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Published in:
Nature Photonics

DOI:
10.1038/s41566-018-0201-9

Published: 01/07/2018

Document Version
Peer reviewed version

Please cite the original version:
Sun, Z. (2018). Electrically tuned nonlinearity. Nature Photonics, 12(7), 383-385. https://doi.org/10.1038/s41566-018-0201-9

Subject codes:

Physical sciences/Materials science/Nanoscale materials/Graphene/Optical properties and devices [URI 639/301/357/918/1054]

Physical sciences/Optics and photonics/Optical physics/Nonlinear optics [URI 639/624/400/385]

Strap: 2D MATERIALS

Title: Electrically tuned nonlinearity

Standfirst: *The demonstration of broadband, electrically-tunable third-order nonlinear optical responses in graphene is promising for a host of nonlinear optical applications.*

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Nonlinear optics is the branch of photonics dedicated to the nonlinear interaction between light and matter, more specifically when the induced dielectric polarization in a material responds nonlinearly to the electric field of light. In 1961, almost one year after the invention of the first laser, Peter Franken and co-workers observed the nonlinear effect of second harmonic generation (SHG) for the first time with a pulsed ruby laser, where two photons with the same frequency are converted into a new photon having twice the frequency of the incident photons [1]. This demonstration marked the birth of nonlinear optics. Currently, various devices enabled by nonlinear optics are extensively used throughout our daily lives [2,3]: for example, ultrafast lasers for micro-machining and micro-surgery; various forms of nonlinear optical spectroscopies for medical diagnostics, bio-sensing and imaging; and systems for optical telecommunications and security.

Now, writing in *Nature Photonics*, Shiwei Wu and co-workers report that a nonlinear optical process called third-harmonic generation (THG) can be widely tuned in graphene using an electric gate voltage [4]. Electrically tunable SHG has already been reported in other two-dimensional (2D) materials, namely monolayer WSe_2 (a well-studied semiconductor in the family of transition metal dichalcogenides) with excitons but the spectral bandwidth was limited [5].

Graphene, a monolayer of carbon atoms packed into a 2D honeycomb lattice, exhibits a strong ultra-wideband light-matter interaction, which has been utilized for a broad range of photonic and optoelectronic devices including photon sources, modulators and photodetectors [6]. For nonlinear optics, second-order optical nonlinearity in graphene is forbidden due to inversion symmetry, but third-order optical nonlinearity in graphene is extremely large [7]. The doping dependent third-order nonlinear optical responses in graphene have been previously reported [7,8]. For example, saturable absorption, where the material absorption reduces at high incident light intensity, is a widely-used third-order nonlinear optical response for ultrafast pulse generation. It has been shown that saturable absorption in graphene is doping dependent and electrically tunable for high performance ultrafast lasers [7,8].

The latest results from Wu and co-workers [4] originate from the ability to adjust the chemical potential (E_f) of graphene in order to selectively switch on or off single and multi-

photon resonant transitions (Figure 1). A maximum tuning strength of up to ~30 times is achieved with a change of ~ 0.74-electronvolt in chemical potential at an excitation wavelength of 1566 nm. Similar results have also been demonstrated by Andrea C. Ferrari et al [9], indicating that gated (or doped) graphene is more suitable than chemically pristine graphene samples for THG applications if no tuning is needed. Benefits are enhanced third-order nonlinear optical susceptibility $|\chi^{(3)}|$ and reduced linear optical absorption which is necessary for devices with a low insertion loss.

Wu et al also demonstrate electrical manipulation of various four-wave mixing (FWM) processes, a third-order nonlinear optical process where two or three photons are mixed to generate one or two photons at new frequencies. The team found that difference-frequency FWM behaves differently from THG and sum-frequency FWM due to phase differences of one- or multi-photon resonant transitions. Note that such gate-tunable nonlinear optical responses with multi-photon resonance selection should exist in other nonlinear optical processes, such as high-order harmonic generation.

The reported operation bandwidth of the gate-tunable THG is from ~1300 to 1650 nm, covering the most common spectral range for optical fibre telecommunications at 1550 nm. Such a broad operation bandwidth comes from the linear energy dispersion of the graphene Dirac fermions (Figure 1). This is fundamentally different from the previous demonstration of the electrically tunable SHG with excitons in monolayer WSe_2, which has a limited operation bandwidth (around few tens of millielectronvolts at low operating temperature) because of the narrow band of the exciton transition energy [5]. In theory, broader bandwidth operation of gate-tunable nonlinear optics in Dirac materials should be possible because a longer operation wavelength (i.e., the mid-infrared spectral region) is naturally covered with smaller doping, while a shorter operation wavelength (i.e., the visible spectral region) is feasible with higher doping. Other interesting Dirac materials such as topological insulators and Weyl semimetals which are less explored for nonlinear optics are also worth further investigation for basic science and potential applications. Nevertheless, the broadband gate-tunable optical nonlinearities in graphene offer a new approach to build electrically-tunable nonlinear optical devices.

In the past decade, studies of nonlinear optics with 2D materials (including graphene and other 2D layered materials) and their mixed-dimensional heterostructures have witnessed significant progress. However, there is a huge variation in the measured nonlinear optical responses, with results for the same material which can be different by a few orders of magnitude, for example as in the case of the $|\chi^{(3)}|$ of graphene and second-order nonlinear optical susceptibility $|\chi^{(2)}|$ of MoS_2 (the most studied semiconductor in the family of 2D transition metal dichalcogenides) [7]. The latest results reported by Wu et al. [4] indicate that the large variation in $|\chi^{(3)}|$ of graphene at different doping levels needs to be considered to obtain a fair comparison of nonlinear optical processes.

The nonlinear interaction between light and a nano-material typically builds up coherently along the interaction length. Graphene and other 2D materials contain only one or few atomic layers, which thus have a very limited interaction length. Therefore, the frequency conversion efficiency of 2D materials is typically low (e.g., $\sim 3\times10^{-10}$ % in the experiment reported by Wu et al [4]) despite having a large $|\chi^{(3)}|$. Future research efforts will likely be dedicated to finding ways to enhance the nonlinear optical interaction in 2D materials using various approaches, including heterostructures, phase-matching methods, waveguide/fibre integration, and optical resonators as displayed in Figure 2a-c. Furthermore, various polaritons (e.g.,

plasmon, phonon and exciton polaritions, Figure 2d) and photonic metamaterials can provide localized enhancement and manipulation of the optical nonlinearity in 2D materials and their mixed-dimensional heterostructures.

The development of electrically tunable nonlinear optical materials will likely play an increasingly important role in every aspect of photonics. It has already enabled diverse and widely-used photonic devices (e.g. pulsed lasers, switches, modulators, and memories), underscoring the unparalleled advantages of photonic techniques over their electronic counterparts. In recent years, exciting nonlinear nanophotonic applications including on-chip photonics, quantum nanophotonics, nonlinear plasmonics, and strong-field nanophysics have also attracted attention. However, the present solutions to these applications using traditional bulk crystals have hit a technical limit imposed by their characteristics such as a relatively small nonlinear optical susceptibility and their complex and expensive fabrication and integration methods.

Graphene and other 2D materials with large nonlinear optical responses offer the benefit of being compatible with chip-integration and thus are very promising for tackling the upcoming challenges of nonlinear nanophotonics and nanophysics. The gate-tunable nonlinear optics mechanisms of graphene and graphene-like materials can, in principle, offer various technical advantages, such as devices with a compact footprint, extremely fast speed (>a few tens of GHz) and compatibility with complementary metal oxide semiconductor technology (CMOS), all of which are desirable for future on-chip photonic and optoelectronic applications. If nonlinear optical interaction enhancement in 2D materials and the production of large-scale and high-quality 2D materials are successful, it is anticipated that 2D materials have the great potential to enable completely different approaches to construct electrically-tunable nonlinear optical nano-devices (e.g. frequency combs, ultrafast lasers, terahertz components, quantum sources, optical parametric sources) for new and emerging applications in metrology, sensing and imaging, quantum technology, and telecommunications.

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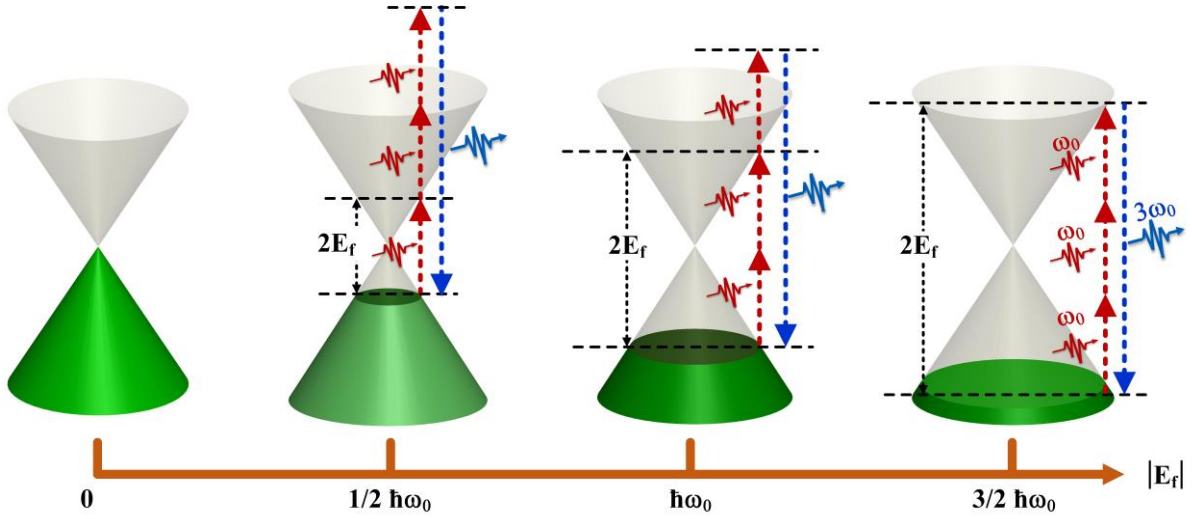

Figure 1 Multi-photon resonance effects in graphene. The increase of $|E_f|$ *can successively switch off one-photon (*$|E_f|>1/2\ \hbar\omega_0$*), two-photon* ($|E_f|>\hbar\omega_0$)*, and three-photon* ($|E_f|>3/2\ \hbar\omega_0$) *inter-band transitions by Pauli-blocking in graphene. Note that two-photon inter-band transitions and one-, three-photon inter-band transitions contribute, respectively, to* $\chi^{(3)}$ *positively and negatively [4]. The red arrows indicate the input photons at* ω_0 *frequency and the blue arrows indicate the generated third harmonic photons at* $3\omega_0$ frequency.

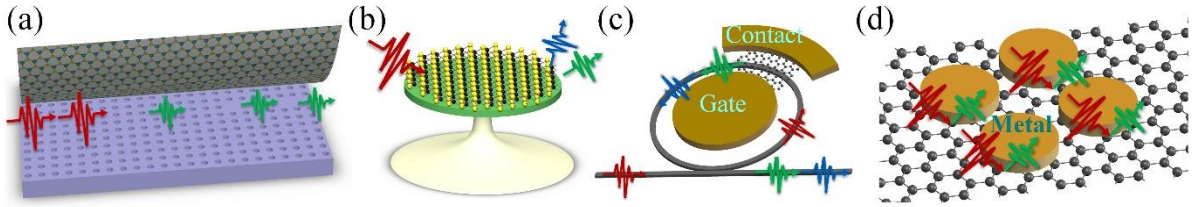

Figure 2 *Enhancement and manipulation of nonlinear optical responses in 2D materials with various methods: (a) Photonic crystal cavity; (b) Micro-disk resonator; (c) Electrically tunable micro-ring resonators; (d) Plasmonic structures. The red arrows indicate the input photons, and the blue and green arrows indicate the generated photons at different frequencies.*