Enhanced second-harmonic generation in Mie-resonant MoS_2 nanodisks

A.A. Popkova[1], I.M. Antropov[1], G.I. Tselikov[2], F. Bedu[3], I. Ozerov[3], A.V. Arsenin[2], V.S. Volkov[2], V.O. Bessonov[1,4], A.A. Fedyanin[1]

[1] *Faculty of Physics, Lomonosov Moscow State University, Moscow, Russia*
[2] *Center for Photonics and 2D Materials, Moscow Institute of Physics and Technology, Dolgoprudny, Russia*
[3] *Aix Marseille University, CNRS, CINAM, Marseille, France*
[4] *Frumkin Institute of Physical Chemistry and Electrochemistry, Russian Academy of Sciences, Moscow, Russia*
popkova@nanolab.phys.msu.ru

Abstract: Enhanced second-harmonic generation in a single MoS_2 nanodisk due to the overlap of Mie resonances at the fundamental wavelength with the C-exciton resonance at the second-harmonic wavelength is observed.

1. Introduction

Molybdenum disulfide (MoS_2) and other transition metal dichalcogenides (TMDCs) are promising two-dimensional materials possessing many interesting properties for optics and opto-electronics applications, such as ability of tuning their carrier densities on demand by field-effect [1], high charge-carrier mobility [2], direct band gap in monolayer crystals [3], photoluminescent emission in the visible-NIR spectral range [4], and high nonlinear optical susceptibilities [5]. These properties make TMDCs excellent candidates for creating optical modulators, switches, frequency converters, transistors [6], and other devices. One of the key features of TMDC monolayers is the presence of a stable exciton states even at room temperature [7], which make them attractive for various optoelectronic and photonic applications, such as phototransistors [8] and heterojunction solar cells [9], where the strong light-matter interaction is required. High refractive index of TMDCs [14] enables creating optical resonators from them that support Mie resonances [10], coupling of which with excitons further enhances the light-matter interaction.

TMDCs monolayers have a large quadratic nonlinear susceptibility and are considered as materials for entangled photon pair generation required for modern quantum technologies. Nonlinear optical properties of the MoS_2 are studied well [5]. It has been demonstrated that the bulk MoS_2 crystal has a centrosymmetric lattice structure [11] and the second-harmonic generation (SHG) is forbidden in the dipole approximation for films with an even numbers of layers [12]. At the same time SHG efficiency strongly depends on the pump wavelength demonstrating resonances corresponding to the A-, B- and C-exciton resonances at the second-harmonic wavelength [5, 13]. In this work, we use the possibility of tuning Mie resonances by selecting the geometrical sizes of MoS_2 nanodisks to enhance SHG due to localization of the pump radiation at a frequency corresponding to the C-exciton resonance at the second-harmonic wavelength.

2. Results and discussion

We designed MoS_2 nanodisks in such a way that the anapole-like mode is excited inside the disk by the pump radiation at a wavelength of about 900 nm, corresponding to twice the C-exciton resonance wavelength. Arrays of MoS_2 disks were fabricated by reactive ion etching of the 110 nm thick exfoliated flake through the mask created by electron beam lithography. The optical image of the sample is shown in Fig. 1(a). The real sizes of the disks were measured using an atomic force microscope (AFM) (NT-MDT NTEGRA) working in a semicontact mode. The diameter and height of resonators are 550 nm and 110 nm, correspondingly. The typical 3D profile of a disk is shown in Fig.1 (a). A non-etched MoS_2 thin film several nm thick between the cylinders was used as a reference. SHG was measured using home-built multiphoton microscope designed for optical harmonics generation in reflection scheme. The 150-fs laser pulses with 80 MHz repetition rate and wavelength tuning in the 680-1080 nm range were used as a pump radiation. The laser beam was focused on the sample by an objective lens (NA=0.95, 50x) into a 2-um spot. The reflected SHG signal was collected by the same objective lens and detected by a monochromator and a CCD camera.

Fig.1 (b) shows the quadratic dependence of the SHG signal on the pump power. The spectra of the nonlinear signal do not contain any features that confirms the absence of the contribution of luminescence and other multi-photon processes. The inset at the top of Fig.1 (b) shows the SHG map obtained for a pump wavelength of 880 nm,

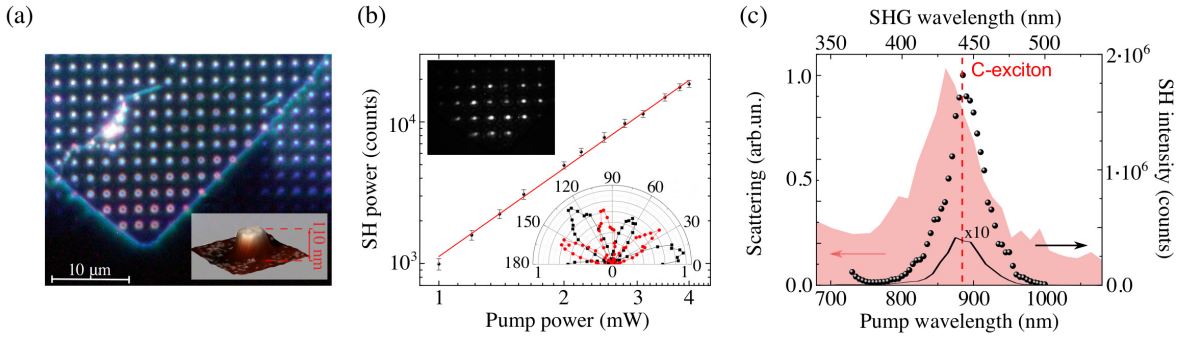

Fig. 1. (a) Dark field image of the sample. Inset shows the AFM image of a single disk. (b) Power-dependent nonlinear optical response of MoS_2 disk (black dots), plotted in double logarithmic scale, and its approximation by power function $y = y_0 + Ax^p$, where $p = 2.06 \pm 0.03$. Up inset is a SHG image of the sample. Down insert is an anisotropy dependence of SHG signal for parallel (red dots) and perpendicular (black dots) orientation of polarizer and analyzer. (c) Scattering cross-section (coral area) and SHG spectrum (black dots) of MoS_2 nanodisk and SH signal from reference sample (black line). The red dashed line shows the position of the C-exciton peak.

where nanodisks are clearly visible. The SHG power from MoS_2 nanodisks is 50 times higher than that from the reference MoS_2 film between the disks. The inset at the bottom of Fig.1 (b) depicts the anisotropy dependence of SHG signal obtained from a single MoS_2 nanodisk. The dependence demonstrates 6-fold symmetry corresponding to the one for MoS_2 monolayer, which indicates that the nanodisk has a single crystal structure.

To assess the effect of Mie resonances on SHG, we simultaneously measured the SHG and pump scattering spectra for a single nanodisk (Fig.1 (c)). The intensity of SHG has sharp resonance near 445-nm wavelength that corresponds to the C-exciton resonance position [5]. At the same time, the maximum efficiency of scattering is observed near 850-nm pump wavelength. Theoretical calculations using multipole decomposition demonstrate that this resonance is the result of electric and magnetic dipole modes superposition that leads to a high localization of the pump electric field inside the disk volume. The black line in Fig.1 (c) shows the SHG intensity spectrum for the MoS_2 film between the cylinders, in which C-exciton resonance is observed with an amplitude 50 times less than for the case of MoS_2 nanodisk.

3. Conclusion

We have demonstrated the possibility of enhancing the nonlinear optical response of MoS_2 by creating nanodisks that support Mie-type resonance modes at the pump wavelength corresponding to the doubled C-exciton resonance wavelength. The maximum value of the SHG intensity for the MoS_2 disk is an order of magnitude higher than that for the unstructured MoS_2 film.

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