Development of ab initio many-body perturbation calculation software RESPACK and its applications to Ta_2NiS_5 and Ta_2NiSe_5

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As progress of this year, I have released a new version of the software RESPACK [1] for many-body perturbation calculation and effective low-energy model derivation. RESPACK is possible to calculate the maximally localized Wannier function, response function with random phase approximation and related optical properties, and frequency-dependent electronic interaction parameters, etc, and the new version includes the GW calculation. RESPACK supports *ab initio* band calculation codes using norm conserving pseudopotentials plus plane wave basis set, and officially supports xTAPP [2] and Quantum ESPRESSO [3] packages. An automatic generation script from these band-calculation output to input files for RESPACK is prepared. An input file for specifying calculation conditions is designed pursuing simplicity and is given in a namelist format. RESPACK has a wide application including simple metals, semiconductors, $3d/4d$ transition-metal compounds, organic and aromatic compounds, etc. It supports OpenMP/MPI and intel/GNU compiler environments. The new version also includes utility such as interfaces to model-analysis solvers mVMC [4] and $\mathcal{H}\Phi$ [5]; users can automatically obtain these inputs via RESPACK.

In this report, as a RESPACK application, I present results for an *ab initio* GW calculation of transition-metal chalcogenides Ta_2NiS_5 and Ta_2NiSe_5 which are actively studied as a possible candidate of excitonic insulators. Using an *ab initio* GW calculation, we studied low-energy electronic structures, especially for the band gap. These materials have a layered structure stacked loosely by a weak van der Waals interaction, and in each layer, Ni single chains and Ta double chains are running along the a axis of the lattice to form a quasi-onedimensional (1D) chain structure. The observed resistivity shows a semiconducting behavior over a wide temperature range with a quasi-1D anisotropic electron conduction at high temperatures. The experimental band gaps at room temperature are estimated as nearly 0.13 and 0.36 eV for the sulfide and selenide, respectively [6]. The crystal structure is body-centered orthorhombic (a=3.415 $\mathring{A}$, b=12.146 $\mathring{A}$, c=15.097 $\mathring{A}$ for Ta_2NiS_5 and a=3.496 $\mathring{A}$, b=15.829 $\mathring{A}$, c=15.641 $\mathring{A}$ for Ta_2NiSe_5) and contains 16 atoms in the body-centered unit cell. We calculate density functional band structure, maximally localized Wannier functions, dielectric properties, and GW spectral functions to understand the low-energy properties of these compounds.

Density-functional calculations were performed with xTAPP with plane-wave basis sets, where we employed norm-conserving pseudopotentials and the generalized gradient approximation (GGA) for the exchange-correlation potential. Maximally localized Wannier functions were used for the interpola-

tion of the self-energy. The experimental structure obtained by an X-ray measurement at room temperature was adopted in the calculations [6]. The cutoff energies in the wave function and the charge densities are 196 and 784 Ry, respectively, and a 7×7×3 k-point sampling was employed. The cutoff for the polarization function was set to 4 Ry, and 100 bands were considered. Nickel pseudopotential was constructed under the semicore configuration $(3s)^2(3p)^6(3d)^9$ by employing the cutoff radius 0.8 bohr. It should be noted that in the GW calculation of $3d$ transition-metal compounds, it is very important in quantitative accuracy to use a pseudopotential parametrized for the semicore configuration.

Figure 1(a) compare the calculated GW spectral function with the density-functional GGA bands (white solid curve) of Ta_2NiS_5. The GW spectra give clear gap in contrast to the GGA ones being the metallic band structure. We also show in Fig. 1(b) the comparison of the GW (green curve) with GGA (red curve) density of states. The GW gap is estimated as nearly 2 eV.

Figure 2 displays the results of Ta_2NiSe_5. The view of the figure is the same as that of Fig. 1. Our calculated GGA band structure is in a good agreement with the past results [7]. The GW band gap is near 1 eV and appreciablly smaller than the sulfide. In the case of insulators, it would be necessary to solve the Bethe-Salpeter equation for a more quantitative argument for the band gap, which is an important issue for the future study.

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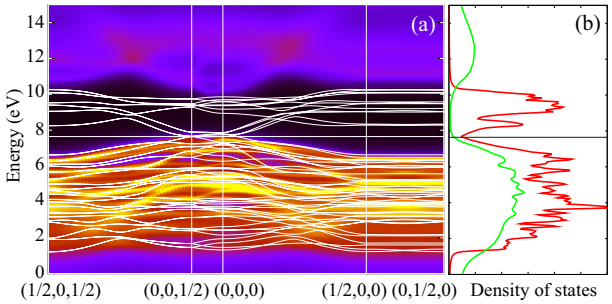

Figure 1: (Color online) (a) Calculated GW spectral function of Ta_2NiS_5, where the GGA bands are superposed with white solid curves. (b) Comparison between the GW (green) and GGA (red) density of states.

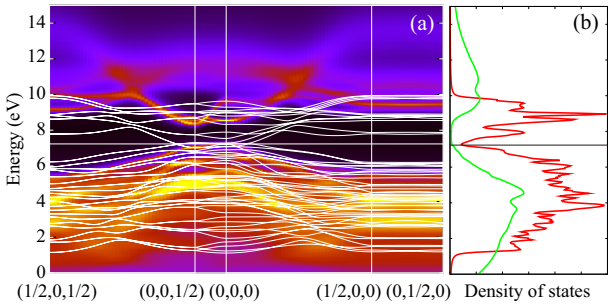

Figure 2: (Color online) (a) Calculated GW spectral function of Ta_2NiSe_5, where the GGA bands are superposed with white solid curves. (b) Comparison between the GW (green) and GGA (red) density of states.

References

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