^{181}Ta ($\leftarrow ^{181}\text{Hf}$) Time-Differential Perturbed Angular Correlation Spectroscopy of Hf/Fe Multilayers

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Hf/Fe multilayers are soft magnetic materials and are a good candidate for magnetic heads of storage devices. We have investigated Hf/Fe multilayers using time-differential perturbed angular correlation (TDPAC) spectroscopy of ^{181}Ta ($\leftarrow ^{181}\text{Hf}$), examining specifically the magnetic nature of the non-magnetic Hf layers sandwiched between two ferromagnetic Fe layers. We prepared multilayer samples with Hf layers of various thicknesses, $[\text{Hf}(x \text{ nm})/\text{Fe}]_n$ ($x = 0.5, 1.0, 2.0, 4.8, 10.0$). Three different hyperfine magnetic fields 53(1) T, 43(1) T and 2(1) T were detected in the samples of $[\text{Hf}(2 \text{ nm}, 4.8 \text{ nm}, 10 \text{ nm})/\text{Fe}]_n$. The hyperfine magnetic field of 53(1) T corresponds to Hf atoms in a turbulent interface between the Hf and Fe layers. The value of 43(1) T corresponds to Hf atoms inside Hf layers within 0.5 nm from the interface. The value of 2(1) T corresponds to Hf atoms inside the Hf layers within 3.5 nm of the interface.

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1. Introduction

Research on soft magnetic properties of multilayers is carried out in the field of magnetic thin films.¹⁻⁴⁾ By making an appropriate multilayer, structure of magnetic walls, size of magnetic domains and magnitude of magnetic anisotropy can be controlled. The crystal grain of a thin film can be controlled by fabricating an appropriate multilayer, in order to make a highly magnetic film. Hf/Fe metal multilayers are soft magnetic materials and research has been carried out on these materials mainly for their use in magnetic heads. In this paper we report investigations into the magnetic nature of non-magnetic Hf layers sandwiched between ferromagnetic Fe layers. We prepared Hf/Fe multilayer samples with varying thicknesses of the Hf layers; Hf thicknesses of 0.5 nm, 1 nm, 2 nm, 4.8 nm and 10 nm were used.

2. Experimental Procedure

We produced all samples by an electron beam evaporation method under high vacuum. The electron beam evaporation method uses an accelerated electron beam as an energy source for the evaporation of atoms and molecules. Electronic energy is concentrated at the surface of the evaporation source. This method is suited to high-speed evaporation and it is possible to make a high purity film.

The metal ingots were placed in copper crucibles, which were cooled with water. A high vacuum was maintained with a turbo molecular pump and a liquid N₂ trap; the degree of vacuum at the time of sample production was about 1×10^{-5} Pa. The current by the accelerated electron beam was 60–80 mA in the case of Fe, and 100–180 mA in the case of Hf. We paid particular attention to stabilizing the

Table 1 Number of layers in the Hf/Fe multilayers. Thickness of the individual layers is given in parentheses.

Sample	Number of layers
Hf (0.5 nm)/Fe (50 nm)	81
Hf (1 nm)/Fe (200 nm)	19
Hf (2 nm)/Fe (200 nm)	17
Hf (4.8 nm)/Fe (200 nm)	5
Hf (10 nm)/Fe (200 nm)	5

evaporation rate in vacuum; the evaporation rate in vacuum is important because the nature of the deposited film depends on it. The evaporation rate in vacuum was 0.05–0.1 nm/s in the case of the Hf layers, and 0.2 nm/s in the case of the Fe layers. We measured the thicknesses of the layers with a thickness monitor with a quartz vibrator method. The thicknesses and number of layers are shown in Table 1. Since used a Fe layer thickness of 200 nm for Hf layers of 1, 2, 4.8 and 10 nm, the Fe layers had sufficient thickness compared to the Hf layers. For the 0.5 nm Hf layers, an Fe layer of 50 nm was used. We would have preferred to use a thickness of 200 nm in this case as well, but because such a large number of layers were used we were concerned that there would not be sufficient Fe in the evaporation source. Even in this case the Fe layer is sufficient thickness compared to the Hf layer. The number of layers was varied from 5 to 81 so that the amount of Hf in each specimen (the activity of a specimen) was almost constant.

The films were irradiated by reactor neutrons at Kyoto University Research Reactor Institute (KUR-RI), Osaka, Japan. This produced ^{181}Hf in the films by the reaction $^{180}\text{Hf}(n, \gamma)^{181}\text{Hf}$. We made time-differential perturbed angular correlation (TDPAC) measurements at room temperature at the Toyonaka Annex of the Osaka University Radioisotope

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Table 2 Parameters of the 482 keV level of ^{181}Ta . I is the nuclear spin, $\tau_{1/2}$ is the life-time of the intermediate state, g is the g factor and Q is the quadrupole moment.

I	$5/2^+$
$\tau_{1/2}$	10.8 ns
g	+1.30
Q	+2.80 barns

Center. We used a conventional four-detector arrangement in a plane at 90° angular separations. We made ^{181}Ta ($\leftarrow ^{181}\text{Hf}$) TDPAC measurements using ^{181}Ta as a probe nucleus. The angular correlations were measured by the 133-482 keV cascade γ decay of ^{181}Ta after β^- decay of ^{181}Hf (γ_1 : 133 keV; γ_2 : 482 keV). Information about the hyperfine interaction in the intermediate state ($I = 5/2$) of ^{181}Ta was obtained. The parameters of the intermediate state are shown in Table 2.

3. Results and Discussion

Hf is a nonmagnetic material. However, Hf atoms at the interface of a Hf layer with a ferromagnetic Fe layer expect a strong magnetic influence from Fe layer. Therefore, for the case $k_1 = k_2 = 2$, where $k_i = 0, 2, \dots$ is the wave vector of γ_i ($i = 1, 2$), the perturbation coefficient $G_{k_1 k_2}$ is expressed as follows.⁵⁾

$$G_{22}(t) = \frac{1}{5}(1 + 2 \cos(\omega_L t) + 2 \cos(2\omega_L t)) \quad (1)$$

Here, t is time and ω_L is the Larmor frequency. In the case when an external magnetic field is vertical to the detector plane, a component $2\omega_L$ appears in the perturbation coefficient. In the case when the angular correlation is the precession of a general axis, the component of the frequency is ω_L . The amplitude of $2\omega_L$ and ω_L depends on the direction of a magnetic field. The ^{181}Ta TDPAC spectra of the [Hf (0.5 nm, 1 nm, 2 nm, 4.8 nm, 10 nm)/Fe] $_n$ samples are shown in Fig. 1. They are analyzed with a model in which the spectrum consists of three components rising from 3 different Hf sites, giving

$$G_{22}(t) = \sum_{m=1}^3 f_m \{1 + 2 \exp[-(\delta_m \omega_m t)^2] \cos(\omega_m t) + 2 \exp[-(2\delta_m \omega_m t)^2] \cos(2\omega_m t)\} \quad (2)$$

Here, ω_m is the Larmor frequency. We suppose that the Larmor frequency has a Gaussian distribution. The factor f_m is the existence probability of each of the three components:

$$\sum_{m=1}^3 f_m = 1. \quad (3)$$

We have analyzed G_{22} with 3 components because it is not possible to fit the experiment result satisfactorily when we analyzed it with 1 or 2 components. An analysis with 4 or more components may fit the experimental results better, but because as the number of parameters increases, the analysis becomes complicated and the physical meaning disappears. An analysis with 3 components fits the experimental results sufficiently; the added complexity of analyzing with 4 or

Fig. 1 ^{181}Ta TDPAC spectra of Hf/Fe multilayers. Solid lines are fitted curves using eq. (2). (a) [Hf (0.5 nm)/Fe] $_n$, (b) [Hf (1 nm)/Fe] $_n$, (c) [Hf (2 nm)/Fe] $_n$, (d) [Hf (4.8 nm)/Fe] $_n$, (e) [Hf (10 nm)/Fe] $_n$.

more components does not add enough accuracy to be worthwhile.

If the amplitudes are small, the Larmor frequencies are not observed clearly. If the magnetic moment is parallel to the plane of detectors, it is difficult to observe the Larmor frequency. From the result of Mössbauer spectroscopy, it was found that most of the magnetic moments of the Fe atoms tend to be perpendicular to the plane of the detectors. The observed wavelength of the Larmor frequency is close to the time resolution of the measurement system and anisotropy is small.

The results of the analysis are shown in Fig. 2, Fig. 3 and Fig. 4. When the Hf layers are less than 2 nm in thickness, the parameters f and ω show a sharp change. Magnitude of the hyperfine magnetic fields determined from [Hf (2 nm, 4.8 nm, 10 nm)/Fe] $_n$ samples are same. These results suggest that the

Fig. 2 Relative occupation probability as a function of Hf thickness.

Fig. 3 Angular frequency as a function of Hf thickness.

Fig. 4 Width of Gaussian distribution of frequency as a function of Hf thickness.

hyperfine fields at Hf nuclei located at the positions over 2 nm apart from a ferromagnetic Fe layer do not show any appreciable difference. On the other hand, the values of the frequency of site 1 and site 2 in the samples of [Hf (0.5 nm, 1 nm)/Fe]_n are clearly different from the other samples and are thought to depend on the state of the Hf atom.

Here, we discuss the component of site 1. In the sample of [Hf (0.5 nm, 1 nm)/Fe]_n, the value of the hyperfine magnetic field was determined to 61(1) T (Fig. 3). This is near the value 59(1) T⁶⁾ of the hyperfine magnetic field of Hf atoms existing as an isolated impurity in an Fe matrix. Therefore, the component of site 1 of [Hf (0.5 nm, 1 nm)/Fe]_n corresponds to the value of a Hf atom that penetrates into a Fe layer. A value of 53(1) T is obtained for the hyperfine magnetic field of the site 1 component in the samples [Hf (2 nm, 4.8 nm, 10 nm)/Fe]_n (Fig. 3). This corresponds to a Hf atom is in the area of a turbulent interface with an Fe atom as the nearest atom. The frequency of site 2 and site 3 of the samples [Hf (2 nm, 4.8 nm, 10 nm)/Fe]_n have a common value. These components correspond to the Hf inside of a Hf layer. Monolayer thickness of Hf atoms is about 0.25 nm from the unit cell size of the hcp Hf crystal. The Hf atoms of the sample [Hf (2 nm)/Fe]_n correspond to 8 monolayers. The relative probability of existence for site 2 and site 3 is 1:1 (Fig. 2). Therefore, site 2 in the samples [Hf (2 nm)/Fe]_n corresponds to Hf at 2 monolayers (0.5 nm) outside from an interface with a Fe layer, and site 3 corresponds to Hf of 4 monolayers (1 nm) inside from the interface. Here, existence probability of site 1 is little (Fig. 2). The hyperfine magnetic field at Hf nucleus located in the 1st and 2nd Hf layer from the interface with an Fe layer (site 2) is 43(1) T (Fig. 3). The hyperfine magnetic field of Hf inside of a Hf layer (site 3) is

2(1) T. In the same way, the ranges of the components of site 2, site 3 of the specimens of [Hf (4.8 nm, 10 nm)/Fe]_n are calculated from relative probability of the existence. Site 2 in the samples [Hf (4.8 nm, 10 nm)/Fe]_n corresponds to Hf at 7, 14 monolayers (1.8, 3.5 nm) outside from an interface with a Fe layer, and site 3 corresponds to Hf at 5, 12 monolayers (1.2, 3 nm) inside of the layer, respectively.

Magnetic influence by the ferromagnetic Fe layer extends to 1.8 nm in the sample of [Hf (4.8 nm)/Fe]_n. This influence extends 3.5 nm in the sample of [Hf (10 nm)/Fe]_n. Therefore, almost all the Hf layers receive the influence of the hyperfine magnetic field from a Fe layer in the samples of [Hf (2 nm, 4.8 nm)/Fe]_n. The range of the component of the weak hyperfine magnetic field reaches 3.5 nm from the interface with a Fe layer. The value 42(1) T of the hyperfine magnetic field of the component of site 2 in the sample of [Hf (1 nm)/Fe]_n corresponds to the Hf inside a layer. However, the value 18(1) T of the hyperfine magnetic field of site 3 in the sample of [Hf (0.5 nm, 1 nm)/Fe]_n differs greatly from the value for the samples of [Hf (2 nm, 4.8 nm, 10 nm)/Fe]_n. This value of 18(1) T is close to the hyperfine magnetic field (16(1) T⁶⁾) that a Hf atom in a cluster state receives from a ferromagnetic Fe atom. Hf atoms in a cluster state exist in the sample of [Hf (0.5 nm, 1 nm)/Fe]_n. Most of the Hf atoms are in a cluster state from the value of the existence probability *f_m*.

Now we consider *δ*, the distribution of the frequency. The component *δ* of site 1 is zero. We think, therefore, that the Hf atoms in [Hf (1 nm)/Fe]_n exist alone in an Fe matrix. The Hf atoms in the samples of [Hf (2 nm, 4.8 nm, 10 nm)/Fe]_n are in various states. The wavelength of the Larmor frequency of site 1 is near the time resolution of the measurement system and hence it is very difficult to evaluate the distribution and difficult to discuss the distribution of the frequency of the samples of [Hf (2 nm, 4.8 nm, 10 nm)/Fe]_n. *δ* of the frequency component for site 3 does not vary much. On the other hand, the distribution of the frequency component for site 2 changes considerably. The component of site 2 corresponds to the 1st and 2nd Hf layers from the interface with a Fe layer. The Hf layers in the samples of [Hf (2 nm)/Fe]_n are as thin as 8 monolayers. A Hf layer receives influence from Fe layers on both sides. Therefore, the distribution of the frequency is broad.

4. Conclusion

We prepared Hf/Fe multilayers, [Hf (0.5 nm, 1 nm, 2 nm, 4.8 nm, 10 nm)/Fe]_n, by using an electron beam evaporation apparatus. We measured ¹⁸¹Ta ($\leftarrow$ ¹⁸¹Hf) TDPAC of these samples at room temperature. The perturbation of the hyperfine magnetic field from the Fe layer was observed. When 3 components were considered, the experimental data could be analyzed well. A large hyperfine magnetic field of 61(1) T was observed in the sample of [Hf (1 nm)/Fe]_n. This corresponds to an isolated Hf atom in an Fe matrix. Most of the Hf atoms are in clusters, like islands.

Three different hyperfine magnetic fields, 53(1) T, 43(1) T and 2(1) T, were observed in the samples of [Hf (2 nm, 4.8 nm, 10 nm)/Fe]_n. Taking into account the proportions of the components, the value of the hyperfine magnetic field 53(1) T can be considered to correspond to Hf atoms existing

at a turbulent interface. The value of the hyperfine magnetic field 43(1) T corresponds to Hf atoms existing inside the Hf layers within 0.5 nm from the interface. The value of the hyperfine magnetic fields 2(1) T corresponds to Hf atoms inside the Hf layers within 3.5 nm from the interface.

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