Using Anomalous Dispersion Effect for Maximum Entropy Method Analysis of X-ray Reflectivity from Thin-Film Stacks

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Abstract. We used the anomalous dispersion effect and the maximum entropy method (MEM) to analyze X-ray reflectivity had been used, a theoretical layer models were not necessary. We measured the reflectivity of a layer stack at two x-ray wavelengths, the near Ru-K edge and pre Ru-K edge, for a sample with a 0.8 nm thick Ru layer at the stack top. We calculated the differential reflectivity between the two wavelengths by subtracting the near Ru-K edge curve from the pre Ru-K edge curve. We analyzed the differential curves using the Fourier transform (FT) method and MEM. The results showed the interface depths of the samples from the surface to the interfaces. The peak width produced by MEM analysis was narrower than the peak width produced by the FT analysis. Furthermore, using MEM analysis, it is possible to analyze the interface of the thin film in the vicinity of the surface. This result suggests the promising candidate for analyzing the layer structure of a sample by using the anomalous dispersion effect and MEM analysis.

1. INTRODUCTION

Thin-layered stack materials with layer thickness in the nanometer range are increasingly being used in modern technology (e.g., in microelectronics and magnetic devices). Giant magneto-resistive (GMR) spin-valve heads, in particular, have been investigated for use in high-recording-density hard drives because, due to their high sensitivity, they can be used for reading magnetic records [1]. Recently, many researchers have studied using perpendicular magnetic-recording media [2] and advanced spin-valve heads with magnetic tunnel junctions for advanced hard drives [3].

Giant magneto-resistive structures consist of two ferromagnetic layers separated by a noble metal spacer with a thickness of a few nanometers. The GMR structures are deposited on antiferromagnetic PtMn thin layers. Their magnetic properties strongly depend on the thickness of each layer.

X-ray reflectivity is used for investigating layer thickness, electron density, and interface roughness [4-6]. For layered materials, x-rays at grazing incidence are reflected and transmitted at each interface. Because of the interference between the x-rays reflected at the various interfaces, fringes are seen in the reflectivity. These oscillations of reflectivity curves reveal the thicknesses of different layers. The least-squares method with theoretical models has usually been used to analyze reflectivity [7-8]. However, to obtain the best-fit result for a measured reflectivity curve is difficult unless the thicknesses of all the layers in a sample are known. Fourier transform analysis of the reflectivity can determine each layer thickness [9], but it cannot determine the exact stacking order. However, it is possible to determine the stacking order using wavelet transform analysis [10]. However, using
Fourier and wavelet transforms to analyze layered stacks with similar thicknesses is difficult. Therefore, we previously investigated layer thicknesses and stacking orders using the fast Fourier transforms method [12].

We investigated layer thicknesses and stacking orders using the MEM. We measured the reflectivity of x-rays that had been affected by anomalous dispersion. We then plotted reflectivity curves from these results and analyzed them using the MEM.

2. DIFFERENTIAL REFLECTIVITY

In this section, we present a brief introduction to the use of frequency analysis of differential reflectivity data. Further information can be found in other papers [7, 8, 12].

Figure 1 is a schematic drawing of x-ray specular reflection by a stacking layer. The scattering vector is shown with $q = (4\pi \sin \theta)/\lambda$. The amplitude of reflectivity ($R_j$) from interface $j$ is written as

$$R_j = a_j \frac{R_{j+1} + F_{j+1}}{R_{j+1} - F_{j+1}}$$

$$\gamma_j = q^2 - 2(\xi_j + i\eta_j)$$

$$a_j = \exp\left[-i \frac{\gamma_j - d_j}{4}\right]$$

$$F_{j+1} = \frac{Y_j - Y_{j+1}}{Y_j + Y_{j+1}} \exp\left[-\frac{\gamma_j - d_{j+1}}{4}\right]$$

$$n_j = 1 - (\delta_j + i\beta) = 1 - \left(\frac{\lambda}{4\pi}\right)^2 (\xi_j + i\eta_j)$$

-------------------- (1)

Thus, reflectivity ($|R_1|^2$) of the stack may be calculated using equation (1). We denote the limit of $q^2 \gg \xi_j$ and $q^2 \gg \eta_j$. The reflectivity of the stack was approximately given by the following equations:

$$R_{j+1} = a_j \frac{R_j + F_j}{R_j - F_j}$$

$$|R_1|^2 = \sum_{j=2}^{N+1} \frac{\Delta \xi_j^2 + \Delta \eta_j^2}{4q^2} + \sum_{j=2}^{N+1} H_{j,k}$$

$$H_{j,k} = \frac{1}{2q^2} \left(\Delta \xi_j^2 + \Delta \eta_j^2\right)^{\frac{1}{2}} \left(\Delta \xi_k^2 + \Delta \eta_k^2\right)^{\frac{1}{2}} \cos(\phi + \psi)$$

$$\phi = \gamma_j \cdot d_j + \gamma_{j+1} \cdot d_{j+1} + \cdots + \gamma_{j+k} \cdot d_{j+k} = q(d_j + d_{j+1} + \cdots + d_{j+k})$$

$$\tan(\psi) = \frac{(\Delta \xi_j \cdot \Delta \xi_k)}{(\Delta \eta_j \cdot \Delta \eta_k)}$$

-------------------- (2)

An anomalous dispersion effect was created using layer 2 (N=2) elements. Wavelength ($\lambda_i$) is near edge and wavelength ($\lambda_2$) is pre edge. The differential reflectivity is defined as subtracting the reflectivity at wavelength ($\lambda_i$) from that at wavelength ($\lambda_2$). The thickness of layer 2 ($d_2$) is limited to about zero. Expression of the differential reflectivity is derived from the equations given by equation (2), as follows.
Equation (3) has the form \((B+C \cos q \cdot z_j)/A\), where \(A\), \(B\) and \(C\) are non-oscillating terms. The fact \(\gamma=q \cdot z_j\) indicates that the differential reflectivity data is converted to the distribution of \(z_j\) by frequency analysis methods.

3. EXPERIMENTS

Two samples were sputtered onto a five-inch diameter silicon disk in a static magnetic field. The first sample (A) was deposited on the silicon disk in the order tantalum (Ta) (10 nm thick), platinum-manganese (PtMn) (10 nm), and ruthenium (Ru) (0.8 nm). The Ru layer being the cap layer of the sample. The other sample (B) was deposited on the silicon disk in the order Ta (10 nm), PtMn (15 nm), Ta (10 nm), and Ru (0.8 nm). The Ru layer of the sample (B) was not able to prevent Ta being oxidized completely. Therefore, the layer of oxides of 2 nm can be done between Ru and Ta layers, and the thickness of Ta layer had decreased to 9 nm. The numbers in parentheses are nominal thicknesses. The reflectivity curves were measured at two energies using a diffractometer (BL16XU, SPring-8) [11].

The incidence x-ray energies were 99.87% of Ru K-edge energy at 22.09 keV (In this paper, this energy is called “near Ru K-edge”) and 98.92% of the energy at 21.88 keV (In this paper, this energy is called “pre Ru K-edge”). The anomalous dispersion factors of experimental energies are shown in Table I. They were measured using a 0-2θ scanning technique and simultaneously transformed from incident angles to scattering vectors. The reflectivity analysis consists of two steps. In the first step, we calculated the differential reflectivity curve by subtracting the near K-edge curve from the pre K-edge one. In the second step, we transformed the differential reflectivity curve using the Fourier transform (FT) method and Maximum Entropy method (MEM). The result of the transformation gives the interface depth of the measured sample. The MEM analysis was used “Auto-Re-greeive Model” of H. Akaike theory [13]. Then the disregard of higher-order terms was decided by the algorithm of the Burg method [14].

| Materials  | Pre Ru K-edge (22.09keV) | Near Ru K-edge (21.88keV) |
|------------|--------------------------|--------------------------|
|            | f’                        | f’’                       |
| Ruthenium  | -5.996                    | 0.5448                    |
| Platinum   | -1.3418                   | 5.737                     |
| Manganese  | 0.2307                    | 0.4712                     |
| Tantalum   | -0.9385                   | 4.422                      |
4. RESULTS AND DISCUSSION

4.1. Sample (A): Ru (0.8nm)/PtMn (10nm)/Ta (10nm)/Si substrate

Figure 2 shows the reflectivity curves of sample (A) at two energies and the differential reflectivity curve obtained by subtracting the pre-edge from the near-edge curve. The two curves of reflectivity look the same. The fringes on the differential reflectivity curve show interference between the surface and interface of PtMn/Ta, the surface and interface of Ta/Si, and the interfaces of PtMn/Ta and Ta/Si. Using the anomalous dispersion that occurs in the surface layer only changes the oscillation of the reflection from the surface layer. The interference fringe that is between the interfaces of PtMn/Ta and Ta/Si is disappeared by subtracting the near K-edge reflectivity from the pre K-edge reflectivity. Figure 3 shows the result of analyzing the differential reflectivity. The solid line shows the result of the FT analysis and the broken line shows the result of MEM analysis. Using FT, peaks are found at 10.8 and 22.2 nm in the results. Using MEM, peaks are found at 10.8, and 21.9 nm. The positions of the peaks agree well with the distance from the Ru layer. The peaks FWHM (Full Width at Half Maximum) given by FT analysis were 0.89, and 1.02 nm. However, the peaks FWHM given by MEM analysis were narrower than the peaks FWHM given by FT analysis. The reflectivity curve of the pre K-edge was analyzed using a best fit to the calculated reflectivity curve. The fitting result revealed the depth of the interfaces (PtMn/Ta and Ta/Si substrate) to be 10.22, and 20.97 nm. This suggests that the differential reflectivity curve reveals the interfaces depth from the cap layer.

4.2. Sample (B): Ru (0.8nm)/Ta oxide (2nm)/Ta (9nm)/PtMn (15nm)/Ta (10nm)/Si substrate

Figure 4 shows the reflectivity curves of sample (B) at two energies and the differential reflectivity curve obtained by the subtraction of two energy curves. These oscillations of the reflectivity curve reveal the distance between the Ru layer and the other interfaces. Figure 5 shows the results of analyzing the differential reflectivity in figure 4. The...
solid line shows the magnitude of the FT of differential reflectivity and the broken line shows the result of MEM analysis of the reflectivity. Peaks are not found at 15 nm. This suggests that the oscillation correlating to the thickness of PtMn in the reflectivity curve has disappeared by subtracting the pre K-edge curve from the near K-edge curve. The reflectivity curve of the pre K-edge was analyzed by best fitting a curve to the calculated reflectivity curve. The results of the best fitting are listed in Table II. The fitting result revealed that the Ta layer was oxidized to a depth of 2.3 nm. Table II shows the result of differential reflectivity obtained using the FT and MEM. The result of MEM analysis shows the Ta oxide layer. It is not possible to use the FT to analyze the oxide layer. However,

| Materials  | Best-fit analysis (nm) | FT analysis (nm) | MEM analysis (nm) |
|------------|------------------------|------------------|-------------------|
| Ru         | 0.75                   | –                | –                 |
| Ta oxide   | 2.33                   | –                | 1.60              |
| Ta         | 9.24                   | 10.66            | 9.74              |
| PtMn       | 13.90                  | 14.21            | 13.73             |
| Ta         | 10.02                  | 9.74             | 10.02             |
| Si         | Substrate              | Substrate        | Substrate         |
the result of differential reflectivity obtained using MEM did not produce a highly accurate analysis of the oxide layer. The information from Table II suggests that the minimum depth of the interface from the surface that can be analyzed is about 2 nm under these experimental conditions using MEM.

5. CONCLUSION

The strong possibility that the layer structure in measured samples can be analyzed using the anomalous dispersion effect and maximum entropy methods. The stack information is given by the distance from the cap layer to the interface. The minimum depth of the interface from the surface that can be analyzed under these experimental and analytical conditions is about 2 nm.

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