The LDA+DMFT Route to Identify Good Thermoelectrics

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Abstract For technical applications thermoelectric materials with a high figure of merit are desirable, and strongly correlated electron systems are very promising in this respect. Since effects of bandstructure and electronic correlations play an important role for getting large figure of merits, the combination of local density approximation and dynamical mean field theory is an ideal tool for the computational materials design of new thermoelectrics as well as to help us understand the mechanisms leading to large figures of merits in certain materials. This conference proceedings provides for a brief introduction to the method and reviews recent results for LiRh$_2$O$_4$.

1 Introduction

Against the background of climate change and the present energy crisis, the quest for alternative, green energy sources is more urgent than ever. In this regard, thermoelectric materials which transform waste heat (gradients) into electrical power through the Seebeck effect [1, 2] are particularly appealing. However, due to a low efficiency we have not yet witnessed a wider technological application almost 200 years after Seebeck’s discovery. Instead, thermoelectrical applications are restricted...
to niche markets such as radioisotope power systems for satellites [3]. A possible first major application is the exhaust heat of cars and trucks, as automobile companies presently test thermoelectrical generators in prototypes [4]. Such efforts could be put on another level if novel materials with a higher figure of merit $ZT$, where $Z$ is the power factor and $T$ the temperature, and hence a higher efficiency, were available. Most present technical applications use semiconductors such as Bi$_2$Te$_3$ [2] where recently power factors $Z$ considerably larger than 1 could be achieved through phonon [5] and bandstructure engineering [6].

Very promising are novel materials on the basis of strongly correlated electron systems (SCES) [7] which are at the core of the present conference proceedings. This class of materials is very diverse, ranging from metals to Kondo insulators and semiconductors, from $d$ to $f$ electron systems, from relative simple crystal structures such as FeSb$_2$ [8] to most complex metallic cage compounds.

Having such a wide field and the additional possibilities to nano- and heterostructure these systems, a better theoretical understanding and reliable tools to compute thermoelectric properties quantitatively are mandatory. Theoretical physicists from the SCES community have analyzed thermoelectric materials mainly on a model level, i.e., on the basis of the Falikov-Kimball, Hubbard and periodic Anderson model [9, 10], often employing dynamical mean field theory (DMFT) [11–13]. These calculations showed, among others, the importance of correlation-induced enhancements of the effective mass generating a high, but narrow density of states—or spectral function to be precise—close to—but not at—the Fermi level. As a consequence, the thermoelectric figure of merits can be strongly enhanced. On the other hand, theoreticians from the density functional theory (DFT) [14] community have been emphasizing the importance of a particularly high density of states (DOS) [15, 16] and of the large group velocities for certain shapes of the bandstructure [17].

Since both, correlations and bandstructure, can substantially contribute to enhanced thermoelectrical figures of merit, we need to deal with both of them on an equal footing. Only if both aspects are optimized we can expect to design materials or artificial heterostructures with really large figures of merit. Taking correlations and bandstructure into account is possible with the merger [18, 19] of DFT in its local density approximation (LDA) [20] and DMFT, for which the name LDA+DMFT was coined [21], see Refs. [22–24] for reviews. While LDA+DMFT has been applied already to many SCES materials, thermoelectrical properties have been calculated rarely in the past. Noteworthy exceptions are LaTiO$_3$ [25] and LiRh$_2$O$_4$ [26]. The main reasons for this is that a wider experimental interest in SCES thermoelectrics emerged rather recently and that the calculation of thermoelectric properties such as the Seebeck coefficient requires some additional postprocessing which is not yet standard in LDA+DMFT calculations.

### 1.1 Outline

In the following, we will give a brief, elementary introduction to the LDA+DMFT approach in Section 2. This section is divided into the three steps LDA (Section 2.1),