THE SUMMARY OF Ph. D. DISSERTATION

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Title

Modeling of condensed-matter systems by the tight-binding method:
from nanostructures to complex liquids
(和文題名: 強結合法による凝縮系のモデリング: ナノ構造から複雑液体まで)


Abstract

The present thesis focuses on the development and applications of the tight-binding (TB) and TB molecular dynamics (MD) methods. In particular, emphasis is put upon applications to real systems. We improve the transferability of tight-binding models, and further implement order-$N$ tight-binding. We use these methods to analyze the properties of nanostructures, complex liquids, and associated amorphous solids.

In chapter 1, we give a general introduction for the present thesis. We study various methods and approximations used to simulate condensed-matter systems by computers, and describe the role of TB methods. Then, we give an overview of the entire thesis.

Chapters 2 and 3 are devoted to the description of TB methods and TBMD methods. In chapter 2, we discuss on the basic formulation of the TB methods, along with a brief review of MD simulations. Then, we describe the way in which we integrate TB methods into MD simulations. In chapter 3, we focus on order-$N$ tight-binding. When applied in a straight-forward manner, calculational costs of TB methods scale as order-$N^3$. We show that, by introducing suitable approximations, it is possible to improve the scaling behavior to order-$N$. In particular, the case of non-orthogonal TB (NTB) and applications to MD simulations are taken into account. We perform realistic test MD simulations for germanium (Ge) by the obtained order-$N$ method. The results obtained show, for the first time, that order-$N$ NTB is in fact applicable to MD simulations.

In chapters 4 and 5, we focus on the study of photoluminescence properties for silicon (Si) nanostructures. Silicon nanostructures, unlike bulk Si, are known to show efficient photoluminescence at room temperature. We introduce new structural models for Si nanostructures, and study their electronic states and optical properties. In chapter 4, our new model, “Si nanostructures devoid of point-group symmetry”, is analyzed by the TB method. The results show that our new model well accounts for the discrepancy found between theoretical calculations and experiments in the behavior of the radiative recombination time. In chapter 5, we introduce another new model, where structural

relaxation of “poorly-passivated” Si nanostructures are taken into account. The effects of structural relaxation for “well-passivated” Si nanostructures have been studied in the past, while those for the “poorly-passivated” Si nanostructures are investigated for the first time in the present work. From our analyses based on the TB method, we find that the behaviors of the so-called ‘F’-band luminescence are well described by this model.

We further study the static and dynamical structure of liquid (*l*-) and amorphous (*a*-) Ge in chapters 6 and 7. In particular, we thoroughly study properties at high density. In chapter 6, we construct a new, transferable NTBMD scheme for Ge, and apply the method to the study of *l*-Ge. We perform NTBMD simulations at low to high density, and calculate static and dynamical structures at each density. We calculate quantities inaccessible by experiments, such as the distribution of bond angles, and deduce the change in the local structure of *l*-Ge in detail. From our analyses, we clearly observe that, with density increase, random configurations characteristic of liquids increase compared to configurations originating from covalent bonds, while the remaining covalent bonds become close to those of the $\beta$-Tin structure. In chapter 7, TBMD simulations on *a*-Ge are performed. We use the order-*N*, NTBMD scheme constructed in the present work. Firstly, we perform glass-transition simulation of Ge. We start from a liquid well above melting point, and quench it to an amorphous solid. The static and dynamical structures of the liquid, super-cooled liquid, and amorphous Ge are extensively studied. We conclude that the large structural change which occurs during the glass transition is addressed to the increase of the covalent bonds. We further increase the density of *a*-Ge obtained in order to study the structural change of *a*-Ge with density increase. At each density simulated, we carefully analyze the static and dynamical structures of *a*-Ge. Our results show that the local structure of *a*-Ge transforms from tetrahedral at low-density to $\beta$-Tin-like at high density. We also find that, at intermediate density, both low-density and high-density amorphous structures coexist. From our work, a comprehensive knowledge concerning liquid and amorphous Ge at low to high density is obtained for the first time.

Finally, we conclude the present thesis in chapter 8. We summarize the achievements obtained in our work, and discuss their consequences. We also comment on work left to be performed in future studies.