High-harmonic spectroscopy of strongly-bound excitons in solids

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Excitons play a vital part in many optical processes of condensed matter systems such as light absorption or photo-generation of currents. Excitons are thereby consequential to material science and photonic technological applications, especially for quasi-two-dimensional materials. An understanding of ultrafast exciton dynamics is needed to outline their influence on monolayer photonics and petahertz electronics.

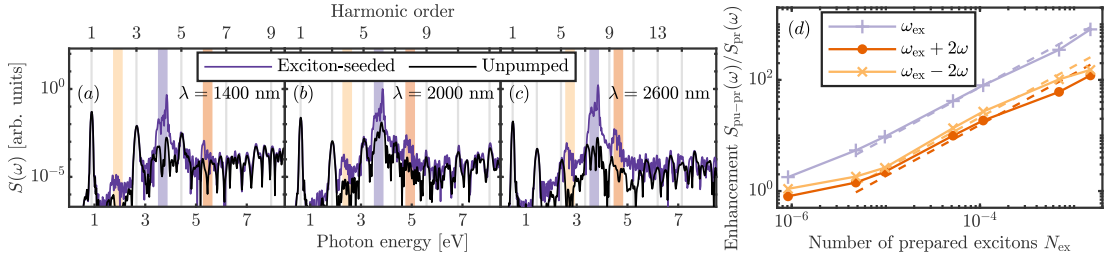

Figure 1: (a)-(c) HHG spectra for the exciton-seeded system and unpumped system at various driving wavelengths for a 100 fs pulse of 10^7 W cm^{-2} peak intensity. The purple, orange and red areas denote the exciton resonance ω_{ex}, and associated sidebands $\omega_{\text{ex}} \pm 2\omega$. (d) Enhancement of the exciton resonance and sidebands for (b) when varying the exciton population. Dashed lines indicate common scaling relations of $N_{ex}^{0.92}$

We explore the nonlinear ultrafast exciton response when driven by a strong electromagnetic field. We utilize time-dependent Hartree-Fock with a one-dimensional hydrogen chain model. Within this model, we identify means of pumping a controlled population of excitons. Following this, we identify signatures of such populations, using high-harmonic spectroscopies. We identify exciton resonances and sidebands in the spectrum shown in Fig. 1. These scale with the exciton population. The exciton wavefunction is visualized and we identify signatures from its evolution in the subcycle emission pattern including excitonic dissociation, recombination and Stark shifts. Our work paves the way towards time-resolved spectroscopy of ultrafast exciton dynamics of solids.

References

[1] S. V. B. Jensen, L. B. Madsen, A. Rubio, N. Tancogne-Dejean, High-harmonic spectroscopy of strongly-bound excitons in solids, (manuscript in preparation).