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Influence of Coulomb correlations on high harmonic generation in monolayer TMDC materials

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The nonlinear optical phenomenon of high harmonic generation (HHG) in solid state materials was employed to study different properties of these materials. In a wide range of semiconducting materials, HHG can be induced by off-resonant excitation with ultrashort strong-files pulses in the THz- and mid-IR-range. The study of HHG in TMDC materials has been shown to be particulary interesting due to their strong non optical non-linearities and the complex interplay of their directional-, spin- and valley-selective emission characteristics (see e.g. [1,2]).

Furthermore, the material thinness of TMDC monolayers causes strong Coulomb interactions that goes hand in hand with a high sensitivity to the dielectric surrounding and the excitation conditions. The presence of excited carriers induces a screening of the Coulomb interaction resulting in a renormalization of the band structure. For pulses that are shorter than the de-coherence time, these carrier distributions vanish with the pulse induced polarizations. Thus, one should expect ultrafast signatures of Coulomb screening and the related field and energy renormalizations in the HHG signal.

Here, we employ a microscopic approach combining DFT with a Semiconductor-Bloch equation approach to investigate the influence of Coulomb correlations on HHG in TMDC monolayers. Based on the DFT single-particle band structure and the phase-corrected dipole matrix elements, we derive an effective two-dimensional model that allows to include all relevant Coulomb effects.

[1] H. Liu, et al., Nature Phys 13, 262–265 (2017).
[2] Y. Kobayashi, et al., Ultrafast Science, 2021, 9820716 (2021).

Category

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