Ab Initio Modelling of Photoinduced Electron Dynamics in Nanostructures

A Thesis

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Doctor of Philosophy

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by

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Chapter 1

Background and Introduction

1.1 Motivation - Solar Technology

A brief discussion about renewable energy and solar technology in particular. We'll mention the various types of latest generation solar cells with a focus on 2D materials.

1.2 2D Materials in Photonics

Properties of 2D materials that make them useful in photonics with experimental and theoretical results.

1.3 Increasing Light Absorption in 2D Materials

Different methods of increasing light absorption with attention towards plasmonic field enhancement.

Chapter 2

Theory

2.1 Background Theory

2.1.1 Electronic Structure Calculations

Brief discussion of various methods for calculating electronic structure, focussing on Kohn-Sham DFT.

2.1.2 Excited State Calculations

Brief discussion of TDDFT, linear response and the (time-dependent) Bethe-Salpeter Equation.

2.1.3 Obtaining Optical Properties

A bit about dielectric functions and absorption spectra and how they are obtained from excited state calculations.

2.2 Projected Equations of Motion Method for Composite Materials

This will include a little about obtaining the polarization from classical electrodynamics. The dipole-dipole model will be presented and the PEOM method will be derived.

Chapter 3

Results

Here we use the SQD-MNP system to validate the method as well as providing new results for femtosecond pulse interaction. The method is further verified by performing calculations on various monolayer systems using TDDFT and BSE theory. We'll then go on to model a monolayer of MoS_2 with gold nanoparticles.

3.1 Semiconducting Quantum Dot-Metal Nanoparticle Hybrid

3.1.1 Describing the System

3.1.2 Energy Absorption Rates

3.1.3 Population Inversion

3.2 Optical Spectra for MoS_2 Composites

3.2.1 MoS_2 Monolayer

3.2.2 MoS_2 / Graphene Monolayers

3.2.3 MoS_2 Bilayer

3.2.4 MoS_2 Monolayer on Gold Surface

3.2.5 MoS_2 Monolayer with Gold Nanoparticles

Chapter 4

Conclusions

We will have shown that the PEOM method provides a way for modelling the plasmonic field enhancement from gold nanoparticles on a MoS_2 monolayer. Such theoretical calculations have not been performed before.

Bibliography