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Narrow-gap signature of $\text{Fe}_x\text{Co}_{1-x}\text{Si}$ single crystals

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Heat capacity, resistance, and magnetic susceptibility have been measured on FeSi, $\text{Fe}_{0.90}\text{Co}_{0.10}\text{Si}$, $\text{Fe}_{0.10}\text{Co}_{0.90}\text{Si}$, and CoSi single crystals. From resistance experiments, the activation energy (Δ) shows a small variation from $\Delta \approx 310$ K in FeSi to $\Delta \approx 325$ K for the $\text{Fe}_{0.90}\text{Co}_{0.10}\text{Si}$ compound. For the 90% Co compound the gap signature disappears completely. Heat capacity experiments performed in the temperature range $1.5 \text{ K} < T < 20 \text{ K}$ reveal an increasing Sommerfeld coefficient (γ) with increasing Co content. A comparison is made with the hybridization gapped material $\text{Ce}_3\text{Bi}_4\text{Pt}_3$.

1. Introduction

Many years have elapsed since the discovery of anomalous magnetic behavior in the small gap semiconductor FeSi (see ref. [1] and references therein). This intermetallic compound presents an unusual behavior of the magnetic susceptibility $\chi(T)$ which exhibits a pronounced maximum in the vicinity of 500 K. Furthermore, neutron diffraction studies [2] show no ordering at low temperatures. A semiconducting band model [1] has been proposed, with a narrow gap, to describe the temperature dependence of $\chi(T)$ and heat capacity. However, the gap model is not completely accepted to describe the thermal properties of FeSi. The possibility of having a temperature-induced paramagnetic moment has been pointed out theoretically [3], and experimentally [4] on the basis of neutron-scattering measurements. In the case of $\text{Fe}_x\text{-Co}_{1-x}\text{Si}$ alloys, weak itinerant ferromagnetism, with a helical spin structure (long period $> 300 \text{ \AA}$), has been reported [5] for $0.3 < x < 0.90$.

In view of the controversy surrounding an interpretation of the physical properties of FeSi, we have investigated the gap formation as a function of Co content. Here we present an account of thermodynamic and transport measurements in a wide temperature range for $\text{Fe}_x\text{Co}_{1-x}\text{Si}$ single crystals.

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2. Materials and experimental techniques

Single crystals were grown via chemical transport. Approximately 1 g of starting material and 60 mg of iodine were sealed in a quartz tube. The tube was placed in a gradient furnace with starting material at the hot end (900°C) and samples growing at the cold (750°C) end. Samples of a few mm^3 grew over the course of one week. Resistance was measured using a conventional AC four-probe method in the temperature range $4.2 \text{ K} < T < 300 \text{ K}$. Magnetic susceptibility measurements were performed on a SQUID magnetometer between 2 and 350 K. Finally, heat capacity experiments were carried out using a quasi-adiabatic thermal relaxation technique in the temperature range $1.5 \text{ K} < T < 20 \text{ K}$.

3. Results and discussion

Figure 1 shows the temperature dependence of the resistance of FeSi, $\text{Fe}_{0.90}\text{Co}_{0.10}\text{Si}$, $\text{Fe}_{0.10}\text{Co}_{0.90}\text{Si}$ and CoSi between 4.2 and 300 K. The resistance of FeSi and the 10% Co alloy increase as the temperature decreases from room temperature to the lowest temperature investigated. The rapid rise in the resistance for these two compounds can be interpreted as a gap formation in the electronic density of states. The activation energy Δ can be estimated by plotting $\ln R$ as a function of $1/T$; a linear regime is observed from

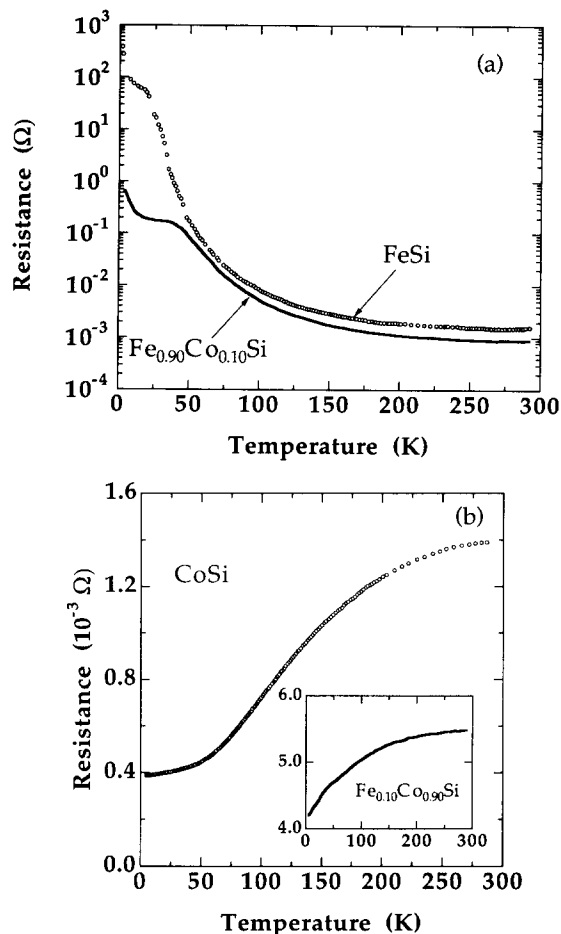


Fig. 1. Resistance as a function of temperature for (a) FeSi and $Fe_{0.90}Co_{0.10}Si$, and (b) for CoSi and $Fe_{0.10}Co_{0.90}Si$ (inset).

33 to 167 K for FeSi and from 33 to 167 K for the $Fe_{0.90}Co_{0.10}Si$ compound. The gap energy (2Δ) estimated from the linear regime is ~ 620 and 650 K for FeSi and $Fe_{0.90}Co_{0.10}Si$, respectively. The low-temperature feature is attributed to impurities incorporated during sample growth. In fig. 1(b) we show the resistance of CoSi and $Fe_{0.10}Co_{0.90}Si$ (inset); metallic behavior is observed for both in the temperature range investigated.

We show in fig. 2 the magnetic susceptibility as a function of temperature for FeSi, CoSi and $Fe_{0.90}Co_{0.10}Si$. A very pronounced rise in $\chi(T)$ is observed for the 10% Co compound at temperatures below 57 K. This rise is consistent with moment formation with increasing Co content. For FeSi a considerable background signal is observed in our susceptibility experiments. This appears to be due to the low sample contribution compared with that of the sample

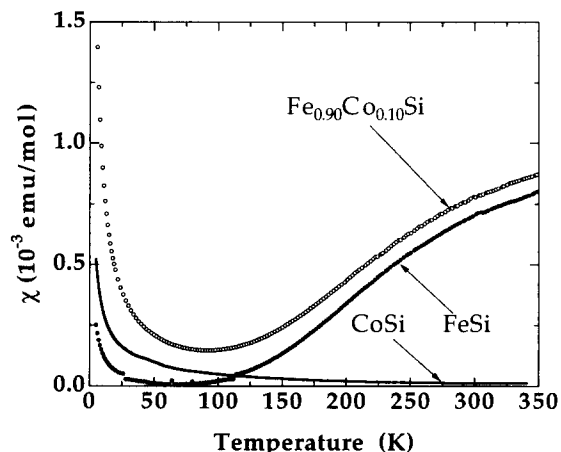


Fig. 2. Magnetic susceptibility, $\chi(T)$, as a function of temperature for FeSi, CoSi and $Fe_{0.90}Co_{0.10}Si$.

holder. Increasing the Co content moves the minimum in $\chi(T)$ towards higher temperatures.

Heat capacity measurements have been performed in a wide temperature range. In fig. 3 we plot C/T as a function of temperature for FeSi, $Fe_{0.90}Co_{0.10}Si$, $Fe_{0.10}Co_{0.90}Si$ and CoSi. The FeSi compound presents the lowest heat capacity compared with all alloys and the CoSi compound. By increasing the Co content an upturn in C/T at low temperature appears. Correspondingly an upturn at low temperatures was also observed in the magnetic susceptibility. The C/T rise with decreasing temperatures is drastically suppressed for the CoSi compound. Using a Debye and electronic ($C = \gamma T + \beta T^3$) fit for the FeSi and CoSi data gives $\gamma = 1 \text{ mJ mol}^{-1} \text{ K}^{-2}$, $\beta = 0.035 \text{ mJ mol}^{-1} \text{ K}^{-4}$ ($\Theta_D =$

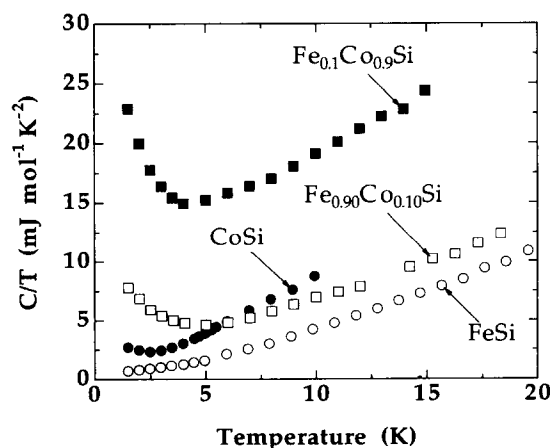


Fig. 3. Heat capacity divided by temperature as a function of temperature for FeSi, $Fe_{0.90}Co_{0.10}Si$, $Fe_{0.10}Co_{0.90}Si$ and CoSi.

480 K) and $\gamma = 2 \text{ mJ mol}^{-1} \text{ K}^{-2}$ and $\beta = 0.056 \text{ mJ mol}^{-1} \text{ K}^{-4}$ ($\Theta_D = 410 \text{ K}$) for the two compounds. In our temperature range, a large variation is observed in the heat capacity of both dilute and highly concentrated Co materials as compared to 'pure' CoSi and FeSi. It is possible that magnon excitations in a helimagnetic phase could be present, since we are close to the known phase boundary for magnetic order [5]. However, we see no evidence for this in our magnetic susceptibility measurements.

In addition to substitutions of Fe and Co, we have substituted P for Si. The advantage of this substitution is that there is no disruption of the Fe sublattice, and phosphorous has no magnetic moment. Figure 4 shows the temperature-dependent electrical resistance and magnetic susceptibility of $\text{FeSi}_{1-x}\text{P}_x$ with nominal $x = 0.05$. These samples were grown using a flux (Sb) growth technique [6]. The magnetic susceptibility is qualitatively similar to that of $\text{Fe}_{0.90}\text{Co}_{0.10}\text{Si}$. This supports the hypothesis that the low-temperature Curie tail in this compound is not due to the Co moment, but associated with the disrupted FeSi lattice itself. The resistance, on the other hand, is very different from the 10% Co substitution. The phos-

phorus substitution ($x = 0.05$) leads to a virtual metallization of the temperature-dependent electrical resistance, although a maximum is observed around 100 K.

FeSi shows striking similar properties to the group of rare earth and actinide compounds known as hybridization gap materials, in which the gap is roughly one order of magnitude smaller. The similarities extend to details of the $\chi(q, \omega)$ as measured via neutron scattering. Dilution studies we have reported [7] on the $\text{Ce}_3\text{Bi}_4\text{Pt}_3$ material in this class show quite similar behavior with doping in both rare earth and non-rare earth sites. The comparison of these results further supports our contention that the underlying physics in the rare earth on the transition metal materials are closely similar.

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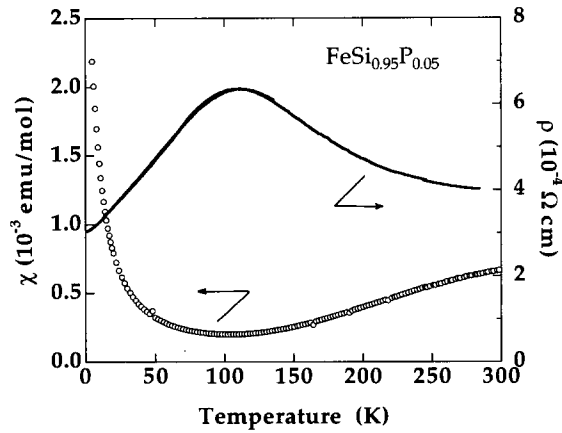


Fig. 4. Resistivity and magnetic susceptibility of $\text{FeSi}_{0.95}\text{P}_{0.05}$.