# Magnetic response of $Fe_{1-x}Co_xSi$ alloys: A detailed study of magnetization and magnetoresistance

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Results of dc magnetization and magnetoresistance measurements are presented for the heliferromagnetic $Fe_{1-x}Co_xSi$ alloys with $x=0.35$ and 0.45. The field and temperature dependence of the magnetization suggests the presence of Stoner-like excitations in these alloys which in turn indicates significant itinerant character of the ferromagnetic state. Similarities with other weak itinerant electron ferromagnets are pointed out. While the relatively large and unusual positive magnetoresistance in the ferromagnetic state of these alloys has earlier been rationalized in terms of quantum interference effect, magnetism probably plays a role in the magnetoresistance of these alloys near the Curie temperature and even at temperatures well inside the paramagnetic regime.

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## I. INTRODUCTION

One of the reasons why the narrow-gap semiconductor FeSi has drawn attention over the last 60 years, is its interesting magnetic properties.[1–3] Magnetic susceptibility shows a broad maximum around 500 K followed by a Curie-Weiss behavior at higher temperatures.[1–3] Previous neutron diffraction,[4] NMR, and Mössbauer measurements[5] could not detect long-range magnetic order in any temperature regime. Jaccarino *et al*.[2] interpreted such a magnetic response of FeSi within a picture of a semiconducting band of negligible bandwidth compared with the band gap. Later on Takahashi and Moriya[6] proposed a spin fluctuation model giving a satisfactory account of the susceptibility and specific heat in FeSi above 100 K. In addition Evangelou and Edwards pointed out the possibility of ferromagnetic correlations in FeSi.[7] Further interest has been generated in the low-temperature magnetic properties of FeSi through a very recent work by Sluchanko *et al*.[8] From a magnetization study involving high-quality FeSi single crystals, these authors have suggested the formation of spin polarons in FeSi at $T<100$ K which are eventually transformed into ferromagnetic microregions at $T_C\approx 15$ K.[8]

It is known for quite sometime now that the Co doping in FeSi gives rise to metalic behavior along with long-range helimagnetic ordering.[9–11] This is in spite of the fact that the end compound CoSi is a diamagnetic semimetal.[12] The discovery of unusual positive magnetoresistance in the $Fe_{1-x}Co_xSi$ alloys has further increased interest in these alloys.[13] In a recent work we have reported magnetic measurements on $Fe_{1-x}Co_xSi$ alloys, highlighting various interesting aspects of the magnetic response in the low-field and low-temperature regime.[14] In the present paper we shall investigate the nature of the induced ferromagnetic states[15] in the $Fe_{1-x}Co_xSi$ alloys in some detail. We shall address to the question of the degree of localization or itinerancy of *d* electrons which are responsible for magnetism in this alloy system. Comparing the present results with the magnetic properties of various other itinerant electron ferromagnets including the isostructural compound MnSi, we shall show the presence of a finite degree of itinerancy in the ferromagnetic character of these Co-doped FeSi alloys. In addition, we shall make an attempt to find a correlation between magnetic properties and interesting magnetoresistance behavior in these alloys.

## II. EXPERIMENT

The samples were prepared by argon arc melting from metals of nominal 99.99% purity and were subsequently annealed for 90 h at 900 °C in vacuo.[14] Magnetization measurements were performed in a commercial superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS-5) using a scan length of 4 cm with 32 data points in each scan. Some important results were checked using a different commercial SQUID magnetometer (Quantum Design) with reciprocating sample option (RSO) and a vibrating sample magnetometer (VSM) (Oxford Instruments). Transverse magnetoresistance measurements were carried out in a 16 T magnet-cryostat system (Oxford Instruments) using a standard dc four-probe technique.

## III. RESULTS AND DISCUSSION

In Fig. 1 we show magnetization ($M$) vs temperature ($T$) plots of the $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ alloys between 2 and 100 K. The applied magnetic field of 50 kOe is well beyond the field for technical saturation ($\approx$1 kOe) in these alloys. The magnetization data could be fitted very well up to temperatures nearing $T_C$ (see Fig. 1) using the following expression incorporating a superposition of collective electron and spin-wave effects:

$$\frac{M(T)-M(0)}{M(0)}=bT^{3/2}+cT^{5/2}+dT^2+\cdots. \qquad (1)$$

Here the leading $T^{3/2}$ term is due to excitations of long-wavelength spin waves,[16] $T^{5/2}$ results from spin-wave–spin-wave interactions,[16] and the $T^2$ term can arise due to Stoner

 

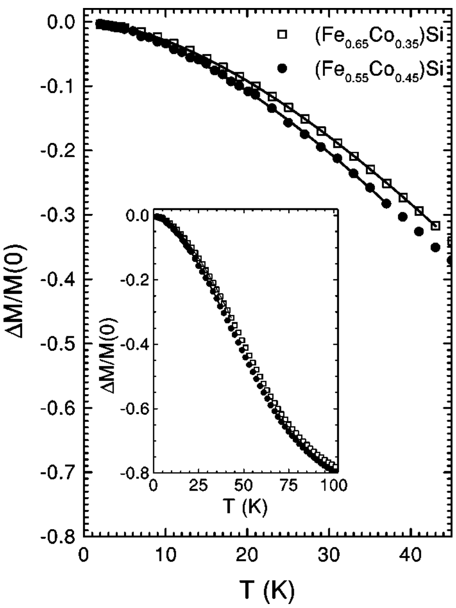


FIG. 1. Temperature dependence of normalized magnetization of $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$. The solid lines represent Eq. (1), and the points represent experimental data.

band excitations.[17] The $T_C$'s of the $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ alloys are 42 and 37 K, respectively (the method of determining $T_C$ is described later on in the paper). We get the magnitudes of *b*, *c*, and *d*, respectively, as 52 $\times 10^{-5}$ $K^{-3/2}$, $1\times 10^{-5}$ $K^{-5/2}$, and $16\times 10^{-5} K^{-2}$ for the $Fe_{0.65}Co_{0.35}Si$ sample and $95\times 10^{-5}$ $K^{-3/2}$, 4.62 $\times 10^{-6}$ $K^{-5/2}$, and $8\times 10^{-5}$ $K^{-2}$ for the $Fe_{0.55}Co_{0.45}Si$ sample. The values of the $T^{3/2}$ term are much larger than the $T^2$ and $T^{5/2}$ terms but the $T^2$ term definitely comes out as the second dominating term. A similar *T* dependence of the saturation magnetization has been observed in the isostructural weak itinerant electron ferromagnet MnSi (Ref. 18) and also in other itinerant ferromagnets like $ZrZn_2$, $Ni_3Al$, Fe-Ni Invar alloys, and NiPt alloys (see Ref. 19 and references therein). The present result indicates that the temperature dependence of the saturation magnetization of the concerned (Fe,Co)Si alloys can be rationalized in terms of spin-wave excitation and a significant contribution coming from Stoner-type excitations.

On the basis of the Stoner model it has been shown[19–21] that for itinerant electron ferromagnets with homogeneous and weak magnetization ($M$), $M$ can be expressed as a function of applied field ($H$) and temperature ($T$) with the help of the following expression:

$$M^2(H,T) = -A/B + B^{-1}H/M(H,T), \qquad (2)$$

where *A* and *B* are the Landau coefficients. Such $M^2(H,T)$ vs $H/M(H,T)$ plots are also known in the literature as Arrott plots.[22] Ideally, for weak homogeneous magnetization, the Landau coefficient *B* should be independent of *T*, leading to parallel $M^2(H,T)$ vs $H/M(H,T)$ plots at different *T*. However, in various real materials—say, for example, Fe-Ni Invar alloys—the Arrott plots are found to be not strictly parallel.[23] The *T* dependence of *B* arises from the fact that the *M* of the Invar alloys was not very weak and thus it was not an ideal case of weak itinerant ferromagnetism.[23] However, it was also argued that as long as the Landau expression for *M* is valid, the slope of the Arrott plots can be expressed as $B^{-1} = 2\chi(0,T)[M^2(0,T)]$, where $\chi(0,T)$ is the magnetic susceptibility.[23,24] Qualitatively similar behavior has been observed in other known weak itinerant ferromagnets like $ZrZn_2$ (Ref. 21), $Ni_3Al$ (Ref. 25 and references within), and Ni-Pt alloys (Ref. 26).

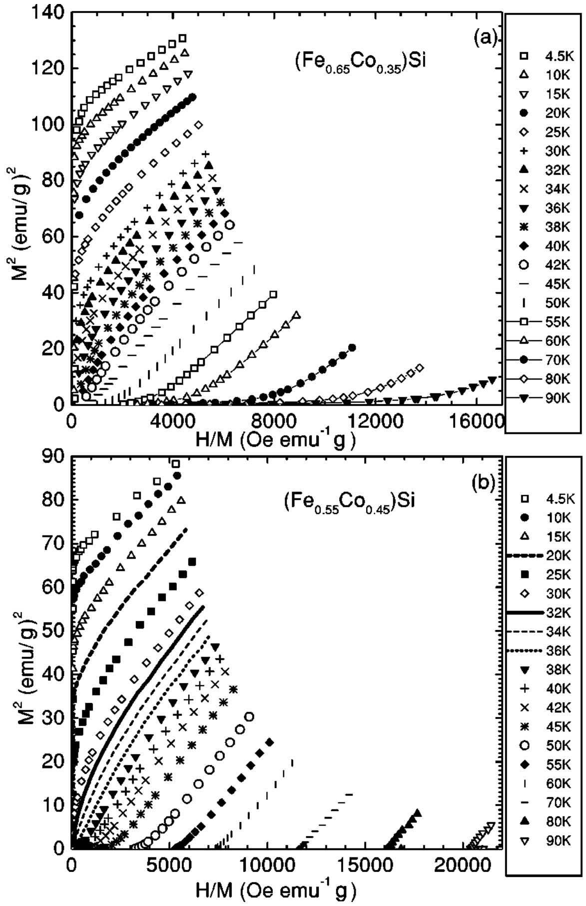


FIG. 2. Arrott plots for $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ obtained from *M* vs *H* data recorded at various constant temperatures.

We shall now relate here Eq. (2) to the experimentally obtained values of *M* for (Fe,Co)Si alloys as a function of *T* and *H* with the aim of assessing the possible role of single-particle excitations. Arrot plots obtained from isothermal *M*-*H* plots (measured at different *T* between 5 and 90 K for magnetic fields 0 Oe$\leq H \leq$50 kOe) for the alloys $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ are shown in Figs. 2(a) and 2(b), respectively. These Arrott plots are straight lines almost parallel to each other over an appreciable range of *T*. They clearly indicate a significantly weak itinerant electron ferromagnetic character of the present alloys. The Curie temperatures ($T_C$'s) determined from these Arrott plots (*T* at which the vertical axis intercept is zero) come out to be 42 and 37 K, respectively, for the $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ alloys. But the $T_C$'s determined from the minima exhibited by $dM/dT$ vs *T* plots are 35 and 30 K (at $H=20$ Oe), respectively, whereas extrapolation from $1/\chi$ vs *T* plots gives $T_C$'s

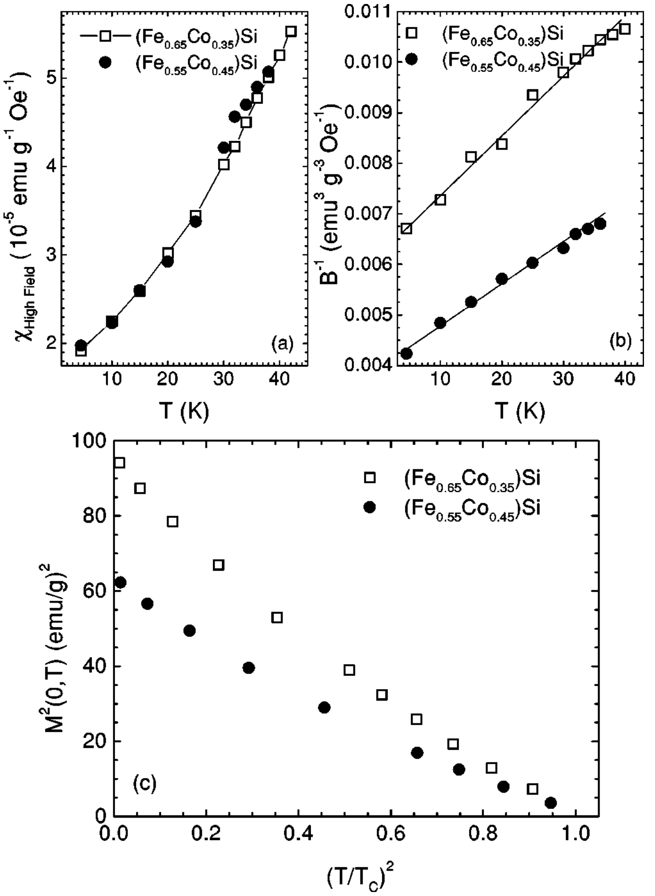


FIG. 3. (a) Temperature dependence of the high-field susceptibility of $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$. (b) Temperature dependence of the slope of the Arrott plots of $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ obtained below $T_C$. (c) Near-linear variation of $M^2(0,T)$ with $(T/T_C)^2$ at temperatures below $T_C$ for $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$.

at 47 and 42 K (at $H=20$ Oe). For the present analysis we choose to use the value of $T_C$ obtained from the Arrott plots.

In Figs. 3(a) and 3(b), respectively, we plot the high-field susceptibility $\chi(H)$ and the slope of the Arrott plots $B^{-1}$ as functions of $T$ for the alloys $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$. While $\chi(H)$ increases quite sharply with increasing $T$, the variation of $B^{-1}$ is roughly linear up to temperatures nearing $T_C$. Very similar characteristics have been observed earlier in Fe-Ni Invar alloys.[23] Again, for ideal weak itinerant ferromagnets, the vertical axis intercepts of the Arrott plots are expected to follow the following relation:[19,20,23]

$$-A/B=M^2(0,T)=M^2(0,0)[1-(T/T_C)^2]. \tag{3}$$

We show the $M^2(0,T)$ vs $(T/T_C)^2$ plots for the present two alloys in Fig. 3(c). $M^2(0,T)$ varies roughly linearly with $(T/T_C)^2$ up to temperatures close to $T_C$. While such a scaling behavior generally points towards the itinerant electron character of the present alloys, the small deviation from linearity might be due to the nonideal $T$ dependence of the Landau coefficient $B$. On a cautious note it can therefore be said that the (Fe,Co)Si alloys are not perfectly weak itinerant ferromagnets. It is to be noted that the evidence of the itinerant electron character in some other $Fe_{1-x}Co_xSi$ alloys was earlier observed in low-$T$ specific heat, NMR, and Mössbauer studies[27] and also in high-pressure magnetization studies.[28]

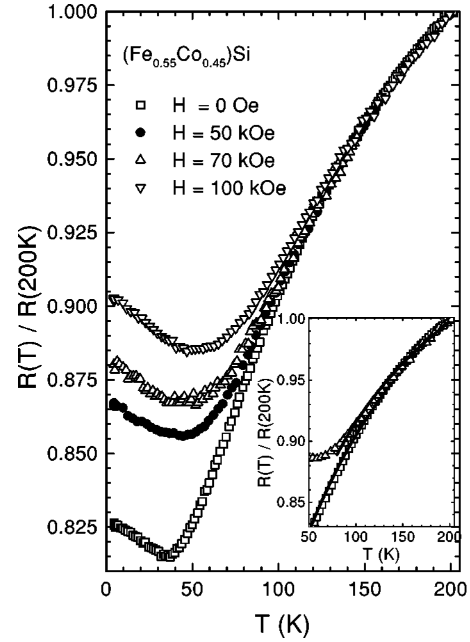


FIG. 4. Temperature dependence of the resistance ratio of $Fe_{0.55}Co_{0.45}Si$ at various applied magnetic fields. The solid lines in the inset represent the results of curve fitting incorporating contributions from spin fluctuations and lattice vibrations (see text for details) for $T>T_C$, for two applied fields $H=0$ and 100 kOe.

Figure 4 shows the $T$ variation of the normalized resistance of the $Fe_{0.55}Co_{0.45}Si$ alloy between 4.5 and 200 K under different applied magnetic fields. For conciseness, we present the results on the $Fe_{0.55}Co_{0.45}Si$ alloy only. The resistance shows typical metallic behavior above $T_C$, but an interesting deviation occurs as $T$ is lowered below $T_C$. In this latter $T$ regime, the resistance tends to rise with a lowering of temperature. Similar behavior in the resistance was reported earlier for the same alloy systems.[10,13] Beille *et al.*[10] attributed this behavior to the *disorder of the repartition of the magnetic moments in the helix due to alloying effects*. Another source may lie in the effect of splitting of the narrow $d$ band (which exists between the Fermi levels of FeSi and CoSi) into two subbands. This might lead to a decrease of the density of states at the Fermi level and cause an increase of resistivity.[10] In our present sample the resistivity has a $T^{1/2}$ temperature dependence well below $T_C$. This might be taken as a signature of a quantum interference effect. A similar resistivity minimum in the *badly metallic* itinerant ferromagnet $SrRuO_3$ was attributed to the inherent tendency of the electronic states to become localized in *bad metals*.[29]

One of the interesting aspects of the magnetic response in (Fe,Co)Si alloys is the relatively large positive magnetoresistance $(MR=[R(H)-R(0)]/R(0))$ observed in these materials.[13] A positive MR has also been observed in $ZrZn_2$ in the $T$ regime below about 10 K which is well below its ferromagnetic transition temperature $(T_C\approx 22\text{ K})$.[30] This

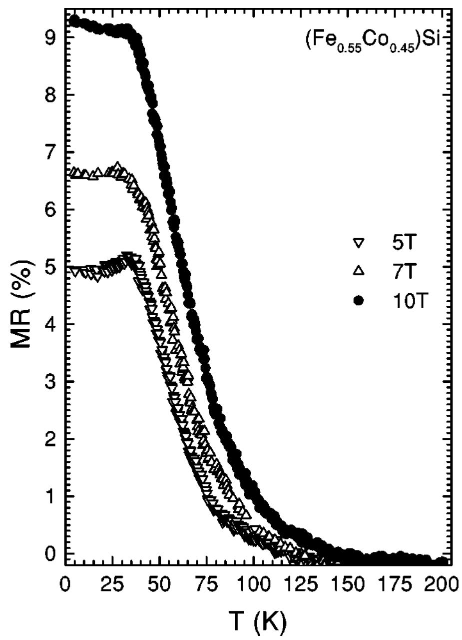


FIG. 5. Temperature dependence of the magnetoresistance of $Fe_{0.55}Co_{0.45}Si$ for different applied magnetic fields.

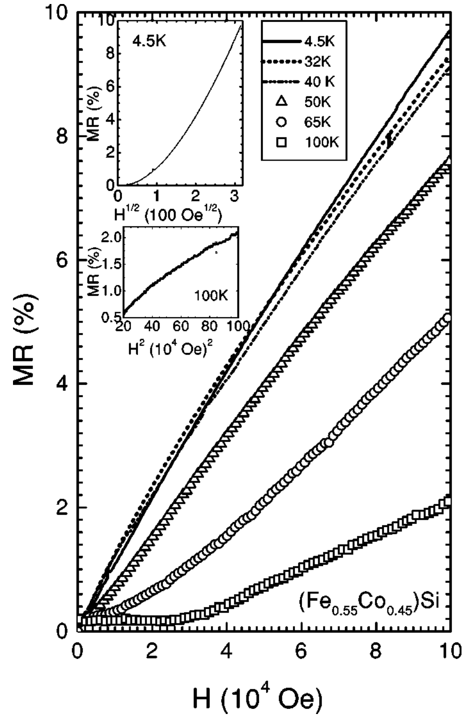


FIG. 6. Magnetic field dependence of the magnetoresistance of and $Fe_{0.55}Co_{0.45}Si$ at different temperatures. The insets show the $H^{1/2}$ and $H^2$ variations of magnetoresistance, in the low- and high-T regimes, respectively.

positive MR in $ZrZn_2$ was thought to be Kohler-type normal MR arising out of the orbital motion of the conduction electrons in a magnetic field. Above 10 K, this is dominated by MR due to spin fluctuations giving an overall negative MR.[30] In the present (Fe,Co)Si system, however, the MR is of a relatively large value and remains positive in the whole $T$ regime right from well below $T_C$ to well above $T_C$ (see Fig. 5). Also the MR shows a small peak and remains weakly temperature dependent in the $T$ regime below $T_C$ [see Fig. 5 and also Fig. 2(b) of Manyala *et al*.[13]]. It is now well known that weak localization and enhanced electron-electron interactions play influential roles in the resistivity and MR of wide variety of disordered conductors.[31,32] A scaling analysis for disordered paramagnets with electron-electron interactions has been successfully applied to understand the field-temperature dependence of the resistivity across the metal-insulator transition in P-doped Si (Ref. 33). Manyala *et al*.[13] extended a similar scaling analysis for (Fe,Co)Si ferromagnets by adding a contribution of the internal field of the ferromagnet to the field parameter and obtained good agreement with their magnetotransport data. We did not meet with much success with a similar scaling analysis of our MR data, but this can probably be attributed to a lack of data in the low-$T$ regime ($T<4$ K). However, in the isothermal field variation of MR for the present $Fe_{0.55}Co_{0.45}Si$ alloy (see Fig. 6), an asymptotic $H^{1/2}$ is observed in the high-field ($H>50$ kOe) and low-temperature ($T \ll T_C$) regime (see the inset of Fig. 6). Positive MR with such high-field asymptotic $H^{1/2}$ behavior is indicative of a quantum interference effect (Refs. 13 and 32, and references therein).

While a negative MR is usually expected in systems with long-range magnetic order due to the suppression of spin fluctuations by the external magnetic field, the occurrence of positive MR in magnetic systems is not very uncommon. Relatively large positive MR has been reported in various ordered antiferromagnetic compounds[34] and in ferromagnetic compounds with hints of antiferromagnetic fluctuations.[35,36] Even in elemental rare-earth Nd, which has a complicated magnetic structure, a positive magnetoresistance has been observed at low $T$.[37] Yamada and Takada[38] and Balberg[39] have theoretically discussed the possibility of positive MR in systems with antiferromagnetic correlations. The helical spin structure of the present (Fe,Co)Si alloys arises via Dzyaloshinski-Moriya interactions[10,11] and indicates some kind of competition between ferromagnetism and antiferromagnetism. Hence the existence of some antiferromagnetic correlations in these alloys is not unexpected,[40] and their contributions to the positive MR cannot be ruled out entirely. Similar antiferromagnetic fluctuations in the band ferromagnet $CeFe_2$ (Ref. 41) is possibly the cause for relatively large positive MR in that compound[36] at low temperatures.

For our present $Fe_{0.55}Co_{0.45}Si$ sample the $H$ dependence of MR in the $T$ regime below $T_C$ (37 K) can roughly be fitted with an expression $aH+bH^{1/2}$ with the linear term dominating more and more with the increase in $T$. A deviation from this behavior takes place with further increase in $T$ across $T_C$. However, a $H^2$ dependence of normal (Kohler) MR is observed only in the $T$ regime around 100 K which is well above the $T_C$ of the alloy in question (see the inset of Fig. 6). It is clear from Fig. 6 that a substantial contribution to positive MR remains above $T_C$ from sources other than the standard Lorentz contribution. With less likelihood of a quantum interference effect taking place above $T_C$, this source may be of magnetic origin.

In various magnetically ordered systems a definite corre-

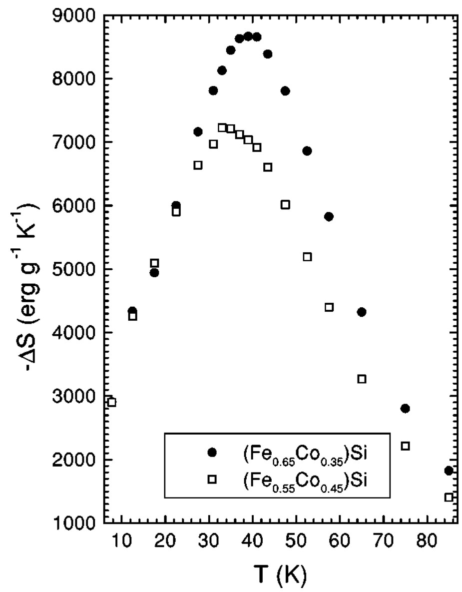


FIG. 7. Temperature dependence of the change of magnetic entropy of $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ estimated from isothermal $M$ vs $H$ data.

lation can actually be found between $M$ and MR (see Ref. 42 and references therein). In our present (Fe,Co)Si alloys no definite correlation between $M$ and MR could be found in the low-$T$ regime below roughly $0.8T_C$. However, as we approach and go across $T_C$ an $M^n$ dependence of MR is observed with $n$ varying from 3 to 1 as $T$ goes from 32 to 65 K. Again, in the higher-$T$ regime, roughly above $1.5T_C$, no straightforward correlation could be found between MR and $M$. This along with the rather slow decrease of MR above $T_C$ (see Fig. 5) further adds to the idea of a possible contribution to the MR coming from magnetic fluctuations in this $T$ regime. It is important to note here that in the same $T$ regime above $T_C$, $M$ shows substantial nonlinearity in its field dependence, indicating the presence of short-range correlation and spin fluctuations. To investigate this matter further we have estimated the change in magnetic entropy $[\Delta S(T)]$ at various $T$ both below and above $T_C$ from our magnetization data. $\Delta S(T)$ is related to the change in bulk magnetization as a function of $T$ and $H$ and is expressed as[43]

$$\Delta S(T) = \int_{H1}^{H2} \left[ \frac{\partial M(T,H)}{\partial t} \right]_H dH. \quad (4)$$

The $\partial M/\partial T$ vs $H$ plots are generated at various $T$ from isothermal $M$-$H$ curves. $\Delta S(T)$'s are obtained by integrating the area under this curve and are plotted for $Fe_{0.65}Co_{0.35}Si$ and $Fe_{0.55}Co_{0.45}Si$ alloys as functions of $T$ in Fig. 7. While $\Delta S$ shows a maximum at $T_C$, it retains a significant value even in the $T$ regime up to $2T_C$. Stoner excitations cannot explain such a magnetic response above $T_C$. However, this is still consistent with the picture of itinerant electron ferromagnetism as the spin fluctuation theory proposed by Moriya[44] has shown that magnetic excitations can persist in weak itinerant ferromagnets in temperatures well above $T_C$. The observation of paramagnetic spin fluctuations in neutron scattering measurements in MnSi (Ref. 45) and in Ni-Fe Invar alloys (Ref. 46) have supported strongly the validity of the spin fluctuation theory proposed by Moriya. It should be noted here that a significantly large value of MR is observed in the paramagnetic regime well above $T_C$ in MnSi as well.[13] However, in contrast to the (Fe,Co)Si alloys the MR in MnSi is negative in nature.[13] It is important to note here that a neutron scattering study has revealed that spin correlations in the paramagnetic state of MnSi can effectively be treated as that of a ferromagnetic material.[47] Hence negative MR in the paramagnetic state of MnSi can be explained in terms of the suppression of spin fluctuations by an external magnetic field. In this respect a non-Kohler-type positive MR of (Fe-,Co)Si alloys remains a puzzle to be solved. It will now be interesting to probe the exact nature of the spin correlations in the paramagnetic state in relation to the helimagnetic state of (Fe,Co)Si alloys with neutron scattering measurements.

The possible presence of spin fluctuations in FeSi and $Fe_{0.95}Co_{0.05}Si$ has actually been suggested in some previous reports.[10,48] The $T$ dependence of the normalized resistance of the present samples above $T_C$ can be expressed in terms of contributions due to spin fluctuations,[49] lattice vibration,[50] and a $T$-independent term. The results of such curve fittings for $H=0$ Oe and $H=100$ kOe, for the $Fe_{0.55}Co_{0.45}Si$ sample, are shown in the inset of Fig. 4. [Data for the other applied fields and for the other sample ($Fe_{0.65}Co_{0.35}Si$) are not presented here for the sake of conciseness.] While the calculated phonon component came out nearly linear in the $T$ regime considered here, the deviation from linearity at temperatures above $T_C$ seems to result from the scattering of electrons by spin fluctuations.

In our previous work[14] we have reported that the $M$-$H$ curves in the (Fe,Co)Si alloys are reversible in the field regime above the field for technical saturation ($H_{sat}$) and an anomalous kind of inverted $M$-$H$ loop opens up in the field regime below $H_{sat}$. However, there are certain questions that remained unanswered.

(1) Can such inverted loops occur due to some contrived history effects of the superconducting magnets?

(2) Since the area of the hysteresis loop represents the energy dissipation per cycle, does the inverted hysteresis loop violate the second law of thermodynamics[51]?

Superconducting magnets can indeed show history effects due to the persistent currents circulating inside the filaments of a superconducting cable.[52] This is now a known problem for accelerator magnets and studied in quite detail during last decade.[52] However, these persistent currents actually cause a damping effect during the change of field. Experimental artifacts caused by this will generate a positive hysteresis loop in a magnetic sample with otherwise reversible response. Another possible source is the proximity effect coupling in the low-field response of the superconducting magnets.[53] This will also generate a positive hysteresis loop as an experimental artifact. Hence such history effects cannot provide an explanation of the low-$H$ low-$T$ inverted hysteresis loop observed in (Fe,Co)Si alloys.

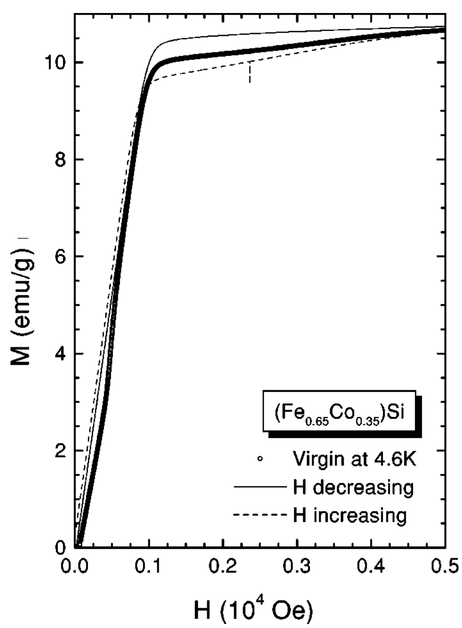


FIG. 8. Magnetization vs field data for $Fe_{0.65}Co_{0.35}Si$ recorded in the VSM showing negative hysteresis in the field regime below technical saturation and positive hysteresis above it.

We have now measured our (Fe,Co)Si samples using a VSM and SQUID magnetometer with RSO option. In the VSM measurements we have used a slow sweep rate of 0.01 T/min and an amplitude of vibration of 1 mm. The inverted nature of the low-field $M$-$H$ loop is clearly observed in these measurements. However, in the VSM measurements after the closure of the inverted loop a positive $M$-$H$ loop is observed in the $H$ regime above $H_{sat}$ (see Fig. 8). In the higher-$H$ regime this positive $M$-$H$ loop closes and the magnetization becomes reversible. The above-mentioned intermediate-field positive $M$-$H$ loop was not observed in our previous measurements[14] with SQUID magnetometers where we have used a scan length of 4 cm. We now find that on varying the scan length, while the lower-field inverted hysteresis loop does not change qualitatively, for intermediate-field values a positive hysteresis loop is observed clearly in SQUID magnetometer measurements with a 2 cm scan length.[54] The low-field inverted hysteresis loop is probably associated with the low-field state of the (Fe, Co)Si alloys which has both clockwise and counterclockwise helixes.[11] On the other hand, a positive hysteresis loop above $H_{sat}$ is characteristic of the field-induced helical state with clockwise helix,[11] and together they make the complete $M$-$H$ loop in (Fe,Co)Si alloys consistent with the second law of thermodynamics. It might not be totally out of place to mention here that an $M$-$H$ loop with small positive hysteresis has recently been observed both in polycrystal[54] and single-crystal[55] samples of pure FeSi below 10 K.

## IV. CONCLUSION

One of the interesting questions on the physical properties of (Fe,Co)Si alloys is whether the magnetic properties of these alloys are related to the electron involved in the transport properties. An early work provided a negative answer to this question.[56] A subsequent study of magnetization, specific heat, Mössbauer, and NMR, however, clearly suggested that the *magnetic electrons are itenerant in nature* and the onset of ferromagnetism in (Fe,Co)Si alloys could be understood in terms of the Stoner model.[27] A more recent study of polarized neutron scattering in (Fe,Co)Si alloys has also emphasized the itinerant electron character of ferromagnetism in this alloy system.[57] Our detailed magnetization and magnetotransport study supports these latter views and suggests that the induced ferromagnetic state in (Fe,Co)Si alloys is indeed itinerant in nature. We have found enough evidence of Stoner-type excitations in the $H$-$T$ dependence of magentization of these alloys. In addition there exist signatures of spin fluctuations and magnetic short-range order in our magnetization, magnetoresistance, and magnetocaloric effect measurements in the temperature regime well above $T_C$, this behavior can be rationalized using Moriya's theory of spin fluctuations.[44] In support of the earlier work of Manyala *et al.*,[13] we have found some evidence of a quantum interference effect in the magnetoresistance measurements at low $T$. However, it seems that magnetic properties also influence the magnetoresistance in the (Fe,Co)Si alloys especially near and above $T_C$, and well inside the paramagnetic regime. Overall it appears that the magnetism of (Fe,Co)Si alloys does play a role in the transport properties, but this role is more complicated than that in the isostructural itinerant ferromagnet MnSi. We believe the present results will provide some pointers towards a proper understanding of the interesting magnetic and transport properties of (Fe,Co)Si alloys.

[1] G. Foex, J. Phys. Radium **9**, 37 (1938); R. Benoit, J. Chem. Phys. **52**, 119 (1955).
[2] V. Jaccarino, G.K. Wertheim, J.H. Wernick, L.R. Walker, and S. Arajs, Phys. Rev. **160**, 476 (1967).
[3] G. Aeppli and Z. Fisk, Comments Condens. Matter Phys. **16**, 155 (1992); G. Aeppli and J.F. DiTusa, Mater. Sci. Eng., B **63**, 119 (1999) (and references therein); K. Koyama, T. Goto, T. Kanomata, and R. Note, J. Phys. Soc. Jpn. **68**, 1693 (1999) (and references therein).
[4] H. Watanabe, Y. Yamamoto, and K. Ito, J. Phys. Soc. Jpn. **18**, 995 (1963).
[5] G.K. Wertheim, V. Jaccarino, J.H. Wernick, J.A. Seitchik, H.J. Williams, and R.C. Sherwood, Phys. Lett. **18**, 89 (1965).
[6] Y. Takahashi and T. Moriya, J. Phys. Soc. Jpn. **46**, 1451 (1979).
[7] S.N. Evangelou and D.M. Edwards, J. Phys. C **16**, 2121 (1983).
[8] N.E. Sluchanko, V.V. Glushkov, S.V. Demishev, A.A. Menovsky, L. Weckhuysen, and V.V. Moshchalkov, Phys. Rev. B **65**, 064404 (2002).
[9] J. Beille, J. Voiron, F. Towfiq, M. Roth, and Z.Y. Zhang, J. Phys. F: Met. Phys. **11**, 2153 (1981).
[10] J. Beille, J. Voiron, and M. Roth, Solid State Commun. **47**, 399 (1983).

[11] K. Ishimoto, Y. Yamaguchi, S. Mitsuda, M. Ishida, and Y. Endoh, J. Magn. Magn. Mater. **54-57**, 1003 (1986).
[12] J.H. Wernick, G.K. Wertheim, and R.C. Sherwood, Mater. Res. Bull. **7**, 1431 (1972).
[13] N. Manyala, Y. Sidis, J.F. DiTusa, G. Aeppli, D.P. Young, and Z. Fisk, Nature (London) **404**, 581 (2000).
[14] M.K. Chattopadhyay, S.B. Roy, and Sujeet Chaudhary, Phys. Rev. B **65**, 132409 (2002).
[15] The (Fe,Co)Si alloys are actually helimagnetic in nature with a very long period (see Ref. 11 and 12). Since the carrier mean paths in these alloys are much shorter than the helix period, for practical purposes these alloys can be treated as ferromagnets. While multiple helixes are observed in the low-field regime in the zero-field-cooled state, a single clockwise helix is stabilized with an applied magnetic field of about 1–2 kOe. We shall mainly be dealing with this field-induced state and henceforth call it a ferromagnet.
[16] B.E. Argyle, S.H. Charap, and E.W. Pugh, Phys. Rev. **132**, 2051 (1963).
[17] C.L. Chien and R. Hasegawa, Phys. Rev. B **16**, 2115 (1977).
[18] M. Matsunaga, Y. Ishikawa, and T. Nakajima, J. Phys. Soc. Jpn. **51**, 1153 (1982).
[19] E.P. Wohlfarth, Physica B **91**, 305 (1977) (and references therein).
[20] D.M. Edwards and E.P. Wohlfarth, Proc. R. Soc. London, Ser. A **A 303**, 127 (1968).
[21] E.P. Wohlfarth, J. Appl. Phys. **39**, 1061 (1968).
[22] A. Arrott and J.E. Noakes, Phys. Rev. Lett. **19**, 786 (1967).
[23] O. Yamada, F. Ono, and I. Nakai, Physica B **91**, 298 (1977).
[24] D.L. Mills, Solid State Commun. **9**, 929 (1971).
[25] P.E. Brommer and J.J.M. Franse, in *Ferromagnetic Materials*, edited by K.H.J. Bushow and E.P. Wohlfarth (Elsevier Science, New York, 1996), Vol. 5, p. 323.
[26] H.L. Alberts, J. Beille, D. Bloch, and E.P. Wohlfarth, Phys. Rev. B **9**, 2233 (1974).
[27] S. Kawarazaki, H. Yasuoka, Y. Nakamura, and J.H. Wernick, J. Phys. Soc. Jpn. **41**, 1171 (1976).
[28] J. Beille, D. Bloch, V. Jaccarino, J.H. Wernick, and G.K. Wertheim, J. Phys. (Paris) **38**, 339 (1977).
[29] L. Klein, J.S. Dodge, C.H. Ahn, J.W. Reiner, L. Mieville, T.H. Geballe, M.R. Beasley, and A. Kapitulnik, J. Phys.: Condens. Matter **8**, 10111 (1996).
[30] S. Ogawa, Physica B **91**, 82 (1977).
[31] P.A. Lee, and T.V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[32] J.S. Dugdale, *The Electrical Properties of Disordered Metals* (Cambridge University Press, New York, 1995).
[33] T.F. Rosenbaulm, R.F. Milligan, M.A. Paalaren, G.A. Thomas, R.N. Bhatt, and W. Lin, Phys. Rev. B **27**, 7509 (1983).
[34] C. Mazumdar, A.K. Nigam, R. Nagarajan, C. Godart, L.C. Gupta, B.D. Padalia, and R. Vijayraghavan, Appl. Phys. Lett. **68**, 3647 (1996).
[35] A.K. Nigam, S.B. Roy, and P. Chaddah, Phys. Rev. B **60**, 3002 (1999).
[36] S. Radha, S.B. Roy, and A.K. Nigam, J. Appl. Phys. **87**, 6803 (2000).
[37] H. Nagasawa, Phys. Lett. **41A**, 39 (1972).
[38] H. Yamada and S. Takada, Prog. Theor. Phys. **48**, 1828 (1972); J. Phys. Soc. Jpn. **34**, 51 (1973).
[39] I. Balberg, Physica B **91**, 71 (1977).
[40] K. Makoshi and T. Moriya, J. Phys. Soc. Jpn. **44**, 80 (1978).
[41] L. Paolasini, P. Dervenagas, P. Vulliet, J.P. Sanchez, G.P. Lander, A. Heiss, A. Panchula, and P. Canfield, Phys. Rev. B **58**, 12 117 (1998).
[42] T.K. Nath and A.K. Majumdar, Phys. Rev. B **57**, 10655 (1998).
[43] V.K. Pecharsky, K.A. Gschneidner, A.O. Pecharsky, and A.M. Tishin, Phys. Rev. B **64**, 144406 (2001).
[44] T. Moriya, J. Phys. Soc. Jpn. **51**, 420 (1982); *Spin Fluctuations in Itinerant Electron Magnetism* (Springer, Berlin, 1985).
[45] Y. Ishikawa, Y. Noda, Y.J. Uemara, C.F. Majkrazak, and G. Shirane, Phys. Rev. B **31**, 5884 (1985).
[46] U. Tajima, P. Boni, G. Shirane, Y. Ishikawa, and M. Kohgi, Phys. Rev. B **35**, 274 (1987).
[47] Y. Ishikawa, Y. Noda, C. Fincher, and G. Shiratic, Phys. Rev. B **25**, 254 (1982).
[48] Y. Takahashi, J. Phys.: Condens. Matter **10**, L671 (1998).
[49] N. Rivier and V. Zlatic, J. Phys. F: Met. Phys. **2**, L99 (1972). P. L. Rossiter, *The Electrical Resistivity of Metals and Alloys* (Cambridge University Press, New York, 1991), p. 339.
[50] J. M. Ziman, *Principles of the Theory of Solids* (Cambridge University Press, Cambridge, England, 1992), pp. 223–225.
[51] Y.Z. Wu, G.S. Dong, and X.F. Jin, Phys. Rev. B **64**, 214406 (2001).
[52] B.J. Holzer (unpublished).
[53] E.W. Collings, K.R. Marken, Jr., M.D. Sumption, E. Gregory, and T.S. Kreilick, Adv. Cryg. Eng. **36**, 231 (1990).
[54] M. K. Chattopadhyay *et al.* (unpublished).
[55] N.E. Sluchanko, V.V. Glushkov, S.V. Demishev, M.V. Kondrin, V.U. Ivanov, K.M. Petukhov, N.A. Samarin, A.A. Menovsky, and V.V. Moschalkov, JETP **92**, 312 (2001).
[56] G.K. Wertheim, J.H. Wernick, and D.N.E. Buchanan, J. Appl. Phys. **37**, 3333 (1964).
[57] K. Ishimoto, M. Ohashi, H. Yamauchi, and Y. Yamaguchi, J. Phys. Soc. Jpn. **61**, 2503 (1992).