Exploring neutral and charged exciton optical properties in a $MoTe_2/SiO_2/Si$-based metal-oxide-semiconductor device

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Molybdenum ditelluride ($MoTe_2$) belongs to a broad family of transition-metal dichalcogenides (TMDs). In its 2H semiconducting phase, as a single-layer crystal, it has a direct optical bandgap at ~1.17 eV at 5 K, translated to ~1.0 eV at 300 K.[1] It makes this material the only TMD with optoelectronic device applicability in the near-infrared spectral range (>1 μm photon wavelength). However, many properties of single-layer $MoTe_2$ crystals are still unknown, including those related to the control of optical bandgap, interactions and excitation dynamics. It is predicted that the excess carrier density, charge polarity and electric field in $MoTe_2$ should effectively control these properties.[2] In this contribution, we examine the role of excess carrier density in monolayer (ML) $MoTe_2$ and examine to what extent one can control the optical gap, interactions in excitonic complexes and excitation dynamics.

We fabricated a metal-oxide-semiconductor (MOS) device allowing control over the excess carrier density and carrier type, switching from electrons to holes, in ML $MoTe_2$. The device consists of large-area exfoliated ML $MoTe_2$ placed partially on a gold electrode pre-patterned on a SiO_2/doped Si substrate. The Si plays a role of a second electrode. The MOS device can be biased with externally applied voltage (V_g), ranging from –30 V to +30 V and operates at cryogenic temperatures. Sweeping the voltage, we achieved a high range of charge tunability tracked by the optical response from the crystal at T = 5 K using the high-spatially-resolved photoluminescence (μPL) and contrast reflectivity experiments (μCR). By increasing V_g, one can observe the conversion of neutral excitons (X) to trions\polarons (T) through the monotonous increase in the T transition-related μPL intensity at the cost of a decrease in the X transition intensity. At roughly +/–20 V, the X population is almost entirely drained, and the T intensity is saturated. This observation is confirmed in the μCR, where at higher V_g, a new T-related resonance appears on a low-energy side of the always present X resonance. Interestingly, the voltage sweeping allows tuning energies of the X-related optical transition observed in μPL, showing a nearly linear increase from ~1.181 eV at ~0 V to 1.185 eV at –20 V and ~1.184 eV at +20 V. Under the same conditions, the T transition (when visible) remains almost unchanged at ~1.158 eV for negative bias and ~1.159 eV for positive. The trion/polaron binding energy ($E_{b,T}$) depends on the V_g polarity. For the negative Vg, $E_{b,T}$ reaches ~27 meV, whereas, for the positive Vg, it amounts to roughly 25 meV.

Finally, we tested the dynamics of X and T excitations using a non-degenerated pump-probe transient reflectivity experiment. We observe at least two fast and slow recombination channels for each species. At Vg~0 V, near the charge-neutrality condition, the X decays within ~9 ps and ~70 ps, whereas T decay with ~12 ps and ~90 ps. Interestingly, the decay channels can be controlled mainly by negative Vg, shortening the short component by a few ps and the long component by a few tens of ps.

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[2] G. Wang, A. Chernikov, M. M. Glazov, T. F. Heinz, X. Marie, T. Amand, and B. Urbaszek, *Rev. Mod. Phys.* **90**, 021001 (2018)