Structure and Magnetic Properties of [Ag/Ni/Au] and [Au/Ni/Ag] Superlattices

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The interface structure and magnetic properties of the two kinds of superlattices, [Ag/Ni/Au] and [Au/Ni/Ag] consisting of the same kind of atomic species and the reverse deposition sequences have been investigated. In the [Ag/Ni/Au] superlattice, the in-plane lattice constant changed continuously in the Ni on Au, and the large lattice strain is contained in the Ni layers and mixing region exists at the interface of Ni on the Au. On the other hand, in the [Au/Ni/Ag] superlattice, the in-plane lattice constant changes discontinuously and little strain is contained. The [Ag/Ni/Au] superlattice containing mixing region shows smaller magnetization than the [Au/Ni/Ag] superlattice. The [Ag/Ni/Au] superlattice containing more elastic strain than the [Au/Ni/Ag] superlattice shows larger magnetic anisotropy.

Key words: perpendicular magnetic anisotropy, RHEED, thin film, superlattice, crystal growth

1. Introduction

Since the structure at/near the interface of superlattices influences magnetic property, it is important to investigate details of the structure. In the crystal growth, continuity of the crystal lattice is one of the interesting points. The reflection high energy electron diffraction (RHEED) allows us to investigate the surface structure during the crystal growth. The RHEED system installed in our molecular beam epitaxy (MBE) system enables us to observe, continuously, the change of the surface in-plane lattice constant on a real-time basis.

The ternary multilayers and superlattices consisting of three elements can clarify the effect of deposition sequence by comparing superlattices with the reverse deposition sequence. However, only a few works to observe in-situ crystal growth of ternary multilayers and superlattices by RHEED have been made previously¹⁾⁻⁴⁾.

In the present study we selected Ni as a ferromagnetic element, and Au and Ag having the very similar atomic radii and a larger atomic radius than that of Ni as non-magnetic elements, and fabricated two kinds of superlattices with reverse deposition sequences: [Ag/Ni/Au] and [Au/Ni/Ag]. Here, the description

[Ag/Ni/Au] means that the deposition sequence is Au the first, Ni the second and Ag the third. We have investigated crystal growth mainly by RHEED and magnetic properties of the superlattices by a vibration sample magnetometer (VSM) and a superconducting quantum interference device (SQUID) magnetometer. Moreover, we discuss the relation between the magnetic properties and the interface structure.

2. Experimental

All the films were prepared under a pressure in the 10^{-9} Pa range using a VG Semicon V80M MBE system. As substrates, [111] oriented Si single crystal wafers were used. In the [Ag/Ni/Au] superlattice, Ag(111) buffer layer was deposited on the Si (111) substrates first and subsequently Au seed layer was deposited on the Ag(111) buffer layer. In the [Au/Ni/Ag] superlattice, only Ag seed layer was deposited on the Si(111) substrate. In the superlattices, several specimens with different Ni thickness were prepared. The substrate temperature was kept at about 323 K. During epitaxial growth of the superlattices, RHEED pattern was recorded in video tapes. In the formation of the superlattices, the unit trilayer of [M1/Ni/M2] were repeatedly deposited on those seed layers. All the deposited films held close-packed atom arrangements. Analyzing the change of the streak distance in detail, continuous in-plane lattice change could be evaluated. The X-ray diffraction analysis was also performed after the deposition in order to confirm epitaxial growth. The magnetization measurement was made at room temperature by the VSM and at 10 K by the SQUID. Magnetic field was applied in two directions, perpendicular and parallel to the film plane.

3. Results and Discussion

3.1 Continuity of the in-plane lattice constants of [Ag/Ni/Au] and [Au/Ni/Ag] ternary superlattices

The changes of the in-plane lattice constants in the [Ag/Ni/Au] and [Au/Ni/Ag] superlattices are shown in Figs.1(a) and 1(b), respectively. In the [Ag/Ni/Au] super-

Fig.1 Change of relative in-plane lattice constant of (a) [Ag/Ni/Au] and (b) [Au/Ni/Ag] superlattices as a function of layer thickness.

lattice, the in-plane lattice constant decreased continuously in the stage of Ni deposition on Au, and the lattice constant of the subsequently grown Ag recovered discretely within a few monolayers. In the [Au/Ni/Ag] superlattice, Ni on Ag did not have the same lattice constant as that of Ag and the two streaks of Ag and Ni coexisted during a few monolayers in the RHEED patterns. Namely, the Ni lattice constant immediately decreased to the bulk Ni value. The lattice constant in the subsequently deposited Au on the Ni discretely returned back to the Au value. The behavior in the changes of the lattice constant was discontinuous. The RHEED pattern of Ni on Ag in the [Au/Ni/Ag] superlattice was spotty at the early stages of deposition, but it became streaky with increasing Ni layer thickness.

Let us discuss the difference in lattice continuity during depositions in terms of two points: (a) mixing and (b) strain. According to the Ni-Ag equilibrium phase diagram, it is a two phase separation system and there is no solubility at both low and high temperatures, so that the both elements do not mix each other even in the liquid state. The fact that there are two streaks in the [Au/Ni/Ag] superlattice indicates that two layers of Ni and Ag exist separately. The discontinuity of the lattice constant means that the [Au/Ni/Ag] superlattice contained little strain.

On the other hand, although the Ni-Au system is a two phase separation system in the equilibrium phase diagrams and there are no solubility at the low temperature, they

can be soluble when the temperature is high. Therefore, in the [Ag/Ni/Au] superlattice, there might exist mixing regions at the interface of Ni on the Au. Moreover, the large lattice strain might be introduced to the Ni layers because the amount of continuous change in the lattice constant is large. The amount of mixing layer and the amount of the strain in the Ni layer will be calculated in the section 3.2.1.

3.2 Magnetic properties

3.2.1 Magnetization

The results of saturation magnetization M_s measurements at room temperature are shown in Fig.2. It is found that the M_s values of the [Au/Ni/Ag] superlattice are larger than those of the [Ag/Ni/Au] superlattice for all t_{Ni} . Similar tendency was found in the results at a low temperature of 10K. This difference is interpreted by the asymmetry of the solubility limit. Namely, the solubility limit of Ni in Au in the phase diagram is larger than that of Au in Ni. Thus the mixing at the interface of Ni on Au is larger than that at the interface of Au on Ni.

When the change of M_s value is observed in detail, in the [Au/Ni/Ag] superlattice the value of M_s shows bulk value in the thicker regions beyond 2nm, while it decreases with decreasing Ni layer thickness t_{Ni} . The decrease of the M_s is caused by the degradation of the layer structure in the thinner Ni regions. In the [Ag/Ni/Au] superlattice the value of M_s increases gradually with increasing thickness of Ni layer t_{Ni} and is smaller than the Ni bulk value even at the thicker region of the 3.3nm thick.

In order to clarify the degree of the contribution of (a) mixing regions and (b) strained regions at/near the inter-

Fig.2 Saturation magnetization of [Ag/Ni/Au] and [Au/Ni/Ag] superlattices as a function of Ni layer thickness.

face, we calculated how much strain are contained in the Ni layer using the method proposed by Kingetsu⁹. Note that there is about 14% difference between the atomic radii of Ni and Au or Ag and the difference between those of Au and Ag is small. As a result of this calculation the mixing layer is determined to be about five monolayers by comparing the experimental M_s value with the bulk M_s value. By excluding the mixing layer, the amount of strain in the Ni layer was evaluated. The amount of the strain is determined to be 2.7% at the sixth Ni layer which corresponds to the first layer except the five mixing layers. Remember that there is little strain in the [Au/Ni/Ag] superlattice. Therefore, it is concluded that Ni in the [Ag/Ni/Au] superlattice contains more elastic strain than that in the [Au/Ni/Ag] superlattice.

3.2.2 Magnetic anisotropy

The effective magnetic anisotropy energy as a function of Ni layer thickness is shown in Fig.3. The magnetic anisotropy energy increases with decreasing Ni layer thickness for the both superlattices. However, even when the thickness of the Ni layer is 1nm, the both superlattices showed in-plane magnetization, i.e. they did not show perpendicular magnetic anisotropy. The amount of the effective magnetic anisotropy energy of two kinds of the superlattices is different, and the effective magnetic anisotropy energy in the [Ag/Ni/Au] superlattice is larger than that in the [Au/Ni/Ag].

Let us consider the difference in magnetic anisotropy of two kinds of the superlattices. We can understand by the following calculation. The effective magnetic anisotropy of the metallic superlattice is expressed phenomenologically by the following equation,

$$K_{\text{eff}} \cdot t = 2K_N + K_E + (K_C + K_D) \cdot t, \quad (1)$$

Fig.3 $K_{\text{eff}} \cdot t$ values of [Ag/Ni/Au] and [Au/Ni/Ag] superlattices as a function of Ni layer thickness t_{Ni} .

where K_N is the Neel-type surface anisotropy which is caused by degradation of symmetry at the surface, K_E is the magnetoelastic anisotropy caused by the strain, K_C is the magnetocrystalline anisotropy, K_D is the demagnetization energy, and t is the thickness of the ferromagnetic layer. Here, for K_C we give the bulk value and for K_E and K_D , we use the values calculated from the present experimental data, and we assume that the K_N is the same. In the calculation of K_E , the magnetstriction constant λ , the Young's modulus Y and the amount of strain ϵ are needed. For the former two, λ and Y , a half bulk value and the bulk value, respectively, are utilized and the ϵ is evaluated from the results of the present RHEED observation. The calculated effective magnetic anisotropy are also shown in Fig.3 by solid lines. The experimental and calculated results agree well. Therefore, it is concluded that the difference in anisotropy is due to the different strained structure caused by stacking sequence.

4. Conclusion

Two kinds of superlattices, [Ag/Ni/Au] and [Au/Ni/Ag] consisting of the same kind of atomic species with the reverse deposition sequences were fabricated, and continuity of the in-plane lattice constants and magnetic properties like magnetization and magnetic anisotropy have been investigated.

- (1) In the [Ag/Ni/Au] superlattice, the in-plane lattice constant changes continuously Ni on Au, while in the [Au/Ni/Ag] superlattice, that is *dis*continuous.
- (2) Therefore, in the [Ag/Ni/Au] superlattice the large lattice strain is introduced to the Ni layers, while in the [Au/Ni/Ag] superlattice little strain is contained.
- (3) The [Ag/Ni/Au] superlattice containing mixing regions shows smaller magnetization.
- (4) The [Ag/Ni/Au] superlattice containing more elastic strain than the [Au/Ni/Ag] superlattice shows larger magnetic anisotropy.

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