Boosting the terahertz nonlinearity of graphene by orientation disorder

In Hyung Baek*(1), Fabian Rotermund(2), and Young Uk Jeong(1)
(1) Center for Quantum-Beam-based Radiation Research, Korea Atomic Energy Research Institute, South Korea
(2) Department of Physics, Korea Advanced Institute of Science and Technology, South Korea
*ihbaek@kaeri.re.kr

1. Introduction

The conical band structure is the cornerstone of graphene's ultra-broadband optical conductivity [1,2]. For practical use of graphene in nonlinear photonics, however, substantial increases of the light-matter interaction strength will be required while preserving the promising features of monolayers, as the interaction of light with a single atomic layer is limited due to the extremely short interaction length and low density of state, particularly for the long-wavelength region. Here, we report that this demand can be fulfilled by random stacking of high-quality large-area monolayer graphene up to a requested number of layers, which leads to the electronic interaction between layers being effectively switched off due to turbostratic disorder.

2. Experimental results

In this work, terahertz (THz) nonlinearities of six graphene samples were characterized by intense THz pulses. Monolayer graphene was synthesized by CVD growth on large-area Cu foils. Multilayer graphene samples were fabricated by two different techniques, which one is an as-grown multilayer graphene (AGMG) and another is a randomly stacked multilayer graphene (RSMG). Figure 1 shows representative results for the comparison between AGMGs and RSMGs. For same number of graphene layers, AGMG and RSMG have a different number of active electronic bands where their chemical potentials are analogous as shown in figure 1b. Therefore, their effective sheet conductivity and corresponding nonlinear transmission characteristics are clearly different. These nonlinear properties originate from a thermo-modulational feedback mechanism through ultrafast free-carrier heating and temperature-dependent carrier-phonon collisions. In addition, we will show the open aperture THz z-scan data for same samples to extract THz nonlinear absorption coefficients in the talk.

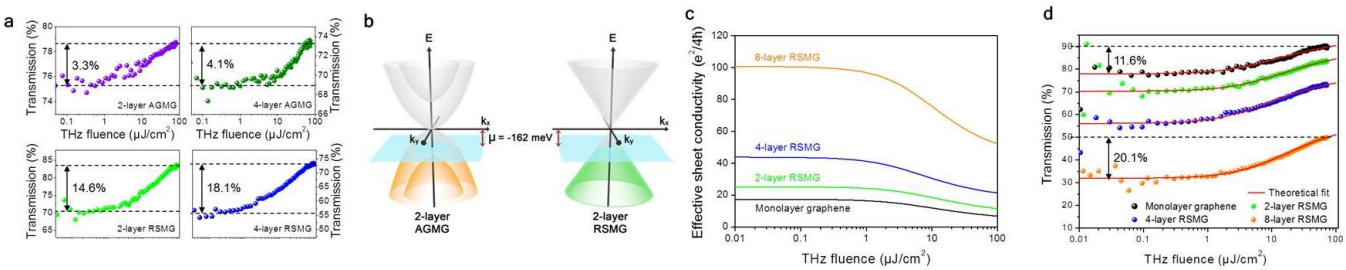

Figure 1. (a) THz nonlinear transmission of RSMG and AGMG films. (b) Schematics of different electronic band structures of 2-layer AGMG and RSMG. (c) Effective sheet conductivity of four RSMGs as a function of incident THz fluence. (d) Experimental results on change in nonlinear THz transmission for monolayer graphene and 2-, 4- and 8-layer RSMGs.

3. Conclusions

The unique nonlinear optical characteristics of RSMGs were systematically investigated in the THz frequency range in comparison with AGMGs. RSMGs can preserve the intrinsic optical properties of monolayer graphene and therefore show controllable nonlinearity with substantial enhancements. These results, including enormously large THz nonlinearity and its easy controllability by the layer-by-layer stacking process, make RSMGs highly suitable candidates for a variety of applications in graphene-based THz nonlinear photonic devices.

4. References

1. A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, "The electronic properties of graphene," *Rev. Mod. Phys*, **81**, 109, 2009.
2. F. Bonaccorso, Z. Sun, T. Hasan, and A. C. Ferrari, "Graphene photonics and optoelectronics," *Nature Photon.*, **4**, 611-622, 2010.