Carrier dynamics in InSe and the impact of terahertz pulses

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***Abstract*—We investigate the electron-hole dynamics in few-layer InSe, a van der Waals semiconductor with promising opt-electronic properties. We show that terahertz pulses modulate the spontaneous emission via absorption by photo-excited carriers. We further study the carrier-phonon interaction in the cooling process of the hot carriers, elucidating the roles of acoustic and optical phonons.**

I. Introduction

InSe is a van der Waals semiconductor with a bandgap energy of about 1.3 eV. This material demonstrates several promising properties for opto-electronic applications such as low electron mass and high-plasticity [1]. Furthermore, InSe has become an attractive material for terahertz applications after the experimental observations of intersubband transitions in the infrared range of the electromagnetic spectrum [2].

To facilitate a further development of the InSe-based technology, it is important to gain a fundamental understanding of the microscopic electron-hole dynamics. To this end, we investigate the photoluminescence (PL) dynamics in few-layer InSe as a function of layer number and study the impact of a terahertz (THz) pumping on the PL emission [3,4]. The experiments have been carried out at the free-electron laser FELBE that delivers intense picosecond THz pulses with a MHz repetition rate.

II. Results

Figure 1(a) shows the time-resolved PL dynamics of InSe crystals of various thicknesses. The PL lifetime increases significantly by reducing the layer thickness. This is due to the direct-to-indirect bandgap crossover when the thickness decreases from bulk to monolayer [3]. The PL lifetime of InSe crystals thinner than 10 nm is dominated exclusively by the defect-assisted recombination and the lifetime reaches hundreds of nanoseconds. The PL emission of thicker flakes is defined by both defect-related emission and band-to-band recombination.

Moreover, we study the impact of THz radiation on the PL dynamics. To this end, we perform two-color time-resolved photoluminescence (PL) experiments. We use visible pulses (770 nm) to excite the electron-hole population and THz pulses from the free-electron laser (25 µm or 12 THz) to perturb the PL emission. Figure 1(b) shows the temporal evolution of the PL spectrum. The arrows indicate the time when the pulses arrive on the sample. We observe up to 50% of PL quenching when the THz radiation illuminates the thin crystal (16 nm thick) and a subsequent cooling and PL recovery on the time scale of 50 ps.

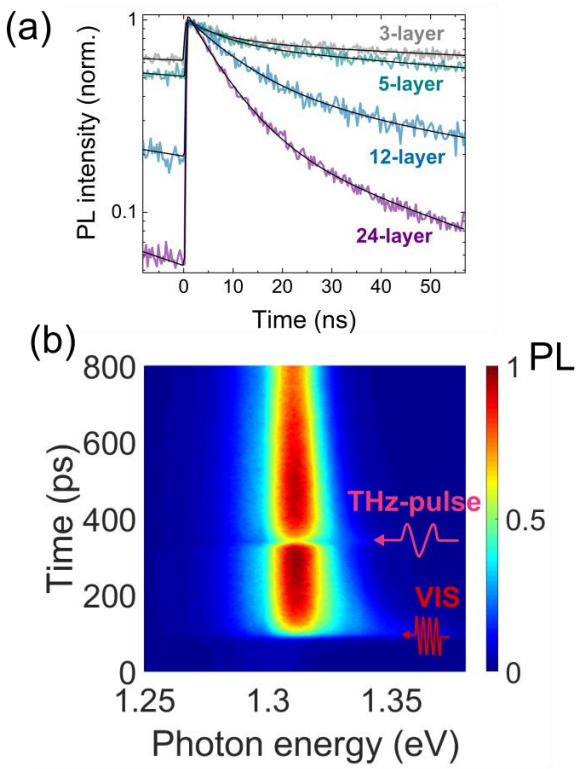

Figure 1 (a) Time-resolved PL as a function of number of layers. (b) Time-resolved PL spectra taken with a high-time resolution streak camera. The additional THz pulse induces a transient quenching of PL emission. A subsequent full recovery of emission is observed. The spectra are measured at 10 K.

To understand the physical mechanism behind the PL quenching, we solve numerically the Boltzmann scattering equation taking into account the scattering of carriers with acoustic and optical phonons. We find that the PL recovery time increases by decreasing the lattice temperature because the optical phonons have too high energy to cool carriers and the cooling at low temperatures is governed solely by the scattering on acoustic phonons, a way less efficient process. Furthermore, we find that the origin of the PL quenching is THz absorption by photo-excited carriers: the broader distribution in the momentum space of the carriers leads to a reduction of the radiative electron-hole recombination rate and, therefore, to a transient quenching of the spontaneous emission.

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