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 and valleytronics devices
- Opto-electronic properties and degree of valley polarization under varying excitation conditions are of significant importance for actual device operation
- We investigate excitation-dependent valley polarization properties using state-of-the-art many-body methods

In this project we investigate carrier dynamics and optical properties in semiconducting two-dimensional transition metal dichalcogenides (TMDCs) with a focus on application in optoelectronic and valleytronic devices. To simulate device operation under realistic conditions, carrier relaxation dynamics driven by the combined action of carrier-carrier and carrier-phonon interaction are essential building blocks. These different interaction mechanisms determine the potential optical gain achievable by TMDC monolayers as well as the valley polarization that can be obtained under different excitation conditions. Since excitons play a crucial role in atomically thin TMDC materials due to strong Coulomb interaction, we will systematically study these effects using state-of-the-art many-body methods that treat free carriers and bound excitons on equal footing.

Nanolasers operate with a minimal amount of active material and low losses. In this regime, single layers of TMDCs are being investigated as next generation gain materials due to their high quantum efficiency. We provide results from microscopic optical gain calculations of highly excited TMD monolayers and specify requirements to achieve lasing with four commonly used TMD semiconductors. Our approach includes band-structure renormalizations due to excited carriers that trigger a direct-to-indirect band-gap transition. As a consequence, we predict a rollover for the gain that limits the excitation regime where laser operation is possible. A parametrization of the peak gain is provided that is used in combination with a rate-equation theory to discuss consequences for experimentally accessible laser characteristics.

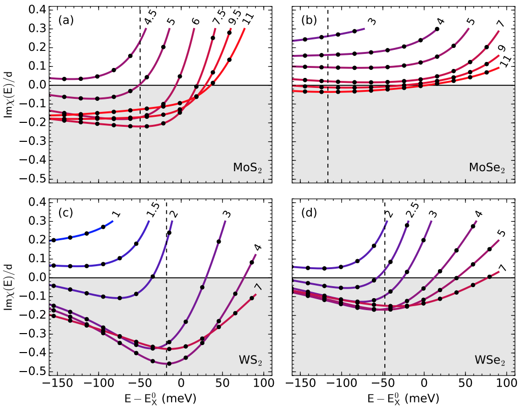

Figure 1: *Imaginary part of dimensionless optical susceptibility for* ***(a)*** *monolayer MoS$_2$,* ***(b)*** *MoSe$_2$,* ***(c)*** *WS$_2$, and* ***(d)*** *WSe$_2$ on a SiO$_2$ substrate at $T = 300$ K and increasing excitation density N given in 10^{13}/cm^2. While a positive value of Imχ implies absorption, optical gain is characterized by a negative imaginary part of the susceptibility (shaded region). The energy axis is chosen relative to the energy E^0_X of the A exciton at zero excitation density. Vertical lines (dotted) indicate the energies at which maximum gain is achieved for each material.*

A key aspect in the application of TMDC semiconductors in laser or valleytronics devices is the presence of excited carriers and their nonequilibrium dynamics, which is governed by carrier-carrier and carrier-phonon scattering processes. To simulate nonequilibrium carrier dynamics and get access to the degree of valley polarization, we solve equations of motion for expectation values such as single-particle occupancies taking into account the different carrier-scattering mechanisms. We also add electron-hole exchange interaction to account for the mixture of excitons in the K- and K'-valleys.

The starting point for the simulation of carrier dynamics is our recently developed description of the carrier-carrier Coulomb scattering in monolayer TMDC semiconductors based on ab-initio determined single-particle band structure and Coulomb interaction matrix elements. These calculations are systematically extended to include also carrier-phonon scattering based on carrier-phonon matrix elements and phonon dispersion relations obtained from ab initio calculations. Since we consider both, resonant excitation of excitons and nonresonant excitation of the band continuum, the theory has to be extended to not only include single-particle quantities describing quasi-free carriers but also exciton populations that are described by two-particle expectation values. The method of choice is the so-called cluster expansion technique that yields a hierarchy of coupled equations of motion (EOM) for arbitrary correlation functions in a solid-state crystal. These correlations are closely connected to the density matrix of the many-particle system of electrons, phonons and photons. The hierarchy can be systematically truncated at a certain level of complexity to get access to the desired quantities such as single-particle and two-particle expectation values. The latter can be shown to correspond to exciton populations, so that the coupled equations enable one to describe unbound carriers and bound two-particle complexes in nonequilibrium on the same footing. We plan to implement the necessary EOM system on top of band structures and matrix elements obtained from first-principle calculations. Since different valleys couple to different polarizations of light, the degree of valley polarization can be concluded by directly calculating the rate of polarized photon emission after optical excitation. As an approximation, we can also calculate the carrier populations in the K- and K'-valleys to estimate the degree of polarization.

WWW

http://www.itp.uni-bremen.de/ag-jahnke/

More Information

[1] Prospects and limitations of transition-metal dichalcogenide laser gain materials, Lohof et al., under review in Nano Letters, preprint: arXiv:1809.08942

Project Partners

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