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Microstructure and electric property of MgO/Fe/MgO tri-layer films forming a nano-granular system

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ABSTRACT

For tunneling magnetoresistance (TMR) devices using ferromagnetic nano particle films, the size, dispersion and number of nano particles are important factors. Relating to this, single layered Fe films (thickness: t = 0.5 – 10.0 nm) sandwiched between two MgO (2 nm thick) layers were fabricated by molecular beam epitaxy. By depositing at T_s = RT (room temperature), the Fe layer had an isolated island structure for less than 1 nm thick. Correspondingly, the negative magnetoresistance effect was observed, which is characteristic of TMR. By increasing T_s, the resistivity and the magnetoresistance (MR) ratio was increased. In this study, it was found that the optimal parameters for the growth of nano particle MgO/Fe/MgO based films are t = 0.5 – 1.0 nm and T_s = RT – 120 °C.

1. Introduction

Magnetic tunnel junctions (MTJs) composed of insulating layers sandwiched between ferromagnetic layers are well known to show the tunneling magnetoresistance (TMR) effect. Because of their large TMR effect, many application works have been carried out, in addition to basic researches, to develop new non-volatile memories (MRAM: magnetic random access memory), read heads for hard disk drives and other spintronic devices [1]. Ferromagnetic (FM) metal-insulator granular films which are studied in this work consist of FM nano particles dispersed in insulating matrices. They form MTJ networks within the films and show TMR effect [2–9]. This type of films has a potential for spinctonic devices as usual MTJs though the TMR effect reported so far was not large enough. Recently, by introducing the single-electron tunneling (SET) effects such as the Coulomb blockade (CB), an enhancement of TMR was reported in Co–Al–O granular films [4]. Relating to this, Ono et al. prepared a three terminal FM-SET transistor Ni/NiO/Co/NiO/Ni by lithography and observed an enhancement of TMR by controlling the gate voltage [10]. By using the gate terminal, the modulation of electronic states of nano particles and of TMR can be effectively controlled. In order to realize FM-SET transistor using nano-granular films, usage of device with current-in-plane (CIP) geometry is required. However, there have been few reports on such investigation [11,12].

The FM metal-insulator granular films are usually prepared by the co-deposition of metals and insulators [4]. Another approach consists in the alternate deposition of a multi-layered structure where metallic nano particle layers and insulator layers stack alternatively [13]. Considering the current flow in the film plane, there are a number of current paths along the thickness. In order to make it the minimum value (i.e. one); a single metallic particle layer covered by insulator must be a candidate. By reducing the number of nano particles, the SET properties may easily be detected and the gate voltage can be effectively applied to the particles. Such a structure was used for measurements with the current-perpendicular-to-plane (CPP) geometry [4,5] but not for CIP geometry [14,15].

Therefore, in this report, we fabricated tri-layer films where one thin Fe nano particle layer was sandwiched between two thin MgO layers. The fundamental electric properties of these "single-layered granular films" were investigated in the film plane, CIP geometry. The influences of the Fe layer thickness and the deposition temperature were studied. SET properties such as CB and CO are briefly demonstrated.

2. Experimental

The films consist of a tri-layer of MgO (2 nm)/Fe (t nm)/MgO (2 nm) structure. Films were deposited onto 16 nm-thick SiN membranes and glass plates for transmission electron microscopy (TEM) and electric measurements, respectively. They were prepared by alternating deposition of Fe (99.95%) and MgO (99.9%) using a molecular beam epitaxy system at base pressure less than

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10^{-8} Pa. These materials were evaporated using two electron beam guns. The average film thickness (t) was monitored by using a quartz thickness monitor placed just near the substrate. For both materials, the deposition rate was 0.01 nm/s. A typical TEM image of such a film is shown in Fig. 1a, where t = 1 nm. A Fe layer showing a dark line contrast is clearly recognized to be sandwiched between two bright layers corresponding to MgO. The Fe layer is composed of nano particles as indicated in Fig. 1b, where the Fe lattice fringe is recognized in a nano region. From electron diffraction patterns, the Fe layer was analyzed to have the bcc structure (Fig. 1c).

In the present work, the influence of the layer thickness of Fe (t) and of the substrate temperature during deposition (T_s) was investigated. First a set of films with different thickness t = 0.5–10.0 nm were prepared at T_s = RT (room temperature). Successively a second set of films were grown keeping thickness fixed at 1.0 nm and changing temperature T_s from RT to 200 °C. The microstructure was studied by means of TEM using a JEM-200CX microscope operated at 200 kV. The electric measurements were performed using the conventional 2- or 4-terminal DC method between RT and 5 K. The size of the tri-layer film was typically 9 mm in length and 2 mm in width. The distance between two voltage terminals was about 2 mm. For the characterization of the films, resistivity-to-temperature (ρT) as well as magnetoresistance (MR) properties were investigated.

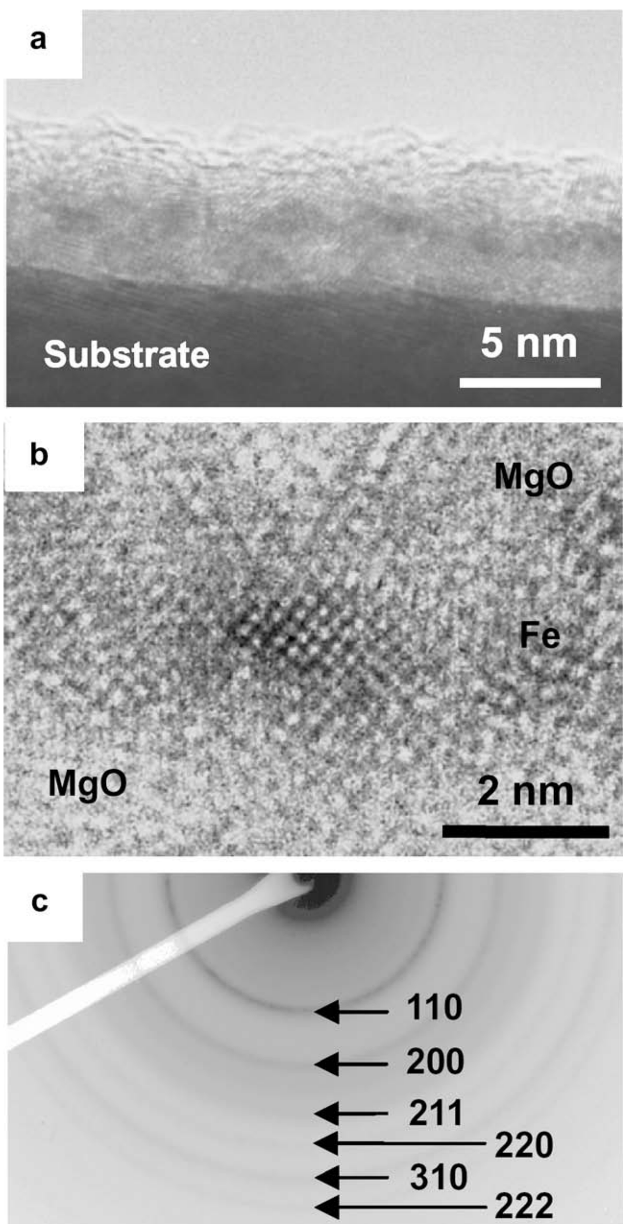

Fig. 1. Typical TEM data of the tri-layer film where the Fe layer is 1 nm thick. (a), (b) Cross sectional TEM images (substrate: Au) and (c) a plane view electron diffraction pattern indicating the bcc structure of Fe (substrate: SiN).

For the study of SET properties, miniaturized devices have to be investigated. They were fabricated by electron beam lithography followed by a lift off process. At first, source and drain electrodes made of Au/Cr (50 nm thick) were formed on SiO_2 (500 nm)/Si wafers where Si was used as the back-gate terminal. Afterwards, MgO (50 nm)/MgO (2 nm)/Fe (1 nm)/MgO (2 nm) pattern was deposited at RT, where the thick MgO is the cap layer to prevent oxidation during the lithography process. The channel size of the current flow was 200–400 nm in length and 400 nm in width. The electric measurements were done at RT and 8 K in a cryogenic probing station (Nagase, GRAIL-20-305-6-LV) using a semiconductor parameter analyzer (Agilent 4156C). The setup allowed to measure the current-to-voltage (I–V) curve between the source and drain terminals and the dependence of the drain current on the gate voltage V_g.

3. Results and discussion

3.1. Dependence on Fe thickness

In Fig. 2a, TEM images of the films prepared at RT with various Fe average thicknesses are compared. The image with t = 10.0 nm indicates that the obtained Fe layer was a continuous film. The dark contrast corresponds to the grains satisfying the Bragg's reflection condition while the brighter part to the grains out of this condition. The grains became small by decreasing Fe thickness. However, the films look continuous down to t = 2.0 nm. By further reducing Fe thickness, the microstructure changed when t = 1.0 nm where the Fe grains were separated from each other forming a nano particle film. The size of these particles became small when t was less than 1.0 nm. The size distributions in these films were measured for 100 particles (or grains) randomly selected from each image. The average particle (or grain) size in the film plane is shown in Fig. 2b as a function of t. For well separated nano particle films (i.e. $t \leqslant 1$ nm), the particle diameter was less than d = 3 nm with which the SET effect may appear even at RT.

In Fig. 3, ρT curves are compared to each other. For the film with t = 10.0 nm, the temperature coefficient was positive showing a metallic conduction. By decreasing t down to 2 nm, the temperature coefficient was still positive while the resistivity increased. When the Fe thickness was t = 1.0 nm, the negative temperature coefficient was recognized. This indicates that the type of current flow changed to tunneling conduction. By further reduction in t, this tendency became clear and the resistivity drastically increased. The threshold thickness to realize the tunnel conduction was between 1.0 and 2.0 nm. A similar threshold value has been reported at the first stage of Al deposition [16]. The resistivity around which the conduction type was changed from metallic to tunneling conductions was $\rho \sim 10^{-3}$ Ωcm. This corresponds to the values reported for Co–Al_2O_3 and Fe–MgO granular films in three dimensions [3,8].

The MR curves of these films measured at RT are presented in Fig. 3(b) where the magnetic field (H) was applied parallel to the sense current for resistance measurements. For the films with thick Fe layers ($t \geqslant 2$ nm), the resistivity around H = 0 Oe showed the minimum value. This is the anisotropic magnetoresistance gener-