

Design and development of a third-harmonic generation setup for the characterization of the nonlinear optical properties of 2D materials

Graphene - a single atomic layer of carbon atoms - is a very promising material, mainly due to its extremely high and broadband nonlinear optical susceptibility [1] and the possibility of occurrence of interband transitions at all optical frequencies. Ultrafast third-harmonic generation (THG) of ultrashort laser pulses in graphene allows not only the temporal characterization of the ultrashort pulses themselves but also the study of carrier dynamics in graphene. The possibility of obtaining an enhanced nonlinear signal when using multi-layer graphene [2] further adds to these capabilities. The technique of dispersion scan (d-scan) [3] developed in our group in collaboration with Lund University enables characterizing ultrashort light pulses using an unprecedentedly simple and fully inline optical setup, based on recording the optical spectrum of a nonlinear signal produced by the pulse for different amounts of dispersion applied to the same pulse. This results in a 2D d-scan trace from which the spectral phase of the pulse can be retrieved using a numerical algorithm and, therefore, by inverse Fourier transform, provides the exact temporal intensity profile and phase of the pulse. The most common nonlinear signal for d-scan has been second-harmonic generation (SHG) produced in non-centrosymmetric nonlinear crystals. In the present work we use THG as the nonlinear signal. Unlike SHG, THG can be produced in any medium, regardless of its symmetry. Also, for very broadband octave-spanning lasers or mid-infrared systems, it is helpful to use higher-order nonlinear effects, like THG [4]. Here we present several examples of THG d-scan measurements of broadband few-cycle laser pulses obtained in graphene coatings produced by different production techniques [5], which enable characterizing the used ultrashort pulses while providing insight on the electronic dynamics in graphene. The possibility of improving the nonlinear optical response of graphene and other 2D materials by chemical or morphological functionalization, will also be presented.

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References

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