

Infrared Pump-Probe Imaging and Spectroscopy with 10nm Resolution

Sergiu Amarie¹, Martin Wagner², Zhe Fei², Alexander S. McLeod², Aleksandr S. Rodin^{2,3}, Wenzhong Bao^{4,5}, Eric G. Iwinski², Zeng Zhao⁵, Michael Goldflam², Mengkun Liu², Gerardo Dominguez^{6,7}, Mark Thiemens⁷, Michael M. Fogler², Antonio H. Castro Neto^{3,8}, Chun Ning Lau⁵, Fritz Keilmann⁹ and Dimitri N. Basov²

¹Neaspec GmbH, Bunsenstrasse 5, 82152 Martinsried, Munich, Germany, ²Department of Physics, University of California, San Diego, La Jolla, California 92093, US, ³Department of Physics, Boston University, 590 Commonwealth Avenue, Boston, Massachusetts 02215, US, ⁴Materials Research Science and Engineering Center, University of Maryland, College Park, Maryland 20742, US, ⁵Department of Physics and Astronomy, University of California, Riverside, California 92521, US, ⁶Department of Physics, California State University, San Marcos, San Marcos, California 92096, US, ⁷Department of Chemistry and Biochemistry, University of California, San Diego, La Jolla, California 92093, US, ⁸Graphene Research Centre and Department of Physics, National University of Singapore, 117542, Singapore, ⁹Ludwig-Maximilians-University and Center for Nanoscience, 80539 Munich, Germany, sergiu.amarie@neaspec.com

Abstract: We introduce pump-probe sub-diffraction infrared imaging and spectroscopy with 100fs temporal and 10nm spatial resolution. On graphene single-layers, we demonstrate time-resolved access to local optical conductivity at technologically relevant mid-infrared frequencies.

OCIS codes: (180.4243) Near-field microscopy; (300.6300) Spectroscopy, Fourier transforms.

1. Introduction to nano-FTIR, Infrared Imaging and Spectroscopy with 10nm Resolution

The performance of the next-generation electronic devices based on Graphene is strongly influenced by the structure-function relationship. A novel technique which combines the best of two worlds, the nano-scale spatial resolution of Atomic Force Microscopy and the analytical power of infrared spectroscopy (nano-FTIR), makes now possible the nanoscale mapping of such nano-devices. Infrared imaging and spectroscopy, which offers direct information regarding molecular and crystal structure, free-carrier concentration and conductivity is now accessible with 10nm spatial resolution [1-3].

2. Ultrafast plasmonics in Graphene

Well-established fiber-based near-field methods are unfortunately limited to visible and near-IR frequencies. In contrast, nano-FTIR described here is capable of probing a broad spectral region from visible to far-infrared frequencies [3]. In our experiment we combine for the first time mid-infrared nano-FTIR and ultrafast 100fs laser excitation to study time-dependent phenomena at the nanoscale (Fig. 1). The pump-probe spectroscopy results revealed ultrafast optical modulation of the infrared plasmonic response of graphene opening thus the gate to ultrafast graphene-based plasmonic devices. In terms of its efficiency, optical control of plasmons in graphene challenges conventional electrostatic gating (Fig. 1), but occurs on a much faster, sub-picosecond time scale as demonstrated in our experiment. Remarkably, modifying graphene's plasmonic behavior requires 2 orders of magnitude less pulse energy than for metal-based structures. For the same pump fluence, the intensity modulation depth of the plasmonic response using ultrafast nano-FTIR on Graphene exceeds 15 times the metal based plasmonics.

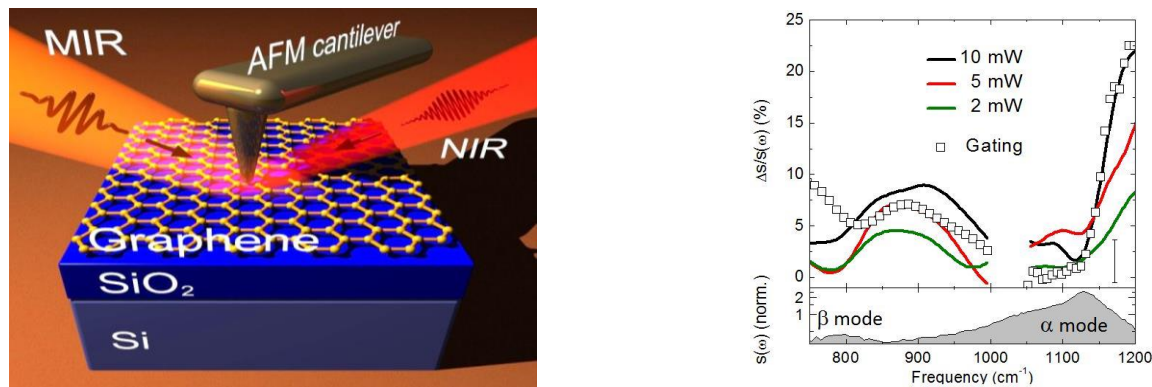


Fig 1. Sketch of the ultrafast nano-FTIR experiment of exfoliated graphene (left), and absolute signal S and pump-induced spectral changes $\Delta S/S$ for 2, 5 and 10mW near-IR pump-power at zero time-delay (right) compared to electrostatic gating.

This first infrared pump–probe experiment beyond the diffraction limit demonstrates the capability of this technique and paves the way to the exploration of a wide range of problems in condensed matter physics, biology, and chemistry.

3. References

- [1] Z. Fei, “Gate-tuning of graphene plasmons revealed by infrared nano-imaging,” *Nature* 487, 82-85 (2012).
- [2] J. Chen, “Optical nano-imaging of gate-tunable graphene plasmons” *Nature* 487, 77-81 (2012).
- [3] I. Amenabar, “Structural analysis and mapping of individual protein complexes by infrared nanospectroscopy” *Nature Communications* 4, 2890- (2013).
- [4] M. Wagner "Ultrafast and Nanoscale Plasmonic Phenomena in Exfoliated Graphene Revealed by Infrared Pump–Probe Nanoscopy" *Nano Letters* [dx.doi.org/10.1021/nl4042577](https://doi.org/10.1021/nl4042577) (2014).