Tsuneyuki Ozaki

INRS-EMT
Varennes, Québec, Canada

ozaki@emt.inrs.ca

Intense Terahertz Field-induced Carrier Dynamics in Graphene

The exceptional quantum properties of graphene, exemplified by its relativistic massless Dirac-fermion physics [1], have great potential for innovative applications in high-speed optoelectronics [2] and photodetection [3]. This has sparked great interest in exploring the ultrafast carrier transport properties of graphene. Noninvasive spectroscopic techniques have been employed to study such properties over a broad range of the electromagnetic spectrum, ranging from the ultraviolet to the far-infrared frequencies [4]. In such experiments, selecting the correct wavelength is crucial for exploring inter- and intra-band carrier dynamics in graphene. Due to the Pauli-blocking effect [5], interband transitions occur linearly only when the exciting photon energy exceeds twice the Fermi-level energy of the graphene sample. Therefore, the optical and infrared frequencies induce interband transitions in graphene [6], while the terahertz (THz) frequencies usually induce intraband dynamics [7], especially in highly doped graphene. Therefore, THz spectroscopy is considered a powerful tool for probing intraband dynamics.

While optical-pump/THz-probe (OPTP) spectroscopy is frequently used for studying the intraband nonequilibrium carrier dynamics in photoexcited graphene [8], recent progress in developing intense THz sources has allowed one to study the *nonlinear* properties of various materials in the THz frequency range [9]. For graphene, it has been theoretically predicted that the relativistic massless Dirac-fermion behavior of its charge carriers and the linear dispersion of its energy spectrum lead to strong *nonlinear* responses to relatively high THz field excitations [10]. Nonlinear processes, such as frequency multiplication [11], intense THz-field induced transparency [12], and impact ionization [13] in graphene, have been recently demonstrated.

In this paper, I will first review the basics of THz technology, and the current status of intense THz sources. Then, I will describe experiments designed to study *nonlinear* THz–induced intraband carrier dynamics in photoexcited graphene, using optical-pump/intense-THz-probe spectroscopy. Through differential transmission measurements, we first observe an enhancement in the THz transmission due to the suppression of the photoconductivity of the graphene after photoexcitation at low THz field strength. This pump-induced transmission enhancement increases when the optical pump fluence of photoexcitation is increased. Remarkably, we observe a reduction in this enhancement as the THz probe field amplitude is increased. We find that the observed nonlinearity does not appear to arise from the intrinsic nonlinearity of graphene that comes from the linear dispersion, but instead is due to the dependence of the carrier scattering rate on the effective lattice temperature, which is dependent on both the optical pump fluence and the THz electric field strength. I will also describe more recent experiments on the *nonlinear* THz spectroscopy of graphene.

References

[1] K. S. Novoselov et al., Science 306, 666 (2004).
[2] T. Mueller et al., Nat. Photo. 4, 297 (2010).
[3] M. W. Graham et al., Nat. Phys. 9, 103 (2013).
[4] K. F. Mak et al., Phys. Rev. Lett. 101, 196405 (2008).
[5] F. Wang et al., Science 320, 206 (2008).
[6] R. R. Nair et al., Science 320, 1308 (2008).
[7] J. M. Dawlaty et al., Appl. Phys. Lett. 93, 131905 (2008).
[8] H. Choi et al., Appl. Phys. Lett. 94, 172102 (2009).
[9] L. Razzari et al., Phys. Rev. B 79, 193204 (2009).
[10] S. A. Mikhailov and K. Ziegler, J. Phys: Condens. Matter 20, 384204 (2008).
[11] P. Bowlan et al., Phys. Rev. B 89, 041408 (R) (2014).
[12] M. J. Paul et al., New J. Phys. 15, 085019 (2013).
[13] S. Tani et al., Phys. Rev. Lett. 109, 166603 (2012).