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Signatures of correlation effects and thermopower in FeSi

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Iron based narrow gap semiconductors such as FeSi, $FeSb_2$, or $FeGa_3$ have received a lot of attention because they exhibit striking similarities to heavy fermion Kondo insulators. Iron silicide, FeSi, which we consider as a prototypical compound in this class, is an ordinary semiconductor at low temperatures. At high temperatures, however, it behaves as a bad metal with a Curie-Weiss like susceptibility. Signatures of this crossover are observed in particular in the thermoelectric properties : The Seebeck coefficient reaches notable values of about 700μV/K at low temperature. Above 100K, however, it is largely suppressed, while displaying multiple sign changes, i.e. a complex balance between electron and hole dominated transport.

Many proposals have been advanced to explain the unusual behavior in various observables. Among these figure Hubbard-like correlations, spin-state transitions, a thermally induced mixed valence, spin-fluctuations, as well as anomalous electron–phonon couplings. However, lacking quantitative methodologies applied to this problem, a consensus remains elusive to date.

Here, we employ realistic many-body calculations[1] to elucidate the impact of electronic correlation effects on FeSi. Our methodology accounts for all substantial anomalies observed in FeSi : the metalization, the lack of conservation of spectral weight in optical spectroscopy, and the Curie susceptibility. In particular we find a very good agreement for the anomalous thermoelectric power. We find the suppression of the Seebeck coefficient to be driven by correlation induced incoherence. The change in the dominant type of carriers is correlated with the non-monotonous evolution of the chemical potential, causing a large temperature dependence of the particle/hole asymmetry. Validated by this congruence with experiment, we further present a physical picture of the microscopic nature of the crossover to the metallic state.

[1] based on the combination of density-functional theory and dynamical mean field theory