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Evaluation of the Dissertation of Miriam Kosik on Tight-binding framework to study optical properties of graphene nanoantennas with adatoms

The doctoral thesis by Miriam Kosik investigates the optical and plasmonic properties of graphene nanoflakes and in particular introduces a mathematical model of a hybrid system consisting of grapheme nanoflakes coupled to adjacent atoms. Graphene flakes acting as photonic nanoantennas may sustain strong electromagnetic field localization and enhancement, which leads to many interesting phenomena. It is therefore very interesting and timely research project.

The thesis is, besides an introduction to the necessary concepts and motivation, separated into four large sections: Electronic and optical properties of graphene, Interaction of light with atoms, Properties of grapheme nanoflakes, and Properties of graphene nanoflakes with adatoms. The first and the second of the above contain fundamental physical features of graphene and the necessary quantum optics formalism to construct the model. In Chapter 3 the new framework is introduced, to describe the optical and electronic properties of finite graphene flakes. The last Chapter is extended to the case of an atom sitting close to the flakes, close in the sense that embedded in the plasmonic fields of the compound structure. It is followed by Final remarks, with the list of the problems already treated by an introduced framework, both these discussed here and the others not included in the thesis, and perspectives for further extension. In closing, we find a series of Appendices.
To the best of my knowledge, scientific studies Miriam Kosik was a coauthor of 6 publications concerning related subjects, all published in high-impact scientific journals.
Hereby I state that:

1) The doctoral dissertation presents the candidate's general theoretical knowledge in a discipline or disciplines as well as the ability to independently conduct scientific work.

2) The subject of the doctoral dissertation is an original solution to a scientific problem.

Below I describe briefly the content of the Thesis and provide justification for my recommendations.

Introduction:

The thesis starts with a general and very accessible chapter that explains the basis and motivation of the dissertation, which is in my opinion to describe properties and possible applications of the plasmonic system, where single additional atom is located in the vicinity of finite graphene flakes.

Electronic and optical properties of graphene

This Chapter offers a concise introduction to graphene physics, tailored to the original studies which are the essence of the study. It starts by presenting general facts about graphene, and its structure starting from a single carbon atom to the lattice. Next, it discusses briefly the tight-binding model that is used to calculate electronic band structure, including honeycomb graphene lattice. The general form of tight-binding Hamiltonian is given and some details of the formalism leading to famous electronic dispersion graphs, including Dirac points are present. Next, the idea of adding electrons (n-doping, p-doping, and gating) is introduced. They offer reliable control of the type and density of the charge carriers in graphene and allow to open bandgap in single-layer and bilayer structures. It is also a simple and effective way to excite plasmons in this material. In the subsection "Exciting plasmons" this issue is extensively discussed, including a comparison with metal surfaces and 3D structures, introducing the subject of surface plasmon polaritons in graphene, tuning of plasmons energies, and nanoantennas. In the last part of the Chapter, the concept of adatoms is introduced as a means to tailor graphene transport properties, with the focus on adsorbates on graphene nanoflakes., as it is related to the thesis's major research problems.

Interaction of light with atoms

This Chapter contains a summary of the basic physics of atom-light interaction with light beam resonant with atom natural frequencies. In principle, it is a standard introduction to quantum optics, but I would like to mention the subsection about spontaneous emission, which is a bit more advanced since it discusses electromagnetic Green Tensor formalism, which is supposed to be later modified and tailored to the case of graphene and especially for the case of adatom placed near the graphene flake.

Properties of graphene nanoflakes

This chapter is presenting components of the framework, which is later used in modeling graphene nanoflakes, and it is where, by the choice of approximations used, the original contribution of the candidate to the research project starts. In the first section, the energy spectrum of the pristine triangular graphene nanoflakes is discussed and the role of edges is discussed. Flakes with different edges, sizes, and doping are considered. Even though they are of finite size, one can recognize the trace of the band structure. In the section Coulomb interaction in the flake, the author discusses the charge distribution due to the electron-electron interactions. And proposes a self-consistent iterative procedure to find it. Next external electric field is included and it is shown how to calculate its influence on charges. Consequently radiative and dissipative and absorption processes are introduced using more heuristic and density matrix Lindblad formalism.

Questions to this chapter:

1. In the chapter I did not find a discussion of the temperature effects. Is it an interesting issue? Is the system always in equilibrium, for instance when we illuminate it with an external short laser beam? Can we then talk about certain temperature?
2. Page 42. Why is the uniform distribution of the final size flake equilibrium state, atoms at the edge feel different Coulomb potential from different sides?
3. On page 43, details of the interaction potential were not described, only reference to paper 66 was given. Can one summarize the ideas of this approach shortly?

4. If I understand it correctly on page 43 after the construction of the Hamiltonian in the iterative procedure one has to find the eigenstate since the density matrix is necessary for further steps?
5. There is a discussion of having different \gamma_ij coefficients, but how to find them? I understand how to get \gamma from the experiment, but particular values of \gamma_ij are more difficult to entangle from experimental data.
6. Do I understand correctly that finding absorption spectra was a task of the group from the Institute of Theoretical Solid State Physics at Karlsruhe Institute of Technology? In this chapter two methods are described and from what I see when interactions are included spectra change dramatically. What is the value of the non-interacting spectrum?

Properties of graphene nanoflakes with adatoms.

In my opinion, this is the essence of the thesis. It is here that adatom in the form of a two-level system is incorporated into the model framework. . I admit that this is a very ambitious task, since one has to account for charge inhomogeneity which modifies charge distribution and fields created by the flake, and consequently dynamics of the atom (for instance Rabi oscillations and emission) are modified. First, the Hamiltonian of graphene nanoflakes with adatom is defined in the tight-binding approximation. The idea here is that the presence of adatom introduces several additional orbitals with its own eigenenergies and appropriate hoping rates between carbon atoms and adatom (adatom is sitting in a top position, above one particular site, at a certain distance). It is estimated from the scaling considerations. Next energy spectra of nanoflakes with adatoms are evaluated using the same iterative procedure as described above. Next, the case of external illumination is investigated, generalizing considerations from Chapter 3, by adding additional orbitals and interaction of the external wave with an atom in two-level approximation. The last additional component is the induced electromagnetic field, which is a form of linear response.
A different set of issues is related to atom dynamics. Rabi oscillations of the two-level atom are modified since the trace of the atom is the self-consistent hybrid structures are HOMO and LUMO states. They are found in the numerical iterative method and they depend on the coupling coefficients. One must also include the fact that under external illumination, the electric charge in the flake oscillates and generate an additional induced electric field. Spontaneous emission from of adatom is modified since it always depends on the surrounding environment. The obvious example is the radiation of atoms in the cavities. Here this effect is even more dramatic since we are dealing with strong plasmonic fields. Details of the derivation are not given, thanks heavens, but I assume it can be done by evaluating the appropriate Green function. And the last issue is to find absorption spectra, which are important for finding resonances.

Questions to this chapter:

1. In figure 4.4 why do we need to show the left panel?
2. Why nonlinear response in this system can be neglected, and also when we change atomic states, they obviously modify flake charge distribution. Is this second order small?

The thesis is written very clearly with a focus on the most important issues and aspects of the model and calculations. One can easily follow the ideas behind various aspects of model construction and corresponding approximations. After all, such modeling must carry a lot of intuitions and to some extent is subjective but I do not see contradictions or elements that I strongly disagree with. Language-wise dissertation is also very good.

Overall recommendation:

Hereby I state that this work fit to serve as a basis for awarding the degree of „Doctor of Philosophy". In my opinion, the thesis presented by Miriam Kosik, entitled "Tight-binding framework to study optical properties of graphene nanoantennas with adatoms" satisfied all the requirements and I recommend further steps of the procedure.

Based on the arguments presented above, I believe that this is an excellent dissertation. I recommend that this dissertation be recognized with distinction. The primary justification for the distinction is the work in Part IV of the dissertation that paves the way for new approaches to solving physics problems of radiation from graphene plasmonic systems with additional atoms located in the strong plasmonic field region.

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