Structure and tunnel magnetoresistance in Fe/MgF_2/Co junctions With an oxide seed layer on an Fe bottom electrode

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Fe/MgF_2/Co magnetic tunnel junctions (MTJs) with an oxide seed layer (MgO, Fe-O) inserted between an Fe bottom electrode and a MgF_2 barrier layer have been prepared through the use of molecular beam epitaxy, and the effects of the seed layers on structure of subsequently deposited MgF_2 barrier layers and tunnel magnetoresistance (TMR) have been investigated. The crystallographic orientation of MgF_2 layers depends significantly on the seed layers, and furthermore, it has been found that the surface roughness of MgF_2 layer is reduced by inserting a MgO seed layer. In Fe/MgF_2 (3-12nm)/Co MTJs without any seed layer, top Fe and bottom Co electrodes contact each other through pin holes in MgF_2 layers. On the other hand, however, Fe and Co electrodes are separated both magnetically and electrically in Fe/MgO (0.3-0.5nm)/MgF_2 (2-5nm)/Co MTJs, i.e., MgF_2-based MTJs have successfully been prepared by inserting a thin MgO seed layer, resulting in TMR of about 10% observed at 4.2 K. (c) 2002 American Institute of Physics.
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Ⅰ. INTRODUCTION

Investigation on growth and microstructure in magnetic tunnel junctions (MTJs) is of importance because structural features, such crystallographic orientation, morphology, and interface roughness and mixing have a crucial influence on tunnel magnetoresistance (TMR). [1-7] For instance, a large TMR is predicted theoretically in an epotaxially grown Fe (001)/MgO (001)/Fe (001) MTJ, suggesting that remarkable TMR effects may appear in single crystal or highly-oriented epitaxial MTJs.6 On the other hand, it is well known that surface roughness of bottom electrodes and/or three-dimensional growth of barrier layers cause pin holes in barrier layers, and the formation of pin holes dramatically diminishes TMR. [1] Due to defects such as pin holes, TMR is observed only for MTJs with limited combinations of ferromagnetic metals and insulators.

TMR is observed not only in MTJs but also in magnetic granular films such as sputter-deposited Co- Al-O films.[8] Recently, the largest TMR at temperature of those observed in granular films to date was found in CoFe-MgF_2 granular films,[9] suggesting that the combination of ferromagnetic transition metals and MgF_2 is suitable for spin dependent tunneling transport. Therefore, it is expected that MTJs having a MgF_2 barrier layer exhibit large TMR effect. However, successful preparation of MgF_2-based MTJ2 has not been reported, probably because structural defects such as pin holes are formed in the MgF_2 barrier layer due to the low chemical affinity of transition metal/MgF_2 interfaces and the large lattice mismatch between MgF_2 and ferromagnetic transition metals. For successful preparation of MTJs consisting of a new and difficult materials combination, such as transition metal/MgF_2, deep consideration of the deposition process and the obtained microstructures of MTJs should be needed.

In this study, considering seeding technique for MgF_2 growth on an Fe layer, we have prepared Fe/MgF_2/Co MTJs with an oxide seed layer inserted between an Fe bottom electrode and a MgF_2 barrier later, as well as conventional Fe/MgF_2/Co

MTJs. The effect of inserting an oxide seed layer on the growth of MgF_2 barrier layer and TMR of the MTJs has been investigated. Schematic illustrations of the MTJ samples with and without an oxide seed layer for barrier growth are shown in Figs. 1(a) and 1(b). Although several kinds of oxides have been examined to find an appropriate seed layer for the interface between Fe and MgF_2, the results of MgO and Fe-O seed layers are shown as typical cases in this article.

Ⅱ. EXPERIMENTAL PROCEDURE

Fe/MgF_2/Co and Fe/I_s/MgF_2/Co (I_s=MgO, Fe-O) MTJs were prepared on MgO (001) single crystal substrates by using molecular beam epitaxy equipment (Eiko EB-5K). Transition metals (Fe, Co) and MgF_2 were deposited by an electron beam gun and a high temperature cell, respectively. MgO seed layers of 0.3-0.5nm in thickness were formed by natural or plasma oxidization of Mg layers deposited on Fe electrodes. The oxygen partial pressures for natural and plasma oxidization are or 0.8 Pa, respectively. Fe-O seed layers were formed on the surface of Fe electrodes at substrate temperature of 200℃ by introducing oxygen gas of 1.3×10^{-5} Pa into the growth chamber. Changing shadow masks for the cross-pattern of TMJs were performed without exposing sample surfaces to air. The junction area $0.5\times0.5mm^2$. Crystal structure and orientation were monitored by *in situ* reflection high energy electron diffraction (RHEED), and surface morphology was characterized by atomic force microscopy (AFM) in a contact mode. Magnetic properties and TMR in the temperature range of 4.2 K-RT were measured by using a superconducting quantum interference device magnetometer and a conventional dc four-probe magnetoresistance measurement system, respectively.

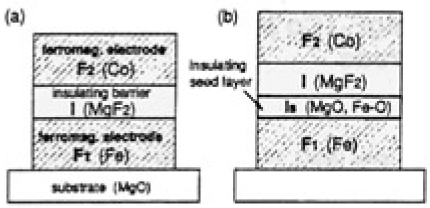

FIG.1 Schematic illustrations of (a) conventional MTL consisting of top and bottom electrodes F_1, F_2 and insulating barrier Ⅰ and (b) MTJ with insulating an insulating seed layer Ⅰs between a bottom electrode F_1 and an insulating barrier 1.

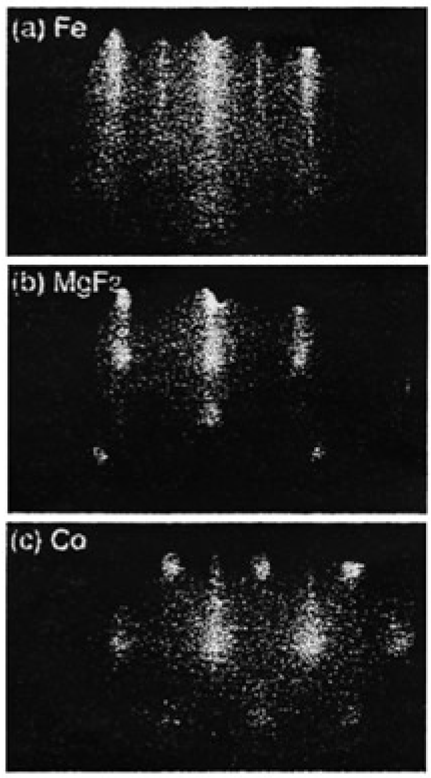

FIG.2 RHEED patterns taken for an Fe (10nm)/MgF_2 (4nm)/Co (20nm) MTJ. (a)-(c) corresponds to Fe, MgF_2, and Co surfaces, and incident electron beam of 10kV is along MgO [100].

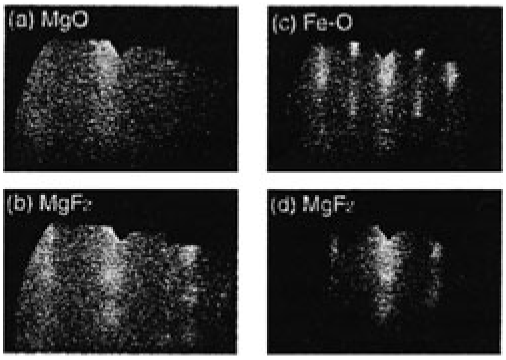

FIG.3. RHEED patterns taken for (a) MgO and (c) Fe-O seed layers grown on Fe (001) bottom electrodes and (b) and (d) MgF_2 barrier layers subsequently grown on the MgO and Fe-O seed layers, respectively, at 200°C. Incident electron beam of 10kV is along MgO [100].

III. RESULTS AND DISCUSSION

Figures 2 (a)-2 (c) show RHEED patterns taken for a typical example of Fe/MgF_2/Co MTJ. A 10-nm-thick Fe bottom electrode was epitaxially grown on MgO (001) at RT, and then was annealed at 200°C. The RHEED pattern for Fe (001) surface in Fig. 2(a) indicates two-dimensional growth of the Fe bottom electrode. Surface roughness of the Fe electrode measured by AFM is comparable to that of MgO substrates. After the substrate temperature was elevated up to 300°C, a 4-nm-thick MgF_2 layer was deposited on the Fe electrode, From the RHEED image for MgF_2 shown in Fig. 2(b), it is found that MgF_2 is epitaxially grown on Fe (001), even though there exists a large lattice mismatch of 14% between bulk Fe and MgF_2. The in-plane lattice parameter of MgF_2 is estimated to be about 5% smaller than that in bulk state. The splitting of streak patterns observed implies existence of mosaic structure in the MgF_2 layer. The subsequently deposited Co top electrode (20nm) is also epitaxially grown. From this result, it is noted that MgF_2 can be grown in crystalline state at relatively low substrate temperatures, compared to Al-O barrier layers, and MgF_2 is considered to be a possible candidate of barrier material that constitutes single crystal MTJs. However, all Fe/MgF_2 (3-12nm)/Co MTJs prepared in various conditions show magnetic and transport properties that was well interpreted by formation of many pin holes in MgF_2 layers, resulting in no TMR effect.

An appropriate seed layer grown on a bottom electrode may change the microstructure of a subsequently deposited barrier layer. MgO and Fe-O were chosen as a seed layer between a Fe bottom electrode and a NgF2 barrier layer. Figures 3(a)-3(d) show RHEED patterns taken for (a) a MgO seed layer (via plasma oxidization), (c) an Fe-O seed layer and (b) and (d) MgF_2 layers subsequently grown on the MgO and Fe-O seed layers at 200°C. MgO and Fe-O seed layers were formed epitaxially on Fe (001) single crystal bottom electrodes, and interestingly, the crystallographic orientation of MgF_2 is different between the MgO and Fe-O seed layers. In-plane orientation relations between MgF_2 (001) layers and Fe (001) bottom electrodes were found to be Fe[100]//MgF_2[110] for a MgO seed layer and Fe[100]//MgF_2[100] for an Fe-O seed layer. Figure 4 shows line profiles taken from contact-AFM images for the MgF_2 surfaces of Fe/MgF_2 bilayers and Fe/MgO/MgF_2 trilayers, where the seed layers are 0.5-nm-thick plasma-oxidized MgO layers. It is shown that inserting a MgO seed layer reduces surface roughness of subsequently grown MgF_2 layers. The RHEED and AFM observation reveals that MgO and Fe-O seed layers grown on Fe bottom layers have a significant influence on crystallographic orientation and morphology of subsequently deposited MgF_2 layers. All Fe/MgF_2/Co MTJs without any seed layer prepared in this study probably have many pin holes in MgF_2 barriers, and show no TMR effect. On the other hand, however, TMR effect was clearly observed for MTJs in which a 0.3-0.5-nm-thick MgO seed layer was formed on a Fe bottom electrode and a subsequent 2-5-nm-thick MgF_2 barrier layer was grown at RT. As a typical result, junction resistance and normalized magnetization at 4.2K as a function of applied magnetic field for a Fe (20nm)/MgO (0.3nm)/MgF_2 (5nm)/Co (20nm) MTJ are shown in Figs. 5(a) and 5 (b). TMR of about 10% appears, roughly corresponding to the magnetization curve with a two-step loop. This result shows that formation of pin holes was suppressed by inserting a MgO seed layer and therefore magnetic and electrical separation required for top and bottom electrodes in MTJs was achieved. It can be concluded that inserting an appropriate oxide seed layers is a much effective method to prepare MTJs with MgF_2 barrier.

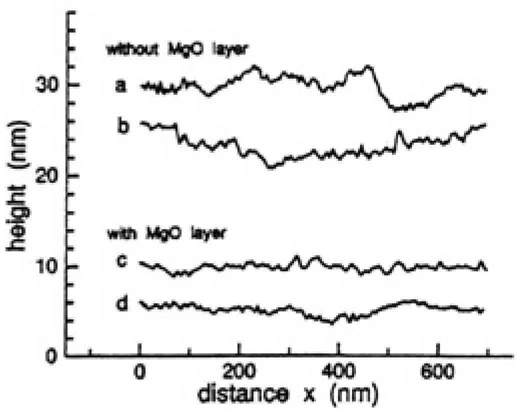

FIG.4. Lineprofiles in AFM images for surfaces of (a) an Fe (10nm)/MgF_2 (4nm) bilayer grown at 300℃, (b) an Fe (20nm)/MgF_2 (3nm) bilayer grown at 100℃, (c) an Fe (20nm)/MgO (0.5nm)/MgF2 (2nm) trilayer grown at 200℃, and (d) an Fe (20nm)/MgO (0.5nm)/MgF_2 (2nm) trilayer grown at RT. The peak-to-peak values that express the surface roughness are 5.0 and 5.1nm for (a) and (b) without seed layers and 2.2 and 2.8nm for (c) and (d) with seed layers, respectively.

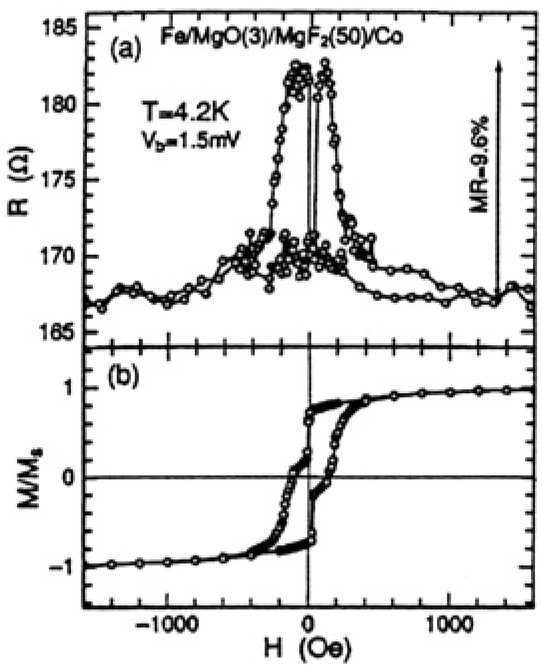

FIG.5. (a) Junction resistance and (b) normalized magnetization at 4.2K as a function of applied magnetic field for an Fe (20nm)/MgO (0.3nm)/MgF_2 (5nm)/Co (20nm) MTJ.

TMR of about 10% remains at 77K for the MTJ, but the TMR ratio is reduced to be less than 1% at RT. Concerning reproducibility, not only TMR ratios but also junction resistances are much different between MTJ samples even though the MTJ samples are prepared by the same deposition process. This is considered to be mainly due to the large junction area ($0.5\times0.5mm^2$) with considerable inhomogeniety. Reducing junction area by a lithography process is required to obtain more reproducible results. For most of MTJs with MgO seed layers, nonlinear I - V characteristics and negative temperature coefficient of junction resistance were observed, and from the T-Vcurves the barrier heights were estimated to be 0.1-2eV. The detailed analysis of transport properties will be published elsewhere.[10] Finally, it is noted that MgF_2 layers in the RT-grown MTJs showing TMR were in an amorphous or polycrystalline state while single crystal MgF_2 layers are epitaxially grown above 200℃ on MgO seed layers. No clear difference between natural and plasma oxidization is seen in the structure and magnetic and transport properties. Optimizing growth conditions to achieve single crystal MgF_2-based MTJs and higher TMR ratios is now in progress.

Ⅳ. CONCLUSION

We have investigated structure and TMR of Fe/MgF_2/Co and Fe/Is/MgF_2/Co (Is=MgO, Fe-O) MTJs prepared on single crystal MgO (001) substrates by molecular beam epitaxy. Crystallographic orientation of MgF_2 layers depends significantly on the seed layers, and the surface roughness of MgF_2 layers is reduced by inserting MgO seed layers. In Fe/MgF_2 (3-12nm)/Co MTJs without any seed layer, Fe and Co electrodes contact each other through pin holes in MgF_2 layers. On the other hand, however, magnetic and electrical separation between Fe and Co electrodes is achieved in Fe/MgO (0.2-0.5nm)/MgF_2 (2-5nm)/Co MTJs, i.e., MgF_2-based MTJs have successfully been prepared by inserting a thin MgO seed layer, resulting in TMR of about 10% observed at 4.2 K.

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