Time-resolved nonlinear optical spectroscopy of basic excitations in 2D materials

Daniel Wigger

School of Physics, Trinity College Dublin, Ireland

Since their discovery in 2010, the study of optically active 2D materials in the form of transition metal dichalcogenides, has grown to a distinguished area of research in solid state optics. In these materials tightly bound excitons dominate the optical response and lead to rich multi-particle physics based on the interplay with light, phonons, single charges, and other excitons. While microscopic descriptions, e.g., based on semiconductor Bloch equations, reveal many effects arising from the different coupling mechanisms, we have employed a handy mean-field extension of the optical Bloch equations. We take a local field coupling and excitation-induced dephasing into account, which account for exciton-exciton interactions [1]. The appeal of this approach lies in the fact that it allows to derive analytic expressions for optical signals even for complex nonlinear wave-mixing experiments in the limit of ultrafast pulses [2]. The local field model has proven to reproduce pump-probe [3], four-wave mixing [4, 5], and even six-wave mixing features [4] surprisingly well, despite its mathematical simplicity. In the first part of my talk I will explain the basic consequences of the local field model and present our recent results on four-wave mixing spectroscopy on gate-controlled $MoSe_2$ monolayers, which reveal the coherence properties and couplings of different exciton complexes when changing the electronic density in the system [5].

In the second part I will move to the van der Waals insulator hexagonal boron nitride (hBN), which is recently gaining significant attention because it can host bright colour centres operating as single photon emitters at room temperature. These quantum emitters also stand out due to their remarkably strong coupling to optical and acoustic phonons. In the second part of my presentation I will discuss our recent coherent control study of phonon-assisted excitation of single hBN colour centres [6]. Through this Ramsey–interferometry-like method we were able to determine the coherence times of bare and optical–phonon-assisted excitations and demonstrate the coherence loss induced by the emission of acoustic phonon wave packets.

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