Ultrafast Manipulation of a Strongly-Coupled Light-Matter System by a Giant ac Stark Effect

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Supplementary Information

Supplementary Discussion

Full 3-by-3 Hamiltonian Diagonalization

A coupled two-level system of an exciton and a cavity photon is described by a 2x2 Hamiltonian of the following form:

$$H_0 = \begin{pmatrix} E_X & \hbar\Omega_R \\ \hbar\Omega_R & E_C \end{pmatrix} \tag{1}$$

where E_X , E_C are the exciton and the cavity photon energies, respectively; $\hbar\Omega_R$ is the Rabi energy, which denotes a two-level coupling strength. The UP and LP energies are calculated by performing diagonalization of the H_0 and are given by

$$\begin{aligned} E_{UP,0} &= \frac{1}{2}\left(E_X + E_C + \sqrt{4\hbar^2\Omega_R{}^2 + \Delta^2}\right) \\ E_{LP,0} &= \frac{1}{2}\left(E_X + E_C - \sqrt{4\hbar^2\Omega_R{}^2 + \Delta^2}\right) \end{aligned} \tag{2}$$

where the symbol $\Delta \equiv E_C - E_X$ denotes detuning or an energy mismatch between the cavity photon and the exciton energies.

In order to describe the Stark photon interaction with exciton-polaritons, we introduce an additional coupling parameter:

$$H_{full} = \begin{pmatrix} E_X & \hbar\Omega_R & \hbar\Omega_P \\ \hbar\Omega_R & E_C & 0 \\ \hbar\Omega_P & 0 & \hbar\omega_P \end{pmatrix} \tag{3}$$

Where $\hbar\omega_P$ is the Stark photon energy and $\hbar\Omega_P$ is the Stark photon-exciton coupling strength.

In order to find modified polariton energies in the presence of the Stark photon, we perform diagonalization of the aforementioned Hamiltonian. The UP and the LP eigenenergies are given by

$$
\begin{aligned}
E_{LP} &= \frac{1}{3}\left\{\left(E_C + E_X + \hbar\omega_P\right) + \left(-1\right)^{2/3} c - \left(-1\right)^{1/3} c^{-1} b\right\} \\
E_{UP} &= \frac{1}{3}\left\{\left(E_C + E_X + \hbar\omega_P\right) - \left(-1\right)^{1/3} c + \left(-1\right)^{2/3} c^{-1} b\right\}
\end{aligned}
\qquad (4)
$$

Where we define several constants to simplify the results

$$
\begin{aligned}
c &\equiv 2^{-1/3}\left(a + \sqrt{a^2 - 4b^3}\right)^{1/3} \\
a &\equiv \left(2E_C - E_X - \hbar\omega_P\right)\left(E_C + E_X - 2\hbar\omega_P\right)\left(E_C - 2E_X + \hbar\omega_P\right) + \\
&\quad + 9\hbar^2\Omega_R{}^2\left(E_C + E_X - 2\hbar\omega_P\right) - 9\hbar^2\Omega_P{}^2\left(2E_C - E_X - \hbar\omega_P\right) \\
b &\equiv E_C{}^2 + E_X{}^2 + \hbar^2\omega_P{}^2 - E_X\hbar\omega_P - E_C E_X - E_C\hbar\omega_P + 3\hbar^2\Omega_P{}^2 + 3\hbar^2\Omega_R{}^2
\end{aligned}
$$

The ac Stark shift expression is given by a difference between the modified polariton energies ($E_{UP,LP}$), and the original polariton energies without the presence of the Stark pump ($E_{LP0,UP0}$).

Perturbative 2-by-2 Hamiltonian Diagonalization

The perturbative treatment is applicable when the Stark photon-exciton coupling is small with respect to the Rabi frequency or, equivalently, when the Stark shift is small with respect to the polariton energy difference. The perturbed exciton energy is obtained from the Hamiltonian describing exciton-Stark photon coupling

$$
H_{X-P} = \begin{pmatrix} E_X & \hbar\Omega_P \\ \hbar\Omega_P & \omega_P \end{pmatrix} \qquad (5)
$$

For small pump coupling, the perturbed expression for the exciton level is

$$E_X' = \frac{1}{2}\left(E_X + \hbar\omega_P + \sqrt{4\hbar^2{\Omega_P}^2 + \left(E_X - \hbar\omega_P\right)^2}\right) \approx$$
$$\approx \frac{1}{2}\left(E_X + \hbar\omega_P + \left(E_X - \hbar\omega_P\right)\left\{1 + \frac{4\hbar^2{\Omega_P}^2}{2\left(E_X - \hbar\omega_P\right)^2}\right\}\right) = E_X + \frac{\hbar^2{\Omega_P}^2}{E_X - \hbar\omega_P} \tag{6}$$

where $\delta = E_X' - E_X = \frac{\hbar^2{\Omega_P}^2}{E_X - \hbar\omega_P}$ is an ac Stark shift for the exciton level.

The perturbed LP and UP energies ($E_{UP,LP}$) are obtained by substituting the perturbed exciton energy (E_X') into the expression of the LP and UP energies without the Stark photon presence ($E_{LP0,UP0}$).

$$E_{UP,LP} = \frac{1}{2}\left(E_X + \frac{\hbar^2{\Omega_P}^2}{E_X - \hbar\omega_P} + E_C \pm \sqrt{4\hbar^2{\Omega_R}^2 + \left(E_X + \frac{\hbar^2{\Omega_P}^2}{E_X - \hbar^2\omega_P} - E_C\right)^2}\right) \tag{7}$$

The significant error between the perturbative and full approaches is noticeable for large Stark photon-exciton couplings as evident from Fig. 4 in the main manuscript. Therefore, in the regime of strong Stark pump coupling, the cavity photon plays a non-negligible role in the dynamical Stark effect, which can no longer be described solely by the perturbed exciton energy.

Supplementary Figure 1

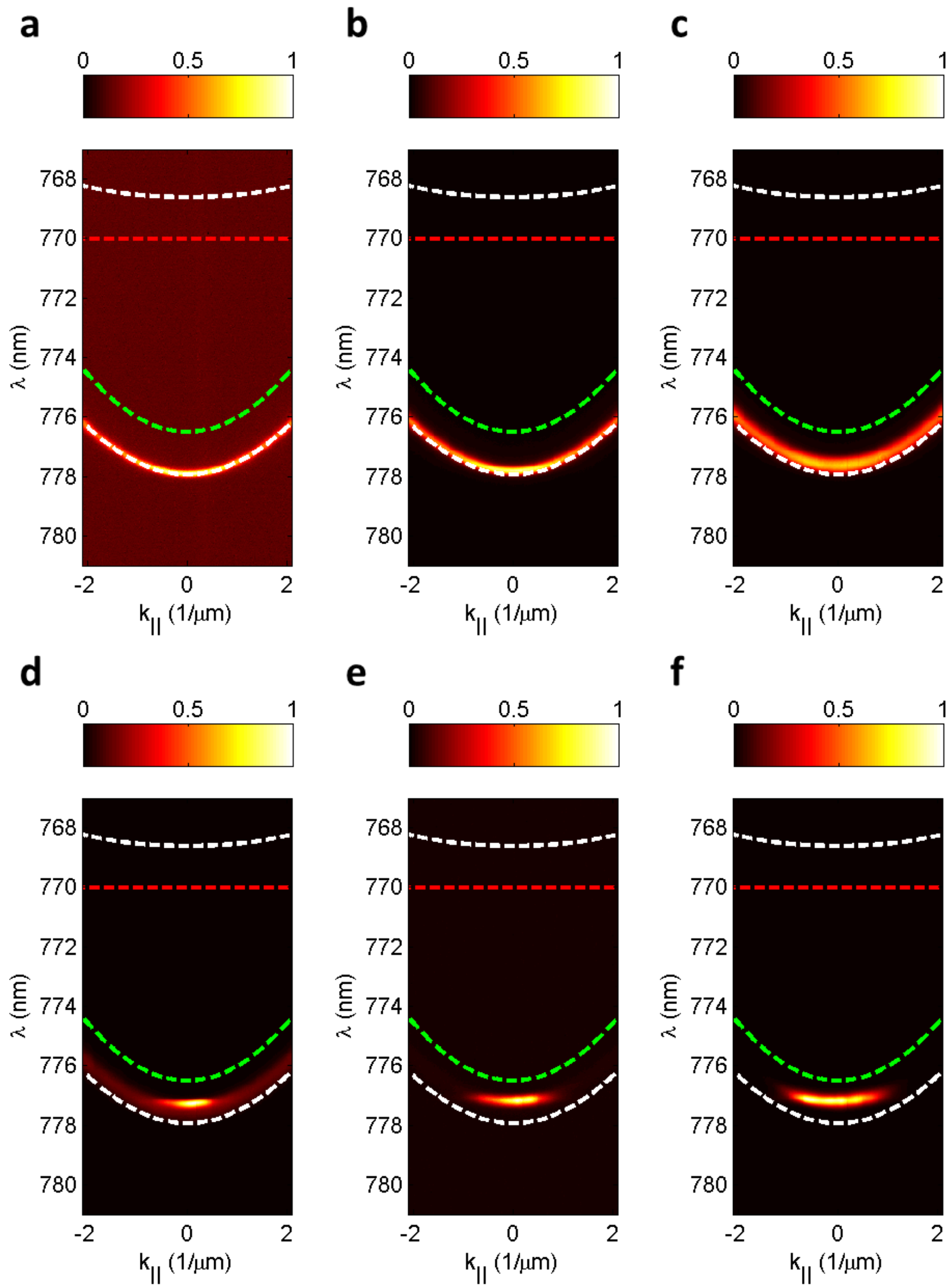

Fig. 1. Sample photoluminescence measurements for a detuning energy of -13.5 meV

a-**f** Angle-resolved sample photoluminescence measurements for excitation powers of 0.08, 1, 2.4, 3, 4.1 and 6.2mW, respectively . Theoretically predicted energies are depicted for the polaritons (dashed white), the bare cavity photon (dashed green) and the exciton (dashed red).

Supplementary Figure 2

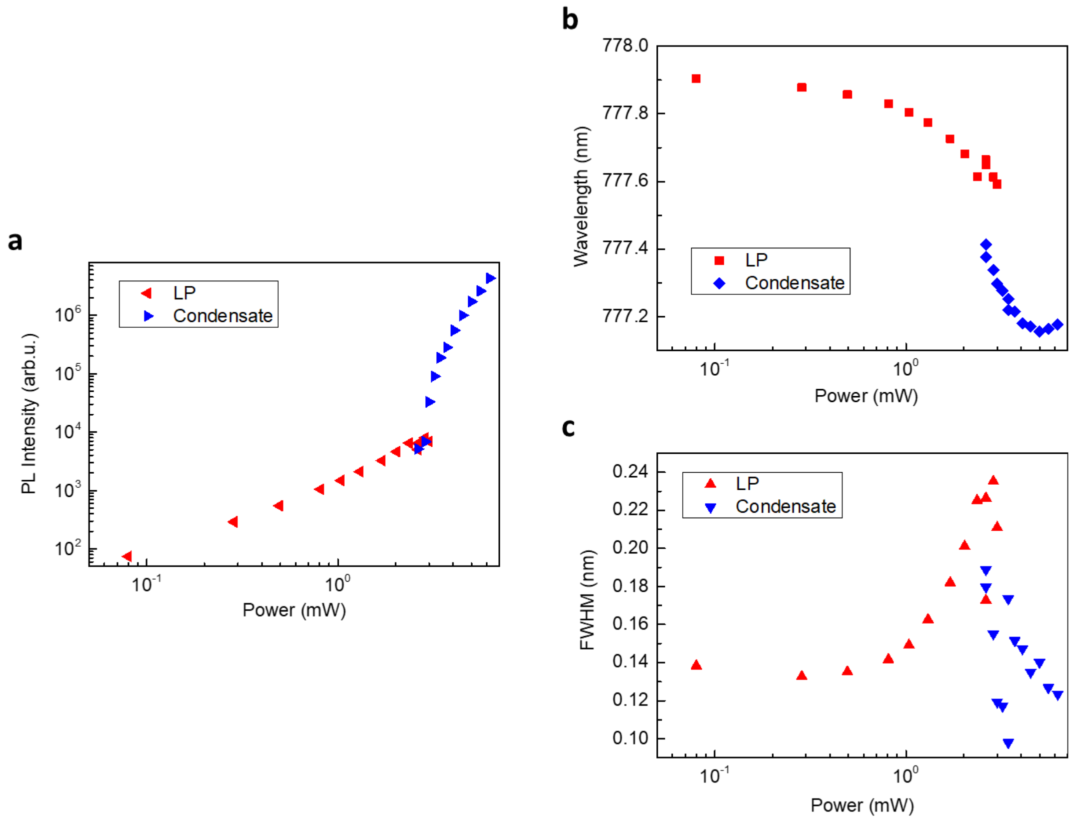

Fig. 2. Sample condensation measurements for a detuning energy of -13.5 meV

a-c Wavelength, line width and integrated intensity of photoluminescence as a function of the injection power. LP points (in red) and condensate points (in blue).

Supplementary Figure 3

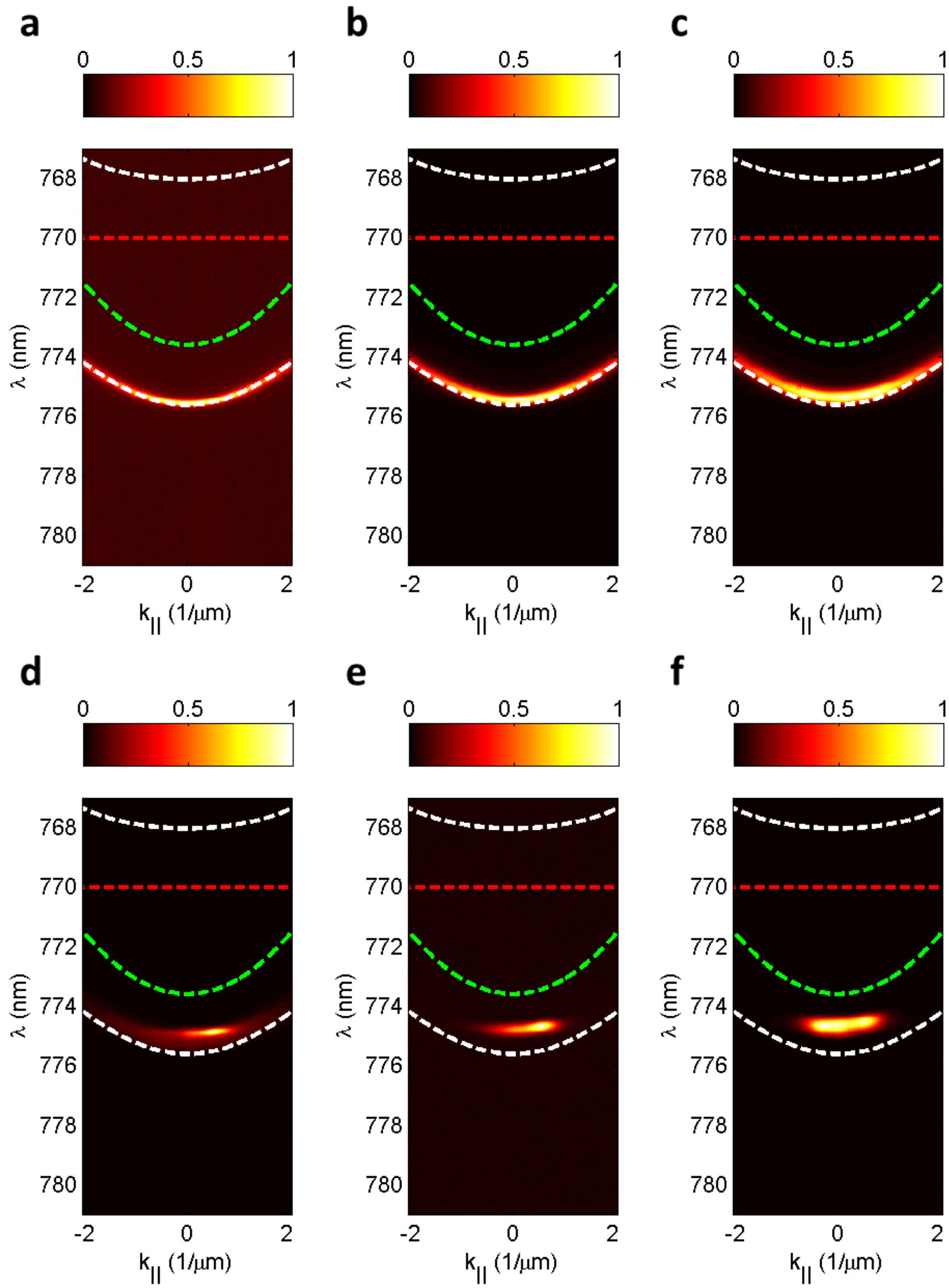

Fig. 3. Sample photoluminescence measurements for a detuning energy of −7.5 meV

a-**f** Angle-resolved sample photoluminescence measurement for excitation powers of 0.09, 0.8, 1.4, 2, 3.1 and 5.2mW, respectively . Theoretically predicted energies are depicted for the polaritons (dashed white), the bare cavity photon (dashed green) and the exciton (dashed red).

Supplementary Figure 4

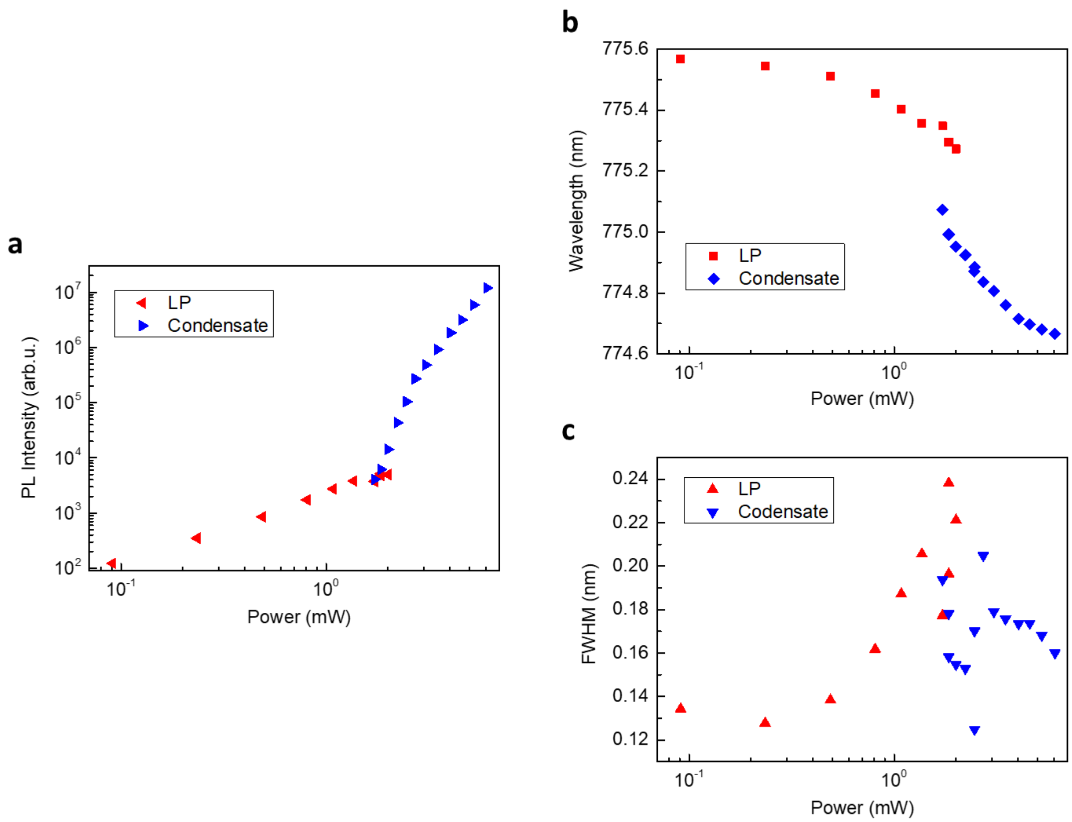

Fig. 4. Sample condensation measurements for a detuning energy of −7.5 meV

a-c Wavelength, line width and integrated intensity of photoluminescence as a function of the injection power. LP points (in red) and condensate points (in blue).

Supplementary Figure 5

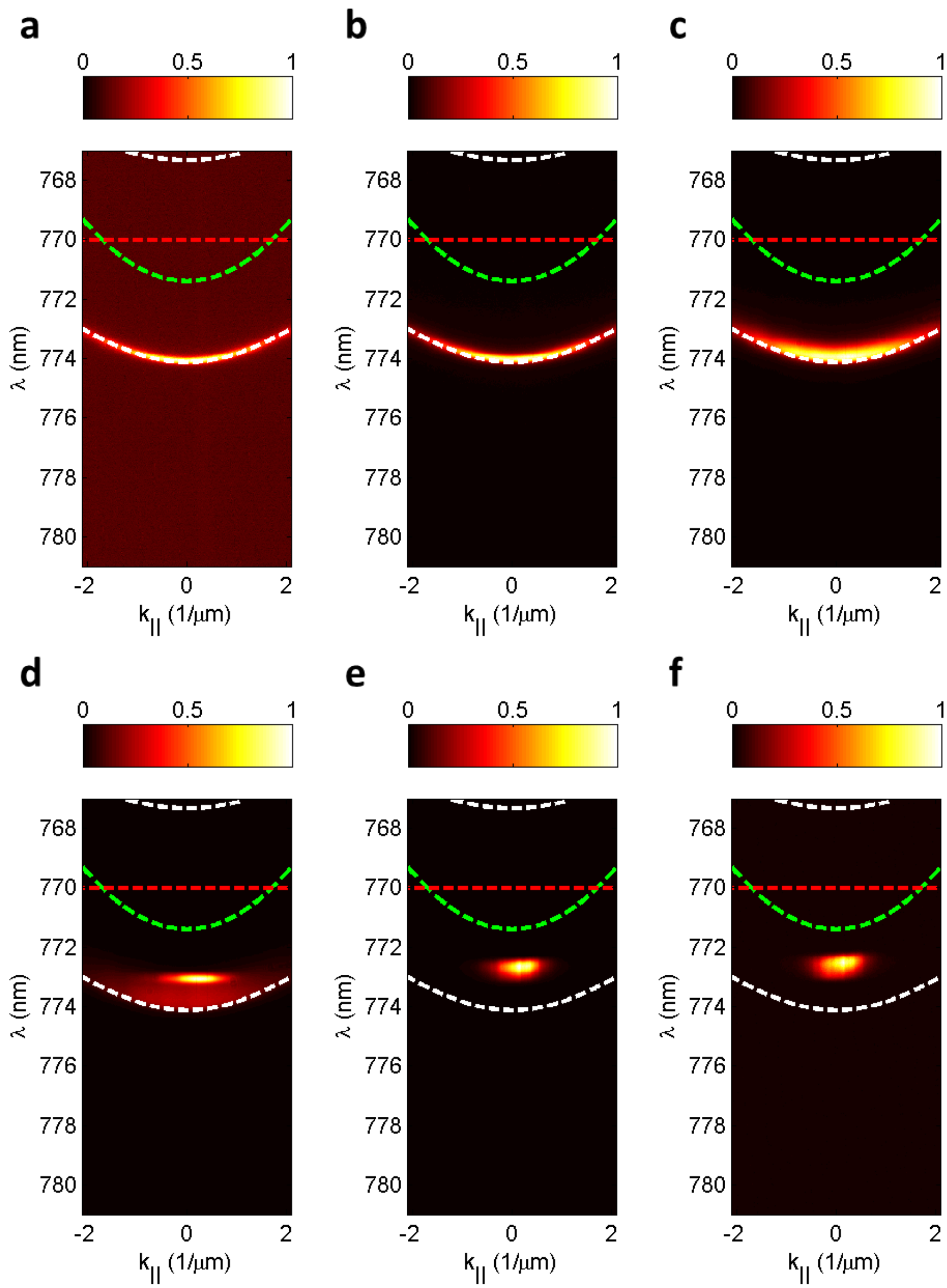

Fig. 5. Sample photoluminescence measurements for a detuning energy of −2.9 meV

a-**f** Angle-resolved sample photoluminescence measurement for excitation powers of 0.06, 0.7, 1.9, 2.7, 3.5 and 5.2mW, respectively . Theoretically predicted energies are depicted for the polaritons (dashed white), the bare cavity photon (dashed green) and the exciton (dashed red).

Supplementary Figure 6

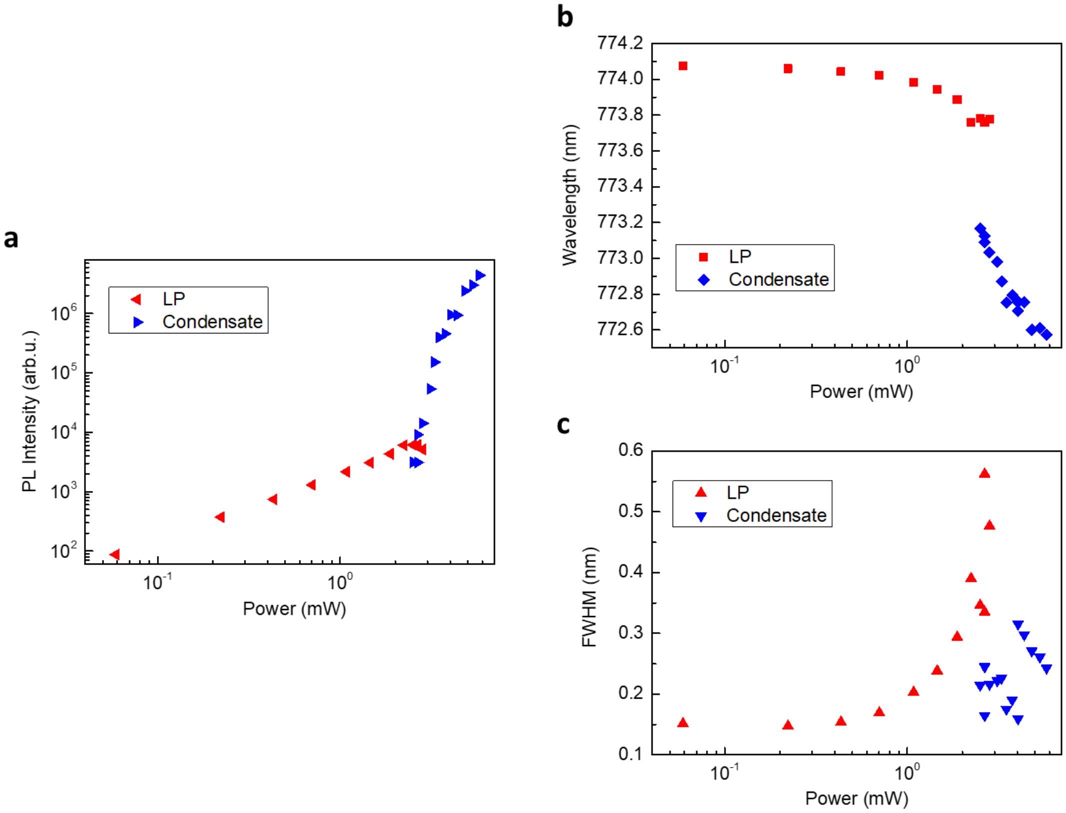

Fig. 6. Sample condensation measurements for a detuning energy of −2.9 meV

a-c Wavelength, line width and integrated intensity of photoluminescence as a function of the injection power. LP points (in red) and condensate points (in blue).

Supplementary Figure 7

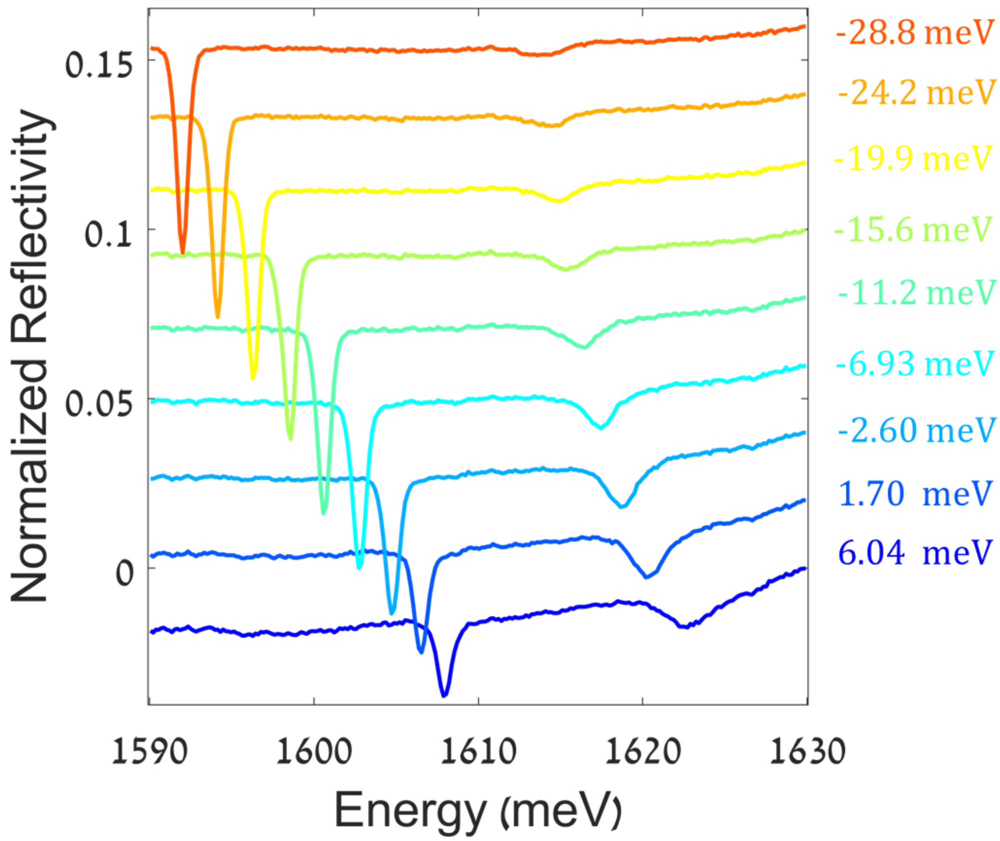

Fig. 7. Sample normalized reflectivity at different detunings as a function of energy showing UP and LP.

Supplementary Discussion: Data Processing and Stark Shift Magnitude Extraction

In the pump-probe experiment, the differential reflectivity is determined by the difference in reflected probe spectra when both pump and probe beams impinge on the sample and the spectrum of the reflected probe beam alone i.e. $\Delta R(\tau) \equiv R_{\text{PumpProbe}}(\tau) - R_{\text{Probe}}$, where τ is the time delay between the pump and probe pulses. Normalized differential reflectivity $\Delta R(\tau)/R_{\text{Probe}}$ is commonly used in order to exclude the spectral dependence of the probe beam. For our sample, the probe spectra alone will reveal two dips belonging to the lower (E_{LP}) and the upper (E_{UP}) polaritons as depicted in Supplementary Figure 8 (a) by the blue trace. When the pump and probe pulses arrive at near zero-time delay, the probe spectra will record a blueshift of the polariton lines (Supplementary Figure 8 (a), red trace).

For Stark shifts less than a half width of the polariton line, the shift will influence the differential reflectivity amplitudes of the characteristic bump and dip trace. In this case, in order to extract the Stark shift, the differential reflectivity spectrum at near zero-delay time should be fitted to the theoretical model with the shift value as a free parameter. For shifts which are much larger than the polariton line widths, however, the differential reflectivity amplitudes of the unperturbed and shifted polaritons are no longer a function of the pump intensity, therefore the extraction process is more straightforward, and is depicted on the Supplementary Figure 8. The Stark shift for each polariton can be determined from the difference in spectral position of the differential reflectivity bump and dip.

Supplementary Figure 8

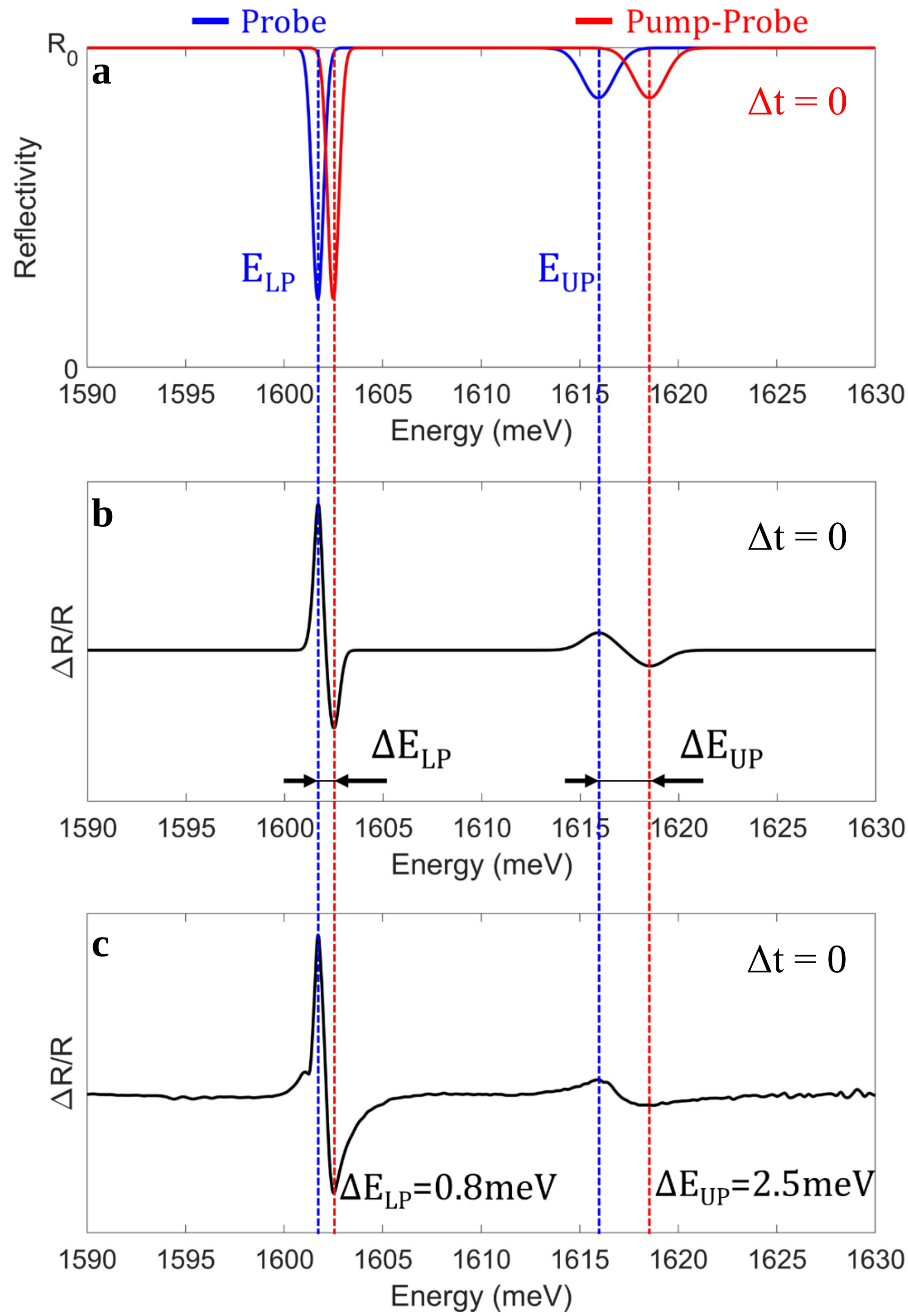

Fig. 8. Schematics of the Stark shift extraction. **a.** Reflectivity of the unperturbed polariton lines (blue) together with the reflectivity at zero pump-probe delay Δt=0 (red) showing shifted polariton energies due to the ac Stark effect. **b.** Normalized differential reflectivity of the spectra shown in **a**, the lower and upper polariton shifts are indicated by ΔE_{LP} and ΔE_{UP}, respectively. **c**. Measured normalized differential reflectivity spectra for a 3.7 GW/cm^2 pump intensity at zero pump-probe delay Δt=0.