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Temperature-dependent exciton-polaritons in perovskite photonic crystal slab

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Abstract. Exciton-polaritons are perspective platform for realizing ultrafast and strong optical modulations, which are necessary for the plant of applications. However, exciton-polaritons are studied mostly for semiconductor quantum wells inside vertical Bragg cavities, which limits it to the cryogenic temperatures and prevents planar realizations, which can be a problem for real-world applications. Recently, perovskites become one of the perspective materials for room-temperature strong light-matter coupling regime due to their unique physical properties. In this work, we experimentally demonstrate for the first-time room-temperature exciton-polaritons in planar halide perovskite photonic crystal slab fabricated by a nanoimprint lithography method. We experimentally measured polariton dispersion from angle-resolved photoluminescence spectra and confirm the strong light-matter coupling regime at room temperature and lower. Also, we studied the temperature dependence of the exciton energy level in MAPbBr₃ and the light-matter coupling coefficient. The obtained dependences can be attributed to polaron effects in this material. The results can become the basis of further research on perovskite exciton-polaritons in planar photonic cavities.

Keywords: halide perovskites, exciton-polaritons, photonic crystal slab

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Материалы конференции

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Температурная зависимость экситон-поляритонов в перовскитной фотонно-кристаллической пластине

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Аннотация. В данной работе мы впервые экспериментально демонстрируем сильную связь свет-вещество в планарной фотонно-кристаллической пластине, сделанной из поликристаллической пленки органо-неорганического перовскита MAPbBr₃ методом наноимпринт литографии. В работе мы рассказываем о методах фабрикации, морфологии образца, а также демонстрируем экспериментально измеренный углоразрешенный спектр фотолюминесценции, содержащий ветки поляритонной дисперсии при комнатной температуре. Варьируя температуру образца, мы получили зависимость экситонного уровня, а также коэффициента связи свет-вещество от температуры.



Ключевые слова: органо-неорганические перовскиты, экситон-поляритоны, фотонно-кристаллическая пластина

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Introduction

One of the most critical problems in photonics is the searching for the material base with high optical nonlinearities in an ultrashort time. Part-light, part-matter quasiparticles exciton-polaritons represent a perspective platform to solve this problem, thanks to its hybrid properties. Since the first works about exciton-polaritons in GaAs quantum well (QW) implemented in a Bragg cavity [1], there already have been shown several concepts of polariton devices, such as polariton transistors, switchers, and lasers [2, 3]. Despite the huge progress in this field at this moment, most of the exciton-polariton designs are limited to cryogenic temperatures, due to low exciton binding energy. Also, large Bragg cavity vertical sizes are still the obstacle for the further development. To overcome it new materials with room-temperature exciton and planar photon cavities designs are needed.

One of the possible implementations of planar photon cavities, which can be experimentally studied, is leaky modes of a photonic crystal slab. This approach was already used for exciton-polaritons with monolayer of MoSe₂ [4]. However, 2D materials are very sensitive to defects and limited to monolayer lateral sizes. In our work, we present a photonic crystal slab based on the halide perovskite MAPbBr₃, fabricated by the nanoimprint lithography method. Such approach has several advantages: large scale of the polariton cavity (> 1 cm²) due to the perovskite synthesis methods and nanoimprint lithography [5]; room-temperature exciton in MAPbBr₃ around 40 meV [6], easy and cheap to fabricate, and huge cavity photon mode localization, because photonic cavity simultaneously plays the role of the exciton material.

In this work, we demonstrate for the first-time room temperature exciton-polariton dispersions in the planar photon cavity. We show the fabrication method of the MAPbBr₃ photonic crystal slab and its morphology. Also, we show the angle-resolved photoluminescence (PL) spectrum at room temperature with exciton-polariton branch, calculated from the two-coupled oscillator model [7], and the dependence of exciton level and Rabi splitting on the temperature, calculated from the noted model.

Materials and Methods

MAPbBr₃ perovskite is one of the most interesting materials for exciton-polariton systems because it has room-temperature exciton with high oscillator strength, defect tolerance, synthesis from solutions, enough softness for nanoimprint lithography and observed polaron effects, which affect exciton-polariton states [5].

First, we prepare the perovskite solution by dissolving 33.59 mg of MABr (Sigma Aldrich) and 110,1 mg of PbBr₂ (TCI) in the mixture of DMF:DMSO in a relation equal to 3:1. Resulting 0.3M solution is steered for 24 hours at room temperature. After we filter it by PTFE filter to avoid small crystallites.

Second, we wash SiO₂ substrates (12×12 cm²) with sonication in the deionized water, acetone and 2-propanol for 10 minutes consecutively, and afterwards clean it in an oxygen plasma cleaner for 10 minutes. Afterwards we transfer substrates in the dry glovebox with nitrogen atmosphere.

Next, we fabricate MAPbBr₃ polycrystalline films by spin-coating method in the nitrogen dry glovebox. (Fig. 1, a). We depose 40 uL of prepared MAPbBr₃ solution on the cleaned substrate

and place it on the spin-coater. After, it is spinning at 3000 rpm for 40 seconds. At the 20 second after the start we drip 300 uL of toluene, which plays the role of the antisolvent, on the top of the rotating substrate.

After the spinning we take out the substrate with MAPbBr₃ polycrystalline film from the glovebox for the nanoimprint lithography. We put a large-scale cleaned DVD disk mold on the top of the MAPbBr₃ film and apply 15 MPa pressure for 10 minutes. After we remove the mold and put the sample in the nitrogen atmosphere the further annealing at 90 °C for 10 minutes. The resulting sample slowly degrades under ambient conditions and therefore it is stored in the dry glovebox with an inert atmosphere (Fig. 1, a).

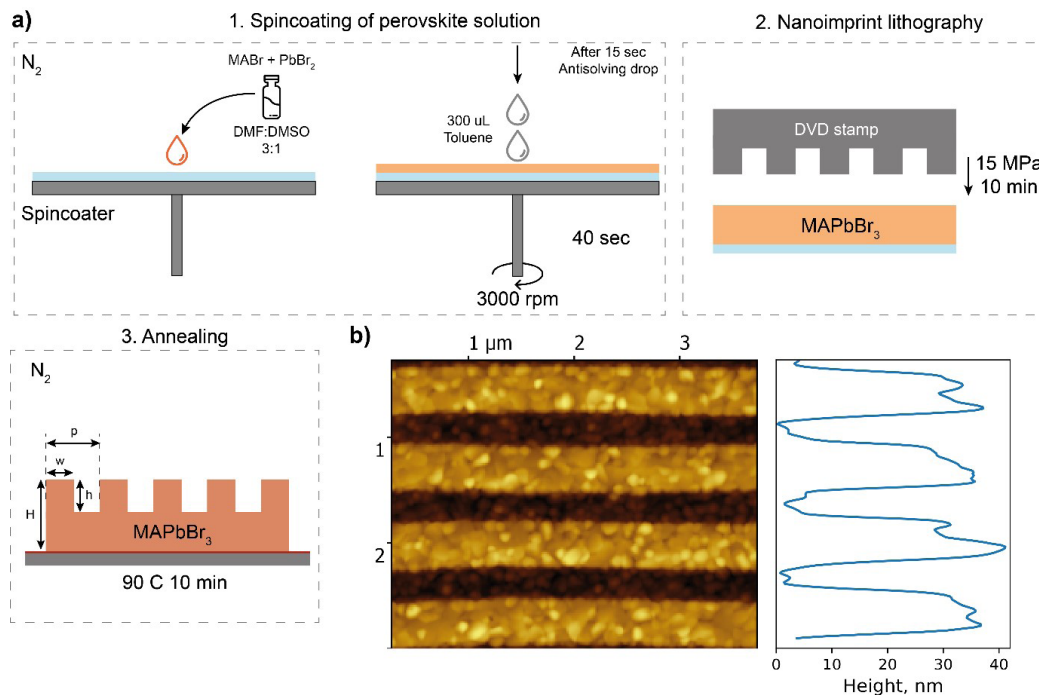


Fig. 1. Sketch of the main steps of the MAPbBr₃ crystal slab fabrication (a) AFM scan of the perovskite crystal slab morphology and its vertical profile (b)

We studied the resulting sample morphology by the atomic force microscopy (AFM) method (Fig. 1, b). The periodic structure has a pronounced rectangular shape with a period of 750 nm, a modulation depth of 35 nm, and a comb width of 450 nm. The roughness of the sample is lower than 5 nm, which points to the high quality of the resulting photonic crystal slab.

To confirm the strong light-matter coupling regime and measure exciton-polariton dispersion we used the angle-resolved spectroscopy method. The experimental setup is shown in Fig. 2. Femtosecond laser (220 fs) at 490 nm (Pharos + Orpheus-F, Light Conversion) was used as an incident pump of PL. A repetition rate of 10 kHz was chosen to avoid thermal effects. Half-wave plate and Glan prism were used to filter laser polarization and attenuate laser fluence. Lens L1 was used to focus incident emission to the back focal plane (BFP) of the objective (Mitutoyo NIR HR 50x with N.A. = 0.65) to realize the pump spot of 30 μm on the sample placed in the closed-cycle helium cryostat (Advanced Research Systems). A large pump spot for a high N.A. objective is required to pump the area with a large number of the grating period.

The emission from the sample gets into the objective and then into the 4f scheme, realized by the lenses L2, L3, and spatial filtering. The information on the emission angle distribution contains in BFP, which is imaged by dashed lines in Fig. 2. Lens L4, located at the focal distance from BFP, makes Fourier transform (focus each particular emission angle to its point on BFP), and then the BFP image goes to the slit spectrometer at $k_y = 0$, which is coupled to the imaging CCD (Princeton Instruments SP2500+PyLoN). Image from the spectrometer, coupled to the imaging CCD represents colormap with angle distribution on the one axis and light wavelength on the other (example in Fig. 3, a).

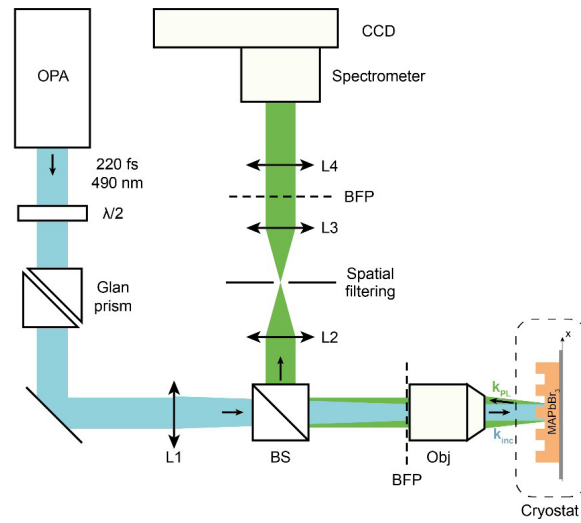


Fig. 2. Scheme of the experimental setup of angle-resolved PL measurements. L – Lens, BS – beam splitter, obj – objective, BFP – back focal plane

Results and Discussion

We measured the angle-resolved spectrum of the MAPbBr_3 photonic crystal slab, fabricated by the nanoimprint lithography method at room temperature (Fig. 3, *a*). We extract the polariton mode from the spectrum, determine an exciton level from the PL and reflection spectra, and estimate uncoupled cavity photon mode by linear approximation of the mode far from the exciton resonance. From the well-known two-coupled oscillator model, described strong light-matter regime (Eq. 1) [7], we fitted polariton dispersion (solid yellow line in Fig. 3, *a*) with varying parameter coupling coefficient g . At room temperature, the coupling coefficient is equal to 109.5 meV, which is much larger than the half-difference of exciton and cavity photon linewidths, which is around 4 meV, that confirms the strong light-matter coupling regime [7].

$$E_{LP}(k_x) = \frac{E_X + E_C(k_x)}{2} - \frac{1}{2} \sqrt{(E_X - E_C(k_x))^2 + g^2} \quad (1)$$

where E_{LP} , E_X , E_C are complex energy of lower polariton, exciton and uncoupled cavity photon respectively; g is coupling coefficient.

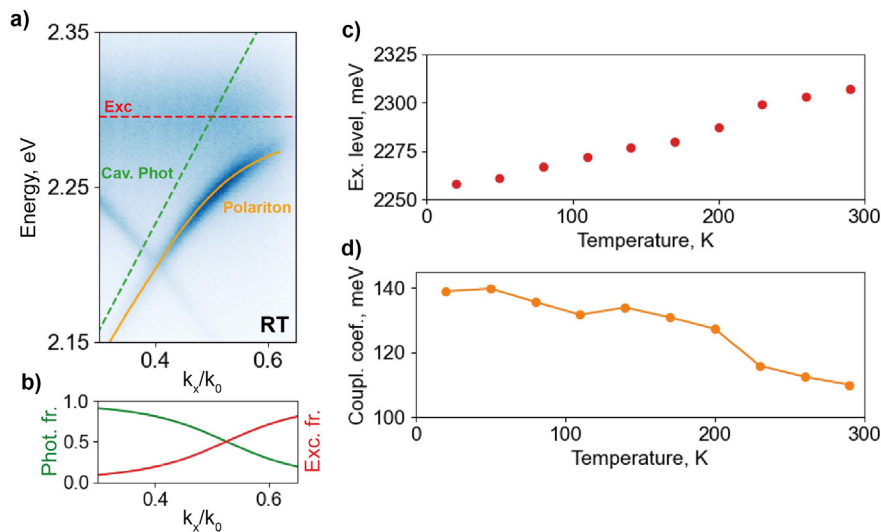


Fig 3. Angle-resolved PL spectrum measured at room temperature. Red dashed line – exciton level, green dashed line – uncoupled cavity photon dispersion, solid yellow line is fitted by two-coupled oscillator model polariton dispersion (*a*). Hopfield coefficients of polariton at room temperature, extracted from the model (*b*); Exciton level as a function of the temperature (*c*); Rabi splitting, calculated from the measured angle-resolved spectra and two-coupled oscillator model (*d*)

In Fig. 3, *b* Hopfield coefficients of the polariton branch are shown, which reveal to the photon and exciton fractions in polariton as a function of angle emission. There should be noted, that when the exciton fraction reaches the value around 0.7, polariton branch dissipates due to the strong exciton absorption. As the exciton and band-to-band absorption is strong, the upper polariton branch does not exist in the system.

With lowering the temperature we measured angle-resolved spectra and extracted from it exciton level and polariton coupling coefficients (Fig. 3, *c*, *d*). Shifting of the exciton level with lowering the temperature can be attributed to polarons in the perovskite [6]. In this work there are no other polaron effects, however, it should play a significant role under a polariton resonance pump [5]. Also, with lowering temperature coupling coefficient increases due to the increase of exciton concentration and decrease of the nonradiative losses in the system.

Conclusion

In our work, we experimentally demonstrate for the first-time exciton-polariton dispersion in the planar photonic crystal slab based on halide perovskite MAPbBr₃. Thanks to the unique properties of perovskites, we succeeded to realize the periodic structure on the polycrystalline perovskite film by nanoimprint lithography and confirm a strong light-matter coupling regime of exciton and photonic crystal slab leaky modes. We also reveal the dependence of the exciton level and light-matter coupling coefficients with temperature. These results confirm the opportunity for room temperature exciton-polaritons in the planar cavity and can form the basis of further perovskite exciton-polariton studies.

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