Supplementary Materials

Room Temperature Non-Equilibrium Bose-Einstein Condensation of Dirac Polaritons in WS_2 Monolayer

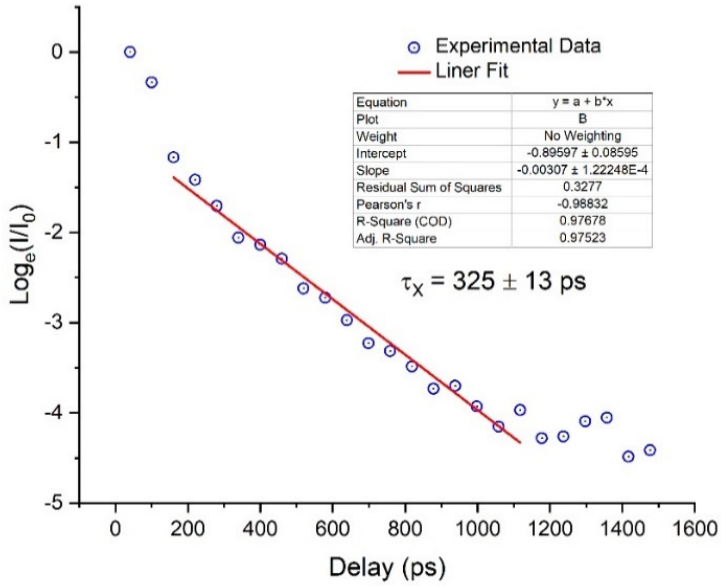
Bruno Miranda, [1]† Silvia Romano,[1]† Scott Duhey,[2] Aidar Kemelbay,[2] Arian Gashi,[2] Vito Mocella,[1] Ivo Rendina,[3] Adam M. Schwartzberg,[2] Gianluigi Zito[1]*

[1]*Institute of Applied Sciences and Intelligent Systems, National Research Council, 80131, Naples, Italy*

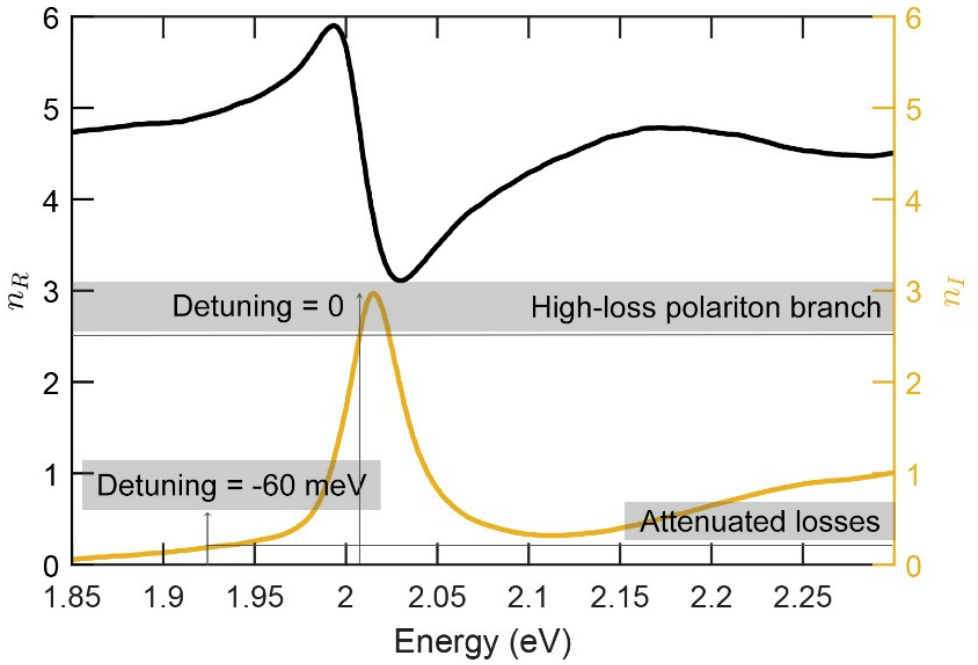
[2]*Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, United States.*

[3]*Institute of Applied Sciences and Intelligent Systems, National Research Council, 80078, Pozzuoli, Italy.*

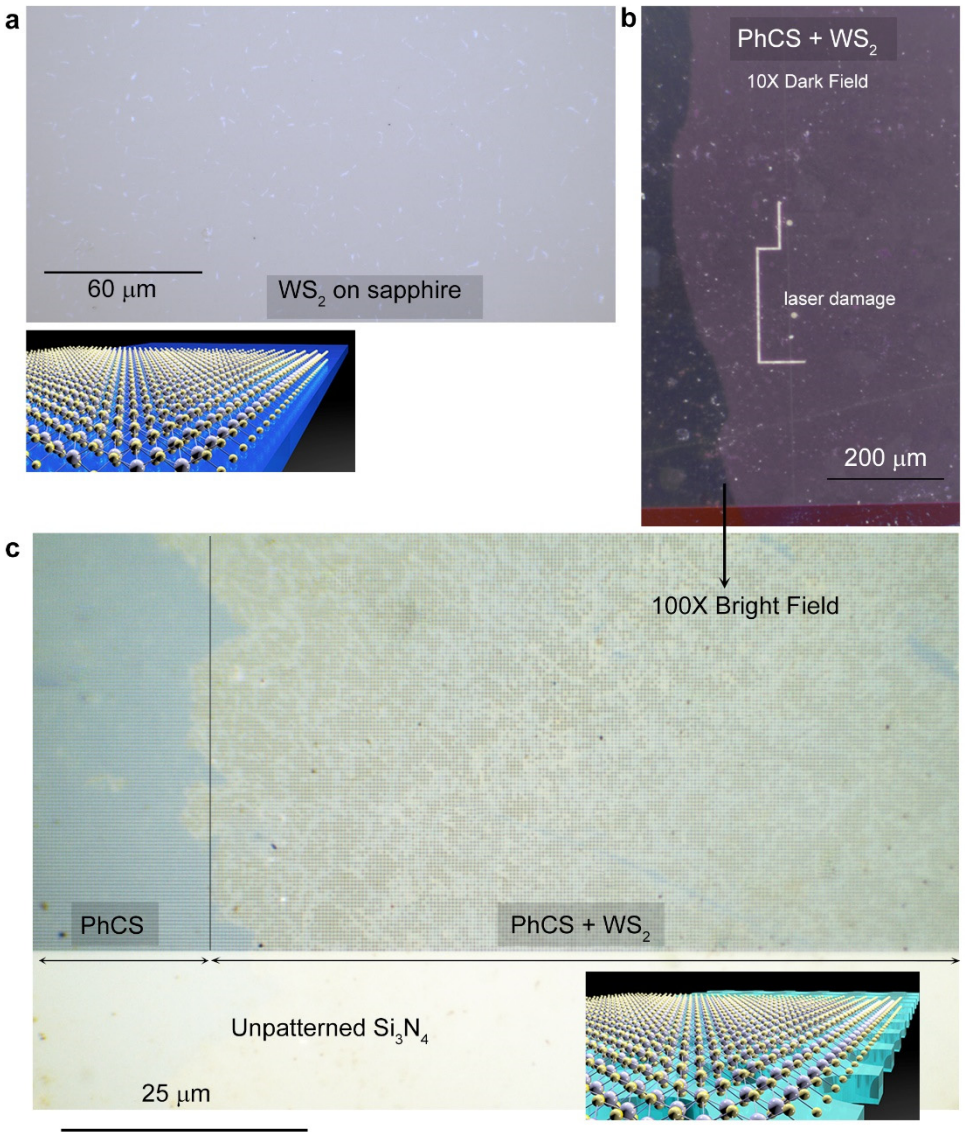
This document contains additional notes and figures supporting the conclusions in the main text.



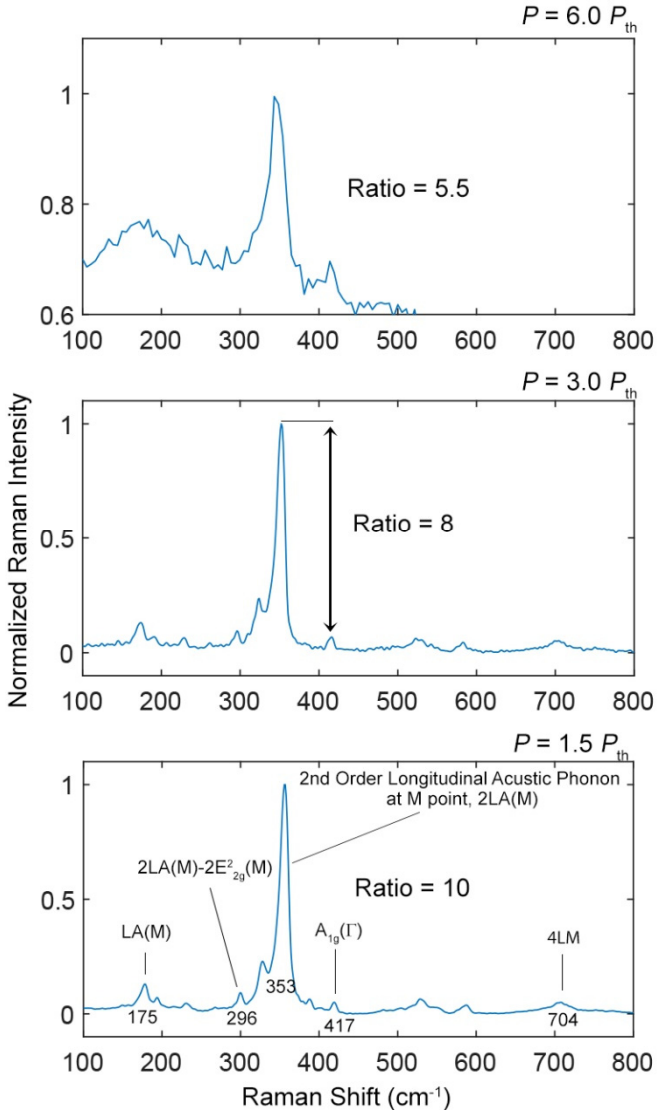
Supplementary Figure 1. Intensity decay as a function of delay from the pump pulse (circles) and fit (solid line).



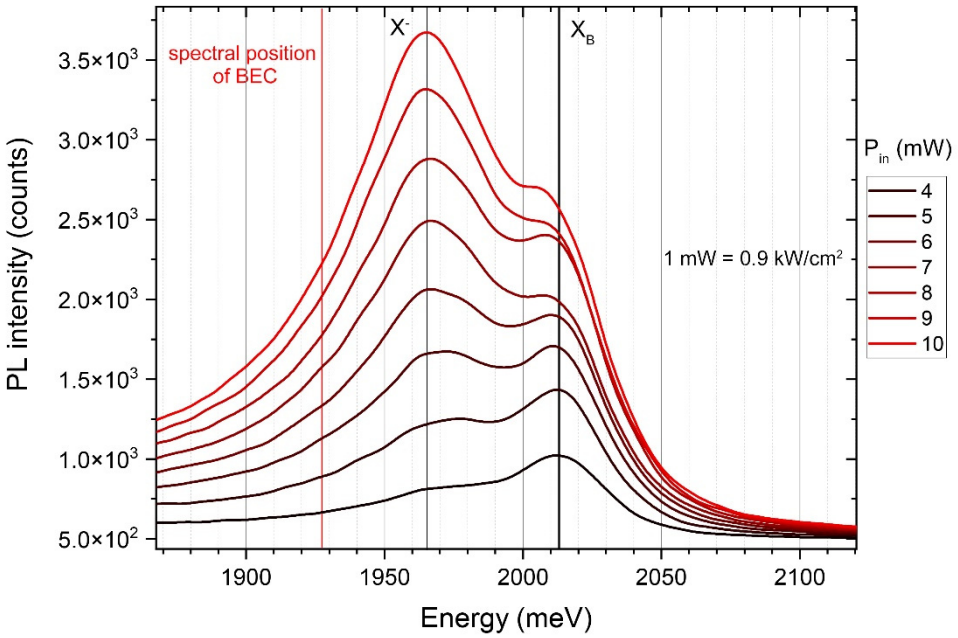
Supplementary Figure 2. Measured complex refractive index $n_R + i\,n_I$ dispersion of WS_2 monolayer: the imaginary part of refractive index reduces significantly with negative detuning.



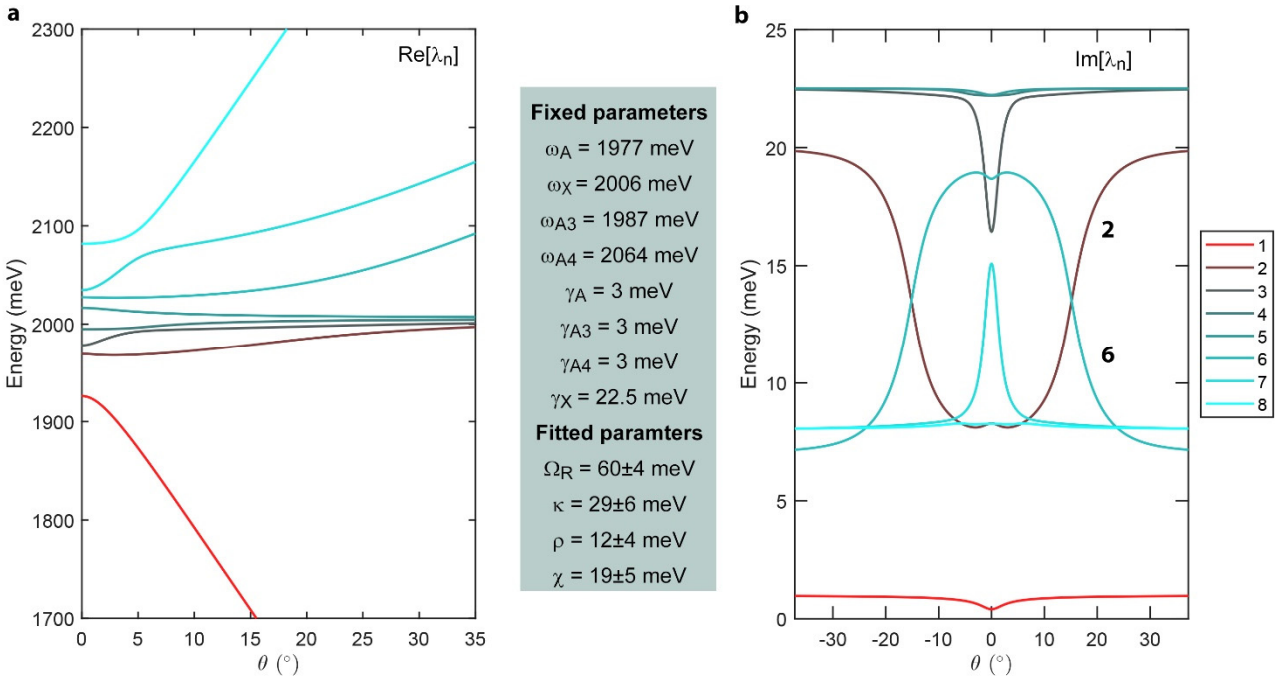
Supplementary Figure 3. Sample inspection at the optical microscope. a, WS2 monolayer on sapphire substrate. b, Dark field microscopy of the WS2 monolayer transferred on a 1-mm^2 square-lattice PhCS, revealing overall good homogeneity of the film. c, Bright-field detail showing the homogeneity of the monolayer over the lattice of the patterned slab. The site is at the boundary between uncovered, unpatterned and covered regions of the PhCS.



Supplementary Figure 4. Measured Raman spectrum of WS_2 transferred on the sample, denoting relative peak intensities characteristic of a single atomic layer according to Ref. [1]. Input wavelength 532 nm. Bottom and middle panels: objective N.A. 0.95, 100×, grating 600 l/mm. Top panel: objective N.A. 0.2, 10×, grating 600 l/mm (polariton BEC excitation/detection conditions). Pump power ranging from 1.5 to 6 P_{th}. According to Ref [2], the evolution of the relative amplitude ratio between 2LA and A_{1g} vibration modes indicate stable operating conditions up to 6 P_{th} with charge carrier photodoping below Mott threshold, and local heating temperature below 320 K.



Supplementary Figure 5. Room temperature photoluminescence (PL) evolution of bare WS_2 after transfer on the sample, denoting charged exciton (X^-) contribution for increasing input power, together with the neutral exciton (X^0) emission. The threshold for BEC is at 5 mW. BEC ground state emerges at circa 1930 meV, then blue-shifting at saturation up to approximately 1960 meV (**Figure 2f** in main paper). At saturation pump level, there might be an interaction of the condensate with charged excitons given the spectral proximity. However the depletion of emission from MP_1 branch, compared to the nonlinear increase of nLP emission, suggests a possible decrease of X^- population as the condensate population increases.



Supplementary Figure 6. a, Eigenfrequencies and **b**, corresponding linewidths of the polariton branches obtained by fitting the experimental angle-resolved PL spectra below threshold. Notably, near the avoided crossing at approximately 17.5°, a clear loss exchange (or linewidth swap) occurs between eigenmodes 2 and 6. This behavior reproduces the experimentally observed PL evolution in Figure 2b: mode 6 exhibits decreasing PL while mode 2 shows increasing PL after the crossing since the intensity is proportional to the radiative decay expressed by the linewidth.

References

[1] Berkdemir, A., Gutiérrez, H., Botello-Méndez, A. et al. Identification of individual and few layers of WS2 using Raman Spectroscopy. Sci Rep 3, 1755 (2013). https://doi.org/10.1038/srep01755

[2] Yu, Y., Yu, Y., Huang, L., Peng, H., Xiong, L., & Cao, L. Giant gating tunability of optical refractive index in transition metal dichalcogenide monolayers. Nano Letters, 17(6), 3613-3618 (2017). https://doi.org/10.1021/acs.nanolett.7b00768