Extreme nonlinear response in graphene

A. Picón[1,*], Óscar Zurrón[1], L. Plaja[1]

[1]Grupo de Investigación en Aplicaciones del Láser y Fotónica, Departamento de Física Aplicada, Universidad de Salamanca, E-37008, Salamanca, Spain

* antonio.picon@usal.es

High-order harmonic generation (HHG) is a remarkable process resulting from the interaction of physical systems with intense electromagnetic radiation. The generation of high-order harmonics is well-established in atomic and molecular gases. However, much progress have been done towards HHG in solids since the first experimental observation [1]. Atomic and molecular gases and crystalline solids emit high harmonics at high intensities and share some common basic principles [2]. In these systems, harmonics are generated by electrons initially bounded, which are first promoted to free or quasi-free states during the interaction with a non-resonant intense laser field. Once freed, the electrons are accelerated by the same electric field and they may be driven back to the parent ion, to subsequently release the acquired kinetic energy in the form of high-frequency radiation. Standard HHG experiments are performed with atomic gases, and a wide range of fundamental processes and applications have been investigated in these simple systems, from imaging and spectroscopy with sub-femtosecond resolution to generation of XUV/soft-x-ray coherent pulses [3,4]. However, in the recent years HHG from solids has burgeoned a great interest, mainly motivated by the quadratic scaling of the harmonic conversion efficiency with the density of the target, as a result of the coherent nature of the process.

HHG spectra are characterized by an extended plateau in the high-harmonic peaks, a feature of the nonlinearity of this process, and the sudden end of the plateau at a particular frequency, the so-called the cut-off frequency. Although the similarities of HHG in atomic gases and solid-state materials, they are fundamental differences in the laws governing the cut-off frequency. These illustrate fundamental differences in the nonlinear response of the system and the dynamics responsible for HHG emission. The induced dynamics have well investigated and understood in these recent years in semiconductor materials [2]. However, other materials, such as graphene or two-dimensional materials, have not been explored. In particular, HHG from graphene have not been experimentally observed.

Recently we have developed a new theoretical approach to describe the nonlinear response of grapheme based on semiconductor like Bloch equations (SBEs) in the reciprocal space. By using this approach, that allows us to treat exactly the nonadiabatic dynamics near the Dirac points, we show the possibility to produce HHG with few-cycle laser pulses at mid-IR wavelengths at intensities below the damage threshold, see Fig 1. The carrier dynamics in SLG is an intricate process, resulting from the superposition of the contributions of interband as well as intraband transitions. Interestingly, we show that the main underlying mechanism of HHG is quite different to semiconductor materials. The first step is dominated by resonant excitation, at variance with tunneling excitation in semiconductor materials, and the cut-off frequency is related to the energy difference between valence and conduction band where electrons are located during the dynamics, this restricts the maximum cut-off frequency. The understanding of the induced dynamics is fundamental to create in the future materials that allow us to tailor and control the emission of HHG by demand.

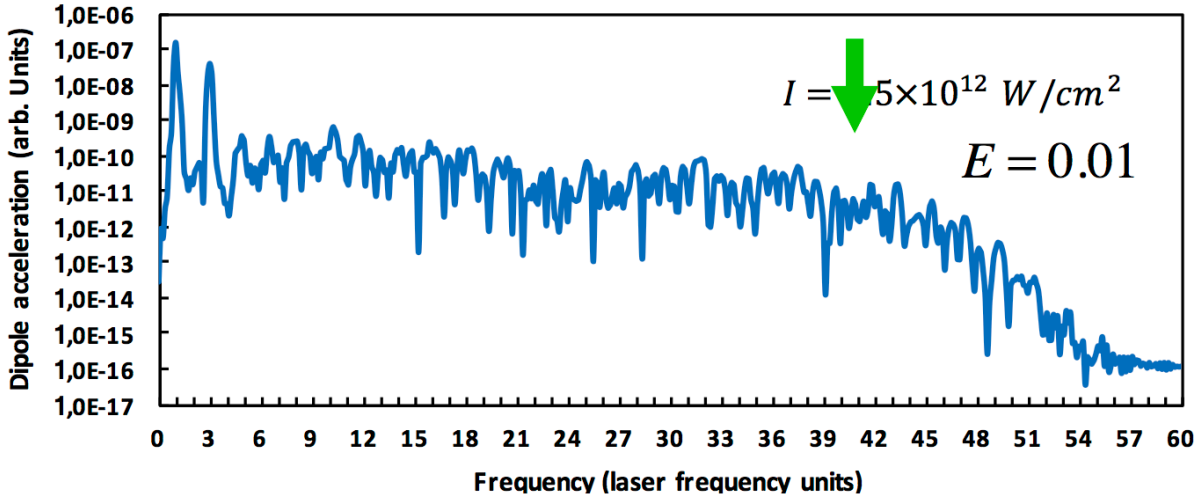

Figure 1: Calculated high-harmonic spectrum for graphene induced by an 8-cycle intense mid-IR pulse. We use a pulse with 3μm wavelength and intensity 1.5x10^{12} W/cm^2.

Acknowledgments

We acknowledge support from Junta de Castilla y León (Project SA116U13, SA046U16) and MINECO (FIS2013-44174-P, FIS2015-71933-REDT) and European Union (FEDER). A.P. acknowledges support from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No. 702565.

References

[1] S. Ghimire, A.D. DiChiara, E. Sistrunk, P. Agostini, L.F. DiMauro, and D.A. Reis, Nat. Phys. **7,** 138 (2011).
[2] G. Vampa, T.J. Hammond, N. Thiré, B.E. Schmidt, F. Légaré, C.R. McDonald, T. Brabec, and P. B. Corkum, Nature **522**, 462 (2015).
[3] L. Plaja, R. Torres, and A. Zäir, Wiley-Vcv Verlag (Weinheim, 2014).
[4] F. Krausz and M. Ivanov, Rev. Mod. Phys. **81**, 163 (2009).