Quasiparticle electronic structures of thermoelectric PbX (X=S,Se,Te)

Axel Svane[1] and Niels E. Christensen[1]

[1]Department of Physics and Astronomy, Aarhus University, DK-8000 Aarhus, Denmark

e-mail: nec@phys.au.dk

A thermoelectric material's potential to convert heat into electricity is quantified by the thermoelectric figure of merit $ZT=S^2T\sigma/\kappa$, where T is the temperature, S the Seebeck coefficient, σ and κ the electrical and thermal conductivities. A high figure of merit requires large S and σ values, but small thermal conductivity. The thermal conductivity has contributions from electrons (κe) and phonons (κl), and for electrons it is difficult to simultaneously maximize σ and minimize κe (Wiedemann-Franz "law" for metals). Therefore materials for thermoelectrics are often semiconductors (doped), and nanostructured (to reduce the thermal conductivity), nanowires, nanodots. Lead telluride, PbTe, has for many years been known to be a good thermoelectric material with ZT up to 1.3-1.5 at high temperatures, but lately it has been realized that new doping and alloying has made it possible to reach even higher values, close to ZT=2 using the lead chalcogenides [1,2]. A materials "design" for such applications requires that the electronic structures, the electron-phonon and phonon-phonon interactions[3] are well understood and accurately modeled. The present work is focused on the electronic structures, and it is shown that conventional density-functional calculations are not sufficiently accurate, and therefore quasiparticle bands are calculated in the quasiparticle self-consistent GW approximation (QSGW, van Schilfgaarde, Kotani and Faleev) including spin-orbit coupling [4]. The semiconducting gap is formed between states of L6 symmetry (L6+ and L6-), which is the reason why these materials are semiconductors even in the local-density approximation (LDA). Closer inspection reveals that the order of the L6 states is reversed in the LDA, and this leads to the wrong sign of the band gap deformation potentials. With QSGW both sign and magnitude of the deformation potentials are in agreement with experiment. Also, the details of the electronic structures which have particular relevance in connection with thermoelectric applications of these materials are discussed.

References

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