Impurity metallic conduction below the critical concentration of metal-insulator transition in Fe$_{1-x}$Co$_x$Si

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Abstract
Analysis on very detailed measurements of resistivity ($\rho$) and thermoelectric power (S) of magnetic impurity (Co) substituted iron silicide (FeSi) has been presented in this report. The impurity valence electrons of Co dominate the whole physical properties at low temperatures below 35 K, below the critical concentration $x_c$ ($\approx 0.02$). The negative thermopower and the positive slope in the resistivity at low temperatures are exciting and show that the system is not entirely insulator below the critical concentration of metal–insulator transition ($x_c$). So, due to the external impurity electrons, the system’s magnetic ground state could change considerably compared to the parent compound FeSi. This report may help unveil the interesting low-temperature transport properties between $x=0$ and $x=0.04$ (Fe$_{1-x}$Co$_x$Si). Two band model and variable range hopping model were employed to explain the low-temperature electrical and thermal transport properties.

Supplementary material for this article is available online

Keywords: impurity band conduction, thermoelectrics, narrow band gap semiconductor, metal insulator transition

(Some figures may appear in colour only in the online journal)

1. Introduction
Semiconductor contribution is enormous in electronics industries, including spintronics, and knowledge of transport properties is necessary. It gives a basic model to theorize transport properties at absolute zero. The scientific community has always had a keen interest in semiconductors due to their unusual transport behavior by external parameters. The physical properties of semiconductors at low temperatures are exciting due to the presence of finite impurities. So it is necessary to study impurity effects on transport properties at low temperatures. Substitution of impurities could change the semiconducting/insulating nature of the system to metals. metal–insulator transition (MIT) is one fascinating physical phenomenon that has been studied over several decades. Physical properties around MIT helps to understand how the transport properties are greatly affected by external parameters such as magnetic field, impurities, pressure, etc. Classical semiconductors such as Silicon (Si) or Germanium (Ge) have been rigorously studied under the above external conditions over the past several decades, and their physical properties are well analyzed with various impurities such as Al, P, As [1–4]. FeSi is a hybridized semiconductor whose physical properties are
exotic at low temperatures. Alloying transition metal iron and metalloid silicon make a unique material whose behavior is neither metal nor completely semiconductor. FeSi crystallizes B20 cubic form with space group P2\(1\)3. Each unit cell of FeSi contains eight atoms. Exotic properties of the ground state are connected with the non-centrosymmetric arrangement of Fe and Si atoms in unit cell [5]. At room temperatures, FeSi behaves like metallic, whereas while cooling to low temperatures, it changes to activated behavior due to a gap opening around the Fermi level [6, 7]. FeSi is paramagnetic metal at high temperatures above \(\sim 400 \text{ K}\), but when decreasing temperature, resistivity increases and saturates below 10 K. Activation energy of FeSi is 50 meV at low temperatures [7, 8].

Researchers suggest that the appearance of non-magnetic singlet ground state at low temperatures is of Kondo type but still debate is going on [9]. The ground state of this system is distinct in the sense that electronic properties cannot be explained with the classical semiconductor model. The impurities make the ground state of the system very sensitive. Cobalt (Co) has one extra electron as compared to the Iron in the 3d-orbital, and adding Co in the FeSi makes the system becomes electron-dominated. The Co-doped FeSi shows MIT above \(x_c \approx 0.02\), helimagnetic behavior above \(x = 0.05\) [10–13] and a non-Fermi liquid can be derived with Mn as impurities [14]. Also, the 20 percentage Co-doped FeSi shows skyrmiom magnetic structure, despite the strong site disorder due to doping [15]. Ge has a similar electronic configuration as Si and, by substituting in Si site makes ferromagnetic metal [16]. FeSi and CrSi are paramagnetic metal [17], MnSi is itinerant ferromagnet and CoSi is diamagnetic metal [14]. All these varieties of ground states can be achieved with the transition from semiconducting to metallic nature at low temperatures with the various dopant. So by understanding the formation of impurity states around the Fermi level will help to gain knowledge on ground-state properties and impurity effects at low temperatures.

2. Experimental methods

Cobalt doped FeSi samples were prepared by using the arc melting technique. High purity constituent elements such as Fe, Co, and Si (of purity better than 99.99\%) were loaded in a double-walled arc melting chamber, and the rod-shaped samples were obtained by melting the constituent elements in an argon environment to avoid oxidation at higher temperatures. Ti ball were used as a gettering element to observe residual impurities. The resultant rod-shaped samples were sealed in a quartz tube which was at a high vacuum better than \(10^{-6} \text{ mbar}\). The sealed quartz tubes were loaded in the furnace for annealing at 1000 °C for a week. The purpose of annealing was to remove the strain from sudden quenching in the preparation of arc melting and improve better homogeneity.

Electrical transport property measurement was carried out in 2 K/14 T PPMS (QD USA) using the dc four-probe method. Moreover, thermoelectric power measurement had been done by using CCR homemade differential dc sandwich method [18].

3. Results and discussions

3.1. XRD analysis of Fe\(_{1-x}\)Co\(_x\)Si

The Bruker D8 x-ray diffractometer (\(Cu - K\alpha\) radiation) was used to confirm the phase purity of the samples. Fullprof software was used to refine the powder diffraction data. The diffraction data shown in figure 1 with Rietveld refinement. The lattice parameters were extracted and tabulated in table 1. The lattice parameters are not varying considerably with Co concentration. The absence of impurity peak in the system and the single-phase nature of samples implies that the Co atoms successfully replaced Fe atoms. The Rietveld refinement, Bragg R-Factor and Chi\(^2\) for Fe\(_{1-x}\)Co\(_x\)Si (including FeSi) are added in supplementary materials.

3.2. Electrical resistivity of Fe\(_{1-x}\)Co\(_x\)Si

Figure 2 demonstrates the electrical resistivity of Fe\(_{1-x}\)Co\(_x\)Si for different level of cobalt substitutions (\(x = 0.0025\) to 0.04) at the temperature interval from 2 to 300 K. The figure 2 inset (a) shows the electrical resistivity of Fe\(_{1-x}\)Co\(_x\)Si in linear versus log plot. The resistivity of FeSi at 300 K is 5.7 milli-\(\Omega\)-cm, which is indicative of metallic nature whereas, as we cool down the system from 300 to 2 K, the resistivity increases by four orders of magnitude due to activation of intrinsic majority electrons. At low temperatures below 30 K, the resistivity saturates due to hopping conduction of extrinsic minority holes [19]. The overall resistivity of Co-doped FeSi decreases with increasing Co concentration. The resistivity of \(x = 0.0025\) changes by four orders of magnitude in the whole temperature range. Below 20 K, the resistivity shows logarithmic increasing behavior. Furthermore, this low-temperature nature is different from the behavior of FeSi. This might be due to the electron–hole recombination; as a result, the number carrier concentration decreases. Hence, the resistivity shows insulating behavior. The resistivity below 40 K shows dramatic changes at higher concentrations above \(x = 0.0025\), which we will see in detail. In \(x = 0.01\), the electrical resistivity shows a hump below/at 40 K, indicating metallic behavior. This metallic conduction is due to the n-type carriers from the external doping.

Further, the resistivity shows a negative slope below 15 K. This implies a competition between the impurity holes from FeSi and the impurity electrons from the Co external doping. With further increasing the Co doping concentration, the positive slope continues, and the low-temperature negative slope disappears, implying the domination of impurity electron conduction. The appearance of the hump like feature (\(x = 0.01\), \(x = 0.0125\)) and positive slope (\(x = 0.0175, 0.02, 0.04\)) in the Co doping concentrations implies that the metallic/impurity-band conduction starts even at a lower concentration, below \(x_c\). The temperature at which the slope changes also move towards the higher temperature. This indicates that the metallic conduction
dominates over the semiconducting nature with increasing Co concentration.

To explain the observed resistivity behavior, we have employed various models, such as the variable range hopping (VRH) and the two-band model (TBM). The VRH model for resistivity in disordered semiconductors is defined as

$$\rho = \rho_0 \times \exp\left(\frac{T_0}{T}\right)^{\beta}$$

(1)

where $\beta = 1/4$ for Mott—VRH and 1/2 for Efros–Shklovskii VRH [20, 21]. The VRH model has been used in the resistivity of Fe$_{1-x}$Co$_x$Si and shown in figure 3. The VRH with $\beta = 1/4$, below 30 K, has been well fitted for $x = 0, 0.0025$ with goodness of fit ($\chi^2$) more than 0.9 and $T_0 = 0.09, 7.75$ K respectively. However, VRH model ($\beta = 1/4, 1/2$) is not fitting well in $x = 0.01$ and the obtained $\chi^2$ value is below 0.3. The $\chi^2 = 0.6$ for $x = 0.0125$ ($\beta = 1/4$). For the concentrations above $x = 0.0125$, resistivity shows positive slopes, implying metallic conduction due to impurities.

The resistivity, at the temperatures between 50 and 200 K, act as semiconducting and the resistivity below 50 K becomes...
metallic conduction (above \(x = 0.0125\)). TBM, which consists of metallic and semiconducting contributions, could be an appropriate model to explain this behavior. The TBM can be used to understand the effect of the semiconducting as well as metallic impurity band and to calculate the activation energies of Fe\(_{1-x}\)Co\(_x\)Si [19, 22].

\[
\sigma(T) = \frac{1}{\rho_{01} + A_p T} + \frac{\sigma_{02}}{\sigma_{01}} \exp\left(-\frac{E_{\sigma p}}{2k_B T}\right) \quad (2)
\]

The first term in the right hand side of the equation (2) is for the metallic band \((\sigma_1 = (\sigma_{01}^{-1} + A_p T^{-1})^{-1})\) responsible for impurity conduction, and the second term is for the semiconducting band \((\sigma_2 = \sigma_{02} \exp(-E_{\sigma p} / 2k_B T))\) responsible for the activated conduction. \(E_{\sigma p}\) is the activation energy, \(k_B\) is the Boltzmann constant and \(\frac{1}{\sigma_{01}}\), \(\frac{1}{\sigma_{02}}\) are the background resistivity due to scattering from the metallic and the semiconducting band respectively. The coefficients derived by using equation (2) tabulated in table 2. The coefficient \(A_p\) increase with increasing Co concentration up to \(x = 0.02\) [19, 22]. The TBM fit with electrical conductivity of Fe\(_{1-x}\)Co\(_x\)Si are shown in figure 4. For \(x = 0.04\), the \(A_p\) is decreased again and the non-linearity could be due to the metallic nature derived by impurity substitutions is different from the ordinary Fermi liquid metals [14]. The activation energy \((E_{\sigma p})\) decreases with increasing Co concentration. Apart from the VRH and TBM, Chernikov et al used a model consists of two rectangular bands of width \(W\), and separated by the energy gap \(E_g\) to explain the conductivity behavior [10]. The conductivity fitting is done from 130 to 300 K, in which the system behaves as bad metal. The fitting in the reference considerably deviates below 130 K, where the extrinsic holes (from FeSi) or impurity electrons (from Fe\(_{1-x}\)Co\(_x\)Si) conduction is established. This manuscripts intended to explain the low temperature transport properties from Co impurities and the TBM is appropriate for this.

### 3.3. Thermopower of Co substituted FeSi

The thermopower data of Fe\(_{1-x}\)Co\(_x\)Si from \(x = 0\) to \(x = 0.04\) shown in figure 5. The measurement is done on the temperature interval between 5 and 300 K. The thermopower of parent compound FeSi shows positive values at room temperature \((S = +0.4 \mu V K^{-1})\), and this is due to the dominance of high mobility holes. These holes are extrinsic, which is created by the slight deficiency of silicon during sample preparation [23]. The decrease in temperature makes the thermopower negative and reaches a maximum in the negative direction around 110 K. This negative thermopower is due to the activation of electrons across the semiconducting band. The thermopower suddenly goes in a positive direction with further decreasing temperature and reaches a positive maximum at 40 K, implying that the carrier conduction changes from electrons to holes. This positive thermopower is due to extrinsic holes. The thermopower goes to zero temperature goes to near zero. From the nature of thermopower of FeSi, two kinds of carriers dominate in the conduction process, namely the minority holes conducting at a high temperature above 300 K as well as at low temperature below 60 K and the majority carriers dominating in the mid-range of temperature interval between 60 and 290 K.

![Figure 4. Electrical conductivity of Fe\(_{1-x}\)Co\(_x\)Si with TBM fit.](image)

![Figure 5. Thermopower of Co substituted FeSi from x = 0 to x = 0.04.](image)

Table 2. Coefficients derived from TBM fit for the resistivity of Fe\(_{1-x}\)Co\(_x\)Si.

| Concentration \((x)\) | \(\frac{1}{\rho_{01}}\) \((\Omega cm)^{-1}\) | \(\frac{1}{\sigma_{01}}\) \((\Omega cm^{-1})\) | \(E_{\sigma p}\) \((meV)\) | \(R^2\) |
|-----------------------|---------------------------------|------------------|------------------|------|
| 0.01                  | 9.1                             | 2.48 \times 10^{-3} | 0.0018           | 27.1 | 0.999 |
| 0.0125                | 173.6                           | 3.34 \times 10^{-6} | 0.000243         | 25.2 | 0.998 |
| 0.0175                | 2102.16                         | 3.74 \times 10^{-5} | 5.4 \times 10^{-5} | 24.6 | 0.997 |
| 0.02                  | 1924.1                          | 5.68 \times 10^{-5} | 9.6 \times 10^{-5} | 24   | 0.996 |
| 0.04                  | 757.57                          | 5.5 \times 10^{-6} | 1.54 \times 10^{-4} | 23.7 | 0.997 |

For the metallic band \((x = 0.04)\), the VRH and TBM are used to understand the effect of the semiconducting as well as metallic impurity band and to calculate the activation energies of Fe\(_{1-x}\)Co\(_x\)Si [19, 22].
The substitution of cobalt makes dramatic changes in the low-temperature thermopower of FeSi. The thermopower at $x = 0.0025$ shows an enhancement at low temperatures compared to the parent FeSi. The value of thermopower for $x = 0.0025$ is $+1350 \mu V K^{-1}$ at 36 K, which is 460 $\mu V K^{-1}$ higher than the parent FeSi. The thermopower is also enhanced at 130 K in the negative direction. It is interesting to note that the dopant thermopower is higher than the parent compound. This is as follows; cobalt has one extra electron as compared to Fe. The substitution of cobalt adds electron-like density of states near the conduction band, and these impurity electrons make recombination with holes. As a result, the number of extrinsic holes and intrinsic electrons decreases. So, the thermopower increases, because it is inversely proportional to the carrier concentration ($S \sim \gamma T/ne$) [24]. The increase in cobalt concentration makes the thermopower negative at low temperatures below 40 K at the concentration above $x = 0.0025$. The negative thermopower is due to the domination of the impurity electrons. The negative peak at 30 K increases with increasing Co concentrations up to $x = 0.02$. At the same time, positive thermopower is drastically suppressed with Co concentration. The 30 K—negative peak and also the over all magnitude of the thermopower in $x = 0.04$ is decreased, due to the $S$ is inversely proportional to the carrier concentration ($S \sim \gamma T/ne$). At $x = 0.04$, the thermopower is fully negative except in the narrow interval between 90 and 150 K with a small positive thermopower. This shows the competitive conduction between impurity electrons and the extrinsic holes. At room temperature, the thermopower of the Co doped FeSi does not vary considerably.

The nature of thermopower in Co-doped FeSi shows that the two kinds of bands dominate: (1) metallic band extrinsic carriers and (2) the semiconducting band due to the intrinsic carriers. The TBM helps find the effect of impurity carriers and the intrinsic carrier contributions. The general two band contributions to $S$ is derived using Nordheim-Gorter rule [19, 25]:

$$ S = \frac{S_1 \sigma_1 + S_2 \sigma_2}{\sigma_1 + \sigma_2} $$

where $S_1 = \pi^2 k_B^2 T/(2eE_F)$ represents diffusion thermopower and $S_2 = C + (k_B/e)(E_{\text{gS}})/2k_B T$ represents semiconducting contribution of TEP. The $\sigma_1$ and $\sigma_2$ is the conductivity contribution described in the section 3.2 and $E_{\text{gS}}$ is the Fermi energy. A more simplified version described in [25] can be used here to fit the thermopower data. $B$ and $C$ are the constants referred in [25].

Figure 6 shows the thermopower from $x = 0$ to $x = 0.04$ with TBM fit. The activation energy ($E_{\text{gS}}$) derived by using two band model listed in table 3. The $E_{\text{gS}}$ decreases with increasing Co concentration monotonically, which implies the closure of the gap. The fitted curve deviation below 50 K is due to the thermopower approaches zero faster than linear T. Also, the deviation above 100 K is due to the closure of the energy gap with the temperature. The values of $E_{\text{gS}}$ are different from the $E_{\text{gS}}$. The reason is that the Co substitution moves the Fermi levels toward the conduction band. The $E_{\text{gS}}$ is referred to $E_C - E_F$, the difference between the bottom of the energy level in the conduction band ($E_C$) and the Fermi energy ($E_F$). The $E_{\text{gS}}$ is due to the activated carriers from the impurity band to the conduction band [25].

Figure 7 shows the power factor ($S^2\sigma$) of Co substituted FeSi from $x = 0.1$ to $x = 0.04$ plotted against temperature. The negative thermopower is due to the domination of the conduction band $E_{\text{gS}}$ from TBM fit. The activation energy ($E_{\text{gS}}$) is derived using Nordheim-Gorter rule [25]. The power factor is high, around 30 and 160 K. The highest
power factor in Co-doped FeSi achieved in $x = 0.0175$, just below the critical concentration of MIT ($x_c = 2\%$). This is due to the good balance between the electric conduction from the Co impurity bands and higher thermopower due to the low carrier concentration. Above the $x_c$ or the metal-to-insulator transition, $S^2/\sigma$ decreases because of the increased carrier concentration from Co substitutions.

4. Conclusion

We have investigated the transport and thermoelectric properties of Co-doped FeSi. We concentrated more on the physical properties, which are below the critical concentration of MIT and below $x_c$, where the system is non-magnetic. As far as our knowledge, we are the first to report the detailed thermoelectric properties of Fe$_{1-x}$Co$_x$Si below $x_c$ at low temperatures. The overall resistivity decreases with increasing Co concentration, and we used the TBM to get an insight into the impurity effects. The activation energy decreases with increasing Co concentration, implying that the impurity states form near the conduction band. The positive thermopower around 40 K dramatically increases at $x = 0.0025$ and then decreases with increasing concentration. The thermopower shows a negative below 35 K from $x = 0.01$; implies the impurity electron carrier domination at low temperatures. The overall activation energy derived by using the TBM in thermopower decreases with increasing Co concentration.

Data availability statement

The data generated and/or analysed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request. The data that support the findings of this study are available upon reasonable request from the authors.

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