Magnetoresistance in canonical spin-glasses

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The transverse magnetoresistance (TMR) $\Delta\rho/\rho$ has been measured in *Au*Fe, *Au*Mn, *Cu*Mn, and *Ag*Mn spin-glasses in the temperature range 1.5 to 77 K and magnetic fields up to 18 kG. All the alloys exhibit a negative TMR at all temperatures and fields and changes with field H as H^n, where $n \simeq 2$ at low fields and $1 < n < 2$ at higher fields. Below the spin-glass freezing temperature T_0 and at higher fields, the TMR is nearly independent of temperature. This behavior is very similar to that of the Hall resistivity. At low fields it varies smoothly around T_0 without showing any anomaly. However, below T_0 the TMR shows a somewhat stronger temperature dependence in a few alloys. Above T_0, $|\Delta\rho/\rho|$ decreases with temperature and becomes undetectable beyond T_m, the temperature of the resistance maximum of the alloy. Also, $|\Delta\rho/\rho|$ increases with the impurity concentration c in *Au*Fe while decreasing with it in *Au*Mn, *Cu*Mn, and *Ag*Mn. However, $|\Delta\rho|$ increases monotonically with c in all the systems. A recent theory of magnetoresistance, based on an Edwards-Anderson-type model for a spin-glass, explains most of the above characteristic features. This theory has been applied to compute the values of the *s-d* exchange parameter "J." They are in reasonable agreement with those found by earlier authors. Effects of field cooling, remanence, aging, and annealing on the TMR are discussed. A fresh annealing and quenching of *Au*Fe alloys significantly reduces $|\Delta\rho/\rho|$.

I. INTRODUCTION

In recent years dilute magnetic metallic systems, crystalline as well as amorphous, have been the subject of renewed interest for the study of magnetic, transport, and thermal properties. These studies have been intended mainly to probe how the magnetic order sets in when the concentration of metallic impurity, e.g., transition or rare earth, is increased in a host metal which does not itself undergo magnetic ordering at any temperature. The study of the transport properties of these systems was rather tempting because of the discovery of the Kondo effect, giant thermoelectric power, etc., in dilute metallic alloys. The two kinds of studies, namely, the magnetic and transport properties, started converging after it was established that the anomalous transport properties are due to the formation of local magnetic moments in dilute alloys. Spin-glass is one of such systems and is of the type $\mathrm{No}_{1-c}\mathrm{Tr}_c$. Here c is the concentration of the $3d$ transition-metal impurity Tr (e.g., Fe, Mn, Cr) which is randomly distributed in the matrix of the noble-metal host No (e.g., Au, Ag, Cu). If $c < 10^{-4}$, one is in the dilute Kondo region, while $c > 0.1$ gives the percolation limit. In the intermediate concentration range (say, $0.005 < c < 0.1$), the distance between the magnetic impurities is such that their indirect exchange energy via the conduction electrons dominates over all other contributions. The canonical spin-glasses are *Au*Fe, *Au*Mn, *Cu*Mn, *Ag*Mn, and *Au*Cr, each of them satisfying the criteria of a favorable solubility of the impurity in the host and a sufficiently low (<0.1 K) Kondo temperature.

The first experimental results containing the spin-glass model were of Cannella and Mydosh,[1] who observed a sharp transition in the low-field (~ 5 G) ac magnetic susceptibility $\chi(T)$ at a characteristic spin-freezing temperature T_0 ($\sim c^{2/3}$) in *Au*Fe alloys. The cusplike peak was found to become rounded even in a field as low as 100 G. Thus the critical behavior of $\chi(T)$ could only be observed at low fields. Below T_0, $\chi(T)$ falls off very much like that of an antiferromagnet below its Néel temperature. Above T_0, it followed a Curie-Weiss law from which p_{eff}, the effective Bohr magnetion number, and Θ, the Curie-Weiss temperature could be obtained. The same type of behavior was found in the other canonical spin-glasses[2] with $0.005 < c < 0.1$. Borg *et al.* and Window observed in *Au*Fe (Ref. 3), *Cu*Mn, and *Au*Mn (Ref. 4) that the magnetic hyperfine field splitting occurred in Mössbauer spectra at T_M (which is close to T_0), indicating a magnetic phase transition. However, the neutron scattering measurements of Sato *et al.*[5] in

*Cu*Mn revealed the absence of any long-range antiferromagnetic ordering. The muon depolarization experiments of Murnick *et al.*[6] indicated at T_0 an abrupt appearance of local fields in *Cu*Mn and *Au*Fe spin-glasses. An external magnetic field smeared the transition due to the creation of larger local fields. McAlister and Hurd[7] found a peak, very similar to that of $\chi(T)$, in the total Hall resistivity ρ_H at T_0, but up to a much higher field of about 1.5 kG. At still higher fields the peak disappeared and ρ_H was relatively independent of temperature around T_0. Ford and Mydosh[8] found that the impurity resistivity in spin-glasses varied roughly linearly with temperature around T_0 and then showed a broad maximum at a temperature T_m which was much larger than T_0. No anomaly was observed around T_0 in magnetic specific heat,[9] thermoelectric power,[1] or ultrasonic velocity.[10]

The Edwards-Anderson[11] (EA) model, in the mean-field approximation, yielded a cusp in $\chi(T)$ at T_0. The physical picture of a spin-glass in this theory is the following: Below T_0, the spins associated with the solute atoms are frozen in random orientations. Even a small magnetic field disrupts this short-range order and unlocks the spins. In the spin-glass regime, the magnetization $M=\langle \bar{S}_i \rangle$ is zero for all temperatures and hence, it could not be an order parameter. Edwards and Anderson introduced an order parameter Q, defined as

$$Q=\lim_{|t_1-t_2|\to\infty} \langle \overline{S_i(t_1)S_i(t_2)} \rangle ,$$

which shows a time-dependent correlation of individual spins at a particular site. Q is zero for $T>T_0$ while it is nonzero for $T<T_0$. This model, however, overemphasized an anomaly which was not observed in the magnetic specific heat at T_0.

From the experimental survey given earlier it is clear that the experiments on spin-glasses could be broadly divided into two classes, one of which exhibits a sharp anomaly at T_0 while the other shows a smeared behavior around T_0. These experiments can be categorized as follows:

Sharp anomaly at T_0	Smeared behavior
Magnetic susceptibility	Resistivity
Remanence	Specific heat
Mössbauer effect	Thermopower
Muon-spin depolarization	Nuclear magnetic resonance (NMR)
Anomalous Hall effect	Ultrasonic velcoity

It is interesting to find that among the transport properties, only the anomalous Hall effect provides a sharp anomaly at T_0, while the resistivity and thermopower do not. The experiments listed in the second column are, indeed, the indirect ones having large nonmagnetic contributions; hence, it is difficult to separate out, correctly, the magnetic contribution. This perhaps might be the cause for not observing a critical behavior around T_0 in the experiments of the second column.

On the basis of the experimental evidence of a magnetic phase transition at T_0 and the fact that the spin correlations are seriously affected by even a small external magnetic field, one might anticipate an anomaly also in the magnetoresistance of spin-glasses as one passes through T_0.

II. MAGNETORESISTANCE IN $No_{1-c}Tr_c$ ALLOYS

A. Earlier experiments

Usually, the resistance of a metallic system increases in a magnetic field. This is referred to as normal magnetoresistance and is due to the Lorentz force acting on the conduction electrons. It varies quadratically in small magnetic fields.

A negative magnetoresistance was observed, for the first time, by Gerritsen *et al.*[12] in some of the dilute noble-metal-transition-metal alloys, as, for example, *Ag*Mn, *Au*Mn, *Cu*Mn, etc. where the noble metal is the host. The negative magnetoresistance was found to be the same for both the longitudinal and the transverse magnetic fields. Later, Schmitt and Jacobs[13] and Muto *et al.*[14] established that the negative magnetoresistance in *Cu*Mn and *Cu*Fe alloys varied as the square of magnetization.

Rohrer[15] had measured the transverse magnetoresistance (TMR) in dilute *Au*Fe and *Au*Mn alloys having the transition-metal impurity concentration in the range of 0.1 to 1.0 at.%. These alloys exhibit a negative TMR up to the highest measuring field of 200 kG. The negative TMR in *Au*Fe alloys had been found to show a strong concentration dependence as against a weak one in *Au*Mn alloys.

The above studies were carried out before the idea of spin-glasses came into existence, and hence

there was no intention to look for a phase transition at T_0, called the spin-freezing temperature in the present nomenclature of spin-glasses. These experiments were meant essentially to probe into the mechanism of the Kondo resistance anomaly and hence were directed to very dilute alloys having magnetic impurity concentration of less than 0.1 at.%.

B. Theory of magnetoresistance

The resistance of an electrical conductor has been found to either increase (normal or positive magnetoresistance) or decrease (negative magnetoresistance) in a magnetic field. The theoretical models of the positive and the negative magnetoresistances relevant to the present work are briefly described below.

(i) Normal magnetoresistance. The normal (positive) magnetoresistance is inherent in all systems having free electrons. A theory based on a free-electron model led to a zero magnetoresistance. A two-band model[16] consisting of two overlapping bands of s and d electrons was therefore proposed. In this model the transverse magnetoresistance, in small magnetic fields, H, is given by

$$\left[\frac{\Delta\rho}{\rho}\right]_n = \frac{1}{2ne}\left[\frac{H}{\rho}\right]^2, \tag{1}$$

where $(\Delta\rho/\rho)_n$ is called the "normal magnetoresistance," ρ being the electrical resistivity in zero magnetic field, n is the number of electrons per unit volume, and e is the electronic charge. The above expression is in fairly good agreement with the experimental results.

(ii) Negative magnetoresistance. Béal-Monod and Weiner[17] calculated the negative magnetoresistivity of dilute alloys containing transition-metal impurities and exhibiting Kondo resistance anomaly. Their calculations were based on a third-order perturbation expansion of the s-d exchange Hamiltonian. The calculations of the conduction-electron scattering amplitude, in the zero-field limit, yielded the famous Kondo logarithmic series in temperature T. The theory of Béal-Monod and Weiner is restricted to alloy systems in which the spins are isolated and hence the spin correlations of the magnetic impurities were ignored.

Recently, Mookerjee[18] has calculated the magnetoresistivity in spin-glasses on the basis of Edwards-Anderson-type model. Here the local spins interact through the conduction electrons via the s-d exchange coupling. Under the following simplified assumptions that

(a) the impurity and the s-d couplings are isotropic,

(b) the magnetic impurity concentration is such that the clustering of magnetic atoms does not take place, and

(c) the effects of spin dynamics have been ignored, the expression obtained for the resistivity $\rho(H)$ in a magnetic field H for $No_{1-c}Tr_c$ alloy could be written in the Born approximation as

$$\rho(H)=cR_0\left[V^2+J^2\left[S(S+1)-M(H)\tanh\left[\frac{g\mu_B H}{2k_B T}\right]\right]\right.$$
$$\left.-2J^2Q(H)\left\{1-\frac{J^2}{V^2}\left[S(S+1)-M(H)\tanh\left[\frac{g\mu_B H}{2k_B T}\right]\right]\right\}\right], \tag{2}$$

where V and J are the Coulomb and the s-d exchange potentials, respectively, $M(H)$ and $Q(H)$ are the order parameters as introduced in the EA model,[19] S is the impurity spin, $g=2$ for conduction electrons, and μ_B is the Bohr magneton. The quantity R_0 is defined as

$$R_0=\left[\frac{3\pi m}{4\hbar e^2}\right]\left[\frac{1}{nE_F}\right], \tag{3}$$

where n is the number of electrons per unit volume, and the quantities m, $\hbar$, e, k_B, and E_F have their usual meaning.

The order parameters $M(H)$ and $Q(H)$ have been found to satisfy the simultaneous equations[18]

$$M(H)=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} e^{-z^2/2}\tanh\left[\alpha+\frac{\Theta M(H)}{T}+\frac{T_0 Q^{1/2}(H)}{T}z\right]dz, \tag{4}$$

$$Q(H)=\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}e^{-z^2/2}\tanh^2\left[\alpha+\frac{\Theta M(H)}{T}+\frac{T_0Q^{1/2}(H)}{T}z\right]dz\ , \tag{5}$$

where Θ is the Curie-Weiss temperature, and $\alpha=\mu H/k_BT$ where μ is the magnetic moment of the impurity atom as seen in the noble-metal host, and is given by $\mu=p_{\text{eff}}\mu_B$, p_{eff} being the Bohr magneton number.

From Eq. (2), the change in resistivity $\Delta\rho$ due to a magnetic field H could be written as

$$\Delta\rho=\rho(H)-\rho(O)=-cR_0J^2\left[M(H)\tanh\left[\frac{g\mu_BH}{2k_BT}\right]+2[Q(H)-Q(O)]\left[1-\frac{J^2}{V^2}S(S+1)\right]+2\left[\frac{J^2}{V^2}\right]Q(H)M(H)\tanh\left[\frac{g\mu_BH}{2k_BT}\right]\right]\ . \tag{6}$$

Thus $\Delta\rho$ can be calculated from Eq. (6) using Eqs. (4) and (5) for $M(H)$ and $Q(H)$, respectively.

Mookerjee therefore concluded that the change in resistivity due to a magnetic field H is

(a) negative at all temperatures and fields, varying as H^2 in low fields and at temperatures near T_0 and

(b) proportional to the magnetic impurity concentration c.

It should be pointed out here that $M(H)$ and $Q(H)$ are also dependent on T_0 and Θ, which in turn, depend on concentration. Thus the dependence of $\Delta\rho$ on c could not be a simple one, as concluded by Mookerjee.

III. EXPERIMENTAL METHODS

A. Preparation of alloys

Four binary systems were studied in the present investigations. They were *Au*Fe, *Au*Mn, *Cu*Mn and *Ag*Mn alloys in the magnetic impurity concentration range of 0.5 to 10.0 at. %. Some of these alloys, viz., *Cu*Mn and *Ag*Mn, were prepared here by induction melting in argon atmosphere. The constituent metals were of 99.999% purity obtained from M/S Johnson-Mathey Inc., England. Each alloy was homogenized at a temperature about 100 °C below its melting point for 24 h in argon atmosphere. Then they were swaged, cold-rolled, and cut into thin rectangular strips ($40\times1\times0.1$ mm^3) for magnetoresistance measurements. The alloys of *Au*Fe and *Au*Mn, obtained from Mydosh and Coles, were already in the homogenized state. Some of the alloys were chemically analyzed and found to be within 2% of the nominal composition of the magnetic impurity.

In order to remove mechanical strains, as well as to provide a random substitutional character to the magnetic impurity, each alloy was finally annealed for 24 h at 900 °C in argon atmosphere. Then they were quenched in water and kept in liquid nitrogen until measurments were made.

B. Experimental setup

A cryostat was designed and fabricated for the measurement of transverse magnetoresistance in the temperature range of 1.5 to 77 K and up to magnetic fields of 18 kG. The resistivity was measured using a four-probe dc method where the current and the voltage leads were spot-welded to the sample. Extreme care was taken to minimize thermoelectric, ground loop, and pickup voltages. A current of about 300 mA was passed through the sample in series with a 1-Ω Leeds and Northrup (LN) standard resistance.

The zero-field voltage across the sample was measured by using a voltage compensation technique in the following manner. An LN K-3 potentiometer, which has a sensitivity of 0.5 μV, was used as a standard voltage source. The specimen voltage due to current I was compensated by adjusting the K-3 potentiometer, connected in series opposition, and using a model 148 Keithley nanovoltmeter as a null detector. In this situation the voltage read on the K-3 potentiometer had been precisely the voltage drop V across the sample. In the same way the voltage drop across the 1-Ω LN resistance gave I. The change in the sample voltage ΔV due to incremental magnetic field was of the order of a few microvolts and hence, it had been read directly on the nanovoltmeter. The ratio $\Delta V/V$ was calculated, which is the same as the magnetoresistance $\Delta\rho/\rho$ of the alloy. By knowing I, V, and the geometrical factor, the resistivity of

the alloy at zero field was obtained.

A Varian 15-in. electromagnet fed by a VFR-2703 power supply provided a highly stable magnetic field. A Honeywell germanium thermometer was used to measure the temperature of the specimen. The temperatures above 4.2 K were achieved by using a Manganin wire heater, while those below 4.2 K were achieved by pumping over the liquid-helium bath with a Kinney pump.

In a few spin-glass alloys the magnetization was measured at 77 K and up to magnetic fields of 16 kG. In these experiments a Princeton Applied Research (PAR) model 155 vibrating sample magnetometer and a PAR model 153 cryostat were used along with the Varian 15-in electromagnet. The details of the complete experimental method are described elsewhere.[20]

IV. RESULTS

The present study is comprised of measurements of transverse magnetoresistance (TMR) in *Au*Fe (2.9, 6.6, and 10.0 at.%), *Au*Mn (1.8 and 4.6 at.%), *Cu*Mn (0.7 and 4.4 at.%), and *Ag*Mn (1.1 and 5.4 at.%) spin-glass alloys in the temperature region of 1.5 to 77 K and up to magnetic fields of 18 kG. The measurements have relative accuracy of about 40 ppm of the resistivity. Each sample had been measured twice under the same sample condition. The data were qualitatively identical in the two runs. However, the reproducibility of $\Delta\rho/\rho$ was within $\pm 5\%$.

The measured TMR $\Delta\rho/\rho$ is plotted as a function of magnetic field H at different temperatures in Figs. 1(a)–1(i) for various spin-glass alloys. It should be noted here that not too many low-field points are shown to enhance readability. All the alloys exhibit a negative TMR in the temperature range of measurement. With the exception of *Au*Fe (2.9 at.%) the alloys had been measured after fresh annealing and quenching as mentioned in Sec. III A. The *Au*Fe (2.9 at.%) alloy had been measured only in the as-received aged condition. It is observed that the magnitude of the negative TMR in *Au*Fe alloys increases with increasing concentration at all temperatures and fields [Figs. 1(a)–1(c)]. In *Au*Mn [Figs. 1(d), 1(e)], the negative TMR slightly decreases by increasing the Mn concentration from 1.8 to 4.6 at.%, e.g., at 4.2 K and at 18 kG, it is 7.5% for *Au*Mn (1.8 at.%) and 6.5% for *Au*Mn (4.6 at.%). In the case of *Cu*Mn and *Ag*Mn alloys, the negative TMR has been found to decrease significantly by increasing the concentration of Mn impurity [Figs. 1(f)–1(i)].

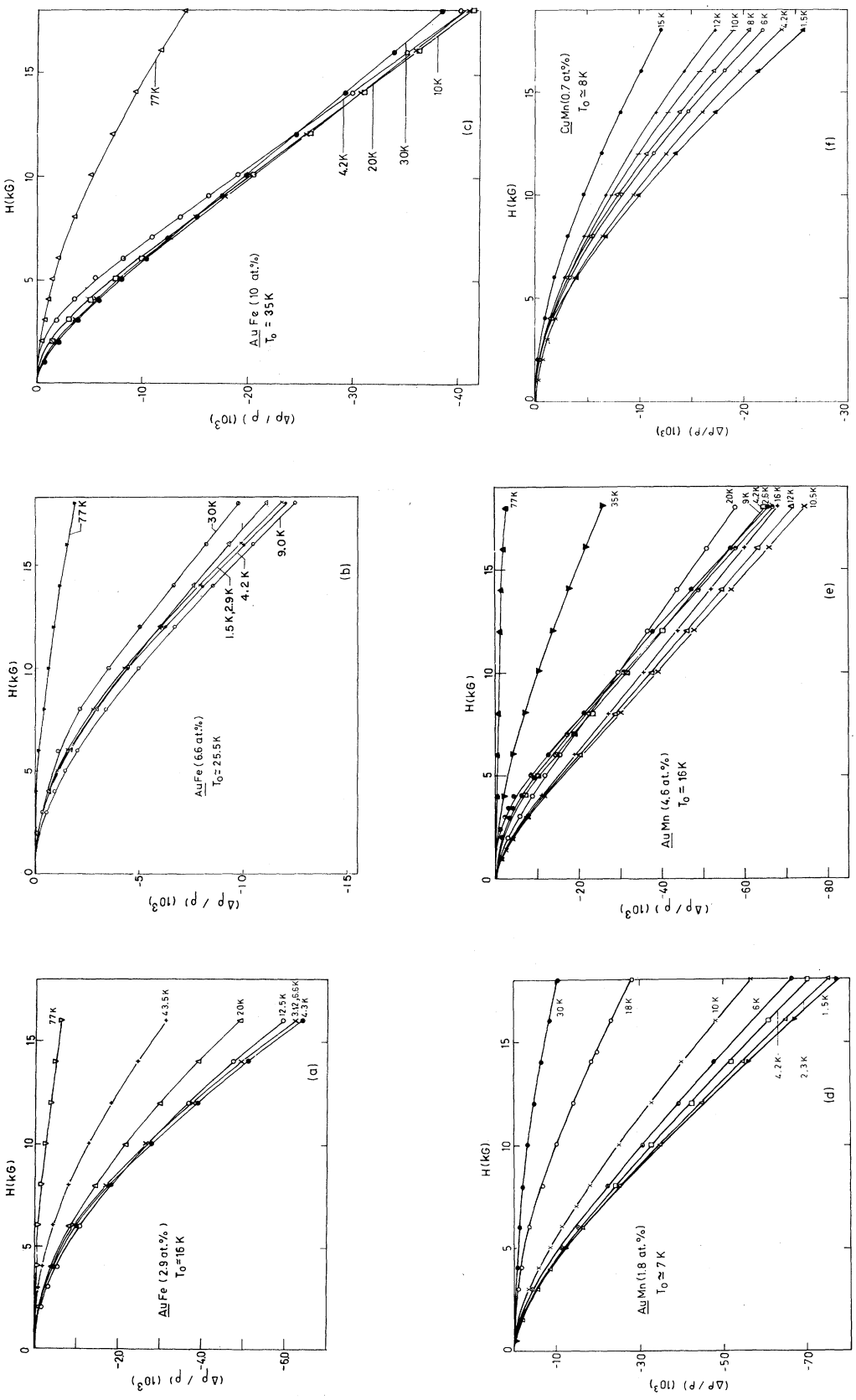
It is observed that at a fixed temperature the negative TMR varies faster than H, i.e.,

$$\Delta\rho/\rho = -b'(T,c)H^n , \tag{7}$$

where $n>1$ and $b'(T,c)$ is a coefficient that depends on the temperature T and the magnetic impurity concentration c of the alloy. To find the value of n, log-log plots of $\Delta\rho/\rho$ vs H were tried for all the alloys, and it has been found that at low fields $n=2.0\pm0.1$ at all temperatures for all the alloys. The maximum field (H_q) up to which the quadratic nature of $\Delta\rho/\rho$ persists at all temperatures is different for different alloys. To show the quadratic dependence of the negative TMR up to H_q, $\Delta\rho/\rho$ have been plotted against H^2 for *Ag*Mn (1.1 at.%) in Fig. 2. In this figure, one can see that H_q is only 3 kG. The slope of these plots gives $b(T,c)=b'(T,c)$ of Eq. (7) when $n=2$. Similar plots have been tried for all the other alloys, and the values of H_q and those of $b(T,c)$ are given in Table I. It is found in all the alloys that at temperatures well above T_0, $\Delta\rho/\rho$ is quadratic in H up to the highest field, i.e., 18 kG. On the other hand, for $T<T_0$ and for higher fields $\Delta\rho/\rho$ is almost linear in H for many of them [especially Figs. 1(c), 1(d), 1(e), and 1(h)]. Nevertheless, $\Delta\rho/\rho$ is quadratic in H up to H_q at all temperatures.

In the temperature region of $T<T_0$, and at low fields, $\Delta\rho/\rho$ is found to be weakly dependent on temperature. However, in *Au*Mn (1.8 and 4.6 at.%), *Ag*Mn (5.4 at.%), and *Au*Fe (10.0 at.%) alloys, the coefficient $b(T,c)$ exhibits a pronounced maximum below T_0, as can be seen from Table I. In other alloys $b(T,c)$ is almost independent of temperature below T_0. Above T_0, $b(T,c)$ decreases fast with increasing temperature and becomes vanishingly small around T_m, the temperature of the resistance maximum of the alloy. As an example, in the *Cu*Mn (0.7 at.%) alloy which has $T_0=8$ K and $T_m=17$ K (Ref. 8), $b(T,c)$ decreases from 10.7 to 8.8×10^{-3} T^{-2} between 1.5 and 8 K while it decreases from 8.8 to 5.0×10^{-3} T^{-2} between 8 and 15.5 K [Fig. 1(f)]. In this alloy, at a temperature of 20 K, which is 3 K above T_m, no TMR could be observed.

At higher fields and in relatively concentrated alloys, $\Delta\rho/\rho$ seems to be almost independent of temperature below T_0. In Fig. 1(g) one could see for *Cu*Mn (4.4 at.%) that at 10 kG $\Delta\rho/\rho$ is almost independent of temperature below 20 K, whereas T_0 is around 16 K for this alloy. The same type of behavior is encountered in *Au*Mn (1.8 and 4.6 at.%), *Ag*Mn (5.4 at.%), and *Au*Fe (2.9, 6.6, and


(a)
Au Fe (2.9 at.%)
T0 = 16 K
77K
43.5K
20K
12.5K
4.2K
H(kG)
(Δρ / ρ) (10³)
(b)
Au Fe (6.6 at.%)
T0 = 25.5K
77K
30K
9.0K
4.2K
1.5K, 2.9K
H(kG)
(Δρ / ρ) (10³)
(c)
Au Fe (10 at.%)
T0 = 35K
77K
4.2K
20K
30K
10K
H(kG)
(Δρ / ρ) (10³)
(d)
Au Mn (1.8 at.%)
T0 ≃ 7 K
30 K
18K
10K
6K
4.2K
2.3K
1.5K
H(kG)
(Δρ / ρ) (10³)
(e)
Au Mn (4.6 at.%)
T0 = 16K
77K
35K
20K
9K
4.2K
2.5K
16 K
12K
10.5K
H(kG)
(Δρ / ρ) (10³)
(f)
Cu Mn (0.7 at.%)
T0 ≃ 8K
15K
12K
10K
8 K
6 K
4.2K
1.5K
H(kG)
(Δρ/ρ) (10³)

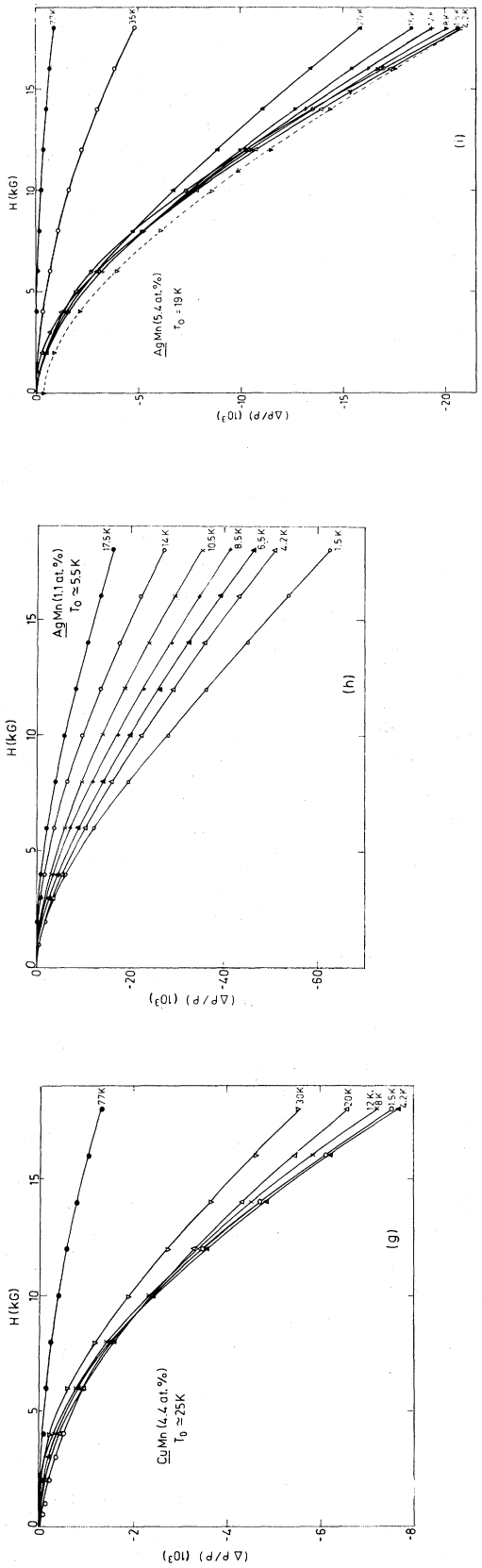


FIG. 1. Transverse magnetoresistance $\Delta\rho/\rho$ vs external magnetic field H at different temperatures for spin-glass alloys.

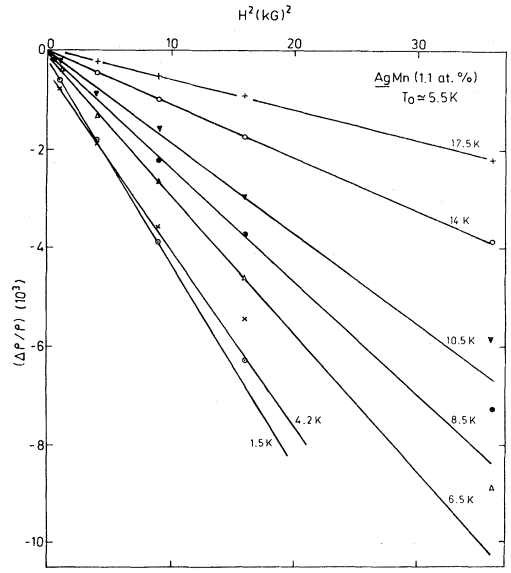


FIG. 2. Transverse magnetoresistance $\Delta\rho/\rho$ vs square of external magnetic field H^2 at different temperatures for *Ag*Mn (1.1 at. %) alloy.

10.0 at. %) samples. This a very similar to the behavior of Hall resistivity as discussed in Sec. I. The values of the resistivity at 4.2 K of all the alloys measured are within $\pm 10\%$ of the literature values.

V. DISCUSSION

The transverse magnetoresistance measured in this work is, in fact, the sum of both normal and negative contributions, that is,

$$(\Delta\rho/\rho)_{\text{meas}} = (\Delta\rho/\rho)_{\text{normal}} + (\Delta\rho/\rho)_{\text{negative}} ,$$

where the second term represents the actual negative contribution to the TMR. It is evident from Eq. (1) that the normal contribution varies inversely as the square of the resistivity, which is a function of temperature as well as composition of the alloy. The alloys of the current study are in a moderately high-impurity-concentration range, and therefore the normal contribution is expected to be insignificant in comparison to the negative contribution to the TMR. For example, at 20 K, the normal magnetoresistance of pure Ag at 3 kG is $\simeq 0.04$ (Ref. 21), while its electrical resistivity is $\simeq 0.005$ $\mu\Omega$ cm.[22] Now, using these data and Eq. (1), if we calculate the normal magnetoresistance of *Ag*Mn (1.1 at. %) [having a resistivity $\simeq 1.7$ $\mu\Omega$ cm (Ref. 8)], we get $(\Delta\rho/\rho)_{\text{normal}} \simeq 10^{-6}$. The observed negative TMR in the present study is of the order of 10^{-3}, and hence the normal (positive) contribution to the TMR is negligible. The validity of this assumption had also been established by Rohrer.[15] We have, therefore, treated the measured negative TMR as the actual negative TMR in the present work.

A. Negative TMR in spin glasses

The existence of a negative TMR in dilute $\text{No}_{1-c}\text{Tr}_c$ alloys had been shown earlier by Gerritsen *et al.*[12] A study of a few alloys of *Au*Fe, *Au*Mn, *Ag*Mn, and *Cu*Mn by McAlister and Hurd[7,23] at 4.2 K in the spin-glass and percolation regimes also showed a negative TMR. The present study confirms the existence of a negative TMR at all temperatures and fields, as described in Sec. IV. Also, the magnitude of the TMR increases with field and decreases with temperature. The occurrence of the negative TMR could be understood in terms of the spin-flip scattering theory of Béal-Monod and Weiner.[17] At low temperatures the external magnetic field modifies the population of the up and down spins of the impurity atom, thus decreasing the spin-flip scattering cross section. This produces a negative TMR whose magnitude increases with field. At higher temperatures thermal fluctuations mix these states, thereby making the external field less effective in reducing the spin-flip scattering. Consequently, the magnitude of the TMR would decrease with increasing temperature.

It was pointed out in Sec. IV that at higher fields the TMR was nearly independent of temperature below T_0 in all the alloys except the dilute ones (≤ 1 at. %). This could be understood as follows: The frozen state of the spin-glass can be disrupted either by increasing the temperature or by applying a magnetic field. At a sufficiently high field this state is already magnetically unlocked well below T_0. So as one increases the temperature thermal unlocking of spins becomes ineffective thereby making the spin-flip scattering insensitive to temperature.

The first prediction of Mookerjee's theory [Sec. II B(ii)] is found to be true in the present experimental study of the TMR. The quadratic dependence has been found to continue up to a field H_q which is different for different alloys, as shown in Table I. However, the present study also shows

TABLE I. Values of $\rho(T)$ (10^{-8} Ω m), $b(T,c)$ (10^{-3} T^{-2}), $a(T,c)$ (10^{-40} J^2K^{-2}), γ (10^{-31} mks units), H_q, T_0, and T_m for spin-glass alloys.

$T(K)$	$\rho(T)$	$b(T,c)$	$a(T,c)$	$T(K)$	$\rho(T)$	$b(T,c)$	$a(T,c)$	$T(K)$	$\rho(T)$	$b(T,c)$	$a(T,c)$
*Au*Fe (2.9 at.%), $\gamma=3.24$, $H_q=1.0$ T, $T_0=16$ K, $T_m\simeq140$ K				*Au*Fe (6.6 at.%), $\gamma=0.57$ $H_q=0.5$ T, $T_0=25.5$ K, $T_m\simeq300$ K				*Au*Fe (10.0 at.%), $\gamma=0.17$, $H_q=0.3$ T $T_0=35$ K, $T_m>300$ K			
4.3	22.8	2.42	1.79	1.5	51.4	4.48	1.32	4.2	53.4	21.0	1.95
6.6	22.9	2.35	1.75	4.2	51.4	4.50	1.33	10.0	53.5	31.7	2.95
12.5	23.0	2.05	1.53	9.0	51.6	4.63	1.37	20.0	53.8	39.0	3.65
20.0	23.3	1.78	1.34	20.0	52.0	4.34	1.29	30.0	54.2	40.4	3.81
43.5	23.4	1.02	0.77	30.0	52.2	4.26	1.28	77.0	57.2	5.7	0.56
77.0	24.1	0.24	0.19	77.0	54.1	0.62	0.19				
*Au*Mn (1.8 at.%), $\gamma=0.84$, $H_q=0.25$ T, $T_0=7$ K, $T_m\simeq77$ K				*Au*Mn (4.6 at.%), $\gamma=0.26$, $H_q=0.3$ T, $T_0=16$ K, $T_m=100$ K				*Cu*Mn (0.7 at.%), $\gamma=44.0$, $H_q=0.4$ T $T_0=8$ K, $T_m=17$ K			
1.5	4.77	47.8	1.90	2.6	13.3	42.0	1.46	1.5	2.61	10.7	12.3
2.3	4.81	49.5	1.99	4.2	13.4	45.5	1.59	4.2	2.69	10.0	11.8
4.2	4.88	56.3	2.29	9.0	13.6	71.0	2.14	6.0	2.71	9.3	11.1
6.0	4.90	50.5	2.06	12.0	13.8	80.0	2.87	8.0	2.75	8.8	10.6
10.0	4.95	30.0	1.24	16.0	13.8	80.0	2.88	10.0	2.77	8.4	10.2
18.0	5.01	10.0	0.42	20.0	14.0	60.0	2.19	12.0	2.79	7.7	9.4
30.0	5.06	5.4	0.23	35.0	14.3	27.0	1.01	15.0	2.80	5.0	6.2
				77.0	15.2	6.5	0.40				
*Cu*Mn (4.4 at.%), $\gamma=0.78$, $H_q=1.2$ T, $T_0\simeq25$ K, $T_m\simeq100$ K				*Ag*Mn (1.1 at.%), $\gamma=3.96$, $H_q=0.3$ T $T_0=5.5$ K, $T_m=43$ K				*Ag*Mn (5.4 at.%), $\gamma=0.55$, $H_q=0.6$ T $T_0=19$ K, $T_m=90$ K			
1.5	14.7	2.33	0.267	1.5	1.66	41.0	2.69	4.2	9.23	7.21	0.368
4.2	14.9	2.49	0.287	4.2	1.73	39.2	2.68	8.0	9.34	8.70	0.450
8.0	15.0	2.36	0.275	6.5	1.74	27.5	1.89	12.0	9.47	9.30	0.487
12.0	15.2	2.47	0.291	8.5	1.76	22.7	1.58	16.0	9.58	10.80	0.572
20.0	15.5	2.19	0.263	10.5	1.77	17.3	1.21	20.0	9.66	8.45	0.451
30.0	15.8	1.95	0.239	14.0	1.78	10.7	0.75	35.0	10.10	2.00	0.111
77.0	16.5	0.41	0.052	17.5	1.78	6.3	0.44	77.0	10.50	0.58	0.034

that well above T_0 the quadratic behavior of the negative TMR continues to the highest field of 18 kG. The second conclusion (an erroneous one) from the theory, namely, the linear proportionality of $\Delta\rho$ on c is nor borne out, at least by our data. This is not a disagreement since the theory does not predict such a simple relationship, as pointed out earlier. Nevertheless, we find that $\Delta\rho$ increases monotonically with concentration in all the systems. Since the essential features of the present experimental data agree qualitatively well with Mookerjee's formulation of the magnetoresistance theory in spin-glasses, it appeared logical to us to apply it in order to attempt a quantitative understanding of the experimental results. Recent measurements by McAlister and Kroeker[24] in *Au*Fe (1.1 at.%) and *Cu*Mn (0.93 at.%) clearly show that the phenomenological model of Li and Paton[25] is too simple to account for the magnetoresistance in spin-glasses. A fit of their data to the above theory yields an unrealistically large Curie-Weiss temperature ($\simeq$40 K). Moreover, they have too few low-field points.

A glance at Table II suggests that at a given temperature (4.2 K) and field (16 kG) $\Delta\rho$ increases monotonically with concentration in each of the spin-glass systems. In this table the data have been compared at 4.2 K because at this temperature the TMR has been measured in all the alloys. The values of $\Delta\rho/\rho$ (second column of Table II) have been found from Figs. 1(a)–1(i) at 16 kG, and multiplied by the corresponding ρ at 4.2 K to obtain $\Delta\rho$ (third column of Table II). Here, one finds that $\Delta\rho$ varies roughly as c^2 for *Au*Fe, as c for *Au*Mn, and slower than c for *Cu*Mn and *Ag*Mn alloys. Therefore, it is clear that the concentration dependence of $\Delta\rho$ is not a unique one. It is interesting to note (Table II) that although $\Delta\rho$ increases with c for all the spin-glass systems, $\Delta\rho/\rho$ has no consistent concentration dependence. For *Au*Fe, $\Delta\rho/\rho$ increases with c while in *Au*Mn it is nearly independent of c. In *Cu*Mn and *Ag*Mn, $\Delta\rho/\rho$ decreases as c is increased. Rohrer[15] as well as McAlister and Hurd,[26] in their high-field measurements of magnetoresistance, also found a similar c dependence of $\Delta\rho/\rho$ in *Au*Fe and *Au*Mn alloys.

TABLE II. Values of $\Delta\rho/\rho$ and $\Delta\rho$ at 4.2 K and 16 kG for spin-glass alloys.

Alloy	$-\Delta\rho/\rho$ (10^{-3})	$-\Delta\rho$ (10^{-11} Ω m)
*Au*Fe (2.9 at.%)	6.4	146
*Au*Fe (6.6 at.%)	10.5	540
*Au*Fe (10.0 at.%)	35.2	1880
*Au*Mn (1.8 at.%)	61.0	298
*Au*Mn (4.6 at.%)	56.5	756
*Cu*Mn (0.7 at.%)	19.8	53
*Cu*Mn (4.4 at.%)	6.1	91
*Ag*Mn (1.1 at.%)	43.5	75
*Ag*Mn (5.4 at.%)	17.3	160

The exact evaluation of the integral in Eqs. (4) and (5) is not simple; hence we have evaluated[27] them in the approximations of low field and $T\to T_0$. In these limits, the simplified equations for $M(H)$ and $Q(H)$ are

$$M(H)\simeq\alpha T/(T-\Theta)\,, \tag{8}$$

$$Q(H)\simeq\frac{\alpha^2T_0^2(1+2\epsilon)}{2\Delta(\Delta+2\epsilon T_0)\epsilon}\quad\text{when } T>T_0\,, \tag{9}$$

and

$$Q(H)\simeq-\epsilon+\frac{\alpha^2T_0^2(1+6\epsilon)}{2\Delta(\Delta+2\epsilon T_0)(-\epsilon)}\quad\text{when } T<T_0\,, \tag{10}$$

where

$$\Delta=T_0-\Theta,\quad \epsilon=(T-T_0)/T_0\,.$$

Here, terms of the order higher than α^2 and ϵ are neglected with the additional restriction that ϵ should be much larger than α^2. In other words, the low-field approximation holds neither very near to T_0 nor very far away from it.

In Eq. (6) the ratio $|J/V|$ is expected to be small [$\simeq$0.16 for *Cu*Mn (Ref. 17)], and hence one could neglect the term containing J^2/V^2. Equation (6) for $\Delta\rho$ could now be simplified, using Eqs. (8)–(10), to the form

$$\Delta\rho\simeq-cR_0J^2\xi^2\left[\frac{1}{p_{\rm eff}}\frac{1}{T(T-\Theta)}+\frac{T_0^2(1+2\epsilon)}{T^2\Delta(\Delta+2\epsilon T_0)\epsilon}\right]\quad\text{for } T>T_0 \tag{11}$$

and

$$\Delta\rho\simeq-cR_0J^2\xi^2\left[\frac{1}{p_{\rm eff}}\frac{1}{T(T-\Theta)}+\frac{T_0^2(1+6\epsilon)}{T^2\Delta(\Delta+2\epsilon T_0)(-\epsilon)}\right]\quad\text{for } T<T_0\,, \tag{12}$$

where $\xi=\alpha T=\mu H/k_B$.

One can now calculate $\Delta\rho$ from Eqs. (11) and (12) provided the values of R_0, ξ, Θ, T_0, and J are known. The value of J had been calculated earlier from the experimental data of $No_{1-c}Tr_c$ alloys[15,17] where c was much smaller than those of the spin-glasses; hence it may be questionable to use such values of J in Eqs. (11) and (12). Therefore, we have applied the above theory in a reverse manner, i.e., rather than calculating $\Delta\rho$ from the above equations, we have substituted our experimental value of $\Delta\rho$ in Eqs. (11) and (12) and, in turn, determined the effective value of J in the spin-glass regime. The procedure for calculating $|J|$ from the present experimental study is here outlined. We first define

$$a(T,C)=-\Delta\rho/(cR_0\xi^2)\ ,$$

which, from Eqs. (11) and (12), takes the form

$$a(T,c)\simeq J^2\left[\frac{1}{p_{\text{eff}}}\frac{1}{T(T-\Theta)}+\frac{T_0^2(1+2\epsilon)}{T^2\Delta(\Delta+2\epsilon T_0)\epsilon}\right] \quad \text{for } T>T_0 \tag{13}$$

and

$$a(T,c)\simeq J^2\left[\frac{1}{p_{\text{eff}}}\frac{1}{T(T-\Theta)}+\frac{T_0^2(1+6\epsilon)}{T^2\Delta(\Delta+2\epsilon T_0)(-\epsilon)}\right] \quad \text{for } T<T_0\ . \tag{14}$$

The value of $a(T,c)$ could also be calculated from the experimental data as follows:

$$a(T,C)=-\frac{\Delta\rho}{cR_0\xi^2}=-\frac{\Delta\rho}{\rho}\frac{\rho(T)}{cR_0p_{\text{eff}}^2H^2}\left[\frac{k_B^2}{\mu_B^2}\right], \tag{15}$$

where $\rho(T)$ and ρ are the same, i.e., the electrical resistivity of the alloy at a temperature T. In Eq. (7) when $n=2$ one gets

$$\frac{\Delta\rho}{\rho}=-b(T,c)H^2\ . \tag{16}$$

Substituting Eq. (16) into Eq. (15), one obtains, in meter-kilogram-second (mks) units,

$$\begin{aligned}a(T,c)&=\frac{\rho(T)b(T,c)}{cR_0p_{\text{eff}}^2}\left[\frac{k_B^2}{\mu_B^2}\right]\\&=\frac{2.3}{cR_0p_{\text{eff}}^2}\rho(T)b(T,c)\\&=\gamma\rho(T)b(T,c)\ ,\end{aligned} \tag{17}$$

where $\gamma=2.3/(cR_0p_{\text{eff}}^2)$.

The values of $\rho(T)$ and $b(T,c)$ for various alloys at different temperatures are given in Table I. The values of R_0 have been calculated from Eq. (3), assuming that the Fermi energy E_F and the number of electrons per unit volume n are the same as those for their host metals. The values of p_{eff} have been obtained from the experimental data of magnetic susceptibility.[1,2,28] Thus one can find the value of $a(T,c)$ from Eq. (17). The values of R_0, and p_{eff} as well as of T_0 and Θ are given in Table III for each alloy.

The values of $a(T,c)$ are given in Table I. They have also been plotted in Figs. 3 and 4 as a function of temperature for the various alloys (*Au*Fe, *Au*Mn, *Cu*Mn, and *Ag*Mn). From these plots the values of $a(T,c)$ corresponding to $\epsilon=1-T/T_0=\pm0.1$ are obtained and have been substituted in Eqs. (13) and (14) to determine $|J|$. The values of $|J|$, thus computed, have also been listed in Table III. It should be noted here that at low fields $\alpha^2<0.01$, and so the restriction that $\epsilon\gg\alpha^2$ holds [see remark below Eq. (10)].

Furthermore, in Figs. 3 and 4, one observes for each alloy that $a(T,c)$ varies smoothly around T_0 without showing any anomaly. However, many of them exhibit peaks at temperatures (always less than T_0) that do not have any specific correlation with respect to T_0. To resolve these peaks and to establish the corresponding temperatures correctly it is necessary to be able to detect very small magnetoresistance ($\simeq10^{-5}$) at fields as low as 100 G. Thus in this work no generalization could be made regarding the peaks observed in $a(T,c)$.

B. Coulomb scattering potential V

From Eq. (2) at $T>T_0$, the zero-field resistivity is given by

$$\rho(0)=cR_0[V^2+J^2S(S+1)]\ .$$

The values of $\rho(0)$, which is the magnetic contribution to the resistivity, have been obtained for the *Au*Mn, *Cu*Mn, and *Ag*Mn alloys from the data of

TABLE III. Values of E_f, n R_0, T_0, Θ, p_{eff}, and $|J|$ for spin-glass alloys.

Alloy	E_F[a] (eV)	n[a] (10^{28} m^{-3})	R_0 (10^{31} Ω mJ^{-2})	T_0[b,c,d] (K)	Θ[b,c,e] (K)	p_{eff}[b,c,e]	$\lvert J\rvert$ (eV) $\epsilon=+0.1$	$\lvert J\rvert$ (eV) $\epsilon=-0.1$	$\lvert J_{\text{av}}\rvert$ (eV)
*Au*Fe (2.9 at.%)	5.5	5.9	1.53	16	−5	4.0	0.53	0.66	0.60
*Au*Fe (6.6 at.%)				25.5	2	6.0	0.52	0.60	0.56
*Au*Fe (10.0 at.%)				35.0	10	9.0	1.04	1.15	1.10
*Au*Mn (1.8 at.%)	5.5	5.9	1.53	7.0	−1	10.0	0.22	0.30	0.26
*Au*Mn (4.6 at.%)				16.0	5	11.2	0.40	0.45	0.43
*Cu*Mn (0.7 at.%)	7.0	8.5	0.83	8.0	3	3.0	0.37	0.37	0.37
*Cu*Mn (4.4 at.%)				25.0	−5	9.0	0.32	0.38	0.35
*Ag*Mn (1.1 at.%)	5.5	5.76	1.57	5.5	−1	5.8	0.20	0.25	0.23
*Ag*Mn (5.4 at.%)				19.0	12	7.0	0.11	0.10	0.11

[a]Reference 29.
[b]Reference 28.
[c]Reference 2.
[d]Reference 8.
[e]Reference 1.

Ford and Mydosh[8] at 300 K. These are given in Table IV. At this temperature a value of $S=\frac{5}{2}$ could be used since the alloys are in the paramagnetic state. Thus the values of V could be calculated (using R_c, c, and $|J|$ from Table III) from the above equation and are given in Table IV. One finds in this table that V is much greater than $|J|$ (given in Table III) and hence, the assumption that $|J/V|$ is small, in applying the theory of Mookerjee to the present experimental data, was justified.

C. Comments on the values of the *s-d* exchange parameter *J*

It is interesting to note (Table II) that, in the *Au*Fe system, though $\Delta\rho$ changes by a factor of 4 when Fe concentration is changed from 2.9 to 6.6 at.%, $|J|$ is nearly the same ($\simeq$0.6 eV) for the two alloys. This implies an almost unchanged strength of the "*s-d* exchange interaction" around T_0 in the spin-glass regime of concentration.

In the case of *Au*Fe (10.0 at.%) the value of $|J|$ is twice ($\simeq$1.1 eV) that of the other two alloys. This is very likely because the *Au*Fe (10.0 at.%) alloy falls in the so-called mictomagnetic regime[8] where ferromagnetic and spin-glass interactions of comparable strength coexist. The existence of ferromagnetic interaction gives an additional contribution to the negative TMR. Since the ferromagnetic contribution to the TMR could not be separated, the obtained value of $|J|$ not only corresponds to the spin-glass indirect exchange interaction but also includes the direct Fe-Fe interaction. This latter interaction parameter ($|J_{dd}|$) had been determined from resistivity by Hedgcock *et al.*[30] in the case of *Cr*Fe alloys, and was found

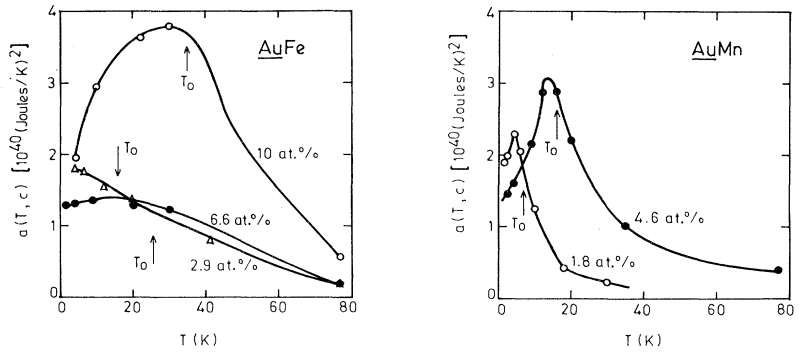


FIG. 3. Plots of $a(T,c)$ vs temperature T for *Au*Fe and *Au*Mn spin-glass systems.

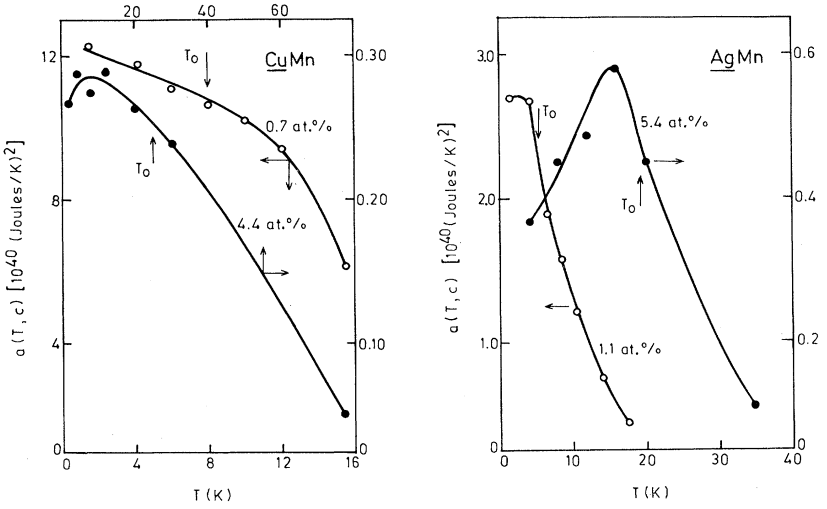


FIG. 4. Plots of $a(T,c)$ vs temperature T for *Cu*Mn and *Ag*Mn spin-glass systems.

to be $\simeq$0.5 eV. If we subtract this numerical value of $|J_{dd}|$ from the effective $|J|$ obtained for our *Au*Fe (10.0 at.%) alloy, we find almost the same value ($\simeq$0.6 eV) of the spin-glass indirect exchange parameter $|J|$ as obtained for the other two alloys with 2.9 and 6.6 at.% Fe. Furthermore, from magnetoresistance measurements, Monod[31] had computed $|J|$ for *Cu*Fe (0.011 at.%) and found $|J| \simeq 0.9 \pm 0.2$eV, which is quite close to that computed in the present study of *Au*Fe alloys.

For *Cu*Mn spin-glasses we obtained $|J| \simeq 0.35$ eV, which is in good agreement with that found by Monod[31] ($|J| \simeq 0.4 \pm 0.1$ eV) for dilute *Cu*Mn ([Mn]=75 ppm). For *Au*Mn spin-glasses $|J|$ is significantly different (0.26 eV for 1.8 at.% Mn and 0.43 eV for 4.6 at.% Mn) for the two alloys of the present study. It is not clear why the strength of the *s-d* exchange scattering has stronger dependence on the magnetic impurity concentration in this system. From high-field magnetoresistance measurements a value of $|J| \simeq 0.18$ eV had been computed earlier by Rohrer[15] for more dilute *Au*Mn alloys.

In the *Ag*Mn system, the values of $|J|$ are also found to be different (0.23 eV for 1.1 at.% Mn and 0.11 eV for 5.4 at.% Mn) for the two alloys. But it is difficult to make any comment, as the values of p_{eff} and Θ could not be reliably ascertained from the available literature data. Kok,[32] from the zero-field resistivity calculations, reported a value of $|J| \simeq 0.29$ eV for *Ag*Mn (5.0 at.%), which is more than twice the value we obtained.

In conclusion, the values of $|J|$ (Table III) computed from the present experimental data of magnetoresistance are of the same order of magnitude as those determined by others through their studies of transport properties. This gives us confidence that the theory of Mookerjee gives a correct qualitative picture as well as a good enough quantitative estimate of the magnetoresistance in spin-glasses.

TABLE IV. Values of $\rho(H=0)$ at 300 K and calculated Coulomb potential V for *Au*Mn, *Cu*Mn, and *Ag*Mn spin-glasses.

Alloy	$\rho(H=0)$[a] (10^{-8} Ω m)	V (eV)
*Au*Mn (1.8 at.%)	4.57	2.5
*Au*Mn (4.6 at.%)	11.25	2.5
*Cu*Mn (0.7 at.%)	2.09	3.7
*Cu*Mn (4.4 at.%)	12.20	3.6
*Ag*Mn (1.1 at.%)	1.62	1.9
*Ag*Mn (5.4 at.%)	8.68	2.0

[a]Reference 8.

D. Magnetization and field dependence of $\Delta\rho$

Since $|J/V|$ is expected to be small, one could write Eq. (6) as

$$\Delta\rho \simeq -cR_0J^2\left[M(H)\tanh\left[\frac{g\mu_BH}{2k_BT}\right] + 2[Q(H)-Q(0)]\right]. \quad (18)$$

From the definition of the order parameter intro-

duced in Sec. I we know that $Q(H)$ varies with the square of magnetization, and therefore one could write

$$Q(H)=Q(0)+q_1M^2(H)\ . \tag{19}$$

Substituting Eq. (19) into Eq. (18) we get

$$\Delta\rho\simeq -cR_0J^2\left[M(H)\tanh\left[\frac{g\mu_BH}{2k_BT}\right]+2q_1M^2(H)\right], \tag{20}$$

where $M(0)=0$. The above Eq. (20) shows that $\Delta\rho$ depends on magnetization which, in turn, is a function of field. It is found that the first term (calculated around T_0 for the determination of J) on the right-hand side is much smaller than the second term, and therefore one could say that $\Delta\rho$ varies roughly as the square of magnetization. This had been recently found by Senoussi,[33] who measured the magnetoresistance in *Au*Fe spin-glasses (when the specimen was zero-field cooled) and computed the value of magnetic susceptibility χ from the expression

$$\Delta\rho/\rho=-\beta M^2=-\beta\chi^2H^2\ , \tag{21}$$

where β is a constant. With the use of Eq. (21) and the magnetoresistance data from his measurements and from those of Nigam and Majumdar[34] for *Au*Fe (2.9 at. %) alloy, Senoussi calculated χ and found surprisingly good agreement with the reported values of χ obtained from direct magnetic measurements. Table V displays the values of magnetoresistance from the present study and the values of χ as calculated by us from Eq. (21) with $\beta=8\times10^{-2}$ (emu/g)$^{-2}$ (Ref. 33) for *Au*Fe (2.9 at. %) and *Au*Fe (6.6 at. %). The literature values of χ from direct magnetic measurement are given in the fourth column of the table for comparison. The agreement is rather good for our *Au*Fe alloys. A quadratic dependence of the negative TMR on magnetization had also been found by Schmitt and Jacobs[13] and by Monod[31] in dilute alloys of *Cu*Mn and *Cu*Fe. These results were interpreted in terms of Béal-Monod and Weiner theory[17] of magnetoresistance.

It has been found experimentally that, in low fields, the dc magnetization[35] of the spin-glasses varies linearly with H, that is, $M(H)=\eta H$, η being the constant of proportionality. Therefore, one could write from Eq. (20) that

$$\Delta\rho\simeq -cR_0J^2\left[\left[\eta H\frac{g\mu_BH}{2k_BT}\right]+2q_1\eta^2H^2\right]$$
$$=-cR_0J^2\left[\frac{\eta g\mu_B}{2k_BT}+2q_1\eta^2\right]H^2\ ,$$

which means that $\Delta\rho$ is proportional to H^2 at all temperatures rather than only around T_0 [as deduced from Eqs. (11) and (12)]. This is what has been observed in the present study (Fig. 2). The measurements of magnetization by McAlister and Freeman[35] in *Au*Fe (4.2 at. %) at 4.2 K revealed an almost linear M vs H plot up to about 5 kG, while in *Cu*Mn (5.5 at. %), Kouvel[36] had observed an almost linear dependence of M on H up to 10 kG. It is worth noting that these values of fields for *Au*Fe and *Cu*Mn are in fair agreement with the fields H_q up to which the quadratic field dependence of the TMR has been observed in *Au*Fe (6.6 at. %) and *Cu*Mn (4.4 at. %) (Table I).

Furthermore, the observed quadratic behavior of $\Delta\rho/\rho$ with H at 77 K up to 16 kG in all our alloys shows that even in the paramagnetic state, $\Delta\rho/\rho$ is proportional to the square of magnetization. This is because in the present work the dependence of M on H at 77 K has been found experimentally to be linear for all the alloys. A few of them, namely, *Au*Fe (6.6 at. %), *Au*Mn (4.6 at.&%), *Cu*Mn (4.4 at. %), and *Ag*Mn (5.4 at. %) are shown in Fig. 5. This implies that $\Delta\rho/\rho$ would have almost the same power dependence on M as it has on H.

E. Effects of field cooling, remanence, aging, and annealing on TMR

It has been found in a few alloys of this work that the TMR is barely affected by cooling the sample through T_0 either in zero field or in a mag-

TABLE V. Magnetoresistance $\Delta\rho/\rho$ and magnetic susceptibility χ of *Au*Fe spin-glasses.

Alloy	$-\Delta\rho/\rho$ at 10 kG	χ_{calc} [(emu/g)/G]	χ_{lit} [(emu/g)/G]
*Au*Fe (2.9 at. %)	2.65×10^{-3} (2.5 K)	17×10^{-6}	12×10^{-6}[a]
*Au*Fe (6.6 at. %)	4.3×10^{-3} (2.9 K)	23×10^{-6}	25×10^{-6}[b]

[a]Reference 28.

[b]Reference 33 [for a *Au*Fe (7 at. %) alloy].

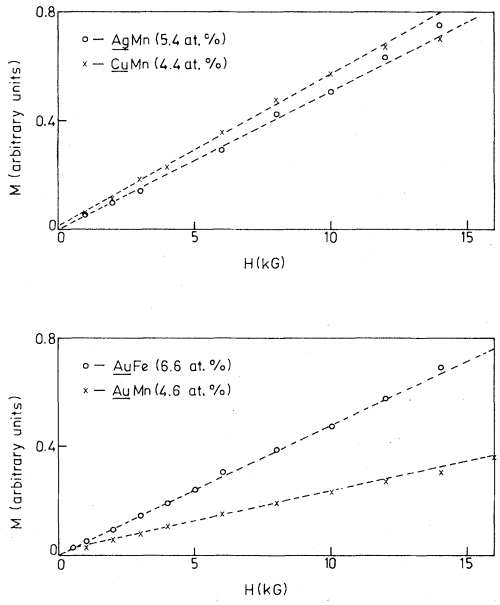


FIG. 5. Magnetization M vs external magnetic field H at 77 K for *Au*Fe (6.6 at. %), *Au*Mn (4.6 at. %), *Cu*Mn (4.4 at. %), and *Ag*Mn (5.4 at. %) spin-glass alloys.

netic field, as, for example, a *Cu*Mn (4.4 at. %) alloy shows a slight increase in the magnitude of the negative TMR when it is cooled through its T_0 ($\simeq$16 K) in a magnetic field of 15 kG. This enhancement, which could be due to the thermoremanent magnetization, however, is not significant. It had been observed by Schmitt and Jacobs[13] that the magnetization of a field-cooled *Cu*Mn sample was initially enhanced, resulting in an open hysteresis loop.

The magnetization in spin-glasses shows the hysteresis effect[37] at temperatures well below T_0. Here the effect has been studied for a few alloys at 4.2 K. It is observed that the relatively concentrated alloys having T_0 greater than about 15 K exhibit remanence effect in the TMR at 4.2 K. This is shown in Fig. 1(i) at 4.2 K for *Ag*Mn (5.4 at. %) and clearly indicates that the magnetoresistance depends on magnetization rather than on magnetic field. No remanence is observed in the TMR for temperatures above T_0.

A fresh annealing and quenching of *Au*Fe alloys with Fe concentrations of 6.6 and 10.0 at. % has shown significant reduction of the negative TMR. The same has not been studied in other alloy systems except in the *Au*Mn (1.8 at. %) alloy. The alloys of *Au*Fe (6.6 and 10.0 at. %) and *Au*Mn (1.8 at. %) were measured first in the as-received and aged condition. Then they were annealed at 900 °C and given the same heat treatment as described in Sec. III A. In Fig. 6, $\Delta\rho/\rho$ has been plotted against H at 4.2 K for *Au*Fe (6.6 at. %), and at 77 K for *Au*Fe (10.0 at. %). It is observed that in the *Au*Fe (6.6 at. %) alloy at 4.2 K and 18 kG the magnitude of $\Delta\rho/\rho$ decreases from 2.1% (for the aged sample) to 1.2% (for the freshly annealed and quenched sample). This quenching of the negative TMR in *Au*Fe (6.6 at. %) was found at all temperatures below and above T_0. In the case of the *Au*Fe (10.0 at. %) alloy, the data have been compared at 77 K (Fig. 6) since the aged specimen was measured only at 77 K and above. It shows a reduction of the magnitude of the negative TMR from 4.2% (for the aged sample) to 1.4% (for the freshly annealed and quenched sample) at 18 kG. The TMR in the *Au*Mn (1.8 at. %) alloy did not show any change on annealing.

Scheil *et al.*[38] and Sundahl *et al.*[39] found that a prolonged aging of concentrated *Au*Fe alloys considerably increased the magnetic susceptibility as well as the transition temperature. Since aging enhances T_0 one finds at a temperature T that the ratio T/T_0 is larger for the quenched alloy than that for the aged sample. Therefore, the magnitude of the negative TMR in the quenched alloy is smaller than that of the aged one.

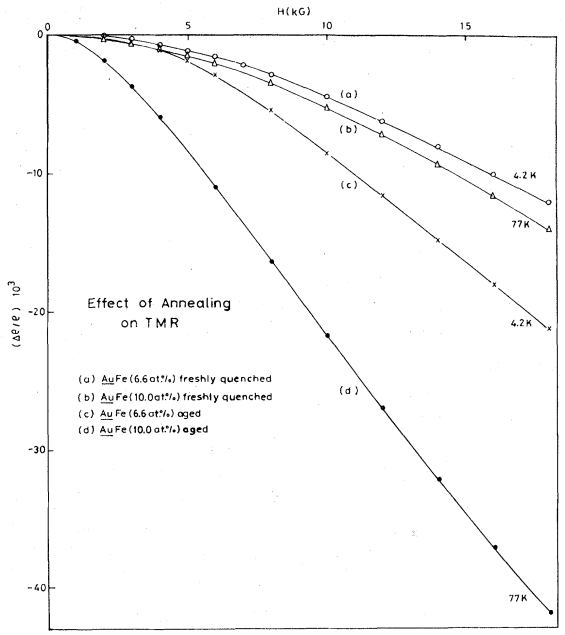


FIG. 6. Transverse magnetoresistance $\Delta\rho/\rho$ vs external magnetic field H in *Au*Fe (6.6 at. %) at 4.2 K and in *Au*Fe (10.0 at. %) at 77 K to show the effect of aging and annealing (followed by quenching) on TMR.

F. Effect of superconducting solder on TMR

A low-field positive magnetoresistance was observed[34,40] in a few spin-glasses when the electrical leads were joined to the specimen with $Cd_{70}Sn_{30}$ solder. Senoussi[33] had pointed out that the observed positive TMR could be due to the superconductivity of the solder material. In these samples no positive magnetoresistance could be detected when the experiments were repeated with leads spot-welded to the sample. The positive contribution, however, in the case of soldered leads, continued up to temperatures (T_c) and fields (H_c) that varied over wide ranges in different spin-glasses ($T_c = 3.5-10$ K and $H_c = 0.5-6.0$ kG). Even in the same alloy they varied considerably in different runs. The superconducting transition temperature T_c of the solder material is found to be 3.52 K (Ref. 41) and the critical field H_c is expected to be around 300 G (Ref. 42). So it is clear that the positive TMR could not be due to the solder material alone. Perhaps new superconducting phases are formed by alloying Cd or Sn with the noble metals from the alloys.[43]

G. Concluding remarks

The present work on the transverse magnetoresistance in spin-glasses does not yield any sharp anomaly at the transition temperature T_0. However, one finds a faster rate of decrease of the TMR above T_0. The magnetoresistance experiments, therefore, seem to fall under that category where a smeared behavior is found around T_0.

The theory of Mookerjee seems to describe most of the essential features observed in the present experiments. Both Mookerjee's theory and the present experimental data indicate that the magnetoresistance may be a unique function of magnetization rather than of the field. The values of the *s-d* exchange parameter J, determined from the present experimental data, have the correct order of magnitude. This favors, at least, an approximate quantitative validity of Mookerjee's theory of magnetoresistance in spin-glasses.

The magnetoresistance in spin-glasses at low fields is very small, and hence it is quite difficult to find its correct temperature dependence in the present experimental setup. It is, therefore, highly desirable to make extremely low-field measurements using a better detection system.

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