Parametric nonlinear optics with two-dimensional materials

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Two-dimensional (2D) materials such as graphene and transition metal dichalcogenides (TMDs) show extraordinarily strong second- and third-order nonlinear optical responses which, thanks to their unique electronic properties and despite their atomic thickness, allow a wide range of fundamental studies and technological applications. In this presentation we will discuss some of our recent results on the study of the nonlinear optical properties of 2D materials.

We show that the third-harmonic generation efficiency in graphene can be increased by almost two orders of magnitude by controlling the Fermi energy and the incident photon energy [1]. This enhancement is due to logarithmic resonances in the imaginary part of the nonlinear conductivity arising from resonant multiphoton transitions. Thanks to the linear dispersion of the massless Dirac fermions, gate controllable third-harmonic enhancement can be achieved over an ultrabroad bandwidth, paving the way for electrically tunable broadband frequency converters.

Using semiconducting TMDs, we demonstrate single-pass optical parametric amplification at the ultimate thickness limit, down to a single atomic layer [2]. Second-order nonlinear interaction at the 2D limit bypasses phase-matching requirements and achieves ultrabroad amplification bandwidths. In agreement with first-principles calculations, we observe that the amplification process is independent of the in-plane polarization of signal and pump fields.

We investigate 3R-stacked TMD crystals which combine broken inversion symmetry and aligned layering. By measuring second harmonic generation (SHG) of 3R-MoS_2 with various thickness, from monolayer (0.65 nm) to bulk (≈1 μm), we obtain the first measurement of the SHG coherence length (≈ 530 nm) at 1520nm and achieve record nonlinear optical enhancement from a van der Waals material, > 10^4 stronger than a monolayer. We find that 3R-MoS_2 exhibits similar conversion efficiency as lithium niobate, but with more than 100-fold shorter propagation lengths [3].

Finally, we demonstrate a novel approach for the all-optical control of SHG polarization in MoS_2 and show that this can be used for all-optical modulation of the SHG efficiency, with modulation depth close to 100% and speed limited only by the fundamental frequency pulse duration [4].

References

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