Inverse and normal tunneling magnetoresistance effects in FeCoGd/FeCo/AlO/FeCo multilayers

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Abstract. FeCoGd/FeCo(t)/AlO/FeCo magnetic tunnel junctions were fabricated with a very thin FeCo layer inserted between the FeCoGd layer and the AlO barrier. At room temperature, inverse tunneling magnetoresistance (TMR) effect and normal TMR effect were observed simultaneously in the low and high magnetic field regions, respectively. The absolute values of both inverse and normal TMR ratios, the saturation magnetic field corresponding to the normal TMR effect have similar dependence on \( t \). Transmission electron microscopy studies indicate that a thin granular film is likely formed between the FeCoGd layer and the AlO barrier, which may result in the normal TMR effect at high magnetic fields.

1. Introduction
Magnetic tunnel junctions (MTJs) have attracted much interest in recent years due to widespread applications in memory and sensor devices [1]. MTJs are often composed of sandwiches of two ferromagnetic (FM) layers separated by thin insulating layers. Besides layered structure MTJs, sizeable tunnelling magnetoresistance (TMR) effect can also be achieved in insulating granular films that consist of magnetic metallic particles dispersed in insulating matrices [2]. Generally, the saturation magnetic field of TMR effect is much higher in granular films than that in MTJs. TMR effect in both MTJs and insulating granular films can be qualitatively described within the framework of Julliere’s model [3].

Although there have been extensive investigations on MTJs and insulating granular films separately, up to now, little has been done on the TMR effect in the hybrid structures combining them together. In this presented work, FeCoGd/FeCo/AlO/FeCo MTJs have been fabricated with a very thin FeCo layer inserted between the FeCoGd layer and the Al-oxide (AlO) barrier. Inverse and normal TMR effects can be observed in the low and high magnetic field regions, respectively. A thin granular film, i.e. small FeCo-rich particles embedded in Gd-oxide matrix, is likely formed between the bottom electrode and the barrier, which may contribute to the normal TMR effect at high magnetic fields.

2. Experimental
MTJs of Ta(5)/FeCoGd(28.7)/FeCo(t)/AlO/FeCo(17)/Ta(5) were fabricated on glass substrates by magnetron sputtering [4]. The unit in the above parentheses is nm. FeCo, FeCoGd and AlO represent...
Fe$_{50}$Co$_{50}$, (FeCo)$_{55}$Gd$_{45}$, and Al-oxide, respectively. After the growth of the Ta buffer layer and the FeCoGd layer, a very thin FeCo layer with its nominal thickness of $t$ was deposited. The sample was exposed in the atmosphere for about half an hour and then put back to the chamber again. After the chamber was re-pumped to a high vacuum with pressure lower than $7 \times 10^{-6}$ Pa, a 1.2 nm thick Al layer was deposited and oxidized by oxygen plasma subsequently to form the barrier.

The MR effect was measured by using a standard dc four-point method under a magnetic field ($H$) up to 14 kOe, which was applied in the film plane and parallel to the direction of the induction field as well. When the resistance increases/decreases with the increase of $H$, inverse/normal MR effect occurs. Magnetic responses of the samples were characterized by a vibrating sample magnetometer (VSM). All measurements were performed at room temperature.

3. Results and Discussion

Figure 1 shows the magnetic hysteresis loops ($M$-$H$ loops) and the MR curves ($R$-$H$ loops) for two representative samples with $t = 0$ ((a) and (b)) and $t = 1.8$ nm ((c) and (d)). Obviously, besides the inverse MR effect at low magnetic fields, normal MR effect can also be observed in the high magnetic field region. The inverse MR effect is due to the fact that the atomic ratio of Gd in the FeCoGd is much higher than the compensation composition and thus the FeCoGd is a ‘Gd dominant RE-TM alloy’ at room temperature [4]. In the high magnetic field region, although $R$ decreases significantly with the increase of $H$, no obvious magnetic moment change can be observed in the $M$-$H$ loops. In addition, in comparison to the easily saturated $M$-$H$ loops, the $R$-$H$ loops are more difficult to be saturated, especially for the sample with $t = 0$.

![Figure 1](image-url)

**Figure 1.** The $M$-$H$ loops and $R$-$H$ loops of the MTJs with $t = 0$ ((a) and (b)) and $t = 1.8$ nm ((c) and (d)). The corresponding low field $M$-$H$ loops and the $R$-$H$ loops are shown in insets (a) and (c).

In order to explain above experimental results, it is essential to get deep insight into the microstructures of the MTJs. Transmission electron microscopy (TEM) with element content analysis has been performed on the sample with $t = 0$. Figures 2(a) and 2(b) show the survey image and the corresponding atom profile along the film growth direction (denoted by the green line), respectively. The left survey image shows clear multilayer structure and fairly good quality of the AlO barrier. From the right atom profile, it is evident that the content of O element in the FeCoGd layer is much larger than that in the FeCo layer, especially near the FeCoGd/AlO interface.

As a rare-earth element, Gd is much easier to be oxidized than transition metals such as Fe and Co. Therefore, at the FeCoGd/AlO interface, after some of the Gd atoms in the FeCoGd layer were oxidized, the left Fe and Co atoms may be aggregated as FeCo-rich particles and isolated by the Gd-oxide matrix. In other words, a very thin granular film, composed of FeCo-rich particles dispersed in the insulating Gd-oxide matrix, is likely to be produced near the FeCoGd/AlO interface. It is noted that in Fig. 1(b), the junction resistance is still far beyond saturation although $H$ has been applied to 14
kOe. This means that those FeCo-rich particles are very small and thus their magnetic moments are hard to rotate towards the direction of $H$. In addition, those small magnetic particles mainly contribute to the TMR effect and almost do not affect the total magnetic moment because they only stay at the FeCoGd/AlO interface and their total amount is quite small, which is verified by the quite large difference between the $M$-$H$ and $R$-$H$ loops shown in Fig.1 (a) and Fig. 1(b), respectively.

![Figure 2. TEM survey image (a) and the corresponding atom profile (b) along the green line for the MTJ sample of FeCoGd (28.6 nm) /AlO/ FeCo(17 nm).](image)

If a discontinuous FeCo thin film with its nominal thickness $t$ is purposely inserted between the FeCoGd and the AlO barrier, followed by the same oxidation procedure described above, a similar granular thin film can also be formed naturally. However, the average size of the FeCo-rich particles may change with $t$ and the magnetic and transport properties of these MTJs will change accordingly. In order to distinguish the TMR effect at both low and high magnetic field regions, two kinds of TMR ratios are defined as $TMR_1 = (R_{min} - R_3)/R_{min}$ and $TMR_2 = (R_1 - R_2)/R_2$, where $R_{min}$ denotes the minimum resistance when $H$ is between 0 and 400 Oe; $R_1$ and $R_2$ represent the resistances when $H$ is 400 Oe and 14 kOe, respectively. Therefore, $TMR_1$ is negative and reflects the low field TMR effect, while $TMR_2$ is positive and reflects the high field TMR effect.

The $t$ dependence of $TMR_1$ and $TMR_2$ is shown in Fig.3 (a). The absolute values of both $TMR_1$ and $TMR_2$ decrease first and then increase with increasing $t$. At almost the same place, i.e. $t \sim 1.5$ nm, they both reach the minimal values. For the presented MTJ samples, the junction resistance decreases with increasing $t$ when $t$ is below 1.5 nm (not shown here). This may be due to the fact that the quality of the barrier became worse after a discontinuous FeCo thin film was deposited on an amorphous FeCoGd layer. Therefore, the lowered junction resistance leads to the decrease of both $TMR_1$ and $TMR_2$ with increasing $t$ first [5]. However, when $t$ is further increased, the size of FeCo-rich particles will become larger and larger and eventually the percolation happens and the FeCo layer becomes continuous. Because the spin polarization of FeCo is much larger than that of FeCoGd [4], the absolute values of $TMR_2$ will increase quickly after the FeCo layer becomes continuous ($t \sim 1.5$ nm). However, even if $t$ is very large ($>1.5$ nm), some small FeCo-rich particles may still reside at the ferromagnet/barrier interface, which causes the existence of the normal TMR effect. As for $TMR_2$, the absolute value increases with $t$ when $t$ is larger than 1.5 nm. One possible reason is that the content of FeCo in the FeCo-rich particle may increase with $t$, which results in the increase of the spin polarization of the FeCo-rich particle and the subsequent enhancement of the TMR effect at high magnetic fields. In order to confirm this, further investigation needs to be done.

In order to compare the saturation field of the $R$-$H$ loop in the high magnetic field region effectively, a parameter $H_s$ is introduced. $H_s$ is defined as the value of $H$ where the maximal value appears in the $dR/dH \sim H$ curve when $H$ is between -14 kOe and -400 Oe, as depicted in the inset of Fig. 3 (b). Obviously, $H_s$ indicates where the resistance drops most abruptly and it can reflect the
magnitude of the saturation magnetic field to some extent. Very interestingly, the \( t \) dependence of \( H_M \) is very similar to that of \( TMR_1 \) and \( TMR_2 \). The variation of \( H_M \) along \( t \) can also be understood by the mean size change of the FeCo-rich particles with \( t \). When \( t \) is very small, very small ferromagnetic particles are produced, leading to superparamagnetic behavior and the fact that their magnetic moments are hard to rotate towards the direction of \( H \). Therefore, \( H_M \) is very large. With the increase of \( t \), the mean size of the FM particles increases, which causes their magnetic moments easier to rotate towards \( H \) and \( H_M \) is decreased consequently. In the mean time, the average magnetization \( (M_1) \) of those FM particles increases. Because the magnetization of FeCo \( (M_2) \) in FeCoGd is anti-parallel to that of Gd and there exists a strong ferromagnetic coupling between \( M_1 \) and \( M_2 \), \( M_1 \) is readily to align with \( M_2 \) (or away from \( H \)) and this will have a tendency to increase \( H_M \). When \( t \) is increased further and even beyond the percolation threshold, the latter contribution to \( H_M \) becomes dominant and \( H_M \) begins to increase with \( t \). Therefore, it is reasonable that in the vicinity of the percolation threshold, i.e. \( t \sim 1.5 \) nm, a minimum of \( H_M \) appears, as shown in Fig. 3 (b).

![Figure 3. Dependence of \( TMR_1 \), \( TMR_2 \) and \( H_M \) on \( t \) for the MTJ samples. Inset of (b) shows the definition of \( H_M \).](image)

4. Conclusions
Inverse and normal TMR effects were observed simultaneously in the MTJs of FeCoGd/FeCo/AlO/FeCo at room temperature. Inverse TMR effect at low \( H \) is due to the fact that the FeCoGd is a ‘Gd dominant RE-TM alloy’. Normal TMR effect at high \( H \) is mainly attributed to a very thin granular film resided near the FeCoGd/AlO interface, which is likely formed by FeCo-rich particles embedded in Gd-oxide matrix. The absolute values of \( TMR_1 \), \( TMR_2 \) and \( H_M \) have the similar dependence on \( t \), which is due to the mean size of the FeCo-rich particles changing with \( t \).

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