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Experimental observation of low threshold optical bistability in exfoliated graphene with low oxidation degree

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ABSTRACT

We have experimentally investigated low threshold Optical Bistability (OB) and multi-stability in exfoliated graphene ink with low oxidation degree. Theoretical predictions of *N*-layer problem and the resonator feedback problem show good agreement with the experimental observation. In contrary to the other graphene oxide samples, we have indicated that the absorbance does not restrict OB process. We have concluded from the experimental results and Nonlinear Schrödinger Equation (NLSE) that the nonlinear dispersion – rather than absorption – is the main nonlinear mechanism of OB. In addition to the enhanced nonlinearity, exfoliated graphene with low oxidation degree possesses semiconductors group III–V equivalent band gap energy, high charge carrier mobility and thus, ultra-fast optical response which makes it a unique optical material for application in all optical switching, especially in THz frequency range.

1. Introduction

Since the last decade, graphene has been proposed as an outstanding nonlinear optical material due to its single layer atomic structure in hexagonal lattice which causes zero band gap energy, independent group velocity, high charge carrier mobility, tunable nonlinear optical conductivity and intense optical nonlinearity [1–9]. These particular aspects have led to the recent researches seeking for the applications in ultra-fast saturable absorption, biochemical sensing, beam splitting, optical power limiting and optical switching [2,9–23]. Primarily, Han Zhang et al. have reported the experimental z-scan measurement of nonlinear refractive index of loosely stacked few-layer graphene to be of order 10^{-11} m^2/W [24]. As well, Rui Wu et al. have experimentally studied the spatial self-phase modulation in graphene dispersion for the visible laser beam; this might imply the huge third order nonlinearity [9]. Subsequently, N. Liaros et al. have experimentally shown that graphene oxide dispersion can exhibit optical power limiting in the visible frequency range. They have deduced that saturable absorption and reverse saturable absorption can be provided by few layer graphene [19,22]. Optical Bistability (OB) as the feasible approach to all optical switching have been also investigated in graphene-silicon waveguide resonator and graphene-silicon photonic crystal cavity [16,25]. Most recently, OB has been reported in graphene nanobubbles [26].

On the other hand, theoretical investigation of optical nonlinearity in graphene invokes precise calculation of nonlinear conductivity by the means of Dirac cone representation which will give an estimation of optical nonlinear susceptibility [7,27–32]. S.A. Mikhailov has proved that the electromagnetic response of graphene in collisionless system is strongly nonlinear, especially for THz frequencies [32–35]. E. Hendry et al. have demonstrated an estimation of remarkable third order optical susceptibility in graphene flakes in the near infrared frequency range [36]. VI. A. Margulis et al. have then theoretically calculated the nonlinear refractive index of moderately doped graphene in the range from mid-infrared to ultraviolet; the results have declared the very high nonlinearity [29]. Today, the universal perception denotes the huge nonlinear response of graphene in THz frequency range resulting from the intraband conductivity in highly doped graphene, decreasing for the higher frequencies due to the interband transitions in lowly doped-undoped graphene. On this base, N.M.R. Peres et al. have investigated OB of monolayer, bilayer and ABA stacked trilayer graphene in THz range by solving Boltzman equation. However, the results have revealed that the attainable OB needs extra-large optical power [37]. Meanwhile, J.B. Khurgin has inferred in his paper that the huge values of nonlinear coefficient

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for interband transitions of graphene are due to the huge absorption coefficient and thus never lead up to the improvement in all optical switching. He has also deduced that all optical switching for intraband transitions requires 1000 doped graphene layers to be realized; but the act of doping will confront a breakdown [38].

In this paper, we show that the sufficient nonlinearity can be achieved in exfoliated graphene with low oxidation degree inside a resonator which will result in low threshold dispersive OB and multi-stability – rather than absorptive – at low optical input power. Performing the experimental results in the visible range, a coincidence will be unfolded as the paraphrase of *tunable OB in dielectric/graphene/dielectric heterostructures* in THz range proposed by Xiaoyu Dai et al. through their inspiration from the "nonlinearity enhancement via stacking several layers of graphene separated by a dielectric medium" implicated by N.M.R. Peres et al. in their paper [37,39]. To provide a theoretical justification, we admit E. Hendry's estimation of graphene third order nonlinearity and the modification presented by J.L. Cheng et al. [36,40]. We start with *N*-layer problem in which the nonlinearity enhancement and the mechanism of OB are expressed by the nonlinear conductivity in *N*-separated graphene flakes dispersed in a dielectric medium; we proceed with a numerical solution of Nonlinear Schrödinger Equation (NLSE) in order to analyze the resonator effect of *N*-separated graphene layers and the nonlinear dispersion/nonlinear absorption contribution in OB and multi-stability process. Then, we result an agreement of the theoretical predictions with our experimental results. Finally we propose exfoliated graphene with low oxidation degree for all optical switching.

2. Theory

2.1. N-layer problem

A system composed of *N* sequential separated graphene layers, dispersed in a dielectric medium is assumed. The system is illuminated by a visible laser beam with TE polarization as the optical input signal. The graphene flakes are not necessarily normal to the incident beam – which are exaggeratedly illustrated in Fig. 1 by the symbol G; D denotes the dielectric medium containing graphene flakes. Also, we note that the graphene layers are not equally displaced; Having recognized three different zones labeled with I, II & III in Fig. 1, the output electric field can be obtained starting from Maxwell equations and introducing the related boundary conditions through the following Eq. (1); where $E_i^{\mathrm{I,II,III}}$ and $E_T^{\mathrm{I,II,III}}$ stands for the incident and transmitted electric fields in three zones I, II & III respectively [39].

$$\begin{cases} \mathbf{I}: & E_i^{\mathrm{I}} = \frac{1}{8}E_T^{\mathrm{I}}\left(1+\frac{k_z^{\mathrm{II}}}{k_z^{\mathrm{I}}}\right)\Theta e^{-ik_z^{\mathrm{II}}d^{\mathrm{II}}} + \frac{1}{8}E_T^{\mathrm{I}}\left(1-\frac{k_z^{\mathrm{II}}}{k_z^{\mathrm{I}}}\right)(2\Lambda-\Theta)e^{ik_z^{\mathrm{II}}d^{\mathrm{II}}} \\ & \Lambda^{\mathrm{I}} = 2e^{-ik_z^{\mathrm{II}}\frac{d^{\mathrm{II}}}{2}} \\ & \Theta^{\mathrm{I}} = 4e^{-ik_z^{\mathrm{II}}\frac{d^{\mathrm{II}}}{2}} - \frac{\mu_0\omega}{k_z^{\mathrm{II}}}\left(\sigma_0+\frac{1}{4}\sigma_3\left|E_T^{\mathrm{II}}\right|^2|\Lambda|^2\right)\Lambda \\ \mathbf{II}: & E_T^{\mathrm{I}} = E_i^{\mathrm{II,1}}, \quad E_T^{\mathrm{II,i}} = E_i^{\mathrm{II,i+1}}, \quad i=1,2,\ldots,N+1 \\ & E_i^{\mathrm{II},i} = \frac{1}{4}E_T^{\mathrm{II},i}\Theta e^{-ik_z^{\mathrm{II}}\frac{d^{\mathrm{II}}}{2}} \\ & \Lambda^{\mathrm{II}} = 2e^{-ik_z^{\mathrm{II}}\frac{d^{\mathrm{II}}}{2}} \\ & \Theta^{\mathrm{II}} = 4e^{-ik_z^{\mathrm{II}}\frac{d^{\mathrm{II}}}{2}} - \frac{\mu_0\omega}{k_z^{\mathrm{II}}}\left(\sigma_0+\frac{1}{4}\sigma_3\left|E_T^{\mathrm{II}}\right|^2|\Lambda|^2\right)\Lambda \\ \mathbf{III}: & E_T^{\mathrm{II},N+1} = E_i^{\mathrm{III}} \\ & E_i^{\mathrm{III}} = \frac{1}{4}E_T^{\mathrm{III}}\Theta e^{-ik_z^{\mathrm{II}}\frac{d^{\mathrm{II}}}{2}} \\ & \Lambda^{\mathrm{III}} = \left(\frac{k_z^{\mathrm{III}}}{k_z^{\mathrm{II}}}+1\right)e^{-ik_z^{\mathrm{II}}d} + \left(1-\frac{k_z^{\mathrm{III}}}{k_z^{\mathrm{II}}}\right)e^{ik_z^{\mathrm{II}}d} \\ & \Theta^{\mathrm{III}} = 2\left(1+\frac{k_z^{\mathrm{III}}}{k_z^{\mathrm{II}}}\right)e^{-ik_z^{\mathrm{II}}\frac{d^{\mathrm{II}}}{2}} - \frac{\mu_0\omega}{k_z^{\mathrm{II}}}(\sigma_0+\frac{1}{4}\sigma_3\left|E_T^{\mathrm{II}}\right|^2|\Lambda|^2)\Lambda \end{cases} \tag{1}$$

$k_z^{\mathrm{I,II,III}} = \sqrt{k_0^2\varepsilon^{\mathrm{I,II,III}} - k_0^2\varepsilon^{\mathrm{I}}\sin^2\theta}$; where $k_0 = \frac{2\pi}{\lambda_0}$ and λ_0 is the free space laser beam wavelength; $\varepsilon^{\mathrm{I,II,III}}$ pertains to the medium dielectric constant; θ is the incident beam angle. d^{II} is the mean dielectric slab distance between flakes; μ_0 is the free space permeability; $\omega = 2\pi c/\lambda_0$ in which c is the light velocity in free space. σ_0 is the isotropic surface conductivity and can be written as the sum of interband σ_{inter} and intraband σ_{intra} expressions given in Eqs. (2) and (3) respectively [32–36,40].

$$\sigma_{\mathrm{inter}} = \frac{ie^2}{4\pi\hbar}\ln\left|\frac{2E_F-(\omega+i\tau^{-1})\hbar}{2E_F+(\omega+i\tau^{-1})\hbar}\right|, \tag{2}$$

$$\sigma_{\mathrm{intra}} = \frac{ie^2k_BT}{\pi\hbar^2(\omega+i/\tau)}\left[\frac{E_F}{k_BT}+2\ln(e^{-\frac{E_F}{k_BT}}+1)\right]. \tag{3}$$

On the other side, σ_3 is the nonlinear conductivity of grahene flakes which can be obtained by Eq. (4) for interband transitions over the visible frequency range [32–36,40].

$$\sigma_3 = -\frac{ie^2}{\hbar}\left(\frac{eV_F}{\hbar\omega^2}\right)^2(1+i\alpha_T) \tag{4}$$

In Eqs. (2)–(4), e is the electron charge, $\hbar$ is the reduced Planck's constant, k_B is the Boltzmann constant, E_F is the Fermi energy, τ is the electron-photon relaxation time, T is the temperature. α_T is two

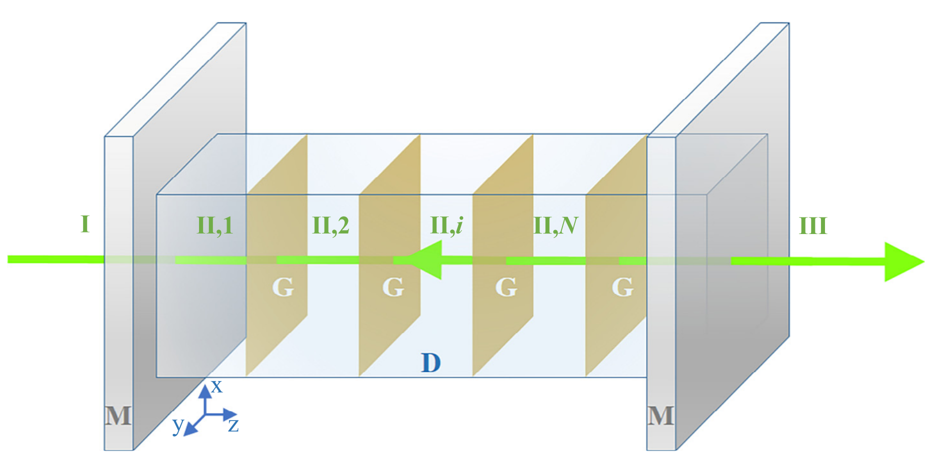

Fig. 1. Schematic illustration of graphic layers dispersed in dielectric system.