Supplementary Information for "Valley addressable exciton-polaritons in atomically thin $MoSe_2$"

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SUPPLEMENTARY NOTE 1: DERIVATION OF THE SCATTERING RATES

In this section, we provide more details on the derivation of the equations of the main text.

First, we would like to describe the scattering between the exciton reservoir (index X) and a polariton state (index 2). In general, the rate equations for the exchange between such states can be written as follows:

$$\begin{aligned} \frac{dn_X}{dt} &= -W_{X\to 2} n_X + W_{2\to X} n_2 \\ \frac{dn_2}{dt} &= +W_{X\to 2} n_X - W_{2\to X} n_2 \end{aligned} \tag{S1}$$

where we have omitted all other terms, e.g. lifetime, spin relaxation, etc. The scattering mechanism can be the exciton-phonon interaction or the exciton-exciton interaction: in presence of a thermalized exciton reservoir, both give the same dependence on the temperature and energy difference:

$$\begin{aligned} W_{X\to 2} &= W_0 x \\ W_{2\to X} &= W_0 x e^{-\frac{E_X - E_2}{k_B T}} \end{aligned} \tag{S2}$$

The excitonic fraction x of the polariton state appears in the scattering rate because only the exciton can efficiently interact with phonons, or the other excitons. The coefficient W_0 is the same for both scattering rates, but since in one case (scattering down) the energy (the phonon) is emitted and in the other (scattering up) – absorbed, the corresponding probability factors are different. Our goal is now to simplify these expressions, in order to obtain an analytical solution. Indeed, if the polariton branch is far below the reservoir, the scattering backwards from this branch to the reservoir is small and can be safely neglected. However, if this branch is close to the reservoir, this scattering is not negligible, and we need to introduce a correction to the scattering rate $W_{X\to 2}$ in order to take this into account. Indeed, in this case the scattering rates $W_{X\to 2}$ and $W_{2\to X}$ become comparable, as well as the populations, and so it is possible to write simply the difference of both terms:

$$W_{X\to 2} - W_{2\to X} = W_0 x \left(1 - e^{-\frac{E_X - E_2}{k_B T}}\right) \tag{S3}$$

which in the first order can be written as

$$W_{X\to 2} - W_{2\to X} \approx W_0 x \frac{E_X - E_2}{k_B T} \tag{S4}$$

On the other hand, at strongly positive detunings $\Delta \gg V$ (V is one half of the Rabi splitting), both the photonic fraction p_2 and the relative energy of the polariton state with respect to the exciton $(E_X - E_2)/\Delta$ scale as V^2/Δ^2, which allows to simplify the expression for the scattering rate by using the photonic fraction instead of the energy difference:

$$W^{eff}_{X\to 2} \approx W_0 xp \tag{S5}$$

and the rate equation now contains a single term with a single effective scattering rate:

$$\begin{aligned} \tfrac{dn_X}{dt} &= -W^{eff}_{X\to 2} n_X \\ \tfrac{dn_2}{dt} &= +W^{eff}_{X\to 2} n_X \end{aligned} \tag{S6}$$

In the main text, we omit the index eff on the scattering rate.

The advantage of all these simplifications is that the final model with a reduced number of terms has a clear analytical solution for the polarization degree.

SUPPLEMENTARY NOTE 2: SPECIAL CASE OF THE UPB

In the main text, we have discussed that one can assume an energy dependence of the polarization degree within the reservoir: fully polarized particles are injected at high energy and they progressively lose their polarization as they relax in energy. Therefore, if the UPB is resonant with some particular energy state within the reservoir, there will be an incoming scattering rate towards this UPB with a certain polarization degree (higher than at the bottom of the exciton reservoir). These polarized polaritons of the UPB will then lose their polarization within this branch, before being emitted out of the system. To obtain their final polarization degree, let us consider the UPB as a separate system with a pumping P (which describes the scattering from the reservoir) carrying a polarization degree ρ. The lifetime of the states of the UPB is τ_2 and the spin relaxation time $\tau_{2\pm}$. The rate equations for this reduced system can be written as follows:

$$\begin{aligned} \frac{dn_+}{dt} &= \frac{P(1+\rho)}{2} - \frac{n_+}{\tau} - \frac{n_+ - n_-}{2\tau_\pm} \\ \frac{dn_-}{dt} &= \frac{P(1-\rho)}{2} - \frac{n_-}{\tau} + \frac{n_+ - n_-}{2\tau_\pm} \end{aligned} \tag{S7}$$

The stationary solution of this system of equations is:

$$\begin{aligned} n_+ &= \frac{\tau(P\tau + P\tau_\pm + P\rho\tau_\pm)}{2(\tau+\tau_\pm)} \\ n_+ &= \frac{\tau(P\tau + P\tau_\pm - P\rho\tau_\pm)}{2(\tau+\tau_\pm)} \end{aligned} \tag{S8}$$

which gives a simple expression for the polarization degree of the UPB:

$$\rho' = \frac{\rho\tau_\pm}{\tau + \tau_\pm} \tag{S9}$$

used in the main text.

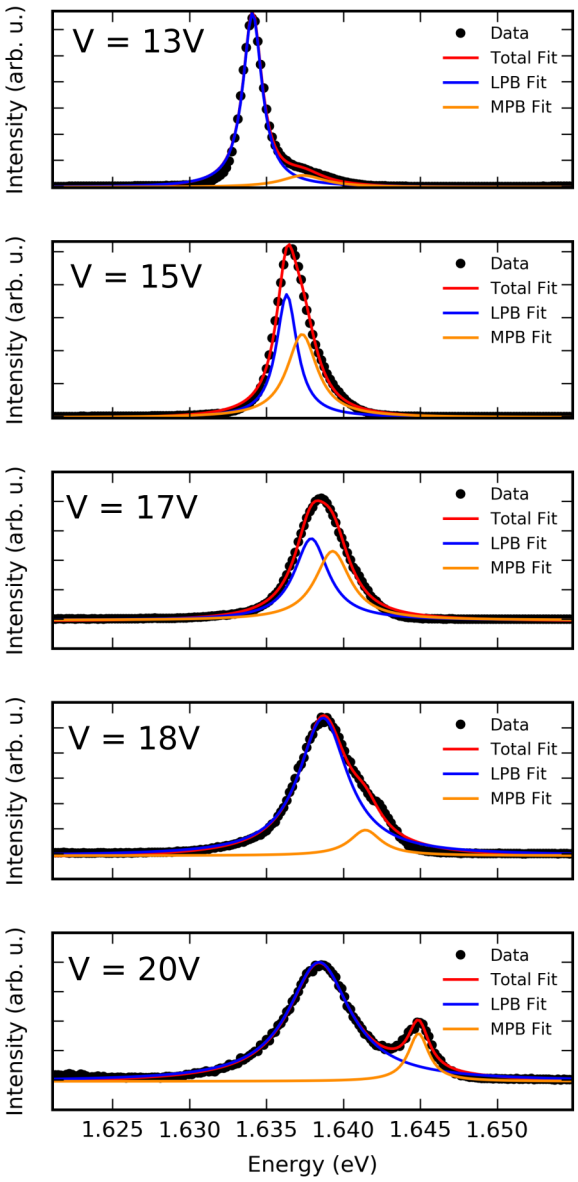

FIG. S1. Spectra as a function of piezo voltage when the cavity is tuned through resonance with the trion emission energy. The presence of two peaks when slightly detuned and a strong asymmetry of the peak at resonance indicates the onset of strong coupling. Here the Rabi splitting of 1.3 ± 0.1 meV is comparable to the inhomogeneously broadened polariton linewidths preventing the polariton branches from being fully resolved at resonance. The fits correspond to those used to extract the polarization degree of the constituent polariton peaks.

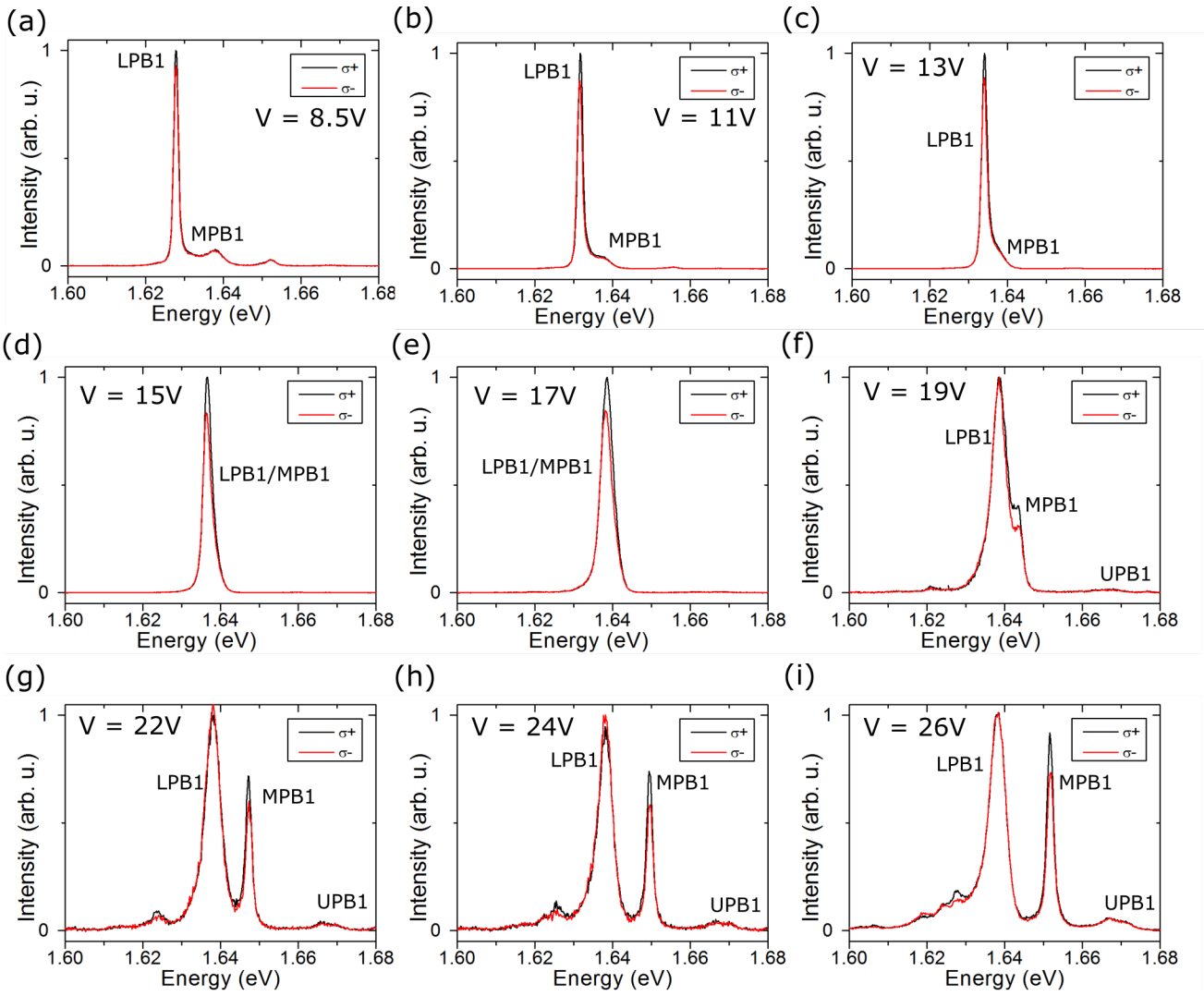

FIG. S2. Polarization resolved spectra when the cavity mode is tuned through the $MoSe_2$ trion resonance (z-piezo voltages from 8.5V to 26V). Piezo voltages correspond to slices from in Figure 2b of main text. (a) 8.5V (b) 11V (c) 13V (d) 15V (e) 17V (f) 19V (g) 22V (h) 24V (i) 26V.

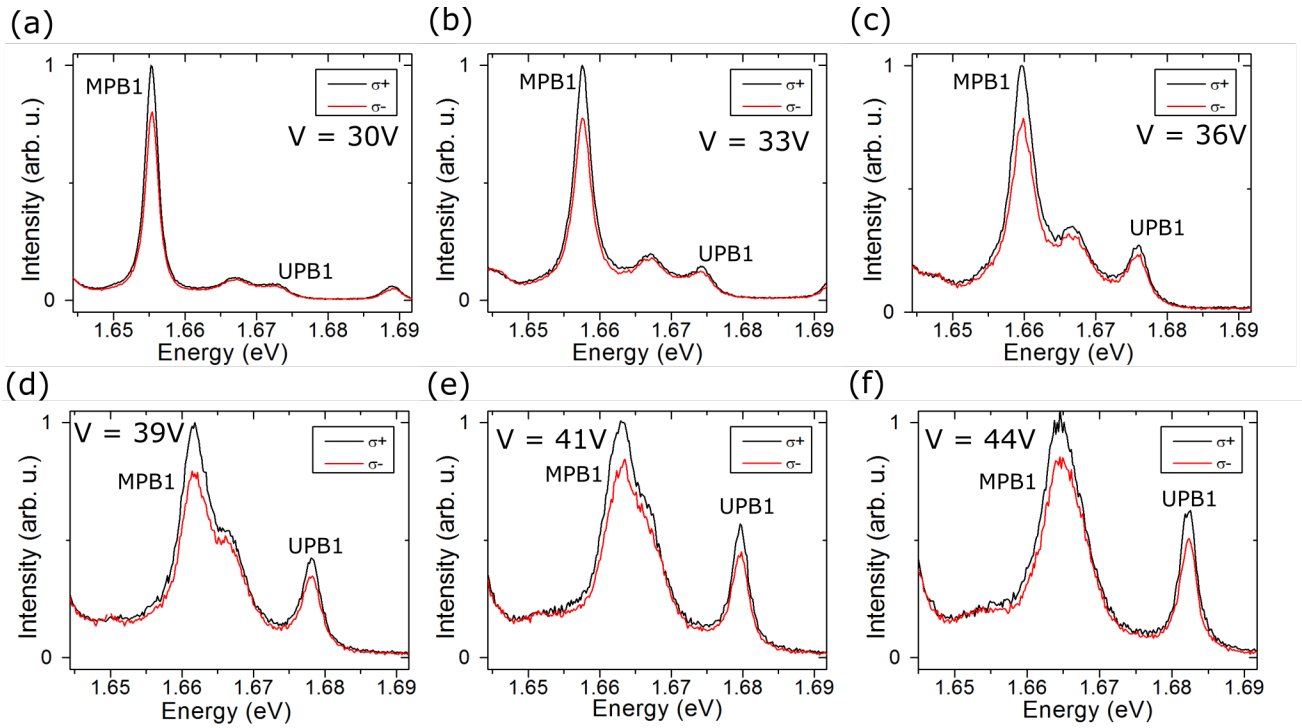

FIG. S3. Polarization resolved spectra when the cavity mode is tuned through the $MoSe_2$ neutral exciton resonance. Piezo voltages correspond to slices from in Fig. 2a of main text. (a) 30V (b) 33V (c) 36V (d) 39V (e) 41V (f) 44V. Polariton branches can clearly be resolved with a Rabi splitting of 15.2 meV which is significantly larger than the polariton linewidths. Varying degrees of retained valley polarisation are present for the polariton branches which strongly depend upon the exciton-photon detuning as discussed in the main text.

SUPPLEMENTARY NOTE 3: DATA FITTING PROCEDURE

Estimate of Rabi splitting and Polarisation Degree

Polariton branches are fitted with lorentzian functions in order to extract the peak positions to estimate the Rabi splitting. The Hamiltonian describing the coupling between the exciton, trion and photon is given by:

$$H = \begin{pmatrix} E_X & 0 & V_X \\ 0 & E_T & V_T \\ V_X & V_T & E_C \end{pmatrix} \tag{S10}$$

where E_X, E_T and E_C are the exciton, trion and cavity energies respectively. V_X and V_T are the exciton-photon and trion-photon coupling strengths.

The peak positions are obtained from a fit of multiple lorentzians to the various peaks using a non-linear least squares method. The peak positions are then simultaneously fitted with the analytical solution to the Eqn. S10 where all parameters are shared in the fitting procedure. The cavity mode energy is assumed to follow the relationship $E_c = y_0 + aV + bV^2$ where the coefficients a and b take into account the linear and nonlinear change in mirror separation as a function of applied piezo voltage (V). The resultant fit is shown in Fig. S8 where the fitted Rabi splittings for trion-cavity and exciton-cavity couplings are 0.4 ± 0.5 meV and 15.2 ± 0.1 meV respectively. LPB and MPB peak positions are poorly reproduced around the trion resonance. A more accurate estimate of the trion-cavity coupling strength is given in Fig. S2 where at resonance the fitted LPB-MPB separation at zero trion-cavity detuning is around 1.3 ± 0.1 meV. The polarization degree is calculated from the peak-intensity extracted from the fits of the co- and cross-polarized data.

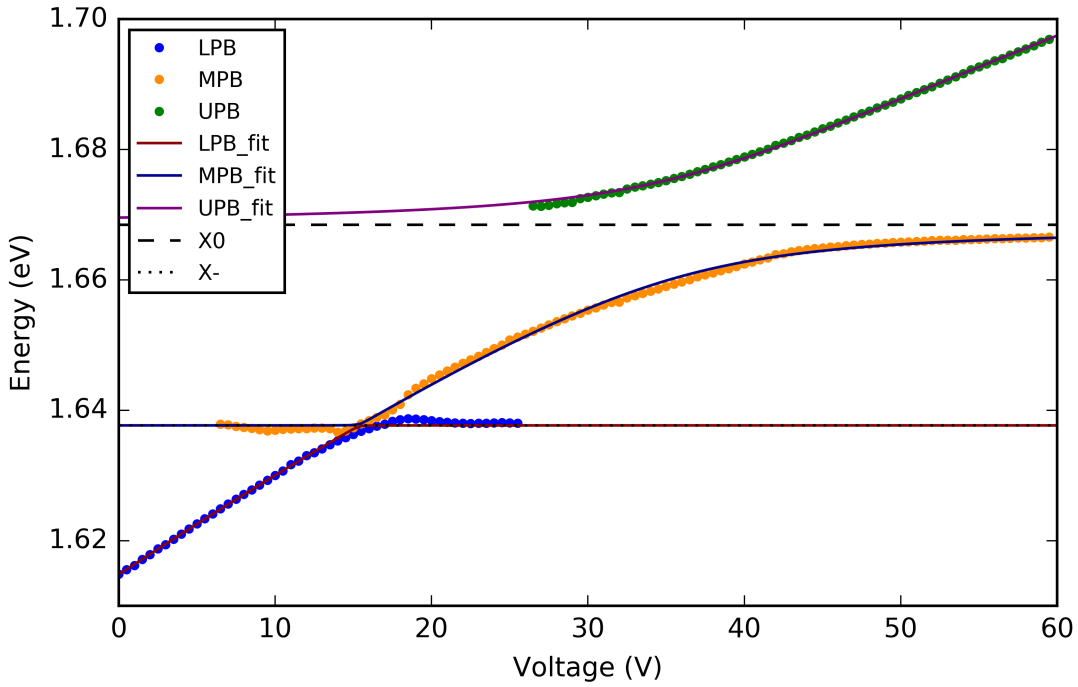

FIG. S4. Polariton peak energies fitted with 3-level coupled oscillator model.

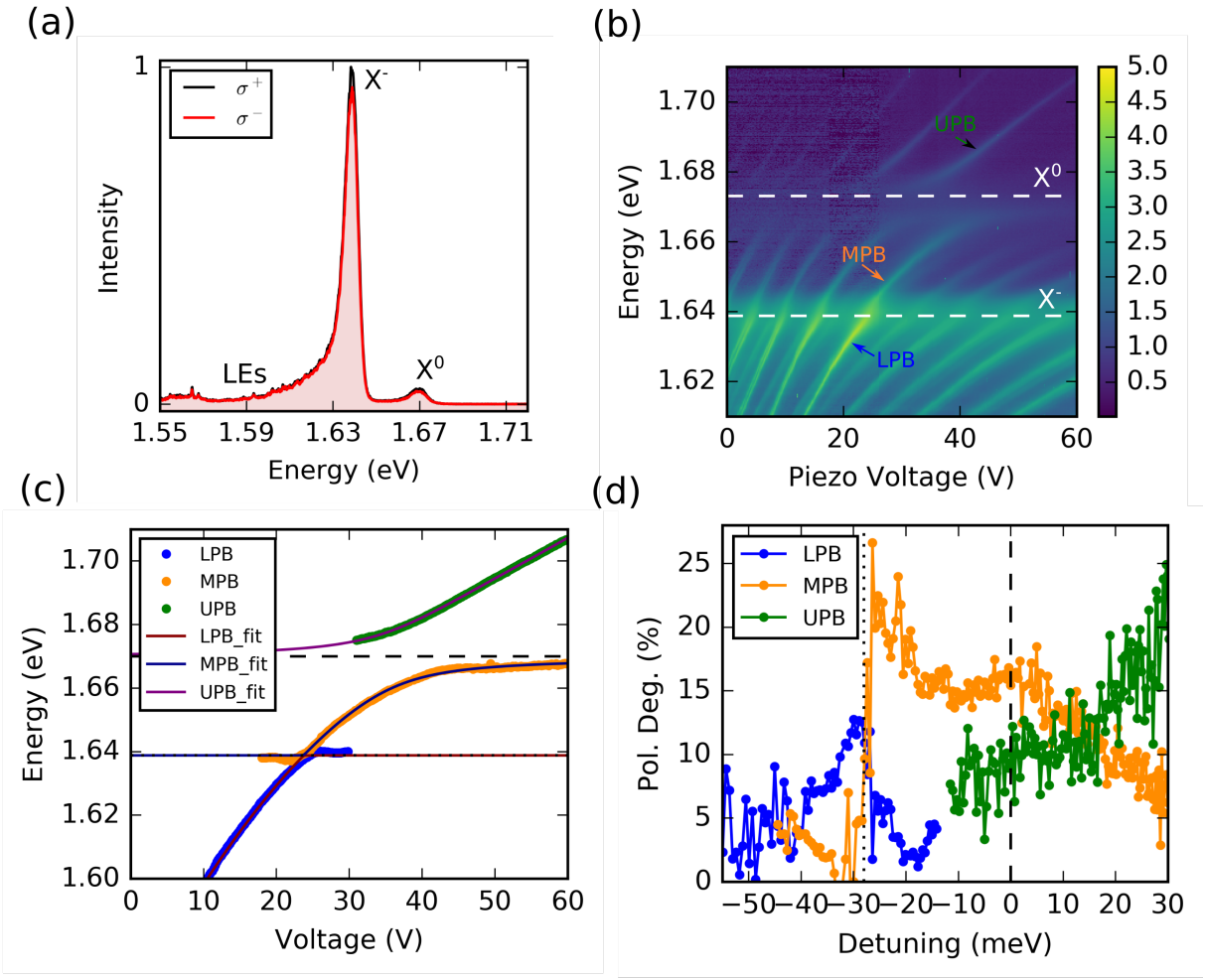

FIG. S5. Data for second sample consisting of hBN/MoSe2 heterostructure. (a) PL spectra of $MoSe_2$ under σ^+ excitation and co- (black) and cross- (black) polarized detection. The polarization degree is 8% and 3% for the exciton and trion respecively. (b) Anticrossing between the ground state LG_{00} mode and the exciton energy. (c) Fitted peak positions giving neutral exciton Rabi splitting of 18.0 ± 0.1 meV. Fitting the LPB/MPB positions around the trion resonance is poorly reproduced. A more accurate estimate of the trion-cavity coupling is given through similar fitting as for sample 1 in Fig. S2 which gives a separation between LPB and MPB at resonance of 1.5 ± 0.2 meV. (d) Polarisation degree of the polariton branches as a function detuning.

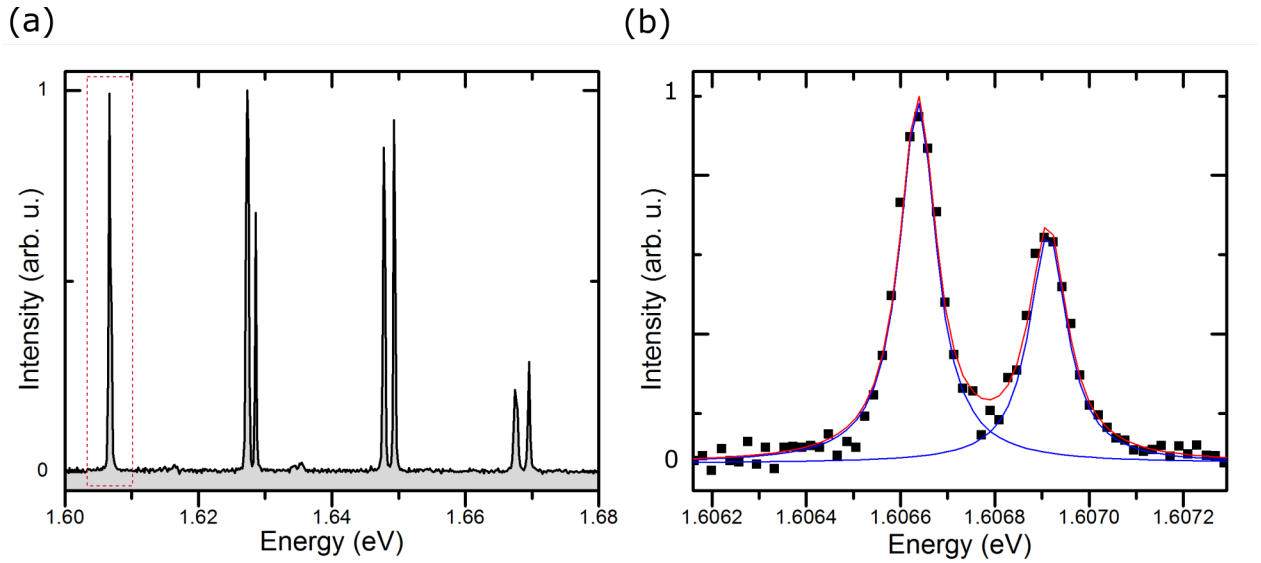

FIG. S6. (a) Cavity mode structure showing ground state (red box) and higher order transverse modes, which arise due to the harmonic like photonic confinement potential, in the weak coupling regime. (b) Spectrum of the TE-TM split ground state mode. The TE-TM splitting is 270 μeV.

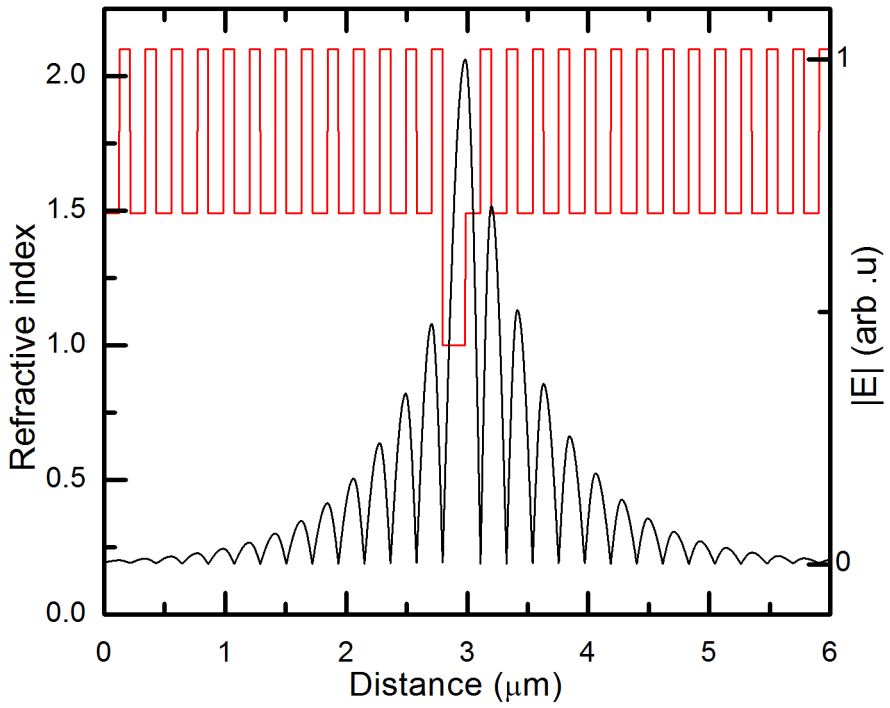

FIG. S7. Electric field distribution of microcavity calculated using the transfer matrix method. The formed tunable microcavity consists of two dielectric DBRs seperated by a $q\lambda/4n$ gap, where q is the longitudinal mode index. The top concave DBR terminates with high index material giving a node at the surface while the bottom planar DBR terminates with low index giving an antinode. The TMD monolayer is placed at the surface of the bottom DBR at an electric-field antinode. The DBRs used in the main manuscript were 10 paired SiO_2/Nb_2O_5.

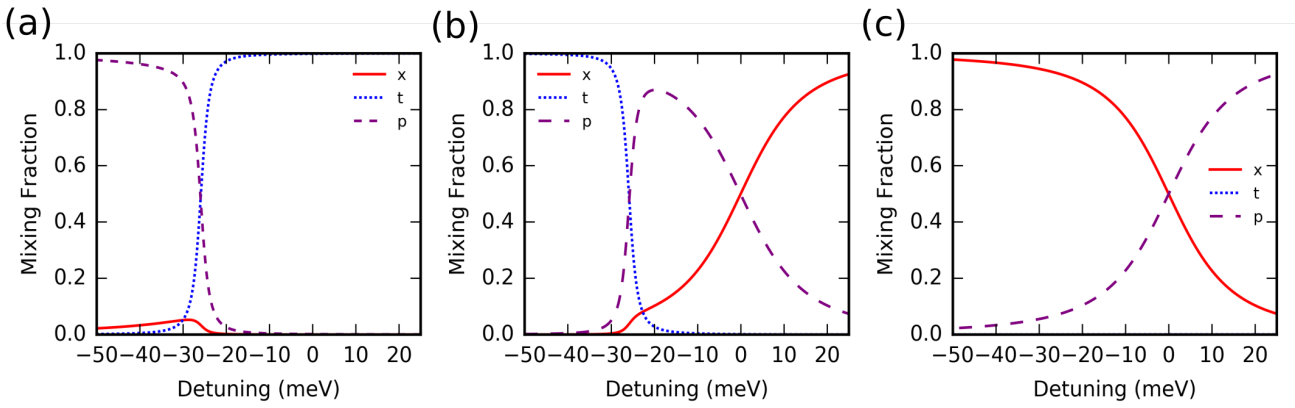

FIG. S8. Hopfield coefficients for the three polariton branches (a) LPB (b) MPB and (c) UPB used in the calculations of polarization degree as a function of detuning shown in the main text.