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著者	Hihara T., Sumiyama K., Wakoh K., Suzuki K.
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Magnetic-Field- and Temperature-Dependent Characteristics of Fe/Cu Granular Films Produced by Sputter- and Cluster-Beam-Deposition*

T.Hihara, K.Sumiyama, K.Wakoh and K.Suzuki

Institute for Materials Research, Tohoku University, Sendai 980-77 , Japan

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The magnetoresistance (*MR*) and magnetization (*M*) have been measured as functions of temperature, *T*, and magnetic field, *H*, in sputter(SP)- and cluster-beam(CB)-deposited Fe_XCu_{100-X} alloys. The *MR* for the SP-deposited $Fe_{20}Cu_{80}$ film exhibits a maximum at around the Curie temperature (T_C= 150 K) and increases rapidly below 100 K owing to a spin-glass transition. For the CB-deposited granular Fe-Cu films with low Fe content, the *MR* is also enhanced at low temperatures being attributable to a cluster-glass behavior of small fcc Fe clusters. At low temperatures, moreover, a $T^{3/2}$ dependence is found for both *MR* and *M* versus *T* curves. For the SP-deposited $Fe_{20}Cu_{80}$ sample, the $T^{3/2}$ coefficient of the *MR* roughly corresponds with that of the *M*. For the CB-deposited $Fe_{22}Cu_{78}$ sample, however, they are inconsistent each other. These results originate from different status of the Fe atoms: the Fe atoms are rather randomly dispersed in the Cu matrices in the SP-deposited sample, while they form clusters in the CB-deposited one.

KEYWORDS: iron-copper, granular film, magnetoresistance, magnetization

1. Introduction

A great deal of experimental and theoretical works have been reported for the granular alloy systems which show such a giant magneto-resistance (*GMR*) behavior as observed in the magnetic multilayers[1,2]. In addition to the *GMR*, they reveal unusual transport properties such as extraordinary Hall effect[3], giant magnetothermal conductivity and magnetothermopower[4]. Spin-dependent scattering of conduction electrons at interfaces between magnetic clusters and conducting matrices plays an important role in these phenomena. Therefore, the *GMR* in the granular films is affected by the size and density of the magnetic clusters, which depend on the chemical composition and the preparation conditions. The ultimate state is the nonequilibrium homogeneous alloy, where the cluster size is about an atomic scale. Fe-Au and Fe-Ag solid solution have been produced by the vapor quenching techniques, e.g. sputtering and thermal evaporation[5]. In the sputter(SP)-deposited Fe_xCu_{100-x} alloys, an fcc structure is stabilized for x < 40 and a bcc structure for x > 60. They show two distinct magnetic characters: a spin-glass state in low Fe concentration alloys and a ferromagnetic state in high Fe concentration ones[6,7]. These phenomena have been discussed in a frame work of a percolation of magnetic exchange interactions[8].

An ionized cluster beam (ICB) technique has been applied in preparing high quality films[9]. The clusters are formed by an adiabatic expansion and subsequent cooling when vaporized atoms are ejected through a crucible nozzle. In previous papers, we have reported the magnetic and electrical properties of Fe/Ag granular films produced by the CB technique[10]. Without any heat treatment, these films exhibit the *GMR*. The *MR* ratio at 4.2 K decreases nonlinearly with increasing a magnetic field and does not saturate even at 140 kOe, although the magnetization saturates easily above 2 kOe. These results indicate the important role of the spin-dependent scattering of conduction electrons at the interface between Fe clusters and Ag matrices.

In the present study, we measure magnetic-field- and temperature-dependence of electrical resistivity and magnetization for CB-deposited heterogeneous Fe/Cu films and SP-deposited homogeneous Fe-Cu films. We analyze their characteristic features using the spin-wave concept and discuss the scattering of conduction electrons by magnetic moments at the interface.

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2. Experimental Procedures

Fe_xCu_{100-x} granular films were prepared on a polyimide film by an ultrahigh-vacuum-type CB equipment. Fe clusters formed by adiabatic expansion of vaporized atoms through the crucible nozzle were deposited on the substrate. Simultaneously, copper vapor was deposited by thermal evaporation from a graphite crucible. The background pressure before deposition was 1×10^{-7} Pa. The chemical composition was controlled using a crystal-oscillation thickness gauge calibrated by an induction-coupled plasma (ICP) analysis.

An Fe-Cu alloy film containing 20at%Fe was also prepared on a water-cooled substrate using a facing-target-type DC sputtering apparatus. The background pressure before deposition was better than 1×10^{-4} Pa. The chemical composition was adjusted by the relative area of a composite target and analyzed by EPMA.

With a standard four probe method, electrical resistivity was measured in magnetic field, *H*, up to 140 kOe at 4.2 K and in *H* up to 50 kOe between 5 and 300 K. Magnetization was measured between 5 and 300 K using a SQUID magnetometer.

3. Results and Discussion

Figures 1(a) and (b) show the magnetoresistance (*MR*) curves for the SP-deposited Fe-Cu and CB-deposited Fe/Cu films[11]. The ordinate represents the magnetoresistance ratio, $\Delta\rho/\rho(0)$, as a function of an applied magnetic field, *H*, up to 140 kOe. Here, $\Delta\rho/\rho(0)$ is defined as follows:

$$\Delta\rho/\rho = [\rho(H) - \rho(0)] / \rho(0) , \qquad (1)$$

where $\rho(H)$ is the electrical resistivities at a magnetic field, *H*. $\Delta\rho/\rho$ increases almost linearly with increasing *H* in the as-deposited state. On the other hand, the magnetization at low magnetic fields for both SP-deposited Fe-Cu and CB-deposited Fe/Cu films are shown in Fig.2(a) and (b). These results are similar to

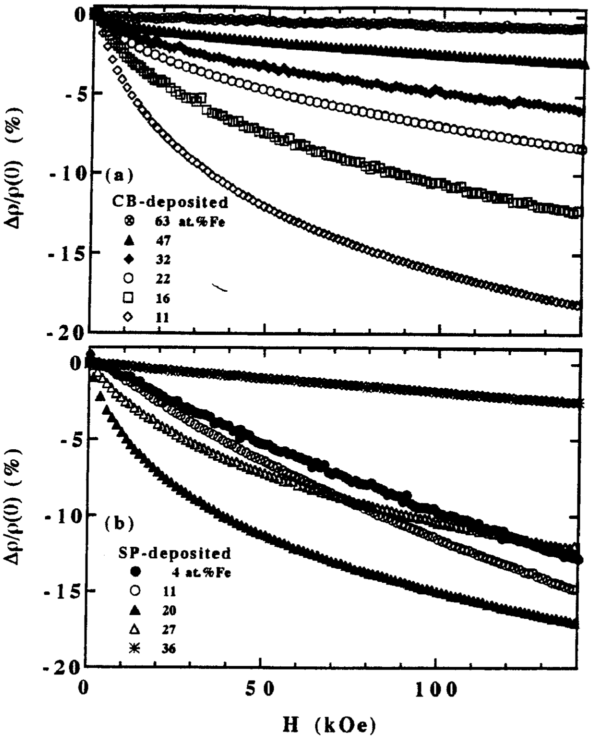

Figure 1 Magnetoresistance ratio, $\Delta\rho/\rho(0)$, as a function of an applied magnetic field, H, at 4.2 K. (a) CB-deposited Fe/Cu films and (b) SP-deposited Fe-Cu films.

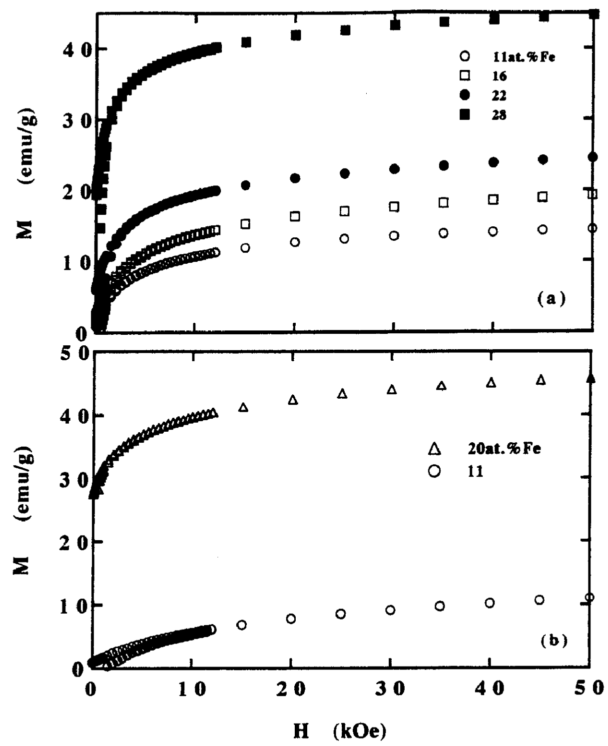

Figure 2 Magnetization, M, as a function of an applied magnetic field, H, at 5 K. (a) CB-deposited Fe/Cu films and (b) SP-deposited Fe-Cu films.

those for the SP-deposited Fe/Ag and the CB-deposited Fe-Ag films.

3.1 SP-deposited homogeneous $Fe_{20}Cu_{80}$ film

Figure 3(a) shows $\Delta\rho(H)/\rho(0)$ versus H plots at different temperatures for the SP-deposited $Fe_{20}Cu_{80}$ film. $\Delta\rho(H)/\rho(0)$ measured at 300 K monotonically decreases with increasing H up to 50 kOe. Below 100 K, its non-linear character becomes more remarkable but does not saturate up to 50 kOe. Figure 3(b) shows temperature dependence of resistivity, $\rho(T)$, in $H = 0$ and 50 kOe for the SP-deposited $Fe_{20}Cu_{80}$ film. $\rho(T)$ at 50 kOe is smaller than that at 0 kOe because scattering of conduction electrons by magnetic impurities is reduced by an external magnetic field. Figure 4 shows $\Delta\rho$ versus T curve for the SP-deposited $Fe_{20}Cu_{80}$ film: $\Delta\rho=\rho(0)-\rho(50\ \text{kOe})$. With decreasing T, $\Delta\rho$ increases rapidly, has a maximum at around 180 K and decreases gradually down to 70 K. With further decrease of T, $\Delta\rho$ increases markedly below 70 K. In previous paper, we reported the magnetic phase diagram based upon the magnetization measurements[11]. In this concentration, the ferromagnetic order appears below Curie temperature, T_C, of about 150 K and the spin-glass phase below freezing temperature, T_f, of about 50 K.

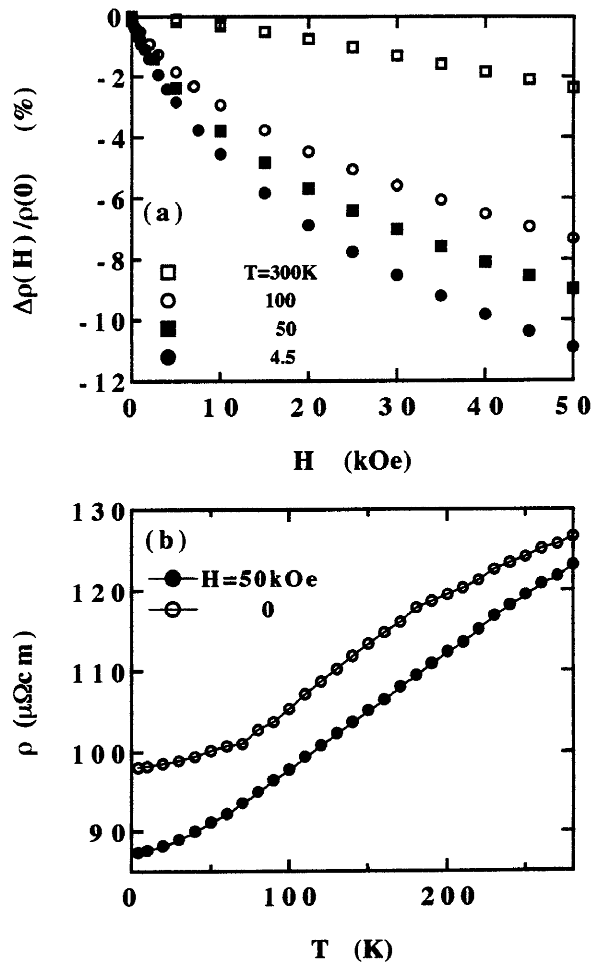

Figure 3 (a)Magnetoresistance ratio, $\Delta\rho/\rho(0)$, as a function of an applied magnetic field, H, and (b)temperature dependence of resistivity in $H = 0$ and 50 kOe for the SP-deposited $Fe_{20}Cu_{80}$ film.

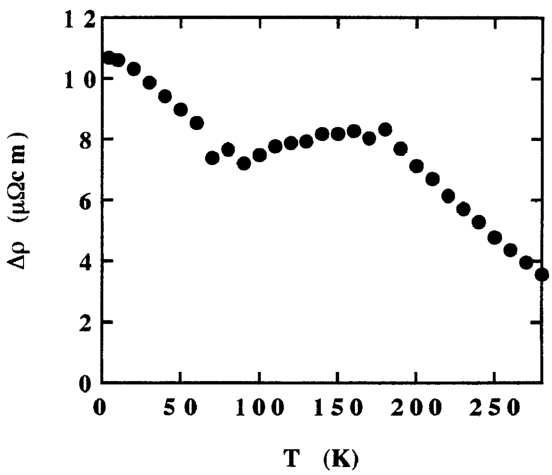

Figure 4 Temperature dependence of $\Delta\rho = \rho(0) - \rho(50$ kOe) for the SP-deposited $Fe_{20}Cu_{80}$ film.

The temperature dependence of $\Delta\rho$ at low temperatures is similar to that of the hyperfine field observed by Mössbauer spectroscopy[12]. In the ferromagnetic region ($T_f < T < T_C$), the hyperfine field reflects only the longitudinal spin component along the direction of the spontaneous magnetization, S_Z. Below T_f, the increase in the hyperfine field has been interpreted as being due to the freezing of the transverse component of local Fe moment, S_T, together with S_Z. The enhanced $\Delta\rho$ at low temperatures ($T < T_f$) shown in Fig. 4 is ascribed to a larger effective spin, $S_T + S_Z$. With increasing T, an inelastic spin-flip scattering, such as an electron-magnon scattering, gradually emerges from the intra-cluster excitations and weakens above T_C since the ferromagnetic correlation length drops rapidly and crosses over an electron mean free path, l [13].

Figures 5(a) and (b) display normalized magnetization, $M(T)/M(0)$, and normalized magnetoresistance, $\Delta\rho(T)/\Delta\rho(0)$, against $T^{3/2}$. For $T < 50$ K, $M(T)/M(0)$ and $\Delta\rho(T)/\Delta\rho(0)$ are well described by straight lines. It is known that temperature dependence of the magnetization for ferromagnetic materials follows Bloch's law at low temperatures.

$$M_S(T) = M_S(0)\,(1 - B\,T^{3/2}), \qquad (2)$$

where B is a constant related to a stiffness constant of spin waves.

MR is an extra electrical resistivity, $\Delta\rho$, due to scattering from nonaligned ferromagnetic entities. The $\Delta\rho$ depends on thermal average of magnetic moments, $\langle\cos\phi_{ij}\rangle$ where ϕ_{ij} is the angle between the magnetic axes of the ferromagnetic clusters. Since $\langle\cos\phi_{ij}\rangle = \langle\cos\theta\rangle^2$ and $M^2 / M_S^2 = \langle\cos\theta\rangle^2$ for a single domain system with the angle, θ, between the magnetization axis of the cluster and the applied magnetic field, $\Delta\rho$ is proportional to the M^2 provided that all the granules have the same size[14]. In this case, temperature dependence of *MR* follows eq.(3) at low temperatures.

$$\frac{\Delta\rho(T)}{\Delta\rho(0)} \propto \left(\frac{M_S(T)}{M_S(0)}\right)^2 = 1 - 2B_{MR}\,T^{3/2} + B_{MR}^2\,T^3$$
$$\approx 1 - 2B_{MR}\,T^{3/2} \quad (B_{MR} \ll 1) \qquad (3)$$

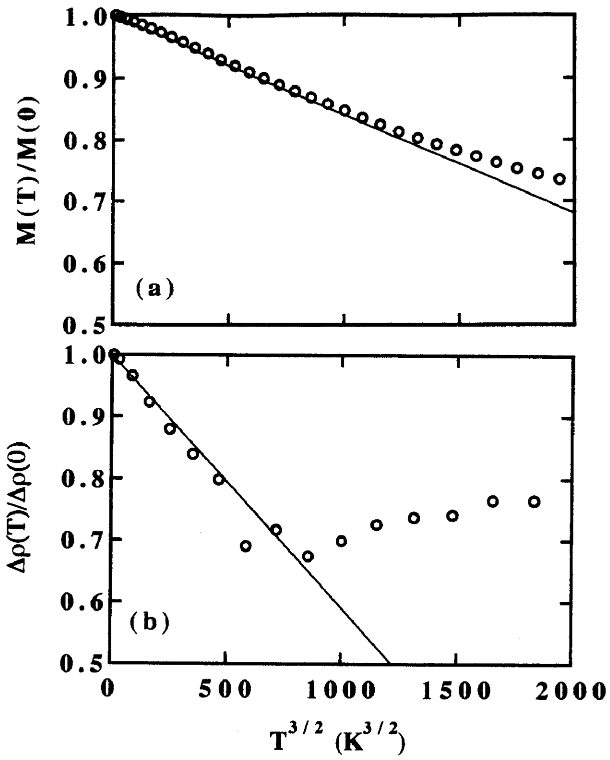

Figure 5 (a)Normalized magnetization, $M(T)/M(0)$, and (b)normalized magnetoresistance, $\Delta\rho(T)/\Delta\rho(0)$, versus $T^{3/2}$ curves for the SP-deposited $Fe_{20}Cu_{80}$ film.

The spin-wave term, B, in eq.(2) and that of the magnetoresistance, B_{MR}, in eq.(3) estimated from the low temperature region ($T < 50$ K) are listed in Table 1. B and B_{MR} are roughly consistent each other. These results suggest that all of spins contribute to the magnetization and the magnetoresistance: the Fe atoms are well mixed with Cu atoms and/or form very small clusters which nearly all of the Fe atoms are located at the surface. Here, B and B_{MR} are, furthermore, considerably larger than the value of 3.3×10^{-6} $K^{2/3}$ for bulk Fe[15]. It is known that the spin-wave constant of surface magnetization is twice as large as that of bulk one[16], because the spin waves become standing waves at the surface, and, the surface layer becomes an antinode since the surface represents a free end. For spin waves of any wavelength, the reduction of magnetization by T at the surface is twice as large as that in the bulk. However, the observed B value is two orders larger than the bulk value. G. Xiao *et al.* also reported large B values for Fe/SiO_2 granular films and pointed out a possibility of the finite size effect[15]: an spin-wave spectrum can be substantially altered from the bulk one, causing a softening of the spin waves.

Table 1. The coefficients of spin-wave term in eqs. (2) and (3), B and B_{MR}, for SP- and CB-deposited Fe_xCu_{100-x} films estimated from the low-temperature part ($T < 50$ K) of $\Delta M(T)/M(0)$ and $\Delta\rho(T)/\Delta\rho(0)$ versus $T^{3/2}$ plots in Figs. 5 and 8, respectively.

	B	B_{MR}
SP-deposited : x=20	1.6×10^{-4}	2.0×10^{-4}
CB-deposited : 11	5.0×10^{-4}	4.6×10^{-4}
22	2.7×10^{-4}	4.2×10^{-4}
35	0.7×10^{-4}	2.5×10^{-4}

3.2 CB-deposited granular Fe/Cu films

Figure 6(a) shows $\Delta\rho(H)/\rho(0)$ versus H curves at different temperatures for the CB-deposited $Fe_{22}Cu_{78}$ film. $\Delta\rho(H)/\rho(0)$ at 300 K shows a linear relationship with H and a very small value in the field of 50 kOe compared with that for the SP-deposited film shown in Fig. 1(a). As temperature goes down to 4.5 K, its absolute value exhibits monotonic increase up to 4 % at 50 kOe. Figure 6(b) shows temperature dependence of resistivity, $\rho(T)$, measured in the field of $H = 0$ and 50 kOe. $\rho(T)$ at $H = 0$ is larger than that at 50 kOe owing to the electron scattering at the interfaces between Fe clusters and Cu matrices. They decrease with decreasing temperature and have a minimum at around 15 K.

Figure 7 shows $\Delta\rho$ versus T plots for the CB-deposited Fe_xCu_{100-x} films with x = 11, 22 and 35. For x=11, $\Delta\rho$ is strongly enhanced at low temperatures, being attributable to the small fcc Fe clusters showing the spin-glass behavior[11]. For x = 22, on the other hand, $\Delta\rho$ is smaller than the other samples because of the antiferromagnetic large fcc Fe clusters, but the enhancement of $\Delta\rho$ at low temperatures is still observable. For x = 35, however, $\Delta\rho$ increases gradually with decreasing temperature because the Fe clusters are linked each other and form the larger bcc Fe clusters showing the ferromagnetic behavior.

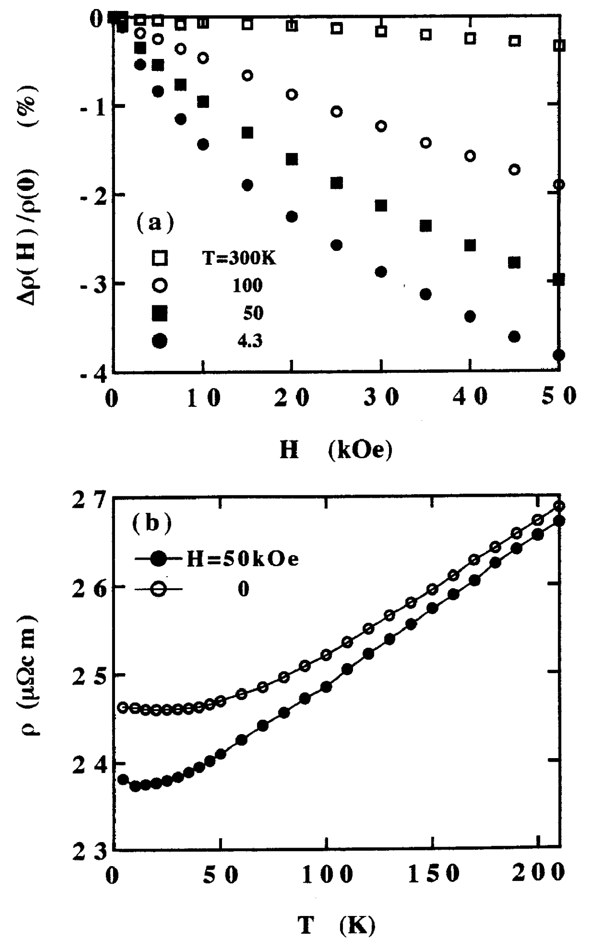

Figure 6 (a)Magnetoresistance ratio, $\Delta\rho/\rho(0)$, as a function of an applied magnetic field, H, and (b) temperature dependence of resistivity in $H = 0$ and 50 kOe for the CB-deposited $Fe_{22}Cu_{78}$ film.

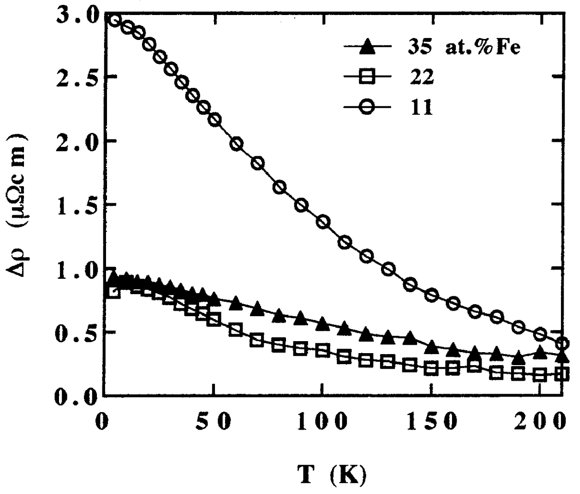

Figure 7 Temperature dependence of $\Delta\rho = \rho(0)-\rho(50$ kOe) for the CB-deposited Fe_xCu_{100-x} films.

Figures 8(a) and (b) display $M(T)/M(0)$ and $\Delta\rho(T)/\Delta\rho(0)$ versus $T^{3/2}$ curves, respectively. As given in Table 1, the coefficient of the spin-wave term in eq.(2), B, and that in eq.(3), B_{MR}, are estimated from the low temperature region ($T < 50$ K). For x = 11, the B value is nearly the same as the B_{MR} value. This result suggests that the Fe clusters are very small and nearly all of the Fe atoms are located at the interface. For x = 22 and 35, on the other hand, B is smaller than B_{MR}, suggesting the formation of the larger Fe clusters in the Cu matrices.

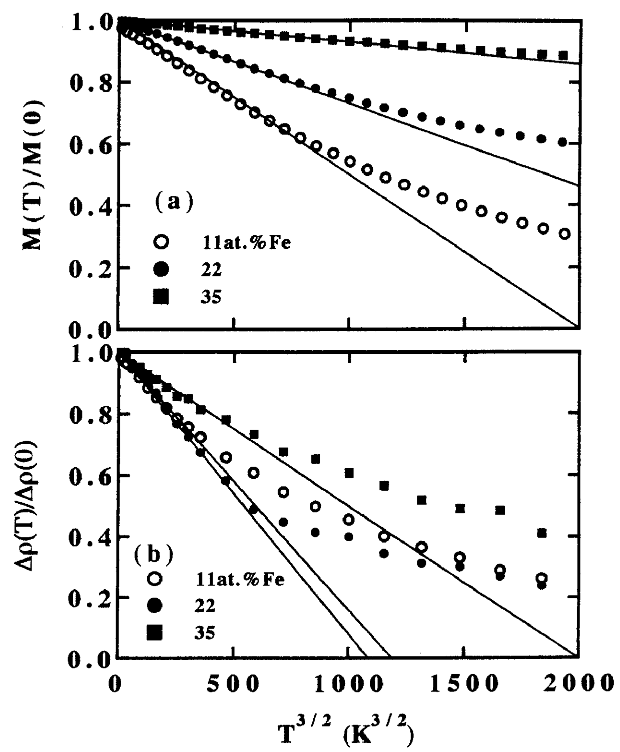

Figure 8 (a)Normalized magnetization, $M(T)/M(0)$, and (b)normalized magnetoresistance, $\Delta\rho(T)/\Delta\rho(0)$, versus $T^{3/2}$ curves for the CB-deposited Fe_xCu_{100-x} films.

4. Conclusion

$\Delta\rho$ for the SP-deposited $Fe_{20}Cu_{80}$ film shows a maximum at around the Curie point (T_C = 150 K) and increases rapidly below 100 K. The enhanced $\Delta\rho$ below the spin-freezing temperature is ascribed to a larger effective spin. With increasing temperature, an electron-magnon scattering gradually increases and weakens above the T_C. For the CB-deposited granular films with low Fe content, $\Delta\rho$ is also enhanced at low temperatures being attributable to the spin-glass-like small fcc Fe clusters. The coefficient of the initial $T^{3/2}$ dependence of $\Delta\rho(T)/\Delta\rho(0)$, B_{MR}, is the same as that of $M(T)/M(0)$, B, for the SP-deposited $Fe_{20}Cu_{80}$ film because, for randomly distributed Fe atoms, this Fe content is near the percolation threshold in the fcc lattice, x_C = 16 - 19, and nearly all of the Fe atoms contribute to the *MR*. For the CB-deposited Fe_xCu_{100-x} film, $B_{MR} = B$ at x = 11, indicating that the Fe clusters are also very small and nearly all of the Fe atoms are located at the interface. On the other hand, for x = 22 and 35, the coefficients are inconsistent each other because the larger Fe clusters are formed in the Cu matrices.

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