Probing guided monolayer semiconductor polaritons below the light line

V. Kondratyev¹, D. Permyakov¹, V. Kravtsov¹, D. N. Krizhanovskii¹,², A. K. Samusev¹

¹ Department of Physics and Engineering, ITMO University, St. Petersburg, 197101, Russia
² Department of Physics and Astronomy, University of Sheffield, Sheffield S3 7RH, UK

E-mail: a.samusev@metalab.ifmo.ru
E-mail: valeriy.kondratiev@metalab.ifmo.ru

Abstract. In this work, we demonstrate an approach to study exciton-polaritons supported by transition metal dichalcogenide monolayers coupled to an unstructured planar waveguide below the light line. In order to excite and probe such waves propagating along the interface with the evanescent fields exponentially decaying away from the guiding layer, we employ a hemispherical ZnSe solid immersion lens (SIL) precisely positioned in the vicinity of the sample. We visualize the dispersion of guided polaritons using back focal (Fourier) plane imaging spectroscopy with the high-NA objective lens focus brought to the center of SIL. This results in the effective numerical aperture of the system exceeding an exceptional value of 2.2 in the visible range. In the experiment, we study guided polaritons supported by a WS₂ monolayer transferred on top of a Ta₂O₅ plane-parallel optical waveguide. We confirm room-temperature strong light-matter coupling regime enhanced by ultra-low intrinsic ohmic and radiative losses of the waveguide. Note that in the experiment, total radiative losses can be broadly tuned by controlling SIL-to-sample distance. This gives a valuable degree of freedom for the study of polariton properties. Our approach lays the ground for future studies of light-matter interaction employing guided modes and surface waves.

1. Introduction

Rapid development of the field of all-optical devices such as optical switches and transistors boosts the search for highly nonlinear optical systems. Those operating in the regime of strong light-matter coupling show great promise. Monolayers of transition-metal dichalcogenides (TMDs) such as WS₂, WSe₂, MoS₂ and MoSe₂ exhibit direct band gaps [1] and are suitable for chip integration [2]. TMD interaction with light is dominated by quasiparticles – excitons with binding energies of the order of 100 meV, large oscillator strength [3] and ability to strongly couple to optical cavities and form polaritons. TMD polaritons were studied in various optical systems, such as plasmonic cavities [4], Bragg mirrors [5] and subwavelength gratings [6, 7]. Most of the systems studied so far require relatively complicated fabrication processes, while their designs allow for radiative coupling to free-space waves with a fixed efficiency. In our work, we realize a simple hybrid planar TMD-based waveguide and demonstrate an approach for the study of polaritons intrinsically uncoupled from free-space waves propagating below the light line.
2. Results and discussion

In the experiment, we employ back focal (Fourier) plane spectroscopy setup combined with solid immersion lens (SIL) [8, 9] schematically shown in Fig. 1(a). We use a high numerical aperture objective lens (Mitutoyo, M Plan Apo HR, 100×, NA_{obj} = 0.9) in combination with ZnSe (refractive index n_{ZnSe} ≈ 2.5 in the visible range) prism coupled to the sample with a precisely controlled air gap in Otto geometry [10]. Such configuration enables the resulting numerical aperture of NA = NA_{obj}n_{ZnSe} ≈ 2.25 and allows for excitation and detection of propagating modes below the light line (see Fig. 1(e)). During the measurement, the sample was excited by a white light halogen lamp (Ocean Optics HL-2000FHSA). The angle-resolved reflectivity spectra were measured by a slit spectrometer (Princeton SP 2500, CCD camera PyLoN 400BRexcelon). The sample was attached to a piezo positioner, which allowed for precise control of the SIL-to-sample gap. The size of the gap was directly related to the radiative losses of the mode induced by SIL and thus allowed controlling mode excitation and detection efficiency.

The sample we investigated in this work consists of a WS\textsubscript{2} monolayer placed on a 90-nm Ta\textsubscript{2}O\textsubscript{5} waveguide on top of SiO\textsubscript{2}/Si substrate. The WS\textsubscript{2} monolayer was exfoliated from a bulk crystal and transferred on top of the Ta\textsubscript{2}O\textsubscript{5} layer. A schematic representation of our device with a SIL (ZnSe prism) attached is shown in Fig. 1(b).

The typical field profile and dispersion of a guided mode is shown in Fig. 1(d) and (e), respectively. For a non-leaky mode, the experimentally available in-plane wavevectors \kappa_{\parallel} satisfy the condition n_{SiO\textsubscript{2}} < k_{\parallel}/k_{0} < n_{ZnSe}, where k_{0} is the absolute value of free-space wavevector (see Fig. 1(c, e)). Since the mode field is exponentially decaying away from the waveguiding layer, it can be only coupled to free space by a high-index prism brought in a close vicinity to the sample.
Figure 2. Simulated (left column) and measured (right column) angle-resolved TE-polarized reflectivity maps for the sample with (bottom row) and without (top row) WS$_2$ monolayer. Red and blue regions correspond to the states above and below the light line in SiO$_2$. The energy splitting between exciton resonance in TMD and waveguide mode is observed at $\approx 2.01$ eV. The rectangle highlights the magnified region shown in Fig. 3.

Fig. 2 shows simulated (a, c) and measured (b, d) angle-resolved reflectivity maps for the sample without (a, b) and with (c, d) TMD monolayer obtained for a 150 nm air gap between sample and SIL. The simulations were performed using Fourier modal method [11]. The central part of the figure marked with red corresponds to small wavevectors $k_x/k_0 < n_{SiO_2}$ and contains strong Fabry-Pérot interference due to reflection from Si substrate. The remaining blue regions correspond to the regime of total internal reflection, while the dips in these regions are associated with guided TE-polarized modes.

The top row in Fig. 2 shows the dispersion of TE waveguide mode, which is efficiently coupled to free-space waves for an air gap between SIL and the bare waveguide of around 150 nm. When coupled to exciton resonance (Fig. 2, bottom row), the waveguide mode experiences Rabi splitting indicating the onset of strong light-matter coupling regime. Fig. 3 shows a magnified reflectivity map containing anticrossing of the modes. From this picture, we can estimate the Rabi splitting to be of the order of few tens of meV.

3. Conclusions
We have demonstrated a new approach for the study of guided exciton-polaritons in TMD-based planar photonic structures at room temperature. We have observed strong coupling between excitons in WS$_2$ monolayer and Ta$_2$O$_5$ waveguide with Rabi splitting of the order of several 10s of meV. Our results provide a basis for future investigations of radiative/non-radiative losses, lifetimes, and nonlinearities of TMD-based guided polaritons.
Figure 3. Magnified exciton-photon splitting region: simulation (a) and experiment (b). White dashed line is a guide for the eye. The same region is highlighted in Fig. 2.

4. Acknowledgments
The theoretical part of this study was supported by Russian Science Foundation, grant no. 21-12-00218. The experimental work was funded by RFBR according to project no. 19-52-51010.

References
[1] Mak K F and Shan J 2016 Nature Photonics 10 216–226
[2] Akinwande D, Huyghebaert C, Wang C H, Serna M I, Goossens S, Li L J, Wong H S P and Koppens F H 2019 Nature 573 507–518
[3] Wang G, Chernikov A, Glazov M M, Heinz T F, Marie X, Amand T and Urbaszek B 2018 Rev. Mod. Phys. 90(2) 021001
[4] Munkhbat B, Baranov D G, Bisht A, Hoque M A, Karpiak B, Dash S P and Shegai T 2020 ACS nano 14 1196–1206
[5] Dufferwiel S, Schwarz S, Withers F, Trichet A, Li F, Sich M, Del Pozo-Zamudio O, Clark C, Nalitov A, Solnyshkov D et al. 2015 Nature communications 6 1–7
[6] Zhang L, Gogna R, Burg W, Tutuc E and Deng H 2018 Nature communications 9 1–8
[7] Kravtsov V, Khestanova E, Beninetskiy F A, Ivanova T, Samusev A K, Sinev I S, Pidgayko D, Mozharov A M, Mukhin I S, Lozhkin M S et al. 2020 Light: Science & Applications 9 1–8
[8] Pidgayko D, Sinev I, Permyakov D, Sychev S, Heyroth F, Rutckaia V, Schilling J, Lavrinenko A, Bogdanov A and Samusev A 2018 ACS Photonics 6 510–515
[9] Permyakov D, Pidgayko D, Sinev I and Samusev A 2020 Measuring full complex dispersion of guided modes and surface waves in planar photonic structures AIP Conference Proceedings vol 2300 (AIP Publishing LLC) p 020096
[10] Otto A 1968 Zeitschrift füür Physik A Hadrons and nuclei 216 398–410
[11] Li L 1997 JOSA A 14 2758–2767