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Structural, Magnetic, and Thermoelectric Properties of Some $CePd_3$-Based Compounds

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Intermediate valence compounds such as $CePd_3$ possess unusual electronic properties such as simultaneously high values of the Seebeck coefficient and electrical conductivity, making them strong candidates for low-temperature thermoelectric applications if a method can be developed for reducing the lattice thermal conductivity without altering these desirable electronic properties. This has motivated us to systematically explore the transport properties of several novel series of $CePd_3$-based compounds. The results of these studies have guided us to a deeper understanding of the factors controlling the various transport properties of the system, and we have successfully used this framework to achieve some modest enhancements in the thermoelectric figure of merit. We present an outline of the approaches we have taken and a summary of our results.

Key words: $CePd_3$, thermoelectric, Seebeck, intermediate valence

INTRODUCTION

The ongoing search for materials with favorable thermoelectric properties at low temperatures has generally required a shift away from the conventional semiconductor paradigm in order to find systems with sufficiently large low-temperature Seebeck coefficient values in order to enable high thermoelectric energy conversion efficiencies, as the incorporation of the Wiedemann–Franz relation into the thermoelectric figure of merit ZT equation shows this property to ultimately limit the maximum possible ZT of any material at any temperature.[1] Intermediate valence (IV) compounds have been proposed as one such candidate class of materials, as the noninteger occupation number of the rare-earth f-shells in these compounds corresponds with a strong interaction of the local atomic states with the conduction band, creating a sharp peak in the electronic density of states near the Fermi level which in turn results in unusually large Seebeck coefficient values in what are otherwise typical intermetallic alloys.[2]

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$CePd_3$ is one particularly well-known IV compound which displays a peak Seebeck coefficient of $\sim$115 μV/K at 150 K, creating the opportunity to achieve relatively high ZT values if appropriate thermal conductivity reduction mechanisms which do not significantly affect the IV state can be introduced.[3] Although the chemical composition and Cu_3Au-type crystal structure of this compound are both relatively simple, the many ways in which the material can be easily modified have allowed for extensive study of how various chemical changes can be correlated with changes in the corresponding structural, magnetic, electronic, and thermoelectric properties of the material.[4–14] The relevant conclusions which have emerged from these studies can be generally summarized as follows: (1) the IV state (and therefore the Seebeck coefficient) is extremely sensitive to any changes in the local bonding environment of the cerium atoms;[4–14] (2) aside from applying mechanical pressures on the order of 10 GPa,[15] no mechanism has ever been reported to enhance the value of the Seebeck coefficient above the $\sim$115 μV/K peak value observed in stoichiometric $CePd_3$; (3) it is possible to sharply reduce the lattice thermal conductivity through palladium site substitutions and/or by partially filling the central 1-b interstitial site with various elements.[16–18]