Local field impact on nonlinear spectroscopy signals from TMDCs

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Monolayers of transition metal dichalcogenide (TMDC) have emerged as an outstanding subject in semiconductor photonics. They show a strong optical response in the visible spectral range that is governed by tightly bound excitonic complexes. Moreover the optical selection rules together with the spin ordering of valence and conduction bands offers the possibility of valley-selective excitations. In this context ultrafast nonlinear spectroscopy techniques are perfectly suited to study the exciton dynamics in TMDC monolayers.

In our model we consider the different excitonic states as few-level systems and the exciton-exciton interaction is incorporated via a local field (LF) and via excitation induced dephasing (EID). While the former affects the exciton's transition energies, the latter changes the decoherence. The strength of both effects is proportional to the exciton occupation. Within this minimal model, one can derive analytical solutions for the nonlinear signal dynamics after ultrafast optical excitations. We will discuss two different spectroscopy signals:

(i) Combining theory and experiment we study pump-probe signal dynamics from a $MoSe_2$ monolayer encapsulated in hBN [1]. When scanning the delay between pump and probe pulse we find that the detected spectrum experiences an energy shift for short delays. The exciton resonance shifts back to the original position within picoseconds. This behavior is thoroughly reproduced by our instructive model. Where the probe pulse arrives before the pump pulse, characteristic spectral oscillations appear. When considering a cross-circular polarization scenario we have additional access on the inter-valley scattering.

(ii) We theoretically study the dynamics of four-wave mixing signals. Depending on the considered series of excitation pulses one can study the dynamics of the exciton coherence or the occupation. As depicted in Fig.1 we find that the FWM spectrum exhibits a characteristic broadening when increasing the applied laser pulse intensities (pulse area). We can trace back this behavior to the local field effect and instructively explain it within a Bloch vector picture.

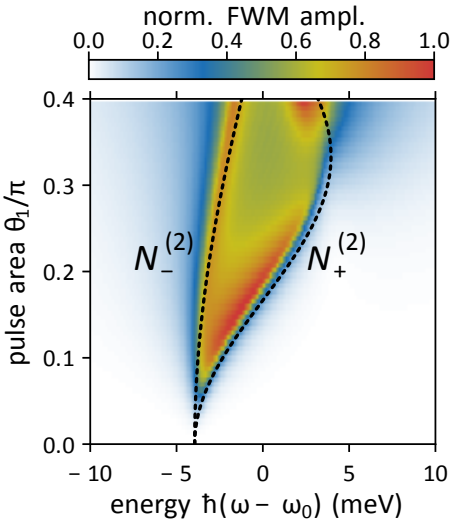

Fig.1: FWM spectra as function of the applied pulse area.

[1] A. Rodek, T. Hahn, et al., arXiv:2103.05328, accepted for publication in Nanophotonics (2021).
[2] T. Hahn, J. Kasprzak, et al., New J. Phys. 23 023036 (2021).