Broadband deep-ultraviolet third-harmonic generation in multilayer graphene and its application to few-cycle pulse measurement by THG d-scan

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Abstract: We report on broadband deep-ultraviolet (DUV) third-harmonic generation (THG) in multilayer CVD-grown graphene pumped by few-cycle pulses from a Titanium:Sapphire laser oscillator. An enhanced nonlinear optical response compared to near-surface THG in the bare (uncoated) substrate is clearly observed. We apply this process to the complete characterization (amplitude and phase) of the low-energy few-cycle (7.1 fs FWHM) pump pulses using the new technique of THG dispersion-scan.

I. Introduction

Graphene is an extremely promising material owing to its unusual properties. Amongst them, it possesses an exceptionally high nonlinear optical susceptibility [1], which has motivated further investigation of its performance in various nonlinear optical processes. Achieving ultra-broadband third-harmonic generation (THG) at low intensities is currently a demanding task and the strong, near-dispersionless nonlinear response of graphene should be very suitable for this purpose. The possibility of using a multilayer graphene film for enhanced THG [2] adds to its interest as a practical material. Furthermore, the promise of graphene retaining its extremely broadband nonlinearity due to lack of macroscopic phase-matching (each layer is sub-nm thick) makes it highly attractive for ultrafast applications. THG in few-layer graphene pumped by 320 fs pulses at 1720 nm resulted in visible pulses at 573 nm, where a quadratic dependence of the THG signal on the number of layers was observed [3]. THG in monolayer graphene pumped with 50 fs pulses at 789 nm enabled probing structural properties of graphene [4]. In a previous communication, we reported on broadband THG in graphene pumped by 7 fs few-cycle laser pulses [5]. In this work we demonstrate THG enhancement from multilayer graphene compared with near-surface THG under loose focusing conditions and over a large bandwidth in the deep-ultraviolet (DUV). Furthermore, we use this signal to make a pulse measurement of a femtosecond oscillator using a THG dispersion-scan (d-scan) setup and further compare these results to a standard SHG d-scan measurement [6]. Pulse characterization through d-scan is based on the fact that when a pulse undergoes a nonlinear conversion process (e.g. SHG), the resulting spectral intensity has a well-defined dependence on the input spectral phase. This way, one only needs to measure the resulting spectrum for different input spectral phases to be able to retrieve the initial spectral phase. THG d-scan is desirable as an alternative to SHG d-scan due to the possibility of gating spectra over an octave in bandwidth without overlapping the nonlinear signal and the fundamental, as well as for its potential for measuring longer wavelength few-cycle mid-IR lasers.

II. Experimental setup and results

For this study, an ultra-broadband Ti:Sapphire laser oscillator (Femtolasers Rainbow CEP) was used. As shown in Fig. 1, the beam was sent through a pair of BK7 glass wedges (8º angle) for variable dispersion adjustment and fine-tuning and then underwent 4 bounces off ultra-broadband double-chirped mirrors (IdestaQE) introducing negative dispersion. The beam was then focused on the sample using a spherical silver mirror with 5 cm focal length. The focal spot was approx. 5 μm ($1/e^2$ width) with a peak intensity of approximately 100 GW/cm^2. The sample is a nickel-grown multilayer graphene (MLG) film transferred to an optical quality fused silica substrate. The film is non-uniform and composed of multilayer domains of graphene having between 1 to 7 layers. To detect the generated third-harmonic we used a prism pair to spatially block the fundamental radiation prior to coupling the THG signal into a fiber-coupled spectrometer (Ocean Optics HR4000). Using this setup we first studied the efficiency of the THG signal. Due to the inhomogeneity of the sample, the signal from MLG varies greatly in intensity across its surface, depending on the number of layers in the location being irradiated. When optimizing for the strongest signal, it is possible to obtain approximately 20 times higher intensity than for locations in the substrate not covered with multilayer graphene, whereas no such enhancement has been observed for a copper grown single-layer graphene sample. This 20-fold enhancement in the THG signal is significant and eases the requirements for some applications that would otherwise need extremely tight focusing. The right plot in Fig. 1 shows the THG spectrum as a function of incident average power, revealing a cubic dependence on the input pulse intensity, as expected. One useful, widespread application of broadband THG generation is femtosecond pulse characterization. Owing to its simplicity, the required parts to make a dispersion-scan measurement were already in place, so we used this exact setup to measure the incident pulses by THG d-scan. Additionally, by simply replacing

the MLG sample with a SHG crystal (5-μm-thick BBO) we were also able to measure the corresponding SHG d-scan traces [6] in order to compare the two methods (Fig. 2). By recording the THG spectrum for varying BK7 wedge insertions we obtained a 2D THG d-scan trace (Fig. 2a) that, like its SHG counterpart (Fig. 2c), contains enough information to fully retrieve the pulse phase using the algorithm described in detail in [6], modified for the THG nonlinearity. An excellent agreement between pulses retrieved using the two methods is observed, as shown in Figs. 2e and 2f.

III. Conclusions

We obtained efficient THG of few-cycle pulses over a broad bandwidth in the DUV (230-305 nm) from nickel-grown graphene multilayer samples transferred to an optical substrate. We found that this signal is readily observable for nJ-level pulses even in loose focusing conditions (i.e. not requiring short and unpractical focal lengths) and subsequently applied this process to few-cycle pulse characterization with zero changes introduced in the previous setup. We demonstrated pulse measurement and retrieval through THG d-scan and compared it to SHG d-scan, achieving excellent agreement. THG d-scan is a promising new technique for characterizing single-cycle pulses and few-cycle mid-IR laser sources.

References

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Figures

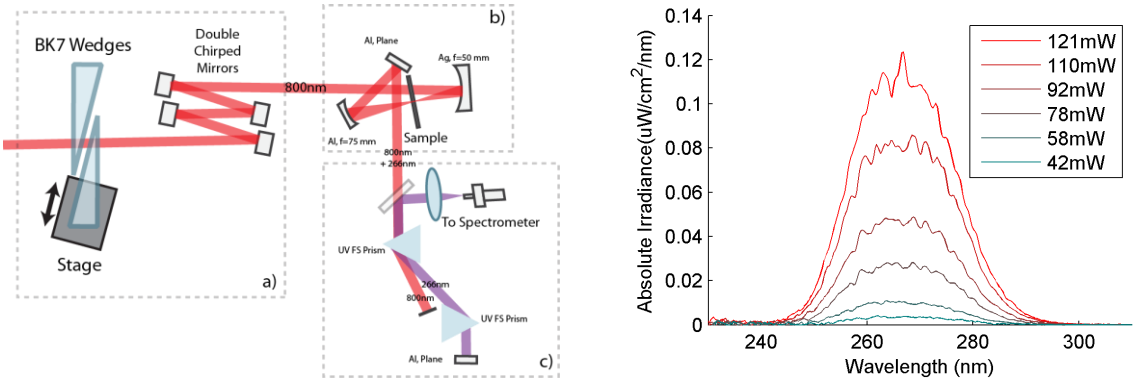

Fig. 1. (Left) THG d-scan setup used in this work, composed of 3 main blocks: a) variable dispersion compensation, b) nonlinear stage (THG/SHG), c) wavelength separation and spectral measurement. (Right) Measured THG spectrum in multilayer graphene as a function of incident average power.

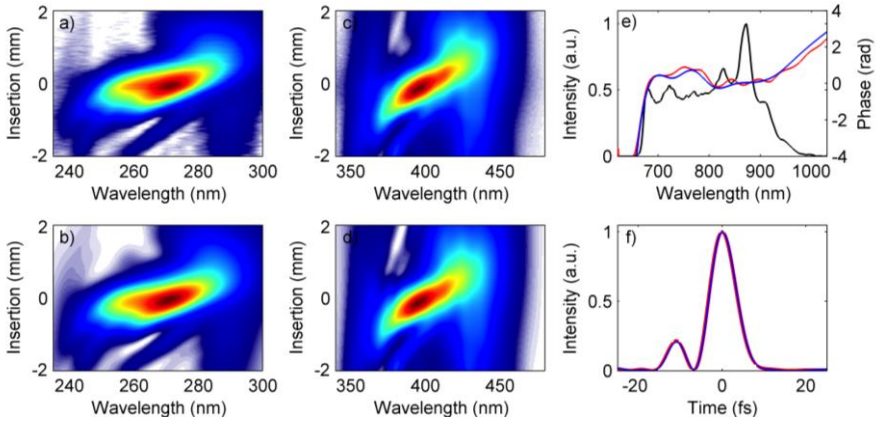

Fig. 2. Experimental results: a) Measured THG d-scan in graphene. b) Retrieved THG d-scan. c) Measured SHG d-scan. d) Retrieved SHG d-scan. e) Black: spectral intensity. Red: Retrieved phase for shortest pulse, SHG-d-scan. Blue: Retrieved phase for shortest pulse, THG-d-scan. f) Temporal intensity corresponding to the spectrum and phases in Fig. 2e. Blue line: THG d-scan (7.1 fs FWHM). Red line: SHG d-scan (7.1 fs FWHM).