Enhancement of Tunneling Magnetoresistance Through a Magnetic Barrier of Granular Fe-Pb-O System

Jen-Hwa Hsu, Ching-Ray Chang, and Yi-Hong Huang

Abstract—A large value of magnetoresistance ratio has been observed in granular Fe-Pb-O solids. The large MR value is associated with the appearance of magnetic Pb ferrite encapsulated on the surface of iron granules. Spin-dependent tunneling picture can qualitatively describe the enhancement of magnetoresistance ratio. Calculated results based on the spin filter effect of the Pb-ferrite layer are reasonably in agreement with the experimental data of TMR enhancement in Fe-Pb-O solids.

Index Terms—Tunneling magnetoresistance, spin-dependent, granular film, spin polarization.

I. INTRODUCTION

TUNNELING magnetoresistance effect (TMR) draws much attention for its fascinating transport properties and industrial applications [1]. Usually the change of resistance with magnetic field in a tunneling junction is much higher than that in multilayers, and the power consumption of the junction is less but with higher sensitivity. Therefore, TMR is particularly utilized in nanostructured memory device [2] for the advantage of heat reduction, e.g., magnetic random access memory. In spin-polarized tunneling studies, polarization of the tunneling current is found to originate from the Zeeman splitting of the density of states of spin-up and spin-down conduction electrons in the ferromagnetic electrode. Since the TMR ratio is expected to be proportional to the product of the spin polarization of both ferromagnets [3], many efforts have been devoted to find ferromagnetic materials with large value of polarization. However, the spin polarization of natural ferromagnetic material at room temperature is at most 0.44 [4] and thus expected TMR ratio is limited to 32%.

After the observation of significant magnetoresistance of a trilayer junction with an insulating spacer [5], some granular systems with insulating matrix also exhibit TMR effect [6]. Electron tunneling between ferromagnetic granules, the electron spin polarization within ferromagnetic granules as well as the relative orientation of the magnetization of different granules determines the value of TMR [7]. Usually a nonmagnetic oxide is the choice for the insulating matrix of the granular

system [6]. Recently some theoretical calculations suggest that a spin-dependent barrier can enhance the polarization and thus the TMR value [8], [9]. Also a granular Fe-Pb-O system has been reported to possess a large TMR value as long as a magnetic phase of Pb ferrite has been formed; however the reason for the significant enhancement of TMR is not well understood. In this work we study the possible mechanism which causes the enhancement of magnetoresistance ratio through the magnetic barrier in the granular Fe-Pb-O system. The observed enhancement of TMR in granular Fe-Pb-O films with the formation of magnetic Pb ferrite, at least a factor of ten, can be explained in the framework of spin-filter effect from a magnetic barrier.

II. EXPERIMENT

The Fe-Pb-O granular films were obtained from a new and unique method. The first step of preparation was to make metastable Fe-Pb alloy films that were fabricated by magnetron rf sputtering with a composite target made from Fe and Pb powder. The substrate of kapton foil was cooled by LN₂. Afterwards, films were annealed in the oven with flowing Ar gas for one hour. The oxidation also occurred in this process with residual oxygen in the oven. The detailed description of the preparation procedures has been reported elsewhere [10]. To compare with other Fe-Pb-O granular films, Fe(PbO) granular films were prepared with a composite target of Fe as well as PbO and by rf sputtering technique [11].

The film structure was determined from X-ray powder diffraction and the film morphology was investigated by transmission electron microscopy (TEM). A conventional four-probe method has been used to obtain the resistivity of the granular film between 5 K and 300 K. The magnetoresistance ratio, $[\rho(0) - \rho(H)]/\rho(0)$, was measured at room temperature with a maximum applied field of 0.9 Tesla. Under this field, the samples are not completely saturated.

III. THEORY

It has been suggested that a magnetic insulator between the ferromagnetic metals will enhance the spin polarization of tunneling electrons [8], [9]. For the ferromagnet-magnetic insulator-nonmagnetic insulator-ferromagnet (FM-MI-I-FM) junction, the ferromagnetic barrier acts as a spin-filter, and it selects the passing polarized electrons. Because of the presence of magnetic barrier, the attenuation lengths, $1/\kappa_{\uparrow}$ and

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Fig. 1. TEM picture of Fe-Pb-O granular film.

$1/\kappa_{\downarrow}$, are quite different for spin-up and spin-down electrons, respectively. Therefore the effective spin polarization through the spin-filter barrier, P_{eff} , is [8]

$$P_{\text{eff}} = \frac{k_{\uparrow'} - k_{\downarrow'} \frac{\kappa_{\uparrow}^2 - k_{\uparrow'}^2}{\kappa_{\uparrow}^2 + k_{\uparrow'}^2}}{k_{\uparrow'} + k_{\downarrow'} \frac{\kappa_{\uparrow}^2 + k_{\uparrow'}^2}{\kappa_{\uparrow}^2 + k_{\uparrow'}^2}}. \quad (1)$$

where $k_{\uparrow'}$, $k_{\downarrow'}$ are for the right ferromagnetic materials. For the nonmagnetic barrier, P_{eff} reduces to Slonczewski's results [4]. For $U_{\uparrow} > U_{\downarrow}$, $\Delta\rho/\rho$ can be expressed as

$$\frac{\Delta\rho}{\rho} = \frac{2P_{\text{eff}}}{1 + P_{\text{eff}}}. \quad (2)$$

It should be noted that the TMR ratio of a conventional spin tunneling junction is proportional to the square of the spin polarization [3] while TMR ratio of a spin-filter junction is only proportional to the spin polarization. Since spin polarization is always less than 1, the spin-filter junction has a much larger TMR ratio than the conventional spin-tunneling junction. Enhancement of the TMR ratio from the spin-filter effect with a ferromagnetic barrier is quite notable, especially when the spin polarization is small. With a FM-MI-I-MI-FM structure, TMR ratio will approach 100% from the spin-filter effect.

IV. RESULTS AND DISCUSSIONS

For films annealed at 300°C, the resistivity of the films decreases with decreasing temperature, typical behavior of a metal. However, when the annealing temperature is above the melting temperature of Pb, which is 327°C, the resistivity of the films exhibits a tunneling behavior. The temperature dependence of the resistivity follows the tunneling behavior, i.e., $\ln\rho \propto T^{-1/2}$. The TEM picture of the granular films annealed at 400°C is shown in Fig. 1. High resolution TEM suggests some granules are core/shell structure where the dark core region is surrounded with other materials of light shell region (Fig. 1). The size of the granules is about 100–200 Å, the area of shell is estimated to be about 40% of the granules and the radius of Fe-core is about 70–150 Å. The distance between granules is roughly 50 Å. In Fig. 2 the X-ray diffraction pattern points out that in the shell a nonmetallic magnetic phase has formed, which is identified to be $\text{PbO} \cdot 3\text{Fe}_2\text{O}_3$ and the granules are embedded in the PbO matrix.

Fig. 2. X-ray diffraction patterns for annealed Fe-Pb-O granular films. (*): $\text{PbO} \cdot 3\text{Fe}_2\text{O}_3$; (γ): $\text{PbO} \cdot 2\text{Fe}_2\text{O}_3$; and (δ): $\text{PbO} \cdot 1/2\text{Fe}_2\text{O}_3$.

Fig. 3. (a) A schematic diagram of the electron tunneling between granules within PbO matrix. The granules are core/shell structure: Fe core surrounded with the magnetic Pb ferrite shell. (b) Mechanism of the enhancement of polarization through a spin-dependent potential of magnetic barrier. Spin-up electrons and spin-down electrons experience different spin-dependent potential barrier $U_{\uparrow}$ and $U_{\downarrow}$ respectively. The dash line is the barrier height for non magnetic PbO insulator. Only one kind of spin electrons emerges from the ferrite shells if the spin-filter effect is strong enough.

The TMR value is about 10% under $H = 0.9$ Tesla at room temperature with $x = 0.4$ for films annealed at 400°C. It drops to 4.5% for films with $x = 0.2$. It is apparent that the TMR ratio is reduced with decreasing amount of $\text{PbO} \cdot 3\text{Fe}_2\text{O}_3$. Furthermore, the TMR ratio becomes less than 1% for Fe(PbO) films where iron granules without $\text{PbO} \cdot 3\text{Fe}_2\text{O}_3$ ferrite shell are embedded in PbO matrix [11].

Since both TEM and X-ray data indicate the possibility that Fe cores are encapsulated within the Pb ferrite shell, the possible schematic structure of the Fe core/ Pb ferrite shell granules embedded in the PbO matrix is shown in Fig. 3(a). With the magnetic barrier, spin-up electron and spin-down

electrons experience different tunneling height of the barrier [Fig. 3(b)] and their attenuation lengths are quite different. From (1), the spin polarization of the electron from the Fe core will be greatly enhanced as the electrons emerge from the Pb ferrite shell. If the tunneling mechanism of the electrons is described as in Fig. 3, the TMR ratio should approach 100% with the FM-MI-I-FM structure. However, some of the Fe granules are not surrounded with the Pb-ferrite shell and most of the electrons are still not tunneling through the magnetic shells on both sides. The experimental results should be described with FM-MI-I-FM structure as in (2) and the TMR ratio only depends on P_{eff} , instead of square of P_{eff} in the Julliere model. No matter what kind of metals next to the ferromagnetic barrier, the electrons emerging from the ferromagnetic barrier have about the same polarization. Instead of the predicted 32% TMR ratio of the Fe-I-Fe junction from Julliere's model [3], we expect a 61% TMR ratio in Fe-MI-I-Fe spin-filter structure from (2). Therefore, the ultimate enhancement of TMR ratio is 61% for Fe granules with 0.44 polarization. However, the predicted TMR ratio of 61% is much larger than the observed 10% in granular Fe-Pb-O film. The main reason for this discrepancy is that the encapsulation of most Fe cores by magnetic Pb ferrite shell is not complete. Other possibilities are the reduction of effective polarization from the barrier height and thickness [4] and the spin-flip during the tunneling [12]. Comparing with Julliere model [3] and (2), the magnetic barrier enhances the TMR value to a factor of the inverse of effective polarization. Since the effective polarization usually is around the order of magnitude of 0.1 [4], it is easy to understand that the TMR value of Fe granules encapsulated with the magnetic Pb ferrite will be 10% if the TMR value of Fe(PbO) granular films is 1% as observed in the experiment.

We have studied Fe-Pb-O granular solids and found that an order of magnitude enhancement of TMR is observed with the formation of magnetic Pb ferrite on the surface of Fe granules. The enhancement of TMR may possibly result from the

spin-filter effect through a magnetic barrier. We also found that it is reasonable for TMR to increase a factor of ten with a spin-filter barrier. Further research on the spin-filter tunneling junction is necessary for possible huge TMR ratio and potential applications in memory device.

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