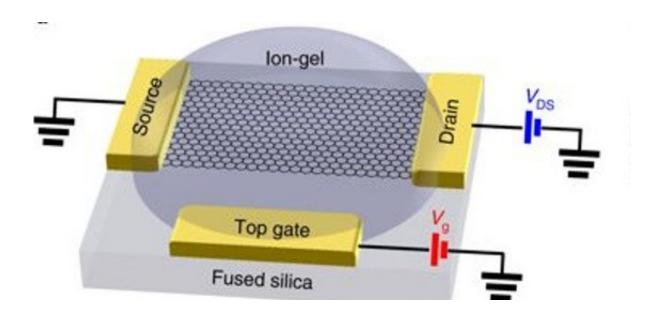


Electrically tunable third-order nonlinear optical response in graphene

August 1 2018, by Thamarasee Jeewandara



Schematic for gate tuning the chemical potential: the ion-gel gating method was adopted in the study using the field-effect transistor structure with graphene (2D hexagonal lattice) supported by fused silica. The device structure measured the linear and nonlinear optical responses of graphene at room temperature and monitored the chemical potential (Ef) versus the gate voltage (Vg) in situ. Credit: *Nature Photonics*, doi: 10.1038/s41566-018-0175-7

The research focus on 2-D materials has intensified with its potential to modulate light for superior performance and realize applications that can enhance existing technologies. Graphene, the best known 2-D material, derived from 3-D graphite, constitutes a monolayer of carbon atoms arranged in a 2-D hexagonal lattice, exhibiting strong ultra-wideband



light-matter interactions, able to operate at an extremely broad spectral range, suited for next-generation <u>photonics</u> and <u>optoelectronic devices</u>. The unique electronic properties of graphene originate from <u>Dirac cones</u>, features in electronic band structures that host charge carriers of zero effective mass, so-called massless Dirac fermions that occur in 2-D materials. Materials scientists are currently at a stage of experimental infancy to realize many interesting properties of the <u>nonlinear optical</u> responses of graphene, to aid its promise to disrupt existing technology and facilitate wide-ranging applications.

The birth of nonlinear optics is credited to an <u>experiment conducted in 1961</u> by Peter Franken and co-workers with a pulsed ruby laser, in which they observed the nonlinear effect of second-harmonic generation (SHG, frequency doubling) for the first time. Dynamic control of optical nonlinearities remains confined to research laboratories as a spectroscopic tool at present.

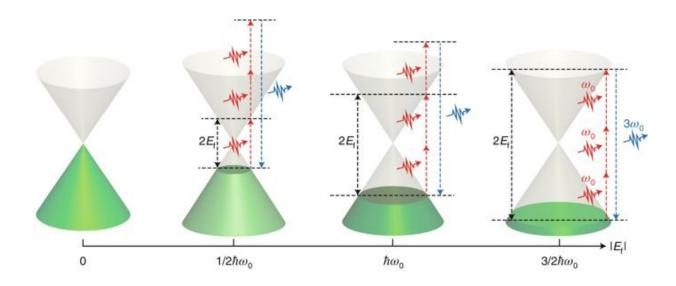
Now writing in *Nature Photonics*, Tao Jiang et al. report that nonlinear third-harmonic generation (THG, frequency tripling) can be widely tuned in graphene using an electric gate voltage. This has many potential applications—gate-tunable, nonlinear optical mechanisms of graphene and other 2-D graphene-like materials are desirable to engineer future on-chip photonic and optoelectronic applications with extremely high speed and complementary metal-oxide semiconductor (CMOS) compatibility for device fabrication. Electrically tunable second-harmonic generation was previously reported in other 2-D materials, such as Tungsten diselenide (WSe₂) with excitons, although the spectral bandwidth was limited. Experimentally, tuning the input frequencies or the chemical potential (E_f) of graphene can provide detailed information about the third-order nonlinear optical response, thus far suggested in theory.

Third-order nonlinear processes are also known as <u>four-wave mixing</u>, as they mix three fields to produce a fourth. The latest results from Jiang et



al. originate from the ability to adjust the chemical potential (E_f) of graphene and electrically switch on or off single photon and multiphoton resonant transitions with ion-gel gating (also known as gate-controlled doping), for a given set of input frequencies. The experimental results matched well with theoretical calculations to provide a firm basis to comprehend third-order nonlinear optical processes in graphene and graphene-like Dirac materials.

The operation bandwidth of gate tunable THG ranged from ~ 1300 nm to 1650 nm, covering the most common spectral range for optical fibre telecommunications at 1550 nm. Such a broad operation bandwidth resulted from the energy distribution of the graphene Dirac fermions. The observation is similar to a parallel investigation published in *Nature Nanotechnology* to electrically control the THG efficiency (THGE) of graphene, likewise attributed to massless Dirac fermions. Overall, the experimentally observed broadband gate-tunable optical nonlinearities of graphene offer a new approach to build electrically tunable nonlinear optical devices in practice.



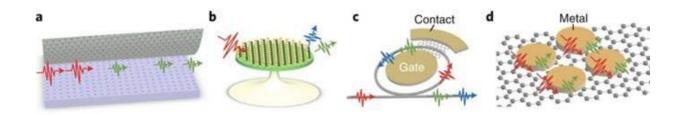
Schematic representation of the multiphoton effects in graphene Dirac fermions:



the increase of chemical potential |Ef| can successfully switch off one-photon (|Ef| > $1/2\hbar\omega0$), two-photon (|Ef| > $\hbar\omega0$), and three-photon (|Ef| > $3/2\hbar\omega0$) interband transitions by Pauli blocking. Two-photon interband transitions contribute to third-order nonlinear optical susceptibility [$\chi(3)$] positively, while one- and three- photon interband transitions contribute negatively. Red arrows indicate the input photons at $\omega0$ frequency and the blue arrows indicate the generated third-harmonic photons at $3\omega0$ frequency. \hbar , reduced Planck constant. Credit: *Nature Photonics*, doi: 10.1038/s41566-018-0201-9.

Existing electronic interconnections (copper cables) for instance, suffer bandwidth loss due to performance restrictions, impeding accelerated information processing required for media streaming, cloud computing and the internet of things (IoT). A growing need exists to regulate light and develop compact, cost-effective, high-performance optical interconnects for higher bandwidth and lower loss.

Future research efforts are likely to enhance the observed effects using a variety of approaches including waveguide/fibre integration and <u>optical</u> <u>resonators</u>. In addition, various <u>polaritons</u> and photonic <u>metamaterials</u> can provide localized enhancement and manipulation of optical nonlinearities in 2-D materials to <u>create surface plasmons</u> and tackle the foreseen challenges of nonlinear nanophotonics and <u>nanophysics</u> device development, with advanced optical solutions.



Methods to enhance and manipulate nonlinear optical responses in 2D materials:



a) photonic crystal cavity, b) microdisk resonator, c) electrically tunable microring resonator, d) plasmonic structure. Red arrows = input photons, blue and green arrows = photons generated at different frequencies. Credit: *Nature Photonics*, doi: 10.1038/s41566-018-0201-9.

The knowledge can be extended to other nonlinear optical processes in graphene, including high-order harmonic generation. The existing technology with traditional bulk crystals has hit a technical limit to realize the envisioned optoelectronic applications, due to their relatively small nonlinear optical susceptibility and the complex and expensive, fabrication and integration methods. The demonstrated nonlinear optical interaction enhancement in 2-D materials should ideally be developed alongside large-scale and high-quality 2-D material production, to enable completely different approaches for electrically tunable nanodevice construction. Such nanodevices may facilitate the proposed advances in metrology, sensing, imaging, quantum technology and telecommunications.

More information: Gate-tunable third-order nonlinear optical response of massless Dirac fermions in graphene www.nature.com/articles/s41566-018-0175-7 Jiang et al, 21 May 2018, *Nature Photonics*.

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