Effect of magnetic impurities on electronic transport of heterogeneous ferromagnets

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Electronic transport in granular Co-Ag films is studied between subhelium and room temperatures in fields up to 16 T. A number of unusual properties are found including a strong variation of magnetoresistance at low temperatures and sublinear ($d^2R/dT^2<0$) temperature dependence of high field resistance above 50 K. We show that paramagnetic impurities or small magnetic clusters embedded in the normal-metal matrix influence significantly the electronic transport in heterogeneous ferromagnets. The effect is strongly enhanced as compared with paramagnetic impurities in homogeneous normal metals, and is comparable and even superior over the phonon scattering up to room temperatures. This mechanism is argued to be responsible for the peculiar properties of resistance and magnetoresistance of granular ferromagnets. [S0163-1829(99)04538-5]

I. INTRODUCTION

Discovery of the giant magnetoresistance (GMR) effect in magnetic multilayers[1] and heterogeneous alloys[2,3] triggered numerous studies on a variety of systems (for recent reviews see Refs. 4 and 5). It is now widely believed that the effect is mainly due to the elastic spin-dependent scattering of conduction electrons at the interfaces between magnetic and nonmagnetic regions and in the bulk of the ferromagnetic regions. The applied magnetic field changes the magnetic configuration of the system, e.g., aligns the magnetic moments of different granules in heterogeneous alloys, and, hence, changes (decreases) the resistance.

One of a few subjects left to be clarified is the temperature dependence of the GMR. Experimentally it was found that the magnetoresistance is strongly suppressed in both multilayer and granular materials with the increase of temperature but the details of this variation are different. In multilayers the magnetoresistance is almost constant at low temperatures ($T<50-60$ K), and decreases by a factor of 2 or 3 towards room temperature.[6–11] This dependence was explained successfully by an enhancement of the spin-independent phonon scattering and the spin-flip scattering by magnons with an increase of temperature.[6,7,9,10,12,13]

In granular materials the variation of magnetoresistance is qualitatively comparable to that of multilayers at high temperatures. At low temperatures however, the magnetoresistance was found to change even stronger than at high temperatures, contrary to the saturation observed in multilayers.[10,14–19] In addition, the resistance $R(H)$, measured in high magnetic fields H, and the magnetoresistance (MR) change at low temperatures almost linearly with temperature, while the zero-field resistance $R(0)$ saturates (up to a shallow minimum, which we will not discuss in this paper) in the low-temperature limit. It was argued first in Ref. 19 that the temperature dependence of $R(H)$ and MR cannot be explained by scattering on phonons and/or magnons. Both these scattering mechanisms lead to a saturation of resistivity at low temperature in zero as well as in high magnetic fields. Moreover, the magnons freeze out at low temperatures in high fields. With a reduction of temperature the contribution of phonons to the resistance decreases faster than that of magnons. One would, therefore, expect that the resistance in high fields will saturate at least not slower than in zero field, in a full contradiction with the experimental results.

In an attempt to solve this puzzle, we extended our measurements[19,20] of granular Co-Ag films to higher temperatures. The measurements where performed at magnetic fields up to 16 T, which would guarantee an alignment of granular magnetic moments up to room temperature. The results presented below reveal unexpected details of the temperature dependence of resistance. The high-field resistance $R(H)$ of as-grown samples with Co volume concentration below approximately 30% is *sublinear* at temperatures above 50 K. It is unnecessary to note that the sublinearity in metals is a very unusual feature.

We show in this paper that all peculiarities mentioned above can be explained consistently if electron scattering on paramagnetic impurities or/and small paramagnetic clusters embedded in the nonmagnetic matrix[14,16] is taken into account. We demonstrate that in heterogeneous magnetically ordered ferromagnets where the macroscopic resistance of spin-up electrons differs strongly from that of the spin-down ones, the effect of such scattering is strongly enhanced in comparison with the spin-dependent scattering on paramagnetic impurities in nonmagnetic metals. While in normal metals the contribution of the spin-dependent impurity scattering to the resistivity is of second order in the (small) parameter J/V (J is the s-d exchange interaction, V is the spin-independent impurity potential), in ordered heterogeneous ferromagnetic materials this contribution is of the first order in J/V. It appears that this scattering governs the temperature dependence of $R(H)$ and MR at low temperatures and is comparable or even dominating the phonon contribution at temperatures as high as room temperature.

II. THEORETICAL MODEL

Granular films are complex systems with an electron mean free path of the order of the inhomogeneity scale. It has been argued in Refs. 4,21,22 that the magnetic granular systems are self-averaging. The global resistivity is proportional to the average scattering sampled in each spin channel by the current line. Therefore, the formalism of the conductivity in

granular films is analogous to that of the current perpendicular to plane geometry in multilayers. The magnetoresistance in this case has been successfully described[23] by the two-current model[24] with volume and interface resistances in a series for each spin orientation. It was shown in Ref. 23 that the two-current model without spin flip holds if the electron mean free path and the thickness of magnetic and nonmagnetic layers are shorter than the spin-diffusion length. Assuming that the spin-diffusion length is the largest length scale also in granular systems, we use the two-current model to calculate the resistance and MR in these systems. As argued in the Introduction, in addition to the interface scattering and bulk scattering on phonons and spin waves, we consider also scattering on magnetic impurities and small magnetic clusters within the nonmagnetic matrix.

Scattering of an electron with spin **s** on a magnetic atom in a cluster with a total spin **S** can be described by the interaction energy of the form:

$$U=V(\mathbf{r})+\frac{2J(\mathbf{r})}{S}\mathbf{s}\cdot\mathbf{S}. \tag{1}$$

Here V is the spin-independent scattering potential and the second term is the s-d exchange interaction normalized to spin one. The conductivity of electrons with spin-up ($\sigma_\uparrow$) and spin-down ($\sigma_\downarrow$) scattered on randomly distributed impurities via interaction energy (1) is given by[25]

$$\sigma_{\uparrow,\downarrow}=\sigma_0\left\{1\pm 2\frac{J}{SV}\langle S_z\rangle+\left(\frac{J}{SV}\right)^2\left[4\langle S_z\rangle^2-S(S+1)+\langle S_z\rangle\coth\frac{E_H}{kT}-\langle S_z\rangle\frac{E_H}{kT\sinh^2(E_H/kT)}\right]\right\}+O((J/V)^3). \tag{2}$$

Here $E_H=g\mu_B H/2$, with g being the magnetic scatter g factor, σ_0 is the conductivity related to the scattering potential V only and is inversely proportional to the clusters volume concentration. $J=J(k_F)$ and $V=V(k_F)$, where k_F is the Fermi wave vector. The average spin component along the magnetic field $\langle S_z\rangle$ is given by

$$\langle S_z\rangle=\left(S+\frac{1}{2}\right)\coth\left((2S+1)\frac{E_H}{kT}\right)-\frac{1}{2}\coth\frac{E_H}{kT}. \tag{3}$$

The last term in Eq. (2) originates from the inelastic scattering of electrons. Generally speaking, the inelastic scattering gives also a spin-mixing term. This term, however, vanishes for the usual model of isotropic scattering at the Fermi surface.[26] The total conductivity of an alloy in magnetic field is equal to $\sigma_\uparrow+\sigma_\downarrow$. Therefore, the terms linear in (J/V) cancel out, and only terms of order $(J/V)^2$ contribute to the total resistance.

Consider now the effect of spin-dependent impurity scattering on the resistance of granular ferromagnet-normal metal mixtures. We assume that the applied magnetic field is high enough to align the magnetic moments of ferromagnetic grains in the entire temperature range in which we are interested. Within the framework of the two-current model, the total resistance in each channel $(\uparrow,\downarrow)$ is a sum of the corresponding intragranular, intramatrix, and interface spin-dependent and spin-independent resistances. It follows from Eq. (2), that the resistance $r_{\uparrow,\downarrow}(T)$ of the matrix due to the magnetic impurity scattering can be written as

$$r_{\uparrow,\downarrow}(T)=r^0_{\uparrow,\downarrow}+r^T_{\uparrow,\downarrow}, \tag{4}$$

where $r^0_{\uparrow,\downarrow}$ is the residual (zero temperature) part of the this resistance,

$$r^0_{\uparrow,\downarrow}=r_0\left(1\pm\frac{J}{V}\right)^2, \tag{5}$$

$r_0=\sigma_0^{-1}$, and $r^T_{\uparrow,\downarrow}$ is the temperature-dependent resistance term given by

$$r^T_{\uparrow,\downarrow}=r_0\frac{J}{VS}\left[\mp 2(S-\langle S_z\rangle)+\frac{J}{VS}\left(S-\langle S_z\rangle\coth\frac{E_H}{kT}+\langle S_z\rangle\frac{E_H}{kT\sinh^2(E_H/kT)}\right)\right]. \tag{6}$$

Note that the second term in this equation is smaller than the first one by a factor J/VS, which is much smaller than J/V if S is large.

The total resistance in each channel is

$$R_{\uparrow,\downarrow}(H,T)=\mathcal{R}_{\uparrow,\downarrow}(T)+\mathcal{R}_N(T)+r^T_{\uparrow,\downarrow}. \tag{7}$$

Here $\mathcal{R}_{\uparrow,\downarrow}$ is the spin-dependent resistance due to scattering on the interfaces and within ferromagnetic grains. The last mechanism in granular films is much less important than the first one.[22,27] We also include into $\mathcal{R}_{\uparrow,\downarrow}$ the residual resistance $r^0_{\uparrow,\downarrow}$ due to scattering on magnetic impurities within the normal-metal matrix. $\mathcal{R}_N(T)$ includes the resistance due to electron-phonon scattering as well as electron-electron and electron-magnon scattering contributions. We shall demonstrate in the following that in our samples the latter two are less significant than the electron-phonon scattering.

It was shown in Ref. 28 that the spin asymmetry of the Co-Cu interface and of the bulk Co is temperature independent. We, therefore, neglect in what follows the temperature dependence of $\mathcal{R}_{\uparrow,\downarrow}$. The total resistance of the sample is now

$$R(H,T)=\frac{R_\uparrow(H,T)R_\downarrow(H,T)}{R_\uparrow(H,T)+R_\downarrow(H,T)}. \tag{8}$$

Experimentally, $R_{\uparrow,\downarrow}(H,T)$ changes in our samples by about 30% between 300 and 1.5 K, i.e., the temperature-dependent part of the resistance is smaller than the residual one. We can, therefore, expand Eqs. (7) and (8) in powers of $\mathcal{R}_N(T)$ and $r^T_{\uparrow,\downarrow}$. This gives

$$R(H,T)=\frac{\mathcal{R}_\uparrow\mathcal{R}_\downarrow}{\mathcal{R}_\uparrow+\mathcal{R}_\downarrow}+\frac{\mathcal{R}_\uparrow^2+\mathcal{R}_\downarrow^2}{(\mathcal{R}_\uparrow+\mathcal{R}_\downarrow)^2}\left[\mathcal{R}_N(T)+r_0\left(\frac{J}{VS}\right)^2\times\left(S-\langle S_z\rangle\coth\frac{E_H}{kT}+\langle S_z\rangle\frac{E_H}{kT\sinh^2(E_H/kT)}\right)\right]+r_0\left(\frac{2J}{VS}\right)\frac{\mathcal{R}_\uparrow-\mathcal{R}_\downarrow}{\mathcal{R}_\uparrow+\mathcal{R}_\downarrow}(S-\langle S_z\rangle). \tag{9}$$

Hence, the leading scattering contribution of magnetic impurities embedded in the nonmagnetic matrix of a granular fer-

romagnetic system is *linear* in the small parameter J/V. This is different from the standard case of nonmagnetic metals with dissolved magnetic impurities, referred earlier. The nonlinear term is of order $(J/VS^{1/2})^2$, and can be neglected.

For $S \gg 1$ the factor $S-\langle S_z\rangle$ can be approximated as

$$S-\langle S_z\rangle \approx \exp\left(-\frac{2E_H}{kT}\right), \quad \text{if } kT \ll E_H,$$

$$S-\langle S_z\rangle \approx \frac{kT}{2E_H}, \quad \text{if } E_H \ll kT \ll 2SE_H,$$

$$S-\langle S_z\rangle \approx S\left[1-\frac{2(S+1)E_H}{3kT}\right], \quad \text{if } kT \gg E_H. \qquad (10)$$

For $\mu_0 H = 16$ T and $g=2$ the energy E_H is equivalent to 10.75 K. Therefore, one should expect the following temperature dependence of the cluster resistance:

(1) At $T<10$ K, the temperature-dependent part of the clusters resistance decreases exponentially with the decrease of the temperature. One should, however, take into account that spins from different clusters and/or grains are coupled via Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange. This random coupling leads to a spread of the E_H values. Therefore, depending on the distribution of E_H, the clusters term decreases at low temperatures much slower than exponentially.

(2) At $T>10$ K, the clusters term is linear in T over a wide temperature range.

(3) At elevated temperatures the clusters term is sublinear and saturates at $kT>2SE_H$.

The phonon resistance increases linearly at $T>\Theta$, where Θ is the Debye temperature. Thus, at sufficiently high temperatures, the total resistance in high magnetic fields can be sublinear in T ($d\ln R/d\ln T<1$ or $d^2R/dT^2<0$).

The model is not restricted to granular metals only. In any system with large asymmetry in the spin-up and spin-down scattering, the effect of magnetic impurities on the resistance is enhanced. The effect should be present, e.g., in layered structures with magnetic impurities or loose spins embedded within the nonmagnetic layers.[29,30]

III. EXPERIMENTAL RESULTS

Granular films of Co-Ag were prepared by codeposition of Co and Ag on room-temperature glass or sapphire substrates using two independent electron-beam sources. It is well evidenced that Co and Ag segregate essentially completely under the proper fabrication conditions.[31] This segregation occurs because the surface free energy of Co (2.71 J/m^2) is more than twice that of Ag (1.30 J/m^2),[32] causing cobalt to coagulate. There is a very poor lattice match since fcc Ag has a lattice parameter (4.086) that is about 15% larger than that of fcc Co (3.544). The heat of formation between Co and Ag is positive (+26 kJ/gr atom),[33] so there is no tendency for formation of the compound CoAg or for alloying. It was, therefore, found that evaporated or sputtered films of Co-Ag condensated at high substrate temperatures ($T_{subs}>400$ K) are completely segregated to Co grains embedded in a silver matrix. However, when films are deposited on a room-temperature substrate, adatoms might lack sufficient mobility to reach complete well-ordered segregation and form an irregular structure of small crystallites observed by the high resolution transmission microscopy.[31,34]

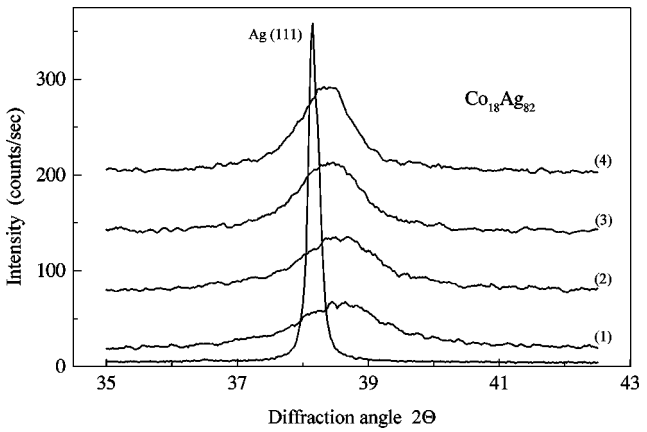

FIG. 1. X-ray diffraction of four simultaneously deposited $Co_{18}-Ag_{82}$ samples. Sample (1) is as-deposited, the rest are annealed as following: sample (2) at 200 °C for 30 min, sample (3) at 300 °C for 30 min, and sample (4) at 400 °C for 180 min. The diffraction of a pure Ag film is shown as a reference.

The x-ray-diffraction patterns for samples with low Co concentration deposited on room-temperature substrates are characterized by the fcc peaks of silver shifted towards higher reflection angles and their intensities suppressed by up to two orders of magnitude as compared to pure silver films. The contraction of the silver lattice must be associated with the partial substitution of Ag atoms by smaller Co atoms or small clusters of Co atoms.[14,16,35] The results of x-ray diffraction are supported by magnetization and susceptibility measurements[16] that indicate the presence of small clusters built of a few Co atoms, coexisting with larger few-nanometer size grains.

The presence of small Co clusters in silver matrix of our samples is confirmed by the x-ray diffraction (Fig. 1) of four simultaneously deposited $Co_{18}-Ag_{82}$ samples (volume concentration of Co is 18% and Ag is 82%) that passed different post-deposition thermal treatment. Sample (1) was left as-deposited, sample (2) was annealed at 200 °C for 30 min, sample (3) was annealed at 300 °C for 30 min and sample (4) was annealed at 300 °C for 180 min. As expected for films deposited on a room-temperature substrate, diffraction peaks of Ag are strongly suppressed and shifted toward higher diffraction angles. Annealed samples show systematically sharper diffraction peaks moving toward undisturbed Ag peak angle. Sharpening of the peaks might indicate an increase of Ag grains, whereas the shift in their location indicates a systematic cleaning of silver matrix from small Co inclusions. Concentration of Co impurities frozen in Ag matrix can be roughly estimated from the position of the peaks. The estimated values are 8, 7.6, 5.5, and 4% for samples (1)–(4), respectively. All other samples discussed in this paper were deposited at similar conditions and, most probably, contain significant amount of cobalt inclusions within silver matrix.

We plot in Fig. 2(a) the resistance of five as-deposited samples with Co volume concentrations 18, 23, 34, 41, and 53% (samples A–E, respectively) measured as a function of

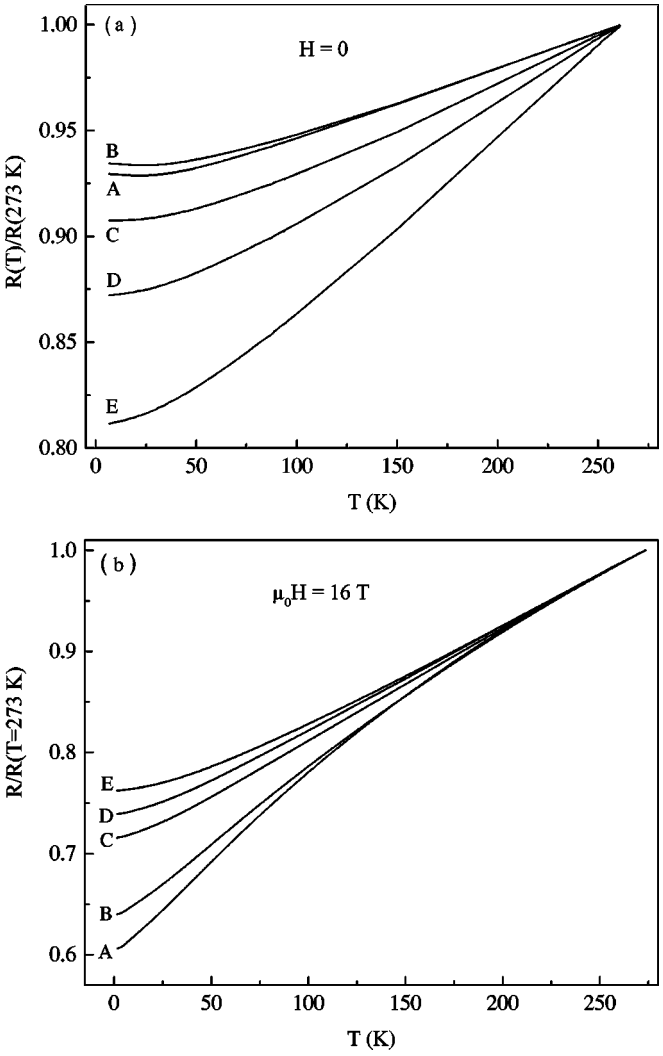

FIG. 2. Resistance of five as-deposited films with Co volume concentrations 18% (A), 23% (B), 34% (C), 41% (D), and 53% (E) divided by their room-temperature values as a function of temperature measured at zero (a) and 16 T magnetic field (b).

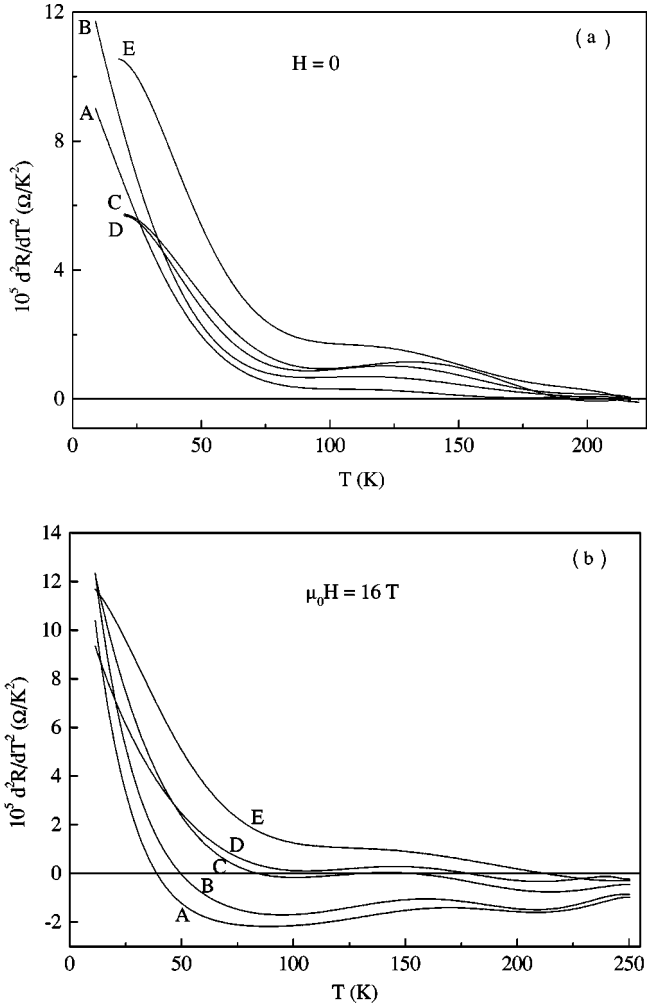

FIG. 3. The second temperature derivative of resistance (d^2R/dT^2) as a function of temperature for the as-deposited samples A–E at zero (a) and 16 T field (b). The derivative is *negative* for samples A and B at high field.

temperature at zero magnetic field. The resistance saturates in the low-temperature limit (we do not discuss here the shallow minimum observed in some samples at $T<20$ K). As will be shown in the next section the resistance of Ag-rich samples follows at low temperatures the dependence

$$R(T)=R(0)+aT^{\gamma}, \tag{11}$$

with $\gamma=3$. Note that the same dependence has been found in *nonmagnetic* $Cu_{20}Ag_{80}$ granular films.[19]

Figure 2(b) presents the resistance of the same samples measured at 16 T applied field. The effective moment of a single Co grain obtained from the magnetization measurements[19] is about 4×10^{-18} emu, i.e., the magnetic energy of a grain in field 16 T is about 0.5 eV. The granular moments are well aligned by this field at temperatures below 300 K.

The overall behavior of the low Co concentration samples is qualitatively different at high fields: there is no resistance saturation at low temperatures, and a ''belly'' is observed at about 100 K. The behavior is better illustrated in Figs. 3(a) and 3(b), where the second derivatives d^2R/dT^2 of the zero field and high field $R(T)$ curves are plotted as a function of temperature. The zero field curves are ''normal'' in the sense that the d^2R/dT^2 is positive at all temperatures. The high-field curves are ''abnormal'' for the low Co concentration samples: d^2R/dT^2 is *negative* at T higher than 50 K, and approaches zero only at about 300 K, i.e., only at high temperatures $R(H,T)-R(H,0)$ starts to increase almost linearly with temperature, as one should expect for phonon scattering.

The temperature dependence of the resistivity observed in high fields differs qualitatively from what is expected from the traditionally discussed mechanisms. At low temperatures the resistivity can usually be well fitted by a combination of the power-law temperature dependences $\Sigma_i a_i T^{\gamma_i}$, where different terms describe different temperature-dependent scattering mechanisms. Scattering on phonons is spin independent with $\gamma=5$ in the clean limit, and follows $\gamma=3$ in dirty, disordered or granular metallic mixtures. Electron-electron scattering with $\gamma=2$ is effective in bulk ferromagnets and ferromagnet rich mixtures. The lowest power-law exponent is $\gamma=3/2$ corresponding to the scattering by magnons in disordered ferromagnets. Application of a high field is expected to suppress the possible magnon-dominated contribution and

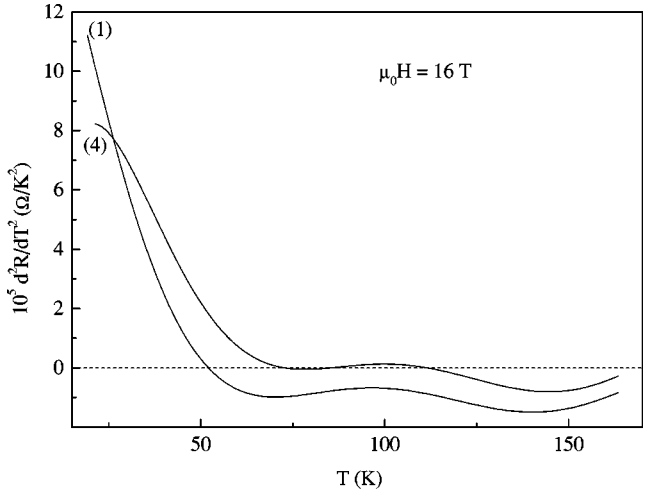

FIG. 4. (d^2R/dT^2) as a function of temperature for the as-deposited sample (1) and the most annealed sample (4) at 16 T field.

leaves γ to be not lower than two. Regardless of the dominating mechanisms all these dependences predict the power-law saturation of resistivity in the low-temperature limit. However, the resistivity of our films in high fields follows Eq. (11) with γ close to 1 for Ag-rich samples.[19]

At elevated temperatures resistivity of metals is usually dominated by the phonon scattering and tends to a linear variation when temperature approaches the Debye temperature. Addition of the magnon or electron-electron scattering can modify the general form of the resistivity-temperature curve, in particular at low temperatures. However, for any common metallic system, including granular mixtures, the resistivity-temperature curve is concave in any temperature range, i.e., the second derivative d^2R/dT^2 is positive. In our samples this derivative in high fields is positive at low temperatures only and is negative in a wide range of elevated temperatures.

Annealing suppresses strongly the ''belly'' effect, as demonstrated in Fig. 4. The belly is pronounced much more in the as-deposited sample than in the most annealed sample (4) in the measured temperature range.

We show in the next section that all these peculiar properties of the high-field resistance can be understood if scattering on both phonons and small magnetic clusters embedded in the nonmagnetic metal are taken into account.

IV. ANALYSIS

The credibility of the model has been tested by the following analysis. We start from the zero-field resistance

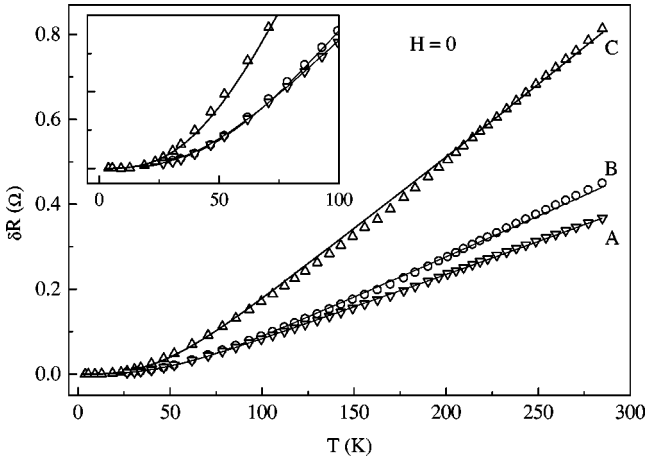

FIG. 5. The measured temperature-dependent part of the zero-field resistance $\delta R = R(T) - R(0)$ of samples A, B, and C and the corresponding fitting curves calculated from Eq. (13).

$R(T,0)$. In the framework of the two-current model $R(T,0)$ is given by

$$R(T,0) = \frac{1}{4}(\mathcal{R}_\uparrow + \mathcal{R}_\downarrow) + \frac{\mathcal{R}_N}{2}. \tag{12}$$

Strictly speaking, the resistances $\mathcal{R}_{\uparrow,\downarrow}$ are different from that in high field, since all the impurity spin scattering is now temperature independent. We neglect this difference since the contribution of the impurity scattering to the residual resistance is supposed to be small. For samples with relatively low cobalt concentration we try to fit the experimental data by the phonon scattering only. As has been shown by x-ray diffraction, the silver matrix is strongly disturbed by cobalt inclusions, and it is reasonable to assume that the noncoherent electron-phonon scattering dominates $\mathcal{R}_N$. The temperature-dependent zero-field scattering can be written as

$$\delta R(T,0) = R(T,0) - R(0,0) = a_1(0)\left(\frac{T}{\Theta}\right)^3 \int_0^{\Theta/T} \frac{x^2 dx}{e^x - 1}. \tag{13}$$

The coefficient $a_1(0)$ and the Debye temperature Θ are the fitting parameters. Evidently, $a_1(0)$ can be excluded from the fitting procedure if the resistance normalized by a chosen temperature value is fitted. We prefer however, to extract the coefficient $a_1(0)$ and its high-field counterpart $a_1(H)$, which will allow us to compare their ratio to that calculated from the two-current model.

We plot in Fig. 5 the resistance of samples A, B, and C measured as a function of temperature at zero magnetic field and the corresponding fitting curves calculated from Eq.

TABLE I. Fitting parameters for samples A, B, and C.

Sample	Co(%)	$\frac{R(0,0)-R(H,0)}{R(0,0)}$	Θ (K)	S	Δ (K)	$[a_1(H)/a_1(0)]_{\text{calc}}$	$[a_1(H)/a_1(0)]_{\text{fit}}$
A	18	5.13	208	13	7.5	1.45	1.5
B	23	5.12	250	7.5	6.3	1.45	1.9
C	34	4.41	220	3.9	4.1	1.40	1.7

(13). The agreement is very good in the entire temperature range. The fitted Debye temperatures (see Table I) are reasonably close to the value $\Theta = 203$ K quoted for pure Ag.[36] The quality of the fit implies that in these samples with relatively low cobalt content the phonon scattering in silver matrix dominates the temperature dependence of the zero-field resistance. Fitting by Eq. (13) is significantly poorer for samples with higher cobalt concentration. Additional scattering mechanisms, like the coherent phonon scattering, electron-electron and electron-magnon scattering have to be considered to fit the resistance of the Co-rich samples. In order to reduce the number of mechanisms in discussion and the number of fitting parameters we shall restrict our future analysis to three samples: A, B, and C.

Following the preceding discussion, the temperature-dependent part of the high-field resistance can be written as

$$\begin{aligned}\delta R(T,H) &= R(T,H) - R(0,H) \\ &= a_1(H)\left(\frac{T}{\Theta}\right)^3 \int_0^{\Theta/T} \frac{x^2 dx}{e^x - 1} \\ &\quad + a_2\left\{ S - \int_{-\infty}^{\infty} d\varepsilon\, w(\varepsilon)\left[\left(S+\frac{1}{2}\right)\right.\right. \\ &\quad \left.\left. \times \coth\frac{(2S+1)(E_H+\varepsilon)}{kT} - \frac{1}{2}\coth\frac{(E_H+\varepsilon)}{kT}\right]\right\}. \end{aligned} \tag{14}$$

Here, $w(\varepsilon)$ is the distribution function of the random molecular fields generated by the RKKY interaction. We choose in the following a Lorenzian distribution with a width Δ, although the correct distribution $w(\varepsilon)$ is not known.[25] With the Debye temperature found from fitting the zero-field resistance, we are left with four fitting parameters $a_1(H), a_2, S$, and Δ.

We show in Fig. 6 the results of the fitting procedure of the high-field data calculated by Eq. (14). The calculated and fitting parameters are presented in Table I. Undoubtedly, the calculated curves fit perfectly the experimental data over more than two decades of temperature range. We found that both S and Δ are sensitive to the distribution function and to the details of the fitting procedure. However, in any case, Δ is lower than 10 K and an effective impurity, defined via the calculated S value, contains between 3 and 10 Co atoms in agreement with Refs. 14,16. At temperature above 20 K, where the RKKY interaction can be neglected ($\Delta = 0$), an equally good fit can be obtained with three parameters $a_1(H)$, a_2, and S only. Moreover, the normalized temperature-dependent part of the resistance $\delta R(H,T)/\delta R(H,270K)$ at $T > 20$ K is fitted by just two parameters: $a_1(H)/a_2$ and S (see Fig. 7).

The ratio α defined as $\alpha = \mathcal{R}_\uparrow/\mathcal{R}_\downarrow$ can be found from the experimental data via the following expression: $[R(0,0) - R(H,0)]/R(0,0) = (1-\alpha)^2/(1+\alpha)^2$. Given α, one can calculate the theoretical ratio $a_1(H)/a_1(0)$, which follows from the two-current model, Eqs. (9) and (12), and compare it with the ratio obtained from the fitting. Both fitted and calculated values for the three samples A–C are shown in Table I. Considering the basic simplification of the two-current model, the agreement is reasonable and confirms the reliability of our analysis.

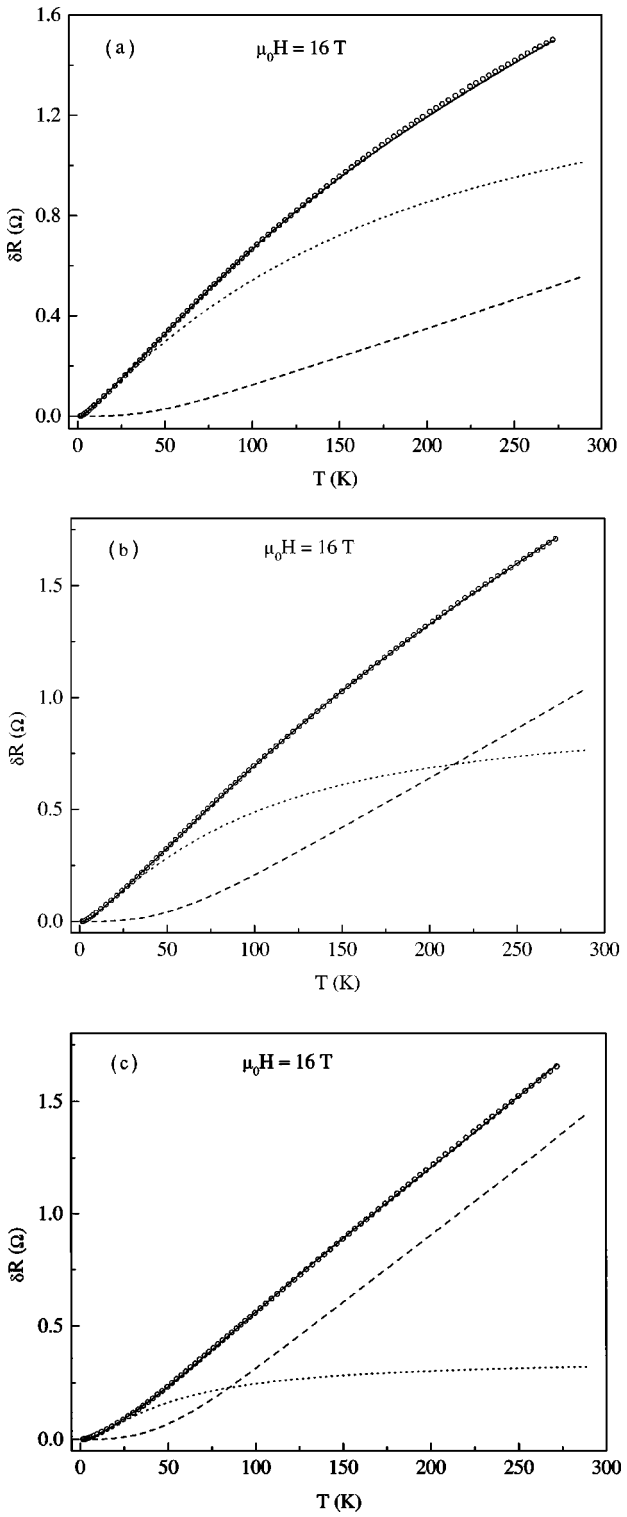

FIG. 6. The temperature-dependent part of the high-field resistance $\delta R = R(T) - R(0)$ as a function of temperature for samples A–C [(a)–(c), respectively]. Symbols are experimental data, solid lines are calculated according to Eq. (14). Dotted curves are also calculated from Eq. (14) with coefficient $a_1 = 0$ and present the impurity scattering contribution only. Dashed curves are calculated from the same equation with coefficient $a_2 = 0$ and present the phonon scattering contribution only.

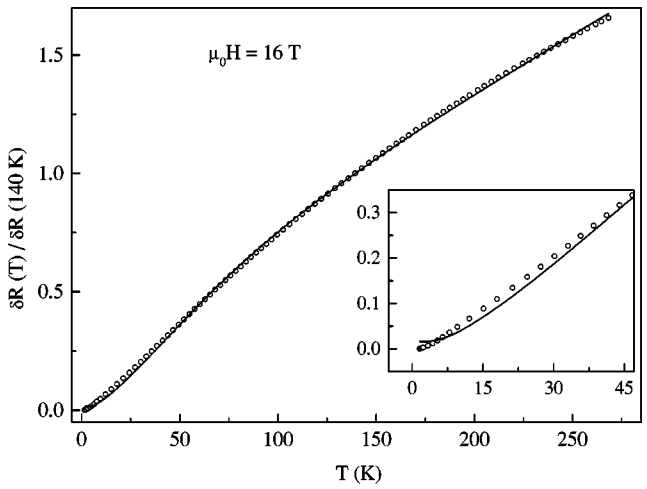

FIG. 7. The normalized temperature-dependent part of the high-field resistance for sample A. Symbols are experimental data. The line is the fit calculated from Eq. (14), with $(\Delta=0)$ and two fitting parameters only: $a_1(H)/a_2$ and S.

It is interesting to estimate quantitatively the effect of the paramagnetic impurity scattering on the temperature-dependent part of resistivity and compare it with the phonon scattering. This is done in Figs. 6(a)–6(c). The dashed and dotted lines are calculated from Eq. (14) for phonon only $(a_2=0)$ and impurity only $[a_1(H)=0]$ terms, respectively. For sample C the temperature variation of resistivity is dominated by the impurity scattering below 80 K and by phonon scattering at higher temperatures, whereas at room temperature the impurity scattering contributes about 15% of the total temperature-dependent value. For sample A the temperature variation of resistivity is dominated by the impurity scattering in the *entire* range from helium to room temperature. At room temperature the impurity contribution is about *twice* that of phonons.

V. APPLICATIONS TO THE GMR EFFECT

Magnetoresistance and its temperature variation depend directly on the resistance at high field and therefore, on the paramagnetic impurity scattering. We show in Fig. 8 the magnetoresistance of samples A and C as a function of temperature. The solid line is the theoretical dependence calculated with parameters given in Table I, whereas the dashed and dotted lines are calculated for the phonon only and impurities only scattering contributions respectively. The temperature dependence of the magnetoresistance of sample A appears to be totally dominated by the impurity scattering in the entire temperature range.

It is natural to wonder how will the magnetoresistance and its temperature variation change if magnetic impurities are removed from the body of granular ferromagnets. Post-deposition annealing is a possible technique to clean the system, as evidenced by the x-ray diffraction. Magnetoresistance of the as-deposited and annealed samples (1)–(4) at 200 and 1.5 K is shown in Fig. 9. The blocking temperature of our films is of order of 30 K.[19] Therefore, superparamagnetic cobalt grains of 3 nm size or larger at 1.5 K should be aligned above the field of order of 0.1 T. This is almost met in the most annealed sample (4). The magnetoresistance of

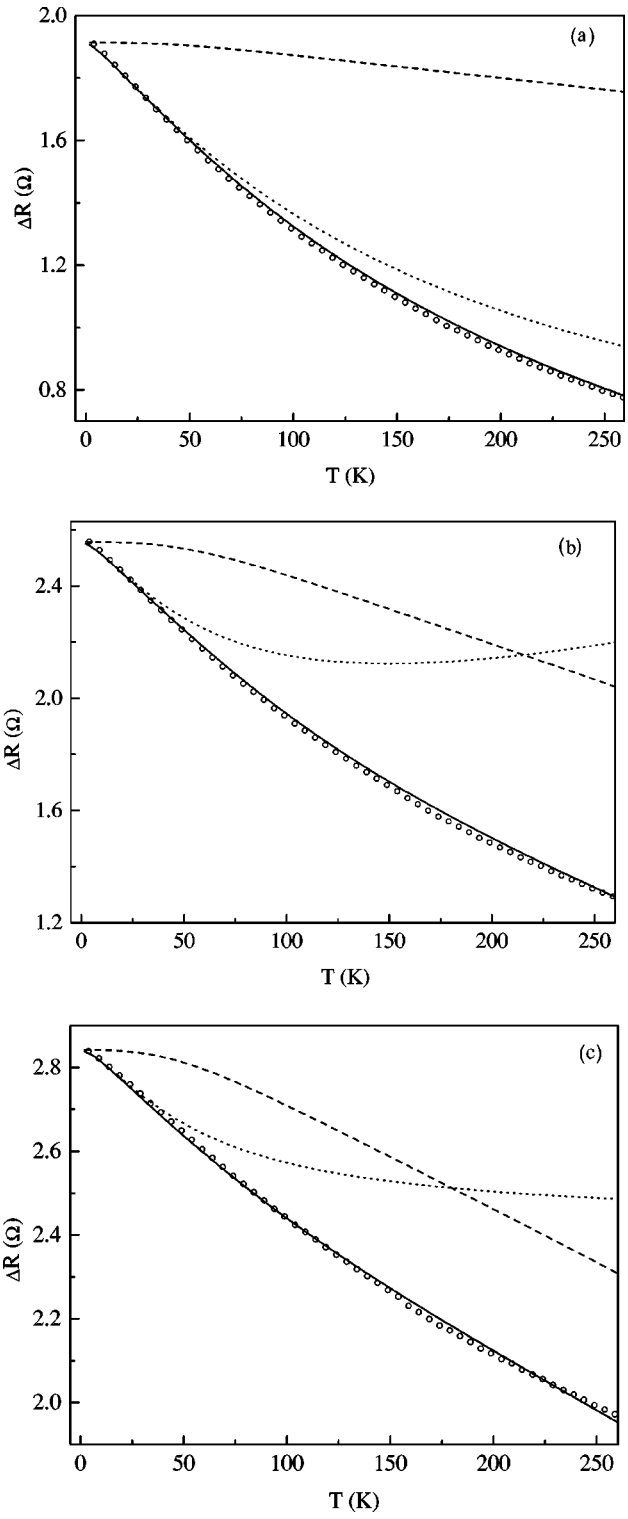

FIG. 8. Magnetoresistance $\Delta R=R(0)-R(16T)$ of samples A–C as a function of temperature [(a)–(c), respectively]. Symbols are experimental data, solid curves are calculated using Eqs. (13), (14). Dotted curves are calculated with coefficient $a_1=0$ and present the impurity scattering contribution only. Dashed curves are calculated with coefficient $a_2=0$ and present the phonon scattering contribution only.

the as-deposited sample (1) is not entirely saturated at 1.5 K even under 16 T field and is very far from saturation at 200 K. This is an explicit demonstration of a significant magnetoresistance contribution of small clusters coupled by the

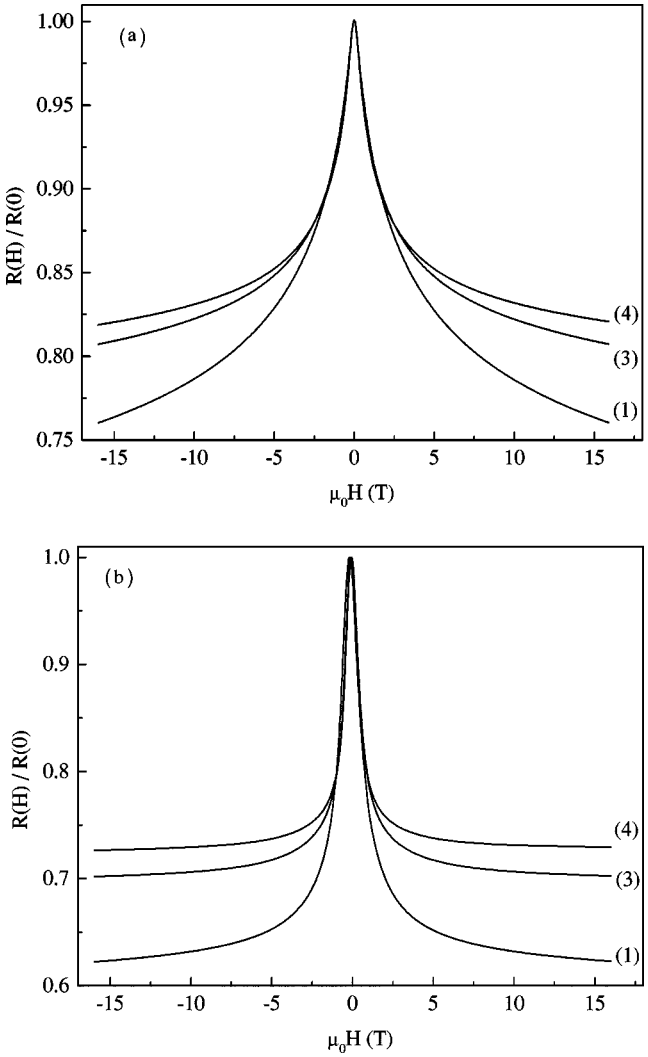

FIG. 9. Resistance of the as-deposited (1) and annealed samples (3), (4) divided by their zero-field values $R(H)/R(0)$ as a function of applied magnetic field at 200 K (a), and 1.5 K (b).

RKKY exchange in a spin-glass-like state.

Reduction of GMR with annealing is a well-known effect which is currently explained by an enlargement of grains and reduction of the granular surface-to-volume ratio.[4] It is difficult to separate this effect from cleaning the matrix of the magnetic impurities. The difference between the two mechanisms can be found by comparing the temperature dependence of magnetoresistance of the as-deposited sample (1) and a slightly annealed sample (2) (Fig. 10). Magnetoresistance of sample (1) is the largest at low temperatures, whereas sample (2) is superior at room temperature. The phenomenon is a result of the impurity scattering. Magnetic moments of 3 nm or larger grains are aligned in 16 T field up to temperatures well above 300 K and the spin-dependent scattering on their boundaries is only weakly temperature dependent. On the other hand, small impurities contribute to magnetoresistance only at sufficiently low temperatures when the applied field is high enough to align their moments. At high temperatures thermal fluctuations exceed magnetic energy of the impurities that remain not ordered at the highest fields applied and therefore, not contributing to the magnetoresistance. Reduction of the impurities content by a light annealing improves the normalized magnetoresistance at high temperature but reduces it at low temperatures, consistently with the result shown in Fig. 10.

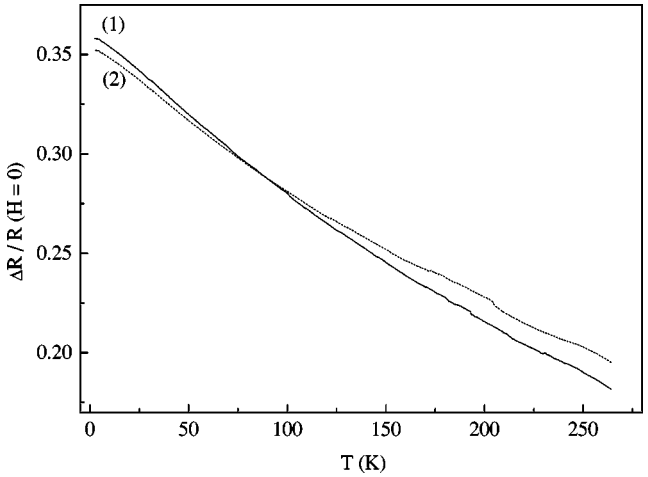

FIG. 10. Normalized magnetoresistance $\Delta R/R(0)$ $[\Delta R = R(0) - R(16T)]$ of samples (1) and (2) as a function of temperature.

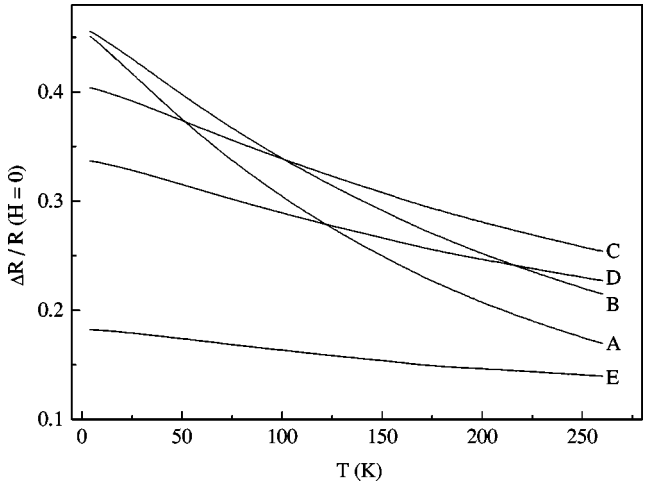

FIG. 11. Normalized magnetoresistance of samples A–E as a function of temperature.

It is tempting to use the same arguments to explain the shift with temperature of the optimum concentration in the series of Co-Ag films. We show in Fig. 11 the normalized magnetoresistance of samples A–E as a function of temperature. At room temperature the largest magnetoresistance is measured in sample (C) with volume cobalt concentration of 34%. At 1.5 K the largest effect is found in samples A and B with 18 and 23 % of cobalt, respectively. The shift of the optimum concentration can be understood assuming that the number of impurities trapped in Ag matrix is larger in samples with low Co concentration.

VI. SUMMARY

We present experimental and theoretical study of electronic transport in granular Co-Ag films that are widely used as a model system for the GMR effect studies. Experiments were performed between room and subhelium temperatures in fields up to 16 T, the field high enough to saturate granular magnetic moments in the entire temperature range. A number of unusual and unexpected effects have been found in

experiments: (1) strong temperature dependence of magnetoresistance, in particular in the low-temperature limit where, following the conventional models, the magnetoresistance is expected to be saturated. (2) Temperature dependence of resistivity of films with relatively low content of cobalt measured under high magnetic field is almost linear at low temperatures and sublinear ($d^2R/dT^2<0$) above 50 K. (3) Optimum concentration (i.e., Co concentration of films demonstrating the highest magnetoresistance value) is temperature dependent and shifts to lower Co concentration at low temperature. We find experimental evidence connecting these phenomena to the presence of small cobalt clusters (built of a few atoms) within silver matrix.

We have developed a theoretical model of the spin-dependent scattering on paramagnetic impurities immersed in a normal metal matrix of a heterogeneous ferromagnet. We have demonstrated that in heterogeneous magnetically ordered ferromagnets where the macroscopic resistance of spin-up electrons differs strongly from that of spin-down ones this scattering is strongly enhanced as compared with the paramagnetic scattering in nonmagnetic metals. The effect of this scattering on the resistivity of heterogeneous ferromagnets is shown to be comparable and even superior over the phonon scattering up to room temperature and can be responsible for all the peculiarities observed in experiments.

We showed also that the temperature dependence of the zero-field resistance in Ag-rich samples can be well described by a simple Eq. (13) with one fitting parameter, the Debye temperature, which appeared to be close to the Debye temperature in pure Ag. This implies that the temperature dependence of the zero-field resistance in these samples is mainly due to phonon scattering in strongly disordered Ag, while the scattering of the electrons on the Co-Ag interfaces causes only the temperature-independent spin asymmetry of the resistance.

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