

Preface

Atomically thin 2D crystals form a distinct and growing class of new materials. The electromagnetic response of a 2D crystal provides direct access to its electronic properties. This thesis presents a series of experimental studies of the electromagnetic response of model 2D crystals as probed by optical spectroscopy. Our aim is to obtain understanding of their intrinsic linear and nonlinear response and the many-body interactions in these materials, as well as to explore the potential to use the 2D materials for sensing applications.

In the two studies of graphene, we either removed contaminations from the environment to reveal the intrinsic response or intentionally applied adsorbates to investigate how the electrons interact with the extrinsic molecules. In the first study, we obtained ultraclean graphene using hexagonal boron nitride as the substrate, which allowed us to probe using Raman spectroscopy the intrinsic electron–phonon and electron–electron interactions free from substrate induced sample inhomogeneity. In a second study, we demonstrate a strong near-field electromagnetic interaction of graphene plasmons with the vibrations of adsorbed molecules. Our results reveal the potential of graphene for molecular sensing.

In our investigations of the monolayer transition metal dichalcogenides, we performed measurements of the linear and the second-order nonlinear dielectric response. From the linear dielectric response, we demonstrate strong light-matter interactions even for a single layer of these materials. Several trends in the excitonic properties of this group of materials were obtained from the measured dielectric function. In the nonlinear optical study, we observed $>10^3$ enhancement of the second-harmonic signal from monolayers as compared to the bulk sample, a consequence of the breaking of the inversion symmetry present in the bulk. In addition to the results for monolayers, we describe the behavior of few-layer materials, where the symmetry properties change layer by layer. For monolayers (and samples of odd layer thickness with broken inversion symmetry), the strong and anisotropic second-harmonic response provides a simple optical probe of crystallographic orientation.

In the magneto-optic study of transition metal dichalcogenide monolayers, we demonstrate the induction of valley splitting and polarization by the application of an external magnetic field. The interaction of the valleys with the magnetic field reflects their nonzero magnetic moments, which are compared to theoretical models. We further clarify the electronic configuration of the charged excitons and important many-body corrections to the trion binding energy through the control of valley polarization achieved by the external magnetic field.

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