Eur. Phys. J. B **5**, 23–28 (1998)

THE EUROPEAN
PHYSICAL JOURNAL B

Well-defined Co clusters embedded in an Ag matrix: A model system for the giant magnetoresistance in granular films

S. Rubin, M. Holdenried, and H. Micklitz[a]

II. Physikalisches Institut, Universität zu Köln, Zülpicher Strasse 77, 50937 Köln, Germany

Received: 21 April 1998 / Received in final form: 17 May 1998 / Accepted: 18 May 1998

Abstract. Well-defined granular Co/Ag films have been prepared by the co-deposition of in-beam prepared Co clusters and Ag atoms. In this way we were able to study the giant magnetoresistance (GMR) as a function of mean Co cluster size $\bar{L}$ for a fixed Co cluster volume fraction v_{cl} as well as a function of v_{cl} for a fixed $\bar{L}$. Mean Co cluster size has been varied between $\bar{L} = 3.3$ and 6.9 nm, Co cluster volume fraction between 5 and 43%. The GMR was measured *in-situ* at $T = 4.2$ K in magnetic fields $B \leq 1.2$ T. The analysis of the GMR data obtained from these studies clearly shows that spin-dependent scattering at the Co-cluster/Ag-matrix interface is the only relevant scattering mechanism causing the GMR in our well-defined samples.

PACS. 72.15.Gd Galvanomagnetic and other magnetotransport effects

1 Introduction

A few years after the discovery of the giant magnetoresistance (GMR) in superlattices [1,2] the same effect has been found in granular systems with ferromagnetic single domain particles embedded in a non-magnetic metallic matrix [3,4]. A large number of experimental papers have been published since then, mostly dealing with either new granular systems showing the GMR, or experimental conditions for improving the magnitude of the GMR. The sample preparation method more or less was the same for almost all systems studied so far: both components A and B which are immiscible in the equilibrium phase diagram (*e.g.* Co and Ag) are evaporated or sputtered onto a substrate where they form a homogeneous metastable alloy; upon annealing this metastable alloy dissociates into the equilibrium phases, *i.e.* ultrafine particles (clusters) of component A (*e.g.* Co) embedded in a metallic matrix B (*e.g.* Ag) are formed during annealing. It is, therefore, quite obvious that such a preparation procedure results in not well-defined samples with a rather broad cluster size distribution and even some magnetic atoms A desolved in the non-magnetic matrix B. Furthermore, the average cluster size strongly depends on the mixing ratio of A and B, *i.e.* increasing volume fraction of component A will result in an increasing average cluster size [5]. Thus, this ill-defined microstructure of the granular systems makes a quantitative analysis of the GMR very difficult [6]. For example, one of the still discussed problems is the question to which extent the following scattering mechanisms contribute to the GMR: spin-dependent interface scattering, spin-dependent scattering within the magnetic grain and spin-flip scattering from magnetic impurities. One approach to this problem from the experimental point of view is the preparation of well-defined granular systems without magnetic impurities and systematic studies of the GMR in such systems both as a function of cluster size at a given cluster volume fraction as well as a function of cluster volume fraction at a given cluster size.

We have prepared such well-defined granular samples in the following way: clusters of component A are prepared in-beam with the help of a so-called cluster source and are simultaneously deposited with atoms of component B on a substrate. We should mention at this point that recently granular samples have been prepared by others in a very similar way [7–11]. In none of these works, however, systematic studies of the GMR have been reported. Dupuis *et al.* [10], for example, measured the cluster concentration dependence of the GMR for Co clusters embedded in an Ag matrix only for one cluster size. In addition, no quantitative analysis of the GMR has been given in this paper.

After a short description of the experimental set-up and sample preparation (a more detailed description already has been given elsewhere [12]) we will discuss the experimental results on the GMR obtained on these samples. We will show that these results can be well described in a simple model in which the interface scattering between the magnetic Co clusters and the non-magnetic Ag matrix is the only scattering process relevant for the GMR in this granular system.

---

[a] e-mail: `micklitz@colorix.ph2.uni-koeln.de`

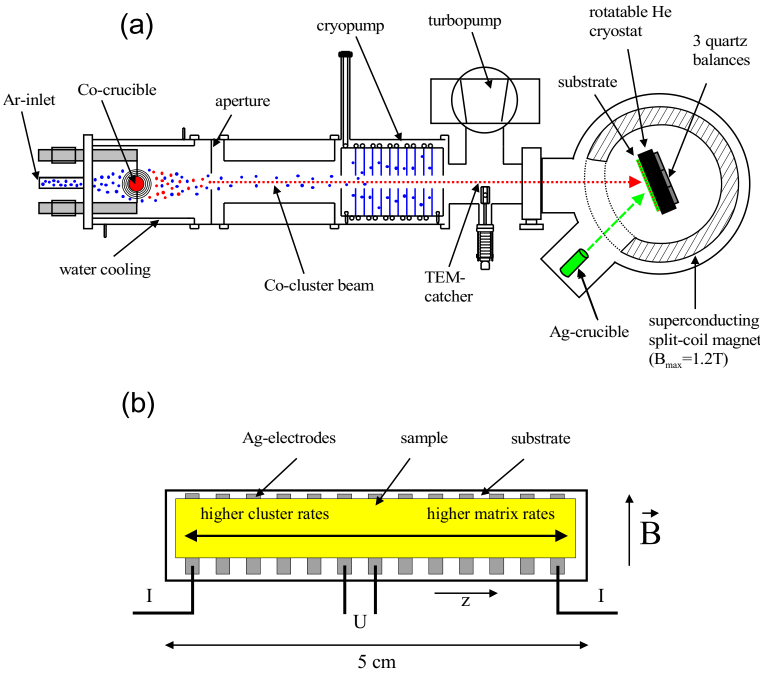

Fig. 1. (a) Experimental set-up of cluster source and He cryostat with built-in superconducting split-coil magnet. (b) Sample substrate with Ag electrodes and sample geometry for 4-probe resistance measurements.

2 Experimental set-up [12]

The experimental set-up shown in Figure 1a mainly consists of two components: the Ar-gas-aggregation cluster source [13,14] and a variable temperature He cryostat in which a crucible for the evaporation of the non-magnetic matrix (*e.g.* Ag) as well a split-coil superconducting magnet ($B \lesssim 1.2$ T) are integrated. The latter one allows *in-situ* measurements of the GMR. The cluster size distribution is obtained with the help of a thin carbon foil catcher, which is brought for a short time ($\lesssim 0.5$ s) into the cluster beam, and transmission electron microscopy (TEM) of this foil after cluster deposition. TEM analysis of the cluster size distributions for Co clusters with an average diameter $\bar{L}$ between 3 and 7 nm showed a distribution width ΔL (FWHM) of $\Delta L/\bar{L} \simeq 0.3$. The Co cluster and the Ag atomic beam have been deposited at 300 K onto a kapton substrate mounted at the cold finger of the rotatable He cryostat. Both deposition rates were controlled by quartz balances. Typical deposition rates were between 0.02 and 0.07 nm/s for the Co clusters and between 0.1 and 0.5 nm/s for the Ag atoms, resulting in films of about 200 nm after 20 min deposition.

Co cluster beam and Ag atomic beam were not parallel and thus created a composition gradient along the substrate in z-direction (see Fig. 1b). In this way different samples with different Co cluster volume fractions are created along the substrate in one sample preparation process. After the deposition and *in-situ* GMR measurements the samples have been analyzed *ex-situ* by an EDX-system for determination of the actual local Co/Ag concentration of each sample. The electrical resistance of all samples has been measured as a function of the applied magnetic field B in a four-probe configuration with an in-plane current. The magnetic field B ($\lesssim 1.2$ T) direction was in-plane and perpendicular to the current (see Fig. 1b). Due to magnetic hysteresis the resistance was measured in a sweeping magnetic field.

3 Experimental results and discussion

The resistivity of granular Co/Ag films was measured as a function of the external magnetic field B for the following 4 different mean cluster sizes $\bar{L} = 3.3$, 4.1, 5.5 and 6.9 nm with Co cluster volume fractions v_{cl} between 5 and 43%. Before presenting and analyzing these data we first have to discuss how the resistivity ρ of our samples in *zero magnetic field* depends on v_{cl} and $\bar{L}$. The resistivity ρ, measured at $T = 4.2$ K in zero magnetic field, is shown in Figure 2 as a function of v_{cl} for different Co cluster sizes $\bar{L}$. One can see that the resistivity (i) linearly increases with increasing Co volume fraction and (ii) is rather independent of Co cluster size $\bar{L}$. Only the samples with Co cluster size $\bar{L} = 6.9$ nm show a deviation from the linear behavior of ρ at large v_{cl} values. This can be explained by the following fact: the Co cluster deposition rate for these samples was ~ 0.07 nm/s and thus much higher than that for the other samples which was typically $0.01-0.02$ nm/s. Such a high deposition rate probably creates more defects in the Ag matrix and thus leads to a higher resistivity. However, we want to point out that the samples with $\bar{L} =$ 6.9 nm and $v_{cl} > 25\%$, *i.e.* those which show the largest deviation in ρ with respect to the other samples (see Fig. 2), have not been used in the further analysis of the $\bar{L}$- and v_{cl}-dependence of $\Delta\rho$.

The linear increase of ρ with increasing v_{cl} is typical for a microscopically inhomogeneous two-component system

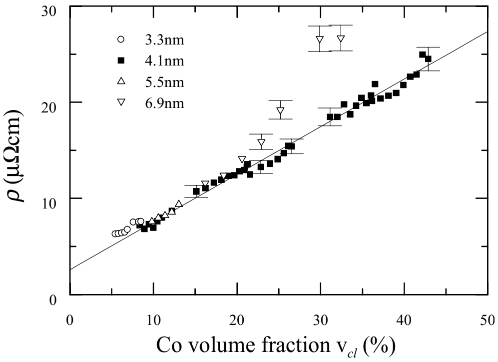

Fig. 2. Sample resistivity ρ at $T = 4.2$ K as function of Co volume fraction v_{cl} for different Co cluster sizes $\bar{L}$. The straight line through the data points is a least-squares fit assuming a linear relation between ρ and v_{cl}.

wherein one of the components (Co clusters) has a much larger resistivity than the other component (Ag matrix) and the volume fraction of the component with the much smaller resistivity (Ag matrix) is far above the percolation threshold [15,16]. Furthermore, according to percolation theory the resistivity of an inhomogeneous two-component system only depends on the volume fractions of the components but not on the length scale of disorder, *i.e.* cluster size. Thus, the resistivity data shown in Figure 2 essentially are in agreement with percolation theory.

Now we come to the main subject, namely the GMR, *i.e.* the *change* of the resistivity ρ due to an applied external magnetic field. In Figures 3a and 3b we can see the relative change $\Delta\rho/\rho$ of the resistivity as a function of external magnetic field measured at $T = 4.2$ K for two samples with the *same* composition ($Co_{21}Ag_{79}$) but for two different Co cluster sizes ($\bar{L} = 4.1$ nm and 6.9 nm, respectively). It is quite evident from these figures that $\Delta\rho/\rho$ strongly depends on $\bar{L}$ (factor 2 difference in $\Delta\rho/\rho$ between $\bar{L} = 4.1$ nm and $\bar{L} = 6.9$ nm). Before we discuss in detail the measured $\bar{L}$- and v_{cl}-dependencies of the GMR we have to ask the following question: is the *relative* change in the resisitvity, *i.e.* $\Delta\rho/\rho$, the correct way for describing the GMR if we want to compare our measured GMR data with theoretical model predictions? In fact, the *absolute* change of the resistivity, *i.e.* $\Delta\rho = \rho(B_c) - \rho(B_s)$ (B_c is the magnetic coercivity at which ρ has its maximum value and B_s is the saturation field at which ρ and the magnetization saturates), is a measure of the change $\Delta\tau^{-1}$ in the scattering rate τ^{-1} between $B = B_c$ and $B = B_s$, *i.e.* $\Delta\rho \propto \Delta\tau^{-1}$. Thus, if one wants to analyze the influence of cluster size $\bar{L}$ and cluster volume fraction v_{cl} on the change in the scattering rate due to alignment of the cluster magnetic moments, one has to discuss the change of $\Delta\rho$ with $\bar{L}$ and v_{cl}. If ρ strongly depends on v_{cl}, as it is the case for our samples (ρ changes by a factor 5 going from $v_{cl} = 5\%$ to $v_{cl} = 43\%$, see Fig. 2), a plot of ($\Delta\rho/\rho$) *vs.* v_{cl} does *not* correctly reflect the change of $\Delta\tau^{-1}$ with v_{cl}, *i.e.* ($\Delta\rho/\rho$) depends on the mean free path λ_{Ag} in the Ag matrix, while $\Delta\rho$ is *independent* of λ_{Ag}. It is for this reason that we will discuss in the following only the *absolute* resistivity change $\Delta\rho$ as a function of $\bar{L}$ and v_{cl}, respectively. The change $\Delta\rho$ can be calculated from the measured resistance change ΔR and the known sample dimensions with an accuracy of about ±5%.

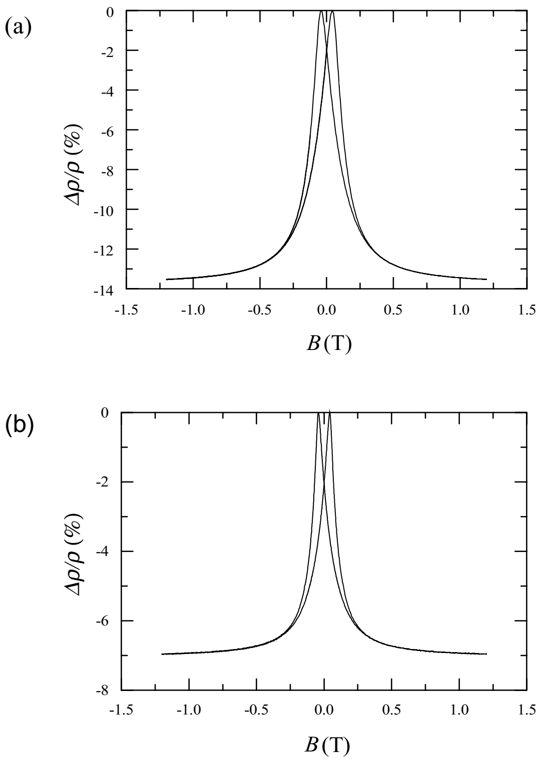

Fig. 3. Relative change $\Delta\rho/\rho$ of resistivity ρ as function of external magnetic field B for sample $Co_{21}Ag_{79}$ at $T = 4.2$ K; (a) mean Co cluster size $\bar{L} = 4.1$ nm; (b) mean Co cluster size $\bar{L} = 6.9$ nm.

In the following we will discuss in detail both the cluster size dependence as well as the cluster volume fraction dependence of $\Delta\rho$ in our samples.

3.1 Cluster size dependence of the GMR

The $\Delta\rho$ values, defined as $\Delta\rho = \rho(B_c) - \rho(B_s)$ (see above), measured for all samples at $T = 4.2$ K for 4 different mean cluster sizes $\bar{L}$ at *small* values of v_{cl} are shown in Figure 4. One can clearly see that for a given Co cluster volume fraction v_{cl} the value of $\Delta\rho$ strongly depends on the Co cluster size $\bar{L}$. The straight lines through the data points in Figure 4 are least squares fits assuming $\Delta\rho = 0$ for $v_{cl} = 0$ and a linear correlation between $\Delta\rho$ and v_{cl} for small v_{cl} values. The assumption of such a linear correlation between $\Delta\rho$ and v_{cl} certainly is justified for small v_{cl} values where each Co cluster scatters independently. Even a detailed theory for the GMR in granular systems, taking into account all possible scattering mechanisms, results in a linear dependence of $\Delta\rho$ with v_{cl} for small v_{cl} [17]. If we plot the slope m of these straight lines *versus* the inverse of the mean cluster size $\bar{L}$ we obtain a linear correlation between m and $\bar{L}^{-1}$ as shown in Figure 5. This linear correlation just indicates that spin dependent scattering at the Co/Ag interface is the only relevant scattering mechanism for the GMR. This can be seen from the following

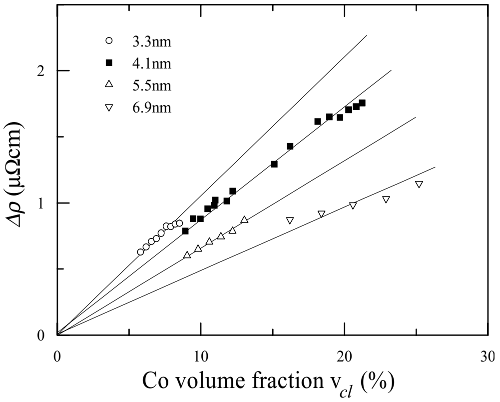

Fig. 4. Resistivity change $\Delta\rho = \rho(B_c) - \rho(B_s)$ at $T = 4.2$ K as a function of Co volume fraction $v_{cl} \lesssim 25\%$ for 4 different Co cluster sizes $\bar{L}$. The straight lines through the data points are least-squares fits assuming $\Delta\rho = 0$ for $v_{cl} = 0$ and a linear relation between $\Delta\rho$ and v_{cl}.

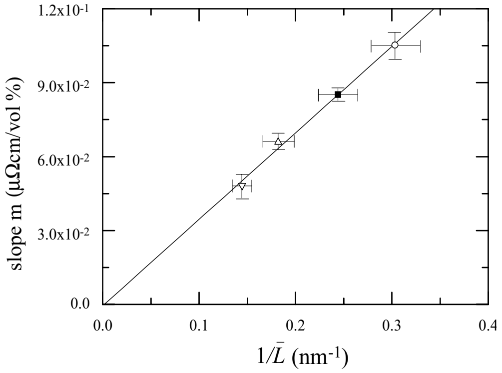

Fig. 5. Slope m of straight lines in Figure 4 as a function of $\bar{L}^{-1}$. The straight line through the data points is a least-squares fit. The horizontal error bar at each data point gives the width (FWHM) of the Co cluster size distribution.

simple arguments: at small v_{cl} values we essentially have isolated clusters, *i.e.* $A_{interface} = N_{cl} \cdot A_{cl}$ (N_{cl} = number of Co clusters, A_{cl} = surface area of single Co cluster); for a given Co cluster volume fraction v_{cl} we have $N_{cl} \propto \bar{L}^{-3}$ and with $A_{cl} \propto \bar{L}^2$ we obtain $A_{interface} \propto \bar{L}^{-1}$. A linear correlation between $\Delta\rho$ and $\bar{L}^{-1}$ in granular Co/Ag has also been observed by Xiong *et al.* [18]; however, this linear correlation (only proven for one single Co cluster volume fraction $v_{cl} = 20\%$) does *not* extrapolate to $\Delta\rho = 0$ for $\bar{L}^{-1} \to 0$. Their extrapolated value of $\Delta\rho$ for $\bar{L}^{-1} \to 0$ is about 50% of that for $\bar{L} = 5$ nm, *i.e.* in their samples other scattering mechanisms than interface scattering significantly contribute to $\Delta\rho$. This is certainly due to the different sample preparation method, *i.e.* their samples had been produced in the traditionally way as described in Section 1.

Two points have to be mentioned here: (i) annealing experiments performed in the first GMR studies of granular Co/Cu [3,4] already indicated spin-dependent interface scattering to be dominant for the GMR; (ii) a detailed *theoretical* analysis of the GMR in granular systems by Zhang and Levy [17], considering all possible scattering mechanisms (spin-dependent scattering at the cluster/matrix interface as well as within the magnetic clusters and spin-independent scattering within the magnetic clusters as well as within the non-magnetic matrix), come to the conclusion that “the GMR comes primarily from the spin dependent scattering at the interfaces between granules and the matrix.”

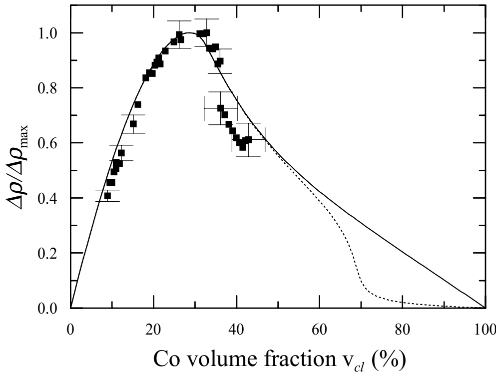

Fig. 6. $\Delta\rho$ at $T = 4.2$ K normalized to $\Delta\rho_{max}$ at $v_{cl} = 29\%$ as a function of Co cluster volume fraction v_{cl}. The solid line is a Monte-Carlo simulation of the percolation aggregate surface (interface area) as described in the text. The dashed line gives the interface area after subtraction of “inner” holes within the precolation aggregates. The interface area has been normalized to the maximal value at $v_{cl} = 29\%$.

Summarizing this part, the observed $\bar{L}$-dependence of $\Delta\rho$ for small v_{cl} (Fig. 5) gives the *experimental* proof that spin-dependent interface scattering is the only relevant scattering mechanism for the GMR in our well-defined granular Co/Ag systems.

3.2 Cluster volume fraction dependence of the GMR

Next we want to discuss how the GMR depends on the Co cluster volume fraction (v_{cl}) for a fixed cluster size. Such experiments have been performed for $\bar{L} = 4.1$ nm in the region $10\% \le v_{cl} \le 43\%$. The experimental data points are shown in Figure 6. In order to interpret these results we will start with a rather simple model based on the results of the foregoing Section. We assume that $\Delta\rho$ is determined by spin-dependent interface scattering in the *whole* Co cluster volume fraction regime. In this case one can immediately understand the shape of the curve formed by the data points in Figure 6: with increasing Co cluster volume fraction the clusters will form percolation aggregates, starting with cluster dimers, trimers, *etc.* These percolation aggregates exhibit a surface, *i.e.* a Co/Ag interface, which is *smaller* than that of the individual clusters. The domain boundaries between the single-domain clusters within these multidomain percolation aggregates will not contribute to the GMR (multidomain bulk Co has no GMR effect). Thus, the linear increase of $\Delta\rho$ with increasing v_{cl} will be continuously reduced with increasing percolation aggregate size. $\Delta\rho$ will reach its largest value at that Co cluster volume fraction which has the largest interface area and with further increasing v_{cl} the GMR effect finally will decrease and goes to zero for $v_{cl} \to 100\%$.

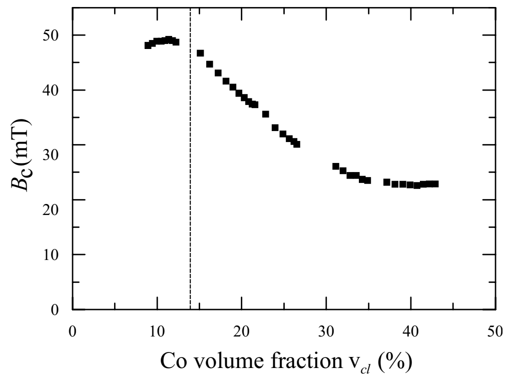

Fig. 7. Coercivity B_c as a function of Co cluster volume fraction v_{cl} for Co clusters of mean size $\bar{L} = 4.1$ nm at $T = 4.2$ K. The vertical dashed line marks the v_{cl} value at which the number of isolated Co clusters has its maximal value (see text).

The formation of such multidomain percolation aggregates can be seen in another measured quantity, the coercivity B_c, as obtained from the $\Delta\rho(B)$-curves. Figure 7 shows that B_c has its maximal value of $B_c \simeq 50$ mT, up to $v_{cl} \simeq 15\%$ which is approximately that volume fraction which, according to percolation theory, gives the largest number of isolated clusters [19]. This means that up to $v_{cl} \simeq 15\%$ we essentially have single domain Co particles with an average coercivity of $\simeq 50$ mT. For higher values of v_{cl} the coercivity B_c starts to drop, which is an indication for the formation of multidomain aggregates. A similar value for v_{cl} at which B_c has its maximal value has been found by Wang and Xiao for granular Fe/Ag [20].

In order to get a more quantitative picture how the surface of percolation aggregates changes with the cluster volume fraction one has to calculate these surfaces. The "surface" or "circumference" of a percolation aggregate is defined as the number of empty sites that are neighboring the percolation aggregate. For small percolation aggregates this can be calculated exactly. Sykes *et al.* [19], for example, have calculated the surface of percolation aggregates in a simple cubic lattice for aggregate sizes up to $n = 11$. However, since we are dealing with much larger percolation aggregates (v_{cl} is going beyond the percolation threshold) one has to use Monte-Carlo simulations in order to get the surface of such aggregates. We have performed such simulations in a simple cubic lattice with $10^2 \times 10^2 \times 10^2$ lattice sites. The validity of our computer program was checked by comparing the results of our Monte-Carlo simulations for the total surface of all percolation aggregates with sizes up to $n = 11$ with that obtained from the exact calculation [19]. The differences in the surfaces obtained by these two different methods always were smaller than 5%. The result of our Monte-Carlo simulations for cluster volume fractions $0 \le v_{cl} \le 100\%$ is shown in Figure 6. The solid line corresponds to the total surface including "inner" surfaces, *i.e.* surfaces which are completely surrounded by a percolation aggregate. After substraction of these "inner" surfaces one obtains the dashed line in Figure 6. Here we want to make two additional remarks to our Monte-Carlo simulations: (i) in order to reduce the statistical error each simulation for a given v_{cl} was repeated 10 times and the results were averaged; (ii) empty lattice sites which were lying at the surface of the finite ($10^2 \times 10^2 \times 10^2$) lattice block have not been counted. Otherwise the fully occupied lattice which per definition has zero interface area would have resulted in a surface of 6%.

If we compare the results of our $\Delta\rho$ measurements (normalized to $\Delta\rho_{max}$ at $v_{cl} = 29\%$) with that of our Monte-Carlo simulations (again normalized to maximum surface at $v_{cl} = 29\%$) the agreement between experiment and simulation is surprisingly good (see Fig. 6). This fact is an additional proof that interface scattering is the only relevant scattering mechanism causing the GMR in our samples.

Now we want to discuss why the rather simple relation $\Delta\rho \propto A_{interface}$ has to be modified for large v_{cl} values. If we look more carefully at the data points in Figure 6 we see that there is a discrepancy between experiment and simulation at large v_{cl} values ($v_{cl} \gtrsim 35\%$). This could be explained by the fact that those samples with large v_{cl} have a relative large error in v_{cl} (the composition was only determined from the deposition rates but not with EDX analysis). However, the reason for this discrepancy certainly is a more fundamental one. If the GMR effect $\Delta\rho$ is caused by spin-dependent interface scattering, the magnitude of $\Delta\rho$ does not only depend on the percolation aggregate surface but also on the orientations of the cluster magnetic moments relative to each other in zero magnetic field [17]. These orientations will change with increasing cluster volume fraction due to increasing magnetic interaction (either *via* RKKY or dipolar) between the clusters. Starting with a complete random orientation of the cluster magnetic moments at very low cluster concentration, the magnetic moments will have the tendency to align parallel to each other and finally form a ferromagnetic multidomain bulk material for $v_{cl} \to 100\%$. Thus, $\Delta\rho$ should decrease faster than the interface area with increasing v_{cl} for large v_{cl} values. The competition between the magnetic anisotropy of the individual clusters, which have a random orientation from the crystallographic point of view, and the magnetic interaction between these clusters determines at which v_{cl} value the initially random orientation of the magnetic moments will start to change. For Co clusters, which have a relative large magnetocrystalline anisotropy, such a change in the magnetic moment orientation will occur at relative high values of v_{cl}. This probably is the reason why for Co clusters in Ag the measured GMR effect $\Delta\rho$ is proportional to the interface area up to $v_{cl} \simeq 35\%$. For Fe clusters, on the other hand, which have a much smaller magnetocrystalline anisotropy, it is expected that $\Delta\rho$ will decrease faster than the interface area already at much smaller cluster volume fractions.

Finally, we want to point out that it is the well-defined granular structure of our samples which allows us to make a comparison between experiment and Monte-Carlo simulations. Only if one has (a) clusters of well-defined size and (b) a mean cluster size which does not depend on the cluster volume fraction v_{cl}, one can calculate the interface area (surface area of all percolation aggregates) as a function of v_{cl} in the above described manner, namely, by simply counting the number of empty lattice sites neighboring the percolation aggregates (occupied sites) as a function of cluster volume fraction (lattice site occupation). In this context we should mention that the cluster volume fraction dependence of the GMR effect in almost all of the granular Co/Ag systems prepared in the usual way (see Sect. 1) is not too much different from the v_{cl} dependence in our samples: there is always a maximum in $\Delta\rho$ in the region $15\% \leq v_{cl} \leq 40\%$ [6,20–24]. However, both, position of maximal $\Delta\rho$ as well as detailed v_{cl} dependence of $\Delta\rho$, strongly depend on sample preparation conditions. Thus, a quantitative analysis of the GMR data of those samples is not possible.

4 Conclusion and outlook

Both, the cluster size dependence as well as the cluster volume fraction dependence of the GMR effect clearly show that the spin-dependent interface scattering is the only mechanism which is causing the GMR effect in our well-defined granular Co/Ag samples. There are, however, still some open questions. It is not yet clear, for example, how the GMR effect changes with the Co volume fraction v_{cl} for large v_{cl} values ($v_{cl} \gtrsim 40\%$). Furthermore, it would be interesting to study the v_{cl} dependence of $\Delta\rho$ in another well-defined granular system, for example, Fe clusters embedded in an Ag matrix. Since Fe clusters have a much smaller magnetocrystalline anisotropy than Co clusters one should expect differences between Co/Ag and Fe/Ag at large v_{cl} values, however, no differences at small v_{cl} values.

Another interesting point regarding future experiments is the following: the usual preparation method of granular systems is limited to systems which are immiscible in the equilibrium phase diagram. Fe and Pt, for example, form intermetallic compounds (Fe_3Pt, FePt, *etc.*) and it is for this reason that until now it was not possible to observe the GMR effect in this system [20]. With our preparation method, however, one can prepare Fe clusters embedded in Pt and study the GMR effect of such samples.

Finally we want to mention that now one can even study the GMR effect in a *monolayer* of ferromagnetic clusters embedded in a non-magnetic matrix, *i.e.* in "two-dimensional" cluster films [25].

Helpful discussions with D. Stauffer on the problem and definition of surfaces of percolation aggregates are gratefully acknowledged. We also want to thank his students R. Küsters and T. Török who have performed for us the exact calculations of the percolation aggregate surfaces for sizes up to $n = 11$. It were these calculations which have motivated us to use Monte-Carlo simulations for the determination of larger percolation aggregate surfaces. This work was supported by the Deutsche Forschungsgemeinschaft (SFB 341).

References

1. M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
2. G. Binasch, P. Grünberg, F. Saurenbach, W. Zinn, Phys. Rev. B **39**, 4828 (1989).
3. A.E. Berkowitz, J.R. Mitchell, M.J. Carey, A.P. Young, S. Zhang, F.E. Spada, F.T. Parker, A. Hulten, G. Thomas, Phys. Rev. Lett. **68**, 3745 (1992).
4. J.Q. Xiao, J. Samuel Jing, C.L. Chien, Phys. Rev. Lett. **68**, 3749 (1992).
5. B. Abeles, P. Sheng, M.D. Coutts, Y. Arie, Adv. Phys. **24**, 407 (1975).
6. K. Ounadjela, S.M. Thompson, J.F. Gregg, A. Azizi, M. Gesler, J.P. Deville, Phys. Rev. B **54**, 12252 (1996).
7. S.A. Makhlouf, K. Sumiyama, K. Wakoh, K. Suzuki, T. Takanashi, H. Fujimori, J. Magn. Magn. Mater. **126**, 485 (1993).
8. D.A. Eastham, Y. Quing, T.H. Maddock, J. Kraft, J.-P. Schille, G.S. Thompson, H. Haberland, J. Phys.: Condens. Matter **9**, L497 (1997).
9. A. Perez, P. Melinon, V. Dupuis, P. Jensen, B. Prevel, J. Tuaillon, L. Bardotti, C. Martet, M. Treilleux, M. Broyer, M. Pellarin, J.L. Vaille, B. Palpant, J. Lerme, J. Phys. D: Appl. Phys. **30**, 709-721 (1997).
10. V. Dupuis, J. Tuaillon, B. Prevel, A. Perez, P. Melinon, G. Guiraud, F. Parent, L.B. Steren, R. Morel, A. Barthelemy, A. Fert, S. Mangin, L. Thomas, W. Wernsdorfer, B. Barbara, J. Magn. Magn. Mater. **165**, 42-45 (1997).
11. F. Parent, J. Tuaillon, L.B. Stern, V. Dupuis, B. Prevel, A. Perez, P. Melinon, G. Guiraud, R. Morel, A. Barthelemy, A. Fert, Phys. Rev. B **55**, 6 (1997).
12. S. Rubin, H. Micklitz, in *Materials Science Forum*, edited by D. Fiorani, M. Magini (Trans. Tech. Publ. Ltd., Switzerland) **235-238**, 711 (1997).
13. F. Frank, W. Schulze, B. Tesche, J. Urban, B. Winkler, Surf. Sci. **156**, 90 (1985).
14. B. Weitzel, A. Schreyer, H. Micklitz, Europhys. Lett. **12**, 123 (1990).
15. S. Kirkpatrick, Phys. Rev. Lett. **27**, 1722 (1971).
16. D.S. McLachlan, M. Blaszkiewicz, R.E. Newnham, J. Am. Ceram. Soc. **73**, 2187 (1990).
17. S. Zhang, P.M. Levy, J. Appl. Phys. **75**, 5315 (1993).
18. P. Xiong, G. Xiao, J.Q. Wang, J. Xiao, J.S. Jiang, C.L. Chien, Phys. Rev. Lett. **69**, 3220 (1992).
19. M.F. Sykes, D.S. Gaunt, M. Glen, J. Phys. A **9**, 1705 (1976).
20. J.Q. Wang, G. Xiao, Phys. Rev. B **49**, 3982 (1994).
21. A. Maeda, M. Kume, S. Oikawa, T. Tanuma, Y. Shimizu, M. Doi, J. Phys.: Condens. Matter **5**, 6745 (1993).
22. A. Tsoukatos. H. Wan, C. Hadjipanays, K.M. Unruh, J. Appl. Phys. **73**, 5509 (1993).
23. F. Conde, C. Gómez-Polo, A. Hernando, J. Magn. Magn. Mater. **138**, 123 (1994).
24. H. Sang, Z.S. Jiang, G. Guo, J.T. Ji, S.Y. Zhang, Y.W. Du, J. Magn. Magn. Mater. **140-144**, 589 (1995).
25. S. Rubin, Ph.D. thesis, University of Köln, 1997.