Excitonic absorption spectra in graphene and carbon nanotubes

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In this contribution, we propose a theoretical model for calculation of absorption spectra of graphene and carbon nanotubes (CNTs) including excitonic and exciton-phonon features. Our approach is based on the density-matrix formalism combined with tight-binding wave functions. It allows the description of graphene and CNTs of arbitrary chiral angle including many-particle contributions, such as electron-electron and electron-phonon interaction [1, 2]. We derive and numerically solve microscopic graphene- and CNT-Bloch equations describing the temporal evolution of the occupation and transition probabilities of the electronic states [3]. Here, we present results on absorption spectra including the formation of excitons and phonon-induced side-bands.

The free-particle absorption spectrum of graphene is shown in Fig. a (green line). We find a strong impact on the spectrum when taking into account the Coulomb-induced renormalization of the band structure cp. Fig. a (red line). Furthermore, we observe clear excitonic features in graphene and CNTs in agreement with the theoretical studies using density functional theory and recent experiments [4, 5]. We find a significant exciton formation around the M high-symmetry point in the Brillouin zone of graphene. In the immediate vicinity of the K point, however, the excitonic influence is less significant.

Furthermore, in the case of carbon nanotubes we investigate the transfer of the spectral weight induced by exciton-phonon interaction. Here, our approach is extended by introducing an excitonic basis set allowing us to calculate directly the impact of the exciton-phonon dynamics. We observe the formation of a phonon-side band 200 meV away from the zero-phonon line, which we ascribe to the process of phonon emission, illustrated in Fig. b. With the increasing temperature its spectral intensity growths and an additional phonon-side band appears below the zero-phonon line corresponding to a phonon absorption processes. The strong exciton-phonon coupling also leads to a polaron shift of some tens of meV. We investigate the chirality and diameter dependence of the spectral weight transfer to the phonon-side bands as well as of the polaron shift. The description of exciton-phonon induced side-bands in the spectra of graphene is work in progress.

References

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Figures

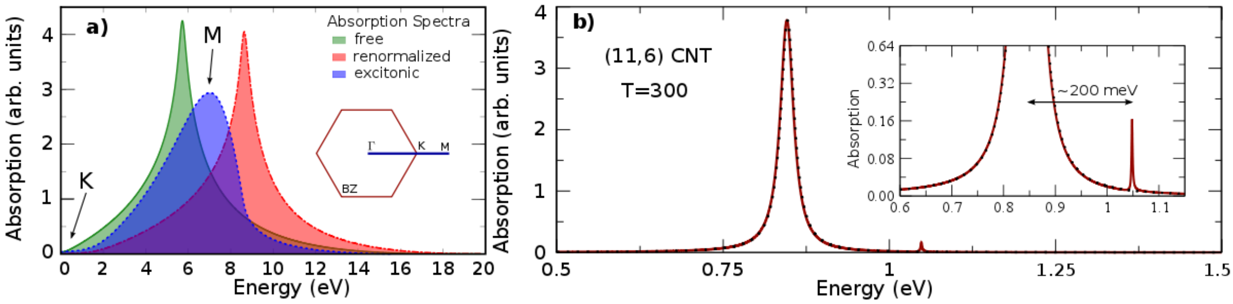

Fig. caption: a) Linear absorption spectra of graphene illustrating the free-particle (green), the renormalized (red), and the excitonic spectrum (blue). The inset figure shows the hexagonal Brillouin zone of graphene within the high symmetry points. b) Excitonic absorption spectrum of the semiconducting (11,6) nanotube including exciton-phonon coupling at room temperature. For comparison, the doted line in the background shows the pure excitonic spectrum. The figure illustrates the formation of a phonon side-band at the higher energy side of the zero-phonon line.