Enhancement of second harmonic generation in a layered MoS$_2$ nanoresonator

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Abstract. Molybdenum disulfide (MoS$_2$) is a layered material with a high refractive index in the visible and infrared spectral range. In this work, we theoretically and experimentally demonstrate Mie-resonant MoS$_2$ nanodisks. We show enhanced second harmonic generation from MoS$_2$ nanodisk resonators due to the overlap of Mie-type resonances at the fundamental wavelength with the C-exciton resonance at the second-harmonic wavelength.

1. Introduction

Unique optical properties of 2D TMDCs materials such as strong photoluminescence [1, 2], valley polarization [3], excitonic valley coherence [4], high refractive index [5], and giant nonlinear response [6], enable these materials to be used as building blocks for integrated photonics. Molybdenum disulfide is a prominent representative of TMDCs. It is a semiconductor with indirect bandgap in bulk form, but monolayer form is a direct bandgap semiconductor [1]. Theoretical and experimental studies show that optical absorption are dominated by excitonic transitions in MoS$_2$, where the exciton binding energy can be close to 1 eV due to dramatically enhanced electron–hole and electron-electron interactions in monolayers [7]. Nonlinear optical (NLO) properties of MoS$_2$ layers are studied well [8–11]. MoS$_2$ has a centrosymmetric lattice structure in bulk form [12] and for an even number of layers, which leads to the elimination of SHG [11]. On the other hand, the strong SHG arises in the MoS$_2$ monolayer or multilayers with an odd number of layers [10,11]. Second harmonic (SH) intensity can be strongly increased by orders of magnitude when excitation photons or SH photons are resonant with the A-, B-, C-excitons in MoS$_2$ monolayers [12, 13]. Another way to enhance SHG is to employ Mie-type resonances for structured objects based on MoS$_2$. MoS$_2$ Layers possess high and anisotropic dielectric constants [14,15], which opens up the possibility of utilizing them to construct nanoantennas supporting distinct Mie-type resonances [16,17] and consider them as a novel platform for high-refractive-index all van der Waals nanophotonics [18]. Nanoresonators based on TMDCs with optical response governed by Mie-type resonances can enhance the efficiency of NLO processes including SHG [17].

In this work, we study the SHG response of an individual MoS$_2$ disk-like nanoresonator. We
observed the strong enhancement of SHG near 900-nm pump wavelength corresponding to the C-exciton resonance at the SH wavelength. Numerical calculations show that this enhancement is associated with the localization of the pump field inside the resonator due to the excitation of the magnetic dipole resonance.

2. Results and discussion
First, we estimated the parameters of nanoresonators so that the magnetic dipole (MD) Mie-type resonance at the excitation wavelength matched with C-exciton resonance in a monolayer of MoS$_2$ at SH wavelength. Then we fabricated an array of disk resonators by electron beam lithography followed by reactive ion etching. The cleaved silicon wafer with a 290-nm thermally grown oxide was used as a substrate. MoS$_2$ flakes were deposited on substrates using mechanical exfoliation from highly oriented synthetic 2H-phase MoS$_2$ crystal. Fig.1(a) shows a sketch of experimental sample. The real sizes of the nanoresonators were determined by scanning electron microscopy (SEM) and atomic force microscopy (AFM). The diameter and height of resonators are 550 nm and 110 nm, correspondingly. Fig.1 (b),(c) demonstrate SEM and AFM images of the nanoresonators, respectively. Non-etched remnants of MoS$_2$ flake approximately 5 nm thick between the nanoresonators was used as a reference.

Figure 1. Images of experimental sample (a) Sketch of the sample. (b) SEM image of the sample. (c) AFM profile of a single nanodisk.

SHG was measured using home-built multiphoton microscope designed for optical harmonics investigation 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, passing through the system of the power and polarization control, was focused in the back focal plane of an objective lens (NA=0.95, 50x), which corresponds to the Kohler illumination scheme with a 12 µm beam size at the sample. Reflected SHG signal from a large area formed a SH image of the nanoresonators on a CMOS camera (Photometrics Prime). Fig.2 (a) shows a SH image of the nanoresonators for Kohler illumination when the excitation wavelength is 900 nm. Each bright spot corresponds to one MoS$_2$ nanoresonator. The power dependences of the nonlinear response were measured to determine the nature of measured signal. For this the signal from sample area corresponded to a single disk was integrated for various pump powers. The obtained dependence is depicted in Fig. 2 (b) in double logarithmic scale and demonstrates the quadratic dependence 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.

To determine the effect of Mie-type resonances on SHG, we measured the SH signal dependence on the pump wavelength (Fig.2 (c)). The intensity of SHG has a sharp resonance near 445-nm wavelength that corresponds to the C-exciton resonance position [13]. A similar signal behavior is also observed for the reference sample with the response value being approximately 50 times smaller. The numerical simulation of scattering cross-section shows the MD resonance in the disk plane near the resonant wavelength for the experimental sample parameters (see Fig. 3 (a)). The corresponding distributions of the components of the electric...
Figure 2. SH characterization (a) SHG image of the sample. (b) Power-dependent nonlinear optical response of MoS\(_2\) disk (black dots), plotted in double logarithmic scale, and its approximation by power function \(y = y_0 + Ax^p\) with \(p = 2.06 \pm 0.03\). (c) SHG spectrum (black dots) of the MoS\(_2\) nanodisk and the reference sample (black line). The red dashed line shows the position of the C-exciton peak.

and magnetic fields in the resonator are shown in Fig.3 (b). The field localization in the disk volume leads to an increase in the efficiency of nonlinear effects which in our experiment is manifested in the form of SHG enhancement.

Conclusion
We have demonstrated that the MoS\(_2\) nanodisk resonators fabricating by electron beam lithography followed by reactive ion etching enhance the nonlinear optical response. This enhancement is associated with the excitation of the 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\(_2\) nanoresonators is 50 times of magnitude higher than that for the unstructured MoS\(_2\) film. This work opens a new route to control and modulate nonlinear response of Mie-resonant nanostructure based on 2D TMDCs materials by tailoring...
their excitation conditions. It may be applied for nonlinear 2D photonics and holographic imaging applications.

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