Metasurface-Enhanced Terahertz Third Harmonic Generation in Double-Layer Graphene

A. Maleki[1], M. Heindl[2], Y. Xin [3], Robert W. Boyd[1,4,5], Georg Herink[2], and Jean-Michel Ménard[1,4]

[1]*Department of Physics, University of Ottawa, Ottawa, Ontario, Canada*

[2]*Experimental Physics VIII – Ultrafast Dynamics, University of Bayreuth, Bayreuth, Germany*

[3]*Iridian Spectral Technologies Ltd, Ottawa, Ontario, Canada*

[4]*School of Electrical Engineering and Computer Science, University of Ottawa, Ontario, Canada*

[5]*Institute of Optics and Department of Physics and Astronomy, University of Rochester, Rochester, NY, USA*

jean-michel.menard@uottawa.ca

The recent demonstration of high harmonic generation (HHG) in graphene with a super-radiant terahertz (THz) source [1] showed the potential of 2D Dirac materials for nonlinear applications in the far-infrared region. These experiments, which also offer a new testing ground for theories on free-electron-driven optical nonlinearities with low-energy photons, are essential to improve our understanding of the extremely high nonlinear optical coefficient reported for graphene. However, a quantitative assessment of THz high harmonics is limited by a low frequency conversion efficiency in a region of the electromagnetic spectrum lacking sensitive detection tools to accurately monitor weak spectral components. This problem arises in part because of the nature of 2D materials, featuring the smallest possible nonlinear interaction length of one atomic layer. The limited access to high-field THz pulses, especially when relying on a table-top source, is another challenge. Here, we combine two techniques to significantly enhance conversion efficiency in graphene and demonstrate third harmonic generation (THG) at intensities that are orders of magnitude above the background nosie. First, we use a stack of decoupled CVD graphene sheets to increase the interaction length. Then, we deposit this sample on a metallic metasurface to locally enhance the field driving the nonlinear effects.

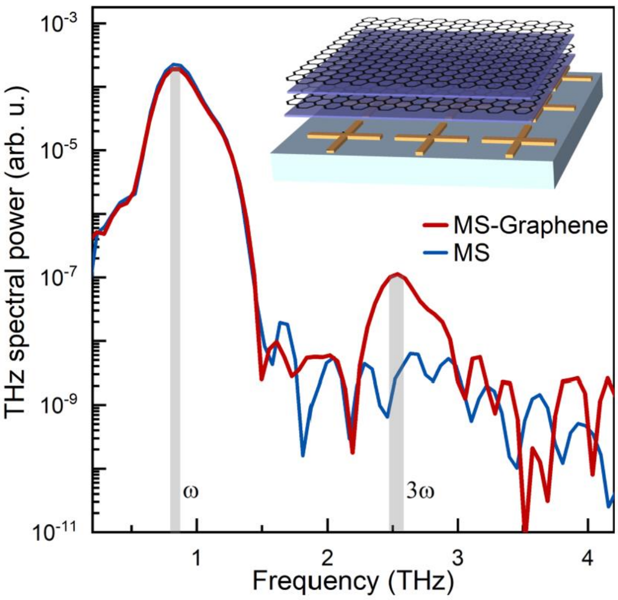

Fig.1. Measured spectrum of the THG at 2.5 THz generated inside two graphene sheets deposited on a metasurface (MS)-based bandstop filter (red line) and compared to the spectrum acquired with the MS alone (blue line). A highpass filter is used to reduce the driving field. Inset shows the sample schematic.

A THz source, based on a tilted pulse-front generation geometry in $LiNbO_3$, delivers high-field pulses that are filtered by a bandpass filter [2] at 0.8 THz to achieve multi-cycle pulses with a 32 kV/cm amplitude. This driving field impinges on two graphene layers that are decoupled by a separating 60-nm-thick aluminum oxide layer. When this sample is deposited on a metasurface-based bandstop filter a distinctive THG spectral peak at 2.5 THz is observed with a peak amplitude of 24 V/cm (see Fig. 1). Note that a high-pass filter [2] located after the graphene-based sample is used to decrease the amplitude of the driving field, which in turn reduces the overall noise in the experiment. In comparison to the THG signal generated inside a single graphene sheet (not shown in Fig. 1), the combined effect of a larger nonlinear interaction length with an enhancement of the local field increases the THG intensity by a factor of 2.5. The resulting nonlinear signal is 20 times larger than the background noise recorded with the metasurface alone, allowing precise monitoring of the nonlinear conversion efficiency.

In conclusion, we demonstrate a large increase of THG in two decoupled graphene sheets deposited on a metasurface. These results highlight the potential of a multi-layered and metasurface-based device architecture to enhance nonlinear responses in 2D materials, paving the way toward practical THz nonlinear applications.

[1] H. A. Hafez, S. Kovalev, J. C. Deinert, Z. Mics, B. Green, N. Awari, M. Chen, S. Germanskiy, U. Lehnert, J. Teichert, Z. Wang, K. J. Tielrooij, Z. Liu, Z. Chen, A. Narita, K. Müllen, M. Bonn, M. Gensch, and D. Turchinovich, Nature **561**, 507 (2018).

[2] A. Maleki, A. Singh, A. Jaber, W. Cui, Y. Xin, B. T. Sullivan, R.W. Boyd, J.-M. Ménard, Photonics Res. **11**, 526 (2023).