Giant magnetoresistance in cluster-assembled nanostructures: a quantitative approach

Simón Oyarzún, Artur Domingues Tavares de Sá, Véronique Dupuis and Matthias Hillenkamp

1 Institut Lumière Matière, UMR5306 Université Lyon 1-CNRS, Université de Lyon, 69622 Villeurbanne cedex, France
2 Instituto de Física Gleb Wataghin, UNICAMP, CP 6165, Campinas, SP 13083-970, Brazil
E-mail: matthias.hillenkamp@univ-lyon1.fr

Abstract. In a recent publication [1] we have discussed qualitatively the applicability of commonly used assumptions and models in the description of giant magnetoresistance in granular media by comparing to well-defined cluster-assembled nanostructures of cobalt clusters embedded in copper thin films. In this article we present a quantitative analysis and discuss the relevant parameters appearing in the model.

1. Introduction
Spin-dependent transport in granular media is a field of contemporary research with high potential as well as many remaining open questions. We have, in a recent publication [1], described the fabrication and study of benchmark cluster-assembled nanostructures consisting of well-defined cobalt nanoparticles around 2.3 nm in diameter embedded in copper matrices. They were prepared from preformed gas-phase clusters following the low-energy cluster beam deposition technique [1, 2]. The thorough magnetic characterization based on the “triple fit” [3] permitted us to unambiguously determine the median magnetic size, its dispersion as well as the effective anisotropy constant of the nanoparticles. Low temperature (2 K) remanence measurements furthermore allowed us to verify that inter-particle interactions are negligible at very low concentrations (0.5 at.% in this case), a crucial prerequisite for the commonly accepted description of giant magnetoresistance (GMR) in granular systems [4, 5].

Besides testing for the first time the model to describe GMR introduced by Zhang and Levy [6] with a truly superparamagnetic sample, we addressed the influence of spin-dependent scattering at the interface and within the volume of the nanoparticle. The literature is controversial [4, 7] and we could, using a parametrized formula, deduce that the stated discrimination between the two influences is highly questionable because of inter-particle interactions.

In order to better describe our data and continue testing the model, we decided to use the whole formula derived by Zhang and Levy and fit our data quantitatively. While this quantitative description is expected to give more insight in the underlying physics, it also introduces a considerable number of free parameters. We have seen before that the magnetic information contained in the room temperature magnetization cycle is highly ambiguous [3, 8], so we have to beware of the same ambiguity also in the magnetoresistance curves. We show in this article a first quantitative fit to our data and a limitation to the applicability of the model.
2. Discussion

The commonly used model to describe GMR in granular media was introduced by Zhang and Levy [6] and leads to the following expression for the field-dependent variation of the resistivity:

$$\frac{\Delta \rho}{\rho} = -\frac{\xi_0^2}{\xi_1}$$  \hspace{1cm} (1)

where

$$\xi_0 = \frac{1 - c}{\lambda_{nm}} + \frac{c}{\lambda_m} (1 + p_b^2) + \frac{a_0(36\pi)^{1/3}}{\lambda_s} \int V_a^{2/3} f(V_a) dV_a$$ \hspace{1cm} (2)

$$\xi_1 = \frac{2cp_b}{\lambda_m} \int V_a f(V_a) m_a(V_a) dV_a + \frac{a_02(36\pi)^{1/3}}{\lambda_s} \int V_a^{2/3} f(V_a) m_a(V_a) dV_a.$$ \hspace{1cm} (3)

The parameters used are the mean free paths $\lambda_{nm,m,s}$ in the non-magnetic matrix, inside the magnetic grains and at the surface of the grain, respectively; $p_s$ and $p_b$ represent the ratios of the spin dependent to spin independent potentials for the interface and the volume of the nanoparticles. The nanoparticle concentration (molar fraction) is denoted by $c$, $a_0$ is the lattice constant of the nanoparticle material, $f(V_a)$ the size distribution function and $m_a$ the corresponding magnetic moment. Field and temperature mainly enter the formula through the Langevin description of the magnetization, the other parameters except $\lambda_{nm}$ show only very weak variations with $H$ and $T$ in the superparamagnetic state (as discussed below).

Additional information can be obtained from the comparison with a pure Cu film of comparable thickness and prepared under identical conditions. The $\rho(T)$ curve nicely overlaps with the one measured for the sample with 0.5 at.% Co concentration. This directly shows that $\lambda_{nm}$ is dominated by scattering events within the thin film, e.g. at grain boundaries, dislocations or the surfaces [9, 10].

Fig.1 shows fits according to the model at different temperatures. $\lambda_{nm}$ at the respective temperature is in agreement with the pure Cu film within the error bars and the Co bulk value was taken for the lattice constant $a_0$. This leaves as free parameters the median size and dispersion of the magnetic size distribution, the mean free paths $\lambda_{m,s}$ and the ratios of the spin dependent to spin independent potentials $p_s$ and $p_b$. At all temperatures shown here...
Table 1. Parameters obtained from fits to GMR data at different temperatures. Typical errors are displayed for 200 K. The median diameter $D_{GMR}^m$ and the diameter dispersion $w_{GMR}^m$ are to be compared to $D_{TEM}^m = 2.3$ nm [1].

|       | 200 K | 150 K | 100 K | 50 K |
|-------|-------|-------|-------|------|
|       | Volume | Interf. Vol. | Interf. Vol. | Interf. Vol. |
| $p_s$ | 0.7±0.3 | 0 | 0.7 | 0 |
| $p_b$ | 0 | 0.7±0.3 | 0 | 0.7 |
| $\lambda_s$ (nm) | 1.9±0.9 | 3.8 | 1.4 | 3.0 |
| $\lambda_m$ (nm) | 25±10 | 3.6±1.6 | 25 | 3.7 |
| $\lambda_{nm}$ (nm) | 26±2 | 31 | 42 | 53 |
| $D_{GMR}^m$ (nm) | 2.4±0.3 | 2.4 | 2.4 | 2.3 |
| $w_{GMR}^m$ (nm) | 0.2±0.1 | 0.2 | 0.2 | 0.2 |

the experimental data are very nicely reproduced by the quantitative model. As for 200 K [1] we obtain identical fits when restricting the scattering to either the surface ($p_b = 0$) or the cluster volume ($p_s = 0$), no definite conclusion about the different contributions can be taken at the moment. It should be noted that even if $p_s$ or $p_b$ are set to zero, a finite term for the corresponding mean free path has to be assumed since both appear in the spin-independent scattering term $\xi_0$.

Looking at the derived numbers shows that above 100 K, when the whole cluster ensemble is in the superparamagnetic state, all the remaining free parameters have reasonable values, no obviously unphysical values are obtained. At 50 K, however, the parameters start to change, and as the temperature gets closer to $T_{max} = 13$ K [1] the median determined size starts to deviate from the correct value of 2.3 nm. The superparamagnetic assumption is no longer valid since a fraction of the ensemble is already blocked.

In conclusion we have shown that it is possible to obtain a quantitative description of the GMR curves at high temperatures using the Zhang and Levy model. In particular, for $T > 5T_{max}$, we obtain the same magnetic size distribution as in the magnetic characterization. Nevertheless, due to the high number of free parameters in the model, it is impossible for now to discriminate between the postulated contributions to spin-dependent scattering from the nanoparticle volume and surface. Further experiments with varying nanomagnet diameters are in progress to illustrate finite size effects in spintronics.

References

[1] Oyarzúin S, Domingues Tavares de Sa A, Tuallon-Combes J, Tamion A, Hillion A, Boisron O, Mosset A, Pellarin M, Dupuis V and Hillenkamp M 2013 J. Nanopart. Res. 15 1968
[2] Perez A, Dupuis V, Tuallon-Combes J, Bardotti L, Prevel B, Bernstein E, Mélinon P, Favre L, Hannour A and Jamet M 2005 Adv. Eng. Mater. 7 475–485
[3] Tamion A, Hillenkamp M, Tournus F, Bonet E and Dupuis V 2009 Appl. Phys. Lett. 95 062503
[4] Allia P, Knobel M, Tiberto P and Vinai F 1995 Phys. Rev. B 52 15398–15411
[5] Battle X and Labarta A 2002 J. Phys. D: Appl. Phys. 35 R15–R42
[6] Zhang S and Levy P M 1993 J. Appl. Phys. 73 5315
[7] Rubin S, Hohenfried M and Micklitz H 1998 Eur. Phys. J. B 5 23–28
[8] Tamion A, Hillenkamp M, Tournus F, Bonet E and Dupuis V 2012 Appl. Phys. Lett. 100 136102
[9] Henriquez R, Cancino S, Espinosa A, Flores M, Hoffmann T, Kremer G, Lisoni J G, Moraga L, Morales R, Oyarzun S, Suarez M A, Zúñiga A and Munoz R C 2010 Phys. Rev. B 82(11) 113409
[10] Henriquez R, Moraga L, Kremer G, Flores M, Espinosa A and Munoz R C 2013 Appl. Phys. Lett. 102 051608