論 文 の 内 容 の 要 旨

論文題目　Quantitative Calculation of Superconducting Transition Temperatures by Density Functional Theory: Method Development and Its Applications
（密度汎関数理論による超伝導転移温度の定量的計算：手法開発とその応用）

氏　　名　明石　遼介

Superconducting transition temperature (T_c) is one of the key quantities of superconductors. Despite the substantial progress in general understanding of the mechanisms of superconducting transition up to date, the accurate quantitative calculation of T_c has long been an unprecedented challenge. This is due to the fundamental difficulty in calculating T_c: Since the interactions contributing to the mechanisms substantially vary depending on the electronic and lattice structure of the material, an accurate quantitative treatment must be carried out on top of the understanding of the mechanisms.

So far, for several conventional phonon-induced superconductors, accurate T_c calculation is established by the recent progress in the density functional theory for superconductors (SCDFT). However, the applicability of the scheme to various superconductors is quite unclear because the examples of the applications are still few. Furthermore, from a theoretical point of view, this scheme should work well only for a limited range of materials because of the following reasons. First, it has been constructed referring to the standard theory of phonon-mediated strong-coupling superconductivity, the Migdal-Eliashberg (ME) theory, and therefore it is not capable of treating other superconducting mechanisms. Second, its formulation includes non-trivial approximations, with which one cannot consider several effects contained in the ME

theory. In order to enable us to carry out accurate T_c calculations for a wider range of materials, further applications and extensions of the current SCDFT are essential.

In this thesis, we study superconductivity in real materials with particular focus on how their T_cs are accurately reproduced by the calculations based on the SCDFT. The structure of the thesis is as follows. We first point out several superconductors whose T_c observed in experiments cannot be accurately reproduced with the SCDFT-based scheme. Next we extend this scheme so that the effects of the following factors can be treated: The particule-hole asymmetric electronic structure and a plasmon-induced superconducting mechanism. Through applications of the extended scheme, we show that it fills the gap between the calculated and experimentally observed T_cs.

We start from a review on how accurate the current scheme is and demonstrate several examples where it yields inaccurate T_cs. There are series of superconductors where the phonon-mediated pairing interaction is considered rather strong but it is unclear whether the phonon mechanism is enough to reproduce experimentally observed T_cs. As representatives, we apply the SCDFT-based scheme to lithium under extreme pressures (T_c: 5—20K), layered nitrides (T_c: 10—26K), and alkali-doped fullerene solids (T_c: 19—40K), showing that the resulting values of T_c are generally less than a half of experimental ones. The present result suggests that the current formulation is omitting some important factors essential to achieve accurate calculations for more general systems.

Next, we consider a factor missing in the current formulation for the conventional phonon mechanism, the particle-hole antisymmetric component in the electronic structure. In that formulation, the electronic structure is assumed to be symmetric in energy with respect to the Fermi level. While this approximation has been introduced to avoid numerical instabilities, we construct a stable formalism without the symmetry assumption. We also demonstrate a model calculations where the asymmetry effects on T_c becomes substantial.

Finally, based on the SCDFT, we formulate an extended scheme including a plasmon-induced superconducting mechanism. The plasmon mechanism exploits the frequency dependence of the screened Coulomb interaction between electrons, and this mechanism favors s-wave pairing. Consequently, it can cooperate with the conventional phonon mechanism. We construct a scheme to treat the screened dynamical Coulomb interaction within the random-phase approximation, which enables us to discuss the effects of the phonon and the plasmon mechanisms on equal footing. We apply this scheme to lithium under pressures and the

layered nitrides and find that the plasmon mechanism substantially enhances the calculated T_c by the cooperation. We also find that the agreement of the calculated and experimentally observed values of T_c becomes better, which indicates that the assistance by the plasmon mechanism is fundamentally important in these systems.

The present results indicate that our new scheme opens the door to accurate calculations of T_c for a wider range of materials than the previously proposed one. Our achievement can be a first step toward the theoretical materials design of novel superconductors.