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Period doubling and route to chaos in reduced graphene oxide, an experimental evidence

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We use Nonlinear Schrödinger Equation (NLSE) to investigate chaotic dynamics inside a graphene-based optical system. Numerical results show that with increasing the control parameters, the character of Modulation instability (MI) is changed from convective to absolute. In consequence, a hyper-chaotic map is more expressive to explain the dynamical state of the system. It is also indicated that a reverse transition from the chaotic regime to the stable state can be resulted if the optical input intensity is further increased. The optical input intensity and feedback strength are distinguished as the two control parameters of the system. We then use Reduced Graphene Oxide (RGO) dispersion with large nonlinear refractive index to verify if a transition to instability and chaotic behavior is experimentally realized for the visible input light wave. The observation begins with saturation in the nonlinear response followed by an Optical Bistability (OB) phenomena. Afterward, a quasi-periodic fluctuations appear expressing the absolute MI. In agreement with our simulation result based on the period doubling bifurcation diagram, this quasi-periodic state is recognized as a route to the chaos. We infer that the graphene Quantum Dots (QDs) which are formed as a result of reduction process are responsible for energy transfer through the optical near-field interactions and thus, strengthen the required feedback.

1. Introduction

Chaotic wave propagation has been contemplated since the 90th decade as a security agent for optical communication. Developing toward the ultrafast pulsed lasers, the need to control the chaos has been seriously taken into account. B. R. Andrievskii, et al. have declared methods and applications of controlling chaos through their review article [1]. L. Illing, et al. have presented a full study of controlling the optical chaos. They have explained that the optical chaos communication can be achieved by electro-optical feedback devices [2].Today, the investigation on chaotic dynamics is expanded to the novel fields of optics and photonics for which the stability considerations seem to be crucial. Chaos is a characteristic feature of the nonlinear optical systems using a feedback mechanism. The latter can be provided by a resonating cavity (e.g. a Fabry-Perot cavity) in which the nonlinear medium is laid. Accordingly, a high power optical beam can interact with the nonlinearity; if the feedback wave is retarded enough to be formed such that the delay time becomes longer than the nonlinear medium response time, the interaction will undergo a complex chaotic procedure [3,4]. The route to chaos may be different from one optical system to another. Previously, it has been shown that the Ikeda instability is a proper chaotic map to describe the dynamical state of an unstable optical system [5–8]. However, it is now revealed that a hyper-chaotic procedure can interfere in. On the other hand, several literature contexts have addressed the effect of Modulation Instability (MI) as a prior state to chaotic regime in a typical nonlinear optical system. MI occurs when the balance between the nonlinearity and dispersion/diffraction is violated. The consequence is the occurrence of a quasi-periodic state. For certain criterion, the character of MI changes from convective to absolute. In this case, the amplitude of the quasi-periodic state will grow fast. Subsequently, an unpredictable state will appear in which tracing the nonlinear dynamics will reveal a chaotic behavior. [8–11]. Before beginning the MI procedure, depending on the feedback strength, the system can either experience a saturation in nonlinearity, known as optical limiting or jump to upper stable states recognized as Optical Bistability (OB) and multistability.

Nanostructure based optical systems exhibit extremely large nonlinearity and can serve as suitable nonlinear media for different applications like ultrashort pulse generation, optical modulation, plasmonics, etc. [12–25]. Herein, the propagation of optical wave is expected to be unstable even if a low input power is illuminated to the nanostructure based nonlinear media; a challenge that intensely influences on the device performance through a desired application. A set of studies have

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theoretically investigated and explained the circumstances of OB and multistability in nanostructure based optical systems [26–31]. In contrast, a few number of papers have experimentally reported the observation of OB and multistabilty [32–35]. Meanwhile, the investigation of chaotic dynamical state is only limited to the theoretical studies with consideration of sub-atomic interactions and quantum coherence [36]; few of them have dealt with the nonlinear medium related measurable/macroscopic quantities required for evaluating an experimental report.

Graphene is a 2D nanostructure that exhibits strong and fast optical nonlinear response [35,37–43]. On this base, graphene - based optical devices and systems have been proposed for nonlinear optical applications [12–15,18,20]. Through the previous article, OB and multistability have been experimentally verified in few layer graphene oxide ink for the low threshold optical power at a visible frequency. It has been theoretically predicted that the nonlinearity can be enhanced for terahertz frequency range inasmuch as the output wave can be unstable and chaotic [35]. In this study, we show that period doubling and a transition to chaos can be experimentally occurred for a visible light inside a medium containing few layer dispersed Reduced Graphene Oxide (RGO). We theoretically explore the procedure of route to chaos in accordance with nonlinear optical characteristics of the medium.

2. Theory

2.1. Graphene nonlinearity

Third order nonlinear optical conductivity of single layer graphene can be written as Eq.(1) for interband transitions if the Third Harmonic Generation (THG) is generally assumed as the prevailing nonlinear phenomena [38,39,41].

$$\sigma^{(3)}(\omega_1,\omega_2,\omega_3)=\frac{(e^2/4\hbar)(\hbar v_F e)^2}{\hbar^4(\omega_1+\omega_2)(\omega_2+\omega_3)(\omega_3+\omega_1)\omega_t}, \tag{1}$$

e is the electron charge, $\hbar$ is the reduced Planck's constant, v_F is the Fermi velocity and $\omega_t=\omega_1+\omega_2+\omega_3$. The third order nonlinear susceptibility can be given by Eq. (2) [38,39,41].

$$\chi^{(3)}(\omega_1,\omega_2,\omega_3)={}^{\sigma^{(3)}(\omega_1,\omega_2,\omega_3)}\Big/_{(-i\omega_t\varepsilon_0 d_{gr})}, \tag{2}$$

where ε_0 is the free space permittivity and d_{gr} is the thickness of single layer graphene. An estimation of the nonlinear refractive index n_2 can be obtained by Eq. (3).

$$n_2=3\chi^{(3)}/\left[4\varepsilon_0 c\left(1+\chi^{(1)}\right)\right], \tag{3}$$

in which c is the free space light velocity and $\chi^{(1)}=\sigma^{(1)}/(-i\omega\varepsilon_0 d_{gr})$ is the linear susceptibility in terms of surface conductivity $\sigma^{(1)}$ given in Eq. (4).

$$\sigma^{(1)}(\omega)=i\left(\frac{e^2}{4\pi\hbar}\ \ln\left|\frac{2\mu-(\omega+i\tau^{-1})\hbar}{2\mu+(\omega+i\tau^{-1})\hbar}\right|+\frac{e^2k_BT}{\pi\hbar^2(\omega+i/\tau)}\left[\frac{\mu}{k_BT}+2\ \ln\left(e^{-\frac{\mu}{k_BT}}+1\right)\right]\right), \tag{4}$$

μ is the chemical potential of graphene; k_B is the Boltzmann's constant; T denotes the temperature and τ is known as relaxation time [38,39,41].

2.2. Reduced Graphene Oxide (RGO)

RGO is an alternative to the pristine graphene for its similar optical and electrical characteristics. However, the preparation of RGO is easier compared to that of graphene considering the simple and low-cost preparation process for GO and the availability of multifarious reduction methods [42–50]. On the side, the degree of reduction determines the optical and electrical characteristics. This will provide a wide range of selection for optical transparency, band gap and nonlinearity [50–53].

2.3. Suspended graphene nonlinearity

If a single layer graphene is suspended inside a dielectric medium, the nonlinear response due to THG will be resonated for certain thicknesses of dielectric slab d given in Eq. (5).

$$d=(\lambda/2\sqrt{\varepsilon})m,\qquad m=0,1,2,\ldots \tag{5}$$

in which λ is the light wavelength and ε is the dielectric permittivity. If this nonlinear system is supported on a metal thin film, m should be replaced with $m+1/2$ in Eq. (5) and a giant enhancement of nonlinear response will be resulted for resonating modes [42]. This implies the increasing in the nonlinear optical conductivity and subsequently the effective nonlinear refractive index.

Eq. (5) is also valid for a suspended RGO sheet in accordance with Section 2.2. If otherwise, it is assumed that multiple RGO flakes are embedded inside a dielectric medium, the nonlinearity enhancement will be further increased. This is an experimental verification [32,35] which will be shown in the experimental section by increasing the RGO suspension concentration.

2.4. Nonlinear dynamics

Using the density matrix formalism, Makoto Naruse et al. have shown that for a nanoscale optical system containing a pair of Quantum Dots (QDs), chaotic oscillation can occur as a result of energy transfer through the optical near-field interactions [36]. When a time delay incorporates between two energy transfers within the nonlinear system, optical pulses will appear even if the system is pumped by a cw radiation. Consequently, QDs are refereed as optical nano-pulsers. These optical pulse trains can grow fast in amplitude if the QDs' pairs are strongly coupled. Solving for nano-pulsers' population, chaotic oscillatory behavior will be obtained for certain criterion [36].

Indeed, the latter mentioned nanoscale optical system stands for a more inclusive concept: the spaser; its nonlinear mechanism is analogous to that of a laser cavity for which the mirrors and coherently generated photons are respectively substituted with the couples of QDs and localized surface plasmon polaritons. In the presence of thousands of QDs' pairs coupled with near-field interactions, surface plasmon polaritons can be intensely amplified. Thereupon, the ultrafast quasi-periodic behavior may be complex and unpredictable and a transition to chaos will be possible. For an exact nonlinear analysis, it is essential to estimate the plasmon resonance modes. This will need information about the compositional/peripheral structure of the nanoscale optical system including the nanoparticles' size, shapes, dielectric function, etc. [54]. Notwithstanding, evolution of instability in a spaser is still an open topic for researchers.

In this study, in order to investigate the nonlinear dynamics inside a mesocopic optical system containing RGO dispersion, we use an alternative formalism i.e. the Nonlinear Schrödinger Equation (NLSE) which is given in Eq. (6).

$$\frac{\partial U}{\partial z}+\frac{1}{2}ik_2\frac{\partial^2 U}{\partial t^2}=i\gamma|U|^2U, \tag{6}$$

U is the slowly varying complex amplitude; t and z determine the time and light propagation direction respectively; $\gamma=2n\varepsilon_0 n_2\omega$ is the nonlinearity whereas n is the refractive index; $k_2=d/d\omega(1/v_g(\omega))_{\omega=\omega_0}$ in which ω is the angular frequency, ω_0 denotes the free space frequency and v_g is the group velocity [55]. If we seeks unstable solutions, we should numerically solve NLSE by a Finite Difference based method like Predictor- Corrector Method (PCM) [56] from which the properties