Influence of excited carriers on opto-electronic properties of transition-metal dichalcogenides

Carrier dynamics and optical properties of transition-metal dichalcogenides

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In Short

- Semiconducting transition metal dichalcogenides are promising candidates for novel nanolaser devices

- Opto-electronic properties under varying excitation and environmental conditions are of significant importance, as they determine response under actual device operation

- We investigate excitation-dependent opto-electronic properties using state-of-the-art many-body methods

Research on atomically thin transition metal dichalcogenide (TMDC) semiconductors has made considerable progress towards the realization of working devices, which involve light-emitting diodes, nanocavity lasers , and single-photon emitters. For conventional semiconductors such as GaAs, substantial experimental and theoretical activities have been devoted to identify and to characterize the intrinsic interaction processes. Only on these grounds, it has become possible to reveal the true device operation potential of these materials. The dominant carrier relaxation processes enabling the conversion of optically or electroni- cally excited carriers into light emission are carrier-carrier and carrier-phonon interaction. While carrier-carrier interaction leads to a redistribution of energy and momentum among the carriers, carrier-phonon interaction additionally provides efficient cooling of the carriers, accompanied by a heating of the lattice itself. A theoretical understanding of the timescales of both interaction mechanisms depending on parameters like excitation density and lattice temperature is crucial for the description of TMDC semiconductors as active material in devices. Carrier-phonon inter- action is also known to provide a channel for the formation of excitons from an optically excited electron-hole plasma. Since excitons and unbound electrons and holes have very distinct physical properties and the expected fraction of bound and unbound carriers after optical exci- tation is a priori not clear, we aim at a microscopic description of exciton and quasi-free carrier dynamics on equal footing. Fascinating prospects arise from the possibility to engineer electronic and optical properties by manipulation of the Coulomb interaction in atomically thin materials via its dielectric envi- ronment. It has also become customary to improve TMD sample qualities by means of encapsulation in hexagonal boron nitride (hBN), which is known to host optical phonons with large oscillator strength in the infrared spectral range. Only recently substrate phonons have been discovered as an additional degree of freedom to tailor the electronic and optical properties of active TMD materials. Besides phonons that are intrinsic to the two-dimensional material, optical phonons in the dielectric substrate can introduce an additional degree of freedom to which TMDC charge carriers can couple. The ever-growing demand for fiber-optical communication evokes a strong interest in semiconductor lasers as active materials with lower power consumption, cost reduction, and higher modulation frequencies. Among the directions to meet these goals is the miniaturization of semiconductor lasers and the search for novel active materials. While todays commercially used semiconductor lasers are based on GaAs quantum wells, a novel and very appealing alternative for the active material consists of atomically thin layers of transition metal dichalcogenide (TMDC) semiconductors. Monolayers of TMDCs can be directly combined with well-developed silicon optical technologies, bringing novel laser devices or other components of integrated photonics with the desired miniaturization within reach. The recently explored strong photoluminescence of various monolayer TMDC materials is the foundation for these developments, thereby initiating strong efforts in the development of TMDC lasers in the past two years. Monolayers of TMDC material have been coupled to various types of nanoresonators: WSe_2 emitting into a GaP photonic crystal cavity, WS_2 on a Si_3N_4 microdisk, MoS_2 on a SiO_2 microdisk and coupled to a SiO_2 microsphere, and $MoTe_2$ on a Si nanobeam cavity. Among the not yet achieved goals is laser operation tuned to the telecommunication wavelength of 1.55 μm for direct coupling into optical fiber networks.

We developed a description for the coupling of TMDC charge carriers to substrate phonons. The geometry of the van der Waals heterostructure (vdW-HS) is schematically shown in the insets of Fig. 1. Our approach uses a mapping of the macroscopic dielectric function of the vdW-HS, which contains optical phonons in the infrared spectral range, to an

effective Frählich Hamiltonian. The Hamiltonian captures the microscopic parameters that characterize both, the heterostructure geometry and the properties of substrate TO phonons as extracted from experiments. Thereby, we transfer the dynamical, frequency-dependent behavior of the substrate dielectric function to a carrier-boson interaction on the level of second quantization. This puts the coupling to substrate phonons on an equal footing with the coupling to intrisic 2d phonons and introduces additional scattering channels for the 2d excitons. We then use 3the augmented coupling Hamiltonian in an equation-of-motion (EOM-) approach to investigate the impact on 2d excitons. Results are shown in Fig. 1. In general, renormalizations of exciton binding energies and quasi-particle band gaps scale as an inverse power law with respect to the substrate TO phonon energy. For monolayer WSe 2 encapsulated in hBN, taking into account the anisotropy of the hBN dielectric response, we find that the coupling to hBN phonons leads to a significant reduction of the 1s-exciton binding energy in WSe 2 . The effect becomes weaker for increasing exciton principal quantum number and is accompanied with additional line broadening.

More Information

[1] Dynamical screening effects of substrate phonons on two-dimensional excitons, A. Steinhoff et al., under review in Physical Review Letters

[2] Charge Carrier Densities in Optically Excited Transition Metal Dichalcogenide Monolay- ers, D. Erben et al., in preparation

Project Partners

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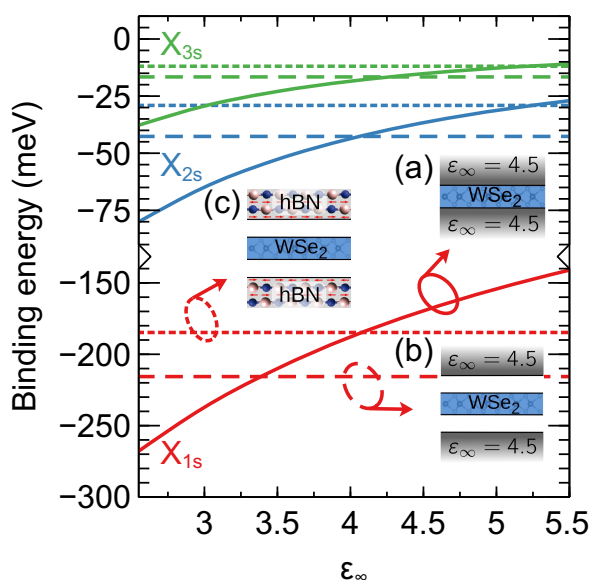
Figure 1: (a) Binding energies of 1s-, 2s- and 3s-excitons as a function of static environmental dielectric constant ε_{∞} in the absence of an inter-layer gap and without dynamical phonon response. This situation corresponds to a Rytova-Keldysh potential. For comparison, binding energies in the presence of an inter-layer gap of 0.2 nm, a fixed static constant $\varepsilon_{\infty} = 4.5$ for hBN (**(b)**, long-dashed lines) as well as additional coupling to hBN phonons (**(c)**, short-dashed lines) are shown.

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