Epitaxial growth of binary and ternary metallic strained superlattices and their magnetic properties

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Abstract

Since the structure at/near the interface of superlattices influences physical properties such as magnetic property, it is important to investigate details of the structure. The interface structure is characterized by the factors like atomic species, strain, mixing and roughness. The reflection high-energy electron diffraction (RHEED) system installed in our molecular-beam epitaxy (MBE) system enables us to observe, continuously, the change of the surface in-plane lattice constant, which is affected by atomic species, strain and/or mixing, on a real-time basis. Ternary superlattices consisting of three elements can clarify the effect of stacking sequence by comparison between the two types of superlattices with the reverse deposition sequences, since the effect caused by the combination of the same atomic species is cancelled out and the effect caused by the different stacking sequences remains. In the present paper, we review growth behaviors of binary and ternary metallic strained superlattices, especially magnetic ones, investigated mainly by our group, and summarize the discussion on their magnetic properties, mainly on the magnetic anisotropy, in terms of their structural characteristics. First, we introduce our RHEED system that works efficiently under a magnetic field arising from evaporation sources for low vapor-pressure materials. Then, MBE-grown binary strained superlattices, Co/Au, Co/Pt and Cu/Au, are discussed, with comparing to incoherent superlattices of Co/Au and Cu/Au having nearly the same lattice mismatch of constituents. Next, we review ternary strained superlattices with immiscible constituents with reverse deposition order, Au/Co/Ag and Ag/Co/Au superlattices, and Au/Co/Cu and Cu/Co/Au superlattices, in relation to the growth behaviors of binary superlattices. Finally, ternary strained superlattices containing both miscible and immiscible constituents, Pt/Co/Ag and Ag/Co/Pt superlattices, and Au/Ni/Ag and Ag/Ni/Au superlattices, are reviewed. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

In the last decade, epitaxial metallic superlattices with controlled orientations have been attracting much attention both from a fundamental viewpoint and for application purposes. For instance, it has been reported that several magnetic superlattices such as Co/Au [1], Co/Pt [2] and Co/Pd [3,4] exhibit perpendicular magnetic anisotropy and that those such as Fe/Cr [5] and Co/Ag [6] have the antiferromagnetic coupling between magnetic layers and show the giant magnetoresistance. These superlattices, in many cases, tend to have more pronounced characteristics, such as larger perpendicular magnetic anisotropy [2,3], than the polycrystalline multilayers with the same constituents. The dependence of these magnetic and magnetotransport properties on the growth orientation of the superlattices is also important for investigating the origins of the properties [2,3,7–9].

One difficulty in clarifying the origin from experimental data arises from the presence of the properties very sensitive to chemical and geometrical qualities of the interfaces, which are strongly affected by the preparation conditions. In particular, a procedure of forming an interface features the properties of the interface and hence the interfaces are different even when the final macroscopic structure is the same. For example, the interface formed by depositing material B onto material A can be different from the interface formed by depositing A onto B. Thus, it is important to study details of the interface structures by an in situ experimental method. Ternary superlattices consisting of three elements with reverse deposition sequences, A/B/C and C/B/A, have the interfaces with the same atomic species in adjacent layers and the reverse stacking sequences, and thus by comparison between these two types of the superlattices, the effect caused by the combination of the atomic species is...
canceled out, in principle. Therefore, the effects caused by strain and mixing arising from the difference in stacking sequence can be clarified, if the roughness is the same. The reflection high-energy electron diffraction (RHEED) can reveal the surface structure during the crystal growth. Especially, 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, even under a magnetic field arising from vaporization sources of low vapor-pressure materials.

In the present paper, we review growth behaviors of binary and ternary metallic strained superlattices, especially magnetic ones, investigated mainly by our group using RHEED, and summarize the discussion on their magnetic properties, especially on the magnetic anisotropy, in terms of their structural characteristics. In Section 2, we introduce our RHEED system that works efficiently under a magnetic field. In Section 3, binary strained superlattices, Co/Au, Co/Pt and Cu/Au are discussed, comparing to incoherent superlattices of Co/Ag and Cu/Ag having nearly the same lattice mismatch of constituents. In Section 4, we review ternary strained superlattices with immiscible constituents with reverse deposition order, Au/Co/Ag and Ag/Co/Au superlattices, and Au/Co/Cu and Cu/Co/Au superlattices, in relation to the growth behaviors of binary superlattices presented in Section 3. In Section 5, ternary strained superlattices containing both miscible and immiscible constituents, Pt/Co/Ag and Ag/Co/Pt superlattices, and Au/Ni/Ag and Ag/Ni/Au superlattices, are reviewed. Finally, we give concluding remarks in Section 6.

2. Continuous in situ RHEED observations under a magnetic field arising from E-guns

The spacing of streaks in RHEED patterns is inversely proportional to the in-plane lattice constant of a deposited film surface. Thereby, the relative in-plane lattice constant can be estimated. In the MBE growth of compound semiconductor superlattices such as GaAs/AlGaAs, in situ RHEED observations are routinely made. In these cases, vapor pressures of materials are relatively high, and hence the evaporation temperatures are not so high. Therefore, effusion cells (K-cells) are usually used as evaporation sources. In this circumstance, neither magnetic field nor electric field that interferes with in situ RHEED observations is produced by the evaporation sources. Furthermore, the radiation of light from the red heated evaporation sources, which also influences RHEED observation with illuminating the RHEED screen, is moderate since the temperature is relatively low. In the MBE growth of metallic superlattices, however, some source materials have low vapor pressures and thus high temperatures are often necessary for evaporation. For evaporation of such materials, electron-beam evaporators (E-guns) are usually used. In the E-guns, electron-beams are focused by a strong magnetic field or electric field. This field, especially a magnetic one, strongly interferes with the RHEED observation. The trajectories of the primary electron beams from a RHEED gun and diffracted or reflected electron beams at the surface of a growing film are changed by the field to form distorted RHEED patterns. This prevents us from accurate determination of surface crystal structures and lattice constants. Moreover, when the E-gun uses beam scanning for uniform evaporation of the source material, the RHEED images are also ‘scanned’, namely, the images oscillate with time. In this case, it also becomes very difficult to measure the time-resolved intensity change of a particular spot such as specular spot. Furthermore, upon operation of an E-gun, strong light emission occurs because of the high temperature radiation, and secondary electron emission occurs because of the bombardment of the primary electron beams of the E-gun. The strong light and the secondary electrons illuminate the RHEED screen to be whitened and this blurs the RHEED patterns. This also prevents us from clear RHEED observations.

In this situation, the in situ RHEED observation of metallic superlattices whose constituents have low vapor pressures is difficult when a conventional RHEED system is used. We have developed a special RHEED system equipped with magnetic, light and electron shield. The system has been installed in a MBE system, VG Semicon special V80M, which is a prototype of V80 S that was commercialized as a Si-MBE system on later date, possessing two magnetically focused and deflected Aircotemescal E-guns with 40 cc hearths, three VG Semicon K-cells and one EPI high-temperature K-cell. Two types of RHEED guns are being used interchangeably. One is of a magnetically focused 30 kV type made by Elko Engineering and another is of an electrostatically focused 30 kV type made by VG Microtech. The primary-beam path from the E-gun is shielded by a mu-metal pipe covered by Ta sheets. The diffracted-/reflected-beam paths are shielded by large-diameter mu-metal pipes with a stepped structure covering a RHEED screen. The outer side of this shield is also covered by Ta sheets for thermal protection. The structure acts as a light and electron shield from light and secondary electron emission from E-guns as well as a magnetic shield, and also protects the RHEED screen from material deposition onto its surface. Above the RHEED gun and RHEED screen, magnets are placed for cancelling the magnetic field from E-guns at the position of the RHEED screen. In between E-gun hearths and E-gun Mo shutters, Ta radiation shields are installed. These shields reduce light and secondary electron irradiation onto the RHEED screen, thermal loads to cryoshrouds of the MBE machine and deposits of materials onto the shrouds. These structures are schematically shown in Fig. 1. This RHEED system allows us continuous in situ observation of long deposition sequences throughout the whole superlattice and buffer layer growth, and surface cleaning and smoothing process of the substrate.

In the next sections, we review epitaxial growth of
metallic superlattices and their magnetic properties mainly studied by our group. In the growth experiments, Co and Pt were evaporated from E-guns, Ag, Au and Cu were done from K-cells and Ni from a high-temperature K-cell. Si(111) wafers were mainly used as substrates [9]. The substrates were heated to a high temperature to obtain clean and smooth surfaces. Prior to superlattice growth, buffer layers of Cu or Ag were grown on the substrates. The growth temperature of the superlattices was 300–500 K depending on material, and the growth rate was around 0.02 nm/s, which was calibrated by a Bayard–Alpert ion gage flux monitor or a quartz film thickness monitor whose sensor heads were positioned at the center of the substrates. All magnetic measurements given in the present paper were performed at room temperature.

3. Binary strained superlattices

3.1. Incoherent superlattices [10–12]

Superlattices with immiscible constituents often have incoherent interfaces. For examples of such superlattices, there are Co/Ag and Cu/Ag binary superlattices. The lattice constants for the closed-packed planes of Co and Cu are about 13% smaller than that of Ag. In the Co/Ag and Cu/Ag systems, the equilibrium phase diagrams are of the two-phase separation type. Especially, in the case of Co–Ag system there is no miscibility even at the liquid state. It is known that the immiscible elements have weak binding to each other. Therefore, the bindings of the Co–Ag and Cu–Ag atoms are weak, and thus the binding breaks and misfit dislocations are easily introduced at the interface. In the next, we show growth behavior of these superlattices.

In the case of Co/Ag superlattices, the crystal structure of Co on Ag, which is determined by RHEED observations and X-ray diffraction analyses, is a hexagonal closed-packed (hcp) structure. The Co/Ag superlattices keep the orientational relationship that their closed-packed planes are parallel. The changes of the in-plane lattice constants in a Co/Ag superlattice are shown in Fig. 2. The lattice constant of Co on Ag immediately decreases to the bulk Co value and does not have the same lattice constant as that of the underlayer Ag. Then the lattice constant in the subsequently deposited Ag on the Co returns to the original Ag value. Thus, the behavior in the changes of the lattice constant is discontinuous.

In the case of Cu/Ag superlattices, both Cu and Ag keep face-centered cubic (fcc) structures during deposition. The Cu/Ag superlattices keep the orientational relationships that the closed-packed planes are parallel. The changes of the in-plane lattice constants in a Cu/Ag superlattice are shown in Fig. 3. The fundamental behaviors are similar to those in the Co/Ag superlattice described above. However, different from the Co/Ag superlattice, the value of the immediately decreases and the Cu lattice constant is not the Cu bulk value, but is a relatively larger value than the bulk value, and then it reaches gradually to the bulk value. The lattice constant of subsequently deposited Ag on Cu discretely returns to the Ag value. The behavior in the changes of the lattice constant is discontinuous.

3.2. Strained superlattices with immiscible constituents [11–15]

Some superlattices with immiscible constituents have semicoherent interfaces and strained layers. For examples of such superlattices, there are Co/Au binary superlattices. In this case, the structure at/near the interfaces of the superlattices can be discussed in terms of strain. The lattice constant for the closed-packed plane of Co is about 13% smaller than that of Au. Thus, the amounts of lattice misfit are the same between Co/Au and Co/Ag discussed in
Section 3.1. However, the behaviors in crystal growth are different as described later. In the Co/Au system, the equilibrium phase diagram is of the two-phase separation type, as in the case of the Co–Ag system appeared in Section 3.1, but the solubility limit expands at high temperatures as compared with the Co–Ag system. This means that the binding between Co and Au is stronger than that of Co–Ag. Therefore, the strains are contained in the Co layers and misfit dislocations introduced at each boundary of adjacent Co atomic layers in the Co layers. This situation is schematically shown in Fig. 13 given later. Detailed discussion will be given in Section 4.1.1.

Examples of epitaxial growth experiments are presented below. Fig. 4 shows RHEED patterns of Au and Co layers in the epitaxial growth of a Co/Au superlattice. Streaks indicate that the layers grow epitaxially and that interfaces between the layers are flat. Fig. 5 shows an XRD profile of a Co/Au superlattice. Satellite peaks around a main peak in the middle-angle range indicate that a well-ordered periodic-structure is formed. In a low-angle range XRD profile, superlattice peaks are observed. The crystal structure of Co on Au, which was determined by electron diffraction analyses during transmission electron microscopy observations, is an hcp structure. The Co/Au superlattices keep an orientational relationship that their closed-packed planes are parallel. This situation is similar to that of the Co/Ag superlattices. However, the structural changes during crystal growth are different between the two. The changes of the in-plane lattice constants in the Co/Au superlattice are shown in Fig. 6. The lattice constant decreases continuously in the stage of Co deposition on Au. Then the lattice constant of subsequently grown Au recovered rapidly within

Fig. 2. Change in the relative in-plane lattice constants of a Co/Ag superlattices as a function of layer thickness, determined from the streak-spacings in RHEED patterns [12].

Fig. 3. Change in the relative in-plane lattice constants of a Cu/Ag superlattice as a function of layer thickness [12].
three monolayers (ML). This is in contrast to the Co/Ag superlattices, where the behavior in changes of the lattice constant is discontinuous.

### 3.3. Strained superlattices with miscible constituents [12,16,17]

Many superlattices with miscible constituents have coherent interfaces and strained layers owing to mixing at the interfaces. For examples of such superlattices, there are Cu/Au and Co/Pt binary superlattices. In the case of these superlattices, the structures at/near the interface can be discussed in terms of mixing. In the Cu/Au superlattices, for instance, the lattice constant for the closed-packed plane of Cu is again about 13% smaller than those of Au. The amounts of lattice misfit are almost the same between Cu/Au and Cu/Ag. However, the behaviors in crystal growth are different. In contrast to the Cu/Ag system, where the equilibrium phase diagram is of the two-phase separation type, in the Cu/Au system, it is of the completely soluble type, so that mixing occurs at the interface region and the gradual change of the lattice constant is due to compositional change. Examples of growth behaviors in Cu/Au and Co/Pt superlattices are given below.

In the Cu/Au superlattices, both Cu and Au keep fcc structures during deposition. The Cu/Au superlattices keep an orientational relationship that the closed-packed planes are parallel. The changes of the in-plane lattice constants in the Cu/Au superlattices are shown in Fig. 7. The lattice constant of Cu on Au decreases gradually, and the lattice constant of subsequently deposited Au recovers within 3 ML. The behavior is continuous. This is in contrast to the case of Cu/Ag superlattices where the behavior in changes of the lattice constant is discontinuous.

In the combination of Co and Pt, Co is about 10% smaller than Pt in atomic radius. The Co/Pt superlattices also keep the orientational relationship that the closed-packed planes are parallel. Fig. 8 shows the change of the relative lattice constant of the Pt and the Co in the first bilayer and the 22nd bilayer as a function of thickness, obtained by in situ RHEED observations. These results clearly demonstrate that through the 22 periods the lattice constant changes continuously and those of Pt and Co match at the interface. The equilibrium phase diagram of the Co–Pt system is of the completely soluble type, so that mixing occurs at the interface region and thus the gradual change of the lattice constant is due to compositional change.

In addition, in another miscible combination of Pt and Cu, a similar coherent interface and a strained layer owing to mixing at the interface can be found on growth of (111) Pt on a (111) Cu layer. Fig. 9 shows the change of the relative lattice constant as a function of thickness of the Pt film up to

![Fig. 4. (110) azimuthal RHEED patterns of 10 ML-thick Au layers and 6 ML-thick Co layers in Co/Au superlattices. (a) and (c) Au layer. (b) and (d) Co layer. (a) and (b) Growth temperature 273 K. (c) and (d) Growth temperature 473 K [15].](image)

![Fig. 5. Middle-angle-range XRD profile of a Co(6 ML)/Au(10 ML) superlattice grown at 373 K [15].](image)
14 ML, which corresponds to about 3.2 nm. This result also demonstrates that the lattice constant changes gradually.

3.4. Magnetic properties [13–15,18,19]

In some magnetic superlattices, it is known that the large uniaxial magnetic anisotropy with the axis perpendicular to the film plane is observed when the Co layer thickness is smaller than several monolayers. The Co/Au and Co/Pt superlattices exhibit the perpendicular magnetic anisotropy, while the Co/Ag superlattices indicate the in-plane magnetic anisotropy at least at room temperature: Fig. 10 shows magnetization curves for applied fields both perpendicular and parallel to the film plane at 300 K of a Co/Ag superlattice. The curves do not exhibit hysteresis loops and the anisotropy is found to be in-plane. Fig. 11 shows magnetization curves for a Co/Pt superlattice. It is clearly found that the superlattice exhibits the perpendicular magnetic anisotropy and that the remanent magnetization is near 100% of the saturation magnetization. The perpendicular coercivity of the superlattice is as large as 3.1 kOe. The magnetization curve of a Co/Au superlattice is given in Fig. 14 in Section 4.1.2.

4. Ternary strained superlattices with immiscible constituents

4.1. Superlattices with largely lattice-mismatched constituents [11,13–15,20]

4.1.1. Growth behavior

In this section, Ag/Co/Au and Au/Co/Ag ternary
superlattices are presented as examples of strained superlattices with largely lattice-mismatched, immiscible constituents. The relative in-plane lattice constant obtained from RHEED observations is shown in Fig. 12 as a function of layer thickness. Here, Fig. 12(a) and (b) are for a Ag/Co/Au and a Au/Co/Ag superlattice, respectively. It is found that in the Ag/Co/Au superlattice the lattice constant of the Co layer decreases gradually towards its own value in the bulk with increasing Co layer thickness. On the other hand, in the Au/Co/Ag superlattice, the Co layer grows on the Ag layer with its own lattice constants in the bulk and the Co/Ag interface is completely incoherent. These growth modes are similar to those in the Co/Au and the Co/Ag superlattices, respectively, which were described in Section 3. It is emphasized that the Au layers on the Co layers grow incoherently in the Au/Co/Ag superlattices, as in the case of Ag on Co in the Co/Ag superlattices, and contrary to the semicoherent growth of Au on Co layers in the Au/Co superlattices. This result means that Au layers can grow semicoherently only on the dilated Co layers, which are caused by depositing Co on the Au layers. Therefore, Co–Au atomic bonding is not to strain the Au layers deposited on the unstrained Co layers, for this type of Au/Co interfaces. This suggests that the Au overlayers do not strain the Co underlayers significantly even in the case of Co/Au superlattices. From the above discussion, it is deduced that only the near-interface regions of the Co layers on the Au underlayers are strained, in the Co/Au superlattices.

From these results, strain distributions in the Co layers are deduced and their strain models are shown in Fig. 13 schematically. The distributions are likely to be featured during growth, because deposited atoms can move freely to adjust the distribution of dislocations on the surface as the growth front. The strain of Co on Au is relieved gradually as the thickness increases with incorporating dislocations, while each Co layer is expected to not be strained at the other side of interface in the Co/Au and Ag/Co/Au superlattices. On the other hand, in the Co/Ag and Au/Co/Ag superlattices, Co layers are basically strain-free and misfit dislocations are localized at the both interfaces.

### 4.1.2. Magnetic properties

Fig. 14 shows magnetization curves of Co/Au and Ag/Co/Au superlattices with Co layer thickness about 6 ML, measured with applied fields both parallel and perpendicular to the film plane. It is clear that the easy axis of magnetization

![Fig. 10. The magnetization curves of a Co/Ag superlattice measured at 300 K [11]. $K_{eff} = -1.2 \times 10^7 J/m^3$.](image-url)

![Fig. 11. Normalized magnetization ($M/M_s$) versus applied field ($H$) curves a Co(0.41 nm)/Pt(7 ML) superlattice measured at 300 K. Magnetic fields were applied up to ±5 kOe, although only ±5 kOe areas are shown in the figure [18].](image-url)
lies on the normal of the film plane for both the superlattices. The curves for the perpendicular fields are nearly the same in the two superlattices, while the curve for the parallel fields in the Ag/Co/Au superlattice reaches the saturation magnetization faster than that in the Co/Au superlattice, indicating that the perpendicular magnetic anisotropy in the Ag/Co/Au superlattice is weaker.

The effective anisotropy $K_{\text{eff}}$ was determined from the area between perpendicular and parallel magnetization curves per unit Co volume. The $K_{\text{eff}}$ is taken to be positive when the magnetization is preferably oriented perpendicular to the film plane. Fig. 15 shows $K_{\text{eff}}$ values of the four types of superlattices, Co/Au (○), Ag/Co/Au (●), Au/Co/Au (□) and Co/Ag (●), as functions of $t$, the thickness of Co layers. It is clearly observed that the $t$–$K_{\text{eff}}$ relationships for the Ag/Co/Au and the Au/Co/Ag superlattices are distinctly different, despite the fact that each Co layer in both types of the superlattices has an interface to Au and another interface to Ag. Especially, as also shown in Fig. 14, the Ag/Co/Au superlattices exhibit perpendicular anisotropy when Co layers are several monolayers in thickness and tend to saturate for the smaller thickness, as in the case of the Co/Au superlattices. In contrast, the shape of the plot for the Au/Co/Ag superlattices is linear and these superlattices show in-plane magnetic anisotropy as in the case of the Co/Ag superlattices, although the $K_{\text{eff}}$ values for the Au/Co/Ag superlattices are somewhat larger. The $K_{\text{eff}}$ values for the Co/Au superlattices are substantially in good agreement with the published data for Co/Au superlattices [1,21], and the linear dependence of $K_{\text{eff}}$ on $t$ in the Co/Ag superlattices also agrees with the results in previous work [21]. The difference between the $t$–$K_{\text{eff}}$ $t$ relationship for the Ag/Au and that for the Au/Co/Ag superlattices arises from the sequence of deposition, i.e. only the Co interface regions of the Co layers on the Au underlayers are strained and contribute to the magnetoelastic interface anisotropy.

In the below, we discuss $K_{\text{eff}}$ based on the two types of magnetic interface anisotropies: the magnetoelastic interface anisotropy [22] and the magneto-crystalline interface anisotropy [23,24] in addition to magnetic bulk anisotropies. The magnetoelastic interface anisotropy originates from the in-plane positive strains of the Co layers, while the magneto-crystalline interface anisotropy arises from degenerate states close to the Fermi energy due to the orbital hybridization of electrons at interfaces. The latter
anisotropy was often called Néel type magnetic surface anisotropy [25] arising from reduced symmetry at the surfaces and interfaces, in earlier phenomenological approaches. \( K_{\text{eff}} \) is expected to originate from volume dependent anisotropy \( K_v \) per unit Co volume and two-dimensional magnetic anisotropy \( K_i \) at interfaces per unit area, and can be expressed as

\[
K_{\text{eff}} = 2K_i + K_v t.
\]

Here, the factor of 2 arises from the two interfaces of each Co layer. If \( K_v \) and \( K_i \) are constants, this equation leads to a linear plot with an intercept of \( 2K_i \) at \( t = 0 \).

In the case of the present four types of superlattices, the inclinations of the linear portions of the four plots in Fig. 15 are the same, \( K_v = -0.63 \text{ MJ/m}^2 \) and hence, the equation is rewritten as

\[
K_{\text{eff}} = [K_{\text{IC(}Au\text{)}} + K_{\text{IE(}Au\text{)}}] + [K_{\text{IC(}Ag\text{)}} + K_{\text{IE(}Ag\text{)}}] + K_v t,
\]

\[
K_{\text{eff}} = [K_{\text{IC(}Ag\text{)}} + K_{\text{IE(}Ag\text{)}}] + K_{\text{IC(}Ag\text{)}} + K_v t,
\]

\[
K_{\text{eff}} = 2K_{\text{IC(}Ag\text{)}} + K_v t,
\]

\[
K_{\text{eff}} = 2K_{\text{IC(}Ag\text{)}} + K_v t.
\]

for Co/Au, Ag/Co/Au, Au/Co/Ag and Co/Ag, respectively. Here the subscripts IC and IE denote the magnetocrystalline interface anisotropy and the magnetoelastic interface anisotropy, respectively, and subscripts (Au) and (Ag) correspond to Co interfaces to Au and Ag, respectively. \( K_{\text{IE(}Au\text{)}} \) denotes the magnetoelastic interface anisotropy arising from the Au/Co interface regions of the Co layers under Au overlayers. It is noted that the magnetoelastic anisotropy in the linear regions are not included in \( K_v \) but included in \( K_i \), unlike the earlier treatments [21]. From Eqs. (2)–(5), the values of \( K_{\text{IE(}Ag\text{)}} \) and \( K_{\text{IC(}Ag\text{)}} \) are estimated to be 0.57, 0.27 and 0.07 mJ/m², respectively. \( K_{\text{IE(}Au\text{)}} \) is approximately zero and this originates from the spacings of the linear plots between Eqs. (2) and (3) and between Eqs. (4) and (5) are nearly the same. This result is consistent with the RHEED results that only interface regions of the Co layers on the Au underlayers are strained, causing the magnetoelastic interface anisotropy. From the above discussion, we conclude that the perpendicular magnetic anisotropy in the Co/Au superlattices consists of contributions from both the magnetoelastic interface and the magnetocrystalline interface anisotropies. The apparent \( K_i \) value is \( [K_{\text{IE(}Ag\text{)}} + 2K_{\text{IC(}Ag\text{)}}]/2 = 0.56 \text{ mJ/m}^2 \) and agrees with the 0.58 mJ/m² reported previously [21].

Next, we discuss the non-linear portions of the \( t-K_{\text{eff}} \) plots. In early reports [21], the deviation from the linear plot at small \( t \) in the Co/Au superlattices was attributed to sample growth problems such as island formation due to poor wetting. However, no appreciable deviation in the plot of the Co/Ag superlattices for similar \( t \) values indicates that this speculation is not appropriate, because similar or severer problems are expected in the Co/Ag superlattices. We will show that this is a more intrinsic property in the Co/ Au system. In the non-linear portions, \( K_v \) and \( K_i \) values are not constant, and \( K_{\text{eff}} \) is expressed as

\[
K_{\text{eff}} = 2K_i + (K_v + K_{\text{IE(}Ag\text{)}}) t + \int_0^t d\tau K_{\text{IE(}Ag\text{)}}(\tau)
\]

(6)

instead of Eq. (1). Here, \( K_v \) is the magnetocrystalline bulk anisotropy of Co, \( K_{\text{IE(}Ag\text{)}} \) the demagnetization energy and \( K_{\text{IE(}Ag\text{)}}(\tau) \) the magnetoelastic anisotropy at depth \( \tau \) owing to the semi-coherency strain. \( K_{\text{IE(}Ag\text{)}}(\tau) \) may be expressed [20] as

\[
K_{\text{IE(}Ag\text{)}}(\tau) = -3\lambda C\varepsilon(\tau)/2,
\]

(7)

where \( \lambda \) is a magnetostriction constant for a strain in a basal plane, \( C(\tau) \) a strain at position \( \tau \) and \( C \) an elasticity constant of Co.

From the RHEED results described above, \( \varepsilon(\tau) \) is deduced as shown below. Fig. 16 shows a strain model for a Co layer. Symbols \( \blacksquare \) and \( \bigcirc \) indicate apparent Co layer strains directly obtained from the RHEED streak spacings. It is noted that apparent in-plane lattice constants obtained from RHEED data are convolution of lattice constants of a few atomic layers near to the surface, since RHEED detects not only the topmost atomic layer but also detects a few near-surface atomic layers because of the electron-beam penetration into the sample by a depth of a few monolayers. The apparent lattice strain \( S(t) \) in the Co layer of thickness \( t \) is given by

\[
S(t) = \int_{-\infty}^t q(\tau - t)s(\tau) d\tau \int_{-\infty}^t q(\tau - t) d\tau,
\]

(8)

where \( s(t) \) is a real lattice strain of the topmost surface layer of Co with a thickness \( t \), and \( q(\tau - t) \) is a decay function due to inelastic scattering of the electron beam at depth \( \tau - t \) from the surface of the Co layer with a thickness \( t \). From the
RHEED results, we assume an exponential decay of the apparent strains

$$S(t) = A \exp(-\alpha t),$$  \hspace{1cm} (9)

where \(A\) and \(\alpha\) are positive constants. For \(q(\tau - t)\), a mean free path model of the electron beam is assumed

$$q(\tau - t) = \exp[(\tau - t)/l],$$  \hspace{1cm} (10)

where \(l\) is an effective mean free path of the electron beam. A solution of the integral equation, Eq. (8) with conditions of Eqs. (9) and (10) is given by

$$s(t) = A(1 - \alpha t)\exp(-\alpha t).$$  \hspace{1cm} (11)

From the RHEED data, an apparent strain curve \(S(t)\) indicated by a thick solid line in Fig. 16 is drawn as the best fit with \(A = 0.09\) and \(\alpha = 1.3\) /nm. The large difference between the curve and the measured points in a range of \(t \leq 0.2\) nm is attributed to the influence of convolution of the underlying Au lattice in RHEED. Next, we estimate \(s(t)\). Here, \(2l = l_0\sin \theta\), where \(l_0\) is a mean free path of the electron beam and the factor of 2 arises from the paths of incidence and reflection. \(\theta\) is an incident angle of the primary electron beam for RHEED. \(l_0\) of electrons with an energy of 20–25 keV in a metal is 20–30 nm, \(\theta \approx 2^\circ\) in the RHEED experiments, and thus \(l = 0.35–0.5\) nm. The estimated real strain curve \(s(t)\) as a function of \(t\) is shown by a dotted line. This curve gives an envelope line of in-plane strain given by the bar graphs in the figure. Total magnetoelastic anisotropy energy per unit Co volume originates from the summation of the in-plane strain energy of each Co atomic layer in whole the Co layer, assuming that the magnetostriction and elasticity constants are independent of strain. Here, to replace the summation with integral, the thin solid curve in the figure is taken: the curve corresponds to the \(\varepsilon(\tau)\) and given by

$$\varepsilon(\tau) = \varepsilon_0 \exp[-\alpha(\tau - d/2)],$$  \hspace{1cm} (12)

where \(\varepsilon_0\) is a strain of the Co atomic layer adjacent to the underlying Au layer, \(d\) is a thickness of a monolayer of Co, and the bulk value of Co(0001)hcp layer (ca. 0.2 nm) is adopted. Then, \(\varepsilon_0 = 3.8–2.4\%\) is obtained. The values of Co layer strains obtained here are not contradictory to earlier reports on high resolution electron microscopy observations on polycrystalline Co/Au multilayers [26] and on epitaxial Au/Co/Au sandwiches [27], and in-plane X-ray scattering measurements of epitaxial Co/Au superlattices [28], where average strains of Co layers were estimated to be around 2–3%.

To calculate the \(K_{\text{eff}}\) values, the term of \(2K_{\text{IC}}\) in Eq. (6) is rewritten as, \(2K_{(\text{IC})}\), \(K_{(\text{IC})} + K_{(\text{IC})}\), \(K_{(\text{IC})} + K_{(\text{IC})}\), \(2K_{(\text{IC})}\) for Co/Au, Ag/Co/Au, Au/Co/Ag and Co/Ag superlattices, respectively, in accordance with Eqs. (2)–(5). Absolute values theoretically obtained of the magneto-crystalline anisotropy energies do not necessarily agree with the experimental values well at present [29]. Hence, experimental values obtained above are adopted, here: \(K_{(\text{IC})}\) and \(K_{(\text{IC})}\) are 0.27 and 0.07 mJ/m², respectively. For \(K_{\text{IC}}\), value of bulk hcp Co (0.56 MJ/m³) is used and \(K_{\text{D}}\) is calculated from \(K_{\text{D}} = -\mu_0M_s^2/2\), where \(M_s\) is the saturation magnetization of Co and \(\mu_0\) is the magnetic permeability of vacuum. The \(M_s\) values for Au/Co/Cr superlattices are somewhat smaller than the bulk value, and we adopt our experimental value (averaged): \(M_s(\text{Co/Au})/M_s(\text{bulk}) = 0.97\). The value of \(K_c\) (\(K_c = K_{\text{IC}}\)) is thus \(-0.63\) MJ/m³. For calculation of the magnetoelastic interface anisotropy energy by Eqs. (7) and (12), we use \(-5 \times 10^{-5} \text{[J]}\) as \(\lambda\) and Young’s modulus \(2.1 \times 10^3 \text{[N/m²]}\) as \(C\) for bulk hcp Co. The calculated \(K_{\text{eff}}\) curves of the Co/Au, and Ag/Co/Au superlattices as functions of \(t\) are shown as upper and lower dotted lines, respectively, in Fig. 15. It is seen that considerable shape difference of the curves from the experimental plots around small \(t\) in addition to the downward shift.

Next, instead of using the bulk parameters of \(\lambda\) and \(C\), we adopt an experimental value for the magnetoelastic interface anisotropy energy, \(K_{(\text{Bi})}\), at large \(t\), 0.57 mJ/m². It is emphasized that this value was obtained from the spacings of straight fitting lines in \(t-K_{\text{eff}}\) plots of the Co/Au, Ag/Co/ Au, Au/Co/Ag and Co/Ag superlattices for large \(t\) values and thus are not related to the saturation of the plots at the small \(t\) region directly in the determination. The calculated results are shown as solid curves in Fig. 15. Straight lines for the Au/Co/Ag and Co/Ag superlattices are also shown as a reference. It is found that the calculated curves for the Co/ Au and Ag/Co/Ag superlattices are in good agreement with the experimental points for smaller \(t\) values. In this case, the magnetoelastic constant, \(-3\lambda\), results in a 1.2–2.0 times larger value as compared with the value expected from the magnetostriction and elasticity constants of bulk Co. This is in agreement with the recent theoretical prediction that
magnetoelastic constants in Co/Au superlattices are much larger than those in bulk Co [22].

4.2. Superlattices with both large and small lattice-mismatched constituents [31]

4.2.1. Growth behavior

In the present section, Au/Co/Cu and Cu/Co/Au ternary superlattices are presented as examples of strained superlattices with both large and small lattice-mismatched, immiscible constituents. The changes of the in-plane lattice constants in the Cu/Co/Au and Au/Co/Cu superlattices are shown in Fig. 17(a) and (b), respectively. In the Cu/Co/Au superlattices, in which Co layers are grown on Au layers, the lattice constant of Co has the bulk value of Au at first and then decreases gradually toward the bulk value of Co. Thus, the behavior in the changes of the lattice constant is continuous. In the Au/Co/Cu superlattices, the lattice constant of Co also has the bulk value of Cu at first and then decreases continuously toward the bulk value of Cu. However, the gradual change in Co lattice constant is small, because the difference in lattice constant between Co and Cu is small. Therefore, it is expected that Co layers in the Cu/Co/Au superlattices contain more elastic strains than those in Au/Co/Cu superlattices.

As for the growth of the Au layer on the Co layer in the Au/Co/Cu superlattice, the lattice constant of the Au increases rapidly in 2 ML growth, but it does not recover its bulk value and gradually increases to reach the bulk value at 15 ML growth as observed in Fig. 17(b). This suggests that the Au overlayer is compressed by the underlying Co layer and that, inversely, the underlying Co layer is expanded by the Au overlayer, leading to the situation that the interface formed here is semicoherent: dislocations are not fully introduced at the interface, namely, retaining strain in the Co layer, the number density of dislocations at the interface is smaller than that for a completely incoherent interface.

In the present Cu/Co/Au and Au/Co/Cu superlattices, the situations are similar to those in the Ag/Co/Au and Au/Co/Ag superlattices described in Section 4.1.1. However, the interfaces between the Co and the Cu layers are coherent. Thus, dislocations are not present at the Co/Cu interfaces. Furthermore, the interfaces to the Au overlayers in the Au/Co/Cu superlattices are not incoherent but semicoherent, and the Au overlayers strain the underlying Co layers to some extent. This is the difference from the interfaces to the Au overlayers in the Au/Co/Ag superlattices.

4.2.2. Magnetic properties

The $K_{\text{eff}}$ as a function of the Co layer thickness in the Cu/Co/Au and Au/Co/Cu superlattices is shown in Fig. 18. The $K_{\text{eff}}$ increases with decreasing Co layer thickness for both types of superlattices. The Cu/Co/Au superlattice exhibits the perpendicular magnetic anisotropy, while the Au/Co/Cu superlattice does not, when the Co layer thickness is 5 ML. The amount of $K_{\text{eff}}$ of the two kinds of the superlattices is different, and the $K_{\text{eff}}$ in the Cu/Co/Au superlattice is larger than that in the Au/Co/Cu. Since Au is much larger than Co, while Cu is only slightly larger than Co in atomic radius, and, moreover, each Co layer in both of the two types of the superlattices is sandwiched by a Au and a Cu layer, it is again concluded that the sequence of deposition gives a large effect on the magnetic anisotropy through elastic strains contained within the ferromagnetic layers, as in the case of the Ag/Co/Au and Au/Co/Ag superlattices.

Next, we discuss quantitatively the difference of the magnetic anisotropy in the two kinds of the superlattices. Since the $t-K_{\text{eff}}t$ plots are linear, the effective magnetic anisotropy of the Cu/Co/Au and Au/Co/Cu superlattice may be expressed by the following Eqs. (13) and (14), respectively

$$K_{\text{eff}}t = [K_{\text{IC(Au)}} + K_{\text{IE(Au)}}] + K_{\text{IC(Cu)}} + K_{\text{v1}}t, \quad (13)$$

$$K_{\text{eff}}t = K_{\text{IC(Cu)}} + [K_{\text{IC(Au)}} + K_{\text{IE(Au)}}] + K_{\text{v2}}t. \quad (14)$$

From the plots, $K_{\text{v1}} = -0.73 \text{ MJ/m}^3$, $K_{\text{v2}} = -0.74 \text{ MJ/m}^3$, $[K_{\text{IC(Au)}} + K_{\text{IE(Au)}}] + K_{\text{IC(Cu)}} = 0.97 \text{ MJ/m}^2$, and $K_{\text{IC(Cu)}} + [K_{\text{IC(Au)}} + K_{\text{IE(Au)}}] = 0.74 \text{ MJ/m}^2$ are estimated. Thus, we obtain $K_{\text{IE(Au)}} - K_{\text{IE(Cu)}} = 0.23 \text{ MJ/m}^2$. This means that the magnetoelastic interface anisotropy caused by the interfaces to the Au overlayers is larger than that to the Au overlayers. If we use the value of the 0.57 mJ/m² obtained by the Au/Co/Ag and the Ag/Co/Au superlattice...
systems described in Section 4.1.2 for $K_{\text{IE(Au)}}$, we obtain $K_{\text{IE(Au)}} = 0.34 \text{ mJ/m}^2$. In this case, $K_{\text{IE(Cu)}} + K_{\text{IE(Au)}} = 0.40 \text{ mJ/m}^2$ is estimated. Furthermore, if we use the value of 0.27 mJ/m$^2$ also described in Section 4.1.2 as the $K_{\text{IE(Au)}}$ value, $K_{\text{IE(Cu)}} = 0.13 \text{ mJ/m}^2$ is obtained. This value is in good agreement with the 0.12 mJ/m$^2$, $K_{\text{IE(Cu)}}$ for Co/Cu multilayers reported by den Broeder et al. [21]. Although the difference between $K_{\text{IE}}$ and $K_{\text{IE(Au)}}$ values are small, the origins of these two values are different, since the crystal structure of the Co layers are different: The crystal structure of Co grown on the Au underlayer is hcp, while that on the Cu underlayer is fcc. $K_{\text{IE}}$ includes the magnetoelastic interface anisotropy but not the magnetoelastic volume anisotropy, while $K_{\text{IE(Au)}}$ includes both the magnetoelastic volume anisotropy and the magnetoelastic interface anisotropy, although the latter is smaller than the magnetoelastic interface anisotropy of hcp Co.

It is reported that, in (111)Cu/Co/Au/Cu quadrilayer films, the perpendicular magnetic anisotropy increases with increasing thickness of the Au interlayer, which is the underlayer of the Co layer [32]. In this situation, epitaxial in-plane strain of the Co layer increases with increasing thickness of the Au underlayer, since the in-plane lattice constant of the thinner Au layer grown on the Cu base layer is smaller than that of the bulk Au because of the smaller lattice constant of the underlying Cu. Hence, this result can be understood that the increase of the magnetoelastic interface anisotropy contributes to the increase of the perpendicular magnetic anisotropy of the film through the increase of the positive in-plane strain of the Co layer [32]. Our results on the Cu/Co/Au and Au/Co/Cu superlattices are consistent with this result and furthermore, are consistent with those for the Ag/Co/Au and Au/Co/Ag superlattices described in Section 4.1.2.

From the above discussions, it is concluded that the nature of the Co-layer interfaces depends on the sequence of deposition and that the magnetoelastic interface anisotropy originates nearly exclusively from the Au/Co interface regions of the Co layers under the Au overlayers in the Co/Au superlattices. Furthermore, it was shown that both the magnetoelastic interface anisotropy (or Neél-type magnetic surface anisotropy) and the magnetoelastic interface anisotropy contribute to the total interface anisotropy that is responsible for the perpendicular anisotropy of the Co/Au superlattices. We have also demonstrated that above two contributions to the interface anisotropy can be estimated separately.

It is noted that the contribution to the total magnetic interface anisotropy form the magnetoelastic interface anisotropy originating from the interface regions of the Co layers under the Au overlayers is much larger in the Au/Co/Cu superlattices than in the Au/Co/Ag superlattices. This is related to the following phenomena: In the Au/Co/Cu superlattices, the Au overlayers strain the Co layers that are strained by the Cu underlayers, while in the Au/Co/Ag superlattices, the Au overlayers do not strain the Co layers that are not strained by the Ag underlayers. Furthermore, in the Co/Au superlattices, the Co layers are strained by the Au underlayers exclusively near the Co/Au interfaces to the Au underlayers, but at the other side of the interface (Au/Co) the strains of the Co layers are small, and thus the Au overlayers formed on these Co layers hardly strain these Co layers. In conjunction with these findings, it is deduced that in a Y/Co/X superlattice, the growth behavior of Y on the Co layers and the effect of Y on causing the strain of the Co layers depend on the element X through the Co layer strains caused by the Co growth on the X underlayer. In fact, we have found the different changing behaviors of in-plane lattice constant in RHEED observations [13]: At the Y/Co interface, the lattice constant of Y (= Au, Ag) is discontinuous for X = Ag, and it is continuous for X = Au. In the former case, the Co layers are not strained by the X underlayers, while they do in the latter case although the strains are small. This situation results in the phenomenon that the magnetoelastic interface anisotropy originating from the Y/Co interface depends on the element X in the Y/Co/X superlattice.

5. Ternary strained superlattices containing both miscible and immiscible constituents

5.1. Co-based superlattices [18,19,33]

5.1.1. Growth behavior

Here, growth of Pt/Co/Ag and Ag/Co/Pt ternary superlattices is presented as examples of Co-based superlattices. In both of the two types of the superlattices, the structures of the Pt and Ag layers are fcc. The streaks arising from the Co layer are rather broad, indicating that the in-plane coherence-length is relatively short. This is presumably due to the presence of dislocation arrangements breaking up the Co layers to small in-plane domains. In
RHEED patterns of Co layers, the intensity modulations along streaks are rather distinct, indicating that the structure of Co is twinned fcc with some stacking faults. The epitaxial relationships of the Ag/Co/Pt and Co/Pt superlattices keep orientational relationships that their closed-packed planes are parallel. In a Pt/Co/Ag superlattice, RHEED patterns of Co indicate that the stacking is nearly random rather than an fcc or an hcp stacking. This crystal structure is only observed in thin Co layers in the Pt/Co/Ag superlattices. These results indicate that the structural coherence and the chemical order of the Pt/Co/Ag superlattices are relatively low.

Fig. 19(a) and (b) shows the relative lattice constant as functions of the layer thickness of Ag/Co/Pt and Pt/Co/Ag superlattices grown at 323 K, respectively. In the Ag/Co/Pt superlattice, the lattice constant of the Co layer decreases gradually toward its own value in the bulk with increasing Co layer thickness. On the other hand, in the Pt/Co/Ag superlattice, the Co layer grows on the Ag layer with its own lattice constant in the bulk and the Co/Ag interface is incoherent. These growth modes are similar to those in the Co/Pt and the Co/Ag superlattices, respectively. In the Pt/Co/Ag superlattice, Pt layers on the Co layers grow incoherently as in the case of Ag on Co in the Co/Ag superlattices, and contrary to the growth behavior of Pt on Co layers at the beginning in the Co/Pt superlattices. Ag layers on the Co layers grow semicoherently in the Ag/Co/Pt superlattice. This growth behavior of Ag on Co is observed only in the Ag/Co/Pt superlattices with thin Co layers. In the Ag/Co/Pt superlattices with Co layers whose thicknesses are larger than about 10 ML, Ag layers grow incoherently on Co layers. These results means that Ag and Pt layers can grow semicoherently only on the diluted Co layers, which are produced by depositing Co on the Pt underlayers. This situation is similar to that in the Ag/Co/Au superlattice system described in Section 4.1.1.

5.1.2. Magnetic properties

Fig. 20 shows \( K_{\text{eff}} \) as a function of \( t \) in the Co/Pt (○), Ag/Co/Pt (●) and Pt/Co/Ag (■) superlattices at 300 K. The dependence of \( K_{\text{eff}} \) on \( t \) is linear for both the Co/Pt and Ag/Co/Pt superlattices, showing that both \( K_v \) and \( K_i \) are constants in Eq. (1). In the Co/Au superlattice system, a deviation of \( K_{\text{eff}} \) value from the straight line, i.e. a saturation of the \( K_{\text{eff}} \) value in a region with \( K_{\text{eff}} > 0 \) exists, as described in Section 4.1.2. The saturation of \( K_{\text{eff}} \) value is due to the strain gradient along the thickness-direction in the Co layers caused by the epitaxial strain upon depositing Co onto Au layers. The absence of the saturation in the Co/Pt and Ag/Co/Pt superlattices, therefore, suggests that the contribution of the magnetoelastic interface anisotropy to the perpendicular magnetic anisotropy in Co/Pt is small or negative, although appreciable strains are observed in the RHEED measurements, as described in Section 5.1.1.

From Eq. (1) the value of \( K_i \) is estimated to be 1.29 mJ/m² for the Co/Pt superlattices. As seen in Fig. 20, the apparent
$K_1$ value for the Ag/Co/Pt superlattices is as large as 0.82 mJ/m$^2$. This is due to the large $K_1$ value of the Co/Pt interfaces in the Ag/Co/Pt superlattices, since the $K_1$ value for the Co/Ag interfaces is small. The values of $K_s$ are very similar: $-0.98$ MJ/m$^3$ for Pt/Co and $-0.96$ MJ/m$^3$ for Ag/Co/Pt. $K_s$ consists of the shape-, the magnetocrystalline- and the magnetoelastic-anisotropies. The crystal structure of the Co layers is fcc in both the Co/Pt and Ag/Co/Pt type superlattices, and the shape magnetic anisotropy energy $-\mu_0 M^2/2$ were comparable with the bulk value or slightly larger. Assuming the values of magnetocrystalline anisotropy energy and the saturation magnetization of bulk hcp Co, the $K_s$ value is estimated to be $-0.72$ MJ/m$^3$. The deviation of the $K_s$ value in the present superlattices from this estimation is presumably ascribed to the fcc structure of Co whose magnetocrystalline anisotropy energy will be much smaller than that of hcp Co, and probably due to the negative magnetoelastic contribution arising from the positive magnetostriction constant of the Co–Pt system [34].

In the case of the Ag/Co/Pt superlattices, Eq. (1) is rewritten as

$$K_{\text{eff}} = K_s + [K_{\text{f(Pt)}} + K_{\text{f(Ag)}}]t,$$

(15)

where $K_{\text{f(Pt)}}$ and $K_{\text{f(Ag)}}$ are the two-dimensional anisotropies at Pt- and Ag-interfaces of a Co layer, respectively. The value of $K_{\text{f(Ag)}}$ can be obtained from Eqs. (1) and (15), and is 0.35 mJ/m$^2$. This value is much larger than the 0.07 mJ/m$^2$ in the Co/Ag superlattices. In the case of the Co/Ag superlattices grown at 323 K, the crystal structure of the Co layers is hcp. As described above, Co layers have an fcc structure, and a possible cause for the difference in $K_s$ value of the Co/Ag interfaces might be the difference in crystal structure, although it is generally considered that the Néel type surface anisotropy (the magnetocrystalline interface anisotropy) predominantly depends on the constituents of the superlattices and not on the crystal structure [3,35]. The $K_{\text{eff}}$ values as a function of $t$ for the Co/Ag superlattices with the hcp Co layers are also shown in Fig. 20. The plot for average values of $K_{\text{eff}}$ for the Co/Pt and the Co/Ag superlattices shown by a broken line in the figure is located very near to the position of the plot for the Pt/Co/Ag superlattices, although the inclination is different. It is noted that the $K_s$ value (inclination) depends on the magneto-crystalline anisotropy, which is sensitive to the crystal structure of Co. Hence, the difference in the structure (fcc in Co/Pt and hcp in Co/Ag superlattices) is responsible for the inclination difference.

In the Co/Au and Ag/Co/Au superlattice systems, in Section 4.1.2, the $K_{\text{eff}}$ value of the Ag/Co/Au superlattices is much larger than the average $K_{\text{eff}}$ value of the Co/Au and Co/Ag superlattices and the difference in $K_{\text{eff}}$ between the Co/Au and Ag/Co/Au superlattices is rather small. The absence of this effect in the Co/Pt and Ag/Co/Pt superlattice systems suggests that the magnetoelastic interface contribution in the Co/Pt system is small. This is consistent with the linear nature of the $t$–$K_{\text{eff}}$ plot in the Co/Pt system and the positive magnetostriction constant of the Co–Pt system, described before. The $K_{\text{eff}}$ value of the Pt/Co/Ag superlattices is much smaller than that of the Ag/Co/Pt superlattices, as seen in Fig. 20. This is presumably due to the more disordered structure of the Pt/Co/Ag superlattices.

5.2. Ni-based superlattices [36]

5.2.1. Growth behavior

Here, growth of Ag/Ni/Au and Au/Ni/Ag ternary superlattices is presented as examples of Ni-based superlattices. In the bulk state, Ni is about 14% smaller than Au and Ag in atomic radius. The changes of the in-plane lattice constants in the Ag/Ni/Au and Au/Ni/Ag superlattices are shown in Fig. 21(a) and (b), respectively. In the Ag/Ni/Au superlattice, the in-plane lattice constant decreases continuously in the stage of Ni deposition on Au, and the lattice constant of the subsequently grown Ag recovers discretely within a few monolayers. In the Au/Ni/Ag superlattice, Ni on Ag does not have the same lattice constant as that of Ag and the two streaks of Ag and Ni coexist during a few monolayers in the RHEED patterns. Namely, the Ni lattice constant immediately decreases to the bulk Ni value. The lattice constant in the subsequently deposited Au on the Ni discretely returns to the Au value. The behavior in the changes of the lattice constant is discontinuous. The RHEED pattern of Ni on Ag in the Au/Ni/Ag superlattice is spotty at the early stages of deposition, but it becomes streaky with increasing Ni layer thickness.

Next we discuss the difference in lattice continuity during
depositions in terms of two points: mixing and 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 two elements do not mix each other even in the liquid state. The fact that there are two types of streaks in the Au/Ni/Ag superlattice indicates that two layers of Ni and Ag exist separately without mixing. The lattice constant discontinuity means that the Au/Ni/Ag superlattice contains little strain. On the other hand, although the Ni–Au system is a two-phase separation system in the equilibrium phase diagram and there is no solubility at low temperatures, they can be soluble when the temperature is higher. Therefore, in the Ag/Ni/Au superlattice, there may exist mixing regions at the interface of Ni on the Au. Moreover, the large lattice strain may 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 estimated in Section 5.2.2.

5.2.2. Magnetic properties

The results of saturation magnetization $M_s$ measurements Au/Ni/Ag and Ag/Ni/Au superlattices at room temperature are shown in Fig. 22. The $M_s$ values of the Au/Ni/Ag superlattice are larger than those of the Ag/Ni/Au superlattice for all $t_{Ni}$, the Ni layer thickness. Similar tendency is observed in the results for a low temperature of 10 K. 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 the bulk value in thicker regions beyond 2 nm, while it decreases with decreasing $t_{Ni}$. The decrease of $M_s$ is caused by the degradation of the layer structure in thinner Ni regions. In the Ag/Ni/Au superlattice, the value of $M_s$ increases gradually with increasing $t_{Ni}$ but is still smaller than the Ni bulk value even at a thicker region as thick as 3.3 nm.

In order to clarify the degree of the contribution of mixing regions and strained regions at/near the interface, we calculated how much strain is contained in the Ni layer using the method presented in Section 4.1.2. 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. By comparing the experimental $M_s$ value with the bulk $M_s$ value, the mixing layer is determined to be around 5 ML, with referring to the above calculation. By excluding the mixing layer, the amount of strain in the Ni layer is evaluated. The amount of the strain is determined to be 2.7% at the sixth Ni layer that 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.

The $K_{\text{effNi}}$ as a function of $t_{Ni}$ in the Au/Ni/Ag and Ag/Ni/Au superlattices is shown in Fig. 23. The $K_{\text{effNi}}$ increases with decreasing Ni layer thickness for both types of superlattices. However, even when the thickness of the Ni layer is 1 nm, both types of the superlattices show in-plane magnetization, i.e. they do not exhibit the perpendicular magnetic anisotropy. The value of $K_{\text{eff}}$ in the two kinds of the superlattices is different, and $K_{\text{eff}}$ in the Ag/Ni/Au superlattices is larger than that in the Au/Ni/Ag superlattices.

Next, let us consider the difference in magnetic anisotropy of the two kinds of the superlattices. We can understand it by the following calculation. The $K_{\text{eff}}$ of the Ag/Ni/Au superlattices may be expressed by the following equation:

$$K_{\text{eff}}t_{Ni} = [K_{IC(Ag)} + K_{IC(Ag)}] + K_{AE} + (K_C + K_D)t_{Ni}.$$  \hspace{1cm} (16)
Here, for $K_C$, we give the bulk value and for $K_{\text{IE}}$ and $K_{\text{D}}$, we use the values calculated from the experimental data. For the Au/ Ni/Ag superlattices, the term $K_{\text{IE}}$ is neglected in Eq. (16) since the Ni layers are not strained. We assume that $[K_{\text{IC}}]_{\text{Ag}} + K_{\text{IC}}(\text{Ni})$ is in both types of the superlattices are the same, although mixing at the interfaces is different between both types of the superlattices. In the calculation of $K_{\text{IE}}$, magnetostriction constant $\lambda$, Young’s modulus $Y$ and the amount of strain $\varepsilon$ are needed. For the former two, $\lambda$ and $Y$, a half bulk value and the bulk value, respectively, are utilized and the $\varepsilon$ is evaluated from the results of the present RHEED observation, using a method [20] given in Section 4.1.2. The calculated $K_{\text{eff}}$ is also shown in Fig. 23 by solid lines. The experimental and calculated results agree well. Therefore, we conclude that the difference in anisotropy is due to the different strained-structure caused by the difference in stacking sequence.

6. Concluding remarks

We have reviewed growth behaviors of binary and ternary metallic strained superlattices investigated mainly by our group, and summarized discussions on their magnetic properties, especially on the magnetic anisotropy, in terms of their structural characteristics. First, we introduced our RHEED system that works efficiently under a magnetic field arising from evaporation sources for low vapor-pressure materials. Then, MBE-grown binary strained superlattices, Co/Au, Co/Pt and Cu/Au, were discussed, with comparing to the incoherent superlattices of Co/Ag and Cu/Ag having nearly the same lattice mismatch of constituents. Next, we reviewed ternary strained superlattices with immiscible constituents with reverse deposition sequence, Au/Co/Ag and Ag/Co/Au superlattices, and Au/Co/Cu and Cu/Co/Au superlattices, in relation to the growth behaviors of binary superlattices. Finally, ternary strained superlattices containing both miscible and immiscible constituents, Pt/Co/Ag and Ag/Co/Pt superlattices, and Au/Nil/Ag and Ag/Nil/Au superlattices, were reviewed. As for the magnetic properties, the magneto crystalline interface anisotropy and magnetoelastic interface anisotropy were discussed as the possible origins of the perpendicular anisotropy in the magnetic superlattices. We emphasized that the structural properties sensitive to chemistry and geometry of the interfaces which are brought about by the deposition sequences strongly affect the magnetic properties: A procedure of forming an interface features the structural and thus magnetic properties of the interface and hence the interfaces are different even when the final macroscopic structure appears the same. For a reasonable theoretical interpretation of the magnetic properties of superlattices, therefore, one should take this asymmetric nature into account.

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