Transactions of the Materials Research Society of Japan 29[4] 1551-1554 (2004)

Improvement of Insulating Characteristics for TMR Granular Multilayers Using Combined Insulators, MgF_2 and Al_2O_3.

M.Nakazumi[1], K.Ono[2], H.Yanagihara[1], T.Koyano[3] and E.Kita[1]
[1]Institute of Applied Physics, University of Tsukuba, Tsukuba, Ibaraki 305-8573,Japan
Fax: 81-29-853-6110, e-mail: nakazumi@riko.tsukuba.ac.jp
[2] ULVAC Tsukuba research center, Toyosato, Tsukuba, Ibaraki, Japan
[3] Cryogenics center, University of Tsukuba, Tsukuba, Ibaraki 305-8573, Japan

In order to improve insulating properties in granular tunneling materials, the combined insulator (CI) layer composed of MgF_2 and Al_2O_3 are examined where MgF_2 helps to grow small Fe particles and Al_2O_3 acquires good insulation. Granular multilayers were produced with alternating deposition. A unit layer consists of Fe and insulating layers which are formed by sequential deposition of MgF_2 / Al_2O_3 / MgF_2. The unit was repeated by 3 times that is enough to prevent an electrical breakdown. Granular TMR with current perpendicular to plane (CPP) arrangements was examined in the sample. Enhancement of the resistivity due to the Coulomb blockade was seen at low temperature only in the combined insulator sample and was not seen in an Al_2O_3 insulator sample. TMR ratio of 7 % was found in the CI sample at low temperature. These facts indicate that Fe particles favorable for TMR are formed only in the combined insulator sample and the sample has larger charging energy than the Al_2O_3 sample. It is concluded that the combined insulator improves the insulating characteristics keeping granularity in Fe layers.

Key words: granular TMR, Fe, MgF_2, Al_2O_3, resistivity

1. Introduction

Novel magnetotransport phenomena such as giant magnetoresistance (GMR)[1] and tunneling magneto-resistance (TMR) have been discovered in magnetic nanostructures. GMR has been intensively studied from both basic and application viewpoints following its discovery in 1988. A pioneer study on TMR was reported about 26 years ago, however, the research has been accelerated by the discovery of large-magnitude TMR by Miyazaki and Tezuka[2] in 1995. In the case of trilayer tunneling devices, barrier materials are mainly reported for Al_2O_3. Control of sample fabrication of the insulator layer has been an important issue in development of these types of devices.

Large TMR in a granular material where fine particles of magnetic metal are embedded in insulating substances was found in Co-Al_2O_3[3] systems. Granular materials were intensively investigated in the 1970s and the basic mechanism of electrical conduction was established[4]. This granular type TMR has also been reported mainly Al_2O_3 as an insulator and the mechanism of tunneling conduction has been precisely investigated[5]. Other insulating materials such as MgO, SiO_2[6], rare-earth oxides[7] and MgF_2[8-10] have been examined.

The sample fabrication techniques of granular materials are mostly co-deposition and co-sputtering to realize the dispersion of metallic grains in an insulating matrix. These techniques suit to form metal particles, however it has disadvantages that the diameter of metal particles often distributes and the numbers of conduction passes cannot be controlled because of randomly embedded metallic particles. Another technique is alternating deposition. If the metal layer thickness is controlled sufficiently thin, Fe layers grow with an islands structure. This technique have advantages that it is possible to control the number of conduction pass due to the number of times of laminating and the diameter of metal particles distribute little. We have been investigating multilayers of ferromagnetic metals and insulating materials and found that island structures were realized in Fe/MgO[11] and Fe/MgF_2[12] multilayers. These multilayers have clear interfaces because Fe and these insulators don't mix at the interfaces[13]. We also reported on Fe, Co / MgF_2 of electrical conduction, which are measured on specimens with current-in-plane (CIP)[9,14] arrangement.

The measurement with current-perpendicular-to-plane (CPP) alignment for granular TMR has two advantages. The first, it is possible to control the number of conduction pass by controlling the number of laminating cycles. At the second, the distance between particles can be controlled by changing thickness of an insulator layer. Additionally very interesting result was reported by using CPP nano geometry. In a measurement of nanometer sized tunneling junction and one laminating ferromagnetic layer, the interplay of spin dependent tunneling and single electron tunneling is represented by Tunnel magnetoresistance oscillations associated with Coulomb staircase[15,16].

Thus the CPP measurement has been revealing interesting property such as nano granular tunneling. However, in the case of MgF_2 system, measurements were difficult. We have obtained preliminary results indicating that MgF_2 layers assist the growth of small metal particles due to the roughness of MgF_2 surfaces. An electrical breakdown often occurs as the thickness decreased in CPP alignment since MgF_2

insulator layers are likely to have pinholes. On the contrary, Al_2O_3 is generally used as a barrier with good insulating property. The reason is that pinholes hardly exist since Al_2O_3 is in an amorphous state. It is noted that Al_2O_3 may not suit to form small particles due to the flat surface.

In this paper we fabricated a combined insulating layer composed of MgF_2 and Al_2O_3, which possibly has both advantages of two insulators. We call this sample combined insulator (CI) sample. Optimum compositions were investigated to obtain good insulating properties. Transport and magnetic properties were measured on the sample and results were discussed with that for the sample composed of Fe and Al_2O_3 as an insulator.

2. Experimental procedure

Granular multilayers were grown with alternating deposition using two e-guns. The structure of samples is shown in Fig. 1. MgF_2 layers were deposited before and after the deposition of the Fe layers. To keep good insulation, Al_2O_3 layer were made between two adjacent MgF_2 layers. It is natural to consider that MgF_2 layer in this combined insulator assists to grow small Fe particles and Al_2O_3 layer contribute to be acquire good insulation. A nominal unit layer consists of Fe particles and insulating layers which are formed by alternating deposition of MgF_2 / Al_2O_3 / MgF_2.

The unit was repeated by N times and $N \geqq 3$ is enough to prevent an electrical breakdown. The samples were expressed as {Fe (15 Å) / MgF_2 (3 Å) / Al_2O_3 (4 Å) / MgF_2 (3 Å)} N=3 (CI sample) and {Fe (15 Å) / Al_2O_3 (10 Å) (Al_2O_3 sample) and were measured in CPP alignment. The thickness of insulating layer is kept at 10 Å, which was most suitable value to obtain large MR and fairly good reproducibility.

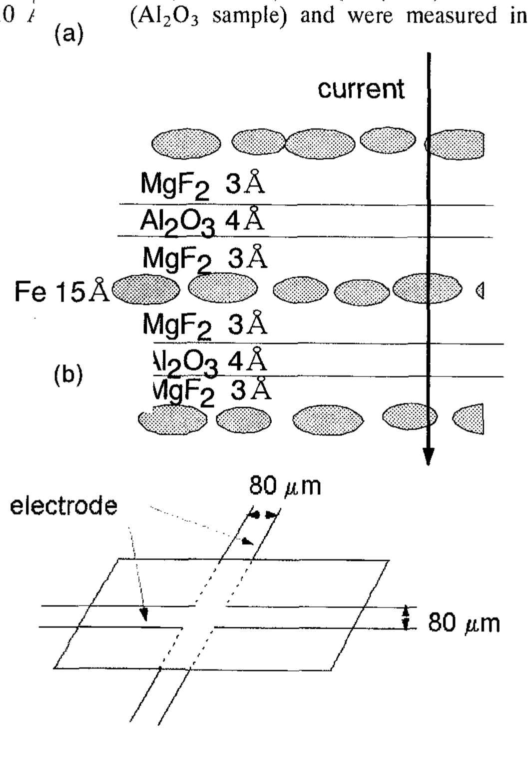

Fig. 1. (a) shows the sample structure composed of combined insulator layers and (b) shows the junction area with cross pattern.

The junction area was 80x80 μm^2 by cross pattern using metal masks as shown in Fig. 1. (b). The resistance measurements were carried out with a two terminal method because of rather high resistance measurement. Magnetization curves were measured by a SQUID.

3. Results and discussion

Figure 2 shows the temperature dependence of resistivity on a logarithmic scale plotted against $T^{-1/2}$. Proportionality of log (ρ) and $T^{-1/2}$ was confirmed for both CI and Al_2O_3 samples and this tendency implies that the electrical conduction is based on the granular tunneling mechanism discussed by Abeles et al[4]. They represented the resistivity in granular systems as follows,

$$\rho = \rho_0 \exp\left(2\sqrt{\frac{C}{k_B T}}\right) \quad \text{and} \qquad (1)$$

$$\log \rho = 2\sqrt{C/k_B} \cdot T^{-1/2} + const \quad . \qquad (2)$$

Here ρ, k_B and C are the resistivity, Boltzman constant and tunnel activation energy, respectively. The values of C/k_B were estimated using eq. (2) from the slope of the plots in Fig, 2. The tunnel activation energy is defined as $C=\chi s Ec$, where, χ is expressed as $\sqrt{2m\phi/\eta^2}$. Here m and ϕ are electron mass and effective barrier height. The charging energy, Ec, is represented by

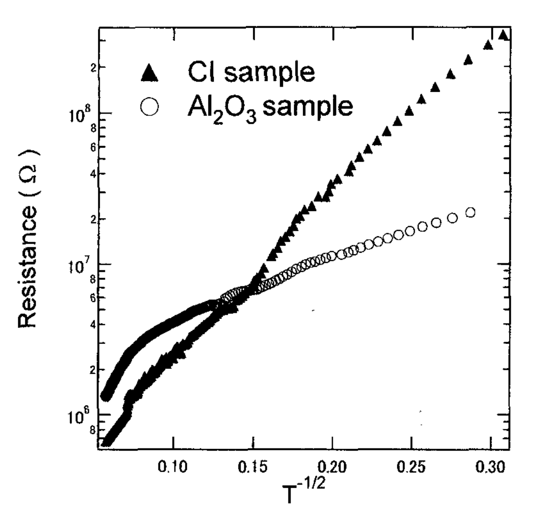

Fig. 2. Logarithmic plots of the resistivity vs. $T^{-1/2}$ for the CI sample and Al_2O_3 sample Date for the CI sample is plotted with solid symbols and open symbols are used for Al_2O_3 samples.

$$Ec = \frac{e^2}{d} \frac{2}{\varepsilon\{1+(d/2\varepsilon)\}}, \quad (3)$$

Here e, ε, d and s are the electron charge, the dielectric constant of MgF_2 (=5.2), the diameter of metal particles and the separation between particles, respectively. Thus Ec can be experimentally estimated using eqs. (2)-(3) and those for the CI and Al_2O_3 samples had 12.7 mV and 1.8 mV. Thus Ec for the CI sample was about 7 times larger than that for Al_2O_3 sample. Large Ec suggested that the particles come under the large influence of Coulomb Blockade, additionally it can explain the enhancement of resistivity at low temperature.

The diameter of Fe particles was evaluated by using Ec via eq. (3) to be 12 nm and 33 nm on the CI and Al_2O_3 samples, respectively. This result confirms that Fe particles formed in the CI sample were smaller than that in the Al_2O_3 samples.

In the Al_2O_3 sample, the slope in Fig. 2 is different between the regions of high and low temperature. Probably because the diameters of Fe particles are distributed, at low temperature electrons selectively tunnel large particles that have small charging energy. This suggests that the diameter of Fe particles in Al_2O_3 sample distributed more than that in CI sample.

This interpretation also explains that magnetization measurement in Figs. 3 and 4. In Fig. 3 although both samples don't show coercivity at room temperature, at 5K the coercivity in CI sample and Al_2O_3 sample are 600 Oe and 450 Oe. Figure 4 shows the temperature dependence of magnetization with field cool at 100 Oe (FC) and with zero-field-cool (ZFC). Blocking temperature at which the thermal fluctuation is quenched in CI and Al_2O_3 samples is 110K and 230K, respectively. These results suggest that the magnetic property of the CI sample is more superparamagnetic like compared with Al_2O_3 sample, probably due to the diameter of Fe particles is smaller.

Since large magnetic moment is required for SQUID measurements, N was increased from 3 to 10 then the compositions of both samples differ from that for the resistivity measurement. Supposing that the difference has little effect on the magnetic properties, the size of Fe particles was estimated with the simple equation concerning superparamagnetism. When blocking temperature is low, the diameter of metal particles is relatively small. Therefore, the result of the lower blocking temperature in the CI sample than that of Al_2O_3 sample suggests that MgF_2 help the structure of Fe layers island-like. We have observed Fe particles in MgF_2 layer by transmission electron microscopy and confirmed that the diameter of Fe particles is about 10nm[13]. The result supports that the estimation of the foregoing agrees with the result of TEM.

MR curves at 10K are shown in Fig. 5. MR ratios in the CI and Al_2O_3 samples at 10K were about 7 % and 1.5 %, respectively. Those at room temperature decreased into 4% and 1% for the CI and Al_2O_3 samples, respectively. MR ratio of CI sample is always larger than that of Al_2O_3 at any temperature. It can be

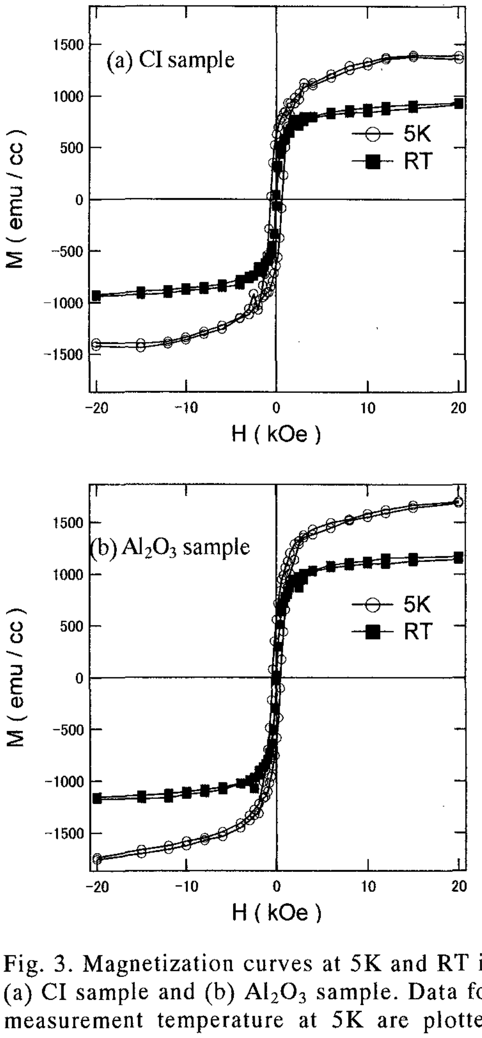

Fig. 3. Magnetization curves at 5K and RT in (a) CI sample and (b) Al_2O_3 sample. Data for measurement temperature at 5K are plotted with solid symbols and open symbols are used for Al_2O_3 sample.

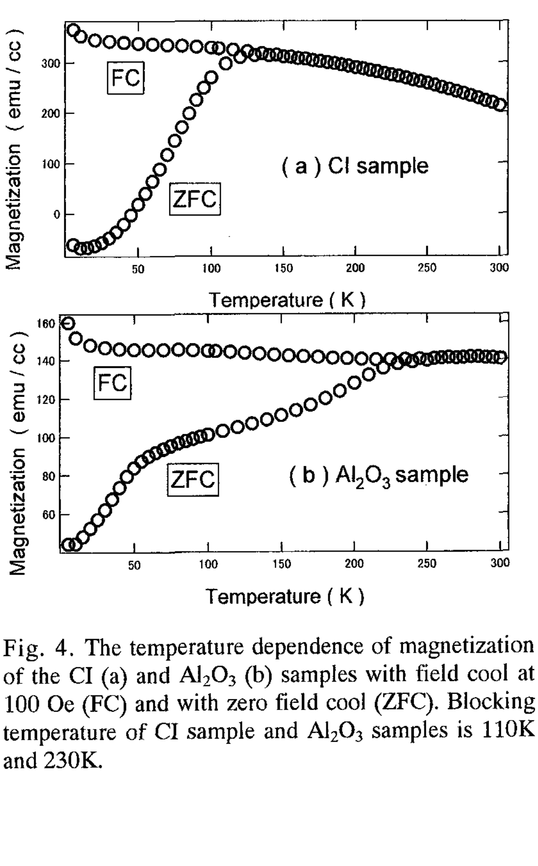

Fig. 4. The temperature dependence of magnetization of the CI (a) and Al_2O_3 (b) samples with field cool at 100 Oe (FC) and with zero field cool (ZFC). Blocking temperature of CI sample and Al_2O_3 samples is 110K and 230K.

explained by the difference in the structure of Fe particles. In the result mentioned above, we found that MgF_2 layers in CI sample help to form small Fe particles, however, Fe particles in Al_2O_3 sample form successive-layer-like, and magnetic moments at zero field don't orient randomly due to strong exchange between Fe particles. Hence the resistance in Al_2O_3 sample at zero field doesn't have the amplitude expected for the ideal random distribution of magnetization.

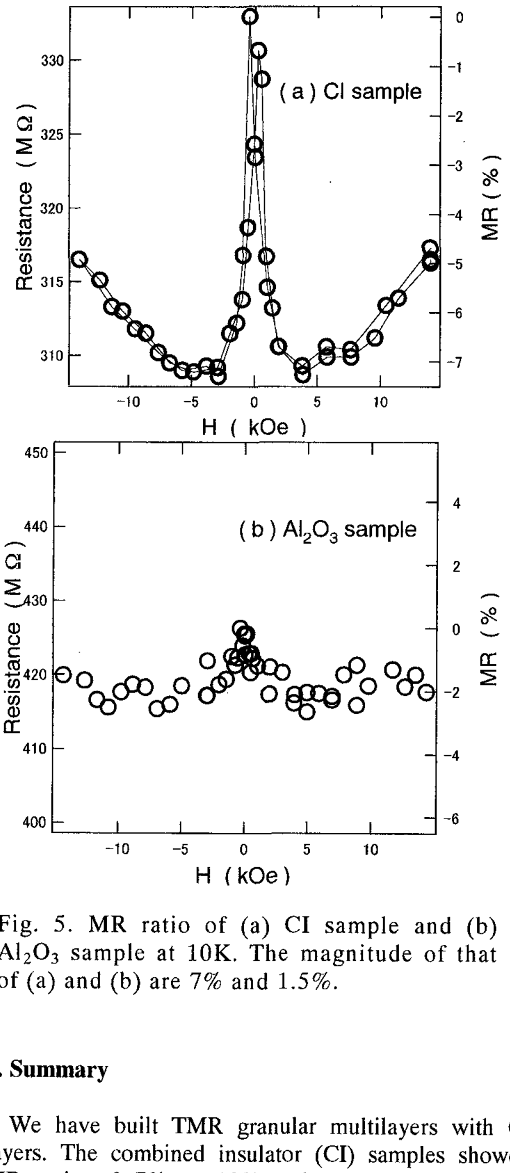

Fig. 5. MR ratio of (a) CI sample and (b) Al_2O_3 sample at 10K. The magnitude of that of (a) and (b) are 7% and 1.5%.

4. Summary

We have built TMR granular multilayers with CI layers. The combined insulator (CI) samples showed MR ratio of 7% at 10K and we got fairly good reproducibility in the resistance measurements, meaning that the probability of electrical break down wasn't so high. In the CI sample, small Fe particles are preferentially formed on MgF_2 layers and have large charging energy. Because of this small particles, combined insulator sample showed larger MR ratio than Al_2O_3 samples.

5. Acknowledgements

We would like to thank Professor Yoshizaki and Dr. Ikeda, for sample fabrication using the deposition system installed at the Cryogenic Center, University of Tsukuba.

Reference

[1] M. N. Baibich, J. M. Broto, A. Fert, N. Van Dau, F. Petroff, P. Etienne, G. Greuzet, A. Friedrich and J. Chazelas: Phys. Rev. Lett. **61**, 2472 (1988).
[2] T. Miyazaki and N. Tezuka: J. Magn. Magn. Mater. **139**, L231 (1995).
[3] H. Fujimori, S. Mitani and S. Ohnuma: Mater. Sci. Eng. B **31**, 291 (1995).
[4] B. Abeles, P. Sheng, M. D. Coutts and Y. Arie: Adv. Phys. **24**, 407 (1975).
[5] S. Mitani, H. Fujimori and S. Ohnuma: J. Magn. Magn. Mater. **165**, 141 (1997).
[6] S. Honda T. Okada, M. Nawate and M. Tokumoto: Phys. Rev. B **56**, 14566 (1997).
[7] N. Kobayashi, S. Ohnuma, T. Masumoto and H. Fujimori: Mater. Trans., JIM **39**, 679 (1998).
[8] T. Furubayashi and I. Nakatani: J. Appl. Phys. **79**, 6258 (1996).
[9] C. Yasui, J. Umehara, K. Ono, T. Matsumoto, H. Yanagihara and E. Kita: J. Magn. Soc. Jpn. **23**, 1345(1999) [in Japanese].
[10] N. Kobayashi, S. Ohnuma, T. Masumoto and H. Fujimori: J. Magn. Soc. Jpn. **24**, 571 (2000) [in Japanese].
[11] T. Koyano, Y. Kuroiwa, E. Kita, N. Saegusa, K. Ohshima and A. Tasaki: J. Appl. Phys. **64**, 5763 (1988).
[12] M. Nishikawa, K. Ono, E. Kita, H. Yanagihara, T. Erata and A. Tasaki: J. Magn. Magn. Mater. **238**, 91 (2002).
[13] S.Hine, T.Shinjo, T.Takada: J. Phys. Soc. Jpn **47**, 767 (1979)
[14] K.Ono, C.Yasui, H.Ynagihara, T.Koyano, K.Ota and E. Kita: Jpn. J.Appl. Phys. **41**, 97 (2002).
[15] K. Nakajima, Y. Saito, S. Nakamura, and K.Inomata: IEEE.Trans. Magn.**36**, 2806 (2000).
[16] K. Yakushiji, S. Mitani, K. Takanashi and H. Fujimori: J.Phys.D **35** , 2422 (2002)

(Received October 8, 2003; Accepted March 1, 2004)