電子相関の強い系に対する第一原理運動量依存局所変分埋論：鉄の電子状態への応用

Form 3

Abstract

Title: First-Principles Theory of Momentum Dependent Local Ansatz to Correlated Electron Systems – Application to Iron –

電子相関の強い系に対する第一原理運動量依存局所変分理論 – 鉄の電子状態への応用 –

We have developed the first-principles momentum-dependent local ansatz (MLA) wavefunction method on the basis of the tight-binding LDA+U Hamiltonian in order to describe quantitatively electron correlations in the real system. The MLA wavefunction expands the Hilbert space for correlated electrons by applying the intra-orbital correlators, the inter-orbital charge-charge correlators, and the inter-orbital spin-spin correlators with momentum-dependent variational parameters to the Hartree-Fock uncorrelated state, so that it reduces to the correct Rayleigh-Schrödinger perturbation theory in the weak correlation limit and can describe quantitatively the ground state as well as related low-energy excitations in solids.

We first performed the lowest-order numerical calculations for paramagnetic bcc Fe in order to clarify the basic behavior of the theory. We found that the MLA yields a reasonable correlation energy gain, and the inter-orbital correlations as well as the intra-orbital correlations plays an important role in the energy gain. The charge fluctuation is suppressed rapidly with increasing the Coulomb interaction strength. Calculated charge fluctuation for Fe is found to be comparable to the result of the local ansatz (LA) in the d-band model. The amplitude of local moment is smaller than the d-band LA value. We also found the strong momentum dependence of the momentum distribution function (MDF), and obtained the mass enhancements m^*/m = 1.4 for Fe from the jump of the MDF at the Fermi level.

We next performed the self-consistent calculations using the new ansatz for the variational parameters which interpolates between the weak and the atomic limit. We obtained the correlation energy −0.076 Ry for bcc Fe, the charge fluctuations for d electrons $\langle(\delta n_d)^2\rangle$= 1.51, and the amplitude of local moment $\langle S^2\rangle$ = 2.61. The latter is in good agreement with the experimental value 2.58. We find that the inter-orbital charge-charge correlations between d electrons make a significant contribution to the correlation energy and charge fluctuations, while the intra-orbital and inter-orbital spin-spin correlations make a dominant contribution to the amplitude of local moment.

We also obtained the MDF along high-symmetry lines for Fe. We found that the MDF for d electrons with e_g symmetry show a large deviation from the Fermi-Dirac function. The analyses of projected MDF indicate that the s and p electrons behave as independent electrons, while d electrons behave as correlated electrons. The average mass enhancement factor m^*/m is found to be governed by the intra-orbital and inter-orbital spin-spin correlations. Calculated m^*/m = 1.65 is shown to be consistent with the experimental data and agrees well with the recent results of theoretical calculations.

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