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Nanoscale Microstructure and Magnetic Transport in AlN/Co/AlN/Co... Discontinuous Multilayers

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Microstructure and magnetic transport phenomina in rf sputtered AlN/Co type ten- layered discontinuous films of nanoscaled [AlN(3 nm)/Co(t nm)]...$_{10}$ with t_{Co}=1.0~2.0 nm have been investigated. The microstructure and tunneling magnetic resistance of the samples are strongly dependent on the thickness of Co layer. Negative tunneling magneto-resistance due to the spin-dependent transport has been observed along the current-in-plane configuration in the samples having the Co layers below 1.6 nm thick. When the thickness of Co layer was less than 1.2 nm, randomly oriented granular Co particles were completely isolated and embedded in amorphous AlN matrix, and the films showed the superparamagnetic behavior with a high MR value of $\Delta\rho/\rho_0$=1.8%. As t_{Co} increases, a transition from the regime of co-existence of superparamagnetic and ferromagnetic behaviors to ferromagnetic behavior was observed. Tunneling barrier called "decay length for tunneling" for the films having the thickness of Co layer from 1.4 to 1. 6 nm was measured to be ranged from 0.004 to 0.021 Å^{-1}.

Key words : Spin-dependent transport, Tunnel magnetoresistance, Discontinuous multilayers

1. Introduction

Negative tunneling magneto-resistance (TMR) originated from spin-dependent transport was found in Co/Ge/Fe trilayers by Julliere *et al.* [1]. According to his model, TMR is determined solely by tunnel conductance which is proportional to the cosine of the angle between the magnetization directions of two ferromagnetic electrodes. However, it has been argued that the barrier and magnetic material may determine the TMR effect as well [2] and its variation follows the relation of $\exp(2\sqrt{C/k_BT})$, where C is an activation energy concerning on the barrier material [3, 4]. Intensive studies of the spin-dependent transport in granular thin films have been stimulated by the discovery of TMR and GMR effects. There have been a lot of publications on TMR granular films recently [5-9], where the magnetic particles embedded in the insulate matrix, and in the most cases, SiO_2 or Al_2O_3 based films have been comprehensively investigated. It is well known that Al_2O_3 is utilized successfully as the barrier material in the magnetic tunneling junction (MTJ). As compared with Al_2O_3, AlN as a barrier in the tunneling junction has been investigated previously as well because AlN has the better physical properties such as low band gap and high thermal conductivity [10]. However no relative work on AlN based granular film has been reported.

Discontinuous metal/insulator films are a new class of granular MR material, which is a multilayer consisting of alternative deposits of discontinuous magnetic and insulator layer and the latter exists as an insulating matrix. It was found that the magnetic layer becomes discontinuous below about 2.5 nm thick in the metal/insulator multilayers. The resultant structure is usually referred to discontinuous metal-insulator-metal (DMIM) films. The DMIM films are easier to fabricate than a MTJ and also more robust since the magnetic particles are well protected by the insulator. Initial work on DMIM films such as CoFe/HfO_2 [11], Co/SiO_2 [12] and CoFe/Al_2O_3 [13] show significantly higher MR values, and the interests are focused on the tunneling MR applications at room temperature.

In this paper, we report that a negative magnetoresistance was observed in the Co/AlN discontinuous multilayers. This magnetoresistance was observed along the current-in plane (CIP) geometry and varied with the thickness of the magnetic metallic layer, Co.

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2. Experimental Procedure

Bi-layered [AlN(3 nm)/Co(t nm)] films were alternatively repeated five times on a glass substrate with rf-sputtering acting alternatively on two separate AlN and Co targets. The insulating layers of AlN were directly sputtered from the AlN target. The base pressure was 2×10^{-6} Torr and a high-purity argon gas under 10 mTorr was used for the sputtering. All the samples consist of discontinuous 10 layers of AlN/Co/AlN/Co ...with the top AlN layer for protective role. No symptom of the formation of Al_2O_3 was observed from the discontinuous films. The nominal thickness of Co layers, t_{Co}, was controlled from 1.0 to 2.0 nm with the AlN layer fixed at 3nm in each sample. The thickness of the sputtered films was measured by α - step (Tencor Instrument, Inc.). Microstructure of the sputtered films was characterized by a field emission transmission electron microscopy(FE-TEM) with a nanoscale resolvable resolution. The magneto-resistance of the samples was measured at room temperature by a conventional four-probe method using indium-painted contacts applying the maximum magnetic field of 4T along the in-plane direction of films. The magnetic measurements were carried out with an Alternating Gradient Magnetometer (AGM) with the magnetic field of 1.4 T applied in the film plane.

3. Results and Discussions

3.1. Microstructure and magnetic properties

Fig. 1(a), (b), and (c) shows the TEM plan view image for the discontinuous films of $t_{Co} = 1.2$, 1.4 and 1.6 nm, respectively, in the formation of AlN (3 nm)/t_{Co}/AlN (3 nm) tri-layers. In Fig. 1(a), the randomly oriented granular Co particles of 2~5 nm size in the dark contrast are completely embedded in the AlN matrix in light contrast. Since those particles are completely isolated by the insulator AlN matrix. Since they are not magnetically coupled and the film exhibits a superparamagnetic behavior causing a tunneling type MR (see below). The aspect and arrangements of the Co nanoparticles are drastically changed when the thickness of the discontinuous Co layer is increased to 1.4 nm as shown in (b). The Co islands tend

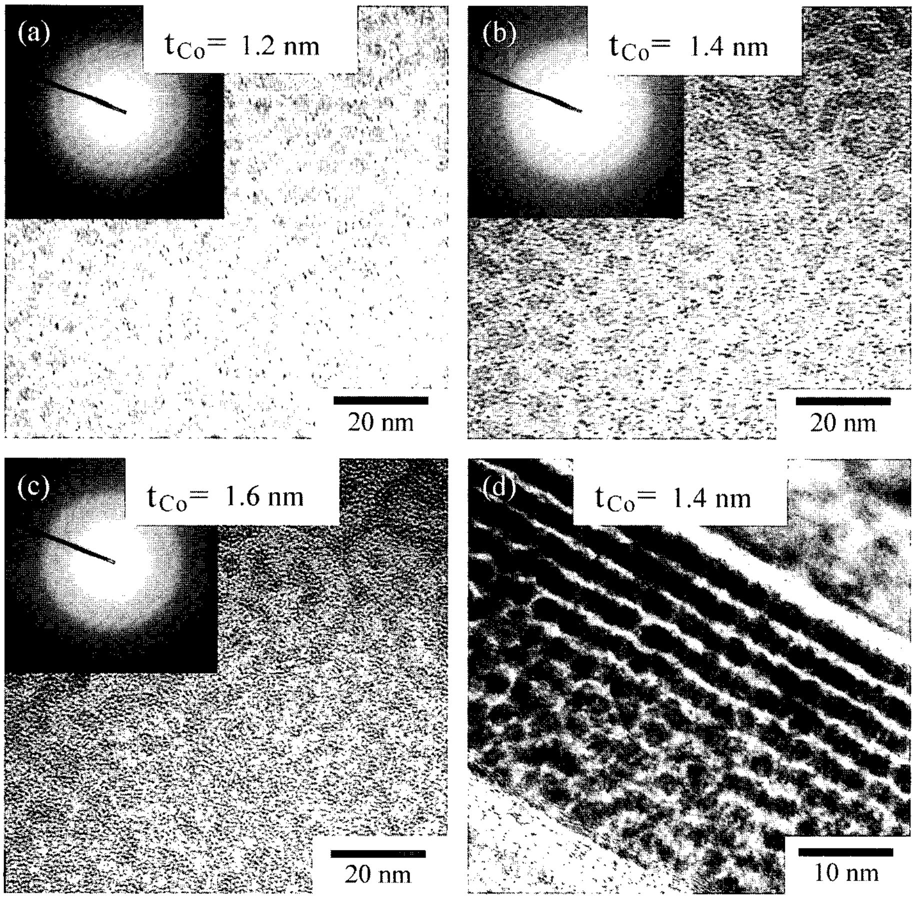

Fig. 1. TEM image of the AlN (3 nm)/Co (t nm)/AlN (3 nm) tri-layers for t_{Co}=1.2 nm (1(a)), 1.4 nm (1(b)), and 1.6 nm (1(c)), respectively. A cross section of $[AlN(3\ nm)/Co(1.4\ nm)]_{10}/AlN(3\ nm)$ is shown in Fig. 1(d). All the EDS specify that the AlN matrix is amorphous.

to agglomerate each other and some of them form a chain-like long particle. Fig. 1(c) shows the image for the films of t_{Co} = 1.6 nm. It clearly shows that many of the Co particles eventually appear as curved long chain-like clusters with a width of approximately 4~6 nm. The electron diffraction pattern (EDP) shown at the inset of each (a), (b), and (c) suggest that the AlN matrix is amorphous. High-resolution TEM micrograph of the cross section of the discontinuous films of ten-layered AlN(3 nm)/Co(1.4 nm)..$_{10}$ are illustrated in Fig. 1(d). It is obvious that the cobalt layers are composed of particles in chain to be formed as discontinuous undulated composite films of multilayer. However, the Co particles are shown to be mostly insulated by AlN matrix.

Fig. 2 shows the hysteresis loop of the films with t_{Co}= 1.2, 1.6, and 2.0 nm, respectively. It can be seen that the behavior depends strongly on the thickness of Co layer. When the thickness of Co layers is 1.2 nm, the magnetization curve exhibits a superparamagnetic nature as indicated in Fig. 2(a), because the granular particles are small enough to be isolated as shown in Fig. 1(a). By increasing the thickness of Co layers from 1.2 to 1.6 nm, a clear transition from superparamagnetic to ferromagnetic behavior appears corresponding to so-called magnetic percolation in a granular system [13, 14]. As t_{Co} is equal to 1.6 nm, the magnetization behavior of the discontinuous film can be understood to show a character having coexistence of both superparamagnetic and ferromagnetic contents, which is similar to the result of Honda *et al.* found in the Co/Ag granular films [15, 16]. The coexistence of both behaviors is possible in magnetic nano clusters embedded in a non-magnetic matrix. In a disordered granular material, the magnetic state below magnetic percolation threshold presents, in general, a mixture of two coexisting phases: a dipolar coupled ferromagnetic phase and residual superparamagnetic phase unless the magnetic layer is very thin. When t_{Co}=2.0 nm, the sample basically shows the typical ferromagnetic behavior.

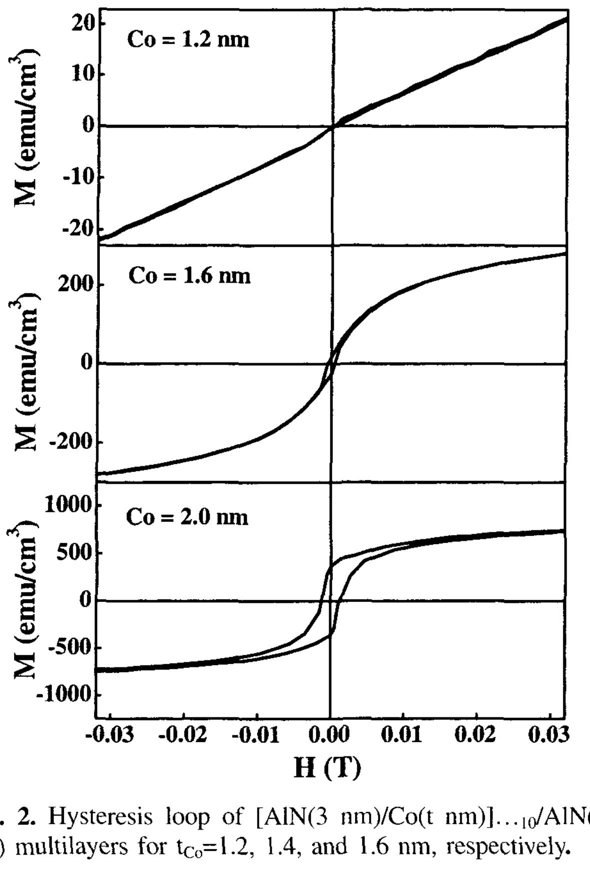

Fig. 2. Hysteresis loop of [AlN(3 nm)/Co(t nm)]...$_{10}$/AlN(3 nm) multilayers for t_{Co}=1.2, 1.4, and 1.6 nm, respectively.

3.2. Magneto-transport properties

Fig. 3 shows the MR ratio, $\Delta\rho/\rho_0$, plotted versus the applied magnetic field for the DMIM films of different thickness of the Co layer. The MR ratio is defined as $\Delta\rho/\rho_0 = (\rho_H - \rho_0)/\rho_0$, where ρ_0 and ρ_H are the resistivity of sample at H = 0 and at an applied field, respectively. With decreasing the thickness of Co layer, t_{Co}, MR significantly increases and MR_{max} is 1.8% at t_{Co} = 1.0 nm. However, since our series of thickness was not extended to below t_{Co} = 1.0 nm, this figure should not yet be considered to be the optimum thickness for the highest MR amplitude in this system. The inset in Fig. 3 indicates the magnetoresistance dependence of the Co layer thickness, and a

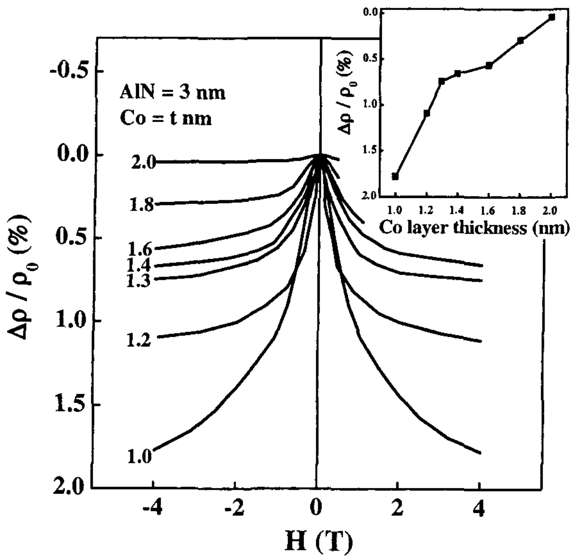

Fig. 3. The curve of $\Delta\rho/\rho_0$ as the function of the applied field H for the samples of [AlN(3 nm)/Co(t nm)]...$_{10}$/AlN(3 nm) multilayers with t_{Co}=1.0, 1.2, 1.3, 1.4, 1.6, 1.8 and 2.0 nm, respectively. The inset shows $\Delta\rho/\rho_0$ vs. the thickness of the Co layers.

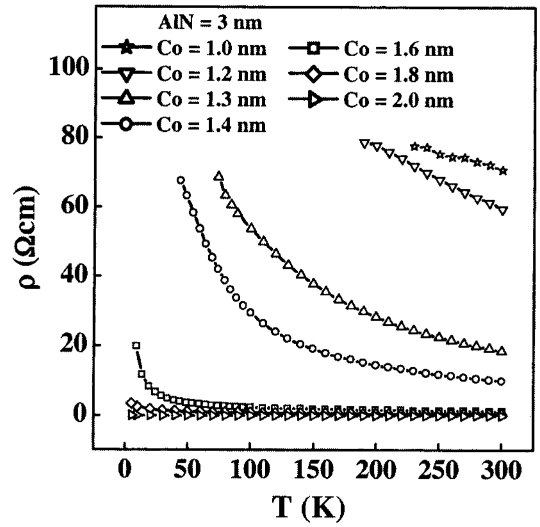

Fig. 4. r as the function of T for the samples of [AlN(3 nm)/Co(t nm)]...$_{10}$/AlN(3 nm) multilayers having different Co-layer thickness.

concave curve was measured. Since TMR is essentially determined by the magnetic correlation between the neighboring granules, the kink at $t_{Co} \approx 1.4$ nm on the curve of MR versus t_{Co} can be related to the certain transitions from superparamagnetic to ferromagnetic, which is an unambiguous indication of cooperative magnetic behavior among the disconnected ferromagnetic granules.

The room temperature electrical resistivity increases sharply with the decrease of Co layer thickness, from 70 Ωcm for $t_{Co} = 1.0$ nm to ≈0.0034 Ωcm at $t_{Co} = 2.0$ nm. The drastic increase of resistivity suggests a change from dielectric to metallic states and this can be understood as follow. For a thin Co layer, the Co grains become electrically isolated by the insulating amorphous AlN matrix resulting in tunnel-type conductivity. For a thick Co layer, the major of Co granules are connected with each other, a metallic conductance can arise. This can be confirmed by the temperature dependence of the resistivity. Fig. 4 shows the temperature dependence of the resistivity ρ(T) at different Co-layer thickness. Also the resistivity dependence on the thickness of Co layer is manifested in Fig. 4 for the comparison of $1.0\,\text{nm} \leq t_{Co} \leq 2.0\,\text{nm}$. Resistivity depends strongly on the temperature and shows a negative temperature coefficient, this negative temperature coefficient associated with the relation between lnr versus $T^{-1/2}$ plotted in Fig. 5 suggest a tunneling resistance behavior.

At a low temperature, due to the Coulomb Blockade effect [17, 18], a tunneling takes place between the largest metallic particles and causes a rapid increase of MR. At a higher temperature, the thermal activation is sufficient to overcome the charging energy and more conduction paths become available, thus generating smaller TMR [3]. This TMR temperature behavior obeys the theory, $\rho \propto \exp(2\sqrt{C/k_BT})$, given by Sheng *et al.* [3, 4], where C is the activation energy proportional to both the tunnel-barrier thickness and the charging energy of the metallic particles. The solid lines in Fig. 5 are the curves fitting to our experimental data. As can be seen for the sample with t_{Co} = 1.4 nm, the data agree well with the theory given by Sheng *et al.* until to the limit of low electric field [3]. The curve of ln(ρ) vs. $T^{-1/2}$ shown in the inset gives the activation energy C ≈ 12.8 meV for Co = 1.4 nm and 2.3 meV for Co = 1.6 nm, respectively. As expected the films of thin Co layer clearly show a higher activation energy for tunneling which is caused by the isolated particles embedded in AlN matrix as shown in Fig. 1(b).

This temperature dependence was predicted by a model in which charge transport proceeds via thermally activated and/or bias assisted tunneling processes [3, 19] for a system with a distribution of particle sizes and separations. The energy barrier is the Coulomb charging energy required in creating a pair of charged particles, which is necessary for conduction. The ratio of the particle separation to the particle diameter, s/d, in the film of t_{Co}=1.4 nm in our study, for instance, can induce the decay length for tunneling "χ" quoting the Sheng-Abeles relation, $C = 8\chi e^2(s/d)^2/(1+2\ s/d)$, with the obtained activation energy C=12.8 meV from the Fig. 5 [19]. The decay length for tunneling "χ" ranging over 0.012 to 0.021 Å^{-1} was obtained by using the s=3~3.4 and d=3~3.1 nm measured from the sample directly. And also the "χ" of the films having

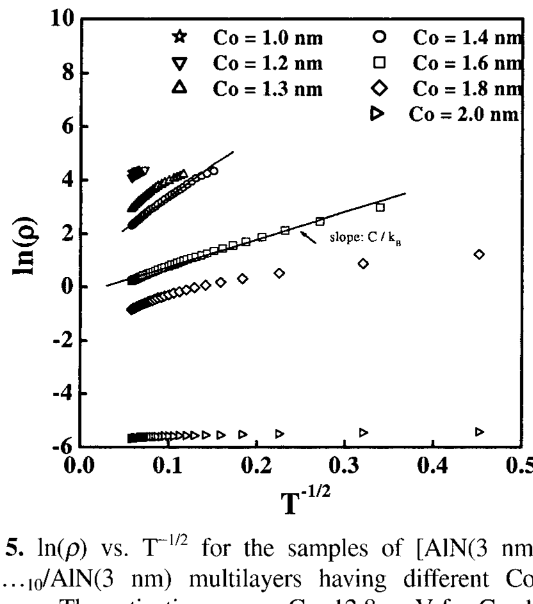

Fig. 5. ln(ρ) vs. $T^{-1/2}$ for the samples of [AlN(3 nm)/Co(t nm)]...$_{10}$/AlN(3 nm) multilayers having different Co-layer thickness. The activation energy C ≈ 12.8 meV for Co=1.4 nm and 2.3 meV for Co=1.6 nm, respectively.

t_{Co}=1.6 nm sample was measured to range over 0.004 to 0.012 $Å^{-1}$ using the S=3~3.4 nm and d=4~4.2 nm, respectively. The obtained estimation of χ=0.004~0.021 $Å^{-1}$ seems to be quite small compared with the theoretically estimated value reported from the reference quoted above [19]. The discrepancy is explained by taking into account of the nano scale particles in Fig. 1. By increasing the thickness of discontinuous Co layer, the Co particles tend to agglomerate becoming a chained long shape. Accordingly the particles may have regions where the separation, s, is smallest. For the films of t_{Co}=1.6 nm, the trend becomes more prominent. Therefore the measured "χ" is quite small. The smaller value of the "decay length for tunneling" of the films having t_{Co}=1.6 nm may indicate that other effect is superimposed on the tunneling effect. As discussed by S. Honda *et al.* in Fe-SiO_2 granular film [6], this phenomenon was contributed by a leak conductivity through the interconnected granules as shown in Fig. 1(c). With increasing the thickness of Co layer, this deviation became more dominant. The more Co clusters were shown to connect with each other, and seemed to increase the fraction of metallic conductivity.

4. Conclusion

A study of AlN(3 nm)/Co(t nm)/AlN(3 nm) type discontinuous MTJ films was reported and a negative magnetoresistance due to spin-dependent transport has been observed in the CIP configuration. The maximum value of $\Delta\rho/\rho_0$=1.8% was obtained for t_{Co}=1.0 nm. The microstructure is strongly dependent on the thickness of Co layer. The t_{Co} below than 1.2 nm, randomly oriented granular Co particles were completely isolated by the amorphous AlN matrix. It responded a superparamagnetic behavior and shows a large MR magnitude originated from the spin-dependence electron tunneling effect. As increasing the Co layer thickness, some particles were connected each other and formed a chain-like shape. This change in nano scale shape induced a magnetic transformation from superparamagnetic to ferromagnetic behavior resulting in a reduced resistivity ratio. Tunneling barrier called "decay length for tunneling" for the films having the thickness of Co layer from 1.4 to 1.6 nm was measured to be ranged from 0.004 to 0.021 $Å^{-1}$.

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