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Low-field magnetoresistance in diverse magnetic phases of γ-$Fe_{80-x}Ni_xCr_{20}$ alloys

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We have measured the low-field (<30 G) magnetoresistance (LFMR) in the spin-glass (SG), ferromagnetic (FM), and the reentrant spin-glass (RSG) phases of fcc $Fe_{80-x}Ni_xCr_{20}$ ($19 \leqslant x \leqslant 30$) alloys. We find close resemblance between the temperature variation of the LFMR and the ac susceptibility in the RSG ($x=26$). Hysteresis effects in the LFMR have been found in the RSG for both increasing and decreasing temperatures. The presence of a dc biasing field at different temperatures alters the sign of the LFMR in the RSG at the lowest temperatures. This supports the existence of mixed FM and SG phases at the lowest temperature in the RSG. In the SG ($x=19$), the LFMR becomes negative, while in the FM ($x=30$), it remains positive. These measurements of LFMR throw new light on our understanding of the various couplings between the moments in such magnetic systems. [S0163-1829(97)05913-4]

I. INTRODUCTION

The application of a magnetic field usually affects the transport properties of a system. The magnetoresistance (MR) of a system depends upon the relative orientation of the electric- and magnetic-field vectors. It is regarded as one of the most powerful tools for probing into the electronic transport processes. The MR depends upon the magnetic state of the system, hence in magnetic alloys it can give greater insight to the understanding of the magnetic phases of such systems.[1]

There are different mechanisms that cause the variation of MR with field and temperature. When the magnetic field $\boldsymbol{B}$ is perpendicular to the current density $\boldsymbol{J}$, the MR is known as the transverse magnetoresistance (TMR), the space charge trajectories are deviated from the paths of least resistance by the Lorentz force proportional to $\boldsymbol{J}\times\boldsymbol{B}$. Since the new paths are no longer necessarily the paths of least resistance, the effective mean free path (l) and the scattering life-time (τ) decrease. This gives a TMR $\Delta\rho/\rho(0)\approx(l/r_L)^2$, where l is the mean free path proportional to τ and $r_L=m^*v/eB$ is the Larmor radius of a charge. This gives the quadratic dependence in B of the TMR. This can be expressed in the general form, known as Kohler's rule, $\Delta\rho/\rho(0)\propto(\omega_c\tau)^2$, where ω_c

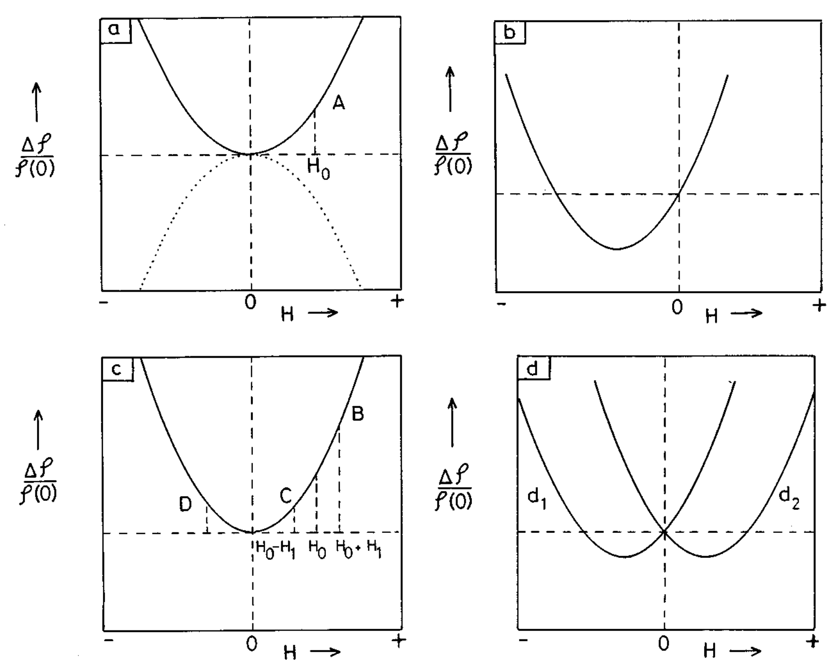

FIG. 1. (a) Variation of MR with field which goes as H^2 with the origin as the static point. (b) Presence of a constant field (H_0) in the sample shifts the static point to A from the origin and the MR becomes asymmetric. (c) If there is an internal field present in the sample and an external field $\pm H_1$ is applied, then the static point will shift from the origin to B and C, respectively. If H_1 is large, then the static point will shift from C to D. (d) d_1 and d_2 are the variation of the MR with field for the static points at C and D, respectively.

($=eB/m^*$) is the cyclotron frequency and for weak magnetic fields $\omega_c\tau<1$. The free-electron theory gives zero MR in the first approximation, whereas in the second approximation, it predicts a quadratic variation for small fields and a saturation in large fields.[2] Similar behavior of the TMR was predicted from the two-band model in cases where two overlapping bands exist as, for example, in transition metals.[3] However, TMR can also be understood in terms of the anisotropic relaxation times and non-spherical Fermi surfaces.[4]

In a metal with magnetic spins, additional quantum-mechanical effects like the suppression of the spin-flip scattering or weak localization can give rise to a negative MR. A conduction electron scatters by exchanging spins with magnetic moments or spin excitations. An external magnetic field increases the energy needed to flip a spin and thus decreases the amplitude of spin-flip scattering. This causes a decrease of resistivity in the presence of a magnetic field, resulting in a negative MR. Abrikosov[5] predicted the variation of MR as $\Delta\rho/\rho(0)=-\alpha(\mu H)^2$, where α is a positive constant and μ is the moment per magnetic scatterer, by considering the weak-field spin-flip scattering amplitude between an electron and a single local magnetic moment. The spin-flip scattering from exchange between conduction electrons and spin excitations in an itinerant ferromagnet was considered by Herring.[6] Mookerjee[7] calculated the MR in spin-glasses (SG's) on the basis of an Edwards-Anderson-type model by considering the fact that local spins interact through the conduction electrons via the s-d exchange coupling. He predicted a negative MR at all temperatures and fields and an H^2 variation in low fields. This was verified experimentally by Nigam and Majumdar[8] in canonical SG's. However, in a ferromagnet (FM) the presence of spontaneous magnetization and domain wall movement tend to further complicate the interpretation.

In this paper we present data on the low-field magnetoresistance (LFMR) of $Fe_{80-x}Ni_xCr_{20}$ alloys where very strong competing exchange interactions produce exotic and diverse magnetic phases within the same crystallographic γ *phase*.[9] These alloys exhibit a compositional phase transition from a long-range antiferromagnetic phase ($x=14$) to a long-range FM one ($x=30$), passing through intermediate phases of SG ($x=19$) and reentrant SG (RSG) ($x=23$ and 26) with increase in Ni concentration. On cooling, the RSG samples pass through more than two magnetic phases and hence the variation of the LFMR of these samples may not necessarily follow any of the above mechanisms. However, they are expected to show some complex interplay of the above mechanisms. The LFMR is closely connected with the state of the magnetization of the alloy and is capable of providing more subtle information than say, direct magnetization measurements. Barnard[10] observed that the LFMR (in Au–8 at. % Mn and Cu–4.6 at. % Mn) showed peaks at the freezing temperature T_g and is negative for both the paramagnetic (PM) and the SG phases, becoming positive when FM ordering is present. Thus, LFMR can provide very useful information about the nature of the coupling between the moments in metallic magnetic alloys. To reveal the intrinsic differences in the magnetic properties of SG's and RSG's, it is necessary to do the measurements in very low fields, for larger magnetic fields can often severely disrupt the rather weak magnetic coupling existing in such alloys. Sometimes the application of larger fields is advantageous in that they might saturate a FM component, enabling weaker SG effects to be revealed. The RSG sample shows a typical PM to FM transition. As the temperature is lowered, the FM phase becomes unstable against further reduction of temperature and reenters a SG-like state in which there is a drop in the ac susceptibility and other magnetic effects show up, like the onset of magnetic viscosity and history-dependent effects. There are many controversial predictions about the nature of this lower-temperature phase. Several important questions still remain unanswered. For example, is there any difference between the SG and the RSG phases at the lowest temperatures? Does FM ordering exist in the RSG phase down to the lowest temperatures? Is the transition near the Curie temperature T_C in the RSG similar to the FM to PM transition? In an attempt to answer these questions we have combined ac susceptibility measurements with very low field magnetoresistance measurements, thereby providing insights into the magnetic behavior of these alloys.

II. EXPERIMENTAL PROCEDURE

All these ternary alloys were prepared by induction melting in an argon atmosphere. The starting materials were of 99.999% purity obtained from M/s Johnson Matthey Inc., England. The alloys were cut to the required size, homogenized at 1323 K for 100 h in an argon atmosphere, and then quenched in oil.

Chemical analysis of Ni and Cr shows that the compositions of the alloys are within ±0.5 at. % of their nominal values. X-ray diffraction data at room temperature in powdered samples reveal that these are single-phase fcc (γ) alloys with lattice parameter $a=3.60$ Å. Neutron diffraction data[9] show the presence of single-phase fcc (γ) structure down to 2 K for the alloy with $x=19$.

In a metallic system, the change of electrical resistivity in the presence of a magnetic field is exceedingly small for a small field (<30 G). To detect the minute change of MR in fields comparable with those used in ac susceptibility experiments, Barnard devised a state of the art instrument which can measure LFMR with a resolution of 10^{-8}.[11] The LFMR is normally obtained by measuring the voltage developed across a current-carrying sample with and without the presence of a magnetic field, applied either in a longitudinal or transverse direction with respect to the current. Now the LFMR can be defined as the ratio of the change of the voltage ΔV ($\propto\Delta\rho$) to the voltage in zero field, $[V(B)-V(B=0)]/V(B=0)=\Delta\rho/\rho(0)$. For low fields ΔV becomes very small ($\Delta V\propto B^2$), and the detection of this against a large background voltage (IR) is very difficult. The fundamental feature of this method is to separate ΔV from V. If a very stable dc current (from batteries) passes through the sample and an unidirectional pulsed or oscillating magnetic field is applied, then the change of the voltage in the presence of the oscillatory field will also be oscillatory in nature. This small ac voltage could then be amplified and measured using a lock-in amplifier. Hence very small LFMR voltages could be detected which are not coupled with the large dc voltage across the sample. Full details of the method used were reported earlier.[11] We have measured the LFMR in two different modes with the field always applied parallel to the direction of the current, i.e., the longitudinal magnetoresistance.

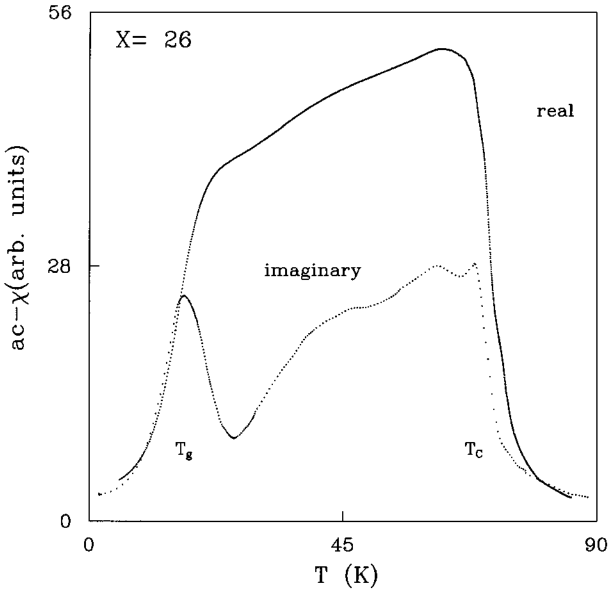

FIG. 2. Temperature variation of the real and imaginary part of the ac susceptibility for $x=26$ measured in a field of 242 Hz and 0.6 G.

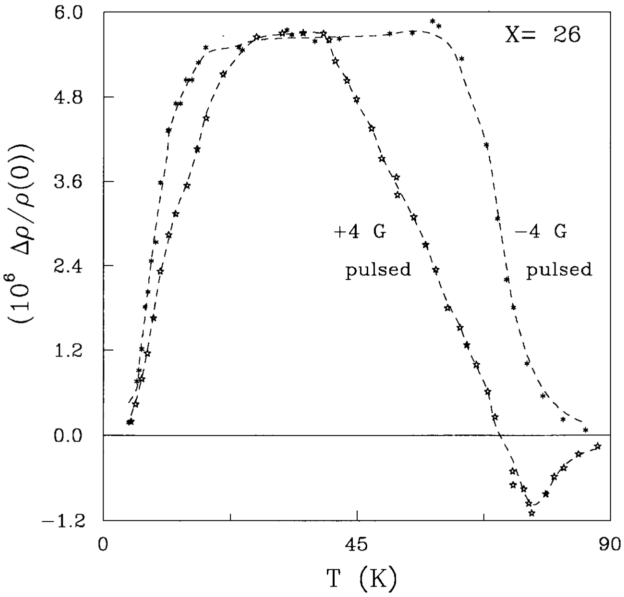

FIG. 3. Temperature variation of the LFMR for $x=26$ measured in $\pm$4-G pulsed square-wave field.

In one case a unidirectional square-wave pulse produced the magnetic field whose amplitude could be varied. In the other case a small constant square-wave pulse was applied with an additional dc biasing field whose magnitude could be varied.

III. RESULTS

Usually when the LFMR is measured for both forward and reverse pulsed magnetic fields, these values are equal, i.e., the LFMR is symmetric for both the field directions. However, if some finite magnetization exists in the sample, which modifies the actual field seen by the electrons, then the LFMR becomes asymmetric with respect to the field directions. In such cases, considerable care must be taken to determine the sign of the LFMR. To understand this, it will be convenient to consider a material in which the intrinsic LFMR is proportional to the square of the magnetization (M). Generally, for small fields $M \propto H$ and hence the LFMR is proportional to H^2. Here whether H is positive or negative, a positive LFMR results. The same variation will also exist when the H field is a pulsed square wave as in our experiments. This is the simplest system where the same variation is observed on both increasing or decreasing H. Under these conditions the sign of the LFMR is unambiguous. This case is shown in Fig. 1(a) where the LFMR is clearly positive, but obviously negative LFMR is also possible (shown by the dotted curve). In the above, the *static point* of the sample is the origin where no field is applied to the sample. However, if we consider an additional constant field H_0 which might be present in the sample, the *static point* is now A, and the pulsed field will be with respect to this point. The observed variation of LFMR will now look like that shown in Fig. 1(b) in which apparently there is a negative LFMR, though no *intrinsic* negative LFMR is really associated with the sample. It should be mentioned here that the field H_0 may not necessarily be an externally applied field; it may be an internal field present in the sample. In such cases, the determination of the sign of the LFMR (intrinsic) can be difficult.

Let us now examine the case where an internal field H_0 is present and an external biasing field H_1 is also applied, which may be positive or negative [Fig. 1(c)]. With H_0 and positive H_1 we observe a *static point* at B, while if H_1 is negative, then point C will be the static point. If H_1 is large and negative, then the static point C could move to D. The variation of the LFMR for the static points C and D will look as shown in Fig. 1(d), curves d_1 and d_2, respectively. So to find the intrinsic LFMR, one has to choose the static point in a way that will enable one to observe the effect of only the external magnetic field which, in our case, is unidirectional pulsed field of frequency 38 Hz, and not of the internal field or any other biasing field present prior to the application of the pulsed external field. If there is no internal field present in the sample, then the LFMR will be symmetric with respect to the origin for small fields and the origin *will* be the proper static point. If the LFMR is not symmetric with respect to the origin indicating the presence of some extra field (internal), then one has to apply an appropriate biasing field H_1 in order to nullify the effect of the internal field on the LFMR. This will *make* the LFMR symmetric with respect to the origin. In other words, one has to shift the axes of the graph $\Delta\rho/\rho(0)$ vs H in such a way that the LFMR becomes symmetric with respect to the new origin, and this point is called the proper *static point*.

The sample with $x=26$ undergoes a double transition, one from PM to FM at 68.89 K and the other from FM to RSG at 16.68 K on further lowering of temperature. These double transitions are clearly seen from the ac susceptibility (χ) measurements. The real part of the susceptibility shows a double knee, while the imaginary part shows two distinct peaks at T_C and T_g, respectively (Fig. 2). The temperature variation of the LFMR also shows somewhat similar behavior (Fig. 3) to the real part of the susceptibility. According to Kohler's rule for MR, $\Delta\rho/\rho(0)=f(B/\rho(0))$. In magnetic

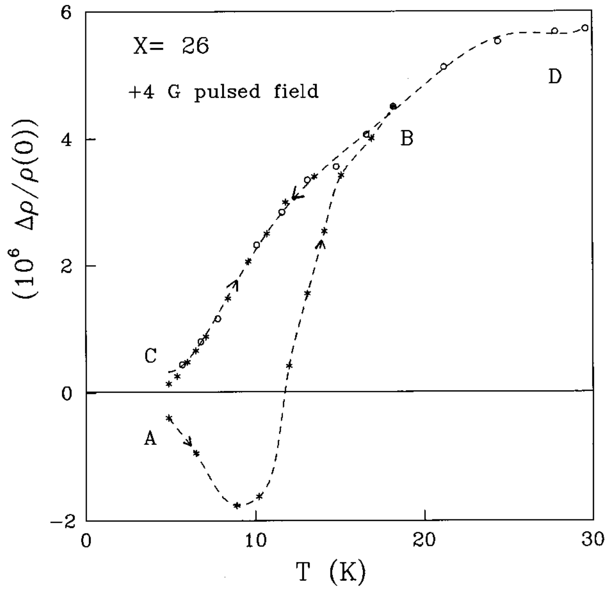

FIG. 4. Temperature variation of the LFMR for $x=26$ for various thermal cycles measured in $+4$-G pulsed square-wave field. Initially the temperature was increased from A to B, then it was decreased to C and then increased again to D. The arrows in the figure show the cycles.

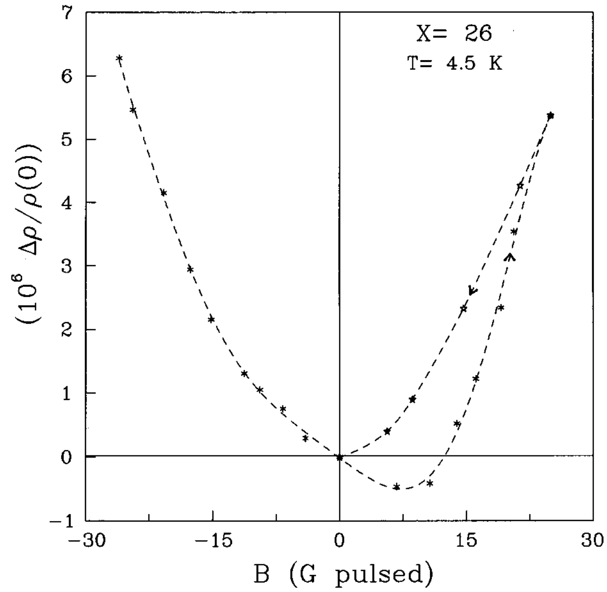

FIG. 5. Variation of the LFMR for $x=26$ at 4.5 K with the increase and decrease of pulsed square-wave fields (the arrow indicating the direction).

samples, B is no longer equal to H, but $B=H+KM=H(1+K\chi)$, where K is a constant. Therefore for constant H, MR should be a function of χ, i.e., $\Delta\rho/\rho(0)=f(\chi)$. $\rho(0)$ has its own temperature dependence which is very weak compared to that of χ in the interval of 4.2–80 K where $\rho(0)$ changes by only $\approx$4%. So the temperature variation of $\rho(0)$ can be neglected. Hence there must be some relation between the temperature variation of MR and χ. We have also observed that in the other RSG sample ($x=23$), the temperature variation of susceptibility and LFMR follow a similar behavior (not shown), indicating a close relationship between the two. Similar behavior was also reported in FeNiMn RSG by Barnard *et al.*[12]

The most interesting feature of the LFMR of $x=26$ is that it shows two different curves for $+4$ and -4 G square-wave fields (Fig. 3). To obtain these curves, we have cooled the sample in unidirectional square-wave pulsed fields of ±4 G and frequency 38 Hz. This occurs because of the presence of some internal field which modifies the field seen by the sample and is discussed later in this paper. The internal field is also a function of temperature. In the temperature interval between 25 and 39 K it seems that the internal field vanishes as the two curves coincide. For $+4$-G field, the LFMR becomes negative around 71 K, which is above the T_C obtained from the ac-susceptibility measurements (details to be published elsewhere). Similar behavior has been observed in other RSG's [(AuFe, $(Fe_{0.65}Ni_{0.35})_{1-x}Mn_x$, $x=11.36$ at. %][10,12] near the PM to FM transition.

The LFMR shows thermal hysteresis effects at lower temperatures for a $+4$-G pulse square-wave field (Fig. 4). To see thermal hysteresis, we have cooled the sample down to 4.2 K in a zero field and then applied a $+4$-G pulsed square-wave field of frequency 38 Hz. Then we increase the temperature up to 20 K ($A\rightarrow B$ of Fig. 4), then cool it down slowly to 4.2 K ($B\rightarrow C$) and again increased the temperature up to 30 K ($C\rightarrow D$). Throughout the operation ($A\rightarrow B\rightarrow C\rightarrow D$), the square-wave pulsed field of $+4$ G was on. So the curve $C\rightarrow D$ of Fig. 4 is essentially the same as shown in Fig. 3 for $+4$ G-field up to 30 K. With the increase of temperature the curve shows a negative LFMR, a feature generally associated with the SG state, then a negative peak around 10 K, followed by a sharp increase, and finally, it becomes positive. On reducing the temperature, it starts bifurcating at 17 K, but retains the positive value to the lowest temperature. Afterwards, it shows a reversible behavior with thermal cycling. It is to be noted that thermal hysteresis is only observed in the RSG phase, and not in the FM phase ($T>T_g$) with this small applied field. The negative peak around 10 K is a finding which we could not trace in our susceptibility measurements. The magneto-thermal-history effect in SG's has also been reported by Rakers and Beck.[13] To further investigate the magnetic hysteresis effect we measured the LFMR with varying pulsed field at 4.5 K (Fig. 5). It shows hysteresis and for the small field the LFMR is negative. We think that the presence of small (negative) internal fields shifts the *static point* and makes the LFMR apparently negative, but the intrinsic MR always remains positive. Here the magnetic history effect is somewhat similar to the thermal history effect. After one cycle, it follows a reversible path. Initially, the LFMR is positive above $B\approx10$ G. On reducing the field, the LFMR always remained positive and reduced to zero as the field was reduced to zero. Then for a negative field, it followed a reversible path. Hysteresis effects were also observed in other RSG samples,[12] where their appearance for smaller fields, followed by a reversible behavior at higher fields, further complicated the situation. Figure 6 shows the variation of the LFMR at 4.5 K in the presence of different static fields on top of the variable pulsed field. In the presence of ±18.2-G static fields, it shows a negative LFMR along with hysteresis effects. The curves are also not symmetric for the positive and negative applied fields. In the

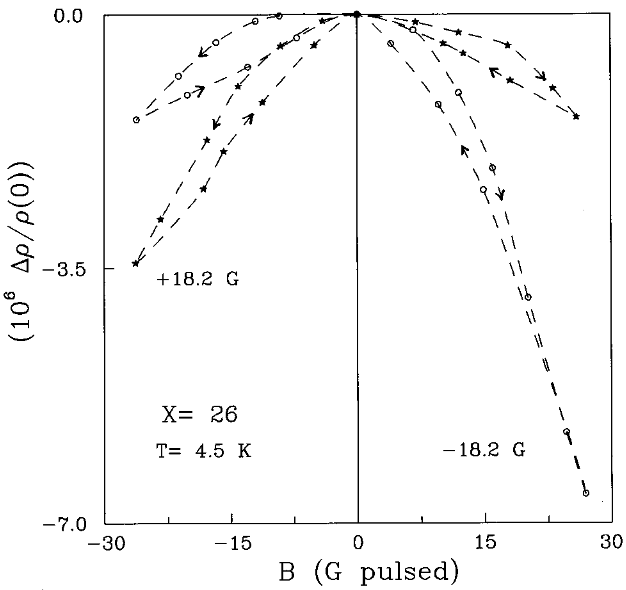

FIG. 6. Variation of the LFMR for $x=26$ at 4.5 K with the pulsed square-wave field in the presence of ±18.2-G static biasing field. The biasing field makes the LFMR negative.

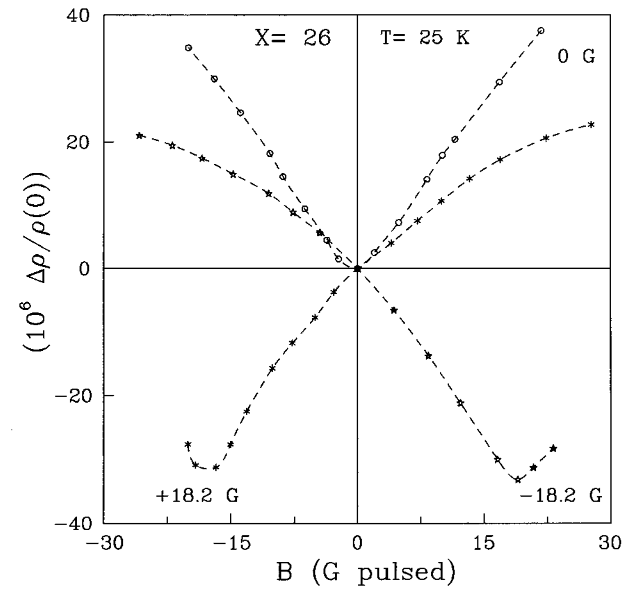

FIG. 7. Variation of the LFMR for $x=26$ at 25 K with the pulsed square-wave field in the presence of 0- and ±18.2-G static biasing field.

RSG phase, both FM and SG types of clusters are present, and the LFMR is the combined effect of all these clusters. In a zero static field, the LFMR arising from the FM clusters predominates and makes the resultant LFMR positive. But the application of a sufficiently high static field effectively locks the FM clusters, and only the SG component can then follow the pulsed field, which reveals itself as a negative MR. Hence, the LFMR provides strong evidence for the presence of FM ordering down to the lowest temperature, a conclusion we had reached earlier from our magnetic relaxation and ac susceptibility measurements.[14] However, the reason behind the asymmetry in the curves for positive and negative pulsed fields and the exact nature of the different types of clusters present and their interactions seems to be quite complicated. One possible explanation is that the ±18.2-G static fields are insufficient to fully saturate the FM component, but nevertheless are sufficient to block the rest of the positive LFMR. That there is still some hysteresis in the curves shown in Fig. 6 strongly suggests near, but not complete, saturation.

We concluded earlier that the internal field was absent at around 25 K. Further measurements on $x=26$ at 25 K of the variation of the LFMR in zero and ±18.2-G biasing fields are shown in Fig. 7. For zero biasing field, the LFMR is always positive, and the curve is symmetrically placed about the origin. With the biasing field of ±18.2 G, the LFMR curves are shifted by equal amounts on either side of the origin. By choosing the proper *static point* through shifting of the graphing axes discussed earlier, we can show that here the LFMR is positive. At 25 K, the sample is in a FM phase, hence a positive LFMR is expected. However, in the presence of ±18.2-G biasing field, the LFMR indicates saturation for higher fields. It may be that at higher fields it reverses its slope and becomes negative. Our high field data (0.1–17 kG) for the other RSG sample ($x=23$) shows negative MR in the FM phase.[15] Therefore it is very important to do low-field measurements to reveal the true nature of the magnetic state. It is only through the positive LFMR that we are able to conclude that the ferromagnetic phase persists even below the lower transition temperature in the RSG. At still higher fields (up to 7 T), Banerjee and Raychaudhuri[16] have shown that the MR can be expressed as $\Delta\rho/\rho(0)=A(T)H^2-B(T)H^m$, where $A(T)$ and $B(T)$ are constants at a given temperature and the exponent, m, is less than 1. They concluded that the negative contribution has a magnetic origin.

The measurements at 78 K, in the PM region, show negative MR (Fig. 8). But to get a symmetric curve we need to

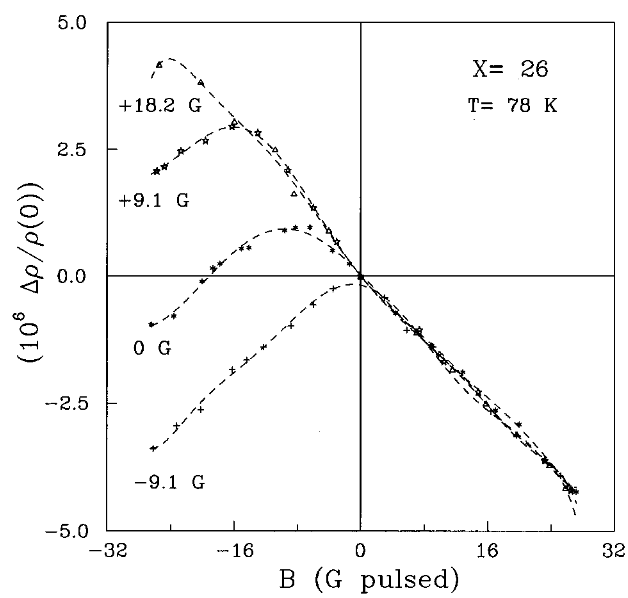

FIG. 8. Variation of the LFMR for $x=26$ at 78 K with the pulsed square-wave field in the presence of +18.2-, +9.1-, 0-, and −9.1-G static biasing fields. −9.1-G biasing field brings the static point to the origin.

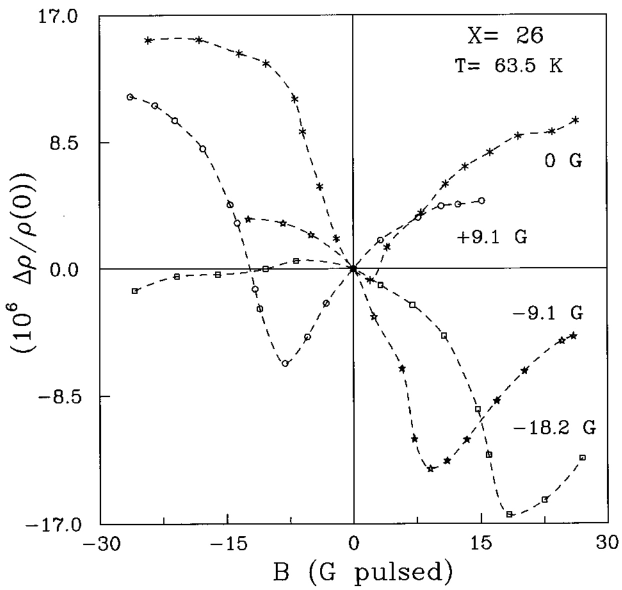

FIG. 9. Variation of the LFMR for $x=26$ at 63.5 K with the pulsed square-wave field in the presence of +9.1-, 0-, −9.1-, and −18.2-G static biasing fields.

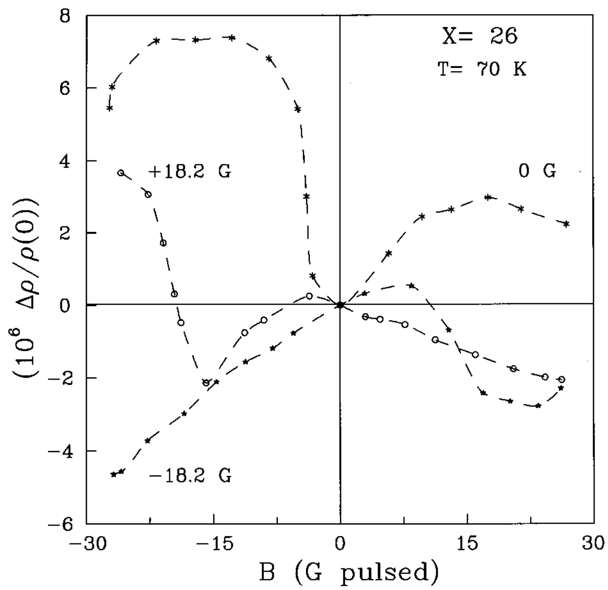

FIG. 10. Variation of the LFMR for $x=26$ at 70 K with the pulsed square-wave field in the presence of 0- and ±18.2-G static biasing fields.

apply a −9.1-G biasing field. For zero and other biasing fields, we have to shift the static points to get the intrinsic LFMR, which is negative in this case. So in the PM region, we observe a negative LFMR and a small internal field of about 9 G. At 4.5 K the LFMR curve (LFMR intrinsically positive) is shifted to the right of the origin for zero bias (Fig. 5), but here it shifts to the left of the origin.

Figure 9 depicts the variation of the LFMR at 63.5 K for zero and some biasing fields. At this temperature, the LFMR starts showing asymmetric behavior. For zero biasing field, the static point is very close to the origin indicating the presence of a very small (negative) internal field. The LFMR initially increases with field, but around 30 G it shows a tendency to saturate. With positive and negative 9.1-G biasing fields, the LFMR shifts by unequal amounts along the $(-\Delta\rho/\rho(0))$ axis, unlike that at 25 K (Fig. 7). For −18.2-G biasing field, the static point is now on the right of the origin and is ≈18 G away along the B axis. With respect to this new origin, the slope of the LFMR curve is positive. But when the field increases along the negative direction, the LFMR reverses its slope and finally becomes negative. As we have already applied a biasing field of −18.2 G, the effective field along the negative direction is still larger. The reason behind this asymmetric behavior of the LFMR for positive and negative fields is not clear.

At 70 K, very close to the critical temperature, the LFMR has a very complicated behavior. Figure 10 shows the variation of the LFMR for different biasing fields. The LFMR is not symmetric, nor can a graphical shift of the static point make it symmetric. Therefore the choice of the static point is very difficult here. Very close to the critical temperature (T_c), the complex interplay of different magnetic constituents and the absence of any pure magnetic phase further complicates the situation. According to our criterion, for zero biasing field, the origin is the so-called static point. Hence for positive biasing fields, the static point is expected to be on the left of the origin and on the right for negative biasing fields. For smaller fields (zero bias case) the slope of the LFMR is positive, but around −20-G field, it reverses its slope indicating a negative LFMR. So, even in the zero bias, the LFMR can have both positive and the negative components. In the presence of a +18.2-G biasing field, the LFMR indeed becomes positive (seen after choosing the proper static point which is now at the left of the origin). But with the increase of the field along the positive direction, the LFMR becomes negative. Actually, when the static point shifts towards the left of the origin, the actual field seen by the sample is larger than the applied field along the positive direction. The nature of the LFMR curves is different for positive and negative 18-G biasing fields. At this point it seems that the LFMR has two components, one positive and the other negative. Starting from the high-temperature side, prior to the establishment of the FM phase, there may be a mixed phase where a PM or even a SG phase coexists with a FM phase. The positive contribution is coming from the FM ordering while the PM or SG ordering gives a negative LFMR. However, the resulting nature of the LFMR curve will depend on their relative contributions. Here the LFMR for zero bias is somewhat similar to that observed in $(Fe_{0.08}Ni_{0.92})_{77}Si_{10}B_{13}$ RSG near T_c.[10]

Figure 11 shows the variation of the LFMR for $x=30$ at 78 K ($T_c=130$ K). $x=30$ is a ferromagnet, and thus the LFMR is positive and symmetric for both the positive and negative pulsed fields. It also does not show any hysteresis effects for the small applied fields used. In the FM phase where long-range order exists, smaller fields do not show nonlinear effects which give rise to hysteresis in magnetization and therefore in magnetoresistance. For small fields, the LFMR in $x=30$ shows reversible paths, unlike the SG phase, where at lower temperatures we have observed that the LFMR in $x=26$ follows irreversible paths for small fields.

Figure 12 shows the temperature variation of the LFMR for the sample with $x=19$, which is a SG. For small applied fields, the LFMR is very small and it is about the limit of our

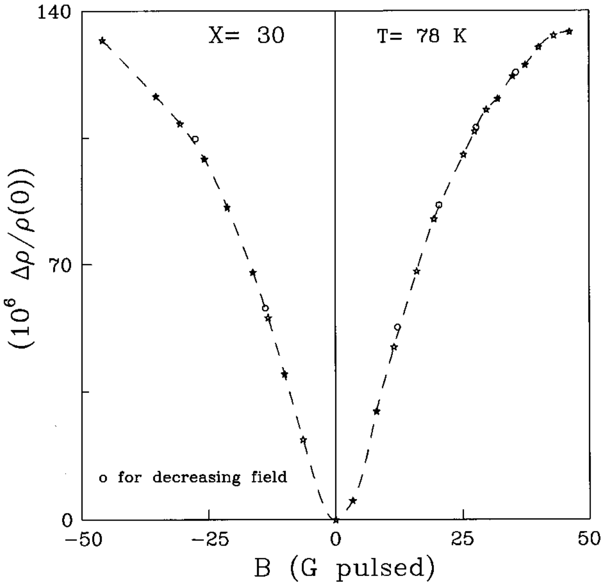

FIG. 11. Variation of the LFMR with pulsed square-wave field for $x=30$ (FM) measured at 78 K in a field of 38 Hz.

experimental resolution. We get rather scattered data, even after applying a pulsed field of 20 G. We can barely resolve a dip at the transition temperature. Nevertheless, we can definitely conclude that the LFMR in $x=19$ remains negative both in the SG and the PM phase (up to $\approx 2T_g$).

IV. DISCUSSION

The LFMR results in the RSG ($x=26$) presented in this paper under very low field conditions (<30 G) show at 4.2 K, a negative LFMR, a characteristic of a SG. It has a negative dip around 10 K, then it increases and finally becomes positive. But the LFMR retains a positive value when the temperature is lowered with the field on. For the RSG ($x=26$) at 4.5 K and zero bias, the intrinsic LFMR is positive. If the lower temperature phase is a pure SG, then the LFMR should be negative. So the presence of FM clusters at the lower temperature is strongly suggested. Now, to see the SG-type of behavior we have to suppress or block the contributions coming from the FM clusters. Application of a biasing field locks the FM clusters allowing the SG component, which is less affected by dc fields, to be revealed. This results in a negative LFMR. We measured the nonlinear ac susceptibility, which shows a peak around T_g.[17] This peak can be observed only if a spontaneous magnetization is present. Moreover, the presence of small internal fields, causing a shift of the static point in the LFMR curve at the lower temperature, supports the above proposition in the RSG. Thus the lower temperature phase of the sample with $x=26$ consists of both SG and FM types of ordering. In this phase small FM clusters are embedded into the matrix of frustrated SG-like spins. We define this phase as the RSG, which is distinctly different from the SG.

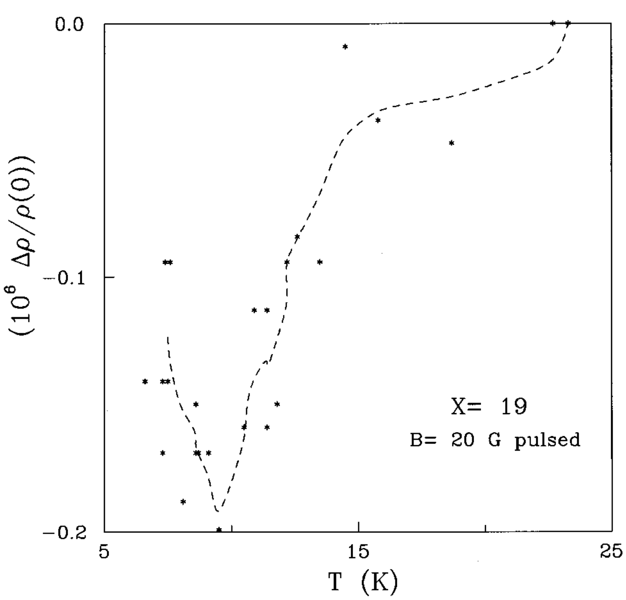

FIG. 12. Temperature variation of the LFMR for $x=19$ (SG) measured in a 20-G pulsed square-wave field.

At temperatures above T_g, the LFMR becomes positive for both nonzero and zero biasing fields, indicating a pure FM phase. However, at higher temperatures, close to the critical temperature T_c, we observed a tendency towards a negative LFMR for higher fields. In the presence of biasing fields, the LFMR can have both positive and negative values. We know that the positive LFMR arises out of the FM clusters, and the negative contribution comes from either the SG or the PM type of ordering. If the FM ordering coexists with the PM (SG) type of random spins, then the resultant LFMR can be of either sign, determined by the relative contributions of the different spin orderings. Normally for smaller fields, the contribution from the FM component dominates, but at higher fields or in the presence of biasing fields, the FM part is locked, and then the negative contribution becomes more prominent. We observed similar behavior around T_C in the RSG ($x=26$). So, near the critical point, both above and below, when the sample ($x=26$) passes from the PM to the FM phase with the lowering of temperature, it passes through an intermediate phase where FM clusters coexist with PM (SG) spins. It is a new kind of phase which exists for a very narrow temperature interval prior to the onset of the FM phase. A similar kind of phase was also predicted earlier near T_C in FeNiMn RSG.[12] At 78 K ($T>T_C$), the LFMR becomes negative, indicative of a PM (SG) phase. So from the present LFMR measurements, we propose a new phase of FeNiCr alloys which exists for a very narrow temperature interval near T_C. This kind of phase is possible where there are very strong competing interactions in a magnetic system. The final nature of the phase depends upon the more dominant interaction. If the strengths of the interactions are comparable, then this kind of mixed phase may be possible. However, we could not trace this kind of phase from our earlier dc and ac magnetic measurements. So from these LFMR measurements, we conclude that the sample with $x=26$ passes through diverse magnetic phases like, PM→[PM(SG)+FM]→FM→RSG(=FM+SG) with the lowering of temperature.

V. CONCLUSION

We have measured the LFMR in different magnetic phases of $Fe_{80-x}Ni_xCr_{20}$ alloys. We observe that the LFMR is negative in the SG ($x=19$) and the PM phases and positive in the FM ($x=30$) phase. In the RSG ($x=26$), at the lowest temperature (4.5 K), the intrinsic LFMR is positive for zero biasing field, but the presence of a 18.2-G static biasing field makes the LFMR negative, a feature generally associated with the SG state. This suggests that the lowest temperature phase of the RSG has mixed FM and SG types of coupling. We believe that small FM clusters are embedded into a matrix of frustrated spins of the SG. We observe hysteresis effects in the LFMR for small applied fields at the lowest temperature for the RSG sample. But such small fields do not produce any hysteresis effect in the FM phase ($x=30$). We find that the LFMR becomes negative around T_C for a $+4$-G pulsed field in the RSG. From the LFMR measurements, we conclude that when the RSG sample ($x=26$) goes from the PM to the FM phase with lowering temperature, it passes through an intermediate phase. Here the PM (SG) phase coexists with the FM phase for a very narrow temperature interval. So with the reduction of temperature, the RSG sample passes through various magnetic phases like, PM→[PM(SG) +FM]→FM→RSG(=FM+SG). We also find a close resemblance between the temperature variation of the LFMR and the ac susceptibility in the RSG ($x=26$).

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