Giant magnetoresistance in the disordered magnetic alloy $(FeNi)_{25}Au_{75}$

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(Received 9 January 1997; revised mansuscript received 12 March 1997)

We present the results of detailed measurements of the field and temperature dependence of the magnetization and the resistivity of the $(FeNi)_{25}Au_{75}$ alloy, belonging to the fcc $(FeNi)_{100-x}Au_x$ ($x=25$, 45, 60, 75) system. Giant magnetoresistance (GMR) has been observed only in the $x=75$ alloy, where the magnetic relaxation, thermoremanent magnetization, and hysteresis data suggest reentrant spin-glass-like behavior at low temperatures. The origin of GMR in $(FeNi)_{25}Au_{75}$ is attributed to the field-induced alignment of Fe moments. The data suggest a subtle link between frustration and GMR in disordered magnetic materials. [S0163-1829(97)06534-X]

I. INTRODUCTION

Frustration due to competing interactions in disordered systems (substitution, chemical) and due to crystallographic structure in nondisordered compounds (*Kagomé*, pyrochlore structure) leads to interesting macroscopic properties. It is worthwhile to investigate such systems for the possibility of spin-glass-like behavior. Reentrant behavior has been found in a variety of disordered magnetic materials in which there is competition between spin-glass and long-range ferromagnetic ordering. When the temperature is lowered in such materials they often exhibit a transition from the paramagnetic (PM) to the ferromagnetic (FM) phase at the Curie temperature T_C, and on further lowering the temperature typical spin-glass [commonly called reentrant-spin-glass (RSG)] behavior appears. It is clear that below T_C the spins become locally *canted* and in zero field a FM domain structure exists at lower temperatures below T_C, so that ferromagnetic and spin-glass order coexist. This interpretation is based on a model for magnetic structure based on the transverse spin freezing approach.[1] Reentrant behavior was explored in detail in the Au-Fe alloys. Recently we proposed the fcc $(FeNi)_{25}Au_{75}$ alloy as a possible reentrant-spin-glass-like system.[2] We chose to study this alloy because only a limited attempt has been made to investigate spin-glass systems with two (or more) local moment species, each of which produces a well-defined spin-glass transition (T_f) when present alone. In this case Au-Fe and Ni-Fe are well-studied systems.[2] Recent observations of giant magnetoresistance (GMR) in spin-glass-like frustrated systems such as Cr-Fe bulk alloys,[3] metamagnetic materials,[4] and Au-rich Co-Au and Fe-Au bulk alloys[5] suggest that GMR can also be observed in conventional bulk materials. It may be mentioned that GMR has also been observed in superparamagnetic systems, where macroscopic properties like field-cooled, zero-field-cooled magnetisation irreversibility, etc., are similar to that of reentrant-spin-glass systems.[6] It has also been shown that in heterogeneous alloy systems consisting of single-domain clusters,[7–9] the resistance is high for a random alignment of magnetic cluster moments and decreases substantially when individual moments are aligned by an external field. Thus the GMR effect seems to be associated with the reorientation of magnetic cluster moments. The above results establish that GMR can be readily observed in magnetically inhomogeneous media with nonaligned ferromagnetic entities.

In order to understand GMR in bulk materials it will be useful to look for an alloy system in multidomain bulk form (not as a thin film or granular ribbon), which shows a lack of saturation due to nonaligned ferromagnetic entities. Such a situation can be realized if one focuses on *frustrated* disordered magnetic systems and their relevance to GMR. This approach is useful as bulk materials can be easily prepared by conventional techniques. Also, from the theoretical point of view, we need to understand the origin of GMR in these system as interfacial regions are absent unlike as in the multilayered samples.

In this paper we present the results of an experimental investigation of GMR along with the low-field field-cooled (FC), zero-field-cooled (ZFC) magnetizations, thermoremanent magnetization (TRM), magnetic relaxation, and field-cooled hysteresis, because the reentrant-spin-glass phenomenon is highly field dependent. An attempt has also been made to show the subtle link between frustration and GMR in bulk materials.

II. EXPERIMENT

Details of the sample preparation have been given elsewhere.[10] The same samples have been used here, as used in the work of Cable and Wollen. The sample is single phase, having fcc structure, and homogeneous to micrometer range as seen in x-ray-diffraction and electron microprobe analysis.[10] Magnetic properties of the sample $(FeNi)_{25}Au_{75}$ ($l=1.275$ cm, $b=0.2$ cm, $w=0.085$ cm, and $Wt=320.6$ mg) have been investigated using our homemade dc magnetometer.[11] The ZFC magnetization in a zero-field-cooled state is the magnetization under a measuring field H after the sample has been cooled from above T_C to the lowest temperature in zero field. For field-cooled magnetization the sample was cooled in the presence of the field. We have

measured FC and ZFC magnetization vs temperature in various dc fields ranging from 2 Oe to 30 Oe. Temperature variation of thermoremanent magnetization has been investigated by measuring the magnetization of the sample after removing the dc field in which the sample was cooled from a temperature above T_C to the lowest temperature for cooling field range 2–30 Oe. ZFC hysteresis experiments were done at different constant temperatures from 15 K to 100 K and the field was varied from -33 Oe to $+33$ Oe. Field-cooled hysteresis curves were obtained in the following way: The sample was cooled to the desired temperature under a low field in the range of 1–5 Oe (H_{FC}) and magnetization was recorded with the field cycle $0 \rightarrow -33$ Oe$\rightarrow 0 \rightarrow +33$ Oe. The coercive field H_C^{FC} in this case is the point at which the curve intersects the negative H axis during the first cycle. This process was repeated for different temperatures in the range 15–100 K. Time decay of the thermoremanent magnetization was measured in the following way: After the sample was cooled in a 10-Oe field from a temperature above T_C to the measurement temperature, the field was reduced to zero after a certain time t_w (called the waiting time) and the magnetization was subsequently recorded with time over a period of 10^3 s. This process was repeated for different temperatures in the range 22–120 K. Magnetization vs dc field (0–20 kOe) at 100 K was measured in vibrating sample magnetometer for different Au concentrations. Magnetoresistance was measured at different temperatures between 48 and 200 K by the conventional four-probe method with a field cycle of 0 to ± 8 kOe.

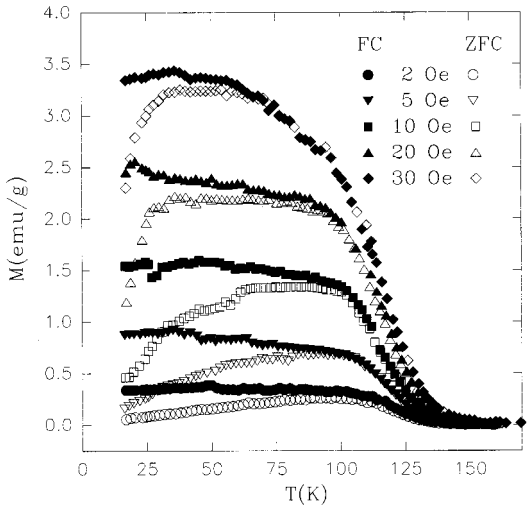

FIG. 1. Field-cooled and zero-field-cooled magnetization vs temperature for different dc fields.

III. SYSTEM

It has been found that for binary alloys such as Ni-Cu, Ni-Au, and Fe-Au, the Fe atom retains its characteristic moment of about $3\mu_B$ even as a dilute impurity in Au whereas the Ni moment decreases and vanishes at sufficiently high dilution. $(\mathrm{FeNi})_{100-x}\mathrm{Au}_x$ ($x=0-75$) is an interesting system, where the magnetic behavior is largely determined by the Fe atoms.[10] The ferromagnetic Curie temperatures decrease from 745 K to 125 K with an increase of Au content from $x=0$ to 75 at.%. Neutron measurements show that μ_{Fe} (μ_B/Fe) is 3.03, 2.94, 2.04 and μ_{Ni} (μ_B/Ni) is 0.53, 0.66, 0.12 for $x=45$, 60, 75 at. % Au, respectively. It is interesting to note that unlike as in the case of the progressive decrease of $3d$ moment observed in Ni or Co when Au or Cu is added, here there is a rapid decrease of moment for $x>60$. It may be recalled that when an Fe atom is present as an impurity in Au it has a moment of $3\mu_B$, where as in the case of the $(\mathrm{FeNi})_{100-x}\mathrm{Au}_x$ alloy, for $x=75\%$ the Fe moment drops to $2\mu_B$. This Fe moment drop does not indicate the disappearance of the local Fe moment but rather the loss of an orderable ferromagnetic Fe moment, suggesting a field-dependent behavior.[10] In fact our earlier high-field data[2] along with low-field data to be reported here confirm this behavior. In other words we believe that by increasing the Au concentration the mean Fe-Fe distance becomes larger, reducing effective $J_{\mathrm{Fe\text{-}Fe}}$ and thereby the ordering temperature. This is consistent with the observed lower $T_C \sim 130$ K for the 75% Au alloy. This suggests the existence of a degree of nonalignment of the ferromagnetic Fe moments, an important criterion to search for, from a GMR point of view. Indeed spinodally decomposed 60% Cu–20% Fe–20% Ni shows a GMR effect of $\sim 6\%$ in the field range 60 kOe.[12] It may be noted here that the GMR effect is due to an extremely-fine-scale microstructure made up of only ferromagnetic phases in this (FeNi)Cu alloy. Due to decomposition, no systematic magnetic study is possible for a higher content of copper,[10] whereas for (FeNi)Au a systematic investigation of the magnetic properties is possible along with GMR data.

IV. RESULTS AND DISCUSSIONS

Our earlier reported low-field ac susceptibility data of $(\mathrm{FeNi})_{25}\mathrm{Au}_{75}$ show a paramagnetic-ferromagnetic transition onset at $T \sim 150$ K with a peak in χ'' (absorption term) and nonlinear susceptibility at $T_C \sim 130$ K. We observed a strong irreversibility at low temperatures of 5–15 K well below T_C depending upon the field of 50 Oe to 1 kOe, as found in other typical reentrant-spin-glass system.[1,2] No hysteresis loop was observed at 5 K, even though there is a weak hysteresis loop at $T<T_C$ (50–100 K) with apparent saturation at 55 kOe. Arrott plot of M^2 vs H/M in the temperature range 2–180 K shows field-induced ferromagnetism for a field >1 kOe. For a field >1 kOe no irreversibility, i.e., drop, in ZFC magnetization was observed, suggesting a highly-field-dependent magnetization in this system. In order to understand the nature of the low-temperature transition, we performed a careful low-field (2–30 Oe) magnetization experiment under FC and ZFC conditions. The results are shown in Fig. 1. For low fields the irreversibility (branching of FC and ZFC data) starts at the $T \leqslant T_C$ onset of ~ 150 K with a systematic drop in ZFC magnetization at temperatures 35–80 K for fields 30–2 Oe, respectively. A broad maximum has been observed in the ZFC data. This observation is different as we already mentioned for the field >100 Oe where T_{irr} starts at a much lower temperature well below T_C. In other words, for fields 2–30 Oe, it is not clear whether true long-range order exists; i.e., the range of field is not sufficient to orient the Fe moments, but short-range ferromagnetic ordering may exist.

The $\mathrm{TRM}(T)=M_{\mathrm{irr}}$ (M_{irr} can be defined as the difference

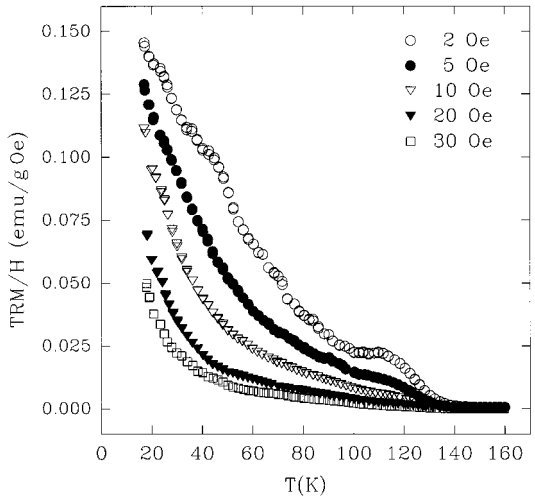

FIG. 2. Experimentally obtained thermoremanent magnetization with temperature at different cooling fields.

of M_{FC} and M_{ZFC}) has been attributed to a distribution of potential barriers in spin glasses.[1] At T_f, TRM(T) is expected to go to zero. On the other hand, for cluster glass the TRM is higher at low fields and it starts at $T \leqslant T_C$, indicating the presence of short-range ferromagnetic ordering within a cluster.[13] For a typical reentrant spin glass, irreversibility starts at low temperatures well below $T < T_C$. Figure 2 shows TRM measured at different temperatures after cooling under various fields (2–30 Oe). TRM(T) goes to zero at $T \sim 130$ K, suggesting the absence of true long-range ferromagnetic order. TRM increases rapidly with a decrease of temperature in the region where a drop in ZFC magnetization occurs. Figure 3 shows the coercive field H_C (obtained from the ZFC hysteresis loop in the field range $\pm$ 33 Oe) vs temperature curve. The coercive field H_C increases rapidly at $T \sim 40$ K, where ZFC magnetization data show a drop. It is expected that for the reentrant spin glass H_C increases rapidly at $T \leqslant T_f$.[1] However, hysteresis is not an essential property of spin glasses. In the same Fig. 3 we have plotted, $dH_C^{\mathrm{FC}}/dH_{\mathrm{FC}}$ with temperature. The $dH_C^{\mathrm{FC}}/dH_{\mathrm{FC}}$ is the slope

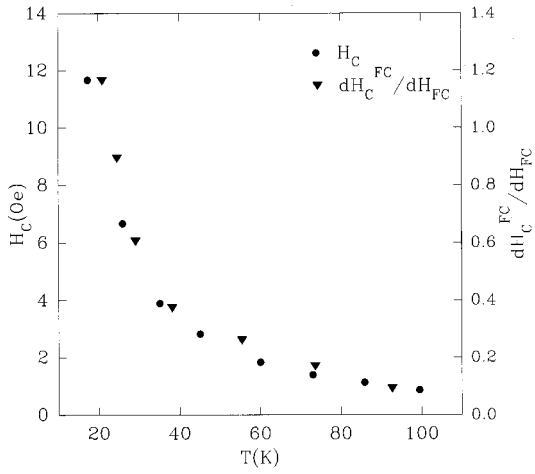

FIG. 3. Coercive field H_C vs temperature estimated from the ZFC hysteresis curve. $(dH_C^{\mathrm{FC}})/(dH_{\mathrm{FC}})$ estimated from the linear plot of H_C^{FC} with H_{FC} vs temperature.

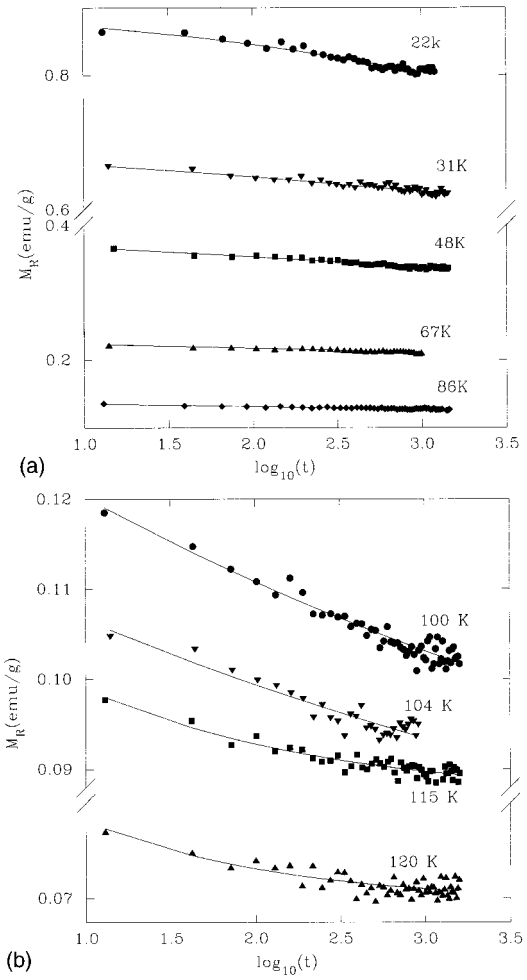

FIG. 4. (a) Thermoremanent relaxation isotherms for a sequence of temperatures $T \leqslant 90$ K. The solid curves are fits to Eq. (1). (b) Thermoremanent relaxation isotherms for a sequence of temperatures $T \geqslant 100$ K. The solid curves are fits to Eq. (2).

of the linear plot of H_C^{FC} with H_{FC}. H_C^{FC} is the coercive field under a field-cooled condition.[13] It has been shown recently that for a typical cluster glass system $dH_C^{\mathrm{FC}}/dH_{\mathrm{FC}}$ increases exponentially with a decrease of temperature, and this suggests that with the increase of temperature blocking of clusters decreases due to thermal activation.[13] A similar analysis for the present data shows no exponential behavior, suggesting no blocking of clusters.

Experimental results of TRM decay with time t at different temperatures at a 10 Oe field are shown in Fig. 4. In this figure we see a gradual and systematic change in curvature from concave downwards at low temperatures to concave upwards at high temperatures (from 100 K onwards) as found in the reentrant-spin-glass CrFe and NiMn systems.[14,15] FC and ZFC magnetization with temperature data at 10 Oe (Fig. 1) shows $T_C \sim 150$ K with a drop in magnetization at 60 K. In the reentrant-spin-glass regime TRM decay is age dependent (waiting time). The relaxation isotherm can be represented by a function consisting of the superposition of the stretched exponential and a constant term[16,17]

$$M_R = M_0 + M_1 \exp\left[-\left(\frac{t}{\tau} \right)^{(1-n)} \right], \qquad (1)$$

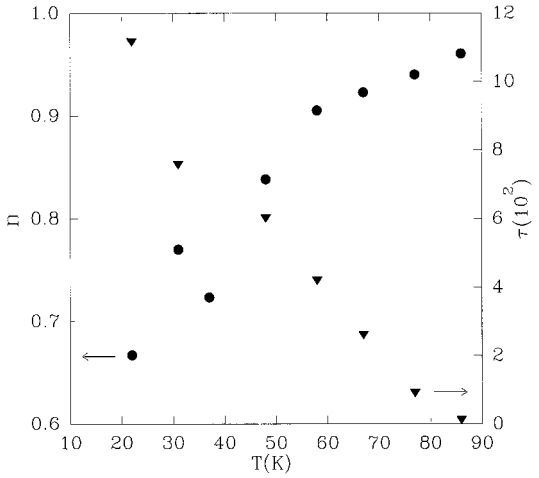

FIG. 5. Stretched exponential exponent n and the characteristic time τ vs temperature.

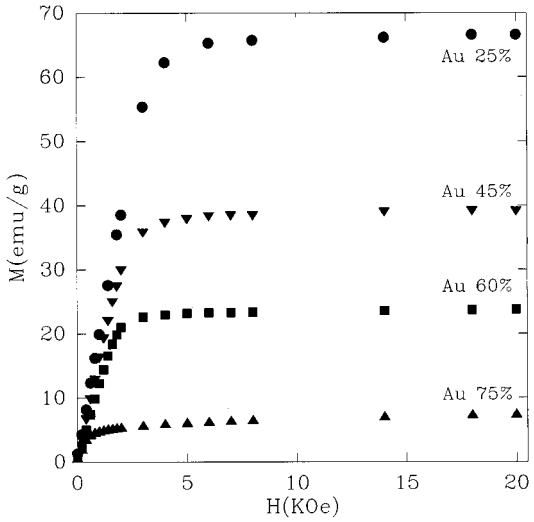

FIG. 6. Magnetization vs dc field for different Au concentrations.

where M_R is the thermoremanent magnetization. M_0 and M_1 are time-independent constants, τ is the characteristic time, and n is the stretched exponential exponent. The time-independent constant term M_0 is consistent with a theoretical model[18] which predicts a longitudinal ferromagnetic spontaneous magnetization to exist with transverse spin-glass freezing. The relaxation isotherms in Fig. 4 in the temperature range $T \leqslant 100$ K are best fitted to Eq. (1) (solid lines). The stretched exponential exponent n and the characteristic time τ are plotted with temperature in Fig. 5. The exponent n increases with an increase of temperature and tends towards unity. This feature is typical of spin glasses. For the temperature $T \geqslant 100$ K, a weak power law decay function describes the characteristic of thermal equilibrium relaxation[16,17] as

$$M_R = M_0 + M_1 t^{-m}, \tag{2}$$

where M_R is the thermoremanent magnetization. M_0 and M_1 are time-independent constants, and m is the power law exponent. This behavior is consistent with that observed in random ferromagnets.[19] Thus TRM decay with time at $T \leqslant T_C$ clearly suggests two relaxation regimes. The thermally driven crossover from the high-temperature ferromagnetic phase $T \geqslant 100$ K to a low-temperature <100 K possibly glassy phase (which is related to the onset of nonequilibrium effects) is clear. This observation is consistent with other reported work on the reentrant-spin-glass CrFe system,[14] etc.

In Fig. 6 as an example the magnetization data at $T \sim 100$ K for the series of $(\mathrm{FeNi})_{100-x}\mathrm{Au}_x$ ($x = 25$, 45, 60, 75) are shown. All concentrations except 75% show saturation behavior for field >5 kOe for the temperature range 100–300K.[2] This clearly suggests that the absence of an orderable ferromagnetic moment ($2.0\mu_B$) plays a significant role in the 75% Au system. Therefore the 75% Au, in the $(\mathrm{FeNi})_{100-x}\mathrm{Au}_x$ alloy system, may be considered as disordered magnet. We wish to stress that such a comparison is not possible with other noble-metal-doped alloys such as (FeNi)Cu at a higher concentration of Cu due to decomposition.

GMR is not a property restricted to multilayers but it can also be found in granular thin films, ribbons, and rapidly quenched alloys such as Co-Cu, Co-Ag, Ag-Fe, Cu-Fe, Au-Fe, Cr-Fe, and NiFe-Ag. These alloys comprise two immiscible metallic components, one magnetic, the other nonmagnetic, which tend to segregate, resulting in the formation of magnetic single-domain clusters embedded in a nonmagnetic matrix. The resistance is high for a random alignment of magnetic cluster moments and it is decreased when individual moments are aligned parallel by an external field.[3,5,7–9,20] As a consequence of the granular nature of the films, systems such as Co-Ag show GMR with superparamagnetic behavior.[6] Also GMR effects are seen in metamagnetic materials undergoing ferromagnetic to antiferromagnetic transitions[4] and in spin-glass and reentrant-spin-glass Cr-Fe bulk granular alloys[3] and spinodally decomposed Cu-Ni-Fe alloys.[12] However, spin-glass alloys[20] (AgMn, AuFe) at a low temperature also exhibit large negative magnetoresistance. In spin glasses and alloys containing small magnetic clusters, a relatively large field is necessary to overcome the anisotropy energy. It is difficult to accomplish ferromagnetic alignment of all the moments, and so the large magnetoresistance cannot be readily saturated, whereas in multilayers and granular systems magnetoresistance can be saturated under a sufficiently large field; i.e., ferromagnetic alignment is achieved.

Figure 7 shows the resistivity $\rho(T)/\rho(300)$ as a function of temperature for the $(\mathrm{FeNi})_{100-x}\mathrm{Au}_x$ ($x = 25$, 45, 60, 75) alloys. We observed a higher ρ_0 for the $x = 75$ sample in comparison with other concentrations and a significant change in ρ(T) is observed near $T_C \sim 130$ K. For other concentrations $\rho(T)$ has been measured at temperatures well below T_C which varies from 700 K ($x = 25$), 530 K ($x = 45$), and 333 K ($x = 60$),[10] respectively. Since a significant change in magnetoresistance has been observed only for $x = 75$ in the ± 8 kOe field in comparison with other concentrations where $\Delta R/R$ is small (for $x = 60$, $\Delta R/R$ is $\sim 0.75\%$ at 79 K and 8 kOe), we focus our magnetoresistance work only on the $x = 75$ sample. Figure 8 shows the magnetoresistance $\Delta R/R$ (%) defined as $[R(H) - R(H = 0)]/R(H = 0) \times 100$ as a function of parallel applied field at different temperatures. At 8 kOe, $\Delta R/R$ is $\sim 4.5\%$ at 48 K to 0.3% at 200 K. This magnitude of $\Delta R/R$ is comparable

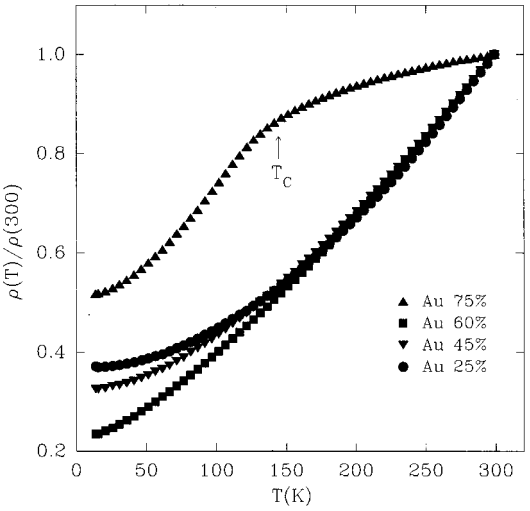

FIG. 7. $\rho(T)/\rho(300)$ with temperature for different Au concentrations.

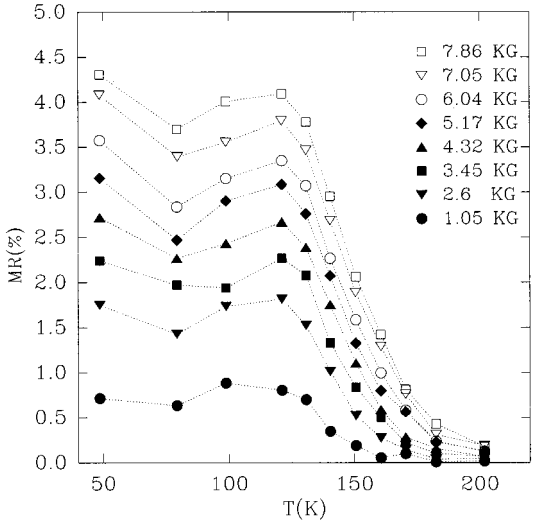

FIG. 9. Parallel magnetoresistance $[R(H)-R(0)]/R(0)$ as a function of temperature for different fields.

with the case of GMR in the bulk granular alloy CoAl, Au-Fe,[5] and (NiFe)Ag alloys.[21]

In Fig. 9 we have plotted magnetoresistance vs temperature for different fields in the range 1–8 kOe. $\Delta R/R$ increases with the increase of magnetic field to about 4.5% for $\sim$8 kOe. Figure 10 shows that no appreciable change in $\Delta R/R$ is observed under FC and ZFC conditions at 98 K. Here the cooling field is $\sim$7 kOe. The present data also show no asymmetry behavior unlike that shown by GMR in superparamagnetic $Co_{10}Cu_{90}$ granular alloys.[8] This means that for the present sample GMR does not depend upon the metastable states that develop when the sample is cooled. It is interesting to note that magnetization data under FC and ZFC conditions does not show any change at these temperatures except that the irreversibility occurs at very low temperatures $\sim$5 K at 1 kOe. Figure 11 shows GMR measured at 98 K, $T<T_C$, 170 K (close to T_C), for $H\|I$ and $H\perp I$ (where I is the in-plane sample current) which is negative. Hysteresis is also not observed and the anisotropy $(\Delta\rho_{\|}-\Delta\rho_{\perp})/\rho_0$ is less than 0.5%. GMR in granular solids is isotropic.[7] In the case of multilayers GMR is negative, but the magnitudes are different due to the demagnetization factor.[20]

According to the two-current model GMR can be expressed as

$$\frac{\rho(H)-\rho(0)}{\rho(0)}=-A\left(\frac{M}{M_S}\right)^2,$$

where the coefficient A is related to the magnitude of GMR, M is the global magnetization, and M_S is the saturation magnetization. The magnitude depends on the spin-dependent scattering, as well as the number and the size of ferromagnetic entities with mean free path (λ). Attempts to fit the above equation to our data and even $(M/M_S)^4$ as in Cu-Mn-Al (Ref. 22) have not been successful. This behavior is probably due to the fact that large fields are required to align the Fe moments. In fact even up to 55 kOe at 98 K full magnetic saturation could not be obtained.[2] For example, the magnetization value at 8 kOe is $\sim$6.8 emu/g compared to 7.3 emu/g at 20 kOe and 14 emu/g at 55 kOe. For other Au concentrations saturation can be easily obtained for fields $\sim$

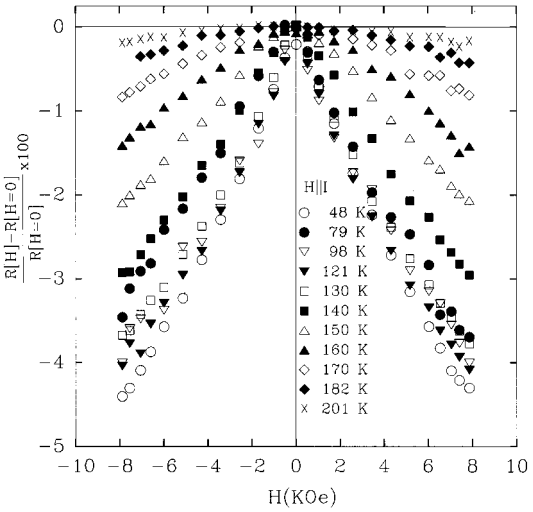

FIG. 8. Parallel magnetoresistance $[R(H)-R(0)]/R(0)$ as a function of external field at different temperatures.

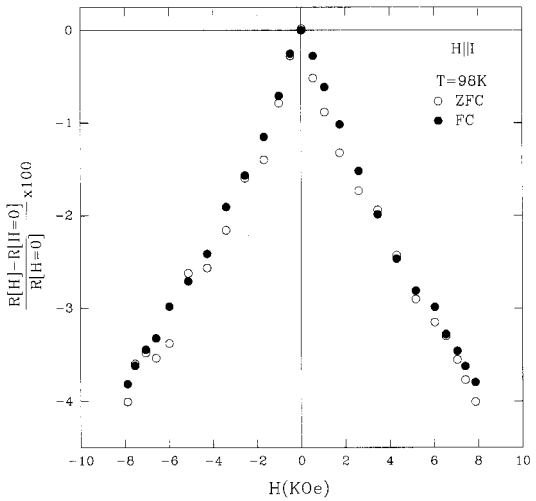

FIG. 10. FC and ZFC parallel magnetoresistance $[R(H)-R(0)]/R(0)$ as a function of external field at 98 K.

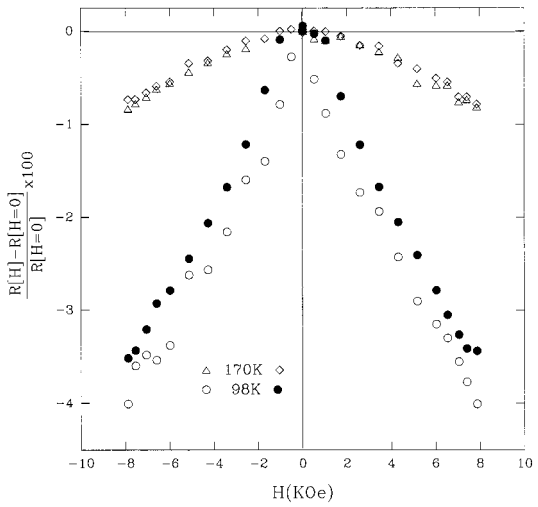

FIG. 11. Parallel (○, △) and perpendicular (●, ◇) magnetoresistance $[R(H)-R(0)]/R(0)$ as a function of external field at 98 K and 170 K.

5 kOe (Fig. 6) for which magnetoresistance is 5 times smaller than that observed in the spin-glass-like sample ($x=75$). For $x=75$ sample GMR may be due to scattering from nonaligned ferromagnetic entities as in the granular magnetic systems.[7] Absence of GMR saturation in 8 kOe data (Fig. 8) is due to this nonalignment of ferromagnetic entities. In fact, depending upon the field strength the alignment of the Fe moment is possible, as shown by our high-field data (1–55 kOe).[2] The present magnetization data suggest the frustration in the system. Therefore we believe that there exists a subtle link between frustration and GMR. It may be mentioned here that for the present study interfacial scattering as in thin films, ribbons, etc., does not arise as the system is a bulk sample. It has been emphasized that GMR occurs in magnetically inhomogeneous media or disordered systems containing nonaligned ferromagnetic entities on a microscopic length scale, roughly equal to the electron mean free path.[20] However, a detailed microstructure parameter is required to further clarify the origin of GMR in the present $(FeNi)_{25}Au_{75}$ bulk alloys.

V. CONCLUSIONS

The present results demonstrate the observation of GMR in $(FeNi)_{25}Au_{75}$ in the series of fcc $(FeNi)_{100-x}Au_x$ ($x=25$, 45, 60, 75) alloys. The origin of GMR in the $x=75\%$ alloy is attributed to the field-induced alignment of Fe moments, for which saturation could not be obtained for fields >20 kOe. The field-cooled and zero-field-cooled magnetization, FC, ZFC hysteresis, thermoremanent magnetization, and magnetic relaxation data show the characteristic features of a typical reentrant-spin-glass-like behavior at low temperatures. For other Au concentrations magnetic saturation can be easily obtained for fields ~ 5 kOe for which no GMR is observed. Thus the present study on $(FeNi)_{25}Au_{75}$ can be considered as an example to investigate GMR in disordered magnetic systems.

ACKNOWLEDGMENTS

We thank A. Chakravarti for his help and Professor J. W. Cable for providing the sample.

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