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Localization microscopy of single photon emitters in locally strained monolayer semiconductor

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Abstract. Integration of single photon emitters with nanophotonic structures on a chip is key for the development of future quantum optoelectronic devices. Here we study the formation of single photon emitters in a WSe_2 monolayer by local nanoindentation with an atomic force microscope probe. Using the bichromatic photoluminescence-imaging approach, we define the spatial locations of single photon emitters with deep sub-wavelength accuracy.

Keywords: single photon emitter, two-dimensional materials, nanophotonics, quantum optics

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Локализационная микроскопия источников одиночных фотонов в локально деформированных монослоях полупроводников

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Аннотация. Интеграция однофотонных излучателей с нанофотонными структурами на оптическом чипе является ключевой задачей в области разработки будущих квантовых оптоэлектронных устройств. Здесь мы изучаем источники одиночных фотонов в монослое WSe_2, сформированные методом локальной деформации с помощью зонда атомно-силового микроскопа. Используя метод бихроматической фотолюминесцентной визуализации, мы определяем пространственное расположение однофотонных излучателей с высокой точностью в глубоком субволновом масштабе.

Ключевые слова: Источник одиночных фотонов, двумерные материалы, нанофотоника, квантовая оптика

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Introduction

Single photon emitters (SPEs) are important elements for applications in quantum communication and computing devices [1]. One of the promising platforms for creating single photon emitters is provided by two-dimensional transition metal dichalcogenides (TMDs) [2]. Deformation of a two-dimensional material, for example, by the probe of an atomic force microscope (AFM), can lead to the formation of SPEs in TMD monolayers [3, 4]. The practical advantage of this approach is the possibility of forming arrays of emitters in specified locations on the chip due to the precise positioning of the AFM probe and the integration of emitters with nanophotonic structures on the chip. However, the integration accuracy is limited by the size of the nanoindent. In this work, we form arrays of single photon sources by the method of local deformation, and then investigate their optical properties and localization. The results of our work provide a broader understanding of the single photon sources in two-dimensional semiconductors and the possibility of their practical application in quantum technology devices.

Materials and Methods

In this work, we study the formation process of SPEs in a WSe_2 monolayer by local deformation with an AFM probe. Our experimental sample is SiO_2(1 μm)/Si substrate with silver alignment marks on the surface, covered with a thin layer of polymethylmethacrylate (PMMA) polymer. We obtain a monolayer of WSe_2 by mechanical exfoliation and transfer it to the polymer using a dry transfer. The array of single photon emitters is fabricated in the WSe_2 monolayer by the method of local deformation by the AFM probe. We modify the tip of the probe by removing the sharp end with a focused ion beam so that the tip is a plane with lateral dimensions of about 500 nm. This allows us to avoid premature rupture of the WSe_2 monolayer. Then we deform the WSe_2 monolayer and create nanoindents with a depth of 100–200 nm and a lateral size of about 500 nm (schematically shown in Fig. 1, *a*). At the same time, the WSe_2 monolayer does not relax and retains its shape due to the adhesion forces with the PMMA substrate. Fig. 1, *b* shows the redshift of exciton states in a WSe_2 monolayer due to lattice deformation, and the process of hybridization of a dark exciton with an atomic defect, which is the most likely mechanism for the formation of a single photon source.

Next, we study the optical properties of the obtained emitters at cryogenic temperatures (7 K). To do this, we use the methods of photoluminescence spectroscopy, bichromatic photoluminescence imaging approach [5, 6] and time resolved spectroscopy. In our work, for pumping we used a continuous HeNe laser with a wavelength of 632.8 nm, or a pulsed laser with filter that regulates the wavelength and width of the laser line, and a pulse frequency of 60 MHz. The collection and pumping of emission was carried out by a 50X objective with a numerical aperture NA = 0.65.

Results and Discussion

Quantum emitters are created and localized at the nanoindents. Fig. 1, *c* shows a typical photoluminescence spectrum of a deformed WSe_2 monolayer near a nanoindent. Bright peaks are clearly visible on the spectrum, they also appear around other nanoindents, and the emission range varies from 720 to 800 nm. Using polarization filtering, we found that they are linearly polarized with a degree of linear polarization from 40 to 80%. We note, that brightness of the formed emitters varies in the range from 0.5 to 3 MHz. We check the single-photon character of the manufactured emitters using measurements of the second-order correlation function, obtaining the value of the second-order correlation function at zero delay $g^{(2)}(0)$ less than 0.2 for most of the studied emitters. Fig. 1, *d* shown a characteristic graph of the second-order function of one of the emitters. Fit function (red curve) gives a value of $g^{(2)}(0) = 0.067 \pm 0.039$.

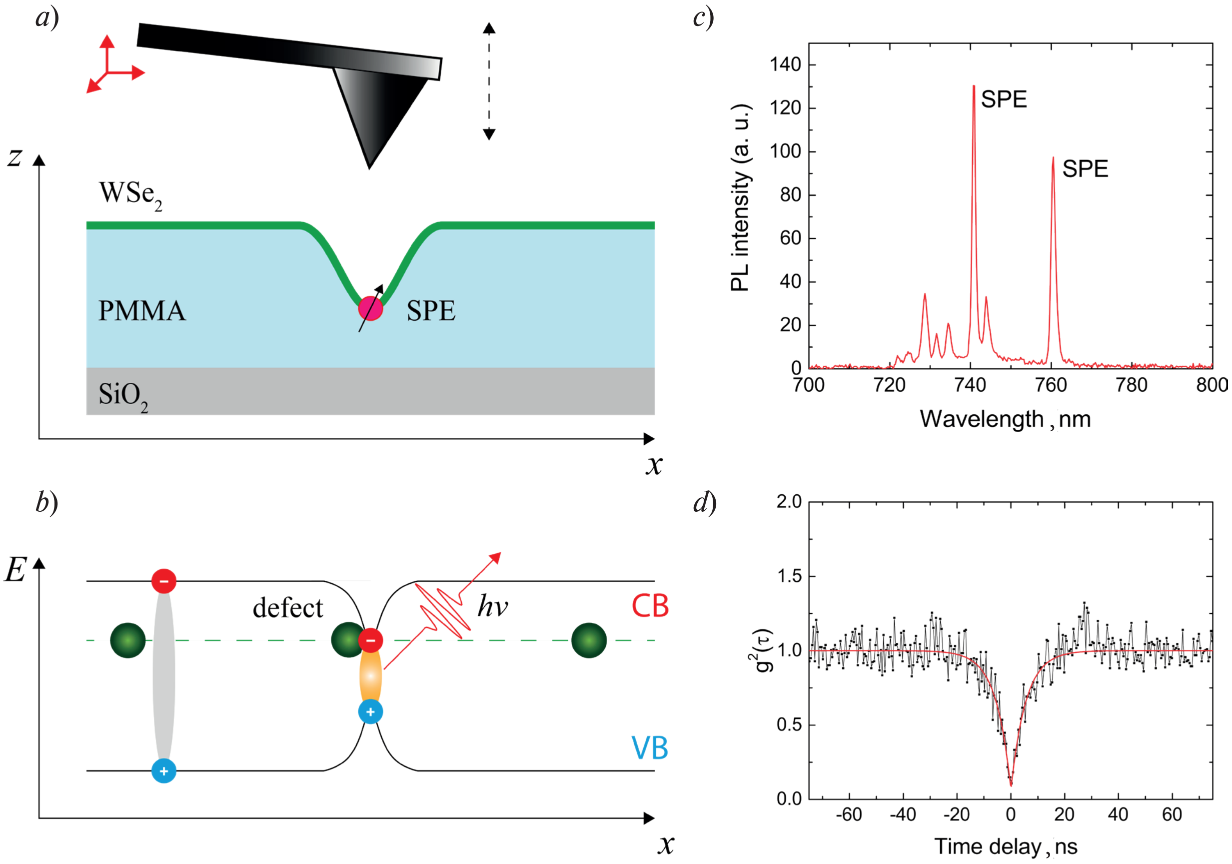

Fig. 1. Concept image of SPE fabrication approach based on nanoindentation of WSe$_2$ monolayer with an atomic force microscope probe (*a*). Diagram showing the process of hybridization of a dark exciton with an atomic defect of the WSe$_2$ crystal lattice (*b*). A typical photoluminescence spectrum near a nanoindent in a region with a diameter of about 1.5 μm (*c*). Experimentally measured second order correlation function for the PL signal from one of SPE. $g^{(2)}(0) = 0.067 \pm 0.039$

To determine the location of quantum emitters, we selected only those emitters whose radiation peaks we were able to completely filter out from a wider background using spectral and polarization filtering. The bichromatic PL-imaging approach consists in simultaneous illumination of the sample with 632.8 nm HeNe laser to excite the photoluminescence of the SPE and with light with a longer wavelength to illuminate the alignment marks. Reflected light and PL of SPE are imaged on the CMOS camera (Fig. 2, *a*). The alignment marks are also visible on the AFM map due to the swelling of the PMMA above them (Fig. 2, *b*). The coordinates of the centers of the alignment marks and the emitters were obtained from orthogonal linear scans of the AFM map and the optical image using Gaussian function fitting (Fig. 2, *c*). The exact position of the SPE is defined by converting the coordinates of the alignment marks and the SPE from the optical image to the AFM map of the structure. On average, the uncertainty of the SPE position obtained from a series of images was less than 60 nm.

Unexpectedly, we found that quantum emitters are formed from the outer edges of the nanoindents, and not inside them. Figure 2, *d* shows an AFM map with the location of one of the SPE. The locations near the vertices of the triangular imprint of the probe is also characteristic of other emitters. We assume that the use of a probe of a different shape may allow us to form emitters in a more controlled way in the WSe$_2$ monolayer.

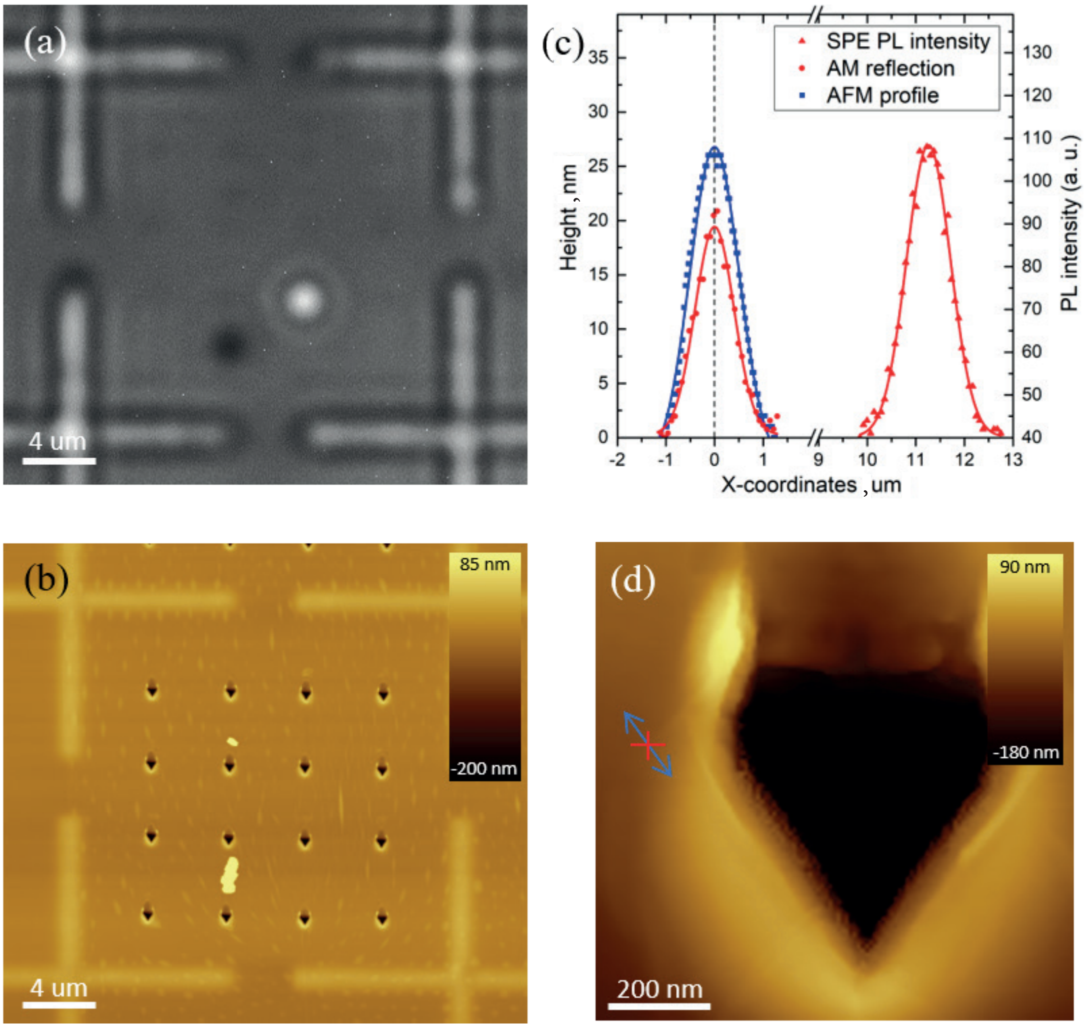

Fig. 2. Optical image of the photoluminescence from a single SPE and reflected light by the alignment marks (*a*). AFM scan of a sample with an array of nanoindents in WSe_2 monolayer and alignment marks (coincides with Fig. 2, *a*) (*b*). Orthogonal line cuts (*x*-axis) of the photoluminescence image, showing the profile of the SPE emission (red triangle symbols) and of the image of the alignment mark (red square symbols) and their Gaussian fits (red lines). Orthogonal line cut of the AFM scan, showing the profile of the alignment mark (blue square symbols) and its Gaussian fit (blue line) (*c*). The exact position of the SPE near the indent (the size of the cross shows uncertainty). The blue arrow shows the polarization of the SPE (*d*)

Conclusion

In this work, we have study the process of forming single photon sources by the method of local deformation. We have determined that the single photon sources obtained have a high degree of linear polarization and a high degree of single photon purity. We also note that in the process of nanoindentation, quantum emitters are randomly formed around the outer edges of the nanoindentation. The results of our research work are important for the practical application of the local deformation method in the creation of single photon generators for quantum communication and computing devices and especially for the following works on the integration of single-photon emitters with nanophotonic structures on a chip.

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