

## Effects of a Au-Cu Back Layer on the Properties of Spin Valves

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We have studied the effect of Au-Cu back layer system  $\sim 10$  Å thick on the properties of a spin valve. The back layers were Cu, Au, co-sputtered  $\text{Cu}_x\text{Au}_{1-x}$ , laminated  $[\text{Au}/\text{Cu}]_n$  and bi-layer  $[\text{Au}/\text{Cu}]$ . When Au was added to the Cu, the resistance of the spin valve abruptly increased most likely due to impurity scattering. The GMR values were not increased significantly for all the structures. In the case of co-sputtered  $\text{Cu}_x\text{Au}_{1-x}$ , the changes in the resistance,  $\Delta R$ , was increased at a composition of  $\sim \text{Au}_{0.5}\text{Cu}_{0.5}$ . This increase in  $\Delta R$  is due to increase in the resistance and not from the enhanced spin-dependent scattering. The structural analyses showed that the orthorhombic  $\text{Au}_{0.5}\text{Cu}_{0.5}$  was formed in the back layer instead of the face-centered tetragonal  $\text{Au}_{0.5}\text{Cu}_{0.5}$  as we expected. Thermal annealing over  $400^\circ\text{C}$  may be required to have face-centered tetragonal in the  $10$  Å thick ultra-thin film. In the case of a laminated or bi-layered back layer, the properties of the spin valve were improved, which may be attributed to the increase in the mean free path of conduction electrons.

**Keywords :** spin valve, back layer, giant magnetoresistance, Au-Cu alloy, XRD

### 1. Introduction

Giant magnetoresistance (GMR) in spin valves have been used as read elements in recording heads for high storage capacity due to their high sensitivity and small noise in reading bits of hard disk drives [1]. The extensive effort to achieve the high performance spin valves during the past decades resulted in a rate of increase of over 60% per year in the areal density of magnetic recording. Because the GMR value (or equivalently the sheet resistance change,  $\Delta R_s$ ) is directly proportional to the performance of a spin valve, the GMR value has been the most important parameter determining the spin-valve head performance, and the aim in the development of the head has been to increase the GMR value. A GMR value of  $\sim 10\%$  is typical in spin valves used in the drive. The ever-increasing need for higher areal density of magnetic disk drives demands a spin-valve read head with a GMR value (or  $\Delta R_s$ ) higher than present read heads.

One attempt in the quest to improve spin valve's performance is to insert a Cu back layer between free and capping layers. The Cu back layer is necessary to keep the soft magnetic properties of a spin valve by separating the free layer from the capping layer. The other

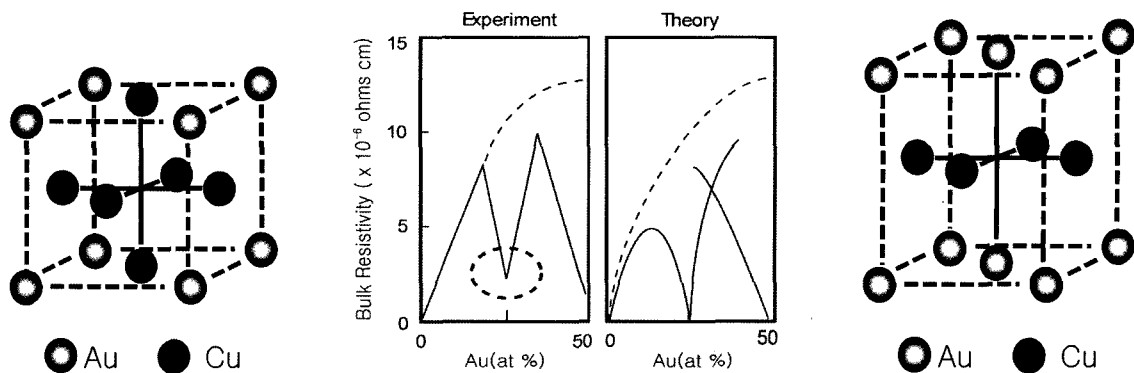
motivation for this Cu back layer is to increase the mean free path of the majority spins in the spin valve, which results in the increase in GMR, since the scattering at the interface free/Cu layers is negligibly small. We have investigated Cu-Au alloy binary system since bulk intermetallic compounds, such as  $\text{Cu}_{0.75}\text{Au}_{0.25}$  and  $\text{Cu}_{0.5}\text{Au}_{0.5}$ , have been reported to decrease the resistance [2, 3]. The Au-Cu alloy has been investigated for a long time as a typical system of disorder-order phase transformation. The ordering can bring an alloy quite close to a translationally invariant configuration in which there is hardly any impurity scattering and therefore its resistivity becomes small. Our motivation was made on the basis of the existence of such an effect. We expected those intermetallic compounds could help increasing a mean free path of conduction electrons so that the parallel resistance would be reduced.

In this paper, we will discuss about Au-Cu binary system as a back layer, and check whether there is indeed the so-called spin-filter effect by replacing Cu with Au-Cu.

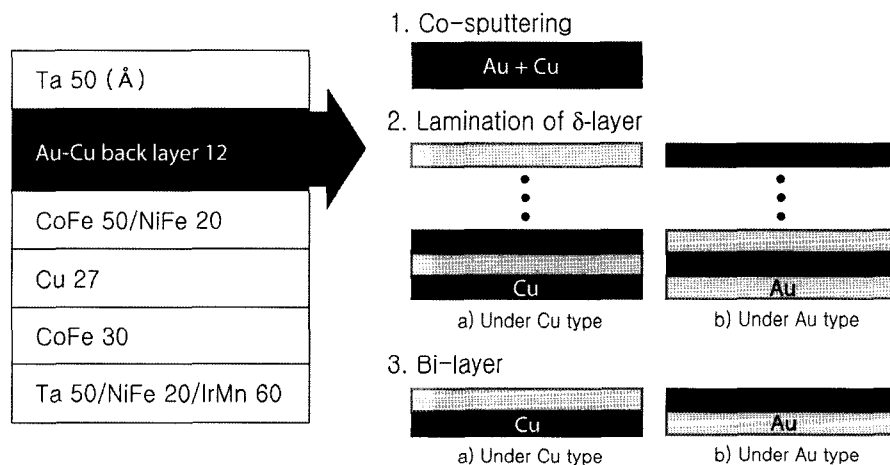
### 2. Experimental Procedures

We have deposited a bottom-type spin valve structure of Ta  $50/\text{Ni}_{80}\text{Fe}_{20}$   $20/\text{Ir}_{81}\text{Mn}_{19}$   $60/\text{Co}_{90}\text{Fe}_{10}$   $30/\text{Cu}$   $27/$

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**Fig. 1.** The structures of inter-metallic compounds of  $\text{Cu}_{0.75}\text{Au}_{0.25}$  (left) and  $\text{Cu}_{0.5}\text{Au}_{0.5}$  (right), and a change in resistivity (experiment and theory) with the content of Au in Cu (center) [4].



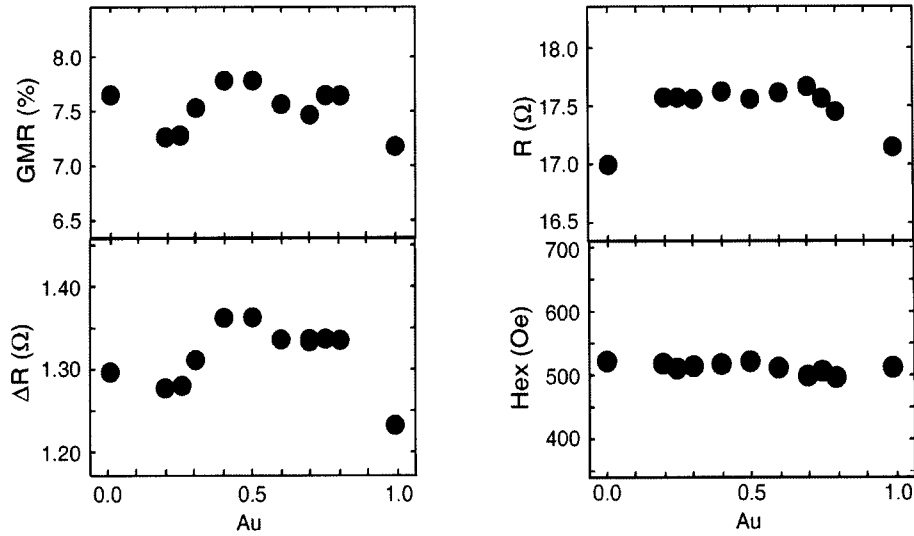
**Fig. 2.** Structures of the Spin Valve with various kind of back layers: 1. a co-sputtered Au-Cu alloy, 2a) lamination of  $\delta$ -layers starting with a Cu layer, 2b) lamination of  $\delta$ -layers starting with an Au layer, 3a) a bi-layer starting with a Cu layer, and 3b) a bi-layer starting with a Au layer.

$\text{Co}_{90}\text{Fe}_{10}$  20/ $\text{Ni}_{80}\text{Fe}_{20}$  20/back layer 10~12/Ta 50 (Å) on a  $\text{Si}/\text{SiO}_2$  (2000 Å) substrate by an UHV DC magnetron sputtering system at room temperature. The base pressure of the chamber was less than  $2 \times 10^{-9}$  Torr. During the deposition process, a magnetic field of  $\sim 90$  Oe was applied to induce unidirectional anisotropy. After deposition, all samples were annealed at 250 °C for 1 hr under a magnetic field of 5 kOe (the base pressure of less than  $3 \times 10^{-7}$  Torr). The back layer was Cu (reference), Au, co-sputtered  $\text{Cu}_x\text{Au}_{1-x}$ , laminated  $[\text{Au}/\text{Cu}]_n$ , or bi-layer  $[\text{Au}/\text{Cu}]$ . Magnetoresistive and electrical properties of spin valves were measured by a DC 4-point probe method by applying a magnetic field in the range of  $-5$  kOe to  $+5$  kOe at room temperature. We used x-ray diffractometry (XRD) for a structural examination of  $\text{Cu}_x\text{Au}_{1-x}$  alloy, and inductively coupled plasma (ICP) mass spectroscopy for a compositional verification.

## 3. Results and Discussion

### 3.1. Au-Cu alloy prepared by co-sputtering

Fig. 3, shows the variations in the magnetoresistive properties of spin valves as a function of Au concentration in the Cu back layer. In particular, we prepared 12 Å-thick alloy layers of Cu,  $\text{Cu}_{0.8}\text{Au}_{0.2}$ ,  $\text{Cu}_{0.7}\text{Au}_{0.3}$ ,  $\text{Cu}_{0.75}\text{Au}_{0.25}$ ,  $\text{Cu}_{0.6}\text{Au}_{0.4}$ ,  $\text{Cu}_{0.5}\text{Au}_{0.5}$ ,  $\text{Cu}_{0.4}\text{Au}_{0.6}$ ,  $\text{Cu}_{0.3}\text{Au}_{0.7}$ ,  $\text{Cu}_{0.25}\text{Au}_{0.75}$ ,  $\text{Cu}_{0.2}\text{Au}_{0.8}$ , and Au, as shown in Fig. 3. As an Au concentration increases in the Au-Cu layer, the GMR ratio and  $\Delta R$  also increases in a similar way. They seemed to have a peak near a CuAu compositional fraction of 1:1. When the back layer was replaced with Au, the GMR ratio and  $\Delta R$  rapidly dropped. For the CuAu alloys, the resistance was higher than that of Cu or Au itself, and stayed at  $\sim 17.6 \Omega$ . The exchange bias field did not change much with Au concentration, since the exchange bias is a



**Fig. 3.** Variations in the GMR,  $\Delta R$ ,  $R$ , and  $H_{ex}$  with a composition of Au in Cu. These spin valves were prepared by co-sputtering.

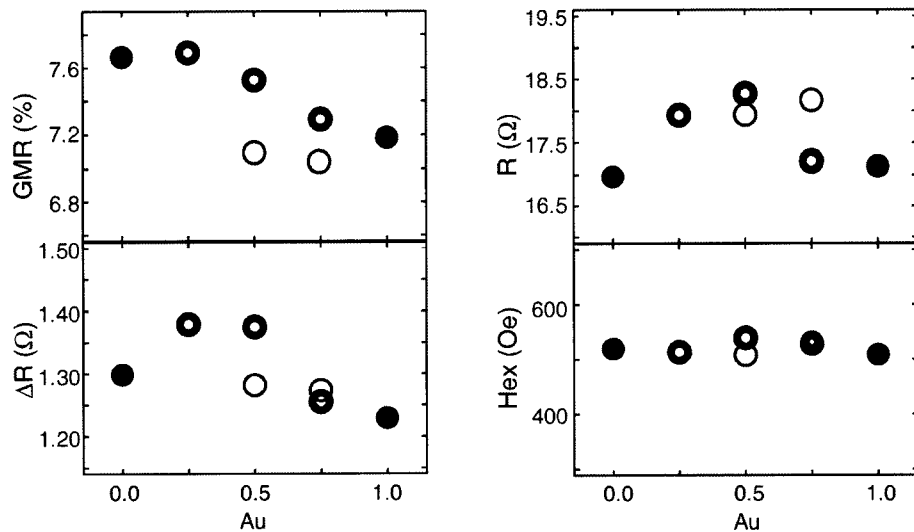
result of atomic-range interaction.

We expected a sudden drop in resistance when we made the inter-metallic compounds, as shown in Fig. 1. However, we could not observe any drops in resistance near the compositions of  $\text{Cu}_{0.75}\text{Au}_{0.25}$  and  $\text{Cu}_{0.5}\text{Au}_{0.5}$ . We have no any way to check whether the structure of an inter-metallic compound was indeed formed or not because 12 Å thickness is too thin to carry out any analysis. However, even though we have no clear evidence on the existence of inter-metallic compounds, judging from the optimum GMR and  $\Delta R$  that were found at the composition of  $\text{Cu}_{0.6}\text{Au}_{0.4}$  and  $\text{Cu}_{0.5}\text{Au}_{0.5}$ , we can conclude that the compositional change led to at least an electronic structural change in a positive way. Considering the high

resistance of the alloys, the little increase in GMR and  $\Delta R$  can be indirect evidence of enhanced spin-dependent scattering by the alloy back layer. The GMR and  $\Delta R$  values of the spin valve with Cu were 7.6% and 1.30 Ω, and those with  $\text{Cu}_{0.5}\text{Au}_{0.5}$  increased to 7.8% and 1.36 Ω, respectively.

### 3.2. Au-Cu alloy prepared by laminations of an Au and a Cu $\delta$ -layer (alternations based on stoichiometric consideration)

These spin valves were prepared by laminating  $\delta$ -layers, an ultra-thin submonolayer, of Au and Cu. There were two types of laminations depending on which layer was deposited first. One is the layer started with Cu



**Fig. 4.** Variations in the GMR,  $\Delta R$ ,  $R$ , and  $H_{ex}$  with a composition of Au in Cu. These spin valves were prepared by laminations of the Au and a Cu  $\delta$ -layer (alternations by stoichiometric consideration). Thin and thick circles correspond to the layer started with Cu (labeled 2a) in Fig. 2.2) and with Au (labeled 2b) in Fig. 2.2) respectively.

(labeled 2a) in Fig. 2. and the other was the one with Au (labeled 2b) in Fig. 2. Prior to prepare the spin valve, we calculated the thickness laminates of  $\delta$ -layers of Cu and Au by considering the densities and the deposition rates of Cu and Au to obtain  $\text{Cu}_{0.25}\text{Au}_{0.75}$ ,  $\text{Cu}_{0.5}\text{Au}_{0.5}$  and  $\text{Cu}_{0.75}\text{Au}_{0.25}$ . To make 12 Å thick, stoichiometric compositions of  $\text{Cu}_{0.25}\text{Au}_{0.75}$ ,  $\text{Cu}_{0.5}\text{Au}_{0.5}$  and  $\text{Cu}_{0.75}\text{Au}_{0.25}$ , we deposited four repetitions of [Au 2.44/Cu 0.57], five repetitions of [Au 1.42/Cu 0.99], and three repetitions of [Au 1.3/Cu 2.74 (Å)]. Figure 4 shows the variations in the magnetoresistive and electrical properties. Interestingly, the properties of spin valves with Cu-started laminations (thin circle) were little worse than those with Au-started (thick circle). The spin valve with an Au-started  $\text{Cu}_{0.75}\text{Au}_{0.25}$  layer did not much gain in GMR and  $H_{\text{ex}}$  but in  $\Delta R$  by 6.2% increase. The resistance was the highest at an alloy of  $\text{Cu}_{0.5}\text{Au}_{0.5}$ , which was consistent with results we described in the previous section.

### 3.3. Au-Cu alloy prepared by laminations of an Au and a Cu $\delta$ -layer (simple alternations)

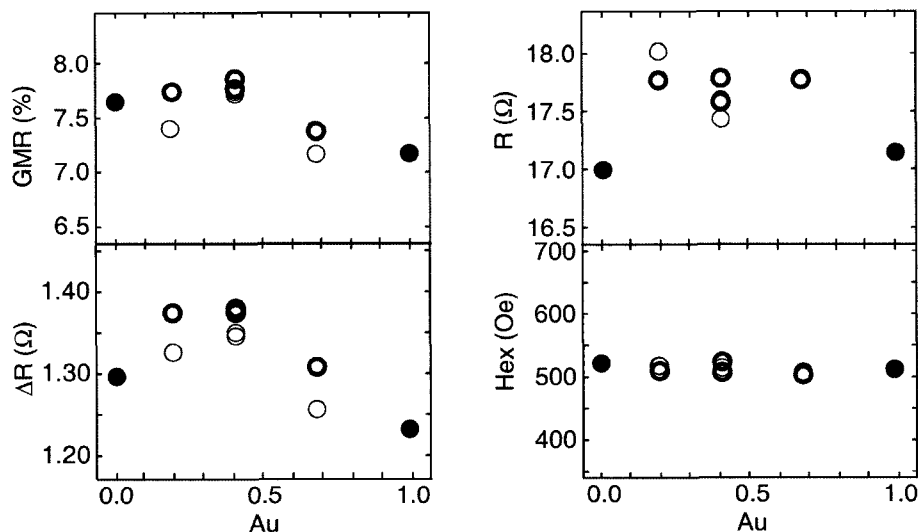
The back layers were fabricated with simple alternations of an Au and a Cu  $\delta$ -layer. Again, we prepared two types of laminations depending on which layer was deposited first. One was the layer started with Cu (labeled 2a) in Fig. 2. and the other was the one with Au (labeled 2b) in Fig. 2. In this time, we did not consider any stoichiometric relationship between thickness and composition. The structures we chose were three repetitions of [Au 1/Cu 3 (Å)], six repetitions of [Au 1/Cu 1 (Å)], three repetitions of [Au 2/Cu 2 (Å)], and three repetitions of [Au 3/Cu 1 (Å)]. The alternations of [Au 1/Cu 3], [Au

1/Cu 1] or [Au 2/Cu 2], and [Au 3/Cu 1] correspond to an atomic composition of  $\text{Au}_{0.19}\text{Cu}_{0.81}$ ,  $\text{Au}_{0.41}\text{Cu}_{0.59}$ , and  $\text{Au}_{0.68}\text{Cu}_{0.32}$ , respectively. Fig. 5. shows the variations in the magnetoresistive and electrical properties of spin valves with Cu-started laminations (thin circle) and with Au-started (thick circle) laminations, respectively.

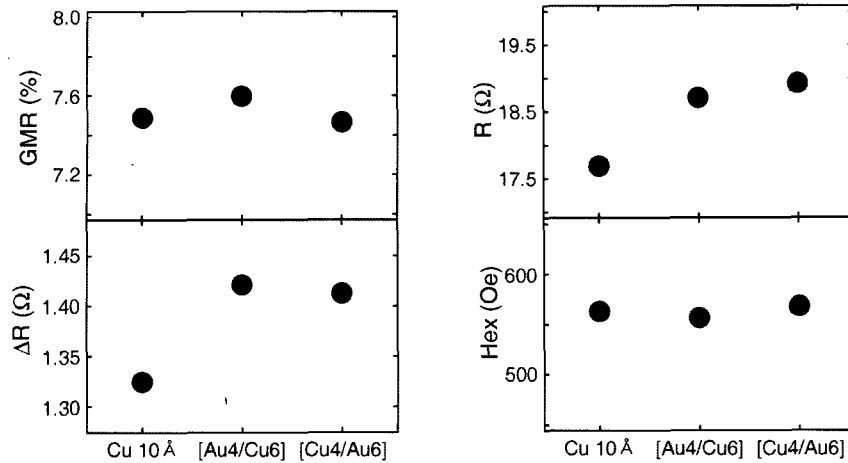
All the properties shown in Fig. 5. are plotted with respect to the calculated stoichiometric compositions of the Au-Cu alloy. The properties showed a similar trend as shown in the spin valves with the laminations described in the previous section. The properties of spin valves with Au-started laminations (thick circle) were little better than those with Cu-started (thin circle). The spin valve with three repetitions of [Au 2/Cu 2 (Å)] showed the best GMR performance; a GMR of 7.9%,  $\Delta R$  of 1.38 Ω. The  $H_{\text{ex}}$  did not show much change for each spin valves.

### 3.4. Au-Cu alloy prepared by a bi-layer of Au and Cu layers

We investigated the spin-filter effect by depositing the back layers with using bi-layers of Au and Cu. Again, we prepared two types of bi-layers depending on which layer was deposited first. One was [Cu 4/Au 6 (Å)] and the other was [Au 4/Cu 6 (Å)], as shown in Fig. 2. The thickness of a bi-layer introduced for a back layer was kept at 10 Å unlike other spin valves described in the previous sections. Fig. 6. shows the variations in the magnetoresistive and electrical properties. The back layer started with Au showed little improved GMR performance. For example, the  $\Delta R$  was increased by ~8%. However, we could not achieve any dramatic improvement in other properties.



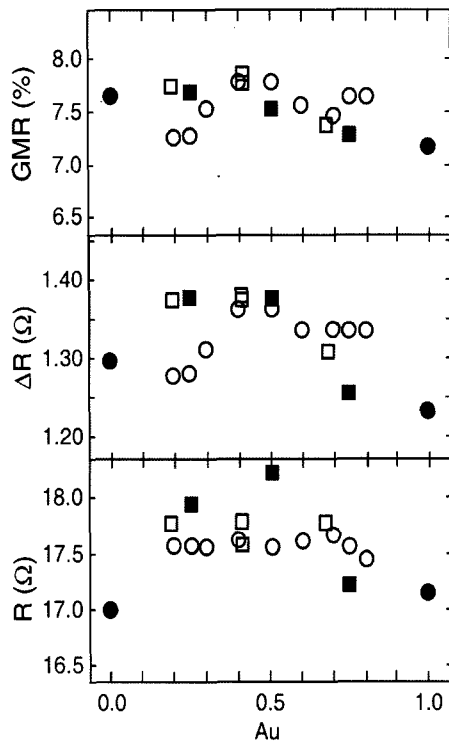
**Fig. 5.** Variations in the GMR,  $\Delta R$ ,  $R$ , and  $H_{\text{ex}}$  with a composition of Au in Cu. These spin valve were prepared by simple laminations of the Au and a Cu  $\delta$ -layer (alternations without stoichiometric consideration). Thin and thick circles correspond to the layer started with Cu (labeled 2a) in Fig. 2.2) and with Au (labeled 2b) in Fig. 2.2) respectively.



**Fig. 6.** Variations in the GMR,  $\Delta R$ ,  $R$ , and  $H_{ex}$  with two kinds of bi-layers of Au and Cu. These spin valves were prepared by bi-layers of Au and Cu. In this case the thickness of bi-layer was 10 Å unlike the other cases where the thicknesses were 12 Å.

### 3.5. Comparison of alloys and laminates started with an Au layer

In this section, we compared the spin-filter effect of all the types of back layers investigated. Since the back layer started with Au showed improved the GMR performance



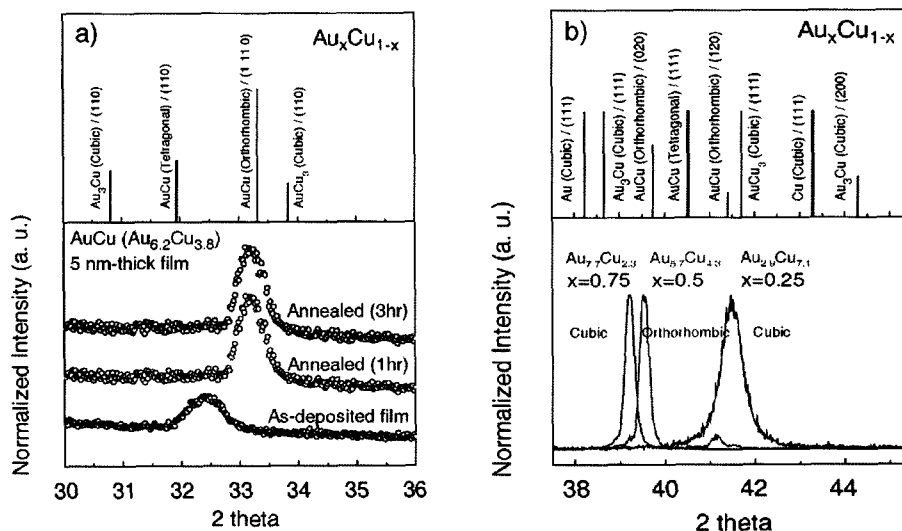
**Fig. 7.** Comparisons of Au-Cu alloys and laminations for the GMR,  $\Delta R$ , and  $R$ . Closed circles (●) indicates the spin valve with back layer of Cu and Au, open circles (○) represents Au-Cu alloy and closed squares (■) and open squares (□) indicates that the lamination started with Au. Closed squares (■) correspond to thick circles shown in Fig. 4. and open squares to thick circles shown in Fig. 5.

as compared to the back layer started with Cu, we focus our discussion on the back layer started with Au from here. When Au was mixed with Cu, the resistance abruptly increased, most likely due to the scattering by impurity, as summarized in Fig. 7. The GMR values were not increased much. They stayed at  $\sim 7.7\%$ . Nevertheless, we could see the increase in  $\Delta R$  by  $\sim 8\%$ . The increase is not from the enhancement of spin-dependent scattering, but from the increase in the resistance. Back layers of  $Au_{0.5}Cu_{0.5}$  alloy,  $Au_{0.4}Cu_{0.6}$ , repetitions of [Au 1/Cu 1] and [Au 2/Cu 2] helped improving the MR performance. However, overall, we could not achieve any dramatic improvement in MR performance out of Au-Cu alloys and laminates.

### 3.6. Structural Analyses of the Cu-Au alloy

X-ray diffractometry was carried out for structural analysis. The alloys were deposited with a thickness of 50 Å and 500 Å, respectively. The compositions of alloys were analyzed by ICP mass spectroscopy. The 50 Å-thick film's composition was  $Cu_{0.38}Au_{0.62}$ , which was little deviated from the stoichiometry. On the other hand, the 500 Å-thick films had compositions close to the stoichiometry:  $Cu_{0.71}Au_{0.29}$ ,  $Cu_{0.43}Au_{0.57}$ , and  $Cu_{0.23}Au_{0.77}$ . We believe that though we could not exactly match the stoichiometry we could extract useful information on the microstructures.

It turned out that the 500 Å-thick films close to the stoichiometry of  $Cu_{0.75}Au_{0.25}$  and  $Cu_{0.25}Au_{0.75}$  were cubic, as expected. In the mean time, the 50 Å-thick and 500 Å-thick films close to the stoichiometry of  $Cu_{0.5}Au_{0.5}$  turned out to be orthorhombic instead of face-centered tetragonal that we want to make a  $Cu_{0.5}Au_{0.5}$  inter-metallic compound with a structure of, as shown in Fig. 8. We believe



**Fig. 8.** XRD patterns for the film a) [Si/SiO<sub>2</sub>/50 Å-thick Au-Cu] and b) [Si/SiO<sub>2</sub>/500 Å-thick Au-Cu] acquired by a synchronous x-ray source with Cu K $\alpha$  source wavelength  $\lambda = 1.54$  Å. Vertical lines in (b) indicate the  $2\theta$  peak positions of inter-metallic Au-Cu compounds.

that the structure is the main reason we failed to decrease the resistance and to improve the MR performance of the spin valves when we tried to make the around 10 Å-thick Cu-Au back layer. In short, we could not obtain a correct phase of the inter-metallic compounds, face-centered tetragonal. We think that thermal annealing over 400°C may be required to get face-centered tetragonal. However, we could not employ this high-temperature process during the fabrication of the spin valves because the other performance including the GMR will be significantly degraded due to inter-diffusion between the layers. Further sophisticated fabrication should be developed to obtain a correct phase to observe the spin-filter effect in the back layers.

#### 4. Summary

When Au was mixed with Cu, the resistance abruptly increased most likely due to impurity scattering. The GMR values were not increased much when we use Cu-Au alloys and laminates for a back layer. However, we could see the increase in  $\Delta R$  by  $\sim 8\%$  for those with a  $\sim \text{Au}_{0.5}\text{Cu}_{0.5}$  composition. This increase is coming from an increase in the resistance and not from the enhancement of spin-dependent scattering. Structural investigations showed that orthorhombic  $\text{Au}_{0.5}\text{Cu}_{0.5}$  was formed in the back layer instead of face-centered tetragonal (FCT)  $\text{Au}_{0.5}\text{Cu}_{0.5}$  that we expected. We believe that thermal annealing over 400°C may be required to have face-centered tetragonal in a 10 Å-thick film. Face-centered tetragonal  $\text{Au}_{0.5}\text{Cu}_{0.5}$  seemed to be difficult to get when

the sputtered alloy was thin and was annealed below 280°C, which likely makes the alloy a non-equilibrium phase orthorhombic. Nevertheless, the spin valve with three repetitions of [Au 2/Cu 2 (Å)] improved the properties, in particular, in GMR and  $\Delta R$ . We strongly recommend three repetitions of [Au 2/Cu 2 (Å)] and a bilayer of [Au/Cu] for the back layer of the spin valve, to take advantage of the spin-filter effect.

#### Acknowledgements

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