Irradiation Effect of Argon Ion on Interfacial Structure Fe(2nm)/Si(t_{Si}=0.5-2 nm) Multilayer thin Film

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ABSTRACT
Investigation includes formation of interfacial structure of Fe(2nm)/Si(t_{Si}=0.5-2 nm) multilayer thin film and the behavior of antiferromagnetic coupling between Fe layers due to Argon ion irradiation was investigated. [Fe(2nm)/Si]30 multilayers (MLs) with a thickness of Si spacer 0.5 - 2 nanometer were prepared on n-type (100) Si substrate by the helicon plasma sputtering method. Irradiation were performed using 400keV Ar ion to investigate the behavior of magnetic properties of the Fe/Si MLs. The magnetization measurements of Fe/Si MLs after 400keV Ar ion irradiation show the degradation of antiferromagnetic behavior of Fe layers depend on the ion doses. The Magnetoresistance (MR) measurements using by Four Point Probe (FPP) method also confirm that MR ratio decrease after ion irradiation. X-ray diffraction (XRD) patterns indicate that the intensity of a satellite peak induced by a superlattice structure does not change within the range of ion dose. These results imply that the surface of interface structures after ion irradiation become rough although the layer structures are maintained. Therefore, it is considered that the MR properties of Fe/Si MLs also are due to the metallic superlattice structures such as Fe/Cr and Co/Cu MLs.

INTRODUCTION
There were many interesting phenomena observed when the spacer between Fe film was substituted by non metallic materials likes Silicon. Fe/Si multilayers(MLs) show an antiferromagnetic couplings (AFC) between Fe layers depending on layer thickness of Si spacers [1]. In addition, atomic mixing in Fe/Si interface regions occurs easily even at room temperature. Fullerton et.al [2] investigate the strong Antiferromagnetic coupling (AFC) in sputtered multilayer for Fe(3nm)/Si(1.5nm) with switching fields of 6 Koe, at room temperature. In this case the interlayer was found to be a crystalline interdiffused Fe-Si alloy. Furthermore, Inomata et.al [3] at 1995 determined the existence of two different types antiferromagnetic interlayer coupling as a function of Si layer thickness. The first kind appeared at multilayer Fe(2.6nm)/Si(1.2nm), with the MR ratio 0.14% at room temperature. The second type was observed at Si thickness t_{Si}=2.5nm with MR ratio value around 0.1% at room temperature. Recently, Tong et.al [4] have observed the dependence of magnetoresistance ratio on the Si thickness t_{Si} on Fe(<2nm)/Si for t_{Si}=0.5~4nm, and on the Fe thickness t_{Fe} on Fe/Si(1.9nm) for t_{Fe}=1~14nm. Also, Sakamoto et.al [5] have grown Fe/Si MLs by using Helicon Plasma Sputtering [6] and studied its structural and magnetic properties.

In this paper, interfacial structure, magnetic and magnetoresistance properties of multilayer Fe/Si grown at ambient temperature by Helicon Plasma Sputtering method and the effect of Argon ion irradiation on the samples of Fe(2nm)/Si(1nm) Multilayer (MLs) are studied.

EXPERIMENTAL METHODS
A series of [Fe(2nm)/Si]30 multilayer (MLs) with t_{Si}=0.5-2 nm were prepared on highly resistivity n-type Si(100) at ambient temperature by Helicon plasma sputtering method in the base pressure of the chamber lower than 1×10^{-7} Torr. The target size is 50 mm in diameter. The deposition rates of Si and Fe were 0.05 nm/sec and 0.068 nm/sec respectively. The Argon gas pressure
was maintained at around $6.7 \times 10^{-4}$ Torr. The multilayer structure was achieved by alternatingly exposing the substrate to Fe or Si target. Structures of multilayers and magnetic properties were observed by X-ray diffraction using CuKα radiation and vibrating sample magnetometer (VSM) up to H=15 kOe. The magnetoresistance (MR) was measured by dc 4 point probe method with field up to H=15 kOe. Rutherford Back Scattering (RBS) experiment was also carried out to confirm the thickness and composition of multilayer. All measurement were carried out at Photonics Research Institute, AIST-Tsukuba, Japan. 400 keV Ar ion irradiation was performed by AIST 400 keV implanter with beam current less than 0.25 μA. The ion range of 400 keV Ar ion in [Fe(2nm)/Si(1nm)]$_{30}$ MLs were estimated to be 364 nm and 338 nm by TRIM code, respectively. Therefore, all Argon atoms stop at the Si substrate.

RESULTS AND DISCUSSION

Pre Argon Ion Irradiation

The first series of [Fe(2nm)/Si(t$_{Si}$)]$_{30}$ multilayer were prepared with various Si layer thicknesses from 0.5 to 2.0 nm for investigating the Si layer thickness dependence on structural, magnetic properties and magnetoresistance.

From Fig. 1, it is clear that the peaks of Fe-bcc strongly depend on Silicon spacer thickness, while the (111) peak of DO$_3$ phase belonging to spacer layer relatively does not show any dependence. The existence of DO$_3$ phase was apparent for all Si thickness might be related to interdiffusion between Fe and Silicon layer as report elsewhere [7].

The peaks of (110), (200) and (211) are belong to Fe-bcc. The (111) peak originates from the DO$_3$ interfacial phase. The crystalline size of Fe/Si MLs estimated from the Fe (110) peak width using Scherer formula [8] are tabulated in Table 1. The large grain size indicates that only few intermixing between Fe and Si occurs.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Grain size (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe(2nm)/Si(0.5nm)</td>
<td>9.4</td>
</tr>
<tr>
<td>Fe(2nm)/Si(1.0nm)</td>
<td>10.8</td>
</tr>
<tr>
<td>Fe(2nm)/Si(1.5nm)</td>
<td>1.5</td>
</tr>
<tr>
<td>Fe(2nm)/Si(2.0nm)</td>
<td>1.8</td>
</tr>
</tbody>
</table>

Figure 2 shows magnetization curves before ion irradiation for [Fe(2nm)/Si(t$_{Si}$)]$_{30}$ MLs at RT. It is clear that the interlayer exchange coupling (IEC) between Fe layer is Antiferromagnetic coupling (AFC) only at t$_{Si}$=1 nm. The characteristic features of antiferromagnetic (AF) coupled multilayer is that the in-plane hysteresis loops have a large saturation field $H_s$ and low remanence $M_r$ relative to $M_s$, as summarized from Fig. 3. There is a peak in $H_s$ and low remanence $M_r$ for Silicon thickness t$_{Si}$=1.0 nm which is clearly due to AF coupling, besides this thickness there is no additional evidence of AF behaviour.

The strength of AF coupling J, which was determined from the following equation $J=H_s.M_s.t_{Si}/4$ is around 0.005 erg/cm$^2$. This value is in agreement with that of P. Bruno et.al [9] for interlayer magnetic coupling between ferromagnetic films separated by an insulating layer.
Si thickness dependence of Magnetoresistance ratio in [Fe(2nm)/Si(tSi=0.5-2nm)30].

Figure 4 was shown Silicon thickness dependence in plane magnetoresistance ratio for [Fe(2nm)/Si(1nm)]30 MLs at room temperature. This phenomenon of the dependence of magnetoresistance ratio in this system may be explained by the phenomenological equation proposed by Dieny et.al [10], as

\[
\text{GMR}(t_{Si}) = \left( \frac{\Delta R}{R} \right)_0 \frac{[1-\exp(-t_{Si}/l_{Si})]}{(1+t_{Si}/t_0)}
\]

The \((\Delta R/R)_0\) represent magnetoresistance (MR) ratio. The factor \([1-\exp(-t_{Si}/l_{Si})]\) is the angle averaged probability for an electron of the type with largest mean free path to be scattered within the Si layer before being scattered diffusely at the outer boundary. This factor explains the fact that if the Si layers are too thin, the contrast between mean free paths for both spin directions will decrease, because the longer mean free paths is lowered by the diffuse scattering at the other boundaries, so that the MR ratio is lowered. The denominator expresses the shunting effect due to the Si layer. The parameter \(t_{Si}\) is represent the Si spacer thickness, while the parameter \(l_{Si}\) is related to electron mean free in the Si layer.

### Post Argon Ion Irradition

Several experiments on Fe(2nm)/Si(1nm) MLs after ion irradiation were performed. The measurements included XRD, VSM and Magnetoresistance measurement at RT. As shown in Fig. 5, there is no difference in the XRD patterns of pre irradiation and post irradiation at 400keV Ar ion dose \(1\times10^{14} \text{ ion/cm}^2\) [Fe(2nm)/Si(1nm)]30 MLs, indicated by almost the same integrated intensity in superlattices peak. The integrated intensity are 27,164 and 27,167 for pre and post irradiation sample at ion dose 1x10^{14} \text{ ion/cm}^2, respectively. From the pattern it is clear that multilayer grown on (110)Fe texture with spacer layer also became crystalline, as indicated by the existence of superlattices peak at low angle scattering. The periods in the layered structure before irradiation were estimated to be 3.069 nm and 3.072 nm, respectively.

These results imply that the surface of interface structures after ion irradiation became rough although the layer structures are maintained. But further data from Small Angle X-ray Scattering (SAXS) is needed to clarify this suggestion.

**Fig. 5.** X ray diffraction pattern of Fe(2nm)/Si(1nm) MLs for as deposited sample (upper), and post 400keV Argon ion irradiation at dose \(1\times10^{14} \text{ ion/cm}^2\) (lower).
irradiated samples showed that the MR ratio of irradiated sample is lower than that of as deposited samples, as tabulated in Table 1. It seems that the MR properties of Fe/Si MLs are similar to the metallic superlattice Fe/Cr [11] and Co/Cu MLs [12], which exhibit Magnetoresistance properties.

![RBS Pattern for Fe2nm/Si1nm MLs (B2)](image)

**Fig. 6.** Rutherford Back Scattering pattern of [Fe(2nm)/Si(1nm)]₃ MLs.

![Evolution of M-H curve](image)

**Fig. 7.** Evolution of M-H curve for original sample of Fe(variation)/Si(1nm) multilayer (blue curve) and post Ar ion irradiation at a dose of 3×10¹³ ion/cm² (red curve) and 1×10¹⁴ ion/cm² (black curve).

<table>
<thead>
<tr>
<th>Sample</th>
<th>not irradiated (zero dose)</th>
<th>Ion dose 3×10¹³ ion/cm²</th>
<th>Ion dose 1×10¹⁴ ion/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe2nm/Si1nm</td>
<td>0.16</td>
<td>0.07</td>
<td>0.05</td>
</tr>
<tr>
<td>Fe3nm/Si1nm</td>
<td>0.22</td>
<td>0.10</td>
<td>0.07</td>
</tr>
</tbody>
</table>

**Table 2.** Magnetoresistance (MR) Ratio for Fe/Si1nm Multilayer (unit: %)

**CONCLUSIONS**

Fe(2nm)/Si(0.5-2nm) multilayer thin film have been developed using Helicon plasma sputtering and effects of Argon ion irradiation on its interfacial structure, magnetic and transport properties has been investigated. It is found that for [Fe(2nm)/Si(tₛi)]₃₀ multilayer the antiferromagnetic properties were observed when Silicon thickness tₛi is 1.0 nm and become ferromagnetically coupled post Argon ion irradiation. 400 keV Argon ion irradiation caused Magnetoresistance (MR) properties to decrease gradually from 0.16% to 0.07% and 0.05% post irradiation fluence of 3×10¹³ ion/cm² and in the 1×10¹⁴ ion/cm², respectively. This phenomenon seems to be related to the roughness of multilayer surface rather than to the layer structure since there is no any difference in the superlattices peak intensity.

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**REFERENCES**

