Wavelength dependence of High Order Harmonic Generation from a single atomic layer TMDC

Viacheslav Korolev[1], Vipin Krishna[2], Alexander Croy[3], Thomas Lettau[4], Giancarlo Soavi[2], Daniil Kartashov[1]

[1] Institute of Optics and Quantum Electronics, Friedrich-Schiller University Jena, 07743 Jena, Germany

[2] Institute of Solid State Physics, Friedrich Schiller University Jena, 07743 Jena, Germany

[3] Institute of Physical Chemistry, Friedrich-Schiller-Universität Jena, 07743, Jena Germany

[4] Institute of Condensed Matter Theory and Solid State Optics, Friedrich Schiller University Jena, 07743, Jena Germany

viacheslav.korolev@uni-jena.de

The Experimental and most of the theoretical investigations on high-order harmonic generation (HHG) in solids up to now were focused on the dependence of the emission spectrum upon the laser polarisation (orientation of the linear polarisation with respect to symmetry directions or ellipticity) and the laser intensity [1]. The dependence of HHG on such important parameter as the laser wavelength remains unexplored. In this contribution, we present a rigorous experimental and theoretical study of the wavelength dependence of HHG in solids. To disentangle the complex contribution of linear and nonlinear propagation effects, the experiments are conducted with different single atomic layer TMDC crystals. We report on two major observables. First, we demonstrate an exponential decrease in harmonic yield with increasing the wavelength. This observation sheds light on the problem of the dephasing time – an empirical and disputable concept introduced originally to match numerically simulated HHG spectra to the experimentally measured ones. As a reference, we show that a similar exponential drop in efficiency is observed in a bulk semiconductor thin film. Our measurements and numerical simulations, based on real-time time-dependent density functional theory and semiconductor Bloch equations, strongly support the concept of dephasing. Second, we investigated the effect of selective enhancement of even order harmonics in single-layer 2D materials. By tuning the laser wavelength at a fixed laser intensity, we show that the enhancement occurs only for even-order harmonics that fall in a specific, relatively narrow spectral range, while odd-order harmonics possess a standard plateau-cutoff behaviour. This enhancement is orientational and material dependent, suggesting the role of multi-band effects.

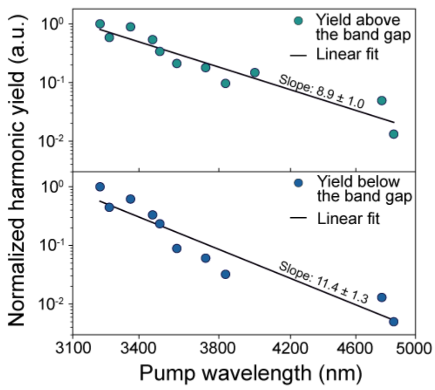

Figure 1: Harmonic yield as a function of pump wavelength for a monolayer WS_2

References

[1] Goulielmakis, E., & Brabec, T. High harmonic generation in condensed matter. *Nature Photonics*, (2022). *16*(6), 411-421.