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Rapid Note

Tunneling magnetoresistance in granular films made of well-defined Co clusters embedded in an inert-gas matrix

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Abstract. We present a new tunneling magnetoresistance (TMR) system. Granular films consisting of well-defined ferromagnetic Co clusters embedded in insulating inert-gas matrices Kr and Xe have been prepared by the co-deposition of in-beam prefabricated Co clusters and inert-gas atoms. Experiments show that the resistance of these films clearly follows an $\exp(C/T^{1/2})$-law and reveal a tunneling magnetoresistance (TMR) of about 7% at 10 K. The TMR is found to be only weakly temperature dependent and independent of the tunneling barrier.

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First reports on the observation of negative magnetoresistance in granular films built of ferromagnetic metallic particles embedded in an insulating matrix date back more than 25 years from now [1]. The theoretical interpretation was given a few years later by Helman and Abeles [2] in terms of tunneling of spin-polarized electrons between ferromagnetic grains with a magnetic field dependent orientation of their magnetic moment directions. This so-called tunneling magnetoresistance (TMR $= -\Delta\rho/\rho_{\rm max} = -[\rho(B_{\rm s}) - \rho(B_{\rm c})]/\rho(B_{\rm c})$ with $B_{\rm s}$ and $B_{\rm c}$ the magnetic saturation and coercive field, respectively), therefore, can give information on the spin-polarization P of the conduction electrons at the Fermi energy $E_{\rm F}$ and for that reason is of fundamental interest. The recent discovery of other large magnetoresistive effects, the so-called giant magnetoresistance (GMR) in magnetic multilayers [3,4] as well as in granular systems [5,6] and the so-called colossal magnetoresistance (CMR) in manganites [7] and their potential technical application for magnetoresistive sensors made the study of spin-dependent transport properties to a very active field of research and renewed the interest in the TMR. Numerous experiments on the TMR in various new granular systems have been reported in the last few years [8–13].

The preparation method of these granular systems, however, did not change too much from those of the early studies: the samples usually are prepared by co-sputtering or co-evaporation of the metallic and insulating component onto a warm substrate or by the reactive sputtering of metallic components (*e.g.* sputtering of a Co/Al target in O_2-atmosphere results in a sample of Co clusters embedded in an Al_2O_3 matrix [8,10]). Metal cluster formation is due to phase segregation by surface migration during deposition and/or following annealing. The morphology of samples obtained in this way naturally is not well-defined both with respect to metal cluster size as well as regarding to the insulating matrix. The latter may even contain isolated magnetic impurities of the cluster material which will influence the spin-dependent tunneling process in an uncontrolled way. For that reason it is highly desirable to prepare *well-defined* granular systems of ferromagnetic metallic clusters in a *pure* insulating matrix. This nowadays can be obtained by the combination of in-beam metal cluster preparation with the inert-gas-matrix-isolation technique, *i.e.* by the co-deposition of well-defined metal clusters and inert gas matrix atoms onto a cold substrate [14]. In this way one obtains granular samples with (i) a well-defined cluster size which is independent of the cluster volume fraction and (ii) no (or negligible) interaction of the insulating matrix with the metal cluster surface. Having such samples one can study in detail how the spin-dependent tunneling process depends on metal cluster size and tunneling barrier thickness, respectively. In this paper we would like to present first measurements of the TMR made on films that were prepared as described above, *i.e.* by co-deposition of well-defined Co clusters and inert-gas matrix atoms onto a cold substrate.

Samples with Co clusters of mean size $L \simeq 5$ nm embedded in either a Kr or Xe matrix and a Co cluster

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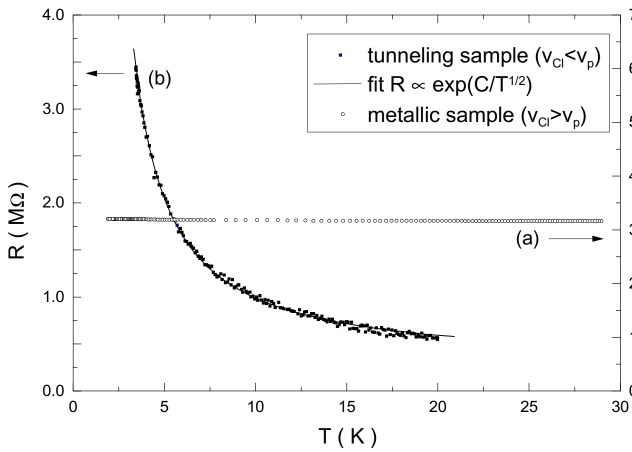

Fig. 1. $R(T)$-dependence for two Co/Kr-samples. The Co volume fraction v_{Cl} of sample (a) is above the percolation threshold v_{p} that of sample (b) below v_{p}. The $R(T)$-curve of sample (b) is fitted with the theoretically predicted exponential law.

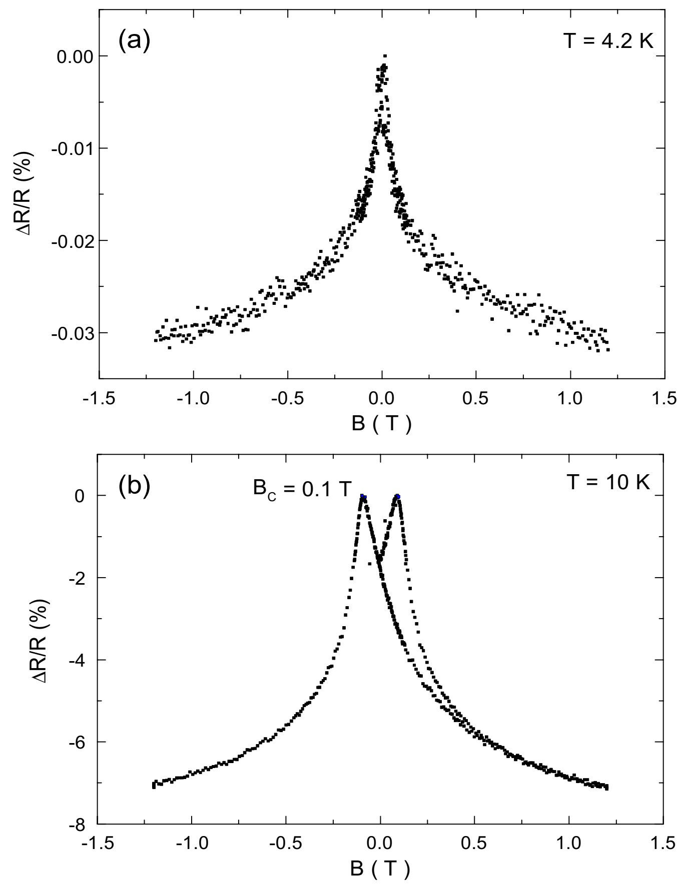

Fig. 2. Magnetoresistance of samples (a) and (b) presented in Figure 1: (a) magnetoresistance of metallic sample (a), (b) tunneling magnetoresistance of sample (b).

volume fraction below the percolation threshold show a large TMR with a value of $\simeq$ 7% at 10 K. The value of the TMR is rather independent of resistivity (6×10^5 $\mu\Omega$cm $\leq \rho \leq 1 \times 10^{11}$ $\mu\Omega$cm) and only weakly temperature dependent (10 K $\leq T \leq$ 40 K). Extrapolating the TMR to $T \rightarrow 0$ K using Julliere's formula for the TMR [15] results in a spin polarization $P \simeq 0.31$ which well agrees with the value found for *bulk* Co [16].

The experimental set-up for sample preparation and measurement will be presented only in brief. A more detailed description already has been given elsewhere [17]. The granular films are obtained by co-deposition of ferromagnetic well-defined Co clusters and inert gas matrix atoms onto a cold sapphire substrate (T = 40 K). The substrate is mounted onto the coldfinger of a ^{4}He cryostat. After the evaporation the magnetoresistance of the films can be measured *in-situ*. The clusters are prepared in a so called inert-gas aggregation cluster source which allows the preparation of metal clusters with diameters between 2 nm and 12 nm and typical cluster size distributions with a width ΔL (FWHM) of $(\Delta L/L) \simeq 0.2$. Cluster- and matrix-beams are not parallel but have an angle of 45° resulting in a concentration gradient within the evaporated film. This is used to get several samples with different cluster volume fractions by one evaporation process which can be measured separately. The deposition rates of matrix- and cluster-material are controlled by quartz balances. Typical deposition rates for the Co clusters are 3 Å/min. The films have a typical thickness of $\simeq$ 50 nm. Due to the inert gas matrix all measurements are limited to a maximum temperature of $\simeq$ 40 K. The measurements in magnetic field are performed with a build-in split-coil superconducting magnet ($B \lesssim 1.2$ T). Due to hysteresis the magnetoresistance is measured in a sweeping magnetic field. All resistance measurements were performed by a dc-four-probe technique. For the high-resistance samples an electrometer was used.

First we want to show that our method embedding prefabricated Co cluster in an inert-gas matrix is very suitable for preparing TMR samples. Therefore, in the following two samples (a) and (b) are compared. The two samples distinguish each other by their Co volume fraction v_{Cl}. Qualitatively speaking the Co volume fractions of the samples (a) and (b) are above and below the percolation threshold $v_{\mathrm{p}} \simeq 34\%$, respectively. Thus, it can be expected that in sample (a) the electronic conductivity is dominated by at least one continuous path of Co clusters while in sample (b) any path is interrupted by at least one tunneling junction resulting in a network of tunneling junctions further below the percolation threshold. In consequence the resistance of the two samples behaves very different with temperature as shown in Figure 1. Not only the absolute value of resistance differs by about five orders of magnitude between the two samples. Furthermore, while the resistance of sample (a) remains nearly unchanged the resistance of the tunneling sample increases very rapidly with decreasing temperature obeying a $R(T) \propto \exp(C/T^{1/2})$-law. This behaviour is in agreement with theoretical predictions for granular tunneling samples, *e.g.* [2,18]. From our results an explicit consideration of tunneling between clusters of different sizes as proposed by Mitani *et al.* [19] to provide a better adjustment of the $\exp(C/T^{1/2})$-law does not seem to be necessary.

The magnetoresistance of the two samples was measured at constant temperatures in a sweeping magnetic field $|B| \leq 1.2$ T. The magnetoresistance curves of both samples are shown in Figure 2. The differences between the two samples are obvious: sample (a) shows a vanishing small magnetoresistive effect of $\sim 0.03\%$ at 4.2 K (the usual magnetoresistance for *bulk* Co) while the effect of sample (b) is more than two orders of magnitude higher and has a value of $\sim 7\%$ at 10 K. Furthermore sample (b) shows a hysteresis with a coercive field $B_c \simeq 0.1$ T. Such a hysteresis is expected if the observed effect is caused by ferromagnetically interacting superparamagnetic clusters below their blocking temperature. For a mean cluster size of 5 nm one can estimate the blocking temperature T_B to be with $T_B > 200$ K [20]. The measured coercive field does not deviate from coercive fields already obtained for Co clusters in an Ag matrix [20].

Similar experiments have been performed with the inert-gas Xe as a matrix material. The $R(T)$-dependence measured for two different Xe samples (A) and (B) also follows the exponential law already observed in the case of the Kr sample. This can be seen in Figure 3a which shows that a plot of $\rho(T)$ *vs.* $T^{-1/2}$ results in straight lines for both Xe and the Kr sample. The TMR was measured for both Xe samples at constant temperatures in the temperature region 40 K to 10 K. The result is shown in Figure 3b: the TMR values for both Xe samples agree quite well which each other and with the TMR value for Kr at 10 K. The temperature dependence of the TMR in this temperature region is rather weak, which is in contrast to the theoretical work by Helman and Abeles [2] who predicted a $1/T$-dependence of the TMR. Inoue *et al.*, on the other hand, neglected the magnetic energy which was considered by Helman and Abeles and they obtained for sufficiently high magnetic fields a TMR expression that is temperature independent [18]. However, at this point one should also mention that Mitani *et al.* measured a considerably rise in the TMR for low temperatures in the system Co/Al_2O_3 [19]. The authors of this work explain their observed increase by sequential tunneling. Another explanation for an enhancement of the TMR at low temperatures is cotunneling [21]. However, in order to draw any conclusion about the origin of the T-dependence of the TMR in *our* samples more data points especially at low temperatures are needed. This will be the subject of further studies.

The most important point of *this* paper is the following: while the resistivities of the three samples differ by more than three orders of magnitude (Kr: $\rho(20\text{ K}) \simeq 1 \times 10^7$ $\mu\Omega$cm, Xe: $\rho(20\text{ K}) \simeq 4 \times 10^{10}$ $\mu\Omega$cm and 1×10^{10} $\mu\Omega$cm for sample (A) and (B), respectively) the TMR essentially is the same (see Fig. 3b). This is in perfect agreement with the simple model developed by Julliere [15]: according to this model the TMR $\Delta R/R = P^2/(1+P^2)$ is independent of the tunneling barrier height or thickness, respectively. The spin polarization P is given by $P = (D_\uparrow - D_\downarrow)/(D_\uparrow + D_\downarrow)$ with $D_\uparrow$ and $D_\downarrow$ being the density of states at the Fermi energy E_F for spin up and down, respectively. In addition to Julliere's simple model there are also other more sophisticated theories that clearly predict that the TMR depends on tunneling barrier height and thickness, respectively [22,23]. Such a dependence is not observed in our measurements. Granular Co/Al_2O_3-films prepared in the usual way (see above) show TMR-values which depend on the resistivities of the samples and which are somewhat higher than our values, *e.g.* between 7% and 12% at 30 K for resistivities in the range from 1×10^5 to 1×10^6 $\mu\Omega$cm [19]. The reason for this difference is yet unclear and will be subject of further studies (see below).

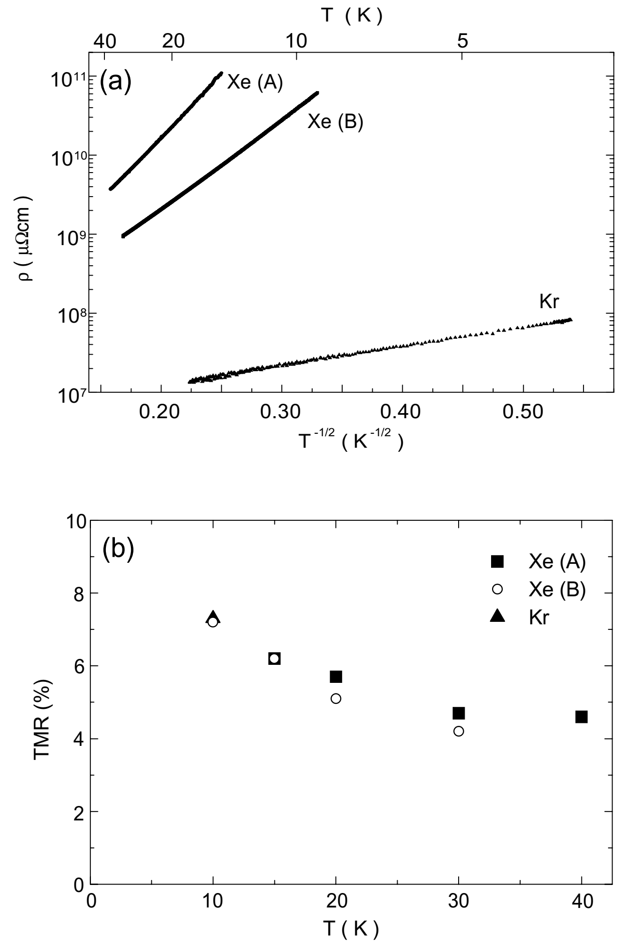

Fig. 3. (a) Temperature-dependence of the resistivity (logarithmic plot) for the two Xe samples (A) and (B) and the Kr sample, (b) temperature-dependence of the TMR for the three samples.

Extrapolating the T-dependence of the TMR in Figure 3 (inset) to 0 K one obtains a TMR value of $\Delta R/R \simeq 0.09$ which according to Julliere results in a spin polarization of $P \simeq 0.31$. This value agrees well with that given by Meservey *et al.* for *bulk* Co ($P_{Co} = 0.35 \pm 0.03$) [16]. However, it should be mentioned that our estimation of P is rather preliminary since the exact temperature dependence of the TMR especially at low temperatures still has to be examined (see above).

Finally it should be mentioned that all samples studied exhibit ohmic behaviour, *i.e.* show a linear I,V-curve for voltages up to 10 V. This indicates that (due to the current-in-plane-arrangement of our measurement) a large number of tunneling barriers are involved, resulting in quite small voltages (probably $\ll 1$ mV) across one single tunneling barrier.

Summarizing, we have found a new granular TMR system that is made of in-beam prepared well-defined Co clusters embedded in an inert-gas matrix (Kr, Xe). Such films show a $R(T)$-dependence that is in agreement with the predicted $R \propto \exp(C/T^{1/2})$-law. The observed TMR seems to depend only weak on temperature and are rather independent of tunneling barrier height and thickness, respectively. However, further measurements especially at lower temperatures are necessary to make more reliable predictions of the TMR(T)-behaviour. In order to investigate the influence of the matrix material on the TMR we plan to prepare films where the prefabricated clusters are embedded in a co-evaporated Al_2O_3-matrix, which is the matrix material usually taken for TMR studies in granular films.

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