Optical Excitations and Nanoplasmonics in Graphene Flakes and Ribbons

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The fabrication of nanoscale graphene ribbons of finite length is now reaching atomic scale accuracy and extreme edge control [1], and further geometries [2] and substrates are being explored experimentally. Here we present a theoretical investigation of the optical excitations of elongated nanoflakes and ribbons. We focus both on intrinsic field enhancement effects and on the modification of the optical properties by means of edge modulation, functionalization and distortions. We employ either quantum chemistry semi-empirical approaches [3-4] or ab-initio techniques [5], depending on the system type.

We find that the optical spectra of elongated graphene flakes are dominated at low energy by excitations with strong intensity, comprised of characteristic coherent combinations of a few single-particle transitions with comparable weight. They give rise to stationary collective oscillations of the photoexcited carrier density extending throughout the flake, and to a strong dipole and field enhancement. This behavior is robust with respect to width and length variations, thus ensuring tunability in a large frequency range. The implications for nanoantenna and other nanoplasmonic applications are discussed for realistic geometries down to the nm and sub-nm, where atomistic and quantum mechanical effects are found to play a key role.

Our work shows that width modulation and edge functionalization [3-4] can be exploited to design both type-I (straddling) and type-II (staggered) all-graphene nano-junctions. In the first case, we find that minimal width-modulations are sufficient to obtain confinement of both electrons and holes, thus forming optically active quantum dots with unique properties [5]. In the second case, we demonstrate that electron affinities and ionization potentials of GNFs can be tuned to form both types of nanojunction [3]. At variance to type-I, type-II GNJs can display indirect excitations with electron and hole densities localized on opposite sides [4]. The effect of functionalization-induced distortions is also discussed.

References

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