THz saturable absorption in graphene

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Abstract

Since the first graphene monolayer device reported by Novolosev et al. in 2004 [1], the number of applications of graphene enormously increased, both in photonics and electronics [2]. This extraordinary rise of graphene-based devices can be attributed to its unique electronic and optical properties that make graphene a remarkable material.

Thanks to its characteristic linear Dirac-like band structure, graphene possesses a huge (2.3% per single atomic layer), flat, wavelength independent linear optical absorption in a broad range of frequencies [3] and an extremely high third-order nonlinear response [4-6]. The combination of such properties with the fast carrier dynamics [7] enables the use of graphene as ultrafast broadband saturable absorber. In fact, once a strong incident light exceeds an intensity threshold, the photogenerated carriers block further absorption (Pauli blocking principle) causing an increase of the material transparency. The bleaching of the absorption with relatively low intensity threshold and its broadband spectral range makes graphene competitive against the commonly used semiconductor saturable absorber mirrors [8] in ultrafast pulsed laser fabrication. Graphene-based mode-locked lasers have been successfully demonstrated at 800 nm [9], in the near infrared [10-14] and at 2500 nm [15].

Interestingly, theoretical studies have predicted further stronger nonlinearity of graphene in the microwave [16] and THz regions [17]. This opens the possibility of novel THz lasers (e.g. graphene-mode-locked THz quantum cascade laser), as well as of detection and amplification in graphene devices [18]. Currently, graphene saturable absorption has been studied in the microwave range [19], but no experimental data have been reported in THz regime yet.

In this work we show experimental results on the saturable absorption properties of graphene in the THz regime. The samples are graphene layers grown on the carbon-face of silicon carbide substrates with approximately 25 (sample 1) and 90 layers (sample 2 and sample 3, fabricated with two different recipes).

Raman characterization (Fig. 1.a,b,c) shows a mainly single lorentzian shape of the 2D peak indicating loosely stacked layers. The presence of defects-induced D peak is expected to influence the non-saturable component of the graphene absorption.

The non-linear properties of the samples are investigated by open-aperture z-scan technique. A laser beam, emitted at 2.9 THz by a quantum cascade laser operating in pulsed regime (15% duty cycle, corresponding to 300 kHz frequency and 500 ns pulse width) and modulated at 4 Hz by a square function (50% duty cycle), is focalized with a beam waist of about 170 μ m onto the sample. The transmitted beam is detected by thin film-based single element thermopile detector (2M from Dexter) for several positions of the sample along the optical axis.

The normalized z-scan trace is then analyzed by assuming the simple two levels saturable absorber model and taking into account the temporal shape of the pulses train:

$$T(z) = \frac{1}{T_0} (1 - \alpha_{NS} - \frac{\alpha_s}{1 + \frac{I_0(z)}{I_s}})$$
(1)

where T_0 is the transmission in linear regime, α_{NS} and α_s are the non-saturable and saturable components of the absorption, respectively. I_0 is the beam intensity along the optical axis and I_s is the saturation intensity threshold, i.e. the intensity value at which the saturable absorption is reduced to 1/2. In all the samples the experiments show an increase of the transmission, as shown in Fig. 1.d,e. This corresponds to about 4% in sample 1 and 10% in samples 2 and 3. The saturation intensity is estimated to be tens of W/cm², that is 4 orders of magnitude lower than that in the telecommunication window [10], as expected from the much lower density of states in the THz. Unlike the shorter wavelengths region, where the non-saturable component is strongly influenced by the number of the layers, here the saturation of the absorption appears to be mainly influenced by the amount of defects in the graphene.

References

[1] K.S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva and A. A. Firsov, Science, **306** (2004) 666-669.

[2] F. Bonaccorso, Z. Sun, T. Hasan and A. C. Ferrari, Nature Photonic, 4 (9) (2010) 611-622.

[3] R. R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres and A. K. Geim, Science, **320** (2008) 1308.

[4] H. Zhang, S. Virally, Q. Bao, L. K. Ping, S. Massar, N. Godbout and P. Kockaert, Optics Letters **37** (2012) 1856-1858.

[5] H. Yang, X. Feng, Q. Wang, H. Huang, W. Chen, A.T. S. Wee and W. Ji, Nano Lett. **11** (2011) 2622-2627.

[6] E. Hendry, P. J. Hale, J. Moger, A. K. Savchenko and S. A. Mikhailov, Phys. Rev. Letter **105** (2010) 097401.

[7] F. T. Vasko, Phys. Rev. B 82 (2010) 245422.

[8] U. Keller, Nature 424 (2003) 831-838.

[9] I. H. Baek, H. W. Lee, S. Bae, B. H. Hong, Y. H. Ahn, D. I. Yeom and F. Rotermund, Appl. Phys. Express **5** (3) (2012) 032701.

[10] Q. L. Bao, H. Zhang, Y. Wang, Z. H. Ni, Z. X. Shen, K. P. Loh and D. Y. Tang, Adv. Funct. Mater. **19** (19) (2009) 3077-3083.

[11] Q. Bao, H. Zhang, Z. Ni, Y. Wang, L. Polavarapu, Z. Shen, Q.-H. Xu, D. Tang and K. P. Loh, Nano Res. 4 (3) (2011) 297-307.

[12] P. L. Huang, S.-C. Lin, C.-Y. Yeh, H.-H. Kuo, S.-H. Huang, G.-R. Lin, L.-J. Li, C.-Y. Su and W.-H. Cheng, Opt. Express **20** (3) (2012) 2460-2465.

[13] Z. Sun, T. Hasan, F. Torrisi, D. Popa, G. Privitera, F. Wang, F. Bonaccorso, D. M. Basko and A. C. Ferrari, ACS Nano 4 (2) (2010) 803-810.

[14] Z. Sun, D. Popa, T. Hasan, F. Torrisi, F. Wang, E. J. R. Kelleher, J. C. Travers, V. Nicolosi and A. C. Ferrari, Nano Res. **3** (9) (2010) 653-660.

[15] M. N. Cizmeciyan, J. W. Kim, S. Bae, B. H. Hong, F. Rotermund, and A. Sennaroglu, Opt. Lett. **38** (3) (2013) 341-343.

[16] S.A. Mikhailov and K. Ziegler, J. Phys. Condens. Matter 20 (2008) 384204.

[17] K. Yang, S. Arezoomandan and B. Sensale-Rodriguez, Terahertz Science and Technology 6 (4) (2013) 223-233.

[18] A. Tredicucci and M. S. Vitiello, Selected Topics in Quantum Electronics, IEEE Journal of, **20** (1) (2014) 130-138.

[19] Z. Zheng, C. Zhao, S. Lu, Y. Chen, Y. Li, H. Zhang and S. Wen, Opt. Express **20** (2012) 23201-23214.

Figures



Figure 1: a, b, c) Raman spectra of the sample 1, 2, 3, respectively; d, e) z-scan traces of the sample 1 and 3 with a pump beam at 2.9 THz and intensity at the focal point of about 223 mW/cm², respectively. The red line is the fitting curve based on Eq. (1).