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Strong exciton–photon coupling with colloidal quantum dots in a tuneable microcavity

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ABSTRACT

Polariton emission from optical cavities integrated with various luminophores has been extensively studied recently due to the wide variety of possible applications in photonics, particularly promising in terms of fabrication of low-threshold sources of coherent emission. Tuneable microcavities allow extensive investigation of the photophysical properties of matter placed inside the cavity by deterministically changing the coupling strength and controllable switching from weak to strong and ultra-strong coupling regimes. Here we demonstrate room-temperature strong coupling of exciton transitions in CdSe/ZnS/CdS/ZnS colloidal quantum dots with the optical modes of a tuneable low-mode-volume microcavity. Strong coupling is evidenced by a large Rabi splitting of the photoluminescence spectra depending on the detuning of the microcavity. A coupling strength of 154 meV has been achieved. High quantum yields, excellent photostability, and scalability of fabrication of QDs paves the way to practical applications of coupled systems based on colloidal QDs in photonics, optoelectronics, and sensing.

Reversible coherent energy exchange between the exciton transition and electromagnetic field modes in an optical cavity leads to the formation of quasi-particles called polaritons when the rate of the energy exchange exceeds the rate of losses in the coupled system.¹ This exciton-photon hybridization of the states within the cavity can significantly alter their intrinsic properties of a quantum emitters, including energy,² lifetime,³ emission spectra, and the possibility of energy transfer.^{4,5} The bosonic nature of polaritons together with their extremely low effective mass have been shown to promote the formation of non-equilibrium condensates.⁶ Superfluidity of the polariton condensate has also been demonstrated.⁷ Since the first demonstration of polariton lasing, many systems have been shown to be suitable for

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operation in the strong coupling regime and to form polariton condensates. Further improvement has been achieved with the use of organic materials. Frenkel excitons in these materials, in contrast to the Wannier–Mott excitons in inorganic systems, having considerably higher binding energies, allow room-temperature operation of strongly coupled systems.⁸⁻¹⁰ Semiconductor colloidal nanoparticles and lead halide perovskites represent unique examples of inorganic materials that can be used to achieve strong coupling at room temperature. Flatten et. al.¹¹ were the first to demonstrate strong exciton–photon coupling using colloidal semiconductor nanoplatelets. The achieved value of Rabi splitting was as large as 66 and 58 meV for heavy and light holes, respectively. A Rabi splitting of more than 300 meV has been demonstrated for exciton–photon coupling in a self-assembled CsPbBr₃ perovskite microcuboid grown by the chemical vapour deposition method.¹² Zero-dimensional colloidal semiconductor nanocrystals, or quantum dots (QDs), although being characterized by lower oscillator strengths,^{13,14} have the advantage of high fluorescence quantum yields and small physical volume,¹⁵ causing a plethora of their practical applications in sensing, optoelectronics and nanomedicine.¹⁶ Therefore, QDs are promising inorganic materials for obtaining room-temperature strong coupling and polariton condensation. Due to a high quantum yield, relatively large transition dipole moment (up to 100-200 D),^{17,18} high photostability, and well-developed technology of their large-scale fabrication, QDs could be used in various polariton-based devices. We have used a tuneable microcavity to investigate the formation of strongly coupled states in an ensemble of QDs placed between two metal mirrors. The tuneable microcavity was developed previously and has already demonstrated the capacity for achieving high values of coupling strength.¹⁹ The lateral localization of the optical mode and several orders of magnitude lower values of mode volume has been achieved with the use of an upper mirror with a convex surface.^{20,21}

In this study, we have obtained strong coupling between exciton transition in colloidal QDs and the optical modes of the tuneable microcavity. We have measured the dependence of the QD PL spectrum on the separation distance between the mirrors of the tuneable microcavity, where emission from the weakly coupled part of the QD ensemble was observed along with the emission from the lower polariton branch. We have analysed the polariton dispersion and extracted the value of the coupling strength of about 154 meV. Furthermore, we have measured the dependence of the emission on the power of non-resonant excitation. At the same time, we have observed neither narrowing of the lower polariton emission peak nor threshold behaviour. Instead, saturation of the emission has been demonstrated as the pump power was increased.

The tuneable microcavity, used in our experiments, has been described in detail previously.^{19,21} Briefly, it consists of two metal mirrors with a reflection of about 87%: convex (curvature radius, 77.3 mm) upper and a flat bottom mirrors, the latter positioned on a piezo stack (Fig. 1). The thickness of the aluminium layer of the mirrors was about 35 nm, with a 20-nm protective SiO₂ layer. This configuration forms an unstable Fabry–Perot microcavity with relatively low Q-factors. The advantage of this approach is that the mode volume can be as small as several units of $\left(\frac{\lambda}{n}\right)^3$, where λ is the mode wavelength and n is the refractive index of the media inside the cavity.²⁰

The rough positioning in all three directions was performed by means of several micrometre screws, and the fine positioning with a precision of several nanometres was done by changing the voltage applied to the piezo stack while simultaneously controlling the transmission spectrum. This configuration allowed us to establish the laterally confined optical mode in the area of the lowest point of the upper convex mirror with a relatively low mode

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volume. The details of the experimental setup consisting of a tuneable microcavity could be found elsewhere.¹⁹⁻²¹ The tuneable microcavity as a part of the unique setup called the *System for Probe-Optical 3D Correlative Microscopy* (<http://ckp-rf.ru/usu/486825/>) was mounted on an inverted Ntegra-base (NT-MDT) confocal microspectrometer with a 100X/0.80 MPLAPON XYZ piezo positioned lens (Olympus). An MCWHF2 white LED (Thorlabs) with an optical condenser was used for transmission measurements. For non-resonant PL excitation, we used emission of a 450-nm CW laser (L450P1600MM, Thorlabs). The recording system included an Andor Shamrock 750 monochromator equipped with an Andor DU971P-BV EMCCD (Andor Technology Ltd.). Typical transmission spectrum of the microcavity is shown in Fig. 1(a). The Q-factor of the mode was about 140, with the mode volumes estimated to be in the range from 15 to 20 units of $\left(\frac{\lambda}{n}\right)^3$. We used the finite elements method for estimating the mode volumes.

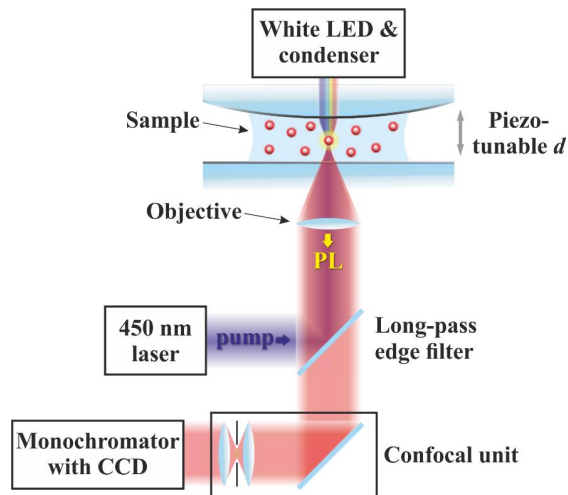


FIG. 1. Scheme of the experimental setup.

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The space between the microcavity mirrors was filled with immersion oil solution containing semiconductor CdSe(core)/ZnS/CdS/ZnS(multishell) QDs at a concentration of about 0.52 mg/ml. QDs were fabricated using the hot injection method.¹⁵ For preparation of a homogeneous solution of QDs in immersion oil, 50 μL of a QD solution in hexane was mixed with the same volume of immersion oil, sonicated for 5 min, and put into a water bath at 80°C for 10 min in order to evaporate hexane from the mixed solutions. After the hexane evaporation, the QD solution in immersion oil was once again sonicated in order to achieve homogeneity. For experiments, 10 μL of each sample was placed on the flat bottom mirror of the microcavity and then covered with the convex upper mirror. During the experiments, the sample solvent evaporation was negligible, and the sample concentration was constant. The absorption and photoluminescence spectra of the QD solution outside of the cavity, as well as their TEM image, are shown in Fig. 2.

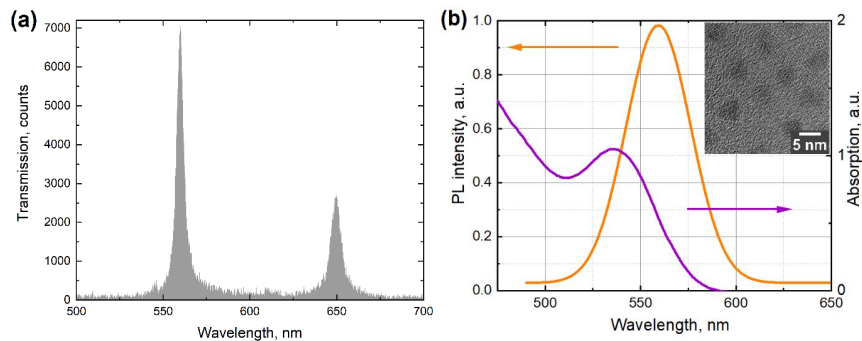


FIG. 2. Tuneable microcavity transmission spectra (a); the absorption (purple) and photoluminescence (orange) spectra of CdSe(core)/ZnS/CdS/ZnS(multishell) QDs in a solution, with a TEM image of QDs in the inset (b).

The absorption spectrum of the QDs consists of a wide absorption band in the short-wavelength region and the first exciton peak at about 535 nm (2.32 eV). The PL spectrum of the QDs represents a symmetrical Gaussian curve with a maximum at 560 nm (2.21 eV) and a

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full width at half maximum (FWHM) of about 40 nm determined by both homogeneous and inhomogeneous broadening.²² It has been reported previously that neither inhomogeneous nor homogeneous broadening influences the spectral positions of polaritonic emission maxima if the strong coupling condition is satisfied.²³ In principle, in an unstable microcavity, used in this study, additional mode localization in the plane perpendicular to the main axis of the cavity is achieved due to the losses determined by the curved surface of the upper mirror. According to the numerical simulation (see Supplementary Material), the spatial region of mode localization at the point of the minimum distance between the mirrors is several micrometres, which is much smaller than the 300- μm focal spot of the objective lens used for collection of the signal in both transmission and PL experiments. Consequently, in the transmission measurements, most of the light passes through the cavity uncoupled, because of the much larger number of photons entering the cavity as compared to the number of strongly-coupled QDs. On the other hand, in the case of PL measurements, the number of emitting QDs cannot exceed the total number of QDs distributed within the optical mode. In photoluminescence measurements, we collected the signal only from the central region with a diameter of about 400 nm, whereas in the transmission measurements, we illuminated with white light, and collected the signal from, a much larger area of the sample (more than 100 μm). Thus, despite the large values of splitting of the PL spectra under non-resonant laser excitation, we should not necessarily expect a parallel observation of splitting in the transmission spectra. In the Supplementary Material, we provide details on the mode distributions and the difference between photoluminescence and transmission measurements.

The dependence of the PL spectra of the hybrid system on the cavity mode detuning is shown in Fig. 3(a). The cavity mode tuning has been performed by changing the distance between the microcavity mirrors from 1178 to 1386 nm. Further decrease in the separation distance between the mirrors led to a significant drop of the emission signal and subsequent

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increase in the signal accumulation time at which the relative drifting of the mirrors became non-negligible.

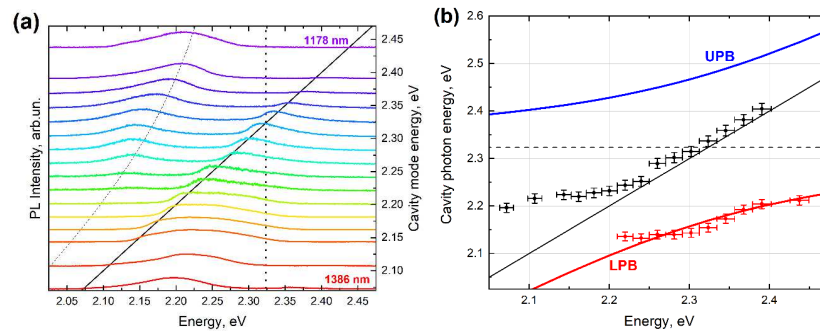


FIG. 3. The PL spectra (a) recorded at distances between the mirrors varied from 1178 to 1386 nm (top–bottom) for a microcavity filled with the solution of CdSe(core)/ZnS/CdS/ZnS(multishell) QDs. The black solid line shows the experimentally controlled position of the cavity mode; the black dotted line shows the positions of the QD first exciton transition; the black dashed line shows the calculated lower polaritonic branch dispersion. The energies of the lower (red), and upper (dark blue) polariton branches at different cavity detunings (b), experimentally derived from the PL spectra (dots) and theoretically calculated (solid lines). The black dots correspond to the emission from weakly coupled QDs. The horizontal dashed line shows the positions of the QD first exciton transition, the solid black line shows the cavity mode energy.

For the maximum cavity detuning from the QD exciton transition, we have observed an almost unchanged PL spectrum. However, it should be noted that, for the largest distance between the mirrors, when the cavity mode is largely negatively detuned from the QD exciton, a small peak arose at about 2.36 eV. This could be attributed to the weak coupling of the highest energy part of inhomogeneously broadened exciton transitions to the higher-order longitudinal optical mode. It is also important that the free spectral range between this mode and the cavity mode was large enough to neglect the influence of the neighbouring longitudinal modes on the

PL spectra of the QDs at all other cavity detunings. When the distance between the mirrors decreased and the cavity photon energy reached the exciton transition energy, two changes occurred. First, the weakly coupled part of the QD ensemble produced emission becomes enhanced at the wavelength corresponding to the microcavity mode and slightly shifted to the blue region due to the presence of higher-order transverse modes in the microcavity. Second, a PL peak corresponding to the lower polariton branch became visible first at 2.13 meV and then shifted towards the bare exciton energy with rising energy of the cavity mode. As the cavity mode shifted to higher energy and the overlap between the bare exciton emission and cavity mode disappeared, the emission from the bare excitonic states merged with the lower polariton branch. Typically, the emission from the upper polariton branch (UPB) in optical microcavities filled with emitters is not observed due to the fast relaxation of the excitation from the UPB to the exciton reservoir.⁹ Thus, the lower polariton branch (LPB) energy displays anticrossing with the emission from weakly coupled emitters, which appears at the bare cavity mode energy.

The light–matter interaction observed in the present research was described in terms of a Jaynes–Cummings Hamiltonian in which quantized electromagnetic field is coupled with a two-level system corresponding to the emitter. It can be written in the following way:

$$H = \hbar\nu_{cav}a^\dagger a + \frac{1}{2}\hbar\nu_x\sigma_z + \hbar g(a^\dagger\sigma_- + a\sigma_+), \quad (1)$$

where $\hbar\nu_{cav}$ and $\hbar\nu_x$ are the energies of the unperturbed cavity mode and emitter absorption transition, respectively; $\hbar g$ is the coupling strength representing the rate of energy exchange between the light and matter constituents; $a(a^\dagger)$ is the annihilation (creation) operator for a cavity photon, $\sigma_{z,-,+}$ are operators acting on the emitter part of the wavefunction; $\sigma_z = |e\rangle\langle e| - |g\rangle\langle g|$, $\sigma_- = |g\rangle\langle e|$, and $\sigma_+ = |e\rangle\langle g|$ ($|g\rangle$ and $|e\rangle$ are the wavefunctions of the

ground and excited states of the emitter, respectively). In a single excitation subspace the diagonalization of the Hamiltonian gives two eigenvalues E_{\pm} :

$$E_{\pm} = \frac{h\nu_{cav} + h\nu_x}{2} \pm \frac{1}{2} \sqrt{(h\nu_{cav} - h\nu_x)^2 + 4(\hbar g)^2}. \quad (2)$$

To obtain the value of coupling strength, we used the experimental frequencies of the lower polaritonic state as E_- and, by varying the free parameter g , found the best fit of our experimental data (Fig. 3(b)). This approach was implemented by means of the differential evolution method.²⁴ The obtained value of coupling constant g is 154 meV. It should be stressed that this value is significantly higher than the coupling strength reported for colloidal nanoplatelets in an open microcavity, despite the considerably higher dipole moment of exciton transition in nanoplatelets. Indeed, previously reported values of coupling strength for ensembles of semiconductor colloidal nanoplatelets in a microcavity consisting of two semi-transparent silver mirrors were about 66 meV.¹¹ Similar values, ranging from 24 to 74 meV for different excitonic transitions, have been observed^{25,26} for the ensembles of colloidal QDs in a bilayer or dielectric slab microcavities. Single-emitter coupling with QDs and plasmonic nanocavities was also studied previously.²⁷⁻²⁹ An extremely low mode volume, about $10^{-5} \left(\frac{\lambda}{n}\right)^3$, made it possible to obtain a coupling strength as high as 230 meV even with a single emitter. Detailed comparison and analysis of the related studies are presented in the Supplementary Material.

However, we would like to emphasise that the tuneable microcavity system used in our study differs significantly from the cavities used in the aforementioned studies. It is known that the coupling strength between light and matter depends on both the cavity mode volume and the number of emitters within the mode. While the conventional optical microcavities consisting of two flat dielectric or metal mirrors¹¹ have the advantage of the collective effect, plasmonic nanocavities²⁷⁻²⁹ are intended to employ the lowest possible mode volumes, making

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the rate of coherent energy exchange so fast that the high losses can be ignored. Our setup was designed to benefit from both effects. On the one hand, due to the convex surface of the upper mirror, we investigated the interaction with the optical mode localized not only in the surface perpendicular to the mirror, but also in lateral directions, providing a several orders of magnitude lower mode volume compared to the typical Fabry-Perot microcavities^{20,21,30}. Typical mode volumes of our microcavity are in the range from 1 to $50 \left(\frac{\lambda}{n}\right)^3$. In the Supplementary Material we provide a detailed description and the calculated spectral and spatial characteristics of the optical modes supported by the cavity. Moreover, the matter placed inside the cavity and its distribution within the mode volume in our experiments are also different from those in the previous studies. Considering these differences, as well as a high photostability and low aggregation of the multishell QDs in the solution with a high concentration, the higher splitting energy looks quite reasonable (see the Supplementary Material for details).

In Fig. 3(a), it can be seen that the PL intensity of the LPB enhanced with an increase in the cavity mode energy, while the exciton fraction of the lower polariton was rising. It is well known that the emission from polaritonic states is proportional to the product of photonic fraction and population.⁴ Thus, observed experimentally increase of the LPB emission with the decrease of the photonic fraction could only be due to a large increase of the LPB population. We consider two mechanisms of LPB population: photonic pumping from the emission of uncoupled states and vibrational scattering.³¹⁻³⁴ The first pathway of LPB population strongly depends on the overlap between the emission spectra of the bare and weakly coupled states and the energy of lower polariton. The second one is proportional to the excitonic fraction of the LPB and depends on the vibrational energy observed in the system^{31,33,34}. In an earlier study on the strong coupling of semiconductor nanoplatelets,¹¹ phonon scattering was shown to be the main mechanism of lower polariton population. However, for the semiconductor

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nanocrystals investigated there, the Stokes shift was about 12 meV, which was much smaller than the observed coupling strength of about 60 meV and, hence, was the main obstacle for efficient population via photonic pumping. In contrast, the observed difference between the first exciton maximum in the absorption and emission spectra of bare QDs in our experiment was as large as 110 meV, which is comparable with the observed coupling strength of 154 meV. Moreover, the maximum emission intensity and the population of the LPB has been observed close to the energy of the emission of bare exciton states. This type of lower polariton intensity behaviour could be attributed to the significant impact of the photon pumping mechanism of the lower polariton population from the excitonic reservoir. However, because the mechanism of phonon-assisted scattering has been shown to be the main mechanism in a similar system of colloidal nanocrystals,¹¹ we cannot fully exclude it despite the fact that the other mechanism is dominant. Thus, it can still provide some of the polariton population for lower polariton states at the energy separated from the bare states with the energy of phonons. In order to investigate the possibility of non-linear behaviour of the observed PL spectra, we measured the dependence of the PL spectra on the excitation power (Fig. 4). The upper limit of the excitation power was due to the photobleaching of the QDs upon excitation at higher pump powers.

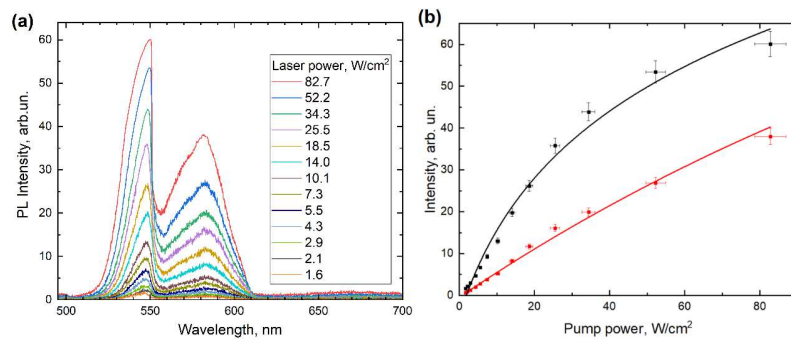


FIG. 4. The PL spectra (a) recorded at various non-resonant pump powers ($\lambda_{\text{ex}}=450$ nm). The extracted values of PL intensity (b) at the wavelengths corresponding to the emission from weakly (black) and strongly (red) coupled parts of the exciton transitions in the QDs ensemble.

Fig. 3(a) shows a rise in both emission maxima with an increase in the excitation power. Minor shifts of the PL maxima occurred at some excitation powers, which was due to the small random instability of the distance between the mirrors. The full width at half maximum was also preserved. We have extracted the intensities of PL maxima and plotted separately their dependence on the excitation power (Fig. 4(b)). As can be seen from Fig. 4(b), there no threshold behaviour was observed for LPB. For excitation powers below 40 W/cm^2 , the emission from both states was found to be linear. For the pump energy above this value, we observed saturation of the emission for both weakly and strongly coupled states. We suggest that the observed behaviour is due to the absorption saturation of exciton transition and photodegradation of the part of the QDs ensemble.

CONCLUSIONS

In this study, we have demonstrated strong coupling between the photonic modes of a tuneable microcavity and localized excitons in colloidal semiconductor QDs at room temperature. A coupling strength as large as 154 meV has been estimated from the fitting of the PL spectra measured at different cavity detunings with the use of the Jaynes–Cummings Hamiltonian. We observed emission from the weakly coupled part of the QD ensemble and LPB emission, while the UPB remained invisible. Analysis of the LPB population dependence on the cavity detuning demonstrated the optical pumping to be the main mechanism of LPB population in our experiments, which is due to the high quantum yields of the bare excitonic states and large Stocks shift. The latter provided sufficient overlap between the LPB and

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emission from the excitonic reservoir. We believe that, due to the high photostability, a quantum yield of up to 100%, wide absorption and narrow emission spectra, and room-temperature operation colloidal semiconductor QDs are promising for practical applications in polaritonics and polaritonic-based devices. Scalability of QD fabrication and the developed technology of coating them with surface ligands, which confers unique chemical properties to QDs and allows targeted linking and control over the space position, pave the way towards the use of colloidal QDs in optoelectronic devices based on light-matter interaction.

SUPPLEMENTARY MATERIAL

See supplementary material for the calculations of the spatial and spectral mode distributions inside the cavity; the details on the difference between photoluminescence and transmission measurements; analysis of the photoluminescence from weakly coupled states; discussion and detailed analysis of the related studies; suppl. Figure S1 with the colour maps of the spatial mode distribution for the lowest-order and a higher-order mode and comparison between the lateral profiles of the two modes in one of the lateral directions; suppl. Figure S2 with schematic configuration of the photoluminescence and transmission experiments; suppl. Figure S3 with comparison of experimental and calculated spectral positions of the peak corresponding to the PL emission from weakly coupled states.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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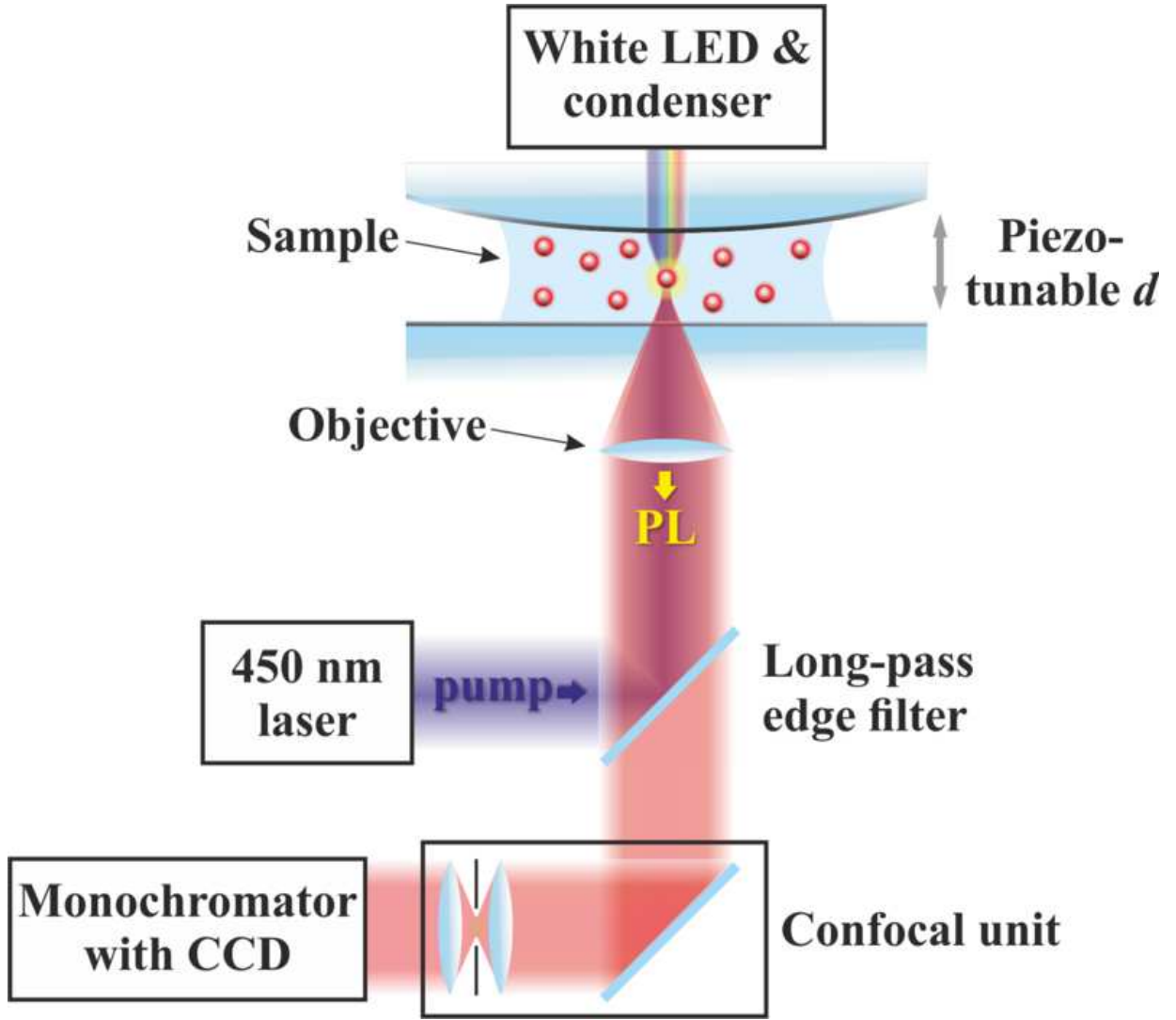
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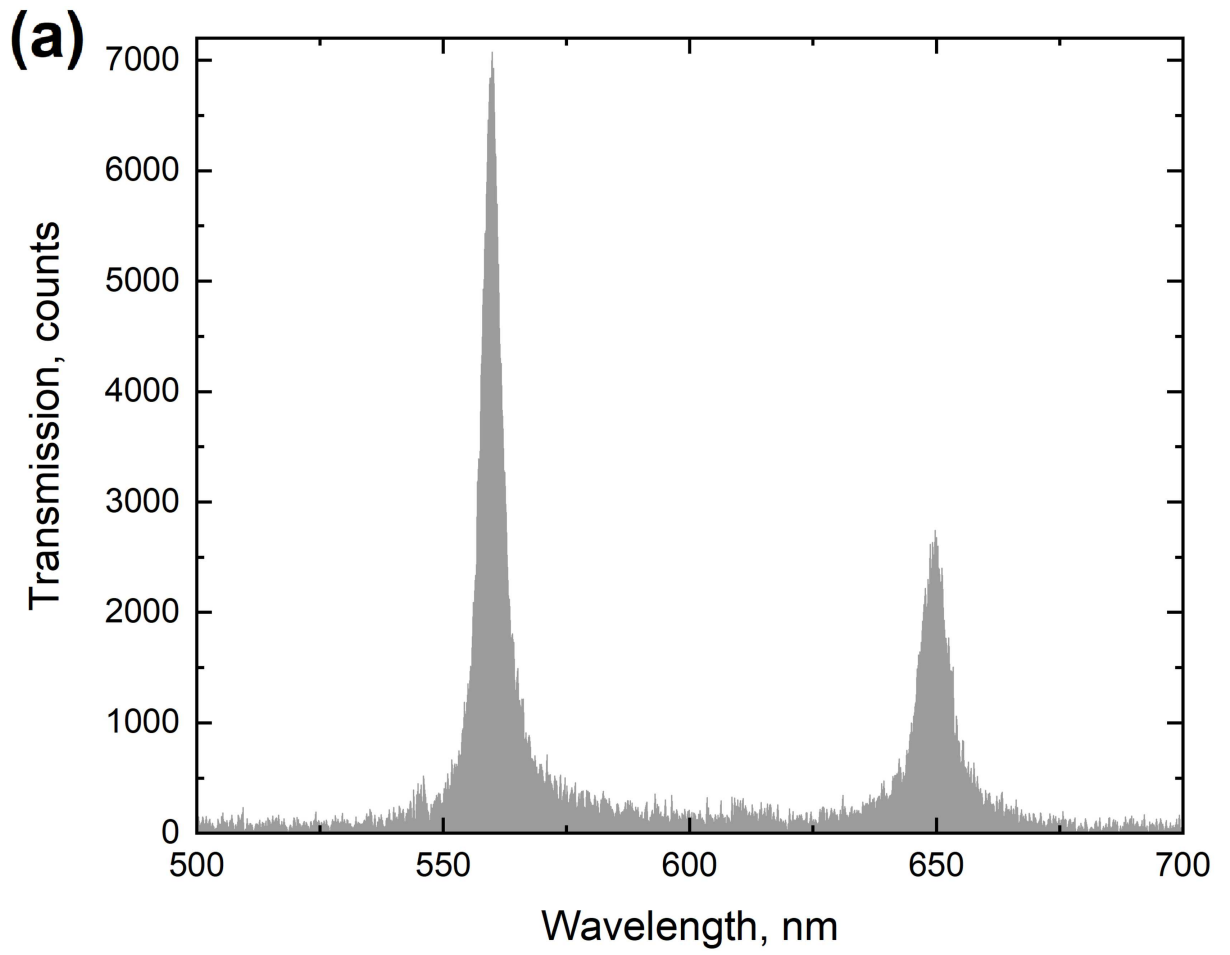
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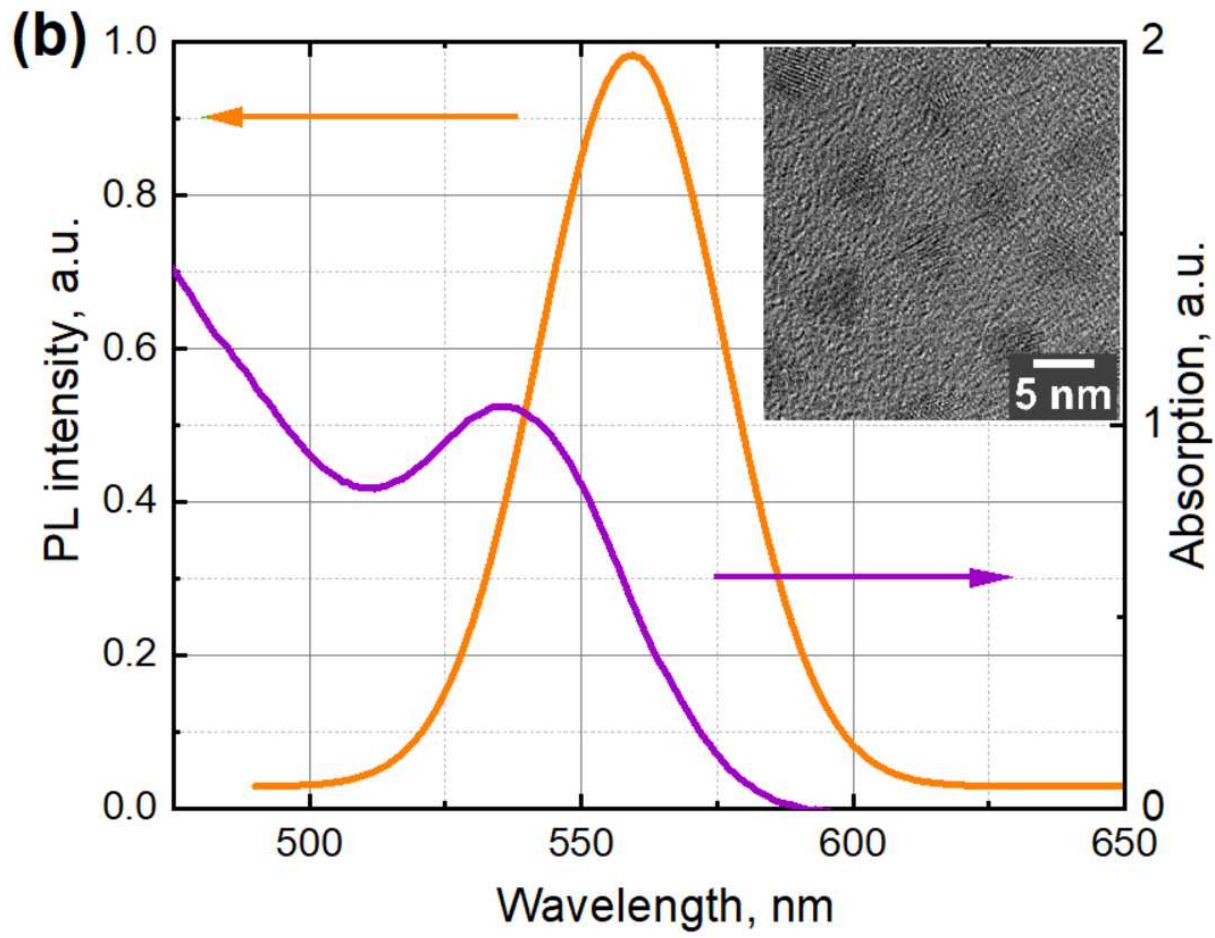
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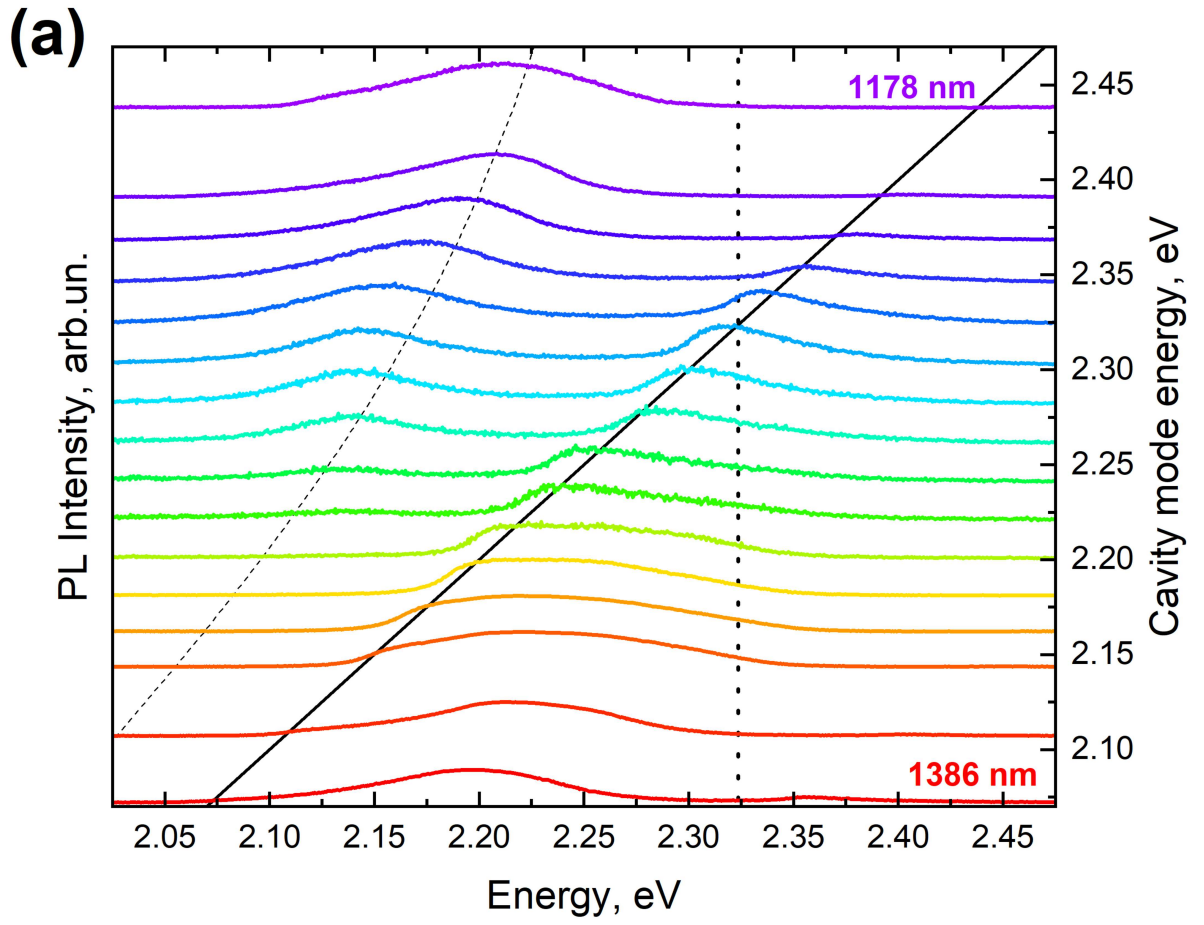
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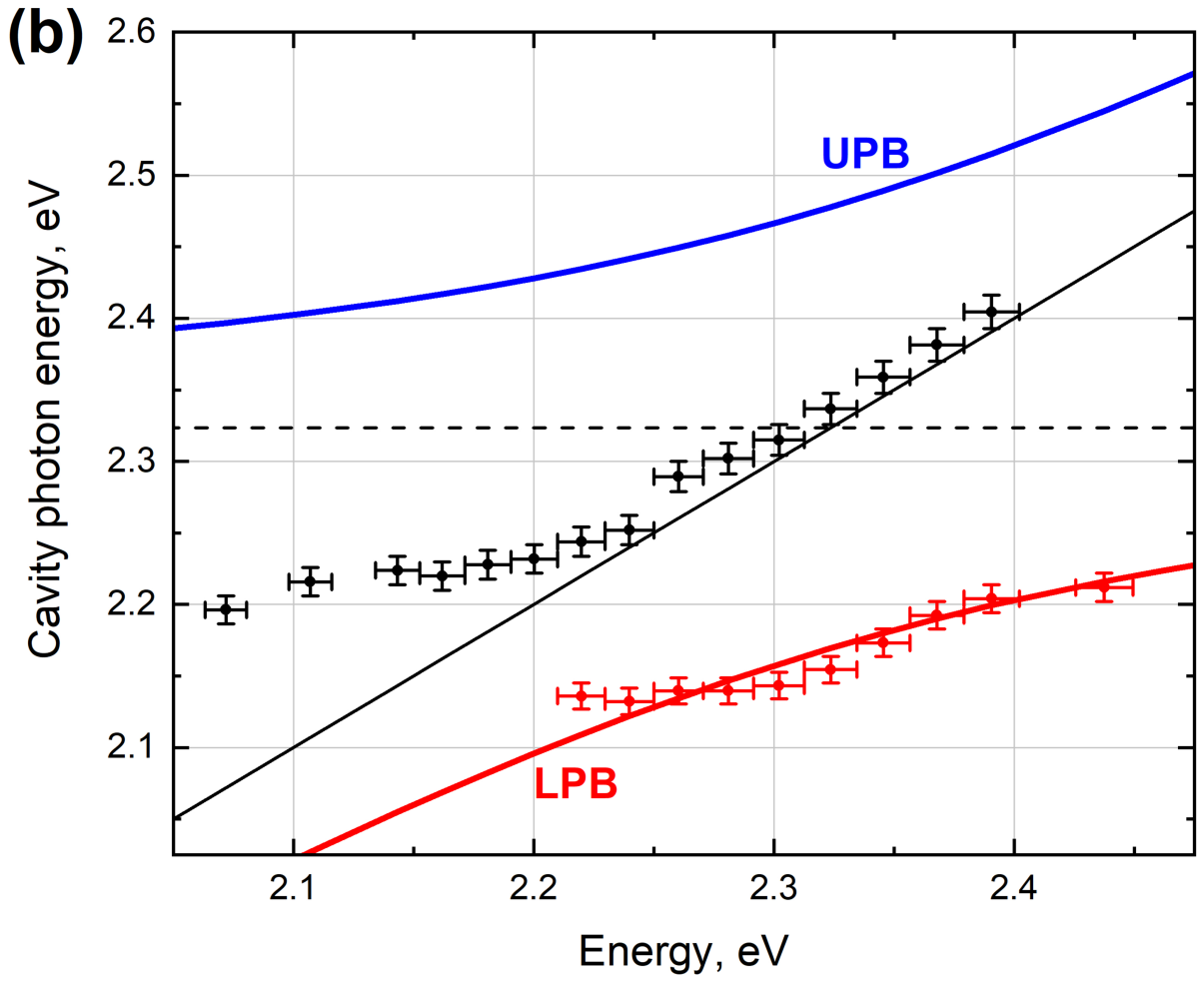
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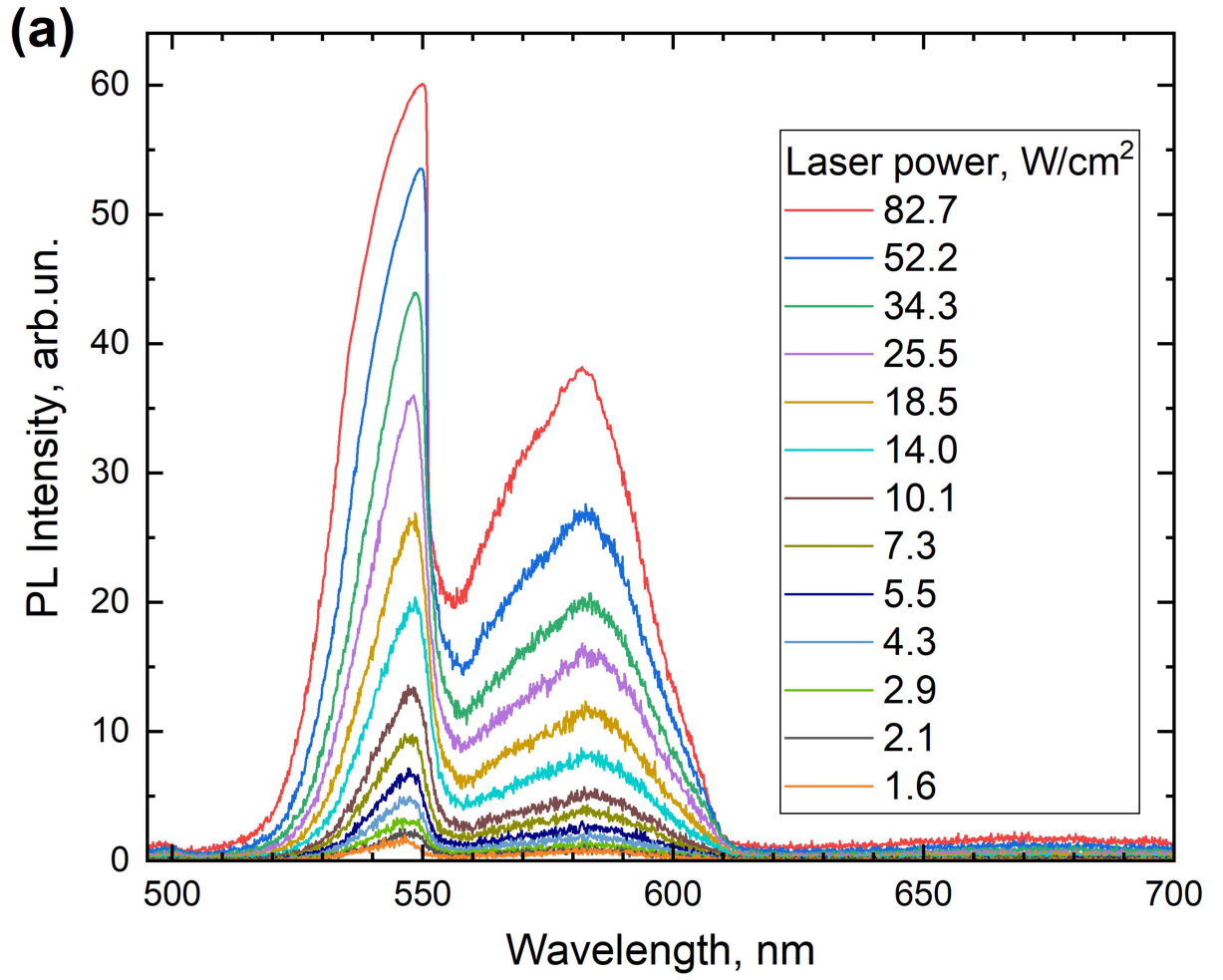
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