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Is FeSi a Kondo insulator?

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Abstract

FeSi exhibits a Curie-like susceptibility accompanied by a moderate electronic conductivity at high temperature. At low temperature the magnetic susceptibility goes to zero, and a small charge gap appears. We discuss phenomenological and theoretical aspects of the relationship between FeSi and various rare-earth correlated systems often referred to as Kondo insulators.

Keywords: FeSi; Kondo insulator

FeSi presents an unusual and interesting phenomenology. A Curie-like susceptibility at high temperature [1] suggests local moment magnetism. At low temperature, however, the susceptibility goes toward zero with an activated temperature dependence. For the best samples activated behavior is obtained for the resistivity as well, and one can infer from these temperature dependencies a small gap of about 60 meV. This phenomenology is in many ways similar to the f-level correlated insulators, known as Kondo insulators [2], however, unlike the Ce and U compounds of that class it contains no narrow f-band in which strong correlation effects consistently occur. The question naturally arises, therefore, as to what the relationship is between FeSi and the Lanthanide and Actinide compounds that exhibit the "Kondo-insulator phenomenology". Here we will address that question by looking at results from several spectroscopic probes, including infrared [3–5], Raman [6], and photoemission [7], and discuss them within the context of theoretical ideas and the phenomenology of several f-level systems.

Infrared measurements show a number of interesting and notable features [3, 4]. First, there is a loss of spectral weight at low energies as the material is cooled, corresponding to the appearance of a gap or pseudo-gap in the infrared conductivity, $\sigma_1(\omega)$ (and to the loss of moment in the susceptibility). The temperature dependence of this gap appearance is, significantly, much too rapid to be explained as an ordinary freezing-out due to the temperature dependence of the Fermi function [3]. Second, one observes anomalously large changes in the linewidths and strengths of infrared active phonons. Third, there is the very unusual observation that the spectral weight which disappears from the low frequency region as the gap forms does not appear just above the gap, as one would expect (Fig. 1(a)). One can infer that it must be shifted to much higher frequency and reappear in a broad and rather high frequency range. A shift in spectral weight from low to high energy was subsequently observed in $CePd_3$ (Fig. 1(b)), which may be a "lightly-doped" Kondo-insulator type material [5].

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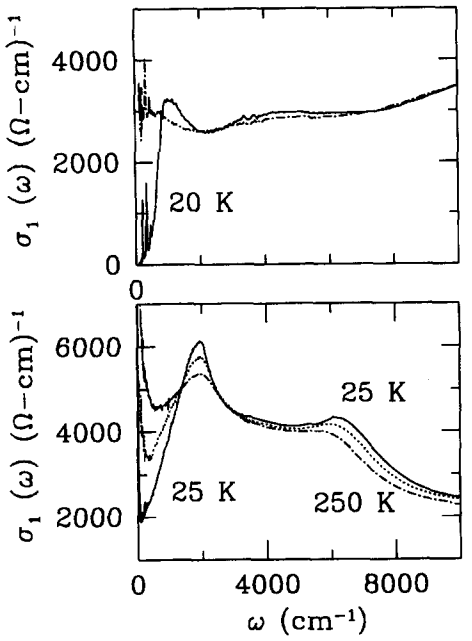

Fig. 1. This figure shows the conductivity as a function of frequency for FeSi (a), at 20 and 250 K; and $CePd_3$ (b), at temperatures of 25 and 150 and 250 K. Both show evidence for a large energy spectral weight shift. For FeSi, the low frequency region of the conductivity is depleted at low temperature as a consequence of gap formation. However, the missing conductivity does not reappear just above the gap as one would expect in ordinary theory, and one must infer a shift of spectral weight to high energy. For $CePd_3$ the low frequency conductivity is similarly depleted at low T, and reappears in the high frequency (6000–10 000 cm^{-1}) region, which is an explicit manifestation of the large energy spectral weight shift.

Raman measurements [6] are similar to the infrared in that they also see a temperature dependence of the gap appearance which is too abrupt to be explained by simple Fermi function origins. They also observe, in Raman active phonons, very substantial changes in phonon linewidths and strengths associated with cooling and gap formation in the electronic response [6].

Photoemission investigations have been recently performed on FeSi as well [7]. The angle resolved studies show an extremely narrow peak close to, but below, the Fermi surface. The width of this peak, seen about 25 meV below the Fermi surface, is approximately 35 meV and is resolution limited. Commensurate with the infrared and Raman observations, they find a remarkably rapid temperature dependence for this peak. They report that it is, to the best of their knowledge, the strongest temperature dependence recorded by photoemission. The photoemission peak disperses very slowly with angle (k) indicating a very narrow bandwidth of roughly 30 meV.

A number of theoretical ideas for FeSi, based on band theory and other approaches including correlation, are discussed in Refs. [8–12]. One finds several things in the phenomenology of FeSi that are not contained in band theory. These include the apparent sharpness of the valence band near the chemical potential, the very abrupt temperature dependence of the conductivity and density of states related probes described above, and the anomalous shift of spectral weight from low to high energy as seen in the infrared conductivity [13].

The behavior of several Ce-based compounds is not very dissimilar [5, 14]. Compounds such as $CePd_3$ and $Ce_3Bi_4Pt_3$ show the same abrupt temperature dependence as well as evidence for a large energy shift of spectral weight from low to high energy as a pseudo-gap appears. This is particularly dramatic in $CePd_3$ [5], where one can explicitly see the loss of spectral weight at low energy, and its reappearance at about 1 eV. The issue of the relationship between $CePd_3$ FeSi and compounds of the Kondo insulator class is discussed in Ref. [5], where it is proposed that $CePd_3$ may be regarded as a doped Kondo insulator.

The issue of a possible difference between the spin and charge gaps is also of interest. For $Ce_3Bi_4Pt_3$, evidence from infrared for a larger charge gap [14] has been discussed based on a comparison with neutron data [15]. For FeSi no difference between spin and charge gaps has been resolved, and, based on existing temperature dependencies (of the susceptibility and resistivity) [1], one would not expect any difference to be very large (i.e., less than about 20 or 30%). This is an important area where further experimental and theoretical input would be helpful to refine and further quantify our understanding.

In the "Kondo limit" of the Anderson lattice model theory (i.e., when f-level occupancy fluctuations are completely suppressed by correlation),

the spin gap is associated with the excitation from the Kondo singlet ground state to the spin triplet configuration. This does not involve any charge motion, and one expects the spin gap to be about 1/2 the energy of the charge gap. On the other hand, in ordinary band theory, or in the simple hybridation approach, the spin and charge gaps are the same. Adding correlation effects, however, will tend to reduce the energy of the triplet exciton, which moves into the gap and evolves (continuously) into the triplet spin excitation of the Kondo limit picture as the correlations strength increases. That there is no phase transition in this evolution, implies that there is no qualitative difference among correlated insulators.

From the experimental results discussed above, including the abrupt temperature dependence, the large energy infrared spectral weight shift, and the evidence for a narrow band, it seems reasonable to conclude that FeSi is strongly correlated insulator. While it is not in the Kondo limit, it is also probably true that the Ce and other f-level based Kondo insulators are not in the extreme Kondo limit. At this juncture it is natural to place FeSi in the same class as $Ce_3Bi_4Pt_3$ and other related f-level systems [2]. Clearly this remains an interesting area for further study, and is one that should be fruitful for our understanding of fundamental problems such as moment compensation and the consequence of correlation effects in small gap semiconductors.

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