Preparation of Magnetic Tunnel Junctions Using Fe_3O_4

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Recently, magnetic tunnel junction (MTJ) has attracted many magnetic researchers due to its large potential for various spintronics devices. Most researchers believe that highly spin polarized conduction electron can significantly improve the MR effect. Since Fe_3O_4 is considered a half-metal, the MTJ using Fe_3O_4 is expected to show very large MR effect. We tried well-defined Fe_3O_4 preparation using in-situ XPS and RHEED analysis methods. It is found that the chemical properties of Fe_3O_4 surfaces are very sensitive to the barrier layer material. Though lithography micro-fabrication technique was applied for a fairly smooth Fe_3O_4 surface sample, no clear tunneling I-V behavior was observed.

Key words: magnetite, half-metal, tunnel junction, XPS

1. INTRODUCTION

Many researchers have become to pay great attention to magnetic tunnel junction (MTJ) because of the large potential for spintronics devices. Especially, the application to magnetic random access memory (MRAM) is expected. However, further large magneto-resistance (MR) is necessary for the MRAM application. Larger spin polarization of magnetic electrodes increase MR due to Julliere's simple model. [1] Half-metallic materials providing 100% spin polarized conduction electron are very attractive for MTJ.

Magnetite (Fe_3O_4) is a member of the half-metals judged from the band structure calculation [2] and shows various desirable characteristics such as chemical stability, high Curie temperature (858 K), non-toxicity. Though several MTJ studies using Fe_3O_4 have been reported, no expected large MR effect has been observed yet. [3,4] We have already tried to prepare MTJs composed of Fe_3O_4 electrode and aluminum oxide barrier. [5] The barrier layer formation method is popular for metallic TMJs. Metal aluminum is first deposited and then oxidized by the exposure to oxygen atmosphere. Measured non-linear current-voltage (I-V) curve indicates actual tunneling phenomenon as shown in Fig.1 (a). Calculated barrier height and width by Simons' equation are 4.6 eV and 1.5 nm respectively. Both calculated values confirm the insulative and uniform barrier layer formation. No significant MR effect above noise level is shown in Fig.1 (b) in spite of the good tunnel barrier formation mentioned above.

Tunnel magneto-resistance (TMR) effect is considered very sensitive to the barrier-magnetic electrode interface properties. We employed in-situ x-ray photoelectron spectrometry (XPS) to examine the chemical condition of the sample surfaces. No metal Fe nor Fe^{3+} satellite between $2p_{3/2}$ and $2p_{1/2}$ peaks observed in the as-prepared fresh surface spectrum (Fig.2 (a)) implies high quality Fe_3O_4 formation. Overlaying 1 nm metal aluminum changes the XPS spectral profile

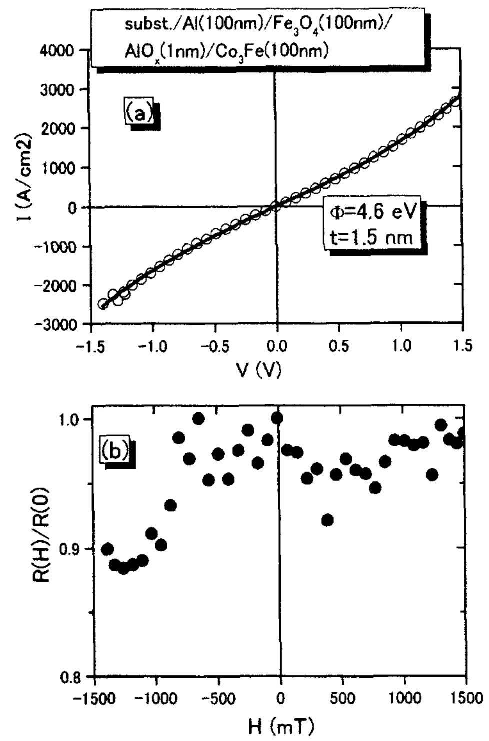

Fig.1 I-V curve (a) and MR result (b) for an aluminium oxide barrier sample. The solid line in (a) is a fitting result using Simons' equation.

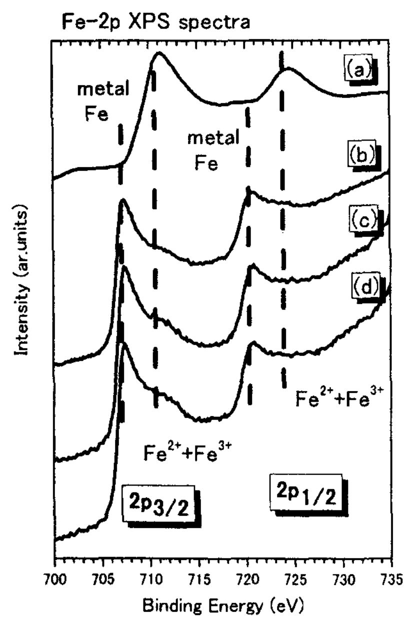

Fig.2 Fe 2p XPS spectra for (a) Fe_3O_4 surface, (b) after Al overlayer, (c) post O_2 exposure at $5x10^3$ Pa for 0.5h, and (d) post O_2 exposure at $5x10^3$ Pa for 9.5h.

drastically. Ionic iron (Fe^{2+}+Fe^{3+}) peaks become faint, instead intense metal Fe peaks appear in Fig.2 (b). Metal aluminum reduces major part of Fe_3O_4 surface to metal iron. The little change in spectra (c) and (d) shows the surface is not re-oxidized by the post exposure to oxygen. Thus the metal aluminum deposition with post oxidation will change the Fe_3O_4 surface magnetic properties.

Magnesium oxide instead of aluminum oxide has been examined as a tunnel barrier and the Fe_3O_4 surface properties have been investigated in this work.

2. EXPERIMENT

Sample films were grown on polished MgO (001) and sapphire (001) substrates by MBE method. Thin Al (1 nm) and successive thick Ag (50 nm) layers were first deposited by K-cells as a conducting under and buffer layer. Metal iron ingot was evaporated in an oxygen atmosphere and a 50 nm Fe_3O_4 layer was reactively deposited on heated substrates. Various oxygen pressures and substrate temperatures have been examined as described later. A barrier layer was formed by depositing MgO in $3x10^{-3}$ Pa O_2 atmosphere. Finally metal Fe as a counter magnetic electrode was grown. All layers without Fe_3O_4 were prepared on room temperature substrates in high vacuum (10^{-6} Pa range).

Epitaxial growth of Al, Ag and Fe_3O_4 layers was confirmed by in-situ RHEED measurements and the detailed structures were investigated by XRD. Sharp and intense Ag and Fe_3O_4 XRD peaks with (111) preferred orientation and sharp rocking curve peaks imply high quality epitaxial growth on sapphire (001) substrates. Though Ag and Fe_3O_4 (200) XRD peaks are not clearly distinguished from the intense MgO (200) peak, streaky RHEED pattern and no extra XRD peak indicate also good epitaxial growth on MgO(001) substrates.

The chemical condition for each layer was investigated by in-situ XPS using non-monochromatic Mg-Kα radiation. The magnetization measurement for a simple Fe_3O_4 film on Al/Ag layer shows clear Verway transition around 120 K implying good stoichiometric composition. Sample surface roughness was evaluated by the contact mode AFM analysis. Both mechanical mask and a lift-off method with a photolithography technique were employed to make MTJ structures. The typical junction size is 0.3x0.3 mm for the former and 10x10 μm for latter, respectively.

3. RESULTS AND DISCUSSION

Successful Fe_3O_4 epitaxial growth on both MgO(001) and sapphire(001) substrates was confirmed by RHEED and XRD at the substrate temperature as low as 250°C. However, higher oxygen atmospheric pressure than $1x10^{-3}$ Pa makes Fe_3O_4 over oxidized a little due to the large oxygen sticking probability at low temperature substrate. Broad satellite between Fe $2p_{3/2}$ and $2p_{1/2}$ XPS peaks characteristic to Fe^{3+} in Fe_2O_3 appears for those over oxidized samples. Such an in-situ XPS analysis is effective to detect a slight change in the surface Fe_3O_4. Actually a little over oxidation of surface Fe_3O_4 by layering MgO barrier layer was detected. [5] No observable satellite in Fe 2p XPS spectra before (Fig.3 (a)) and after (Fig.3 (b)) MgO deposition confirms stoichiometric Fe_3O_4 surface after overlaying barrier layer. Thus we can believe the Fe_3O_4 hold half-metallic properties at the interface.

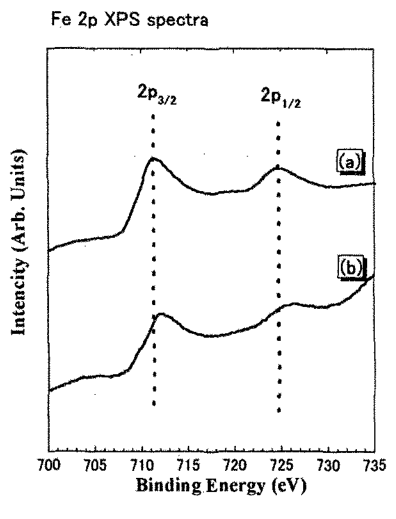

Fig.3 Fe 2p XPS spectra for Fe_3O_4 surface before (a) and after (b) MgO (3 nm) layer deposition in $3x10^{-3}$ Pa O_2.

Flat and uniform interfaces between magnetic electrodes and barrier are important to get good tunneling properties. Various surface roughness of a cross stripe shape sample was examined by a contact mode AFM as illustrated in Fig.4. Actual junction surface of this sample shown in Fig.4(c) is fairly rough (average roughness (Ra)=8.1 nm). The roughness of MgO surface on Fe_3O_4 (Fig.4 (a)) is the same level (Ra=9.7 nm), while the Fe surface on MgO is very smooth (Fig.4 (b), Ra=1.9 nm). This result implies the Fe_3O_4 surface governs the interface roughness of this series of MTJs. Possible preparation conditions of Fe_3O_4 have been discussed to

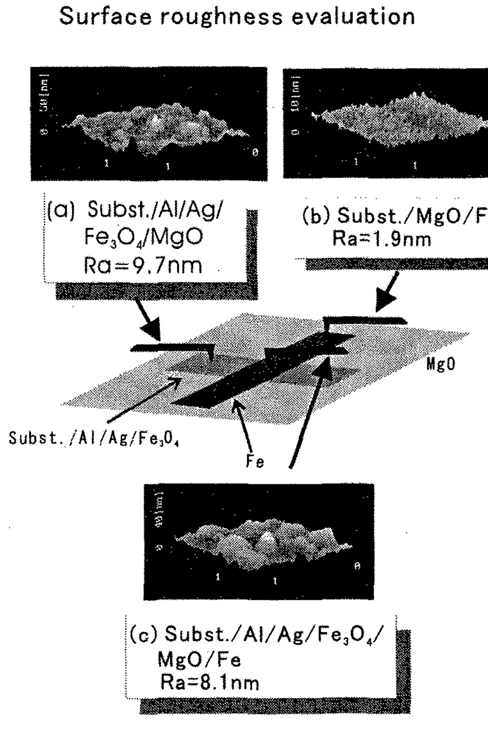

Fig.4 Various surface morphologies and average roughness (Ra) measured by a contact mode AFM. 1.5x1.5 mm area is scanned for a subst./Al(1 nm)/ Ag(100 nm)/Fe_3O_4(50 nm)/MgO(5 nm)/Fe(50 nm) sample.

minimize the surface roughness. Several MTJs were prepared on polished MgO(001) substrates with the optimum conditions of Ts=250°C and $P(O_2)=3x10^{-3}$ Pa and the measured Ra is less than 2 nm. Since the probability of pinholes in barrier layers is proportional to the junction area, the micro-fabrication technique with a combination of photolithography and lift-off was employed instead of a mechanical mask. Only one junction showed a tunneling like non-linear I-V curve, however no reproducibility and no MR effect suggest it suspicious. We are investigating the micro-fabrication experimental problem now.

4. SUMMARY

High quality Fe_3O_4 layers with stoichiometric interfaces were prepared using MgO barrier layers instead of aluminum oxide and confirmed by both in-situ XPS and RHEED measurements. A very flat junction surface (Ra < 2 nm) was obtained by optimizing Fe_3O_4 preparation conditions. However, reliable MTJ behavior has not been observed yet.

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