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Origin of pseudo gap and thermoelectric signatures of semimetallic Ru_2TaGa: Structural stability from phonon dynamics, mechanical, and thermodynamic predictions

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ABSTRACT

We have studied the origin of the semimetallic pseudo gap and the role of site preference in the L_21-type Heusler compound Ru_2TaGa. A detailed account of the structural stability, phonon-dependent properties, electronic structure, and thermoelectric properties of the host compound is presented herein. Stability assessments have been made on the basis of phonon band structure, formation energy, mechanical criteria, and hull energy calculations. From the calculated density of states (DOS) and band structure, a semimetallic scenario is evident, with the Fermi level trapped in the pseudo gap. HSE calculations provide a decisive picture, with an indirect overlap between electron and hole pockets near the Fermi level of Ru_2TaGa being responsible for the formation of a negative or pseudo gap. Comparison of the Ru e_g and Ta t_{2g} states reveals the role of d-d hybridization in pseudo gap formation in Ru_2TaGa-like compounds. Additionally, we endeavor to discuss the transport coefficients of Ru_2TaGa. The room temperature Seebeck coefficient calculated for Ru_2TaGa is 66.42 μV/K, considerably larger than those of other semimetallic compounds of the same family. Considering the lattice thermal conductivity ($\kappa_L = 2.5$ W/mK) at 800 K, we obtain a significant figure of merit of $ZT = 0.025$ in Ru_2TaGa. Although the absolute thermoelectric figure of merit (0.002) is relatively low in Ru_2TaGa, it is still the highest among the non-doped full Heuslers of the same family hitherto reported.

1. Introduction

Heusler materials have decisively topped the list of materials for smart devices and spintronics due to their magnificent properties ranging from half-metallic ferromagnetism to superconductivity [1–3]. The exhibition of remarkable transport and magneto-electronic properties by materials of this kind has attracted the attention of researchers since their discovery by F. Heusler in 1903 [4]. Heuslers of the type X_2YZ (where X and Y are transition metals; Z is a p-block element) generally crystallize in an L_21 structure (space group: *Fm*-3*m*) or an inverse X-type structure (space group; *F*-43*m*). The atomic coordinates in the former case are X (0.75, 0.75, 0.75) and (0.25, 0.25, 0.25), with Y and Z at the (0, 0, 0) and (0.5, 0.5, 0.5) positions, respectively. However, in the X-type structure, the X atoms are located at the (0, 0, 0) and (0.75, 0.75, 0.75) positions, the Y atoms at (0.5, 0.5, 0.5), and the Z atoms at (0.25, 0.25, 0.25) [5,6]. This class comprises semiconductors, half-metals, semimetals (spin-gapless semiconductors, Dirac or Weyl semimetals, spin semimetals), ferromagnets, ferrimagnets, antiferromagnets, half-metallic ferromagnets, and so on [7–11]. Non-magnetism in Heusler semiconductors or semimetals is best described by the Slater–Pauling rule ($M = N_v - 24$ or 28), which relates the valence electron count (N_v) with magnetic moment (M). Relevant systems include Fe_2TaX (X = Al, Ga, In) [12], Fe_2VZ (Z = Al, Ga) [13,14], Fe_2NbGa, Fe_2HfSi, and Fe_2TiSn [15], which have proved to be promising candidates for efficient thermoelectric designs.

Recently, Ru-based Ru_2NbAl has been reported as a ferromagnetically correlated and clustered semimetal with a pseudo gap at the Fermi level [16,17]. The 24-valence electron nonmagnetic semimetal Ru_2NbGa has also been synthesized with similar characteristics, and has been confirmed as having a stable cubic L_21 structure by X-ray analysis [18]. Temperature-dependent electrical resistivity measurements on the experimentally reported Ru_2TaAl have indicated semiconducting

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