Energy levels and THz optical properties in Graphene Quantum Dots

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Abstract— **Owing to their energy level splitting in the meV range, large graphene quantum dots (size ~100 nm) are very attractive candidates for THz technology. Whereas their electronic properties have been widely studied by transport measurements, only very few works have been focused on their interaction with THz radiation. Here, we report a theoretical and experimental investigation of the optical properties at THz frequencies of large graphene quantum dots. Using a tight-binding modeling, we show the existence of spatially extended mixed-states that should couple efficiently to THz photons. Furthermore, we experimentally demonstrate THz optical absorption of an array of circular 75 nm-diameter graphene quantum dots at 4K and 300K.**

I. INTRODUCTION

Graphene quantum dots (GQD) of few tens of nanometer size are very attractive candidates for THz technology. For instance, large GDQ possess long relaxation time of electronic excitations (60ns) [1], extensive bolometric effects [2] and a transport gap that prevents from large dark current in THz graphene-based photodetectors [3]. However, the optical properties of large GQD remain elusive due to computing limitations with conventional computers (up to ~10^4 atoms) and technological issues in order to match the large diffraction-limited size of focused THz beam.

Here, we combine theoretical and experimental investigation of the optical properties of large GQD. Our study reveals the existence of mixed states that could couple to THz radiation. We further experimentally demonstrate absorption at THz frequencies of an 1mm^2 size array of GQD.

II. RESULTS

We perform tight-binding modeling on GQD, with diameters ranging from 35 to 50 nm to calculate the discrete energy levels and their associated electronic density distributions. We do not impose any condition on the GQD edges (armchair or zigzag) for a more realistic modeling of our samples.

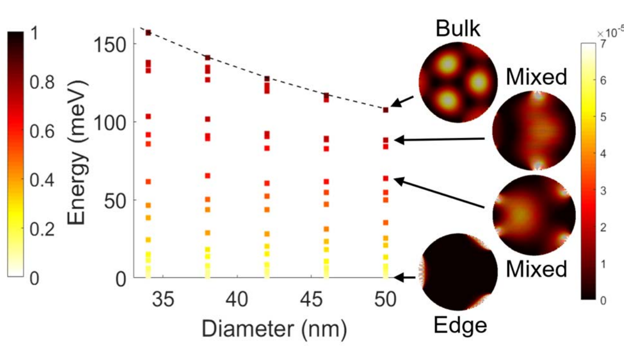

Fig. 1. (Left) GQD spectra as function of diameter. (Right) Probability density for bulk, mixed and edge states.

Figure 1 reports the discrete spectrum as function of the GQD diameter, up to the first expected bulk state (dashed line) [4]. The color code indicates the integrated wavefunction *I* for each state on 85% of the diameter. The expected value for a wavefunction with a uniform density ($U_{85\%}$~0.72) is used as a criterion to identify if a state lies more in the bulk or near the edges. We observe the existence of wavefunctions in the meV range that have a non-negligible spatial extension (*I*~0.66-0.7) in the bulk. These states are different from the expected zero-energy states localized on zigzag edges (*I*~0.02) and have lower energy than the bulk ones. They could arise from the orthogonality between allowed wavefunctions that naturally increases their spatial extension and the existence of armchair edge states, which can survive away from the edges [5]. These mixed-states could couple efficiently to a low frequency (THz) excitation.

For probing optical properties, we fabricate multilayer GQD of quasi-spherical 75 nm diameter in an array of ~1mm^2 size, which contains ~10^7 quantum dots. The GQD array is fabricated by nanostructuring multilayer epitaxial graphene (MEG), using electron beam lithography [3]. We use THz time-domain spectroscopy experiment based on 15fs optical pulses at central wavelength 800nm to measure the transmission of the GQD. Figure 2 reports the transmittance spectra of the GQD with respect to frequency and for 4K and 300 K. These transmittance spectra strongly differ from the Drude-like response of MEG [6] (see Fig. 2 insert). Moreover, the GQD transmittance spectra show opposite dependence with temperature. Calculation of the oscillator strengths between the mixed-states in GQD at THz frequencies are on-going and will be presented to support the interpretation of transmittance spectra.

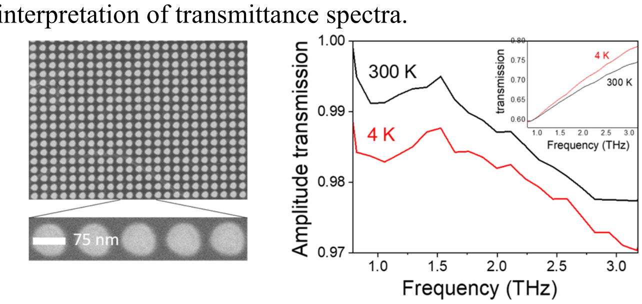

Fig. 2. (Left) SEM image of the GQD array. Dots spacing is 110 nm. (Right) GQD and MEG (insert) transmittance at 4K and 300K.

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