Impact du magnétisme sur les propriétés thermoélectriques des thiospinelles

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Abstract

The study focused on the analysis of the couplings between magnetoresistance (MR) and magneto-Seebeck thermopower (MTEP) in three chalcogenide-based compounds: $FeCr_2S_4$, $CuCr_2S_4$ and $Fe_{0.52}In_{2.32}S_4$. These materials were selected for their interesting magnetic and transport properties, particularly in the presence of a magnetic field. The $FeCr_2S_4$ compound, ferrimagnetic with a magnetic transition at 176 K, exhibits a significant negative magnetoresistance (MR ≈ -20% at TN). On top of this MR, a notable and positive MTEP, which is not directly associated with entropy loss under magnetic field, but rather with band structure modification effects has been observed. By applying Mott's formula, a clear relationship between MR and MTEP was established, similar to the one observed in metallic systems such as metallic multilayers or nanowires. An important feature was the increase in power factor (PF) under magnetic field conditions, with an improvement of over 80% around the magnetic transition temperature. As for the ferromagnetic $CuCr_2S_4$ compound, it shows a transition at around 375 K, but no significant change in resistivity or Seebeck coefficient was observed at the Curie temperature. Magnetoresistance is low, and transport properties follow classic metallic behavior. However, a negative MTEP was recorded below 100 K, with a value of -25 to -30%, which could be linked to a transition to a non-collinear magnetic state, recently reported in the literature. This preliminary result suggests that specific entropic effects could be at the origin of the negative MTEP. Finally, $Fe_{0.52}In_{2.32}S_4$ exhibits a spin-glass-like behavior, with a low-temperature transition (Tf ≈ 4.5 K). This semiconductor material shows a maximum resistivity at 5 K, which drops rapidly at room temperature. The Seebeck coefficient shows saturation at around 200 K, and then decreases with increasing carrier ratio. At lower temperatures, it follows a typical Variable Range Hopping behavior. A significant positive magnetoresistance (+67% under 9 T at 5 K) is observed below 50 K, accompanied by a large and positive MTEP. As with $FeCr_2S_4$, a close relationship between MR and MTEP was identified, validating Mott's model. The results obtained for these three materials highlight interesting couplings between MR and MTEP in these thiospinels.