Equivocal role of relativistic effects in thermoelectric conversion. Theoretical study of Mg_2X (X = Si, Ge, Sn) compounds

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A theoretical study of the influence of relativistic effects on the electronic band structure and thermopower in Mg_2X (X = Si, Ge, Sn) semiconductors using the full potential Korringa-Kohn-Rostoker (KKR) method [1-2] is presented. A detailed comparison between the fully relativistic and semirelativistic electronic structure features allows to elucidate that the spin-orbit (SO) interaction splits the valence bands at specific points of the Brilloiun zone, which remains in excellent agreement with the experimental data. As expected this effect strongly depends on X atom and the SO modifies the topology of the hole-like Fermi surface pockets, which leads to a change in electron transport properties. The thermoelectric properties are investigated using the Boltzmann approach, which has been recently implemented to the KKR and KKR-CPA methods [3].

It is found that the SO coupling of the valence bands reduces the effective mass and therefore signifcantly lowers the thermopower, primarily in Mg_2Sn, but also in Mg_2Ge. A detrimental influence of the SO interaction on the thermoelectric performance of $p-$type Mg_2X is analyzed as a function of temperature and carrier concentration.

Interestingly, similar calculations, e.g. combining electronic structure calculations and Boltzmann transport theory, in $n-$type Mg_2X, show a negligible effect of the SO interaction on the lowest conduction bands, and consequently, also on the Seebeck of Mg_2X semiconductors.

It is worth noting that the $Mg_2Si_{1-x}Sn_x$ alloys have recently attracted a particular interest due to the so-called convergence of conduction bands near the Fermi level, which appears near the $x \sim 0.7$ composition and significantly enhances thermoelectric efficiency of these materials [4,5]. So, it seems really important to evidence that the relativistic effects do not destroy this subtle feature of electronic structure and should not decrease the thermoelectric properties of $n-$type Mg_2(Si-Sn-Ge) materials.

Paradoxally, the $n-$type thermopower at lower carrier concentrations benefits from the degradation of the $p-$type Seebeck coefficient due to the reduction of the bipolar effect, which is well seen in Mg_2Ge and Mg_2Sn compounds [6].

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