Supporting Information

Coherent Coupling of WS_2 Monolayers with Metallic Photonic Nanostructures at Room Temperature:

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1. Scanning electron microscope (SEM) image of the hole array sample

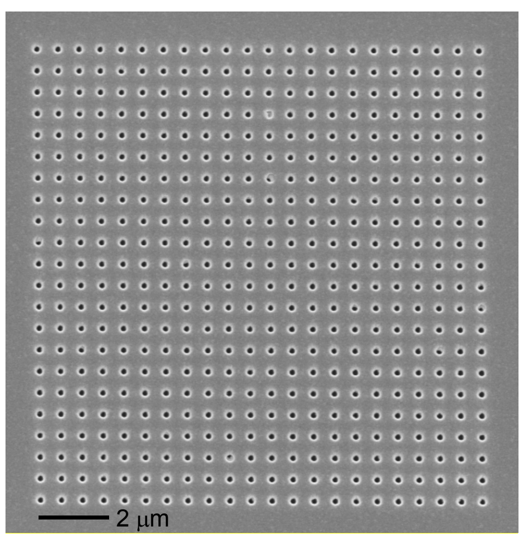

Figure S1. (a) SEM image of the plasmonic hole array. The array, milled through a 260 thick Au film, is a square array of period P = 530 nm and hole diameter D = 120 nm.

2. Dispersion of 2D materials

In Figure S2 b,d are shown the refractive indices of the WS_2 and MoS_2 monolayers respectively, extracted from the transmission spectra in Figure S2a and c. The imaginary part of the refractive indices emphasizes the exceptional absorption characteristics of the exciton band in the 2D materials. The full details of the fitting for the visible spectra are included in Table S1 and S2.

Table S1 Monolayer WS_2 dispersion parameters

Oscillator No. j	***Oscillator strength*** f_j	***Resonance energy*** $E_j(eV)$	***Damping constant*** $\Gamma_j(eV)$
1	1.59	2.0195	0.028
2	0.70	2.2379	0.20
3	2.95	2.4087	0.15
4	2.80	2.5996	0.30
5	12	2.850	0.23

Table S2 Monolayer MoS_2 dispersion parameters

Oscillator No. j	***Oscillator strength*** f_j	***Resonance energy*** $E_j(eV)$	***Damping constant*** $\Gamma_j(eV)$
1	0.65	1.9001	0.040
2	0.25	1.9315	0.050
3	1.2	2.0516	0.080
4	5	2.3065	0.8
5	12	2.4	1
6	24	2.87	0.35

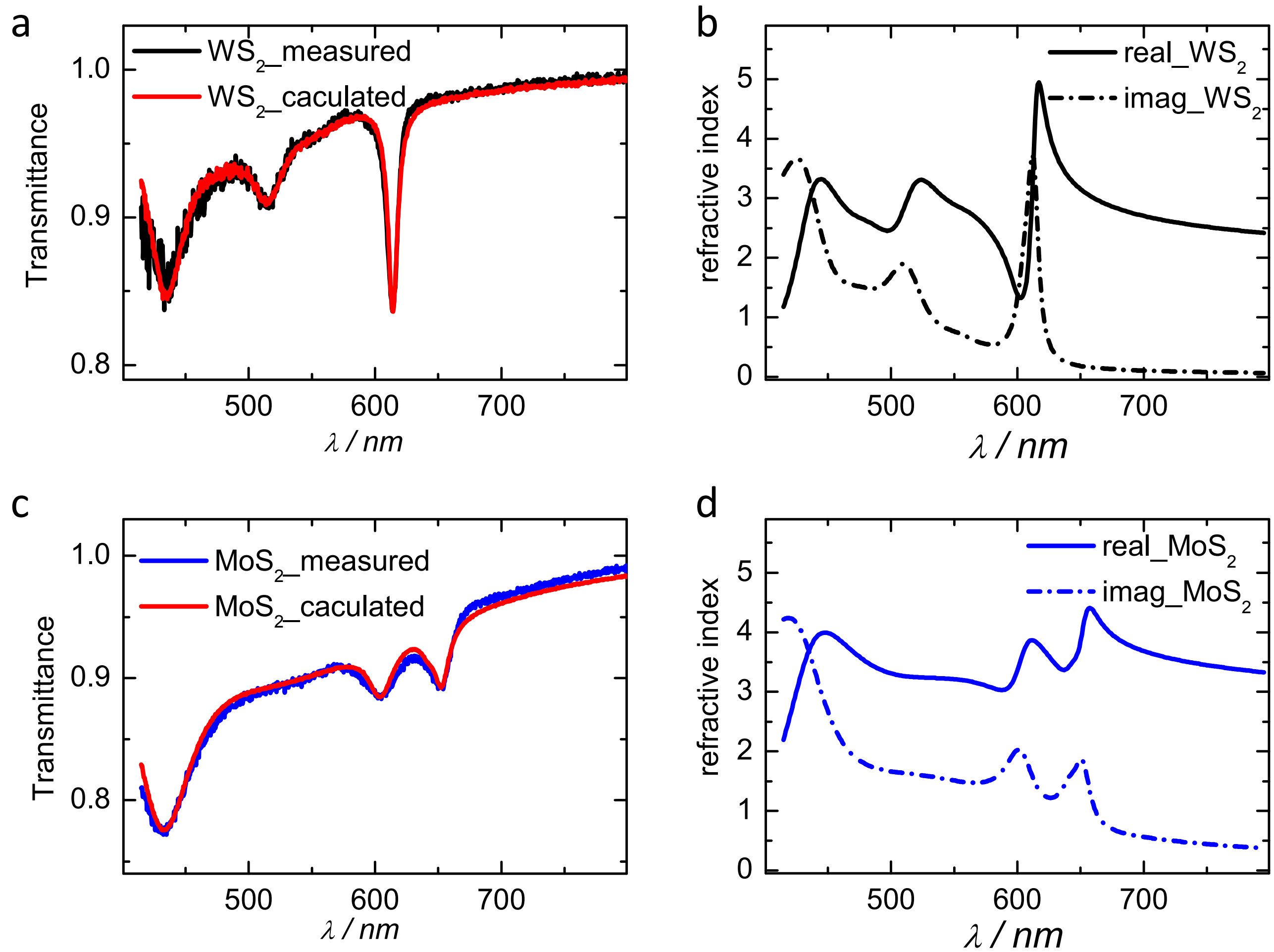

Figure S2. The real and imaginary refractive indices of monolayer WS_2 (b) and MoS_2 (d) calculated from the transmission spectra of the monolayer flake on quartz shown in (a) and (c) respectively.

3. Comparing transmission, reflection of FP cavities simulated by transfer matrix method

In order to model the spectral properties of the FP cavities, we used the standard formalism of the transfer matrix method (TMM).[1] It relies on solving Maxwell's equations at each interface of a multilayer stack, each layer being characterized by a complex refractive index. The refractive index of the silver mirrors (50 nm thick) and of the LiF spacer layers were obtained from regular data bases.[2-3] The complex refractive indices of monolayer WS_2 and MoS_2 are taken from the data in Figure S2. The thickness of the two LiF spacers were tuned to 86 nm for WS_2 and 92 nm for MoS_2 respectively and those of the WS_2 and MoS_2 monolayers were considered as 0.618 nm and 0.646 nm respectively.[4] All the parameters of the stack having been determined, the TMM was used to model the transmission/reflection spectra of WS_2 and MoS_2 cavities at normal incidence conditions. The splitting in reflection is compared to that in transmission as shown in Figure S3. The splitting in reflection $\hbar\Omega_{R\_R}$ = 67 meV is slightly smaller than that in transmission $\hbar\Omega_{R\_T}$ = 70 meV for the case of WS_2 which has very sharp *A* exciton absorption band. The predicted Rabi-splitting is smaller than that of experimental results (101 meV) likely due to errors in the assumption of the thickness of the monolayer and the refractive index of the silver mirrors. For MoS_2, the splitting in reflection ~ 41 meV and ~50 meV in transmission. The average splitting ratio (1.51) observed for WS_2 compared to MoS_2 cavity is comparable with the ratio of exciton transition dipole moment in each case $\sqrt{\frac{1.59}{0.65}} = 1.56$.

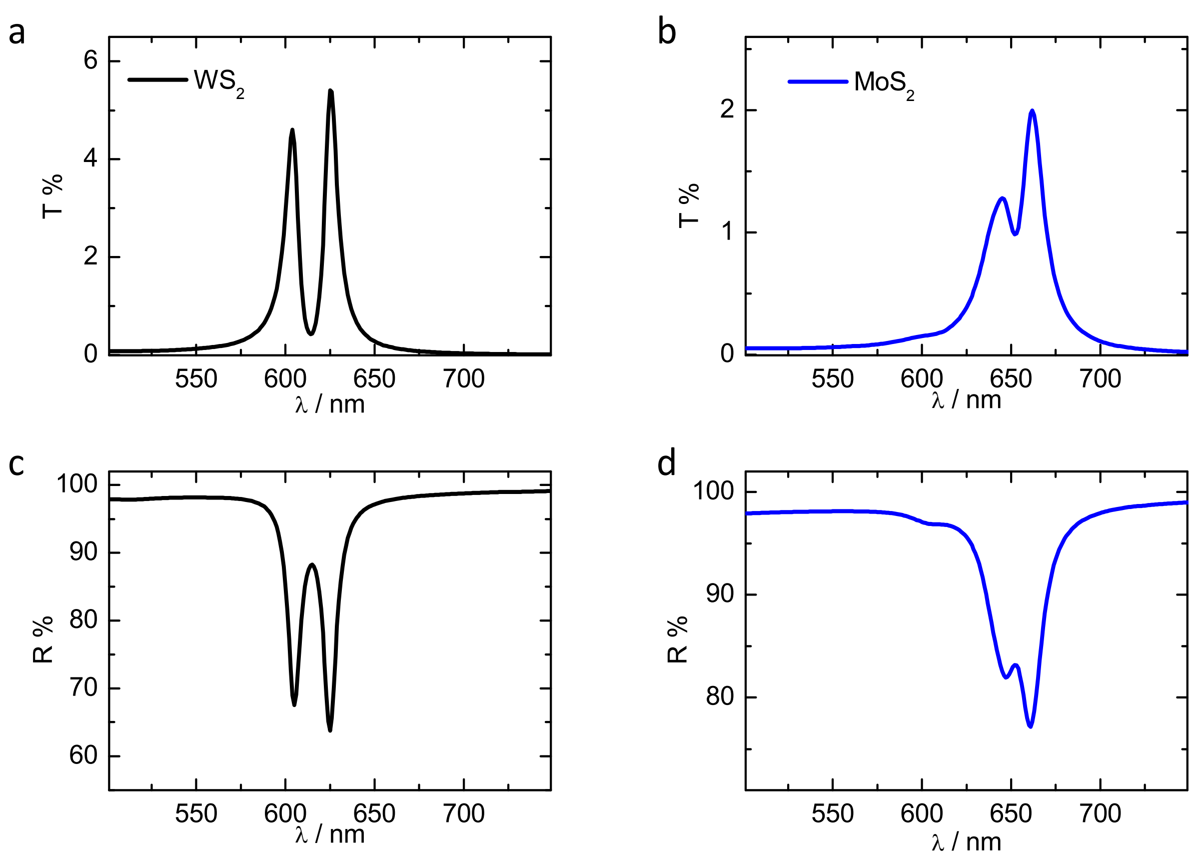

Figure S3. Simulated transmission (a) (b) and reflection (c) (d) of monolayer WS_2 (a) (c) and MoS_2 (b) (d) sandwiched in the middle of FP cavities tuned such that the fundamental mode is resonant with the *A* exciton transition for each case.

4. Bare optical modes

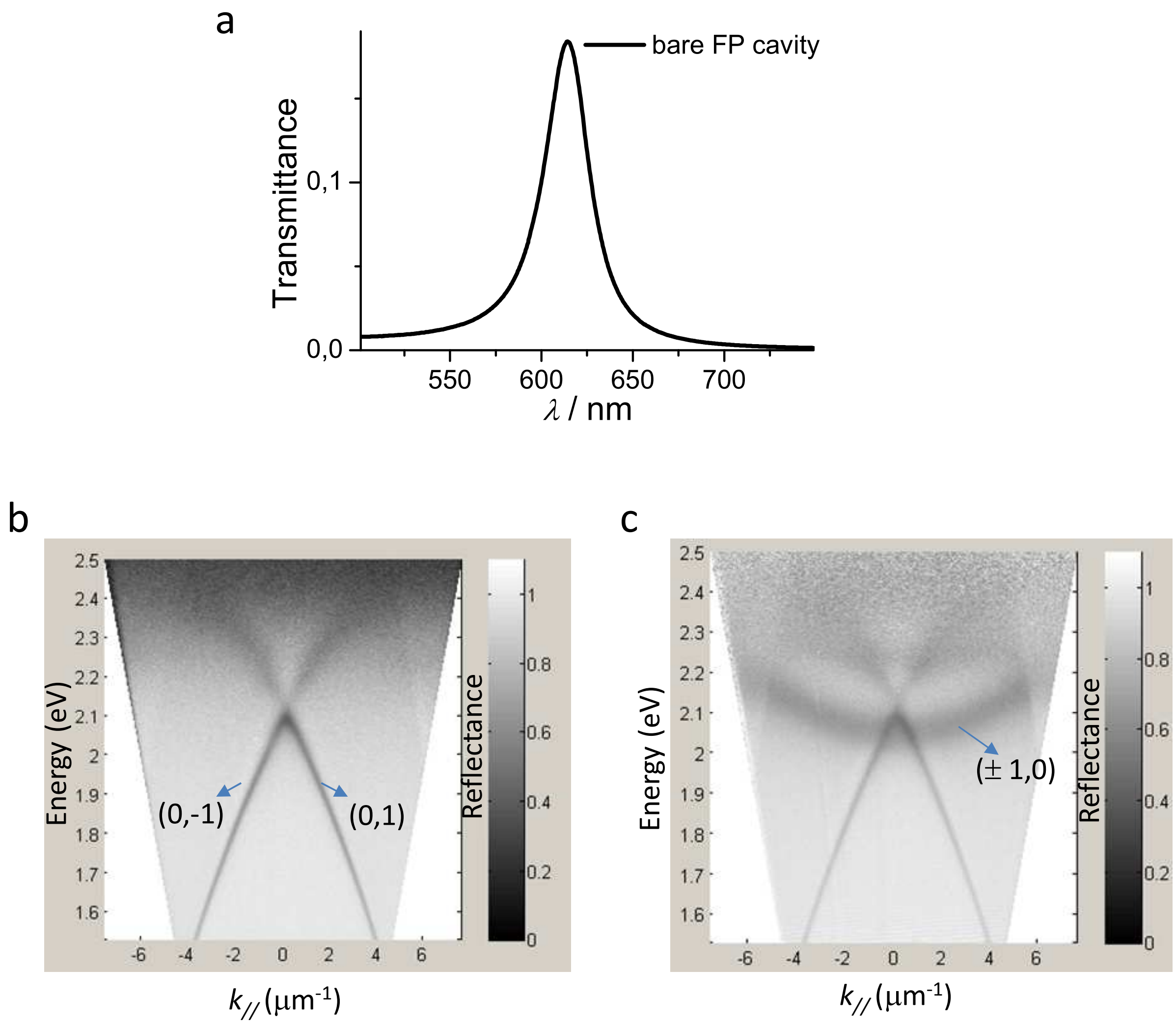

Figure S4 Measured transmission spectrum of the bare cavity (a) and reflection dispersion of the plasmonic hole array in TM polarization (b) and without polarization selection (c).

5. Mixing coefficients for strong coupling of monolayer WS_2 with the TE FP cavity mode

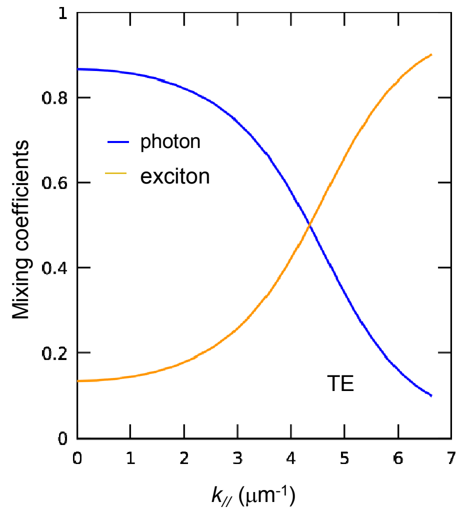

Figure S5 The mixing coefficients of the P- band for coupling of the WS_2 monolayer with the TE FP cavity mode. Blue and yellow curves represent the photonic and excitonic content of the P- band respectively.

6. PL spectra at resonance for the strong coupling of monolayer WS_2 with the TM/TE modes of the plasmonic hole array

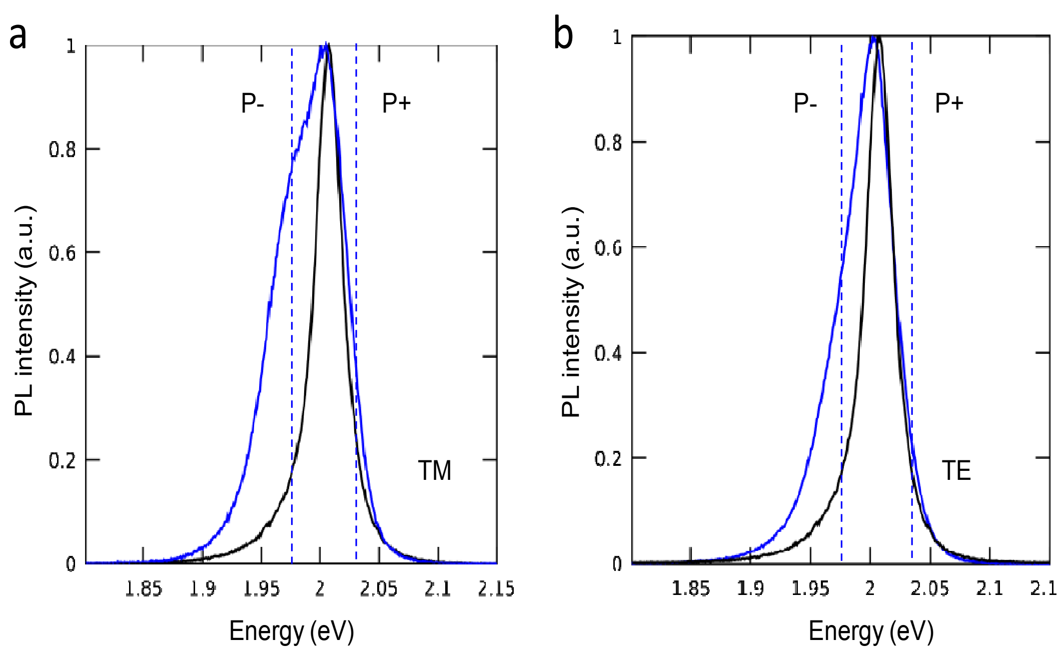

Figure S6. PL spectra (solid blue curve) of the plasmonic hole array with monolayer WS_2 analyzed in (a) TM and (b) TE polarization. Both spectra were obtained at the resonant condition. The black solid curve is the PL spectrum of uncoupled exciton. The vertical dashed lines represent the energy of P+/-, measured in reflection.

REFERENCES

1. Schwartz, T.; Hutchison, J. A.; Genet, C.; Ebbesen, T. W., *Phys. Rev. Lett.* **2011,** *106*, 196405.

2. Johnson, P. B.; Christy, R. W., *Phys. Rev. B* **1972,** *6*, 4370-4379.

3. Li, H. H., *J. Phys. Chem. Ref. Data* **1976,** *5*, 329-528.

4. Li, Y. L.; Chernikov, A.; Zhang, X.; Rigosi, A.; Hill, H. M.; van der Zande, A. M.; Chenet, D. A.; Shih, E. M.; Hone, J.; Heinz, T. F., *Phys. Rev. B* **2014,** *90*, 205422.