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Optimal doping and entropic origin of giant thermopower in doped Mott insulators [1]

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Empirically, strong correlations can lead to giant thermoelectric power. It is however necessary to develop our theoretical tools to better understand this physics. We thus study the Seebeck coefficient of the Hubbard model on the 3-dimensional FCC lattice at various fillings and interaction strengths using dynamical mean-field theory (DMFT) for the one-band Hubbard model.

To solve the impurity problem of DMFT, we use two different methods. 1) The numerically exact continuous time quantum Monte Carlo method (CTQMC) [1], that relies on the Monte Carlo summation of all diagrams obtained from the expansion of the partition function in powers of the hybridization. 2) Iterated Perturbation Theory (IPT), an approximation method that relies on an interpolation from 2nd order perturbation theory for the Anderson impurity problem [2]. It was realized during this work that, for large couplings, the popular condition of Refs.[3] to fix the bath chemical potential gives unphysical results at low temperature. We thus developed a new condition for IPT. Indeed, at large enough coupling and in the paramagnetic state, the double occupancy (D) becomes a simple function of the density and is practically independent of the temperature. We thus fix this condition instead.

We show how the high frequency limit [4] and the Kelvin approach [5] of the thermopower give reliable estimates of the DC limit at weak to intermediate couplings and in the strongly interacting case without the need for analytical continuation. For the former, we look at the infinite limit of an AC thermopower to obtain $S^* = \lim_{\omega\to\infty} S_{Kubo}(\omega)$. While, for the latter, by first considering $\omega \to 0$ and, afterward, letting the system going to the thermodynamic limit we obtain $S_{Kelvin} = \left(\frac{\partial\sigma}{\partial n}\right)\Big|_T = \left(\frac{\partial\mu}{\partial T}\right)\Big|_n$, where σ is the entropy. This expression looks very much like the Mott-Heikes result which states that at large enough temperature (or large enough incoherence) the thermopower is given by $S_{MH} = \left(\frac{\partial\sigma}{\partial n}\right)\Big|_E$. Fixing temperature is a much more natural process.

This also allows us to discuss to what extent, in doped Mott insulators, the enhancement of the thermopower can be understood on entropic grounds. The enhancement of thermopower is particularly important at strong coupling in this highly frustrated FCC lattice. These results will be useful in the quest for finding optimal conditions for large Seebeck coefficients.

[1] P. Werner, A. Comanac, L. de Medici, M. Troyer, and A. J. Millis Phys. Rev. Lett. **97**, 076405 (2006)
[2] H. Kajueter and G. Kotliar Phys. Rev. Lett. **77**, 131 (1996)
[3] A. Martin-Rodero, F. Flores M. Baldo, and R. Pucci, Solid State Commun. **44**, 911 (1986)
[4] B. S. Sriram Shastry Rep. Prog. Phys. **72** 016501 (2009)
[5] M. R. Peterson and B. S. Shastry Phys. Rev. B **82**, 195105 (2010)

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