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Optimal thermoelectric materials: beyond the Mahan-Sofo approach [1]

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Much of the current literature on optimal thermoelectrics is based on the Mahan-Sofo (MS) approach [1], according to which the transport coefficients can be expressed as integrals of the transport function $\Sigma_{MS}(\varepsilon)$, where $\Sigma_{MS}(\varepsilon) = \frac{2}{\Omega}\sum_{\mathbf{k}} v_{\mathbf{k}}^2 \tau_{\mathbf{k}} \delta(\varepsilon - \varepsilon_{\mathbf{k}})$. The MS approach is based on the standard transport theory formulated in terms of the semi-classical Boltzmann equation. MS made two additional assumptions: (i) the relaxation time approximation was adopted and (ii) the drag effects between electrons and phonons were neglected. The purpose of this talk is to discuss thermoelectric phenomena within the Boltzmann equation approach, but without invoking the additional assumptions (i) and (ii).

In the first part of this talk I will discuss the purely electronic contribution to the figure of merit ZT. Considering the simplest variational solution to the Boltzmann equation which goes beyond the relaxation time approximation and which describes both, the heat current and the charge current [2], I will show that the role of the MS function $\Sigma_{MS}(\varepsilon)$ is played by the function $\Sigma(\varepsilon) = \frac{2}{\Omega}\sum_{\mathbf{k}} v_{\mathbf{k}}^2 \delta(\varepsilon - \varepsilon_{\mathbf{k}})$ that does not contain the relaxation time $\tau_{\mathbf{k}}$ of the electrons. After presenting a simple physical explanation of the optimal shape of the function $\Sigma(\varepsilon)$ I will consider several special cases. First I will show that a nondegenerate doped semiconductor whose chemical potential μ is sufficiently far from the band edge represents an (electronically) ideal thermoelectric material. Then I discuss several metallic systems with a structure in $\Sigma(\varepsilon)$ close to μ caused by band-structure effects: weak hybridization with a set of localized levels, van Hove singularities in layered systems, and resonant interlayer tunneling in layered systems. I will argue that none of these systems leads to a promising thermoelectric material.

In the rest of this talk I will present preliminary results of a study of the coupled system of electrons and phonons. I will consider the simplest variational solution to the coupled Boltzmann equations for the electrons and phonons [2] and I derive a full formula for the figure of merit of such a system. In order to concentrate on novel effects, I then specialize to a particle-hole symmetric system and discuss under what conditions it can exhibit a large figure of merit. Relation of such an approach with the recent experimental results on silicon nanowires [3] will be discussed.

[1] G.D. Mahan and J.O. Sofo, Proc. Natl. Acad. Sci. USA **93**, 7436 (1996).
[2] J.H. Ziman, *Electrons and Phonons* (Clarendon, Oxford, 1960).
[3] A.I. Boukai *et al.*, Nature **451**, 168 (2008).

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