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Possible Enhancement of Thermoelectric Properties by Use of a Magnetic Semiconductor: Carrier-Doped Chalcopyrite $Cu_{1-x}Fe_{1+x}S_2$

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We focus on the chalcopyrite $CuFeS_2$ to utilize the interaction between carriers and magnetic moments of Fe as a possible source to achieve high power factor. Polycrystalline samples of $Cu_{1-x}Fe_{1+x}S_2$ were synthesized, and their thermoelectric properties are reported. Electrical resistivity decreased by two orders of magnitude with increasing x, while the Seebeck coefficient showed large values of -200 μV/K at room temperature. Thermal conductivity also decreased with the increase of x. As a result, the power factor and the figure of merit, zT, of the carrier-doped samples are about 10 times larger than those of $CuFeS_2$. These observations suggest that magnetic semiconductors can make good thermoelectric materials.

Key words: Thermoelectric property, power factor, magnetic semiconductor, chalcopyrite

INTRODUCTION

In the last decade, thermoelectric materials have attracted a significant amount of attention because they can generate electric power from waste heat. The performance of thermoelectric materials can be measured by the dimensionless figure of merit, $zT = S^2T/\rho\kappa$, where S, T, ρ, and κ are the Seebeck coefficient, temperature, electrical resistivity, and thermal conductivity, respectively. Recently, great progress has been achieved in materials exploration. Many new materials have been discovered that show zT close to or higher than 1.[1] Those include inorganic clathrates,[2] Zintl compounds,[3,4] and materials with nanostructures.[5,6] The key strategy here has been to reduce the thermal conductivity κ while leaving the electrical conduction intact.

On the other hand, it is still challenging to increase the power factor S^2/ρ, which is also an important figure for power generation. High power factors of the order of 10^{-3} W/K^{2}m are reported in materials such as cobalt oxides,[7] Heusler alloys,[8] and intermetallic compounds with narrow energy gap.[9,10] Since the power factor is sensitive to the electronic structure around the Fermi level, material explorations with precise calculation techniques have to be developed to find materials with high power factors.

In this paper, we suggest that the power factor may also be enhanced by use of magnetic semiconductors. Magnetic semiconductors are a series of materials where magnetic ions such as Mn, Fe, and Co are incorporated into the host semiconductor. They have been intensively studied for spintronics applications, but their thermoelectric properties have rarely been investigated. One characteristic feature of magnetic semiconductors is the strong coupling between carriers and the spins of magnetic ions. This strong interaction may lead to a large effective mass of carriers, which can enhance the Seebeck coefficient with good carrier conduction.

We focus on the natural magnetic semiconductor $CuFeS_2$, known as chalcopyrite. $CuFeS_2$ shows semiconducting behavior with an energy gap of about 0.5 eV observed by optical absorption measurements.[11] The crystal structure is tetragonal with space group $I\bar{4}2d$, where Cu and Fe ions

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