Ultrafast coherent polariton oscillations in a WS_2 monolayer-based microcavity at room temperature

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Strong coupling between excitons in semiconducting materials and photons in optical resonators can lead to the formation of new hybrid states called upper and lower polaritons (UP, LP). A periodic, coherent energy exchange between the coupled modes can be observed through ultrafast optical experiments as quantum beats between the polariton states (polariton Rabi oscillations), with an oscillation frequency given by the UP-LP energy separation. Coherent polariton oscillations have been studied in a variety of systems[1, 2] for their potential use in all-optical switches[3] and quantum information[4]. Excitons in transition metal dichalcogenides (TMDs) monolayers exhibit spin-valley locking and display very large binding energies even at room temperature[5], making them a promising platform for novel applications of strong coupling regime. However, observations of Rabi oscillations in such systems are still elusive. In this work, we show ultrafast coherent polariton oscillations in a planar microcavity made of a large area WS_2 monolayer. Through k-space imaging, we retrieve the photonic dispersion of our cavity, clearly displaying the polariton branches. The dispersion is reported in Fig.1a alongside a coupled oscillators model fit, resulting in a Rabi splitting value of $2\hbar\Omega = 37meV$, leading in theory to Rabi oscillations with a period of 112 fs. Then, by using transient reflectivity (TR) and two-dimensional electronic spectroscopy with sub-20-fs temporal resolution, we characterize the ultrafast dynamics of this sample, observing oscillations at room temperature with a period of about 87 fs, as shown in Fig.1b. To demonstrate the polaritonic nature of these oscillations, we performed TR measurements by changing both the pump fluence and the angle of incidence of both pump and probe with respect to the sample. Figure 1c reports the oscillating contributions to the fitted dynamics, from which it is clear that the oscillation period changes with either the fluence or the angle, following the variations of UP-LP energy separation. Such results can pave the way for room-temperature all-optical micro and nano-photonic devices, for applications in quantum information and quantum coherent transport, also exploiting the valley pseudo-spin degree of freedom of TMDs.

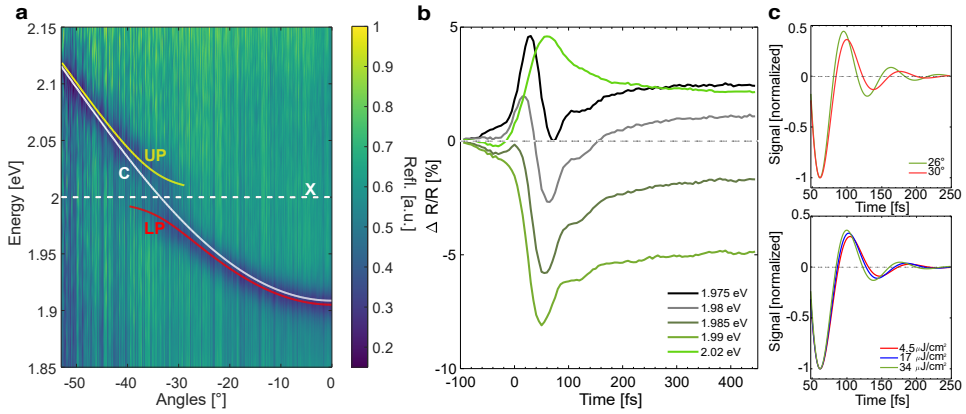

Figure 1: (a) Reflectivity map of the WS_2 microcavity as a function of angle and energy. The fit of the UP and LP are reported in yellow and red, respectively, alongside with the exciton (X, dashed white line) and the cavity (C, solid white line) energies. (b) Temporal dynamics of the microcavity measured at specific energies, extracted from the TR measurements. (c) Damped oscillator contributions of the fitted dynamics, showing different periods upon changing the pump fluence or angle.

References

[1] Trifonov A. V., et al. *Physical Review B* **95**, 15, 155304 (2017).
[2] Timmer D., et al. *Nature Communications* **14**, 1, 8035 (2023).
[3] Ridolfo A., et al. *Physical Review Letters* **106**, 1, 013601 (2011).
[4] Pashkin Y. A., et al. *Nature* **421**, 6925, 823-826 (2003).
[5] G. Wang, et al., *Reviews of Modern Physics* **90**, 2, 021001 (2018).

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