Thesis Abstract:

Two-dimensional (2D) layered materials, such as graphene and group VI transition metal dichalcogenides (TMDs), have attracted significant attention for their promising applications in photonics and optoelectronics. Recently, interest has shifted toward anisotropic 2D semiconductors due to their polarization-sensitive optical and electronic properties. Among these, the relatively less explored group VII TMD rhenium disulfide (ReS_2) exhibits pronounced in-plane anisotropy arising from its low-symmetry, distorted triclinic (1T) crystal structure. This anisotropy gives rise to a well-defined crystallographic direction aligned with the Re–Re atomic chains (the b-axis), which governs many of its optical and electronic responses. These 2D materials can host tightly bound electron–hole pairs, known as excitons. Due to reduced dielectric screening, two-dimensional confinement, and large carrier effective mass, the exciton binding energy can reach values on the order of 100 meV, making excitonic effects prominent even at room temperature. In ReS_2, two distinct excitonic species have been identified: the X_1 exciton, nearly aligned along the b-axis, and the X_2 exciton, oriented approximately perpendicular to it. Despite these unique properties, the electronic band structure of ReS_2 remains not fully understood due to its low symmetry, giving rise to a long-standing debate regarding whether its bandgap is direct or indirect.

To address this, temperature-dependent photoluminescence (PL) measurements were performed, revealing a non-trivial variation in PL intensity with temperature. Using a rate equation model, the observed trend was successfully reproduced and attributed to the presence of a momentum-forbidden dark exciton state, supporting the conclusion that ReS_2 exhibits a quasi-indirect bandgap. The coexistence of in-plane anisotropy, low crystal symmetry, and a quasi-indirect bandgap suggests that ReS_2 is a promising platform for Raman-based photonic applications.

The role of excitons in Raman scattering in ReS_2 is further investigated, revealing a two-order-of-magnitude enhancement in Raman intensity under excitonic resonance conditions when either the pump or the Stokes photon energy matches an excitonic transition. Upon embedding few-layer ReS_2 in a microcavity, stimulated Raman scattering is observed under a double-resonance condition, where both pump and Stokes photons are resonant with polaritonic energy levels. This strong light-matter interaction gives rise to hybrid quasiparticles known as exciton-polaritons. By exploiting polariton dispersion, Raman scattering is enhanced by nearly five orders of magnitude, leading to thresholdless Raman lasing, a phenomenon absent in the bare system.

Furthermore, the polarization state of specific Raman modes in the microcavity-integrated ReS_2 system can be actively controlled by tuning the pump energy or its polarization. This polarization tunability, observed even for Raman modes with fixed emission energies, is interpreted within the framework of PT-symmetric non-Hermitian physics. These findings demonstrate several nontrivial phenomena, including thresholdless lasing, polarization-tunable Raman emission, and the potential for controlling polarization coherence.

Altogether, this work lays the foundation for polarization-sensitive Raman photonics, with promising applications in quantum optics, integrated photonic circuits, and advanced optical sensing technologies.

Keywords: Anisotropic two-dimensional materials, Rhenium disulfide (ReS_2), Resonance Raman scattering, Stimulated Raman scattering, Raman laser, Tunable Polarization of Raman modes