Magnetoresistance and its correlation with magnetization in γ-$Fe_{80-x}Ni_xCr_{20}$ ($14 \leqslant x \leqslant 30$) alloys near the multicritical point

T. K. Nath and A. K. Majumdar
Department of Physics, Indian Institute of Technology, Kanpur 208016, India
(Received 21 May 1997; revised manuscript received 19 August 1997)

A comprehensive study of low- (0–16.5 kOe) and very high-field (0–200 kOe) magnetoresistance $\Delta\rho/\rho(H,T)$ and magnetization $M(H,T)$ has been carried out between 4.2 and 300 K on substitutionally disordered γ-$Fe_{80-x}Ni_xCr_{20}$ ($14 \leqslant x \leqslant 30$), austenitic stainless-steel alloys near the multicritical point. Interpretation of magnetoresistance (MR) data and correlation between MR and magnetization have been emphasized in this paper. All the alloys in the low-field regime show a negative (anomalous) MR below 50 K, except for the antiferromagnetic (AFM) alloy with $x=14$. In the latter the MR is positive at lower fields but becomes negative beyond the *spin-flop* transition due to the canting of the AFM spins. Above 50 K all of them show Kohler-type (quadratic in H) normal MR. In the low-field regime below 50 K, the MR data of all the alloys (except the AFM) are well described by an empirical relation $\Delta\rho/\rho = -\alpha(T)H^n$ with distinct values of α and n for the different magnetic phases. In the case of the spin-glass (SG) alloy ($x=21$), the MR correlates very well with M as $\Delta\rho/\rho \propto M^{2.5}$ over a wide range of temperature and field. For the SG alloys ($x=19$ and 21) with no long-range ordering the isotropic MR $[(\Delta\rho/\rho)_{\rm iso}]$ and M satisfy an empirical relation $[(\Delta\rho/\rho)_{\rm iso}] = -\alpha[M^2(H)]$. As the system approaches the long-range ferromagnetic (FM) ordering passing through the re-entrant spin-glass (RSG) regime, a distinct deviation from the above relation has been observed. Finally, in the case of the long-range FM alloy ($x=30$), the relationship of $[(\Delta\rho/\rho)_{\rm iso}] = -\alpha[M^2(H) - M^2(0)]$ holds well. The observation of the upturn in the negative MR in these Fe-Ni-Cr alloys at low temperatures and in very high fields beyond 40–50 kOe has been attributed to the positive (normal) contribution having a quadratic field dependence $[(\Delta\rho/\rho)_n \sim aH^2]$. The negative (anomalous) contribution is found to appear mainly from the suppression of the spin-flip scattering in all the magnetic states. At very high fields, deviation from the simple quadratic dependence of the negative MR on M has been observed in the concentrated SG ($x=19$) alloy. The contribution to MR from *quantum interference effects* is found to be very small compared to the other contributions. [S0163-1829(98)00514-1]

I. INTRODUCTION

The magnetotransport study [magnetoresistance (MR)] of metallic alloys is a powerful probe of its electronic transport processes. Generally in metals and alloys with weak potential scattering, the dominant contribution to the MR is essentially a band effect. This is positive, quadratic in field, and follows within certain limitations Kohler's rule[1] [$\Delta\rho/\rho_0 = f(H/\rho_0)$]. In nonmagnetic amorphous alloys one generally sees a negative MR arising from the suppression of *weak localization*.

The magnetoresistance of a magnetic system is one such probe where the transport properties are coupled with the magnetic ones. It is a convenient tool to investigate local spin correlations which otherwise get averaged out in the bulk magnetic measurements such as susceptibility, magnetization, etc. It is also to be noted that unlike the bulk magnetic measurements, *MR is less sensitive to domain effects if their size is greater than the mean free path*. The isotropic MR of magnetic alloys is predicted to be sensitive only to changes in the magnetic correlations on a scale of the order of the electron mean free path (l), whereas the magnetization depends on both short and long-range ferromagnetic (FM) orders as well as on the rotation of the FM domain in response to an applied field. Therefore, the study of MR of magnetic materials can provide very useful and complementary information in relation to magnetization on a semimicroscopic scale ($\sim l$).

In the case of *canonical* spin glasses [CuMn,[2,3] AuFe,[4] AuCr, and AgMn (Refs. 2,5) with (0.5–10) at. % of $3d$ impurities] a general correlation

$$\frac{\rho(H,T)-\rho(0,T)}{\rho(0,T)} = \frac{\Delta\rho}{\rho_0} = -\beta M^2 = -\beta\chi_0^2 H^2 \qquad (1)$$

was obtained in a wide range of magnetic field (H) and temperature (T) below their spin-freezing temperatures. It has been argued by Senoussi[4] that an M^2 dependence of the MR supports a homogeneous model for the magnetization in spin glasses (SG). In an investigation of the correlation between MR and magnetization M in AgMn and AuMn at intermediate fields, Majumdar[5] obtained a good agreement of the data with Mookerjee's model[6] for the MR of spin glass, given by

$$\frac{\Delta\rho}{\rho_0} = -\left(\frac{J^2}{V^2}\right)\left[M\,\tanh\left(\frac{g\mu_B H}{2kT}\right) + 2\gamma M^2\right]. \qquad (2)$$

Here V and J are the Coulomb and s-d exchange potentials, g is the splitting factor, μ_B is the Bohr magneton, and γ is a constant. This model was derived within the framework of the Edwards-Anderson model where the local spins interact through the conduction electrons via the s-d exchange cou-

 © 1998 The American Physical Society

pling, neglecting the complication of spin dynamics. Sherlekar *et al.*[7] computed the MR of ternary spin glasses by including higher-order terms and obtained a better agreement with experiments as compared with Mookerjee's theory.

Generally the alloys around the percolation threshold have strong competing interactions and they are found to pass on to a phase where long-range order coexists with the spin-glass ordering, as predicted by Gabay and Toulouse.[8] The irreversible effects associated with the magnetization of spin glasses are also observed in this phase. MR studies reveal that the slope of MR $[(1/\rho)(d\rho/dH)]$ increases as the system enters a mixed phase[9,10] from a FM one. This increase is associated with the increase of random, uncorrelated components and the breakdown of FM order which results in an increase of high-field susceptibility. A number of reports on AuFe alloys with higher Fe concentration (>13 at. %) (Refs. 2,9,11–13) confirmed the coexistence of both the SG and the FM phases in the re-entrant phase through studies of MR, susceptibility, etc. NiMn alloys[10,14] also show the re-entrant behavior below 25 at. % of Mn (percolation threshold).

The commonly observed low-field ferromagnetic anisotropy of resistivity (FAR) in different crystalline and amorphous FM alloys have been explained well by Smit's model[15] which is based on spin-orbit interaction and s-d scattering incorporating parallel current conduction. This model has been extensively used by Cambell, Fert, and Jaol[16] to derive a relation which holds for many crystalline Ni-based alloys. At high fields the suppression of scattering due to spin waves leads to a reduction in resistivity and consequently a negative MR above their technical saturation.

There have been very few MR measurements in antiferromagnets, possibly because of their small values, which in some cases may be beyond the resolution of measuring instruments. Nagasawa[17] had observed that Nd, which has a complicated spin ordering with two antiferromagnetic phases at low temperatures, shows a positive magnetoresistance at sufficiently low temperatures whereas as the temperature rises it becomes negative at the temperature where one of the antiferromagnetic phases disappears. This suggests that in such metals with oscillatory spin orderings the magnetic field does not simply suppress the fluctuations of spins, but may as well increase it leading to a positive MR. Yamada and Takada[18] have calculated the MR of antiferromagnetic (AFM) metals due to electron-spin scattering with localized spins through s-d interaction, using the molecular-field approximation. For AFM's the direction of external magnetic field is taken to be parallel or perpendicular to the *staggered magnetization*, and the latter (perpendicular) case includes the *spin-flop state*. The external magnetic field is assumed to be weak ($\omega_c\tau \leqslant 1$). They have shown that the MR is positive in the AFM state when the field is parallel to the sublattice magnetization in contrast to the negative MR in the FM and paramagnetic (PM) cases. In the FM and PM cases the magnetic field increases the effective field acting on the localized spins and suppresses the fluctuation of spins which leads to a negative MR. On the other hand, in an AFM state with parallel field, the fluctuations of the spins of one sublattice may be suppressed whereas it might increase in the other. The MR is determined by the net spin fluctuations in the two sublattices, and this may lead to a positive MR. For an AFM the MR is zero when the applied magnetic field is perpendicular to the sublattice magnetization.

In this work we have carried out systematic measurements of longitudinal $[(\Delta\rho/\rho)_{\parallel}](H,T)$ and transeverse $[(\Delta\rho/\rho)_{\perp}](H,T)$ magnetoresistances and magnetization $M(H,T)$ in low (0–16.5 kOe) and very high (0–200 kOe) field ranges at temperatures between 4.2 and 300 K in γ-$FE_{80-x}Ni_xCr_{20}$ ($14 \leqslant x \leqslant 30$) austenitic stainless-steel alloys in the fcc γ phase. The magnetic phase diagram[19,20] had been established in this alloy system through dc magnetization,[19] magnetic neutron scattering,[21] and ac-susceptibility measurements. Due to the strong competing exchange interaction between different kinds of $3d$ transition-metal magnetic atoms (nearest-neighbor interaction is either FM or AFM with positive or negative values of exchange integral, J_{ij}), this system of alloys undergoes a compositional phase transition from long-range AFM ($x = 10-14$) to SG (17–21), to mixed FM and SG (23–26), to long-range FM ($x \geqslant 30$) order within the same crystallographic γ phase. In the mixed phase alloys ($x = 23-26$), the coexistence of long-range FM and SG ordering was confirmed through magnetization[19,22] and magnetoresistance[22] measurements. This is in agreement with the Gabay-Toulouse[8] model of mixed phase.

Magnetoresistance (MR) measurements are supposed to throw new light on or confirm the proposed magnetic phase diagram in the sense that each of the above phases has characteristic field, temperature, and orientation dependence of MR. Conversely, the present system of concentrated alloys forms an interesting set where one could study the detail MR behavior of almost continuously varying magnetic phases originating from competing interactions within the same chemical constituents and crystal structure.

We have reported earlier preliminary low-field magnetoresistance measurements[22] on a few of these alloys ($x = 30$, 23, and 21) in the field range of 0–16.5 kOe without any detailed analysis. In this report we extend the low-field MR and magnetization measurements to $x = 26$ [another re-entrant spin glass (RSG)], 19 (pure SG) and 14 (AFM). Also, high-field (till 20 T) magnetoresistance data in the RSG ($x = 26$) and SG ($x = 19$) are presented, analyzed, and also correlated with the corresponding magnetization data already reported.[23] The motivation and emphasis of the present work are on meaningful correlations between magnetotransport and magnetic properties in different magnetic phases. Some important new results are as follows:

(a) Functional relations $\Delta\rho/\rho = -\alpha(T)H^n$ between MR and H with distinct values of α and n have been found for different magnetic phases (except the AFM).

(b) For the AFM ($x = 14$), the positive LMR at lower fields is correlated with the *spin-flop* transition observed in $M(H)$ at 10 kOe.

(c) For the SG ($x = 21$), $\Delta\rho/\rho \propto M^{2.5}$ over a wide range of H and T. The isotropic MR for the SG's ($x = 21$ and 19) goes as $[(\Delta\rho/\rho)_{\rm iso}] = -\alpha[M^2(H)]$ whereas for the FM ($x = 30$) $[(\Delta\rho/\rho)_{\rm iso}] = -\alpha[M^2(H) - M^2(0)]$ holds well. The RSG's ($x = 23$ and 26) satisfy the latter only roughly.

(d) The very high-field (20 T) MR has normal positive and anomalous (magnetic) negative contributions. The magnetic part of the MR in the SG ($x = 19$) does not correlate

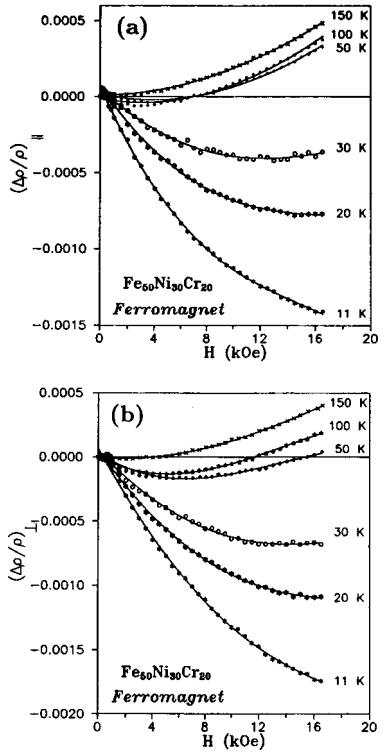

FIG. 1. Magnetic-field (H) dependence of (a) longitudinal $[(\Delta\rho/\rho)_{\parallel}]$ and (b) transverse $[(\Delta\rho/\rho)_{\perp}]$ magnetoresistances at various temperatures for $Fe_{50}Ni_{30}Cr_{20}$ (FM) alloy.

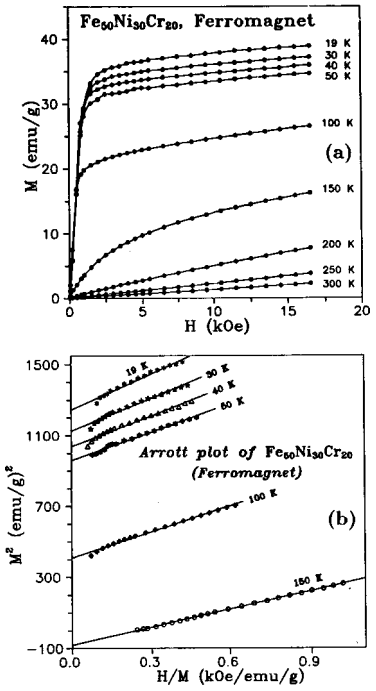

FIG. 2. (a) M-H isotherms of $Fe_{50}Ni_{30}Cr_{20}$ (FM) alloy at various temperatures in the field range of 0–16.5 kOe. (b) Arrott plots at various temperatures for $Fe_{50}Ni_{30}Cr_{20}$ (FM) alloy confirming the presence of large spontaneous moment.

with M^2 (as in *canonical* SG's). The contribution to the MR from quantum interference effects is found to be negligible.

II. EXPERIMENTAL DETAILS

Sample preparation and characterization, low-field magnetoresistance and dc magnetization measurement techniques have been described elsewhere.[22] The very high-field (till 20 T) magnetoresistance $[\Delta\rho/\rho(H)]$ measurements were carried out in the temperature range between 4.2 and 60 K using the standard ac technique and a water-cooled Bitter coil magnet (2-T and 2-Y types) of 52.5 mm bore and a 10 MW power supply at the Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology. The data acquisition is done through a PC/AT in both continuous scanning increasing and decreasing field modes. The temperature of the sample was measured by a calibrated carbon resistor and was controlled by a glass-ceramic capacitance sensor and matching electronics. The stability of temperature during the measurements was within ± 100 mK and the resolution in $\Delta\rho/\rho$ was about 1 part in 10^4.

III. RESULTS AND DISCUSSION

A. General features and comparative study of low-field magnetoresistance and magnetization

(a) In Figs. 1(a) and 1(b) we have shown, respectively, the longitudinal (LMR) $[(\Delta\rho/\rho)_{\parallel}]$ and transeverse (TMR) $[(\Delta\rho/\rho)_{\perp}]$ magnetoresistances of the alloy with $x=30$ (FM) in the temperature range of 11–150 K and up to a field of 16.5 kOe. The MR in both the orientations is negative and varies slower than H below 50 K. $(\Delta\rho/\rho)_{\perp}$ at 11 K and a field at 16.5 kOe is only $\approx 0.17\%$. Above 50 K the MR becomes positive and shows a normal $(\Delta\rho/\rho \sim H^2)$ magnetoresistance behavior (Kohler type) till 300 K. It can be clearly seen that $|(\Delta\rho/\rho)_{\perp}|$ is always greater than $|(\Delta\rho/\rho)_{\parallel}|$, i.e., a small but finite anisotropy is indeed present till $T=50$ K. The typical value of the anisotropy of the MR (FAR) here $(=[|(\Delta\rho/\rho)_{\parallel}|-|(\Delta\rho/\rho)_{\perp}|])$ is 0.01% at $T=11$ K and at $H_{\rm int}=0$. This value of anisotropy of MR in $x=30$ (FM) is very small in contrast to the much larger values for a homogeneous FM. The origin of such an anisotropy term $(\Delta\rho_{\parallel}-\Delta\rho_{\perp})/\rho_0$ at the lowest fields and temperatures can be associated with the domain orientation in ordinary FM's like Ni and also in NiMn (21 at. % Mn) alloy near the percolation threshold.[10]

The field dependence of magnetization at several temperatures (isotherms) for $x=30$ has been presented in Fig. 2(a) up to a field of 16.5 kOe. We observe a clear tendency towards saturation but true saturation is not achieved even at the highest field (16.5 kOe) at the lowest temperature (19 K). The field dependence of magnetization at 19 K is found to be given by $M \propto H^{0.05}$ in the field range of 2–16.5 kOe. With the increase of temperature, this alloy becomes a PM $(M \propto H)$ beyond 200 K as clearly seen from Fig. 2(a). The Arrott plots $[M^{1/\beta}$ vs $(H/M)^{1/\gamma}]$, keeping $\beta=0.5$, and $\gamma=1$ (the mean-field values) for $x=30$ is shown in Fig. 2(b). We find a set of parallel lines having both positive and negative intercepts on the M^2 axis from which T_c can be estimated to be 135 K.

(b) In the case of the alloy with $x=26$ (mixed phase or re-entrant phase) both the LMR and TMR in the temperature range of 11–75 K and up to fields of 16.5 kOe are negative until 75 K and varies slower than H (not shown). $(\Delta\rho/\rho)_{\parallel}$ at

11 K and a field of 16.5 kOe is $\approx 0.18\%$. Several isothermal M-H measurements were taken in the range of 19–250 K. The plot at 19 K and fields up to even 16.5 kOe has a lot of curvature indicating lack of saturation of the magnetization. The functional dependence of this plot was obtained from the fit to $M \propto H^{0.1}$ in the field range of 2–16.5 kOe. The Arrott plots (not shown here) show large positive intercepts on the M^2 axis which confirm the FM ordering below 60 K.

(c) In a similar fashion the alloy with $x=23$ (another mixed-phase alloy) show a negative MR, almost isotropic (FAR$\simeq 0$) and its magnitude increases with field and decreases with temperature in the range 11–50 K. The MR at 100 K and above becomes positive (normal magnetoresistance). The value of the negative TMR at 16.5 kOe and at 11 K is 0.35%. It varies slower than H and depends strongly on temperature below 50 K. This kind of behavior is similar to that of concentrated AuFe (18 at. % Fe) (Refs. 11,13) and NiMn (21 at. % Mn) alloys[10,14] in the mixed-phase regime. This is in contrast to the low-field H^2 dependence of MR in AuFe (2.9 at. % Fe).[2] This is also different from those of conventional FM's where MR is linear in H and shows FAR. With the decrease of Ni concentration (x) the value of the FAR decreases (i.e., as the composition moves away from the critical concentration for long-range FM ordering, the anisotropy vanishes).

The isothermal M vs H plots for the alloy with $x=23$ (mixed phase) hardly shows any saturation even at 19 K and at 16.5 kOe. This behavior is comparable to those of AuFe (17 at. % of Fe) (Ref. 24) mixed-phase alloys. Toulouse had found theoretically that in the coexistence regime (mixed phase), the field dependence of magnetization at low fields is given by $M \propto H^{3/7}$. This was derived considering the infinite range Ising model of Sherrington and Kirkpatrick and including an average FM interaction. In contrast, from our M-H measurements, it is found that for $x=23$ (mixed phase) at 19 K, $M \propto H^{0.22}$ even at fields extending up to 15 kOe. The positive intercepts of Arrott plots for $x=23$ (not shown) as well confirm the presence of FM ordering in the coexistence regime. Had it been a pure SG there would not have been any positive intercept. The SG freezing of this mixed-phase alloy has already been established from the thermomagnetic history dependence of dc magnetization.[19] So we conclude that in the mixed-phase regime, SG and FM orderings indeed coexist.

(d) The field dependence of the LMR and TMR at several temperatures for the alloy with $x=21$ (SG) show identical (completely isotropic, FAR$=0$) behavior within the resolution of our experiment. The curves at 100 K and above show positive normal magnetoresistance. Below 50 K, the MR varies slower than H. The value of the TMR is $\approx 0.4\%$ at 11 K and at 16.5 kOe and is rather large in comparison with those of the other alloys (x). From the field dependence of the magnetization of $x=21$, we find that at the lowest temperature (19 K), the M-H plot does not show any tendency of saturation due to the strong competing interactions between FM and AFM exchange couplings (also seen in, say, AuFe with 4–8 at. % Fe impurity).[25] At 19 K, $M \propto H^{0.4}$ till 15 kOe. The Arrott plot of this alloy at 19 K (no positive intercept on the M^2 axis) rules out any FM ordering at this temperature.

(e) In Fig. 3(a), the low-field LMR $[(\Delta\rho/\rho)_\parallel]$ at several temperatures for the alloy with $x=19$ (SG) are shown. Its value at 11 K is $\approx 0.26\%$. At the lowest temperature $\Delta\rho/\rho$ varies slower than H. However, at higher temperatures it varies faster than H. This kind of behavior has also been found in AuFe alloys (8–13 at. % of Fe).[12] The MR above 100 K becomes positive (normal MR).

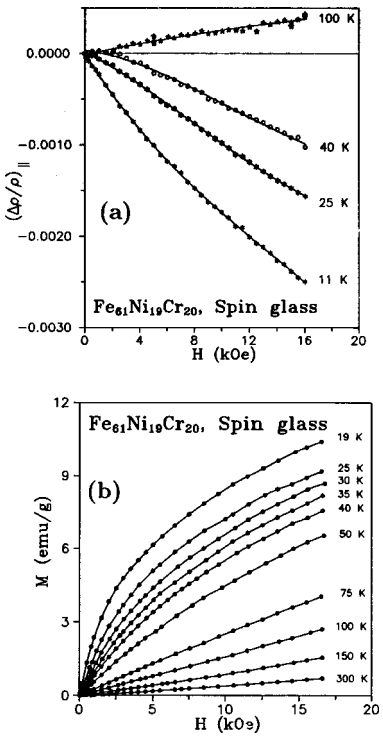

FIG. 3. (a) Magnetic-field (H) dependence of longitudinal $[(\Delta\rho/\rho)_\parallel]$ magnetoresistance at various temperatures for $Fe_{61}Ni_{19}Cr_{20}$ (spin glass) alloy. (b) M-H isotherms of $Fe_{57}Ni_{19}Cr_{20}$ (spin glass) alloy at various temperatures in the field range of 0–16.5 kOe.

The M-H plots [Fig. 3(b)] of this alloy ($x=19$) resembles those of $x=21$. At the lowest temperature (19 K) $M \propto H^{0.45}$. The Arrott plots of this alloy do not have any positive intercept on the M^2 axis even at the lowest temperature confirming the absence of any FM ordering.

(f) In Fig. 4(a) we have shown the field dependence of both the LMR and TMR [$(\Delta\rho/\rho)$ vs H] for the alloy with $x=14$ (AFM) at 4.2 K. Initially the LMR increases with field followed by a maximum and ultimately it becomes negative. In contrast, the TMR is negative for all fields from 0 to 14 kOe. As mentioned earlier, as the magnetic field is increased, the LMR of polycrystalline AFM metals should change from positive to zero and finally to negative values and the curve is rounded by domain-wall and polycrystalline effects. In this AFM the field dependence of the LMR exhibits a similar behavior which is consistent with studies on other AFM alloys, viz. Nd by Nagasawa.[17] The development of a negative LMR indicates that this alloy undergoes a field-induced *spin-flop* transition from an AFM state to a canted FM state.

The M-H curve of the alloy with $x=14$, which has an AFM ordering below $T_N=26$ K, shows a striking change of slope at around 10 kOe at 4.2 K [Fig. 4(b)]. This abrupt change of slope is more pronounced in the dM/dH vs H plot [Fig. 4(b)] which shows a peak at 10 kOe. However, it vanishes completely at 35.3 K (not shown). This change of slope

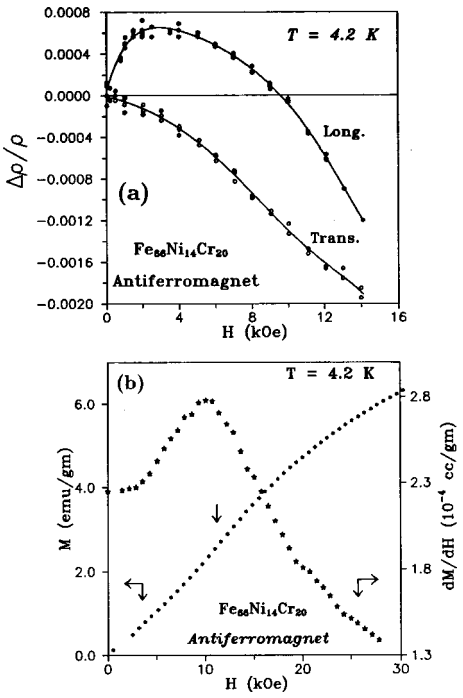

FIG. 4. (a) Magnetic-field (H) dependence of longitudinal (Long.) and transverse (Trans.) magnetoresistances at 4.2 K for $Fe_{66}Ni_{14}Cr_{20}$ (antiferromagnetic) alloy. (b) M-H and dM/dH vs H plots for the same alloy at 4.2 K. The M-H plot shows a striking change of slope around 10 kOe (shown by an arrow) where dM/dH shows a peak. This has been attributed to *spin-flop* transition at this field.

in the M-H plot and the development of a negative LMR at 4.2 K should have the same origin, namely, the *spin-flop* transition arising from the canting of the AFM spins.

1. Magnetic field, temperature, and composition dependence of magnetoresistance

The magnetic-field dependence of $\Delta\rho/\rho$ of all the alloys under investigation have been fitted to the relation

$$\frac{\Delta\rho}{\rho} = -\alpha(T)H^n$$

using a nonlinear least-squares-fit program. We have also estimated the contribution to the MR from Lorentz force normal MR and find that it is negligible compared to the magnetic term for all the concentrations in the field range of 1–16.5 kOe and temperature range of 11–50 K. Above 100 K the MR of all the alloys are positive where the normal MR dominates. The normal MR (Kohler) is essentially due to band effects and is given by $\Delta\rho/\rho \sim (\omega_c\tau_e)^2$, where $\omega_c = eB/m$, is the cyclotron frequency and τ_e, the elastic-scattering time. At a magnetic field $B=1$ T, $\omega_c \sim 10^{11}$ s^{-1}. Using various data for these Fe-Ni-Cr alloys from our earlier reports,[26,27] namely, conductivity at 10 K ($\sigma_{10\,\mathrm{K}} \sim 10^4$ s/cm), mean free path ($l \sim$ a few tens of Å), average electron velocity ($v_0 \sim 10^7 - 10^8$ cm/s), and elastic scattering time ($\tau_e \sim 10^{-14}$ s) we expect the MR in such a field to satisfy the condition, $\omega_c\tau_e \ll 1$. In our alloys the normal (Kohler) MR in magnetic field $B=1$ T at 10 K is going to be negligible $[\Delta\rho/\rho \sim (\omega_c\tau_e)^2 \sim 10^{-6}]$, compared to the observed negative MR $[\sim 10^{-3}]$.

TABLE I. Values of the parameters n, and $\alpha(T)$ and also χ^2 for the fits to $\Delta\rho/\rho = -\alpha(T)H^n$ at various temperatures for $Fe_{80-x}Ni_xCr_{20}$ ($14 \leqslant x \leqslant 30$) alloys in the field range of 2–16.5 kOe.

Alloy (x)	T (K)	n	$\alpha(T)$ (10^{-4} kOe^{-n})	$\chi^{2\,a}$ (10^{-6})
19	11	0.80	2.8	5.0
($T_{SG}=12$ K)	25	1.07	0.8	3.9
	40	1.40	0.2	6.0
21	11	0.74	4.9	15
($T_{SG}=10$ K)	15	0.77	4.0	31
	19	0.76	3.7	9
	30	0.78	2.6	18
	40	0.73	0.6	82
23	11	0.66	6.0	6.4
(T_c, $T_{SG}=35$, 20 K)	17	0.66	5.5	7.0
	20	0.66	4.6	2.4
	25	0.65	4.7	9.8
	30	0.64	3.9	1.8
	35	0.65	4.6	8.7
	45	0.65	3.7	12
26	11	0.54	4.3	6.0
(T_c, $T_{SG}=60$, 7 K)	15	0.53	4.0	9.0
	19	0.48	3.6	8.5
	40	0.52	2.2	8.8
	60	0.27	2.1	12
	75	0.19	1.4	21
30	11	0.74	2.4	25
($T_c=135$ K)	20	0.67	6.7	19
	30	0.60	1.4	50
	50	0.10	1.3	11

[a] $\chi^2 = 1/N\sum_{i=1}^{i=N}[Y_i(\mathrm{data}) - Y_i(\mathrm{fit})]^2/[Y_i(\mathrm{data})]^2$.

In Table I we have summarized the values of n, $\alpha(T)$, and χ^2 till 50 K for all the alloys in the magnetic-field range of 2–16 kOe. The values of $\chi^2 (\approx 10^{-6})$ are consistent with our experimental accuracy. From this table we observe that

(1) for the alloy with $x=19$ (SG phase), the exponent of H, $n=0.8$ at 11 K. The value of n increases with temperature. Above the ordering temperature (T_{SG}), n increases continuously to values between 1 and 2. This indicates the existence of short-range correlations since in a completely paramagnetic state, n is 2.[13,28] In canonical SG's (Ref. 2) where spin freezing takes place because of random bond frustration (magnetic impurities are indirectly ferro- or antiferromagnetically coupled to each other via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions), the exponent (n) was found to be 2 with almost temperature-independent coefficient α below their freezing temperatures. In this alloy ($x=19$) where the SG phase arises not due to RKKY exchange interaction mediated by conduction electrons but due to the direct competing FM and AFM exchange interactions (concentrated SG), the value of $n<1$. This kind of observation was also made in NiMn alloys[10] (only qualitative study) in the SG phase near the critical composition for ferromagnetism ($25 \leqslant x \leqslant 28$).

(2) In the case of $x=21$ (spin-glass or cluster-glass phase), we find that $n\simeq 3/4$ and is temperature independent as well. Here also we find n different from 2 unlike the canonical SG's.[2] There are some recent reports on $Au_{87}Fe_{13}$,[11] $Au_{85}Fe_{15}$,[12,29] and $Au_{82}(Fe_{1-x}Cr_x)_{18}$ (Ref. 29) alloys with $15\leqslant x\leqslant 25$ which find the same value of the exponent, n ($\approx 0.75\pm 0.04$) below the $T_{SG/CG}$ phase. Moreover, they have referred our value[22] as the only available one for comparing their data for the concentrated alloys in the SG/CG regime.

Since this alloy ($x=21$) is very near the mixed-phase or re-entrant SG phase regime ($x=23-26$), it must have cluster-glass-like (CG) ordering below $T_{SG/CG}$. Hence the MR will have contributions from both intercluster interaction which is of spin-glass-type and intracluster interaction which is ferromagnetic. The unique value of the exponent of H ($n\approx 3/4$) results from the above combined effects for all the alloys in the SG/CG regime.

(3) For the alloy with $x=23$ (mixed or re-entrant phase), the exponent of H is 2/3 and is independent of T up to 50 K which is above both T_{SG} and T_c. This value of n is different from the value of 6/7 found for AuFe (18 at. % Fe) (Ref. 13) for $T<T_{SG}$. There it depended strongly on temperature for $T>T_{SG}$. However, a recent study on $Au_{82}(Fe_{1-x}Cr_x)_{18}$ alloys for $0.05\leqslant x\leqslant 1$[12] which show RSG behavior, the value of n is found to be 0.67 (± 0.02) or $\simeq\frac{2}{3}$ in the temperature range of 4.2–200 K and in the field range of 2–45 kOe. It is interesting to note that in all these alloys the range of fit of $H^{2/3}$ dependence of $\Delta\rho/\rho$ extends beyond the respective Curie temperatures. Moreover, in the same study,[29] they have rechecked the data for $Au_{82}Fe_{18}$ (RSG) alloy and found that the value of $n\approx\frac{2}{3}$ only between 140 and 160 K which is close to its T_c (150.7 K). Below T_{SG}, $n\approx 0.8\pm 0.2$. They have also referred our value[22] as the only available one for comparing their data[29] in the case of concentrated alloys in the mixed-phase (or RSG) regime.

According to Balberg, the field dependence of MR of a weak ferromagnet at high fields is given by[28]

$$\Delta\rho\propto -H^{(1-\alpha)/\beta\delta},$$

provided $(\mu H/kT)\gg|1-T/T_c|$, where μ is the magnetic moment of the ion, α the specific heat exponent, β and δ are the critical exponents for spontaneous magnetization and critical isotherm, respectively. In the case of AuFe (RSG) alloys, magnetization[30] and nonlinear susceptibility[31] measurements near the critical temperature show that the critical exponents are similar to those of $3d$ Heisenberg systems.[32] Putting the values of these critical exponents, n [$\approx(1-\alpha)/\beta\delta$] is found to be 0.64 which is very close to the value found for our RSG alloys ($\approx\frac{2}{3}$).

So our present study along with the studies mentioned above reveal that $\Delta\rho/\rho\propto -H^{2/3}$ could be claimed as a characteristic feature of re-entrant spin glasses or mixed-phase alloys in the field range above their technical saturation.

(4) For the alloy with $x=26$ (RSG), the value of n is found to be $\frac{1}{2}$ in the temperature range of 11–50 K and is temperature independent. This particular alloy is very close to the critical concentration for long-range FM ordering. The different value of the exponent n compared to that in the RSG regime ($n\approx 2/3$) can be attributed to the fluctuations due to the formation of large clusters along with the spin-glass freezing.

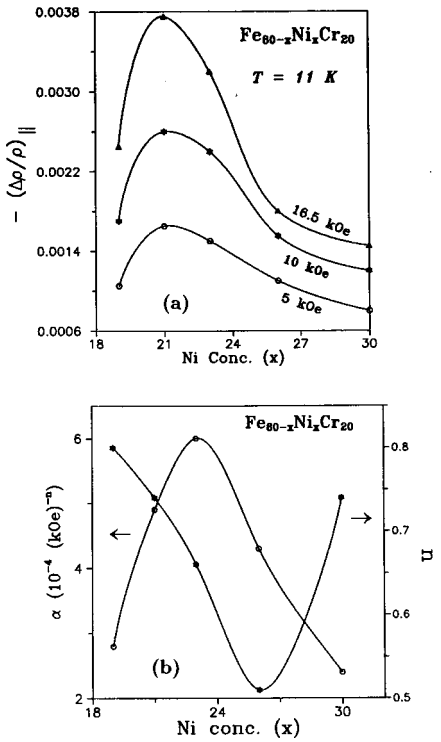

FIG. 5. Ni concentration (x) dependence of (a) longitudinal MR at 5, 10, and 16.5 kOe magnetic fields and (b) α and n for $Fe_{80-x}Ni_xCr_{20}$ alloys at 11 K.

(5) In the case of the alloy with $x=30$ (FM phase), n varies strongly with temperature. $\alpha(T)$, the coefficient of H^n, is the strength of the MR. It decreases with temperature in all the samples. As mentioned in the earlier section, a small but finite anisotropy (FAR of 0.01%) is found in the very low-field region (0–200 Oe) and at high fields (16.5 kOe) as well.

In Fig. 5(a) the composition dependence of $(\Delta\rho/\rho)_\parallel$ has been shown at 11 K for fields of 5, 10, and 16.5 kOe. It can be clearly seen from the figure that the magnitude of the MR is maximum in the SG regime (close to the critical concentration region $x\approx x_c$). In Fig. 5(b) we have shown the composition (x) dependence of the coefficient α (Table I) and the exponent, n at 11 K. The value of α increases with x, reaches a maximum at $x=23$ and then decreases as the alloys approach long-range FM. The value of n decreases with x, reaches a minimum at $x=26$ and then increases with x.

The temperature dependence of $(\Delta\rho/\rho)_\parallel$ for $x=30$, 26, 23, 21, and 19 at fields of 5, 10, and 16.5 kOe have shown that above 50 K the sign of $(\Delta\rho/\rho)_\parallel$ changes from negative to positive (not shown here). The decrease of the $|(\Delta\rho/\rho)_\parallel|$ with the increase of temperature indicates an enhancement of the amplitude for spin-flip scattering of conduction electrons from localized moments of the magnetic ions.

So we observe a definite trend of MR, α and n with composition as the Fe-Ni-Cr system evolves from long-range AFM ordering to long-range FM ordering passing through the critical concentration region ($x\approx x_c$) where the maximum change of all the three quantities occurs.

There are reports on MR of NiMn alloys[10,14] with about 21–30 at. % Mn (which are very near the critical concentration regime for FM ordering) in SG, RSG, and FM phases. In all these phases they have also found the field dependence of $\Delta\rho/\rho$ to be slower than linear. However, no quantitative study has been made on those alloys.

In this low-field region ($g\mu_B H/k_B T<2$, where $H\sim 10$ kOe) below the respective transition temperatures the most rapidly varying part of the negative MR of these Fe-Ni-Cr magnetic alloys is caused by the freezing out of the spin-flip s-d scattering due to the alignment of localized magnetic spins with the applied magnetic field.

2. Correlation between low-field magnetoresistance and magnetization

In this section we have studied the magnetization dependence of the magnetoresistance of the Fe-Ni-Cr series. To find out any meaningful correlation between $\Delta\rho/\rho$ and M, we have carried out $\Delta\rho/\rho(H,T)$ and $M(H,T)$ measurements under identical conditions (at the same H and T) for each composition. Eliminating H between the two sets, namely,

$$\left|\frac{\Delta\rho}{\rho}\right|\propto H^n \quad \text{and} \quad M\propto H^m,$$

one gets

$$\left|\frac{\Delta\rho}{\rho}\right|\propto M^{n/m}, \quad \text{at any temperature } T. \tag{3}$$

Thus we can find $\Delta\rho/\rho$ vs M at any temperature T without using any theoretical model. Figure 6(a) shows $\ln|\Delta\rho/\rho|$ vs $\ln|M|$ plot for $x=21$ at various temperatures from 11 to 30 K. Fitting the data to a straight line ($\chi^2\approx 10^{-6}$) yields $\Delta\rho/\rho\propto M^{2.5}$ over a wide range of temperature and field. However, the data above 30 K do not fall on this curve. In the case of $x=23$ (mixed phase) such plots yield the values of the exponent of M varying from 3.1 to 2.0 as T changes from 19 to 45 K, thus showing no correlation between $\Delta\rho/\rho$ and M. For $x=30$ (FM), $\Delta\rho/\rho$ (H) is strongly dependent on temperature, while $M(H)$ has a weak temperature dependence for $T\ll T_c$ making the attempt of correlation meaningless. So for the FM alloy and also for the alloys in the RSG phase such a dependence of $\Delta\rho/\rho$ on M [Eq. (3)] is not observed. This is a very important observation because it shows that the MR of the RSG is more like that of a FM than that of a SG.

For canonical spin glasses (e.g., AuFe, CuMn, etc.) where indirect RKKY interaction (J_{RKKY}) between magnetic impurities dominates, it is known that

$$\frac{\Delta\rho}{\rho}\propto M^2\propto\chi^2H^2, \quad \text{for zero-field-cooled} \tag{4}$$

and

$$\frac{\Delta\rho}{\rho}\propto(m_r+\chi H)^2, \quad \text{for field-cooled states,} \tag{5}$$

where χ, H, and m_r are, respectively, the reversible susceptibility, the applied field, and the remanent magnetization (irreversible part).

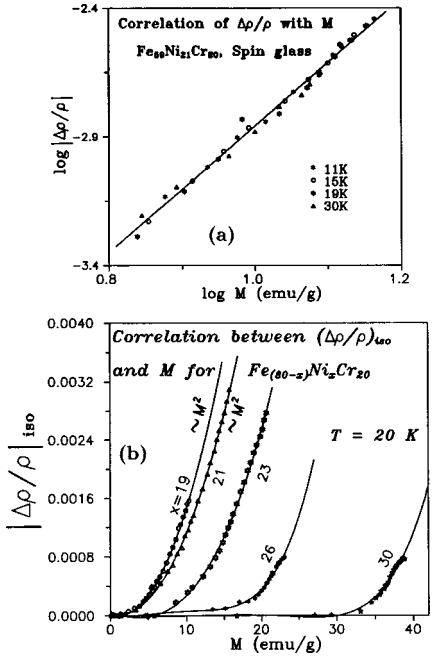

FIG. 6. (a) Log-log plot of transverse magnetoresistance and magnetization at different temperatures for $Fe_{59}Ni_{21}Cr_{20}$ (spin-glass) alloy. The slope of the plot gives $|(\Delta\rho/\rho)_{\parallel}|\sim M^{2.5}$. (b) Correlation between $(\Delta\rho/\rho)_{\mathrm{iso}}$ and M for $Fe_{80-x}Ni_xCr_{20}$ alloys at $T=20$ K clearly showing a gradual evolution of MR as one goes from the long-range FM ordering towards the critical region ($x\approx x_c$) close to the SG regime.

Equation (4) can be rewritten as

$$\frac{\Delta\rho}{\rho}(H)=-\alpha[M(H)]^2 \quad \text{for homogeneous SG alloys.} \tag{6}$$

In a similar way, for homogeneous randomly canted FM's with nonzero spontaneous magnetic moment, Senoussi[14,11] suggested that

$$\frac{\Delta\rho}{\rho}(H)=-\alpha[M^2(H)-M^2(0)], \tag{7}$$

where $M(0)$ is the spontaneous magnetization in units of μ_B/ion (estimated by extrapolation from measurements in strong enough fields to align the domains).

We have seen in earlier sections that in all our alloys (FM, RSG, SG, etc.) in the vicinity of the critical composition ($x\approx x_c$), the anisotropy $[|(\Delta\rho/\rho)_{\parallel}|-|(\Delta\rho/\rho)_{\perp}|]$ is weak and there is a comparatively large isotropic contribution to the MR. We have made an attempt to correlate the isotropic part of the MR of each alloy with their magnetization. In any polycrystalline magnetic material with random initial domain orientations, the isotropic MR is defined by

$$\left[\frac{\Delta\rho}{\rho}\right]_{\mathrm{iso}}=\frac{(\Delta\rho/\rho)_{\parallel}+2(\Delta\rho/\rho)_{\perp}}{3}. \tag{8}$$

In Fig. 6(b) we have plotted $[\Delta\rho/\rho]_{\mathrm{iso}}(H)$ vs $M(H)$, eliminating H from both, for all the alloys under investigation at $T=20$ K. For the lower concentrations corresponding to the SG alloys ($x=19,21$), $[\Delta\rho/\rho]_{\mathrm{iso}}$ is exactly propor-

TABLE II. Ni concentration dependence of α, $M(0)$, and αM_0^2 as obtained from the fit to Eq. (7).

Ni concentration (x)	α 10^5 (emu/g)$^{-2}$	$M(0)$ (emu/g)	αM_0^2 (10^{-4})
19	1.40	0	0
21	1.19	0	0
23	0.84	10.1	8.5
26	0.36	18.2	11.1
30	0.23	34.0	26.1

tional to $M^2(H)$ showing that the alloys behave more or less as homogeneous spin glasses confirming the results of Senoussi.[4] However, for $x=23$ (mixed phase or RSG), there is a weak departure from the M^2 behavior. This becomes more severe for $x=26$ (RSG). This change of behavior is a precursor to the FM ordering. Finally, in the case of $x=30$ (FM) one observes a behavior which is almost characteristic of a homogeneous canted FM state [Eq. (7)] or at least the state has started to set in. Fitting the data to Eq. (7), one obtains a value of $M(0)$ which is very close to the value of M where one observes technical saturation in the M-H curve [Fig. 2(a)].

At low fields alignment of domains produces a rapid increase in M without affecting the resistivity as the domain size is much greater than the electron mean free path. At higher fields (beyond technical saturation) once a single-domain structure is established, the progressive alignment of individual moments of canted spins [which is manifested in the high-field slope of the M-H curve, Fig. 2(a), beyond the technical saturation] dominates the MR behavior and the M^2 dependence [Eq. (7)].

We have fitted the data to Eq. (7), viz $\Delta\rho/\rho(H)=-\alpha[M^2(H)-M^2(0)]$ for all the samples at $T=20$ K [shown in Fig. 6(b)] and the best fitted parameters are summarized in Table II. We find that for $x=19$ and 21 (SG), the values of $M(0)\approx 0$ as expected. For $x=23$ and 26 (mixed phase or RSG) and 30 (FM), the $M(0)$ values are very close to their values at technical saturation. The values of α decrease with the increase of Ni concentration as the system approaches long-range FM ordering.

We thus conclude that the RSG's ($x=23$ and 26) are the precursor to the FM ordering. In the concentrated SG ($x=19$ and 21) alloys, $[\Delta\rho/\rho]_{\rm iso}$ varies as M^2 as expected (supporting the homogeneous model for the magnetization as found in canonical spin glasses). For $x=30$, the homogeneous canted FM phase sets in. For the RSG alloys there is a short-range FM ordering on the scale of electron mean free path ($\sim 10-20$ Å). Figure 6(b) clearly shows a gradual evolution of MR as one goes from the long-range FM ordering towards the critical region ($x\approx x_c$) close to the SG regime.

B. High-field (till 200 kOe) magnetoresistance and magnetization

We have also carried out longitudinal magnetoresistance $[(\Delta\rho/\rho)_{\parallel}(H)]$ and magnetization $[M(H)]$ (Ref. 23) up to fields as high as 200 kOe in the temperature range of 4.2–60 K for $Fe_{80-x}Ni_xCr_{20}$ ($x=14$, 19, 26, and 30) alloys. In Fig. 7 we have shown $[(\Delta\rho/\rho)_{\parallel}]$ vs H for $x=26$ (mixed phase) in

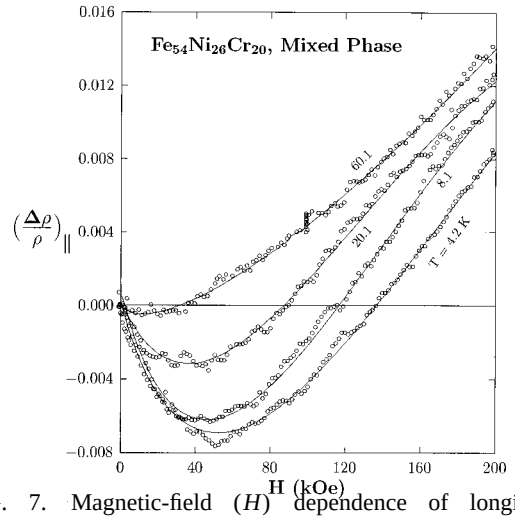

FIG. 7. Magnetic-field (H) dependence of longitudinal $[(\Delta\rho/\rho)_{\parallel}]$ magnetoresistance at various temperatures for $Fe_{54}Ni_{26}Cr_{20}$ (mixed phase) in the field range of 0–200 kOe.

the temperature range of 4.2–60 K and up to a field of 200 kOe. In the earlier section, the MR plot for the same alloy until 16.5 kOe was found to be negative having a typical value of 0.18% at 16.5 kOe at 11 K. Figure 7, where the same plot has been extended up to 200 kOe, shows that $[(\Delta\rho/\rho)_{\parallel}]$ is negative at low fields, passes through a minimum at $H_{\rm min}$ and becomes positive at higher fields. At higher temperatures the depth of the minimum reduces, the value of $H_{\rm min}$ shifts towards lower fields and the MR $[(\Delta\rho/\rho)_{\parallel}]$ becomes more and more positive. It should be noted that above 75 K, $[(\Delta\rho/\rho)_{\parallel}]$ becomes completely positive (as found in the earlier low-field study) at all fields. This observation is also consistent with the current high-field studies at higher temperatures. In Table III, we have given the values of the depth of minima and $H_{\rm min}$.

To the best of our knowledge no theory has been developed for any magnetic system so far which could explain the observation of an unknown positive contribution to magnetoresistance dominating at higher fields. Generally, with the application of a magnetic field, the amplitude of the spin-flip scattering (s-d interaction) of the conduction electrons by the magnetic ions gets suppressed because of the alignment of the magnetic spins. As a result all magnetic systems (except AFM) show negative $\Delta\rho/\rho$ (H). With a higher field, more of the canted spins will be aligned. But beyond a certain field this anomalous (negative) contribution will be smaller compared to the normal (Kohler) MR which originates from band effects. With a higher B, ω_c and hence $(\Delta\rho/\rho)_{\rm normal}$ will be higher and ultimately the latter dominates at the highest field.

So the MR can be thought of as composed of one positive and one negative term which can be expressed as

$$\left[\frac{\Delta\rho}{\rho}\right](H)=\left[\frac{\Delta\rho}{\rho}\right]_n+\left[\frac{\Delta\rho}{\rho}\right]_a=aH^2-bH^n. \qquad (9)$$

The first term is due to the normal (Kohler) MR and the second term is the anomalous one (as discussed earlier). a, b, and n are temperature-dependent adjustable parameters. In the low-field studies (up to 16.5 kOe) we have already observed the dominating H^2 dependence of the normal MR

TABLE III. Temperature dependence of depth of minimum of MR, strength of field at which minimum observed (H_{min}), and the parameters obtained from the fit to Eq. (9) for Fe-Ni-Cr alloys.

Sample	Depth of minimum $[\Delta\rho/\rho(\%)]$	H_{min} (kOe)	T (K)	a (10^{-6}) $(\text{kOe})^{-2}$	b $(10^{-(2+n)})$ $(\text{kOe})^{-n}$	n	χ^2 (10^{-4})	Range of field (kOe)
$Fe_{54}Ni_{26}Cr_{20}$ (mixed phase)	0.702	50	4.1	0.585	0.375	0.433	5.1	0–200
	0.620	45	8.1	0.605	0.358	0.375	9.7	0–200
	0.315	35	20.1	0.448	0.229	0.173	7.0	0–200
	0.046	13	60.1	0.380	0.002	0.100	7.5	0–200
$Fe_{61}Ni_{19}Cr_{20}$ (spin glass)	0.50	60	8.1	0.279	0.294	0.386	8.7	0–190
	0.21	45	20.1	0.312	0.138	0.458	1.7	0–190

for all the alloys above 100 K. In this high-field investigation it is found that the positive contribution (H^2 dependent) dominates only at higher fields (>50 kOe at 4.2 K) at low temperatures. In Table III, we have listed the values of the fitting parameters (a,b,n), χ^2, and the field range of the fit to Eq. (9) for $x=26$. Figures 8(a) and 8(b) show the field dependence of the individual contributions from the normal and anomalous magnetoresistances, respectively at various temperatures.

In Fig. 9, the high-field $[(\Delta\rho/\rho)_\parallel]$ plot has been shown for the alloy with $x=19$ (SG) at 8.1 and 20.1 K. These are the best fits to Eq. (9). The field dependence of individual contributions (normal and anomalous MR) are shown in Figs. 10(a) and 10(b), respectively. The fitting parameters are given in Table III.

So we find that the high-field MR data for $x=26$ and 19 can be well described by Eq. (9) within the experimental resolution. As it stands we can assign the negative contribution essentially to some magnetic origin.

In Figs. 11(a) and 11(b) we show the correlation of anomalous (negative) contribution of MR $|(\Delta\rho/\rho)_a|$ with magnetization M at several temperatures in the field range of 30–200 kOe, eliminating H from both M vs H (Ref. 23) and $|(\Delta\rho/\rho)_a|$ vs H curves for the alloys with $x=26$ (mixed phase) and $x=19$ (SG). Employing a nonlinear least-squares fit to all the curves [Fig. 11(a) and 11(b)] using a function $|(\Delta\rho/\rho)_a|=\beta M^n$ we obtain the exponents, $n=2.65$, 2.4, and 1.06 and $\beta=(0.8,\ 1.6,\ \text{and}\ 83)\times10^{-6}$ $(\text{emu/gm})^{-n}$ at 4.2, 8.1, and 20.1 K, respectively for $x=26$ (mixed phase). In the case of the alloy $x=19$ (SG), the above parameters are found to be $n=1.5$ and 1.3 and $\beta=(4.4,\ 13)\times10^{-6}$ $(\text{emu/gm})^{-n}$ at 8.1 and 20.1 K, respectively. From the above results one can conclude that at very high fields the simple homogeneous model of quadratic dependence of the negative (anomalous) MR on M is not strictly obeyed for the concentrated SG ($x=19$) alloy.

Béal Monod and Weiner[33] had suggested that for very high magnetic fields and low temperatures ($g\mu_B H/k_B T \geqslant 4$), the localized magnetic spins get completely aligned with the magnetic field; the conduction-electron scattering amplitude becomes the main source of variation in the MR

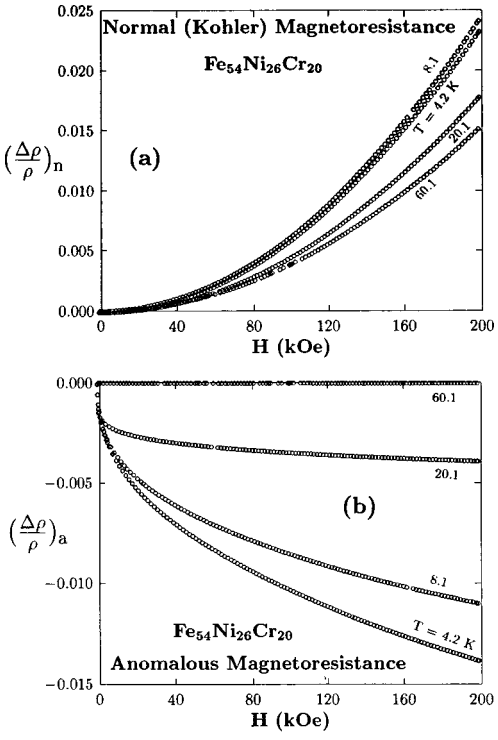

FIG. 8. Magnetic-field dependence of (a) normal and (b) anomalous contribution to MR for $Fe_{54}Ni_{26}Cr_{20}$ (mixed phase) alloy at various temperatures in the field range of 0–200 kOe.

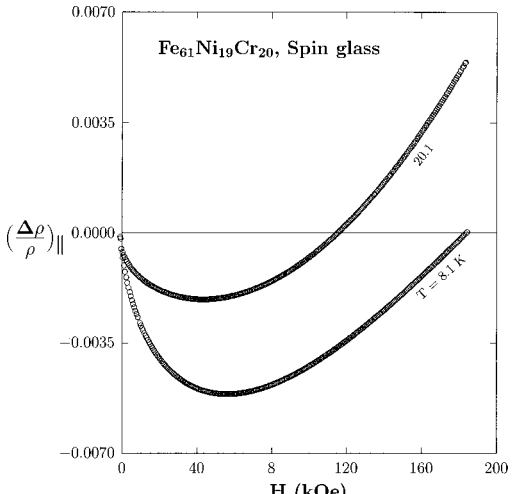

FIG. 9. Magnetic-field (H) dependence of longitudinal $[(\Delta\rho/\rho)_\parallel]$ magnetoresistance at 8.1 and 20.1 K for $Fe_{61}Ni_{19}Cr_{20}$ (spin glass) in the field range of 0–200 kOe.

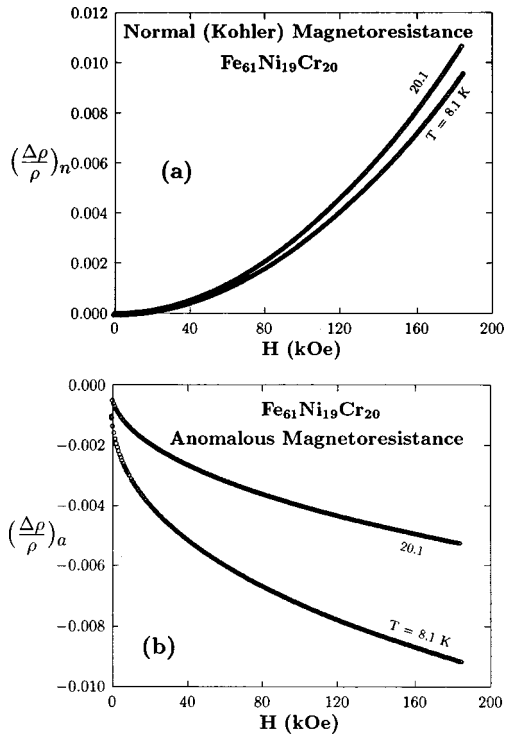

FIG. 10. Magnetic-field dependence of (a) normal and (b) anomalous contribution to MR at various temperatures for $Fe_{61}Ni_{19}Cr_{20}$ (spin glass) in the field range of 0–200 kOe.

with H but the exact theory is yet to emerge. At high enough fields the anomalous contribution to MR may be expected to saturate [Figs. 8(b) and 10(b)] in all these magnetic Fe-Ni-Cr alloys.

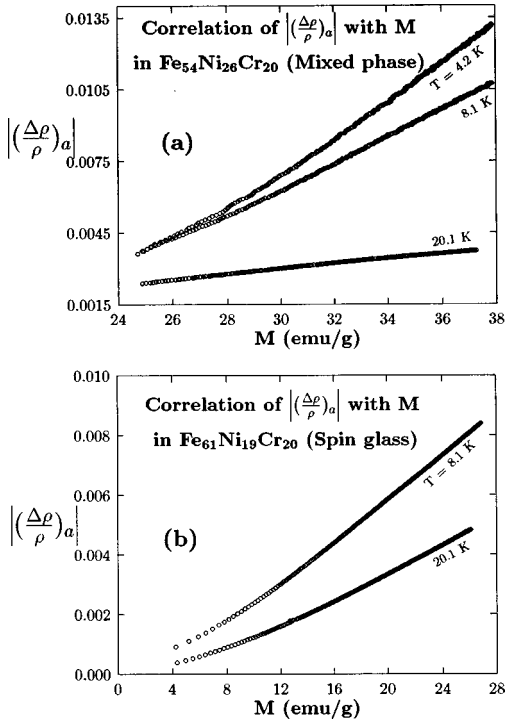

FIG. 11. Correlation of negative (anomalous) MR $[(\Delta\rho/\rho)_a]$ with magnetization (M) at various temperatures for (a) $Fe_{54}Ni_{26}Cr_{20}$ (mixed-phase) and (b) $Fe_{61}Ni_{19}Cr_{20}$ (spin-glass) alloys in the high-field (30–200 kOe) regime.

As mentioned earlier, a part of the negative MR contribution can come from *quantum interference effects* (QIE) at low temperatures as observed in many structurally disordered metallic glasses. So one might argue that the negative MR in these disordered alloys [$\rho_{300\ \mathrm{K}} \sim (100-130)\ \mu\Omega$ cm] (Ref. 26) might arise from QIE. The magnetoresistance due to electron-electron interaction effects (EEI) is positive[34] and so our observation of a negative magnetoresistance can not be explained through EEI effects.

The MR due to weak localization is generally negative (in the absence of any spin-orbit interaction). In the presence of a magnetic field the time-reversal symmetry between the two time-reversed paths (weak localization picture) breaks down. The electron waves traversing the two otherwise equivalent paths acquire a phase difference proportional to the enclosed area and the strength of the field. This destroys the constructive interference between the two counter-propagating waves giving rise to a negative MR just as inelastic scattering produces a negative TCR at finite temperatures.

The magnetoconductance due to weak localization effect can be estimated using relations theoretically predicted by Kawabata[35] and Altshuler and Aronov.[36] It is given by

$$[\Delta\sigma]_{\mathrm{WL}} = \frac{e^2}{2\pi^2\hbar}\left[\frac{e}{\hbar}\right]^{1/2}\sqrt{\mathbf{H}} \tag{10}$$

$$\approx 0.9\sqrt{H}\ \text{s/cm}, \quad (H \text{ in kOe}). \tag{11}$$

We estimate the contribution to the magnetoconductance due to QIE at 50 kOe using the above relation and get $\Delta\sigma_{\mathrm{WL}} \approx 6.3$ s/cm. At 50 kOe and $T = 4.2$ K, the typical observed change in conductivity $\Delta\sigma$ for $x = 26$ is 77 s/cm. Thus the contribution from the quantum correction term to the MR is very small (if present at all) compared to the magnetic contribution. Moreover, a very important point should be mentioned here that the presence of any small ferromagnetic impurity can destroy weak localization. In all these random magnetic alloys Ni is ferromagnetic and there are also strong FM interactions among some other pairs as found from inelastic small-angle neutron scattering by Men'shikov *et al.*[37] It is very unlikely that weak localization can persist in these magnetic alloys. So one has to be very careful in invoking QIE in these Fe-Ni-Cr alloys.

IV. CONCLUSIONS

In this work we have performed systematic studies of low (0–16.5 kOe) and very high field (0–200 kOe) $\Delta\rho/\rho(H,T)$ and $M(H,T)$ in the temperature range of 4.2–300 K. The most important observation made in the low-field magnetoresistance (MR) studies is that all the alloys show negative (anomalous) MR (with large isotropic part) [except $x = 14$ (AFM) alloy) below 50 K. Above 50 K all of them show Kohler-type (quadratic in H) normal MR. The magnitude of the negative MR reaches a maximum near the critical concentration region [$x = 19,21$ (SG)]. At the lowest field and temperature the FM ($x = 30$) alloy does show a small FAR because of the domain orientation. The magnitude of the anisotropic MR $[(\Delta\rho/\rho)_{\parallel} - (\Delta\rho/\rho)_{\perp}]$ decreases as this sys-

tem moves away from the long-range ordered phases [$x=30$ (FM) or $x=14$ (AFM)].

The low-field magnetization studies (Arrott plots) in the mixed-phase alloys ($x=23$ and 26) show conclusively that the long-range ferromagnetism coexists with the spin-glass ordering below T_{SG}.

In the low-field regime below 50 K, the MR data of all the alloys have been well described by an empirical relation $\Delta\rho/\rho=-\alpha(T)H^n$ with distinct values of α and n for different magnetic phases. In the case of the concentrated SG alloy ($x=21$), n is found to be $\frac{3}{4}$ whereas for the mixed-phase alloy ($x=23$) it is $\frac{2}{3}$ and they remain temperature independent below 50 K. From these results we claim that these are the characteristics of the field dependence of MR in concentrated SG and re-entrant SG phases, respectively. For $x=26$, $n\simeq\frac{1}{2}$ but this RSG is too close to the FM regime. Definite trends of MR, α and n with composition (x) have been obtained as the Fe-Ni-Cr system evolves from the long-range AFM ordering to the long-range FM ordering passing through the critical concentration ($x\approx x_c$) where the maximum change of all the three quantities occurs.

In the case of the SG alloy ($x=21$), the MR correlates very well with M as $\Delta\rho/\rho\propto M^{2.5}$ over a wide range of temperature and field. The isotropic part of MR $[(\Delta\rho/\rho)_{iso}]$ correlates with M in distinct fashions in different magnetic phases in the presence of either short or long-range ordering. For the SG alloys with no long-range ordering it correlates very well with M satisfying an empirical relation $[(\Delta\rho/\rho)_{iso}]=-\alpha[M(H)]^2$. As the system approaches the long-range FM ordering from the SG and RSG regimes, a distinct deviation from the above relation has been observed and finally in the case of long-range FM alloy ($x=30$) the relationship $[(\Delta\rho/\rho)_{iso}]=-\alpha[M^2(H)-M^2(0)]$ holds. We thus conclude that the RSG ($x=23$ and 26) phase is the precursor to the FM ordering. For the RSG alloys there is a short-range FM ordering on the scale of electron mean free path ($\sim 10-20$ Å). For the FM alloy ($x=30$), a homogeneous canted FM phase sets in. *So a gradual evolution of MR occurs as one goes from the long-range FM ordering towards the critical region* ($x\approx x_c$) *close to the SG regime.*

At low temperatures and very high fields beyond 40–50 kOe the observed upturn in the negative MR in these Fe-Ni-Cr alloys has been attributed to the positive (normal) contribution having a quadratic field dependence $[(\Delta\rho/\rho)_n \sim aH^2]$. The negative (anomalous) contribution mainly arises from the suppression of the spin-flip scattering in these alloys in different magnetic states. At very high fields the simple quadratic dependence of negative MR on M is not strictly followed for the concentrated SG ($x=19$) alloy.

The contribution to MR from QIE is found to be very small compared to the other contributions in all these Fe-Ni-Cr alloys. To our knowledge, very few MR studies have been done on similar type of highly resistive crystalline alloys. More work is needed to understand anisotropies, QIE, magnetic effect, etc. in these magnetic alloys.

ACKNOWLEDGMENTS

Financial assistance from Project No. SP/S2/M-24/93 of the Department of Science and Technology, Government of India and NSF grant INT-960 2975 is gratefully acknowledged.

[1] J.-P. Jan, in *Solid State Physics: Advances in Research and Applications*, edited by F. Seitz and D. Turnball (Academic, New York, 1957), Vol. 5, p. 1; J. M. Ziman, *Electron and Phonons* (Clarendon, Oxford, 1960).
[2] A. K. Nigam and A. K. Majumdar, Phys. Rev. B **27**, 495 (1983).
[3] A. K. Majumdar and V. Oestreich, Phys. Rev. B **30**, 5342 (1984).
[4] S. Senoussi, J. Phys. F **10**, 2491 (1980); J. Phys. (France) Lett. **42**, L35 (1981).
[5] A. K. Majumdar, Phys. Rev. B **28**, 2750 (1984).
[6] A. Mookerjee, J. Phys. F **10**, 1559 (1980).
[7] G. Sherlekar, C. M. Srivastava, A. K. Nigam, and G. Chandra, Phys. Rev. B **34**, 498 (1986).
[8] M. Gabay and G. Toulouse, Phys. Rev. Lett. **47**, 201 (1981).
[9] A. Hamzic and I. A. Campbell, J. Phys. (France) Lett. **42**, L309 (1981).
[10] S. Senoussi and Y. Öner, J. Appl. Phys. **55**, 1472 (1984).
[11] H. Rokoto, J. C. Ousset, S. Senoussi, and I. A. Campbell, J. Magn. Magn. Mater. **46**, 2112 (1984).
[12] A. K. Nigam, S. Radha, and Girish Chandra, J. Phys.: Condens. Matter **5**, 9197 (1993).
[13] A. K. Nigam, G. Chandra, and S. Ramakrishnan, J. Phys. F **16**, 1255 (1986).
[14] S. Senoussi and Y. Öner, J. Magn. Magn. Mater. **40**, 12 (1983).
[15] J. Smit, Physica (Amsterdam) **17**, 612 (1951).
[16] O. Jaol, I. A. Campbell, and A. Fert, J. Magn. Magn. Mater. **5**, 23 (1977).
[17] H. Nagasawa, Phys. Lett. **41A**, 39 (1972).
[18] Hiroshi Yamada and Satoshi Takada, J. Phys. Soc. Jpn. **34**, 51 (1973).
[19] A. K. Majumdar and P. v. Blanckenhagen, Phys. Rev. B **29**, 4079 (1984).
[20] A. Z. Men'shikov and A. Ye. Teplykh, Fiz. Met. Metalloved. **44**, 1215 (1977).
[21] A. K. Majumdar and P. v. Blanckenhagen, J. Magn. Magn. Mater. **40**, 227 (1983).
[22] T. K. Nath and A. K. Majumdar, J. Appl. Phys. **70**, 5828 (1991).
[23] T. K. Nath, N. Sudhakar, E. J. Mcniff, and A. K. Majumdar, Phys. Rev. B **55**, 12 389 (1997).
[24] T. Sakakibara, T. Goto, and Y. Miyako, Solid State Commun. **58**, 563 (1986).
[25] J. J. Smit, G. J. Nienwenhuys, and L. J. de Jough, Solid State Commun. **32**, 233 (1979).
[26] T. K. Nath and A. K. Majumdar, Phys. Rev. B **53**, 12 148 (1996).
[27] T. K. Nath, Ph.D. thesis, Indian Institute of Technology, Kanpur, India, 1997.
[28] I. Balberg, Physica B & C **91**, 71 (1977).
[29] S. Radha, Ph.D. thesis, Tata Institute of Fundamental Research, Bombay, India, 1995.

[30] A. K. Gangopadhyay, S. B. Roy, and A. K. Majumdar, Phys. Rev. B **33**, 5010 (1986).
[31] T. Bitoh, T. Shirane, and S. Chikazawa, J. Phys. Soc. Jpn. **62**, 2837 (1993).
[32] J. C. Le Guillou and J. Zinn-Justin, Phys. Rev. B **21**, 3976 (1980).
[33] M. T. Béal Monod and R. A. Weiner, Phys. Rev. **170**, 552 (1968).
[34] B. L. Altshuler, A. G. Aronov, A. I. Larkin, and D. E. Khemel'nitskii, Sov. Phys. JETP **54**, 411 (1981).
[35] A. Kawabata, J. Phys. Soc. Jpn. **49**, 628 (1980).
[36] B. L. Altshuler and A. G. Aronov, *in Electron-Electron Interaction in Disordered Systems*, edited by A. L. Efros and M. Pollak (Elsevier, New York, 1985), p. 1.
[37] A. Z. Men'shikov, N. N. Kuz'min, V. A. Kazantsev, S. K. Sidorov, and V. N. Kalinin, Phys. Met. Metallogr. **40**, 174 (1975).