Supplementary Information (SI) for Nanoscale.

Supporting information file for:

Thermotransmittance spectroscopy of layered crystals using lab on fiber

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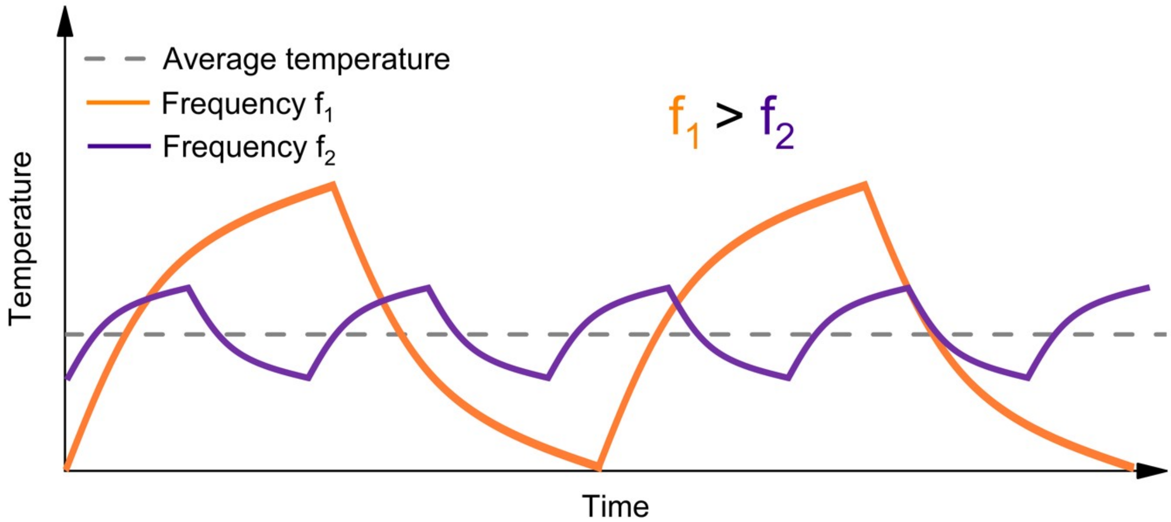

Figure S1. The diagram illustrates the temperature variations over time at two different frequencies. At lower frequencies, the fluctuations have a larger amplitude, leading to stronger resonance, while at higher frequencies, the amplitude decreases, resulting in weaker resonance. The average temperature stays consistent, maintaining the same energy position.

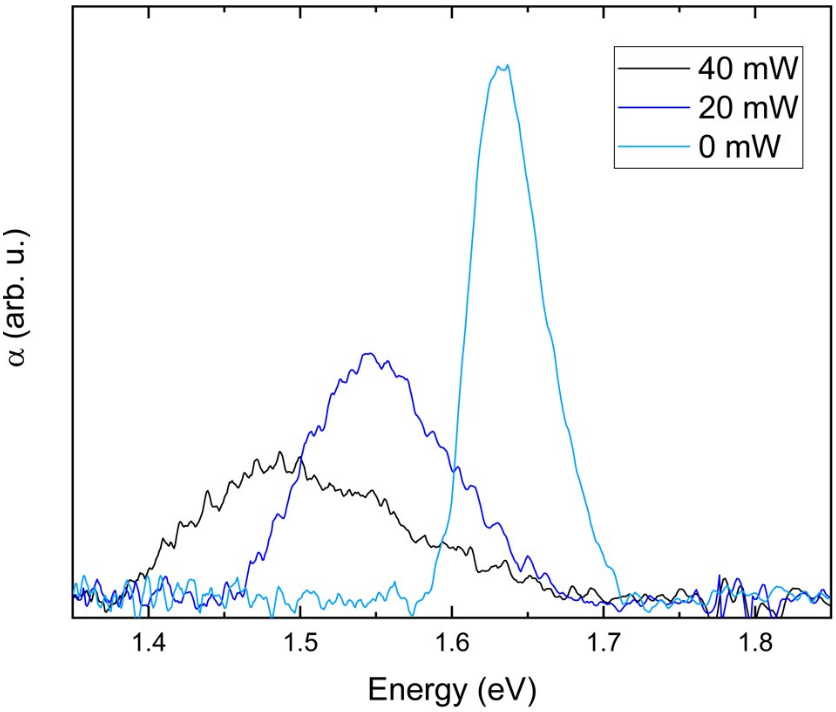

Figure S2. Variation of broadening in the WSe_2 sample with the change of laser power, at 0 mW, the linewidth is 84 meV, increasing to 107 meV at 10 mW, and further widening to 153 meV at 40 mW.

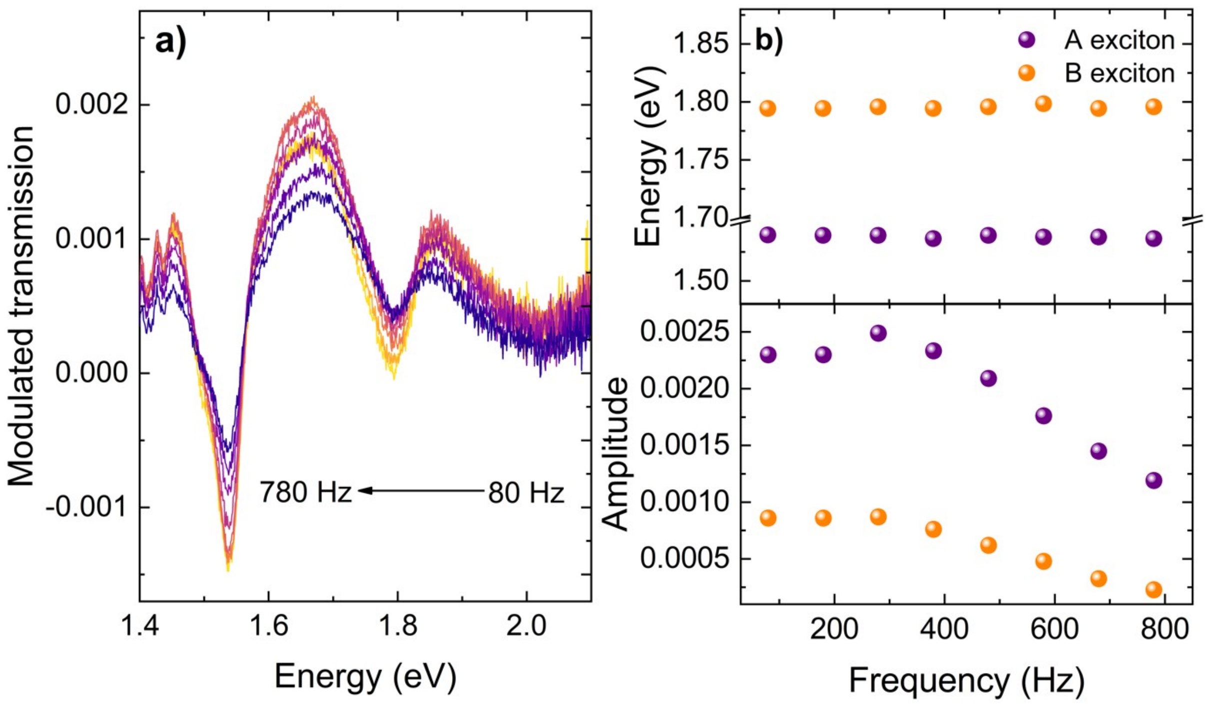

Figure S3. (a) Photoreflectance spectra of $MoSe_2$ obtained at different frequencies in the one-beam experiment, (b) present the results of the analysis for the spectra included in panel (a), i.e., energy and amplitude of optical transitions in the function of frequency.

Detailed Discussion on the Thermal Effects in the TMDC Samples

1. **Low Heat Capacity and Rapid Heating**

The thermal behavior of the TMDC sample is strongly influenced by its low heat capacity, given its small volume due to the thinness of the material and limited lateral size. The heat capacity (C) of the material can be described by:

$$C=c_p m=c_p \rho V$$

where c_p is the specific heat capacity, ρ is the material density, and V is the volume. Due to the extremely small volume of the TMDC layer, the energy required to raise its temperature is minimal. As a result, even small amounts of absorbed laser radiation can cause significant temperature fluctuations in the material. This relationship can be further understood by the equation:

$$Q=C\Delta T$$

where Q is the energy absorbed by the sample, and ΔT represents the temperature change. Since, as already mentioned, the TMDC layer has a low heat capacity, even a small amount of absorbed energy leads to a large temperature increase, resulting in rapid heating.

2. **Limited Thermal Conductivity and Heat Diffusion**

The other important factor is the limited thermal conductivity in TMDC materials, which significantly contributes to the thermal effects observed during the experiments. Heat conduction is described by Fourier's law:

$$q=-k\nabla T$$

where q is the heat flux, k is the thermal conductivity, and ∇T is the temperature gradient. In the case of TMDC materials, the thermal conductivity is much lower in the direction perpendicular to the plane compared to the direction parallel to the plane due to the weak van der Waals bonds between the layers. This affects the heat dissipation from the spot illuminated by the laser. The last factor that also plays a critical role in the thermal effect is the quality of the interface between the studied crystal and the fiber, which, together with the poor thermal conductivity of the fiber, leads to heat accumulation within the TMDC material.

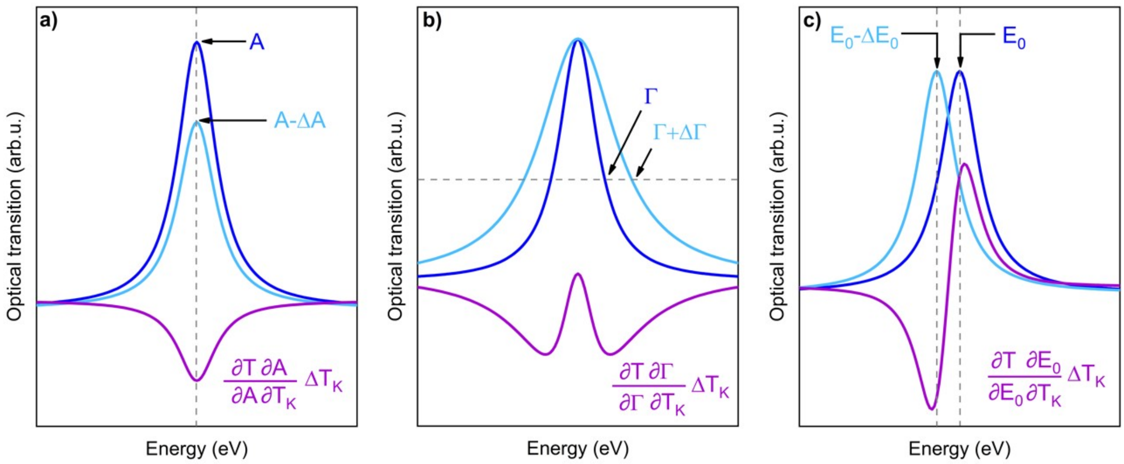

Figure S4. The impact of varying individual parameters of the Lorentzian function on the spectral shape related to temperature variation, along with their corresponding differential spectra (purple line). Results of: (a) a decrease in amplitude A, (b) an increase of broadening Γ, which progresses with temperature, and (c) a change in energy position, specifically, a redshift of the resonance energy E0.

In Fig. S4, we present the impact of varying individual parameters of the Lorentzian function o the spectral shape as a function of temperature, along with their corresponding differential spectra. This analysis helps to illustrate how each parameter contributes to the observed changes in the measured transmission spectra under varying thermal conditions. The first parameter, shown in Fig. 5a, is the amplitude A. As temperature increases, a gradual decrease in the amplitude of the optical transition (light blue line) is observed, resulting in a higher resonance intensity (purple line). This behavior is accompanied by the thermal broadening of the optical transitions and is attributed to a weakening of oscillator strength at elevated temperatures. The second parameter (Fig. 5b), which, as mentioned, is the broadening Γ, increases significantly with temperature. This broadening arises from enhanced phonon interactions and other scattering mechanisms at elevated temperatures, which effectively smear the resonance peak and reduce its sharpness. Finally, Fig. 5c illustrates the shift in the resonance energy E0 with temperature. In our measurements, this parameter exhibits a redshift as the temperature rises, corresponding to the temperature-dependent bandgap narrowing commonly observed in van der Waals crystals and other semiconducting materials.[1,2]

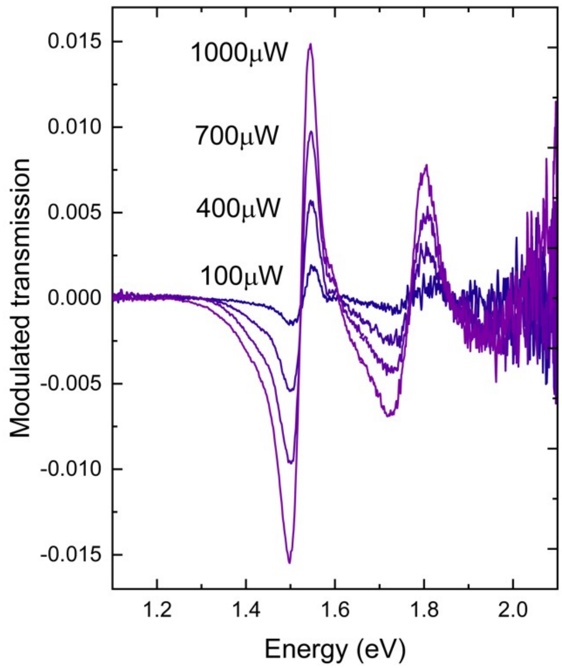

Figure S5. Modulated transmission spectra collected for the WSe_2 crystal at low laser power, showing that even at lower powers, the changes in transmitted light can be significant (e.g., ~3% for 100 μW and ~15% for 1000 μW).

References:

1 D. E. Aspnes, *Surface Science*, 1973, **37**, 418–442.

2 J. N. Hilfiker and T. Tiwald, in *Spectroscopic Ellipsometry for Photovoltaics: Volume 1: Fundamental Principles and Solar Cell Characterization*, eds. H. Fujiwara and R. W. Collins, Springer International Publishing, Cham, 2018, pp. 115–153.