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Symmetry of Wavefunction at the Interface of Fe/MgO Magnetic Tunneling Junction

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Abstract: We measured the anisotropy of Magnetic Compton profiles (MCP) in the Fe/MgO multilayers and compared it with band structure calculations. At the Fe/MgO interface, the $|m| = 1$ state in the minority band is suppressed, which in turn promotes the spin-polarized occupation of the $|m| = 1$ state. At the Fe/FeO interface of intentionally oxidized Fe/MgO multilayers, the occupations of the magnetic quantum number are almost equal (spherical) in the majority band and minority band, and therefore, the spin-polarization occupancy is also almost equal (spherical). These results contribute to the material design for MTJs as high-performance spintronic devices.

Keywords: Fe/MgO; magnetic tunneling junction; magnetic Compton scattering; band structure calculation

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1. Introduction

Fully epitaxial Fe/MgO/Fe magnetic tunneling junctions (MTJs) have an extremely high MR ratio because of the coherent tunneling effect [1]. These phenomena are important for the recent spintronics technique. For the coherent tunneling effect, an electronic structure is required to conserve the wavefunction coherency at the interface [2]. The theoretical calculation by Butler et al. [2] showed that the Δ1 symmetry (Fe4*s*, 4*p* and Fe3*d* of $|m| = 0$, here m denotes the magnetic quantum number) at the Fe/MgO interface contributes to the coherent tunneling effect. Furthermore, an MTJ with perpendicular magnetic anisotropy (PMA) is required because of the high thermal stability and low critical current for current-induced magnetization switching [3]. The PMA arises from a symmetry of the wavefunction at the MTJ interface [4]. Therefore, direct observation for the symmetry of the wavefunction at the Fe/MgO interface is important.

Considering the symmetry of the wavefunction, a magnetic Compton profile (MCP) measurement is a good probe to measure the symmetry of wavefunctions [5–9]. We reported magnetic Compton profiles (MCPs) of Co/Pd, Co/Pt and Co/Au multilayered films from a viewpoint of the PMA and symmetry of a wavefunction [5–9]. We analyzed the anisotropies of the MCPs to obtain spin-projected occupation number ratios of each one of the magnetic quantum numbers, $|m| = 0$, 1 and 2. The analysis showed that the symmetry of the wavefunction contributes to the PMA properties of the Co/Pd, Co/Pt and Co/Au multilayered films. Therefore, an MCP measurement will probe the symmetry of wavefunctions at the Fe/MgO MTJ interface.

In this paper, we report measurements of anisotropies of MCPs in Fe/MgO MTJs. The anisotropies of the MCPs are decomposed into the spin-projected occupation number ratios of each one of the magnetic quantum numbers, $|m| = 0$, 1 and 2. The obtained results are

compared with the band structure calculations [10]. From the MCP measurements and theoretical calculations, the symmetry of wavefunctions is discussed from a viewpoint of the chemical states at the Fe/MgO interface, such as the oxidization of the Fe layer.

2. Materials and Methods

Fe(x nm)/MgO(1 nm) (x = 2.5, 4, 10) multilayered films were fabricated by radio frequency sputtering (ULVAC MB96-1011, ULVAC, Chigasaki, Japan) in Ar gas at a working pressure of 1.0 Pa, on an Al foil substrate (thickness: 12 μm) and Si substrates. A buffer layer of 100 nm of Fe was fabricated on the substrate and a buffer layer of 100 nm of MgO was fabricated on the Fe buffer layer before the fabrication of the multilayers. The total thickness of the multilayers was adjusted to about 1000nm. Intentionally oxidized samples at the Fe/MgO interface were also fabricated as Fe(x nm)/FeO/MgO(1 nm) (x = 4, 10). The list of fabricated samples is summarized in Table 1. A Si substrate sample was used for the X-ray diffraction measurement, while an Al foil substrate sample was used for the magnetization measurement and MCP measurement. The Al foil substrate was chosen to suppress the X-ray background in the MCP measurements.

Table 1. Fabricated samples for measurements.

Sample Name	Fe nm	MgO nm	Λ nm	Number of Layers	Fe Oxide (FeO)
sample1	10.0	1.0	11.0	150	nothing
sample2	4.0	1.0	5.0	300	nothing
sample3	2.5	1.0	3.5	400	nothing
sample4	4.0	1.0	5.0	300	exist
sample5	10.0	1.0	11.0	150	exist

The crystal structure and the multilayer period were confirmed by X-ray diffraction measurements using CuKα1 radiation (40 kV and 30 mA). A transmission electron microscope (JEM-4000EX, JEOL, Akishima, Japan) was also used to confirm the multilayer structure. The macroscopic magnetization curve measurements were performed using a SQUID magnetometer (MPMS-5SW, Quantum Design, San Diego, US). The MCP measurements were carried out on a beamline BL08W at SPring-8, Japan [11]. Elliptically polarized X-rays were monochromatized to be 182.6 keV and were focused onto a spot of 1 × 1 mm^2 on the sample. The X-ray beams were parallel to an applied magnetic field. Magnetic fields of ± 2.5 T were applied parallel or perpendicular to the film plane. To achieve a larger scattering volume, the films were folded 5 times to be stacked to 32 sheets. The scattered X-rays were detected by a ten-segment Ge solid-state detector (Canberra GL0115S, Mirion Technologies, Atlanta, GA, USA) with a scattering angle of 178 degrees. Therefore, an observed electron momentum, p_z, was parallel to the applied magnetic field. The overall momentum resolution was Δp_z = 0.43 au. The intensity of the incident X-ray beams was monitored by an ion chamber for data normalization. The measurements were performed in a vacuum at room temperature.

The band structure calculations (the full potential linear augmented plane wave (FLAPW) band structure calculation based on the local spin density approximation) were performed by the all-electron band structure calculation package (ABCAP), developed by N. Hamada [10].

3. Results and Discussion

Figure 1 shows the X-ray diffraction pattern for the Fe/MgO multilayered films in the middle angle region. Fe (002) and MgO (002) textures are confirmed around 65 and 42 degrees, respectively. The intensity ratios of MgO (002) to Fe (002) correspond to the thickness ratios of the MgO layer to the Fe layer, as shown in Figure 1a,b. On the contrary, the intensity around 42 degrees enhances in the intentionally oxidized Fe/MgO

multilayered film because both the MgO (002) and FeO (002) have peaks around 42 degrees, as shown in Figure 1c.

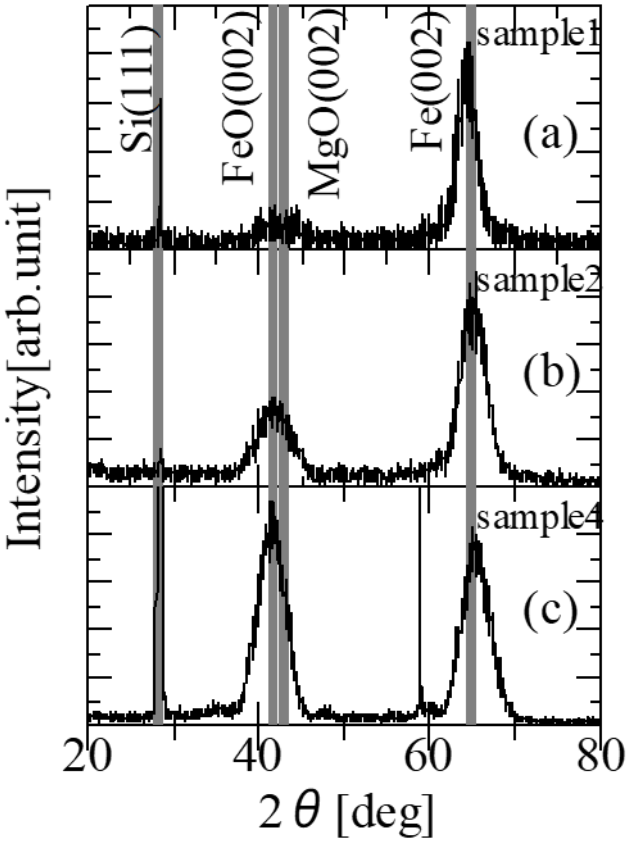

Figure 1. X-ray diffraction pattern for (**a**) Fe(10 nm)/MgO(1 nm), (**b**) Fe(4 nm)/MgO(1 nm) and (**c**) intentionally oxidized Fe(4 nm)/MgO(1 nm) multilayered films. The gray lines indicate PDF data (Si(27-1402), Fe(06-0696), MgO(45-096) and FeO(89-0686)).

The designed multilayered structure is confirmed by the TEM observation, as shown in Figure 2. Lattice spacing corresponding to the FeO layer is confirmed at the Fe/MgO interface in the intentionally oxidized Fe/MgO multilayered films, as shown in Figure 2b.

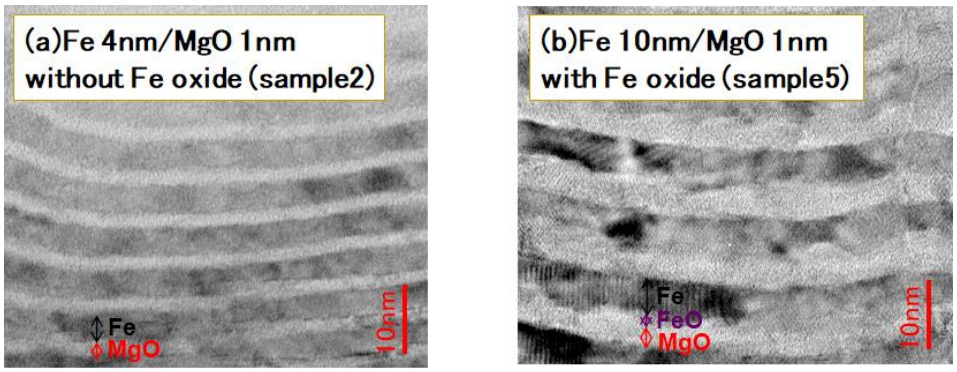

Figure 2. Images by transmission electron microscope (JEM-4000EX, JEOL) for (**a**) Fe(4 nm)/MgO(1 nm) (sample2) and (**b**) intentionally oxidized Fe(10 nm)/MgO(1 nm) (sample5).

Figure 3 shows the saturation magnetizations for the Fe/MgO multilayered films. The magnetization enhances with the decrease in the Fe volume ratio. This indicates the enhancement of the magnetization at the Fe/MgO interface. The decrease in the magnetization when the Fe volume ratio is less than 0.7 comes from the decrease of the

Curie temperature in the nano-cluster. The intentionally oxidized Fe/MgO multilayered films show the decreased magnetizations because of the oxidization of the Fe layer, such as FeO, which is shown in Figures 1 and 2. The exchange bias field was observed with zero field cooling in the intentionally oxidized sample (Fe(4 nm)/MgO(1 nm) (sample 4)), which indicates the antiferromagnetic phase, such as FeO at the Fe/MgO interface. On the contrary, no exchange bias field was observed with zero field cooling in the Fe/MgO multilayered film (Fe(4 nm)/MgO(1 nm) (sample 2)).

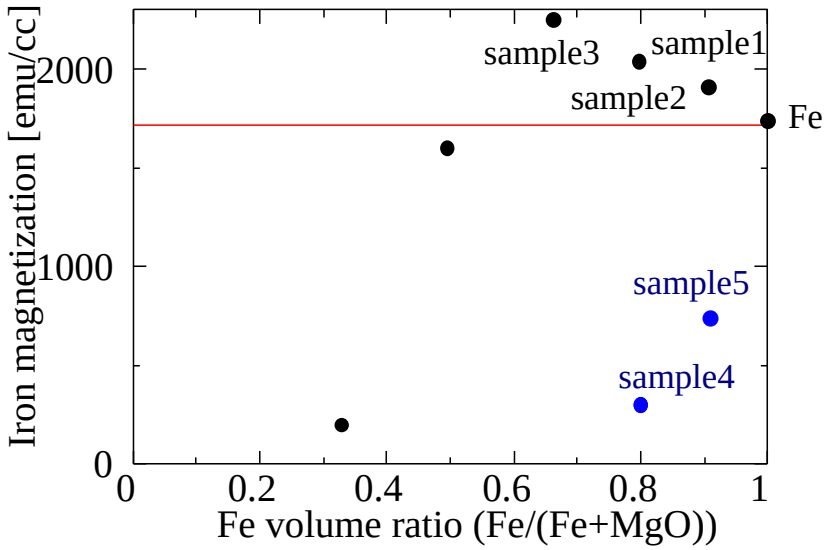

Figure 3. The magnetizations of Fe layers for Fe/MgO multilayered films. Samples 1, 2 and 3 denotes Fe(10 nm)/MgO(1 nm), Fe(4 nm)/MgO(1 nm) and Fe(2.5 nm)/MgO(1 nm), respectively. Sample 4 and 5 denote intentionally oxidized Fe(4 nm)/MgO(1 nm) and Fe(10 nm)/MgO(1 nm), respectively.

Figure 4 shows the MCPs of the Fe/MgO multilayered films with an applied magnetic field being parallel and perpendicular to the film plane. Differences in the MCPs between the parallel and perpendicular applied magnetic field, in other words, anisotropy, are observed in the momentum of less than 2 au, as shown in Figure 4. To highlight the anisotropy, a difference of MCPs, $\Delta J_{mag}(p_z)$, is obtained as the following:

$$\Delta J_{mag}(p_z) = J_{mag\_out\_of\_plane}(p_z) - J_{mag\_in\_plane}(p_z) \quad (1)$$

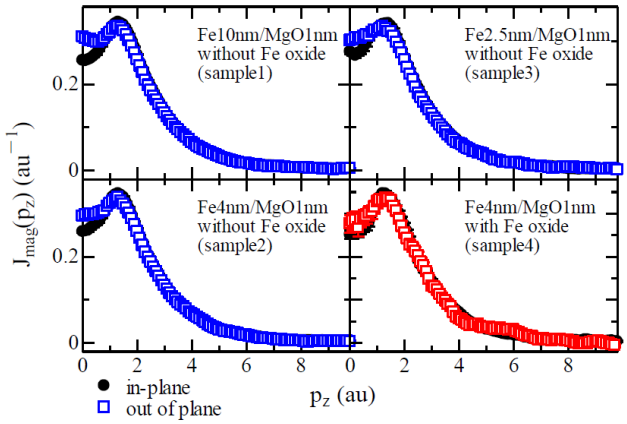

Figure 4. Magnetic Compton profiles (MCPs) for Fe/MgO multilayered films. Open squares and closed circles denote the MCPs in the applied magnetic field perpendicular (out of plane) and parallel (in-plane) to the film plane, respectively.

Here, $J_{mag\_out\_of\_plane}(p_z)$ and $J_{mag\_in\_plane}(p_z)$ denote MCPs with an applied magnetic field perpendicular and parallel to the film plane, respectively. The obtained anisotropies, $\Delta J_{mag}(p_z)$s, are shown in Figure 5.

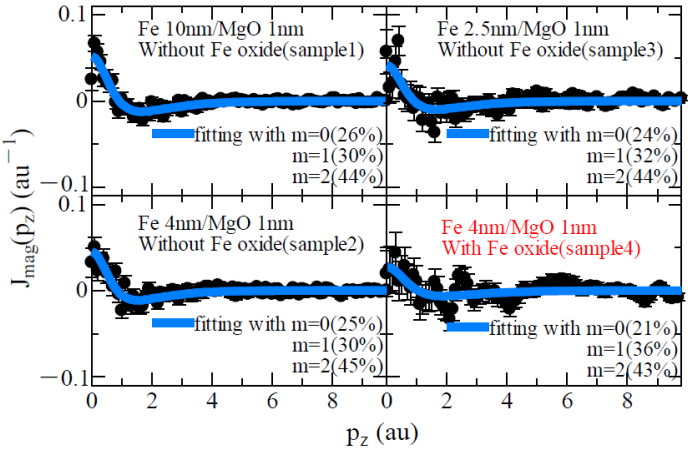

Figure 5. Anisotropies of Magnetic Compton profiles for Fe/MgO multilayered films obtained from Figure 4. Solid lines denote the fitting results explained later.

To analyze the anisotropy of the MCPs, we calculate the model MCPs for the magnetic quantum numbers, $|m| = 0$, 1 and 2 states, assuming the atomic wavefunction of Fe 3*d* under a uniaxial crystal field, whose quantization axis is perpendicular to the film plane. Figure 6 shows the model calculation of MCPs in the applied magnetic field (a) parallel (in-plane) and (b) perpendicular (out-of-plane) to the film plane for the magnetic quantum numbers, $|m| = 0$, 1 and 2 states. The anisotropies $\Delta J_{mag}(p_z)$s are also obtained by using Equation (1), as shown in Figure 6c. The anisotropies depend on the magnetic quantum numbers, $|m| = 0$, 1 and 2 states. Therefore, the anisotropies in Figure 5 can be decomposed into each contribution of the magnetic quantum number by fitting the analysis with the basis functions shown in Figure 6c. The fitting results are shown in Figure 5. The experimental anisotropies can be fitted well by the model calculation in Figure 6c, as shown in Figure 5.

Each contribution extracted by the fitting analysis is summarized in Table 2. The contribution of the magnetic quantum number, $|m| = 1$ states, slightly increases with a decrease in the Fe layer thickness, in other words, with an increase in the Fe/MgO interface contributions. This suggests the symmetry of the wavefunction of 3*d* electrons at the Fe/MgO interface is dominated by $|m| = 1$ states. On the contrary, the contributions of $|m| = 0$, 1 and 2 states in the intentionally oxidized Fe/MgO multilayered film have an almost spherical distribution of 20%, 40% and 40%, respectively. This is consistent with the reduced anisotropy in Figure 5.

This behavior can be reproduced by the band structure calculation [10]. We carried out the full potential linear augmented plane wave (FLAPW) band structure calculation based on the local spin density approximation for the MgO(9)Fe(7) and MgO(8)FeO(1)Fe(7) super cell models. Figure 7 shows the occupation of the magnetic quantum numbers of Fe3d for the majority and minority band in the MgO(9)/Fe(7) and MgO(8)/FeO(1)/Fe(7) multilayers. Spin-polarized occupation numbers are obtained as differences of the occupation numbers between the majority and minority bands. The occupations of the magnetic quantum numbers are almost equal (spherical) in the majority band. However, the occupation of the magnetic quantum number in the minority band show asphericity. For the Fe/MgO interface in MgO(9)/Fe(7), the $|m| = 1$ states of the minority band are suppressed, and hence the spin-polarized occupation of the $|m| = 1$ states is enhanced. For the Fe/FeO interface in MgO(8)/FeO(1)/Fe(7), the occupations of the magnetic quantum numbers are almost equal (spherical) in both the majority and minority bands, and hence the spin-polarized occupations are also almost equal (spherical). These calculated results are

consistent with the experimental results discussed in Table 1, which mainly comes from the antibonding repulsing among the O2*p* and Fe3de_g orbitals.

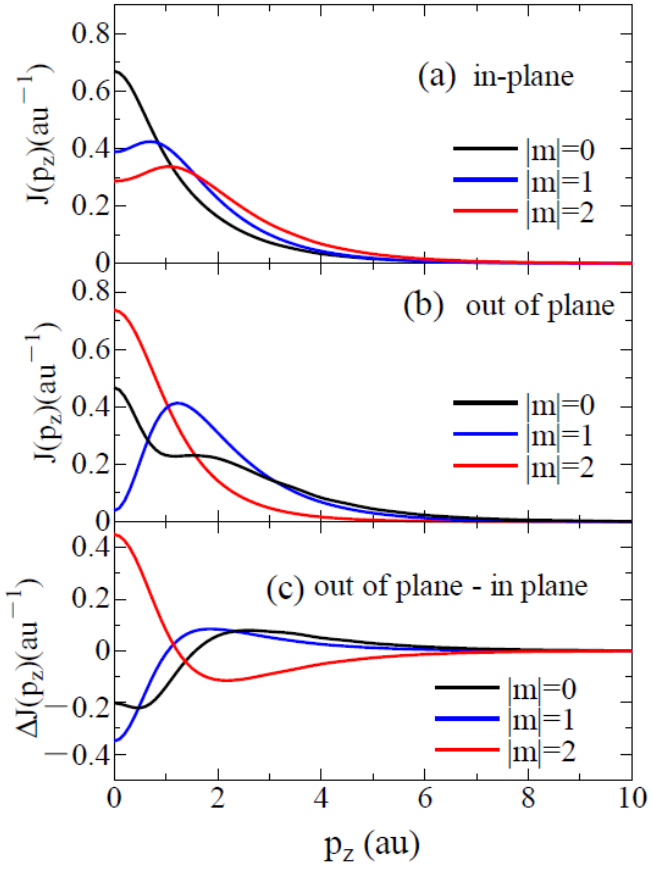

Figure 6. Model calculations of MCPs for Fe3d under the uniaxial crystal field with an applied magnetic field (**a**) parallel (**b**) perpendicular to the film plane. Anisotropies are also shown in (**c**). The model calculations are convoluted by the experimental resolution of 0.43 au.

Table 2. Occupation number of magnetic quantum numbers obtained by magnetic Compton profile measurements. Notice that $|m| = 1$ states have two states of $m = \pm 1$, and $|m| = 2$ states $m = \pm 2$.

Sample Name	$\|m\| = 0$	$\|m\| = 1$	$\|m\| = 2$
Fe 10 nm/MgO 1 nm Without Fe oxide (sample 1)	25%	30%	45%
Fe 4 nm/MgO 1 nm Without Fe oxide (sample 2)	26%	30%	44%
Fe 2.5 nm/MgO 1 nm Without Fe oxide (sample 3)	24%	32%	44%
Fe 4 nm/MgO 1 nm With Fe oxide (sample 4)	21%	36%	43%

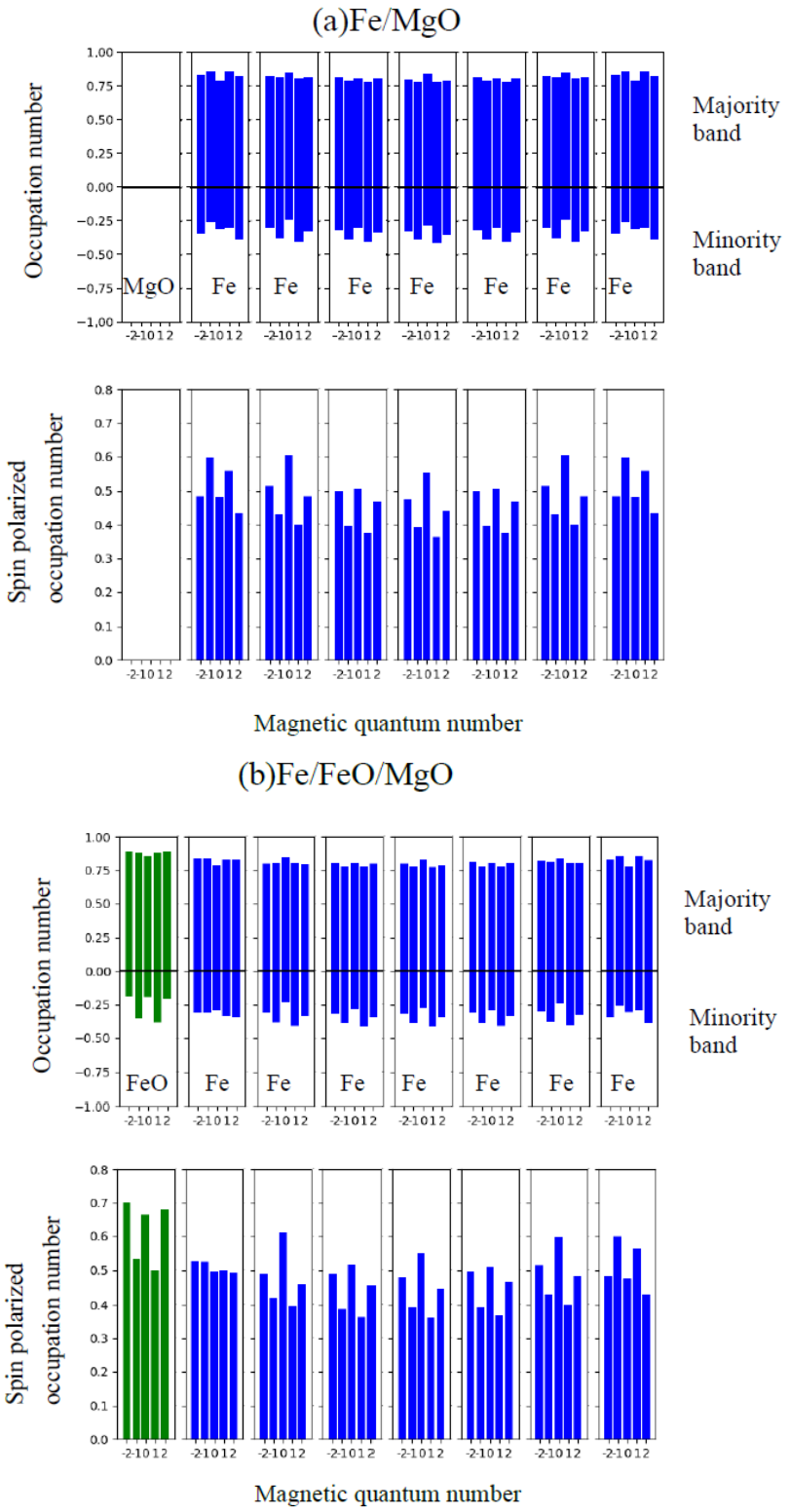

Figure 7. Occupation number and spin-polarized occupation number of magnetic quantum numbers for (**a**) Fe/MgO and (**b**) Fe/FeO/MgO obtained by band structure calculations.

4. Conclusions

In conclusion, we measured the anisotropy of the MCP in the Fe/MgO multilayers and compared it with the band structure calculations. At the Fe/MgO interface, the $|m| = 1$ state in the minority band is suppressed, which in turn promotes the spin-polarized occupation of the $|m| = 1$ state. At the Fe/FeO interface of intentionally oxidized Fe/MgO

multilayers, the occupations of the magnetic quantum number are almost equal (spherical) in the majority band and minority band, and therefore, the spin-polarization occupancy is also almost equal (spherical). These results contribute to the material design for MTJs as high-performance spintronic devices.

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Conflicts of Interest: The authors declare no conflict of interest.

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