Effect of the density of structural vacancies on the thermoelectric properties of Cu-Ga-Te ternary compounds

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Thermoelectric (TE) materials can convert waste heat into electrical power, which is an effective way to reduce greenhouse gas emissions and contribute substantially to future power supply and sustainable energy management. The main obstacle to the widespread use of TEs in diverse industries is the low efficiency of materials in converting heat to electricity. The efficiency of a material used in TE devices is determined by the dimensionless figure of merit, $ZT = S^2T\rho^{-1}\kappa^{-1}$, where S is the Seebeck coefficient, T is the absolute temperature, ρ is the electrical resistivity, and κ is the total thermal conductivity ($\kappa = \kappa_{lat}+\kappa_{el}$, where κ_{lat} and κ_{el} are the lattice and electronic contributions, respectively). Since the S, ρ, and κ_{el} in bulk materials are interrelated, it is very difficult to control them independently. Therefore, the reduction of κ_{lat} is essential to enhancing ZT. The ZT value of the materials currently used in commercial cooling devices is still limited to about 1 or less over the entire operating temperature range, corresponding to a device efficiency of several percent.

Our group has focused attention on the reduction of the κ_{lat} by manipulating the spatial distribution and density of structural vacancies. In the Cu-Ga-Te ternary system, there exist various compounds with zinc-blende or chalcopyrite structure containing structural vacancies, viz., $Cu_3Ga_5Te_9$, $Cu_2Ga_4Te_7$, $CuGa_3Te_5$, and $CuGa_5Te_8$, in addition to $CuGaTe_2$ containing no vacancies. In the crystal of $Cu_3Ga_5Te_9$, $Cu_2Ga_4Te_7$, $CuGa_3Te_5$, and $CuGa_5Te_8$, one-ninth, one-seventh, one-fifth, and one-fourth of the cation sites are structure vacancies. Here we investigated the effect of the density of structural vacancies on the TE properties of the Cu-Ga-Te ternary compounds. The Hall mobility (μ_H) and the κ_{lat} decreased with increasing the amount of the vacancies, showing that the vacancies scattered both carriers and phonons. It was found that the decreasing rate of the μ_H was larger than that of the κ_{lat}, indicating that the vacancies scattered carriers much more than phonons. Therefore, the existence of the vacancies degraded the TE performance of the Cu-Ga-Te ternary compounds. In other words, $CuGaTe_2$ without structural vacancies might show the best ZT.