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Thermoelectric Properties of Bi_2Te_2Se Compensated by Native Defects and Sn Doping

M.K. FUCCILLO,[1,2] SHUANG JIA,[1] M.E. CHARLES,[1] and R.J. CAVA[1]

1.—Department of Chemistry, Princeton University, Princeton, NJ 08544, USA. 2.—e-mail: mkfuccil@princeton.edu

In Bi_2Te_2Se the defect chemistry involves native defects that compete such that they can either exchange dominance or else significantly compensate each other. Here we show how the net carrier concentration, $n - p$, which depends on the relative amounts of these defects and is readily obtained from Hall data, can be used as a fundamental materials parameter to describe the varied behavior of the thermoelectric properties as a function of compensation. We report the effects of tuning this parameter over multiple orders of magnitude by hole-doping the n-type material $Bi_2Te_2Se_{0.995}$, which is already significantly compensated because of its Se deficiency. Crystals with different levels of hole doping were achieved by two separate approaches, namely by selecting pieces from different locations in an undoped crystal in which a systematic carrier concentration gradient had been induced by its growth conditions, and alternatively by doping with Sn for Bi. The thermoelectric power factors for $Bi_{2-x}Sn_xTe_2Se_{0.995}$ for $x = 0$, 0.002, 0.005, 0.010, and 0.040 are reported, and the dependence of the transport properties on the extent of compensation is discussed.

Key words: Thermoelectrics, defect chemistry, bismuth telluride selenide, doping

INTRODUCTION

Thermoelectric materials based on the tetradymites $(Bi,Sb)_2Te_3$ and $Bi_2(Te,Se)_3$ (BTS) have long been the best materials for use in Peltier coolers because they have the highest known figures of merit near and below room temperature.[1–3] However, these materials are still under investigation as researchers continually try to drive the thermoelectric figures of merit even higher. The material properties encompassed by the figure of merit $ZT = S^2T/\kappa\rho$ (where S is the Seebeck coefficient, T is the temperature, κ is the thermal conductivity, and ρ is the electrical resistivity) are generally interrelated and competing, so that it is difficult to tune a single parameter as desired without negatively affecting the others. In response to this challenge, several well-established avenues for improving ZT in tetradymites have evolved, such as compositional tuning (e.g., alloying Bi_2Te_3 with Sb_2Te_3)[1–3] and mechanical processing (e.g., introducing grain boundaries and making nanostructures).[4–6] Within a given compositional system, the former approach essentially adjusts the carrier concentration. This, in turn, affects all of the parameters in ZT to reveal the optimal point at which the properties counterbalance each other and yield the highest figure of merit. In particular, too many carriers lead to high κ and low S and therefore a small ZT, whereas too few carriers lead to a small ZT as ρ becomes too large. Tuning the carrier concentration between these two extremes is the key to finding a good balance between the transport properties. In the present study on Bi_2Te_2Se, the carrier concentration has been varied through multiple orders of magnitude using two different methods and consequently the thermoelectric properties have been dramatically affected.

Manipulating the carrier concentration in BTS compounds to achieve a desired result requires an understanding of the defect chemistry of the host

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