Pressure Dependence of the Magneto-transport Properties in Fe/MgO Granular Systems

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The effect of hydrostatic pressure at room temperature on the resistance and magnetoresistance (MR) of a discontinuous metal-insulator multilayer (DMIM) of nominal composition $[\text{Fe}(t_{\text{Fe}} = 0.7 \text{ nm})/\text{MgO}(t_{\text{MgO}} = 3 \text{ nm})]_{15}$ has been studied. The resistivity of the DMIM, ρ , decreases linearly with pressure indicating an increase in conduction via tunneling effect. The value of coefficient $(1/\rho_0)d\rho/dP = -3.9 \times 10^{-2} \text{ kbar}^{-1}$ is higher than reported values in other granular films implying that the electronic state of the DMIM is close to the iron percolation threshold. At the maximum applied magnetic field (3 kOe) the MR ratio increases from 0.6% at ambient pressure to 1.1% at 7 kbar. This result can be explained by a reduction of the tunnel barrier width induced by the hydrostatic pressure.

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I. INTRODUCTION

Discontinuous metal-insulator multilayers (DMIMs) are a special example of granular thin films consisting of metallic, granular-like layers, intercalated between insulating layers. The size and shape of the nanosized granules can be controlled by the nominal thickness of the metal layer. The DMIMs based on ferromagnetic metals showed moderate room temperature magnetoresistance originating from spin-dependent tunneling between adjacent metallic granules [1–5]. During the last years we have performed an exhaustive study of Fe/MgO DMIMs prepared by pulsed laser deposition (PLD) on glass and MgO single crystal substrates [6–9]. We have shown that the iron percolation threshold for this system

is around Fe nominal thickness, $t_{\text{Fe}} = 0.8 \text{ nm}$. Samples below percolation threshold exhibit isotropic MR due to spin-polarized electron tunneling between nanometer-sized superparamagnetic Fe grains [8].

The temperature dependence of the MR in these materials can be explained according to the theoretical model developed by Mitani *et al.* [10] which takes into account higher-order processes of spin-dependent tunneling between large granules through intervening small ones with strong Coulomb blockade. In this model the width of the tunnel barrier determined by the mean separation of the Fe particles, s , is a key factor to determine the tunneling MR ratio, $|MR| \propto (s\chi)^{-1}$. The hydrostatic pressure, P , is an external parameter that can modify s , which strongly affects the MR value. Despite this fact, the studies of the effect of P on the magneto-transport properties in metal-insulator granular systems are scarce. To our knowledge, only some pioneering works performed

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Fig. 1. Dependence of electrical resistivity, ρ , versus hydrostatic pressure for $[\text{Fe}(t_{\text{Fe}} = 0.7\text{nm})/\text{MgO}(t_{\text{MgO}} = 3\text{nm})]_{15}$ DMIM at room temperature. The solid line corresponds to the best linear fit.

by *S. Kaji* and co-workers in Co-Al-O films have been done [11–13]. The authors observed a decrease of the tunneling magnetoresistance (TMR) at room temperature and an increase of TMR at 4.2 K with increasing the hydrostatic pressure. These results were explained by considering higher order tunneling effect [10]. In order to continue these studies we present in this work some preliminary results of the effect of P on resistivity and TMR at room temperature in a DMIM with nominal composition $[\text{Fe}(t_{\text{Fe}} = 0.7\text{ nm})/\text{MgO}(t_{\text{MgO}} = 3\text{ nm})]_{15}$.

II. EXPERIMENT AND DISCUSSION

The selected polycrystalline $[\text{Fe}(t_{\text{Fe}} = 0.7\text{ nm})/\text{MgO}(t_{\text{MgO}} = 3\text{ nm})]_{15}$ multilayer was grown by pulsed laser deposition (PLD) on a Corning® glass substrate. The preparation procedure and the structural, electrical and magnetic characterization of the whole series of the multilayers was reported in detail elsewhere [6–8]. Electrical resistance and MR measurements were carried out using the four-point method at room temperature under hydrostatic pressure ranging from ambient pressure to 7 kbar using a standard Cu-Be cell. MR measurements were performed in transversal (magnetic field in the substrate plane but perpendicular to the electrical current) and perpendicular (magnetic field perpendicular to the substrate plane and to the electrical current) configurations.

Figure 1 shows the electrical resistivity dependence with pressure at room temperature. The resistivity decreases approximately linearly with pressure indicating an increase in conduction via tunneling [11]. The value of the coefficient $(1/\rho_0)d\rho/dP = -3.9 \times 10^{-2} \text{ kbar}^{-1}$, being ρ_0 the resistivity at ambient pressure, is much higher than that obtained in metals and also significantly higher than the reported value in $\text{Co}_{52}\text{Al}_{20}\text{O}_{28}$ granular films, $(1/\rho_0)d\rho/dP = -1.87 \times 10^{-2} \text{ kbar}^{-1}$ [12]. This result implies that the electronic state of the sample with $t_{\text{Fe}} = 0.70\text{ nm}$ is near the percolation threshold. This is in good

Fig. 2. Room temperature magnetoresistance for $[\text{Fe}(t_{\text{Fe}} = 0.7\text{ nm})/\text{MgO}(t_{\text{MgO}} = 3\text{ nm})]_{15}$ DMIM at ambient pressure (open circles) and 7 kbar (full circles) for (a) transversal and (b) perpendicular configurations.

Table 1. Isothermal bulk modulus, K_T , at room temperature for Fe, Co, MgO and Al_2O_3 .

	K_T (kbar) at T=295 K	Reference
Fe	~ 1700	[14]
Co	~ 1800	[14]
MgO	~ 1600	[15]
Al_2O_3	~ 2500	[15]

agreement with our previous results, where we found that the electrical percolation threshold of Fe/MgO multilayers grown by PLD is $t_{\text{Fe}} \approx 0.80\text{ nm}$ [8].

In Fig. 2 we present the TMR ratios measured at room temperature in transversal and perpendicular geometries at ambient pressure and under an applied pressure, $P = 7\text{ kbar}$, and magnetic field up to 3 kOe. The measurements show an increase of the TMR ratio in the transversal configuration from 0.6% to 1.1% at ambient pressure and $P = 7\text{ kbar}$, respectively (see Fig. 2(a)). The opposite behaviour has been found by *Kaji et al.* in Co-Al-O granular materials, where a decrease of TMR ratio with the applied pressure was observed at room temperature [12,13], and explained as a result of the dependence of the mean intergranular distance of the Fe particles, s , with P .

The important physical quantity that dominates the resistance of a material that is compressed under an applied hydrostatic pressure at constant temperature is the isothermal bulk modulus (K_T) defined by the expression:

$$K_T = -V(\partial P/\partial V)_T$$

K_T values for Fe, Co, MgO and Al_2O_3 are listed in Table 1. In the case of Co-Al-O granular films the bulk modulus of the insulating matrix (Al_2O_3) is much higher than the K_T of Co. As a consequence of increasing P the oxide matrix is less compressed than the Co grains, yielding enhancement of s and a decrease of the TMR ratio. The $[\text{Fe}(t_{\text{Fe}} = 0.7\text{ nm})/\text{MgO}(t_{\text{MgO}} = 3\text{ nm})]_{15}$ multilayer ($K_{T,\text{Fe}} > K_{T,\text{MgO}}$) is the opposite case where increasing

P will result in more compressed MgO matrix than the Fe grains, leading to a decrease of s and an increase of the TMR ratio according to Mitani's model [10]. Additional measurements in metal-insulator granular systems are needed to confirm this hypothesis.

The absolute value of TMR that was measured for perpendicular configuration (Fig. 2(b)) is slightly lower than that obtained for the transversal one. This behavior has been already observed in other Fe/MgO DMIMs at ambient pressure [6] and is probably caused both by the shape anisotropy of the sample and possibly oblate spheroid shape of the magnetic particles. The higher difference observed at $P = 7$ kbar could be caused by an enhancement of the oblate spheroid shape originated by a preferential deformation of the Fe granules in the film plane caused by the applied pressure.

III. CONCLUSION

We have determined the dependence of the resistivity of an $[\text{Fe}(t_{\text{Fe}} = 0.7 \text{ nm})/\text{MgO}(t_{\text{MgO}} = 3 \text{ nm})]_{15}$ multilayer on the applied hydrostatic pressure. The obtained value of the $(1/\rho_0)d\rho/dP$ coefficient is higher than that observed in metals and other metal-insulator granular systems, indicating that our DMIM is near the percolation threshold.

The TMR ratio strongly increases under an applied hydrostatic pressure of 7 kbar at room temperature. To our knowledge it is the first time that this behavior has been observed in a metal-insulator granular system. We explain this result assuming a decrease of the tunnel barrier width (mean separation between adjacent Fe grains) with pressure.

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