Incident angle and photon energy dependence of polariton lasing in organic microcavity

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Organic semiconductor microcavities forming strongly-coupled exciton polaritons are interesting system due to their nonlinear coherent emission observed even at room temperature. Here we investigate the incident angle and photon energy dependent photoemission from a planar organic microcavity to evaluate the relaxation efficiency toward the bottom of the lower polariton branch. We observe nonlinear increase of the vertical emission from the microcavity when the sample is pumped resonantly in the lower polariton branch with the incident angle of 30 to 55 degrees. The observed results support the fast and efficient relaxation of organic microcavity polaritons within the lower polariton branch.

1. Introduction

Designing and manipulating the interactions between a semiconductor material and confined electromagnetic field has been a topic of intensive study for the last 30 years. When the light-matter interaction is strong so that the Rabi frequency is larger than the decay rates of cavity photons and excitons, optical properties of the system is described within the strong-coupling regime. Under the strong-coupling regime, we cannot treat the light-matter interaction perturbatively, and new bosonic quasi-particles called cavity exciton-polaritons are formed as a mixture of the cavity photons and excitons. Among the various kinds of microcavity structures, a planar Fabry-Pérot microcavity composed of a pair of high-reflectivity mirrors called distributed Bragg reflectors (DBRs) is widely used for its simple structure and easy fabrication process. The most interesting characteristics of the DBR microcavity polaritons are their in-plane light mass. This property makes it possible to achieve the polariton condensation.

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and succeeding polariton lasing where the macroscopic portion of the polariton distribution is accumulated in the bottom of the polariton states (\(k_{\parallel} = 0\) state), and emits coherent light.\(^2\)

The observation of polariton condensation and polariton lasing has been achieved in recent decades, utilizing semiconductor microcavities based on GaAs and CdTe.\(^3,4\) These phenomena are considered very similar to the Bose-Einstein condensation and atom laser in atom physics, although the polariton condensate is a "non-equilibrium system" due to the large decay rate. After pioneering works, polariton lasers have been investigated by many research groups aiming for new quantum light sources or ultra-low threshold lasing devices. Most of these studies have been conducted using inorganic semiconductor microcavities based on GaAs, GaN, and ZnO.\(^5-11\) One problem with the GaAs-based devices is that a cryogenic environment is necessary for operation. There are studies on GaN- and ZnO-based devices which can be operated at room temperature, but the fabrication of high-quality microcavity devices remains an important area of study for GaN and ZnO.

Compared to those inorganic semiconductors, organic semiconductor materials have much larger exciton binding energy and vacuum Rabi splitting energy. The strong-coupling of an organic semiconductor in a microcavity was first reported by Lidzey et al.\(^12\) Several years later, Kéna-Cohen et al. have reported room-temperature polariton condensation and following polariton lasing behavior using anthracene single crystal\(^13\) and blue-light-emitting spiro compounds as an active layer.\(^14\) Plumhof et al. have reported a similar phenomena using a ladder-type conjugated polymer layer.\(^15\) In both cases, the observation of nonlinear increase of the photoemission intensity, narrowing of the photoemission bandwidth, and blue shift of the photoemission peak energy supports the macroscopic occupation of the \(k_{\parallel} = 0\) state and the spontaneous formation of coherence.

The formation mechanism of the polariton condensate in inorganic microcavity is explained by the bottleneck effect\(^16\) where the relaxation rate of polaritons in the lower polariton branch reduces significantly near the bottom of the dispersion curve due to the inefficient polariton-phonon scattering process. Instead, the occupation of the bottom of the lower polariton branch is achieved by the bosonic stimulation of the polariton-polariton scattering process from the bottleneck states.\(^17\) This relaxation scheme is supported by the observation of the angle-resonant amplification of the stimulated emission process.\(^18\) For organic semiconductors, however, such parametric scattering process has not been reported.\(^19\) This may be attributed to the inefficiency of the polariton-polariton scattering or the shorter lifetime of the polaritons in the organic semiconductors. Until now, only limited kinds of organic semiconductor materials have been reported to show the polariton condensation and polariton
Since strong-coupling organic microcavity polaritons (OMPs) have a shorter lifetime compared to the inorganic ones, the efficient relaxation towards the \( k_{||} = 0 \) state is crucial for the realization of polariton lasing behavior. For the investigation of organic semiconductor materials suitable for the formation of cavity exciton polaritons and subsequent polariton condensation, it is important to understand the relaxation processes under the resonant and non-resonant excitation of OMPs. With this strategy, Coles et al. have reported the observation of the bottleneck effect in OMP by monitoring the photoluminescence along the \( k_{||}=0 \) direction while scanning the incident angle of the pump laser pulse.\(^{20}\) They have shown that the direct relaxation from the upper polariton branch to the bottom of the lower polariton branch occurs more efficiently compared to the scattering process via the bottleneck state. This experiment has been performed using J-aggregated dye 5,5′,6,6′ -tetrachloro-1,1′ -diethyl-3,3′ -di(4-sulfobutyl)-benzimidazolocarbocyanine (TDBC) as the active layer, but the polariton condensation and lasing behavior from the bottom of the lower polariton branch have not been observed.

In this paper, we have adopted the spiro compound 2,7-bis[9,9-di(4-methylphenyl)-fluoren-2-yl]-9,9-di(4-methylphenyl)fluorene (TDAF) as the active layer, which has been well studied by Daskalakis et al. and known to show polariton lasing behavior.\(^{14, 21, 22}\) Since the relaxation dynamics can be influenced by the formation of polariton condensate, it is important to observe the photoemission spectrum in a wide range of pump pulse fluence covering below and above the polariton lasing threshold. We have chosen several different excitation conditions (excitation photon energy, incident angle) based on experimentally determined polariton dispersion curves to discuss the polariton relaxation efficiency within the OMP. This paper is organized as follows. The preparation of the DBR samples and their evaluation by angle-resolved reflectivity and transmittance measurement are described in Sec. II. The excitation photon energy and incident angle dependence of momentum space photoemission spectrum is described in Sec III together with the discussion on the relaxation behavior of OMPs. Sec. IV offers concluding remarks.

2. Method

2.1 Sample preparation

We prepared planar DBR microcavities for the following cavity exciton-polariton experiments. First, 7 pairs of SiO\(_2\) and Ta\(_2\)O\(_5\) dielectric layers (thicknesses of 66 nm for the SiO\(_2\) layers, 42 nm for the Ta\(_2\)O\(_5\) layers) are sputtered on a fused silica substrate (\( \phi = 12 \) mm, thickness=1 mm). The center of the stop band is measured to be \( \sim 3.30 \) eV, with a band width of \( \sim \)
130 meV at normal incidence. As an active layer, a thin layer of TDAF (>98%, Lumtec) is deposited on top of the DBR layer by thermal evaporation under a vacuum of $2 \times 10^{-4}$ Pa. The thickness of the TDAF layer is evaluated to be $\sim 115$ nm by a surface profilometer. On top of the TDAF layer, we fabricated the top side DBR structures composed of 7 pairs of SiO$_2$ and Ta$_2$O$_5$ layers by using the same sputter. The thickness of each layer is designed to be the same as those of the bottom DBR. The same sample is used for the angle-resolved reflectivity and transmittance measurement to evaluate the dispersion curves, and for the momentum space photoemission measurement.

2.2 angle-resolved reflectivity / transmittance measurement

The dispersion curve of OMPs is determined from the angle-resolved reflectivity and transmittance spectrum. The setup for the reflectivity measurement is shown in Fig. 1(a). The fiber-coupled white light (DH-2000, Ocean Optics) is focused on the surface of the sample by a planoconvex lens. The reflected light is collimated and picked up by another fiber cable and introduced to the CCD camera (ProEM+512B, Princeton Instruments) attached behind the f=150 spectrometer (SP2156, Princeton Instruments). The groove density of the grating is 300 l/mm. The spectrum is taken with vertically binned mode so that the central 100 pixels along the vertical axis are accumulated to give one spectral data point. The incident angle $\alpha$ is scanned from $35^\circ$ to $70^\circ$. With this method, we cannot obtain the reflectivity spectrum around the normal incidence ($\alpha \sim 0^\circ$) due to the interference of the optical components. Instead, we have observed the transmittance spectrum by positioning the collection optics on the opposite side of the sample as shown in Fig. 1(a).

2.3 Momentum space photoemission measurement

The photoemission spectrum pumped by femtosecond UV laser is measured using the momentum space imaging setup depicted in Fig. 1(b). Briefly, the output of a Ti:Sapphire regenerative amplifier (repetition rate 1kHz, central wavelength 800 nm, Legend Elite, Coherent) is frequency converted to near infrared signal and idler pulses using a commercial OPA system (Opera Solo, Coherent). The temporal full-width at half maximum (FWHM) of the fundamental laser pulse is measured to be $\sim 35$ fs. We prepare wavelength tunable UV pulses using the second harmonic generation (SHG) of the SHG output of the signal pulse using two nonlinear crystals. The typical output bandwidth of the UV pulse is $\sim 62$ meV at the photon energy of 3.14 eV and 48 meV at 3.72 eV. This pulse is focused on the sample
with a planoconvex lens (f=150) at a particular incident angle $\theta_{pu}$. The lens is mounted on a rotational stage (RBB300A/M, Thorlabs) so that $\theta_{pu}$ can be manually tuned while keeping the distance between the lens and the sample constant. The spot size of the pump pulse at the sample surface depends on $\theta_{pu}$, with a typical spot size of $45 \times 62 \ \mu m$ at $\theta_{pu}=35^\circ$.

Since the reflectivity changes as we scan $\theta_{pu}$, we monitor the input and reflected pulse energy using a power meter for each $\theta_{pu}$. We have assumed that the transmitted pump pulse energy is negligibly small, so that the difference of the input and reflected pulse energy corresponds to the net absorption by the microcavity sample. The photoemission signal is collected from the opposite side of the microcavity with a high NA objective lens (NA=0.9, UPLFLN 60X, Olympus), and momentum space image is generated by another plano-convex lens (f=100) on the entrance slit of the spectrometer (MS-3501i, Solar TII). For this purpose, the emitted beam is rotated by 90° using a periscope before arriving at the spectrometer. The wavelength of the emitted light is dispersed by a grating (1800 l/mm). The energy-dispersed angle dependent photoemission image is mapped on the CCD camera (Newton 940, Andor) attached on the output port of the spectrometer.

3. Results and discussions

3.1 Observation of exciton-polaritons and polariton lasing

Figure 2 (a) shows the angle-resolved transmittance spectrum taken from $\alpha=0$ to $30^\circ$. Clear peaks around 3.1 eV are observed within the stop band of the DBR mirror. Figure 2(b) shows the angle-resolved reflectivity spectrum taken from $\alpha=35^\circ$ to $70^\circ$. In both measurements, the pump pulse has transverse-magnetic(TM) polarization. At $\alpha = 35^\circ$, we can observe a dip structure around 3.15 eV. As $\alpha$ increases, the dip position is blue shifted up to 3.24 eV at $\alpha = 50^\circ$. At higher $\alpha$ values, we can observe another dip around 3.70 – 3.80 eV. Each of the dip and peak positions observed in Figs. 2(a) and (b) are fitted with Gaussian function with center positions plotted in Fig. 2(c) with black square dots. The straightforward analysis of the angle-resolved reflectivity / transmittance spectrum can be done based on the simple coupled oscillator model taking the cavity photon mode and the exciton mode as base functions. The corresponding $2 \times 2$ Hamiltonian matrix is given as

$$H = \begin{pmatrix} E_{ph}(\alpha) & V_{int} \\ V_{int} & E_{ex} \end{pmatrix}$$

(1)

where $E_{ph}(\alpha)$ and $E_{ex}$ are the energy of cavity photon and exciton, respectively, and $V_{int}$ is the parameter representing the exciton-photon interaction strength. Here we assume the constant
exciton energy of $E_{\text{ex}} = 3.50 \text{ eV}$, and $E_{\text{ph}}(\alpha) = E_{\text{ph}}(0) \left( 1 - \left( \frac{\sin \alpha}{n_{\text{eff}}} \right)^2 \right)^{-1/2}$, where $E_{\text{ph}}(0)$ is the energy of cavity photon without transverse momentum, and $n_{\text{eff}}$ is the effective refractive index of the cavity.\textsuperscript{24} The angular dependence of $V_{\text{int}}$ is also neglected for simplicity. After diagonalizing eq. (1), we obtain two anti-crossing dispersion curves which are typical for the strong coupling system. Hereafter, we call the lower energy curve as lower polariton branch (LPB) and higher energy curve as upper polariton branch (UPB). The observed angle dependent reflectivity and transmittance spectrum is fitted by these polariton curves, and the resulting fitted parameters are $E_{\text{ph}}(0) = 3.21 \text{ eV}$, $V_{\text{int}} = 234 \text{ meV}$, and $n_{\text{eff}} = 2.10$, respectively. The fitted LPB and UPB dispersion curves are plotted in Fig. 2(c) together with $E_{\text{ph}}(\alpha)$ and $E_{\text{ex}}$. From the dispersion curve of the LPB, the effective mass of the LPB polariton at $k_{\parallel} = 0$ is derived to be $3.45 \times 10^{-35} \text{ kg}$. This value should be compared with the effective mass of the cavity photon which is derived to be $2.25 \times 10^{-35} \text{ kg}$ at $k_{\parallel} = 0$. The 1.53 times heavier mass of the polariton is the direct evidence of forming exciton-polaritons in our experiment. The obtained exciton-photon interaction parameter is smaller than the reported value of $293 \text{ meV}$,\textsuperscript{14} which may be due to the quality of the fabricated DBR cavities.

Figure 3 shows the angle-resolved photoemission spectrum from the same sample at two different pump fluences. The incident angle and the photon energy of the excitation laser pulse is fixed at $\theta_{\text{pu}} = 55^\circ$ and $E_{\text{pu}} = 3.26 \text{ eV}$, respectively. When the fluence of the pump pulse is $15.7 \mu\text{J/cm}^2$, we observe the angle-resolved photoemission spectrum between $\pm 25^\circ$ as shown in Fig. 3(a). When the fluence is increased to $31.6 \mu\text{J/cm}^2$, we observe strong directional emission along the $k_{\parallel} = 0$, as shown in Fig. 3(b). The pump fluence dependence of the photoemission intensity spectrum is plotted in Fig. 4(a). We selected $0 \pm 1.0^\circ$ part of the photoemission spectrum to obtain the emission along the $k_{\parallel} = 0$. To evaluate the peak intensity and its width, the spectrum is fitted with a Gaussian function. The integrated area and its full width at half maximum (FWHM) are defined as the peak intensity and FWHM of the photoemission along $k_{\parallel} = 0$. These are plotted in Fig. 4(b). Both plots show clear nonlinear behavior around the fluence of $\sim 25 \mu\text{J/cm}^2$. In Fig. 4(c) the emission peak photon energy along the $k_{\parallel} = 0$ direction is plotted. As the pump fluence increases, the peak energy shifts towards the higher photon energy.

Although we have not checked the spatial coherence of the above threshold emission along the $k_{\parallel} = 0$ direction, the nonlinear increase of this directional emission component above the threshold accompanied by the sudden decrease of the emission FWHM and blue shift of the peak photon energy is the typical characteristics of the polariton lasing behavior.\textsuperscript{14,15} These
phenomena are explained by the increase of occupation in $k_{||} = 0$ state, and the elongated temporal coherence due to the formation of macroscopic coherent state. The blue shift at the higher polariton density is explained due to the repulsion among the polaritons and exciton-polaritons. Thus, we conclude that it is reasonable to say that the room temperature polariton lasing behavior similar to the one reported in the previous papers$^{14,22}$ is observed in our microcavity sample.

3.2 Pump angle and energy dependence of polariton emission

Based on the results obtained in the previous section, we measured angle-resolved photoemission spectrum at different excitation conditions to clarify the relaxation efficiency in the momentum-energy space. Figure 5 shows the UPB and LPB dispersion curves of the microcavity determined by the fitting procedure. These are the same as those plotted in Fig. 2(c). Along the LPB dispersion curve, we choose six excitation conditions with incident angles of $\theta_{pu} = 30$ to $55^\circ$ with the step of $5^\circ$, labeled as #1 – #6 in Fig. 5. The corresponding photon energies of the pump pulse are 3.15, 3.17, 3.19, 3.21, 3.24, and 3.26 eV, respectively. In each case, the pump pulse has the energy FWHM of 40–60 meV. Hereafter, these six excitation conditions are called "excitation points" within the two-dimensional energy-angle plot shown in Fig. 5. At each excitation point, we scanned the pump pulse fluence to evaluate the pump fluence dependence of the polariton lasing behavior. For comparison, we selected one additional point, $(\theta_{pu}, E_{pu})=(15^\circ, 3.65 \text{ eV})$, labeled #7 in Fig. 5. At this point, slightly above the UPB is excited. To avoid possible damage to the active layer during the measurement, the focused position is slightly shifted for each excitation point #1 – #7. The local homogeneity of the sample is checked separately by monitoring the sample position dependence of the photoemission peak energy. The deviation of the emission peak photon energy is less than 2 meV when we scan the laser spot position over 1.2 mm. In this way, we have assumed the influence of the inhomogeneity to the following analysis is negligibly small.

The photoemission peak intensity along the $k_{||} = 0$ direction and its FWHM are plotted in Figs. 6 (a) and (b) for each of the excitation points #1 – #7, as the function of the absorbed fluence. In each case of #1 – #6 where the LPB is resonantly excited, we could observe polariton lasing behavior with the threshold around 13 $\mu$J/cm$^2$ at #2, and around 22 – 28 $\mu$J/cm$^2$ for other points (#1, #3 ~ #6). The peak photoemission intensity is enhanced more than two orders when the absorbed fluence is above the threshold. At the same time, the emission FWHM is sharpened to about 2 – 4 meV which is about one-fifth compared to the
FWHM of incoherent photoemission observed below the threshold. The peak photon energy is blue shifted by about 3 – 4 meV as the absorbed fluence increases. Note that the spectral width of the pump pulse (40 to 60 meV) is much larger than the observed blue shift so that the resonant condition of the pump pulse is not affected by this blue shift. On the other hand, we can not observe clear threshold behavior for the polariton lasing at #7 where the pump pulse energy is tuned slightly above the UPB. The absorbed fluence dependence of the emission FWHM also shows the same tendency. From these results, we may infer that the relaxation from the UPB to the bottom of the LPB is not efficient in our sample. Comparing the lasing behavior at points #1 – #6, we conclude that the excitation angle dependency of the relaxation efficiency toward the \( k_{||}=0 \) states is rather small except at #2, with no signature of the bottleneck states at higher angle side. The minimum threshold observed at point #2 may be related to the parametric scattering process as reported in the previous paper. More detailed studies based on the pump-probe type measurement are planned to clarify this phenomenon.

Next, we checked the pump pulse energy dependence of the angle-resolved photoemission spectrum at a fixed incident angle of 35°. We took 7 excitation points, labeled #a – #g in Fig. 5. The photon energy of the pump pulse was tuned to 3.14 (#a), 3.16 (#b), 3.29 (#c), 3.39 (#d), 3.49 (#e), 3.60 (#f), and 3.72 eV (#g), respectively. At point #a, \( E_{pu} \) is slightly detuned below the LPB, and point #b is on-resonant with the LPB. Points #c to #f are off-resonant with their photon energies between LPB and UPB. At point #g, the excitation point is set slightly above the UPB. In Figs. 7(a) and (b), the peak intensity, FWHM of the emission peaks are plotted at each excitation point #a – #g as the function of the absorbed fluence. From Fig. 7(a), it is clearly seen that the threshold for the polariton lasing is lowest when the pump pulse is resonant to the LPB (#b). The threshold is estimated to be \( \sim 13 \, \mu \text{J/cm}^2 \). As the photon energy of the pump pulse is detuned from the LPB, the threshold increases. The reason why the lasing behavior can be observed, even under negatively detuned condition (#a), is probably due to the overlap of the broad bandwidth of the pump pulse and the broad absorption lineshape of the LPB. Unfortunately we cannot set the \( E_{pu} \) lower than #a, due to the unwanted direct scattering components observed by the CCD camera. For excitation point #g, polariton lasing is not observed even with the absorption fluence of 100 \( \mu \text{J/cm}^2 \). Figure 8 shows the energy-momentum photoemission image taken with the excitation point #g, with the absorption fluence of 4.78 \( \mu \text{J/cm}^2 \). We observe the photoemission from the LPB between ±20° around 3.06 eV. For higher angles around ±40°, we detect broad weak emission covering the photon energy from 2.80 eV to 3.10 eV. The angle-dependent shift of the edge of the stop band of DBR cavity is clearly seen. These higher angle emission components overlap nicely.
with the broad photoemission spectrum from the bulk exciton reservoir, so that we assign these components due to the leakage of the photoemission of bare exciton from the cavity. Thus Fig. 8 directly indicates that the non-resonantly excited energy in the UPB flows to the bare exciton state and considerable amount of the energy in the exciton reservoir leaks out of the cavity instead of being scattered to the LPB. The higher lasing threshold observed at excitation points #d – #f compared to #a – #c can be explained by the same mechanism.

Studies on the relaxation scheme in the strong-coupling OMP have been reported by several groups. Most studies have been performed using J-aggregates as the active medium. Coles et al. performed the photoluminescence measurement in an OMP composed of J-aggregated TDBC layer using a tunable narrow-band laser excitation at different incident angles. They have observed efficient and fast energy transfer from the UPB to the exciton reservoir, followed by the scattering to the high angle region of the LPB assisted by the intramolecular vibrational modes. They also found that the relaxation efficiency within the LPB towards the bottom is dependent on the excitonic fraction of the polariton state. This is very similar to the bottleneck effect reported in the inorganic microcavity polaritons. From these results, they have concluded that the non-resonant excitation above the exciton energy level is the most efficient means to accumulate population in the bottom of the LPB to realize polariton lasing.

Virgili et al. performed femtosecond transient absorption measurement of an OMP using TDBC as an active medium. The fast energy transfer from the UPB to the exciton reservoir and subsequent energy flow from the exciton reservoir to the LPB have been directly observed by femtosecond transient absorption measurements. Although the fast relaxation toward the bottom of the LPB is confirmed in J-aggregated TDBC, the coherent emission from the \( k_{||} = 0 \) state has not been reported in TDBC, except in the composite layer structure.

In contrast, in our OMP composed of TDAF active layer, such efficient relaxation from the UPB to the \( k_{||} = 0 \) state in the LPB could not be observed. As shown in Fig. 8, the energy scattering from the UPB to the exciton reservoir was observed in our sample, so that the difference will be the scattering efficiency of the excitons in the reservoir towards the lower polariton branch. Thus it is expected that the vibrationally assisted scattering from the exciton reservoir to the high energy LPB band is not efficient in TDAF layer compared to the J-aggregates. On the other hand, the relaxation within the LPB towards the \( k_{||} = 0 \) state occurs efficiently in our TDAF microcavity within the observed excitation angle range of 30 – 55°. These results indicate that the resonant excitation along the LPB will be the most efficient method for the formation of polariton lasing in OMP composed of TDAF. These results are consistent with the preceding experiments of TDAF microcavity performed by Daskalakis et
al., in which the pump photon energy was mainly tuned to the LPB.$^{14,22}$ The difference of the efficiency for the polariton relaxation within the LPB may explain why polariton lasing behavior has been reported in OMP composed of TDAF, while no reports of polariton lasing have appeared for OMP composed of TDBC. These findings suggest that the efficiency of relaxation from the exciton reservoir to the LPB should be checked carefully for the future investigation of organic materials suitable for the polariton lasing. If the relaxation towards the $k_{||}=0$ state within the LPB is efficient, it is also possible to circumvent this issue simply by resonantly exciting the LPB at intermediate incident angle.

4. Conclusion

We have observed the angle-resolved photoemission spectrum from an organic semiconductor microcavity showing polariton condensation and polariton lasing behavior at room temperature. When the excitation is performed along the LPB at incident angles between 30 and 55°, the polariton lasing from the $k_{||} = 0$ state is observed without any increase of the relaxation efficiency at the higher angle region. From the observed pump photon energy dependence of the polariton lasing behavior at fixed incident angle, we notice that the direct excitation of the UPB does not result in the efficient accumulation of population in the $k_{||}=0$ state of LPB. This result suggests that for the polaritons in the UPB, the efficiency to relax towards the $k_{||}=0$ state of LPB and the leakage out of the exciton reservoir is comparable, which hinders the low threshold polariton lasing. This is a clear difference from the preceding relaxation dynamics observed in organic microcavity J-aggregates. For the efficient polariton lasing from the bottom of the LPB in the OMP composed of TDAF active layer, we conclude that the resonant excitation of the LPB will be the most efficient process.

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Fig. 1. (a) A schematic view of angle-resolved reflectivity and transmittance measurement setup. (b) Momentum space image detection setup used for the incident angle dependent photoemission measurement.
Fig. 2. (a) Angle-resolved transmittance spectra taken at low incident angle. Each spectrum is vertically shifted for clarity. (b) Angle-resolved reflectivity spectra taken between $\alpha=35$ to $70^\circ$. Blue lines labeled $E_{LP}$ and $E_{UP}$ are the manually drawn supporting lines connecting the dip position in each spectrum. (c) The plot of the dip and peak positions observed in (a) and (b). Dashed lines indicate the energy of a bulk exciton of TDAF, $E_{ex}$, and the dispersion of cavity photons, $E_{ph}$. Solid lines indicate the fitted result of $E_{LP}$ and $E_{UP}$ curves by the model calculation.
Fig. 3. Angle-resolved photoemission spectra taken at two different absorption fluence of (a) 15.7 $\mu$J/cm$^2$ and (b) 31.6 $\mu$J/cm$^2$. The incident angle and photon energy are set as $\theta=55^\circ$ and $E_{pu}=3.26$ eV, respectively.
Fig. 4. (a) photoemission spectra taken at different pump fluence. (b) Absorbed fluence dependence of the photoemission peak intensity along $k_{\parallel} = 0$ and its FWHM. (c) Absorbed fluence dependence of the peak photon energy for the photoemission along $k_{\parallel} = 0$ direction.
Fig. 5. Map of the excitation conditions (Incident angle, center energy) of the pump pulse examined in the angle-resolved photoemission measurement.
Fig. 6. Absorbed fluence dependence of (a) the photoemission peak intensity, (b) the photoemission peak FWHM, along $k_{\|} = 0$ when the excitation is performed at different excitation points #1~#7 in Fig. 5.
Fig. 7. Absorbed fluence dependence of (a) the photoemission peak intensity, (b) the photoemission peak FWHM, along $k_{||} = 0$ when the excitation is performed at different excitation points #a~#g in Fig. 5.
Fig. 8. Angle-resolved photoemission image taken with \((\theta_{pu}, E_{pu})=(35^\circ, 3.72 \text{ eV})\) (excitation point #g). The absorbed fluence is 4.78 \(\mu\text{J/cm}^2\). The photoemission from the LPB and leaked emission from the bare exciton reservoir are observed.
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