaks show a zero-phonon line (ZPL) at the room temperature where purely electronic transition occurs. In addition, the defect when

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excited by green light (532 nm) gives broad fluorescence emission in the spectral range from about 550 nm to 800 nm and a maximum around 700 nm. The result is in good agreement with the typical emission wavelength spectrum of NV-centers [8] and it shows that the NV-centers are distributed evenly over the sample.

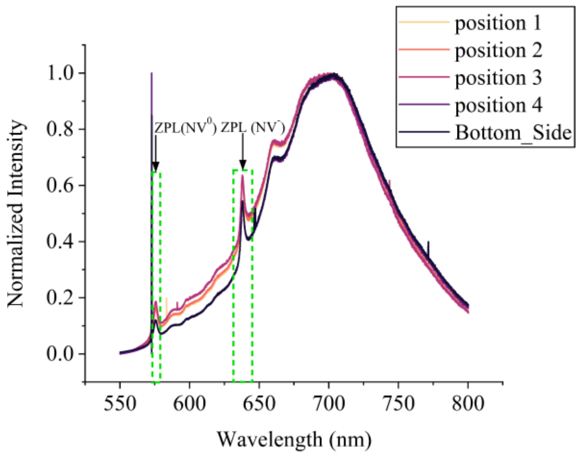

Fig. 1. PL spectra for different positions on the sample surface as well as the bottom of the sample. Fluorescence emission from NV-center showing two peaks at wavelengths 575 nm (NV^0) and 637 nm (NV^-) for different positions. Green dotted boxes show ZPL for (NV^0) and (NV^-) separately.

As a very first test, we used the STED microscopy system to image a random feature on the substrate surface. Figure 2 shows the image of the features in confocal and STED mode. The arrow in the figure shows the center of each feature and frame size of image is $4.5 \times 2\ \mu m^2$ with pixel size 10 nm.

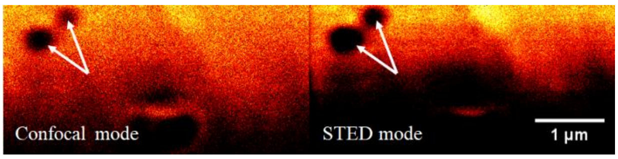

Fig. 2. Image of the random features in confocal mode (left) and STED mode (right).

Figure 3 shows the intensity profiles of a cross-section through the features. There is a drop in fluorescence emission intensity around border of features in STED mode. Dotted lines in STED mode diagram show the areas where the intensity drops to zero. These areas in the confocal mode diagram are not clear with compared to STED mode diagram. Thus, two features that are close to each other can be resolved in the STED mode. Moreover, our results show that the border of the features has a proper contrast in STED mode. Therefore, this feature of STED mode is promising for the dimensional metrology of nanostructure applied to the substrate with NV-centers.

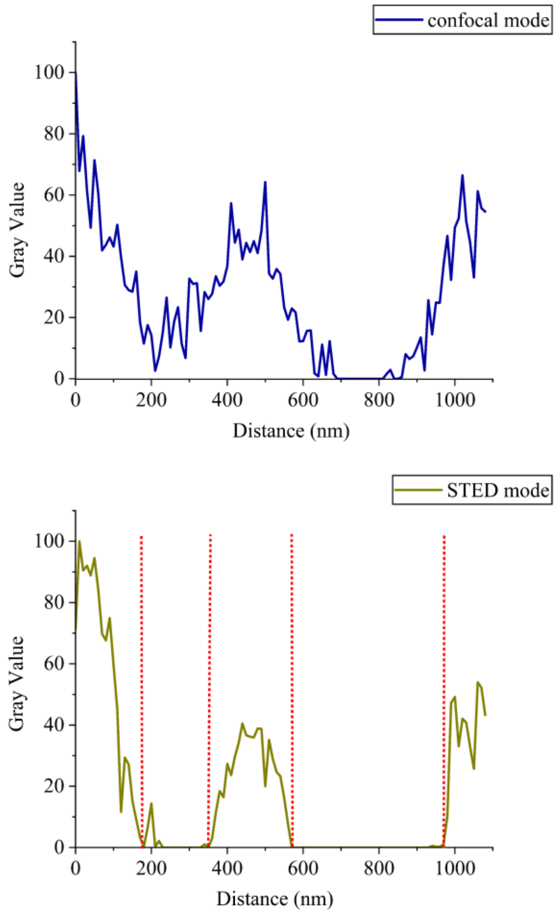

Fig. 3. Intensity profile of the random features for confocal mode (top) and STED mode (bottom). Dotted lines in STED mode diagram show the approximate border of each feature.

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