

Fabrication of Magnetic Tunnel Junctions with a Single-Crystalline LiF Tunnel Barrier

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Abstract

We fabricated Fe/LiF/Fe magnetic tunnel junctions on MgO(001) substrates by molecular beam epitaxy, where the LiF has the same crystal structure as MgO. Crystallographical studies revealed that the LiF is single-crystalline with LiF(001)[100] || bottom Fe(001)[110]. The observed tunnel magnetoresistance (MR) ratios were about 6 % (1%) at 20 K (room temperature). Such a low MR ratio implies that the thermally excited carriers, which are not spin-polarized, in the LiF barrier play a major role on the transport process.

1. Introduction

In magnetic tunnel junction (MTJ), great efforts have been devoted to develop single-crystalline tunnel barriers because the epitaxial MTJs can yield giant magnetoresistance (MR) ratios due to the so-called coherent spin-polarized tunneling [1]. To date, giant MR ratios have been observed at room temperature (RT) in epitaxial MTJs with oxide barriers such as MgO [1-3] and Mg(Al, Ga)₂O₄ [4,5], and GaO_x [6]. LiF is an insulating halide (band gap ~13 eV) having the same crystal structure as MgO (rock salt type). It is noteworthy that the lattice constant of LiF (3 nm) gives a quite small lattice mismatch (~0.4 %) when the Fe unit cell is turned by 45° with regard to the LiF unit cell. A recent *ab initio* calculation for the epitaxial Fe(001)/LiF(001)/Fe(001) structures has predicted the existence of coherent spin-polarized tunneling, and thereby very high MR ratios [7]. However, only a few studies on the LiF-based MTJ have been reported [8]. In the present study, we report on structural and magneto-transport properties of Fe/LiF/Fe MTJs.

2. Experimental methods

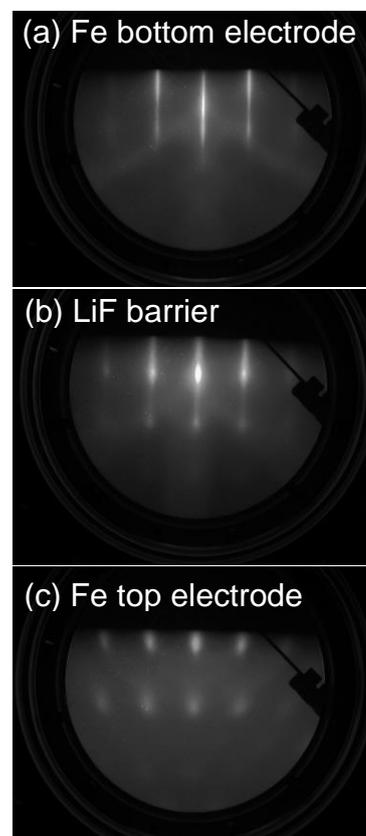
MTJ films were grown by molecular beam epitaxy. The structure of the MTJs is Au (10 nm) cap / Co (10 nm) pinned layer / Fe (5 nm) upper electrode / LiF (3 nm) tunnel barrier / Fe (30 nm) bottom electrode / MgO (10 nm) buffer layer on MgO(001) substrates. The Fe bottom electrode was grown at 100°C, followed by an *in situ* annealing at 350°C for 10 min to improve the surface morphology. The LiF tunnel barrier was deposited at RT under the vacuum below 2.5×10^{-10} Torr. Then the Fe top electrode, Co-pinned and Au-cap layers were grown at RT. The films were patterned into tunnel junctions (3 × 12 μm) using a conventional micro-fabrication technique. The MR curves were measured

with a conventional two probe method.

3. Results

Figures 1 show reflection high-energy electron diffraction (RHEED) patterns of the (a) Fe bottom electrode, (b) LiF tunnel barrier, and (c) Fe top electrode, respectively. The Fe bottom electrode had sharp streak RHEED patterns, indicating an atomically flat surface. It is clear that LiF barrier layer also revealed streak patterns, suggesting that the LiF is single-crystalline. For the Fe top electrode, on the other hands, broad spots appeared in the RHEED image. By a careful observation, we could see faint rings which were superimposed on the broad spots. This suggests that the Fe top electrode is not single-crystalline but has still highly (001)-oriented structure.

A cross sectional transmission electron microscopy (TEM) image of the MTJ is given in Fig. 2(a). The TEM image revealed both LiF tunnel barrier and Fe bottom electrode are single-crystalline as expected from the RHEED observations. From the image in the range of about 50 nm, the in-plane lattice mismatch ($\Delta a/a$) between the LiF barrier and Fe bottom electrode was estimated to be about 0.5%, where the lattice constant of Fe is (smaller or larger). The $\Delta a/a$ value is in good agreement with that expected from the lattice constant of bulk Fe and LiF (0.4%). The crystal orientation between the LiF barrier



Figures 1 RHEED images of the (a) Fe bottom electrode, (b) LiF tunnel barrier, and (c) Fe top electrode Fe, respectively. ([100] azimuths of MgO substrate).

and the Fe bottom electrode was determined to be LiF(001)[100] || bottom Fe(001)[110] ([100] azimuth of the MgO substrate) by nano-beam diffraction (NBD) analyses (Figs. 2b and 2c). Remarkably, the LiF unit cell is turned by 45° with regard to the Fe unit cell in the same manner as the MgO(001) on Fe(001) [1]. We also confirmed by TEM and NBD observations that majority part of the Fe top electrode has (001) crystal orientation.

Typical MR curves of the Fe/LiF/Fe MTJ at temperature (T) of 20 K and RT are given in Figs. 3(a) and 3(b), respectively. MR ratio is defined as $(R_{AP} - R_P)/R_P$ where R_{AP} and R_P are the junction resistances between two Fe electrodes at anti-parallel and parallel magnetization states, respectively. Despite the single-crystalline LiF(001) barrier, the observed MR ratios were low of about 6% even at a low T of 20 K. With increasing T , the MR ratio rapidly decreased and was about 1% at RT. We also observed significant T dependence in the resistance-area (RA) products ($2 \times 10^6 \Omega \mu\text{m}^2$ for 20 K and $9 \times 10^4 \Omega \mu\text{m}^2$ for RT). The results imply that the thermally excited carriers, which are basically not spin-polarized, in the LiF barrier play a major role on the transport process.

Recently, Xue *et al.* fabricated fully epitaxial FeCo(001)/LiF(001)/FeCo(001) MTJs, and also observed a rapid decrease of the MR ratio with temperature (90% and 17% at 77 K and RT, respectively) [8]. They pointed out that the observed large T dependence is attributed to imperfections in the LiF barrier. It is necessary for achieving a giant MR ratio at RT to improve a crystal quality of the LiF tunnel barrier by optimizing its growth conditions.

4. Conclusion

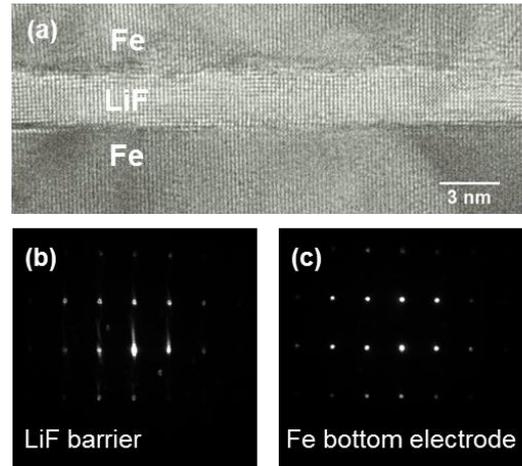
We fabricated Fe/LiF/Fe MTJs on MgO(001) substrates. The LiF barrier layer was confirmed to be single-crystalline with LiF(001)[100] || bottom Fe(001)[110] crystal orientation in the same manner as the Fe(001) on MgO(001). The $\Delta a/a$ between the LiF barrier and Fe bottom electrode was obtained to be very small of $\sim 0.5\%$. Despite the single-crystalline LiF barrier, the observed MR ratios were low of about 6% (1%) at 20 K (RT), respectively. The result implies that the thermally excited carriers in the LiF barrier play a major role on the transport process.

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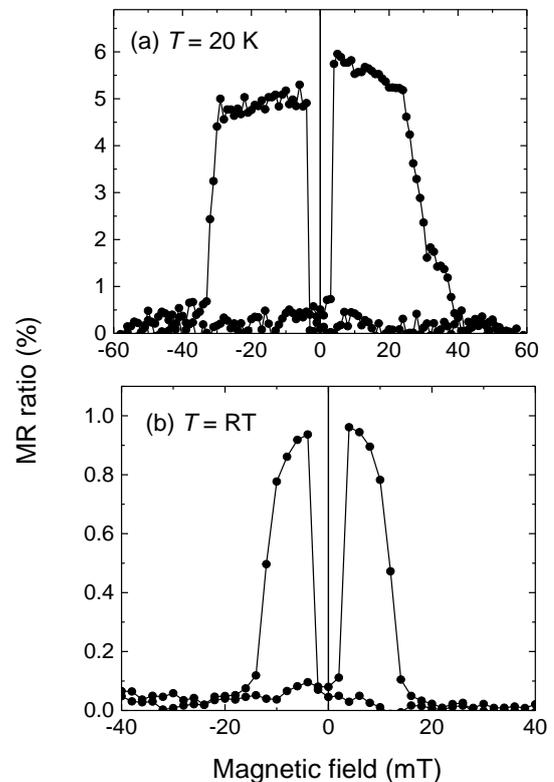
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Figures 2 (a) Cross sectional TEM image of the Fe/LiF/Fe MTJ, NBD patterns of the (b) LiF barrier and (c) Fe bottom electrode, respectively ([100] azimuths of MgO substrate).



Figures 3 MR curves of the Fe/LiF/Fe MTJ at (a) 20 K and (b) RT, respectively.