Ab initio phonon calculation for $Ca_5Ir_3O_{12}$

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The title compound has recently attracted attention as a thermoelectric material because it has a relatively high Seebeck coefficient. In this study, we report an ab initio study about electronic and phononic properties for $Ca_5Ir_3O_{12}$. In particular, we investigated the effect of spin-orbit interaction (SOI) on the low-energy properties [1].

Density functional calculations with plane-wave basis sets were performed using the xTAPP code [2], where the ultrasoft pseudopotential and the generalized gradient approximation (GGA) of the exchange correlation potential were employed. The cutoff energies in the wavefunction and charge densities were 64 and 900 Ry, respectively, and the SOI was explicitly considered. To study the effects of SOI, we performed the usual GGA calculation and compared it with the result including the SOI. Below, we refer to the former as GGA and to the latter as SO-GGA. The atomic and lattice parameters were optimized with an 8×8×8 k-point sampling, and we found that SO-GGA reproduces the experimental crystal structure quite well. The Fermi-surface calculations were performed with the dense 21×21×63 k-point sampling to obtain the detailed surface structure [3]. Phonon calculations with a frozen phonon approximation were performed using the PHONOPY for 1×1×3 and 2×2×1 supercells [5, 6]. For the k-point sampling, 11×11×11 was used for 1×1×3 supercell calculation, and somewhat coarse 9×9×5 was used for the 2×2×1 supercell calculation.

Figure 1(a) shows our calculated band structure. To see the SOI effect, the SO-GGA band

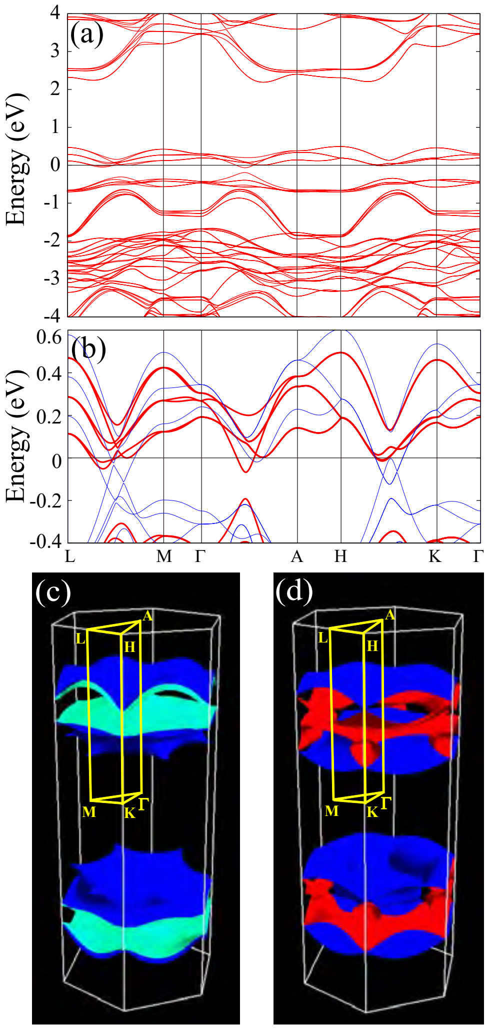

Figure 1: (Color online) (a) Ab initio electronic band structure considering the SOI in $Ca_5Ir_3O_{12}$. The energy zero is the Fermi level. (b) A zoom of the low-energy band structure, where the SO-GGA result (thick red curves) is compared with the usual GGA result (thin blue curve). (c) Calculated Fermi surface for SO-GGA and (d) GGA.

(thick red curves) is compared with the GGA band (thin blue curves) in Fig. 1(b). An appreciable difference can be observed in the low-energy bands; the GGA result exhibits metallic bands, particularly along the L-M or H-K lines. When the SOI is switched on, the metallic bands are split and a pocket-like band structure appears. The gap size due to the SOI is about 0.3 eV, which is comparable to the valence bandwidth 0.5 eV.

Figures 1(c) and 1(d) show the Fermi surfaces based on the SO-GGA and GGA, respectively. We see that the GGA Fermi surface is contributed from the two bands (indicated in dark-blue and bright-red colors), while the SO-GGA Fermi surface is basically formed by the one band; the SOI makes the bright-red colored GGA Fermi surface disappear. In the SO-GGA Fermi surface, since the SOI resolves the band degeneracy, the Fermi surfaces are seemingly two (dark-blue and bright-blue colored surfaces); however, these two are originated from the same band. Also, in the SO-GGA Fermi surface, we see a sheet structure along the c*-axis (the Γ-A line), which indicates a nesting trend along this direction. We note that the SOI is relevant to the narrowing of the sheet separation between the blue colored Fermi surfaces.

Figures 2(a) and 2(b) are our calculated phonon dispersions with SOI for 1×1×3 and 2×2×1 supercells, respectively. It was found that the experimental results at the room temperature are better reproduced by the 1×1×3 supercell calculation. The panel (b) is a preliminary phonon calculation on a 2×2×1 supercell. This supercell includes the displacement degree of freedom in an in-plane 2×2 superlattice, and we can investigate the phonon instability within this superlattice. On the other hand, the 1×1×3 supercell can clarify the triple-period phonon instability along the *c*-axis. Within these two supercells, we observe phonon softening in the 2×2×1 supercell calculation; there is a sign of softening at the K and M points. However, since the calculation conditions for the 2×2×1 calculation are rough, it is necessary to perform the calculation under more severe conditions.

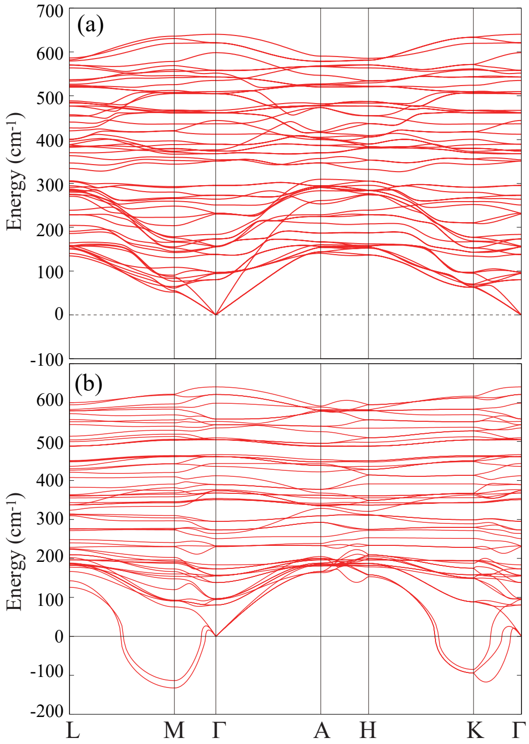

Figure 2: (Color online) Ab initio phonon dispersion with the SOI in $Ca_5Ir_3O_{12}$. Panels (a) and (b) show results based on 1×1×3 and 2×2×1 superlattice, respectively.

References

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