Proceedings Paper:

Benimetskiy, F.A., Kravtsov, V., Khestanova, E. et al. (7 more authors) (2020) Strong coupling of excitons in 2D MoSe2/hBN heterostructure with optical bound states in the continuum. In: Journal of Physics: Conference Series. METANANO 2019, 15-19 Jul 2019, St. Petersburg, Russian Federation. IOP Publishing .

https://doi.org/10.1088/1742-6596/1461/1/012012

PAPER • **OPEN ACCESS**

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To cite this article: F. A. Benimetskiy *et al* 2020 *J. Phys.: Conf. Ser.* **1461** 012012

IOP Conf. Series: Journal of Physics: Conf. Series **1461** (2020) 012012 doi:10.1088/1742-6596/1461/1/012012

Strong coupling of excitons in 2D $MoSe_2$/hBN heterostructure with optical bound states in the continuum

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Abstract. We experimentally demonstrate strong exciton-photon coupling in a $MoSe_2$/hBN heterostructure interfaced with an all-dielectric metasurface supporting high-Q bound states in the continuum. The resulting exciton-polaritons are probed by means of temperature- and angle-resolved reflectivity and photoluminescence. Our findings pave the way towards new-generation nonlinear planar polaritonic devices.

1. Introduction

Over the last decade, the field of two-dimensional layered materials has seen rapid progress. Monolayers of transition-metal dichalcogenides (TMDCs) such as WS_2, MoS_2, WSe_2 and $MoSe_2$ have attracted strong interest as they exhibit direct bandgaps [1] and can be used as basic elements in compact optoelectronic and all-optical devices. The realisation of latter systems requires strong nonlinearity, which can be dramatically increased, when realized in materials supporting strong coupling between photons and solid state excitations. One of the most promising ways to achieve efficient photon-photon interactions at low pump intensities is to exploit the strong exciton-photon coupling, resulting in the emergence of so-called polaritons, being half-light half-matter quasiparticles [2]. In this case effective optical nonlinearity is mediated by strong exciton-exciton interactions in the solid state.

Excitons in TMDCs are characterized by large oscillator strength and binding energies (200-500 meV for neutral excitons, and 30 meV for trions) [1]. Those properties of two-dimensional semiconductors and possibility to create compact planar structures integrated with them pave the way towards a wide range of practical applications.

Strong coupling in nanostructures with semiconducting two-dimensional layered materials has been observed in different systems such as all-dielectric microcavities, metal-based microcavities, and plasmonic structures [3]. Most common polariton system both for traditional and for two-dimensional semiconductors are based on various realizations of Fabry-Perot cavities [4, 5, 6, 7, 8]. However, in bulk microcavities not all advantages of the two-dimensional nature of TMDC are not employed. One of the alternative approaches that may unleash the potential of these materials is polariton systems based on dielectric photonic crystals (PCs) or metasurfaces [9]. In particular, structures supporting high-Q modes called optical bound states in the continuum (BICs) are of special interest [10]. Here we focus on an experimental investigation of exciton-polaritons in PCs-TMDC system where strong coupling occurs between the exciton and at-Γ BIC mode.

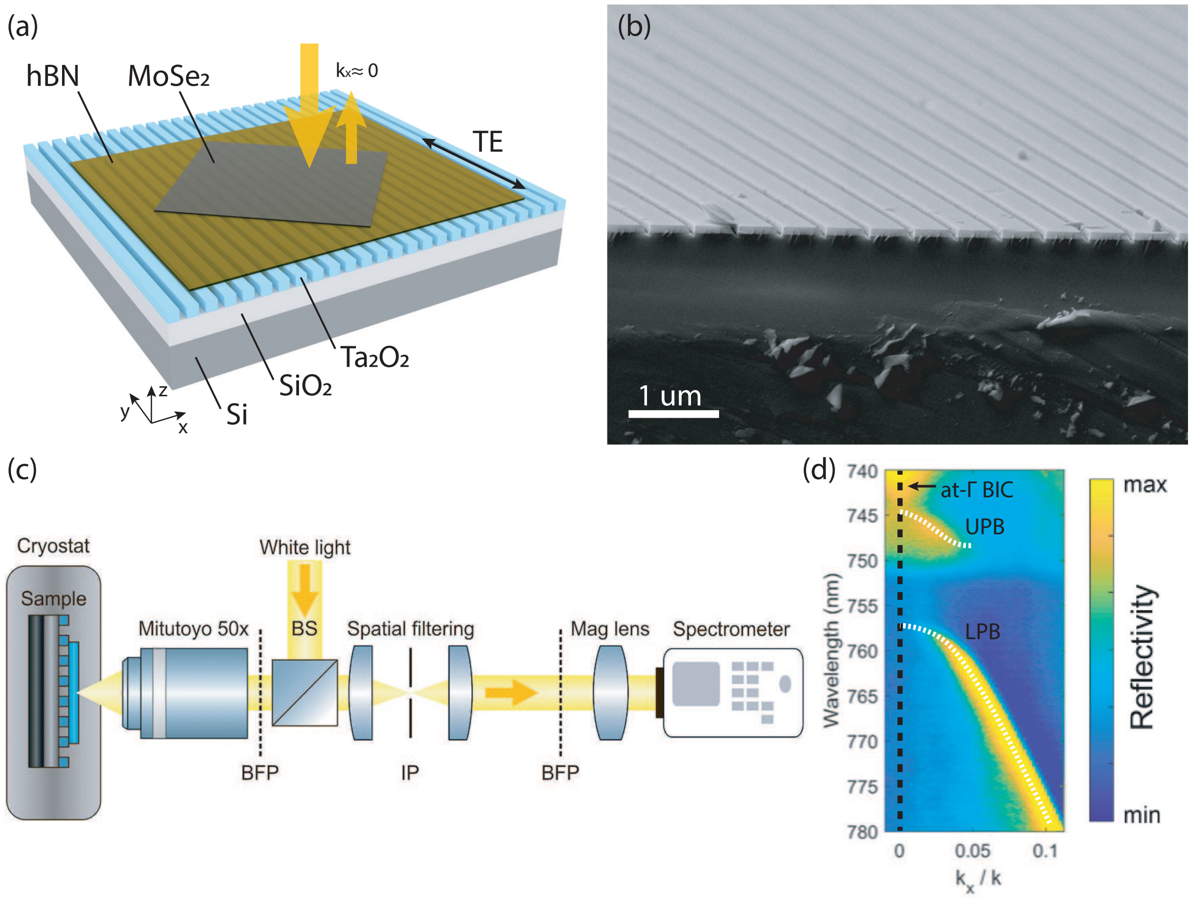

Figure 1. (a) Schematic of the Ta_2O_5 subwavelength grating with a $MoSe_2$/hBN heterostructure placed on top. (b) A side-view SEM image of the bare grating. (c) Schematic of the experimental setup for microphotoluminescence and microreflectance and Fourier plane imaging combined with a closed-cycle helium cryostat. BFP: Back focal plane. IP: image plane. The slit placed in the image plane stands of a spatial filter. Strong coupling between TMDC exciton and BIC mode measured by angle-resolved reflectance at 10 K.(d) Angle-resolved reflectance spectra of the PCs-TMDC structure, showing splitting of the mode into upper and lower polariton branches (UPB and LPB).

2. Results and discussion

In our experiment, $MoSe_2$/hBN heterostructure was integrated with a tantalum pentoxide (Ta_2O_5) sub-wavelength grating structure. The schematic of the structure is shown in Fig. 1a. The substrate containing grating has three layers: Ta_2O_5 - 90 nm, SiO_2 - 1 um and bottom layer is Si. The grating period is equal to 480 nm, the fill factor is around 0.6 and etching depth is 90 nm (Fig. 1b).

To demonstrate exciton-photon coupling in PCs-TMDC structure, we use the setup for microphotoluminescence spectroscopy in Fourier plane imaging configuration (Fig. 1c). The setup is integrated with a closed-cycle helium cryostat allowing to perform optical measurements at cryogenic temperatures down to 6.5K. Here, the incident white light of a tungsten halogen lamp (Ocean Optics HL-2000, polarized along the grating that corresponds to transverse-electric (TE)) is focused at the sample surface be means of an objective lens (Mitutoyo Objective 50x/0.42). The reflected radiation is then collected by the same objective lens and projected to the entrance slit of the spectrometer (Princeton SP 2558, CCD camera PyLoN 400BR_eXcelon).

First, we characterized a bare grating without a heterostructure. The BIC mode quality factor Q near Γ point is about 550 ($\gamma_{BIC} = 3$ meV), twice higher than in recent experimental observation photonic-crystal exciton-polaritons in monolayer semiconductors [9]. Strong coupling between exciton in $MoSe_2$/hBN heterostructure and TE-polarized BIC mode supported by Ta_2O_5 grating becomes apparent in angle-resolved reflectance spectra. The anticrossing of two modes is clearly seen (Fig. 1d). Also, we characterised the heterostructure photoluminescence $\gamma_{exc} = 10$ meV. The sample is excited by a He-Ne laser beam linearly polarized across the grating. In this case high-Q PCs modes are not excited. To confirm the observation of strong coupling regime, we compare the measured dispersion curves with finite-difference time-domain simulation results . The achieved Rabi splitting is 17 meV.

3. Conclusions

In summary, we have experimentally demonstrated exciton-photon coupling in a $MoSe_2$/hBN heterostructure interfaced with a Ta_2O_5 subwavelength grating structure supporting high-Q optical bound state in the continuum. The resulting exciton-polaritons are characterized by means of angle-resolved reflectivity at 10 K. We systematically investigate the temperature and polarization dependence of the polariton dispersion and associated Rabi splittings in the hybrid metasurface/TMDC structures through complementing the experimental data by numerical simulations. We further discuss the role of high-Q bound states in the continuum modes supported by sub-wavelength gratings in the formation of strongly-coupled exciton-polaritons. Our results suggest a new platform for future TMDC-based polaritonic devices.

4. Acknowledgments

This work was supported by the Ministry of Education and Science of Russian Federation (project 14.Y26.31.0015).

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