

Ab Initio Calculation of Linear and Nonlinear Optical Properties of Semiconductor Structures

F. Bechstedt, B. Adolph and W. G. Schmidt*

Friedrich-Schiller-Universität, Max-Wien-Platz 1, 07743 Jena, Germany

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The theoretical and numerical approaches are discussed for *ab initio* calculations of optical properties. The density-functional theory (DFT) combined with the local-density approximation (LDA) allows the calculation of the geometry of crystalline solids and their surfaces with a precision of about one percent. The DFT-LDA band structure and single-electron states therefore provide a reasonable starting point for the calculation of linear and nonlinear susceptibilities within the independent-particle approximation. However, this approach has to be improved by taking into account many-body interactions: self-energy effects, local-field corrections, and electron-hole attraction. Three types of optical spectra are studied: the frequency-dependent dielectric function, the second-harmonic generation, and surface reflectance anisotropy spectra. The systems considered are two-atomic semiconductors, their polytypes and their surfaces.

I Introduction

The recent major advances in the quantitative computation of ground-state properties of solids are essentially related to the development of density-functional theory (DFT) in local-density approximation (LDA) [1]. However, spectroscopic properties, as optical spectra, are not accessible in this theory, since its single-particle eigenvalues or differences of them cannot be directly identified with electronic excitation energies. For instance, energy gaps of semiconductors and insulators are remarkably underestimated within DFT-LDA [2]. Instead, quasiparticle (QP) energies have to be computed. The basic approach to quasiparticles is Hedin's GW approximation [3], in which the spatially nonlocal and energy-dependent exchange-correlation (XC) self-energy Σ is approximated by a convolution of single-particle Green function G and dynamically screened Coulomb interaction W . Vertex corrections are neglected. Full numerical calculations of the single-particle excitations can now be performed on real crystals starting from the DFT-LDA results [4,5].

Despite of the complications with the inclusion of the many-body effects, DFT-LDA has been used to determine the atomic structure and to define a reasonable starting point for the description of the electronic structure. Optical spectra have been calculated [6] within the independent-particle approximation [7]. Meanwhile, quasiparticle shifts have been included also

in calculations of the complete dielectric function [6,8]. Moreover, the effects of XC beyond the independent-particle approximation have been demonstrated within DFT-LDA for bulk semiconductors, and local-field effects are taken into account [9]. Very recently the compensation effect of the dynamical screening in quasiparticle and vertex corrections has been found [10]. Furthermore, the Coulomb attraction of excited electrons and holes has been indeed calculated in the limit of static screening [11].

In the last years such kind of *ab initio* calculations in the framework of DFT-LDA and the independent-particle approximation has been also applied to nonlinear optical properties of semiconductors, e.g. the second-harmonic generation (SHG) [12,13], and to semiconductor surfaces [14-16]. The surface activities have been focused on the reflectance anisotropy (RA) spectra [17]. The inclusion of the many-body self-energy and excitonic effects is an unsolved problem. However, at present many activities occur in this field. They mainly concern the inclusion of the QP shifts.

After a brief description of the calculational methods and basic expressions, the status of the *ab initio* description of the optical properties is demonstrated for three examples, the frequency-dependent dielectric function, the second-harmonic generation and the surface reflectance anisotropy. The semiconducting materials considered are the group-IV semiconductor Si as

*Present address: Department of Physics, North Carolina State University, Raleigh, U.S.A.

Nonlinear Optical Diagnostics Of Laserexcited Semiconductor Surfaces

**Sergej Akhmanov, Vladimir Il'ich
Emel'janov, Nikolaj Koroteev**

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