THz absorption in Graphene Quantum Dots

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Abstract— **We study the optical response of multilayer graphene quantum dots at THz frequencies. We fabricate 73 nm-diameter graphene quantum dots in an array of ~1mm^2 size. We demonstrate optical absorbance of these graphene quantum dots from 0.85 to 4.7 THz and study the absorption dependence with the temperature from 4K to 300 K.**

I. INTRODUCTION

The THz spectral range remains one of the least exploited spectral regions, mainly due to the lack of compact powerful THz sources. The development of the typical semiconductor-laser scheme emitting at THz frequencies has been seriously hampered by the absence of an appropriate material with a sufficiently small bandgap. Graphene is potentially an excellent candidate for a THz semiconductor-laser model owing to its 'zero' bandgap and its large optical phonon energy (200meV). However, non-radiative Auger recombination process reduces the lifetime of the optical gain to few hundreds of femtoseconds that drastically limits the feasibility of a THz laser [1]. In this context, graphene quantum dots are extremely attractive since Auger processes could be reduced in this material system by the suppression of final states for Auger carriers [2].

Here, we study the optical response of array of multilayer graphene quantum dots at THz frequencies. We fabricate graphene quantum dots of 73 nm diameter in an array of ~1mm^2 size. We demonstrate optical absorbance of these graphene quantum dots from 0.85 to 4.7 THz. We study the dependence of this absorption with the temperature from 4K to 300 K.

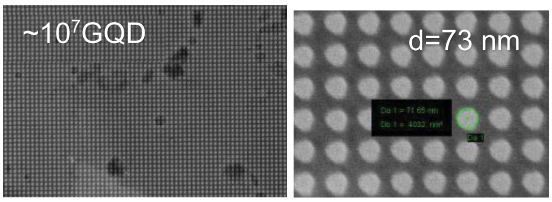

Fig. 1. SEM images of the graphene quantum dot array. The dots of 73 nm diameter are spaced by a distance of 130 nm. The array dimension is ~1mm^2.

A main challenge for investigating optical absorption in quantum dots at THz frequencies is the large THz beam spot limited by the diffraction limit to ~300 μm at 1 THz. It requires processing large array of graphene quantum dots. We fabricate the graphene quantum dots array by nanostructuring multilayer epitaxial graphene (MEG) by using negative electron beam lithography, with HSQ resist acting as hard mask, followed by plasma O_2 etching. Raman spectroscopy is performed in order to optimize the position of the array on the substrate and validate the etching step. The array of ~1mm^2 dimension contains ~10^7 quantum dots. The initial MEG sample is produced by thermal desorption of Si from the C-terminated face of single-crystal 4H-SiC(000-1) [3] and contains four first doped layers near the substrate and a few tens of independent quasi-neutral layers with non-Bernal rotated graphene planes.

II. RESULTS

The graphene quantum dots and a bare SiC substrate used as a reference are mounted in a cryostat with polyethylene windows, allowing to vary the temperature from 4K to 300K by He cooling. Transmission measurements have been performed using THz time-domain spectroscopy experiment based on 15fs optical pulses at central wavelength 800nm. Figure 2 reports the absorbance spectra of the graphene quantum dots with respect to frequency and temperature. We observe that the absorbance spectra strongly differ from the Drude-like response of multilayer 2D graphene [4]. At low temperature, the absorption show large absorption lobes centered at some specific frequencies. These lobes tend to vanish when the temperature is increased, resulting at 300K to a broad absorption up to 3 THz followed by a plateau.

The dependence of the absorption with the temperature clearly indicates that the observed behavior comes from the quasi-neutral layers of the graphene quantum dot. Indeed, the optical response of the doped layers is insensitive to temperature since their Fermi levels are high compared to thermal energy. To interpret these absorbance spectra, we currently calculate the electronic states distribution and the oscillator strengths in these large graphene quantum dots at THz frequencies.

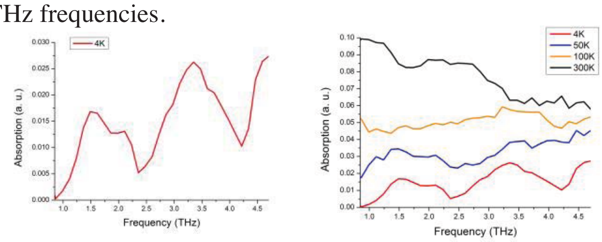

Fig. 2. Transmissions with respect to frequency for different temperatures. *Right :* Curves are slightly vertically shifted for clarity of the figures.

REFERENCES

[1] K. J. Tielrooij *et al.*, Nat. Phys. **9**, 248 (2013)
[2] M. Mittendor *et al.*, Nature Phys. **11**, 75 (2015)
[3] W. A. de Heer *et al.*, Proc. Natl. Acad. Sci. USA **108**, 169000 (2011)
[4] S. Massabeau *et al.*, Phys. Rev. B **95**, 085311 (2017)

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