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Charge Kondo effect in molecular quantum dots and $Pb_{1-x}TeTl_x$ and a mechanism for large thermopower

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The negative-U Anderson model is believed to be a relevant low energy effective model for molecular quantum dots with strong local electron-phonon interactions, for valence skipping Tl impurities in semiconducting PbTe and possibly also for many other systems, e.g. $H^{+/-}$ at interstitial sites in Si. The negative-U Anderson model supports a charge Kondo effect, in which the role of spin up and spin down states in the conventional spin Kondo effect is played by the empty and doubly occupied states. Dynamic valence fluctuations between these lowest pseudospin states results in the charge Kondo effect. This has some remarkable thermoelectric properties which we elucidate here in the context of two systems, (1), molecular quantum dots, and, (2), semiconducting PbTe doped with Tl impurities.

1. For molecular quantum dots described by the negative-U Anderson model we show that the charge Kondo effect provides a mechanism for enhanced thermoelectric power via a correlation induced asymmetry in the spectral function close to the Fermi level. A dramatic enhancement of the Kondo induced peak in the thermopower is found with Seebeck coefficients exceeding $50\mu V/K$ over a wide range of gate voltages [1].

2. For PbTe doped with a small concentration x of Tl impurities acting as acceptors we use the numerical renormalization group method to show that the system self-tunes to a charge Kondo state upon increasing x above a critical concentration $x^* \approx 0.5\,\%$. The resulting charge Kondo effect naturally accounts for both the observed superconductivity at $x > 0.3\%$ [2, 3, 4] and the unusual low temperature and doping dependence of normal state properties, including the self-compensation effect in the carrier density and the non-magnetic Kondo anomaly in the resistivity, which we find to be in good qualitative agreement with experiment [5]. Our results for the Tl s-electron spectral function provide a new interpretation of point contact data [6].

We conclude that Tl impurities in PbTe constitute the first convincing physical realization of the charge Kondo effect, albeit a complex one due to the strong dependence of the chemical potential on temperature, and the presence of local disorder, phonons etc. Hence, we suggest that negative-U molecular quantum dots may eventually offer cleaner realizations of the charge Kondo effect with enhanced thermopowers and figures of merit of potential use in low temperature thermoelectric applications [1].

[1] S. Andergassen, T. A. Costi, and V. Zlatić, Phys. Rev. Lett. (2011 submitted); arXiv:1101.4124.
[2] I. A. Chernik and S. N. Lykov, Sov. Phys. Solid State, **23**, 817 (1981).
[3] Y. Matsushita, H. Bluhm, T. H. Geballe, and I. R. Fisher, Phys. Rev. Lett. **94**, 157002 (2005).
[4] M. Dzero and J. Schmalian, Phys. Rev. Lett. **94**, 157003 (2005).
[5] T. A. Costi and V. Zlatić, Phys. Rev. Lett. (2011 submitted); arXiv:1109.xxxx.
[6] H. Murakami, W. Hattori, Y. Mizomata, and R. Aoki, Physica C **273**, 41 (1996).