Electron-phonon coupling in a WSe_2 monolayer unveiled by two-photon photoluminescence excitation spectroscopy

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The "all-surface" character of two-dimensional semiconductors leads to strong interacrions of the electronic and vibrational degrees of freedom of a monolayer semiconductor to its surroundings [1]. Here, we investigate this aspect by performing a detailed investigation of the two-photon photoluminescence excitation (PLE) spectrum of a high quality WSe_2 monolayer, which we show in Fig. 1(a).

The low-temperature photoluminescence (PL) and reflection contrast spectra, shown in Fig. 1(b), show resonances of neutral and charged exciton species, as well as higher order excitonic transitions, like the 2s state of the A exciton. Figure 1(c), is a false-color map of the two-photon PLE experiment, with the horizontal axis corresponding to the emission energy and the vertical axis corresponding to two times the excitation energy. From the PLE spectrum, we extract the intensity of the two-photon PL of the exciton and plot it in Fig. 1(b), where we see pronounced resonances related to the absorption of the 2p and 3p states. We show in Fig. 1(d,e) selected PL spectra and close-up views, excited at the energies indicated in Fig. 1(c), respectively. In Fig. 1(e), we note a peak, separated by 108 meV from the doubled energy of the laser. Its shifts by the exact same amount as the doubled energy of the laser is a distinctive sign of its phonon-related nature. Ongoing work is devoted to a more precise understanding of this phonon line.

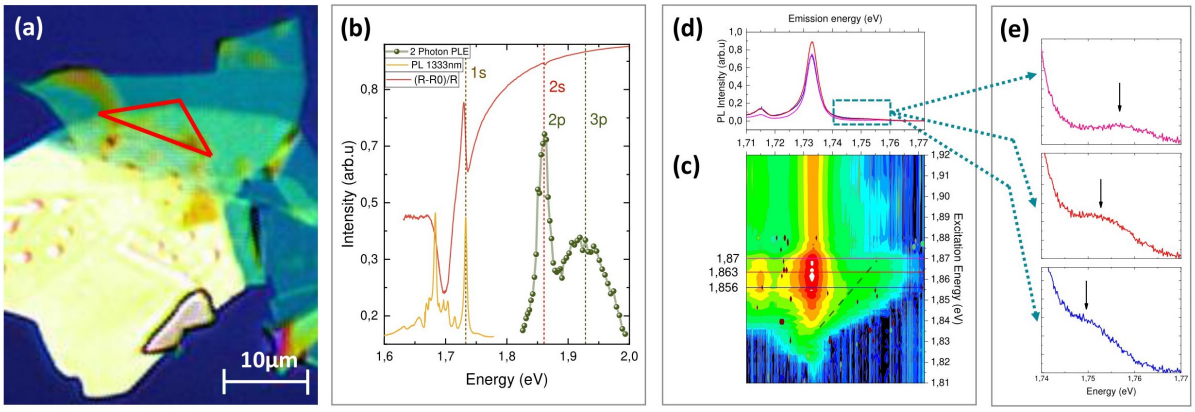

Figure 1: (a) Micrograph of the hBN-encapsulated $WSe2_2$ flake. The monolayer region is framed by the red triangle. (b) PL and reflectivity contrast and two-photon PLE spectra of the WSe_2 monolayer. The relevant excitonic resonances are indicated by dashed vertical lines. (c) False color map depicting two-photon PLE spectra. (d) Two-photon PL spectra extracted at selected excitation energies. (e) Close-up of the PL spectra. The phonon line shifting with the excitation laser is indicated by arrows.

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