Structure and exchange coupling in (100) oriented Co/Ir artificial alloys

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Co/Ir artificially ordered alloys which do not form any inter-metallic composition at equilibrium state have been fabricated by controlling layer-by-layer growth of each element. A structural long range order parameter of 0.26 was obtained for the [Co(1 ML)/Ir(1 ML)](100) epitaxial multilayered films and it reveals that the $L1_0$ structure is partly realized. Significant differences between both Co/Ir(100) and (111) multilayers were found in magnetic properties, namely, small magnetic anisotropy in (100) samples and strong one in (111) oriented multilayers with thicker layer thickness than 3 ML. The amplitudes of peak interlayer coupling are almost the same in both alloys with different growth direction.
Key words: magnetic multilayer, magnetic coupling, Co, Ir, (100) growth

1. INTRODUCTION

Developments of highly controlled vacuum deposition technique has realized fabrications of artificial materials such as superlattices and nano layered structures. Inter-layer coupling through non-magnetic layers was reported on an Fe/Cr multilayer.[1] Following to this, Giant Magneto-Resistance(GMR) was discovered and magnetic multilayers have been attracted much attention because of the interest of physical origin and high potential for application in such materials.

Oscillatory inter-layer coupling between ferromagnetic layers has been reported in many systems including nonmagnetic 3d, 4d and 5d metals experimentally and is highly dependent on their thickness and growth orientation.[2] Co/Ru superlattices show the strongest inter-layer magnetic coupling of 5 erg/cm^2 so far.[3] In such a system, the amplitude of the coupling constants becomes larger as the layer thickness of the nonmagnetic elements decreases. The limit for the thinnest spacer make the system to be an artificial alloy. Growth of metals by mono-layer (ML) units can be carried out by using this approach. Recently Takanashi *et al.* reported Fe/Au artificial alloy which is epitaxially grown along fcc [100] Au direction.[4,5] They deposited two metallic elements with controlling the layer thickness on the single crystal substrate with proper buffer layers and artificial $L1_0$ structure, where the layer-by-layer heterogeneous structure along fcc [100] axis. Clearly defined ferromagnetism has been found for Fe/Au mono-layer alloy. Very recently, a Co/Ru system has been tested as a artificial material by the same authors.[6]

Co/Ir layered structures also show strong coupling after the Co/Ru system which has the largest interlayer coupling. Several have been reported for the magnetic coupling[7,8] and transport properties[9] of Co/Ir multilayer and trilayer systems.[10,11] The peak magnitudes of coupling were almost same as the value first reported by Parkin.[2] We have investigated epitaxial multilayers preferentially grown along (110) of fcc Ir by sputtering technique and the Co layers of the

films were found to be in the hcp phase, whose [001] axes aligned parallel to in-plane Ir[001].[12] Epitaxial growth of Co/Ir was performed by the use of an MBE technique for Ir (100) plane parallel to the substrate surface and it was found that the Co layer thickness less than 10 Å is essential for the layer-by-layer growth. We have reported 2.2 erg/cm^2 of AF exchange coupling in a [Co(7 Å)/Ir(5 Å)]N=50 (100) multilayer and its saturation field was more than 90 kOe.[7]

It is of interest that even the thickness of Ir less than 3 ML can realize stable AF coupling between Co layers. We have investigated Co/Ir(111) artificial alloys with equal layer thickness.[13] Co-existence of strong anti-ferromagnetic inter-layer magnetic coupling and perpendicular anisotropy was found and spin flip transition was observed.

In this paper, we report fabrication and magnetism of artificial alloys of Co/Ir grown along fcc Ir [100] direction (see Fig. 1). Magnetic properties will be described for $n = 1$ to 5 for the repeated multilayers of bi-layer of Co(n ML) and Ir(n ML). Relation between growth direction and magnetic anisotropy will be also discussed together with magnetic inter-layer coupling.

2. EXPERIMENTAL PROCEDURE

Multilayers and artificial alloys of Co/Ir were grown by vacuum deposition under ultra-high vacuum using a conventional MBE system.[14] Two e-guns independently controlled were used for the evaporation of metals. Base pressure of the growth chamber was 1 ~2 x 10^{-9} and growth pressure was order of 10^{-8} Torr. The significant change in vacuum was due to the high melting temperature of Ir. Detailed condition was described in the former report.[7]

Samples were epitaxially grown on single crystal MgO and sapphire. Prior to growth, the substrates were heated to 700 °C for three hours or more in a growth chamber. In the case of (100) growth, layers of 50 Å Cr and 100 Å Au were subsequently grown on the MgO(001) substrate as seed layers at the substrate temperature of 250 °C. It is noted that the buffer layers

are different from the previous report in order to eliminate additional diffraction peaks around $2\theta = 23°$ and $73°$ for Cu-$K\alpha$ radiation, which were originated from Fe and Pt.
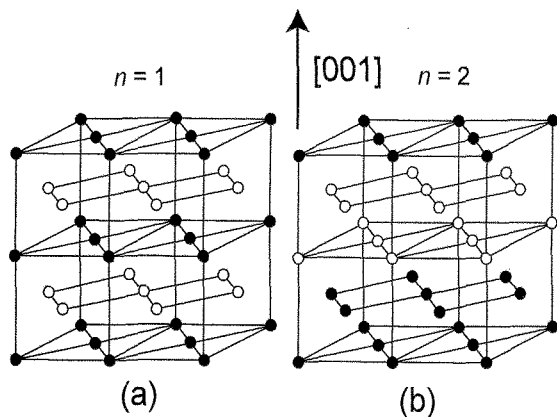

(a)   **(b)**

Fig. 1. Crystalline structures of artificial structure realized in this work. Closed and open circles represent different sorts of atoms, Co and Ir, $n = 1$ and $n = 2$ structures are shown in (a) and (b). Growth direction is shown by an arrow in the figure.

Growth rates were 0.1 Å /sec for Cr and Pt, and 0.2 Å /sec for Au and Ir. Co/Ir superlattices were grown at ≈ 60 °C to suppress alloying and inter-diffusion. The deposition rate was controlled to be $0.15 \sim 0.2$ Å /sec. Here, 1 ML for fcc (100) and (111) planes are 1.92 and 2.2 Å for Ir, and 1.77 and 2.05 Å for Co, respectively. Mono-layer thickness, n, was varied from 1 to 4 for the (100) growth and 1 to 5 for (111). Superlattices were designed where numbers (n) of ML of Co and Ir were equal in a bilayer. The repetition number(N) of bilayers was chosen as the total Co layers were constant to be 200 ML's and a final Ir layer of 50 Å thick was deposited in order to prevent oxidation. The sample with layer thickness of n ML is referred as [Co(n ML)/Ir(n ML)](100) for (100) growth.

The structural analysis of superlattices was examined *in situ* with Reflection High Energy Electron Diffraction (RHEED) and *ex situ* with x-ray diffractometry by Cu-$K\alpha$ radiation. Magnetization measurements were performed at room temperature with a commercial SQUID magnetometer with magnetic fields up to 55 kOe applied mainly along the in-plane MgO[100] for Ir(100) samples and in-plane. Magnetization per unit volume of Co was deduced assuming that only Co layers contributed to the total magnetization.

3. RESULTS

RHEED patterns during the deposition were recorded to examine the growth state of the films. From Fig. 2, in the case of (100) multilayer growth, clear streak patterns were observed through the entire growth process (Fig.1(c) and (d)), while diffraction spots in RHEED pattern appeared for the case of co-deposition alloy growth(see Fig. 1(b)). This means that the multilayer growth is in layer-by-layer mode with atomically flat surface, however co-deposition films

were grown in 3 dimensional island growth mode with the coarse surface compared with that of the multilayers. Although there is remarkable difference in growth mode
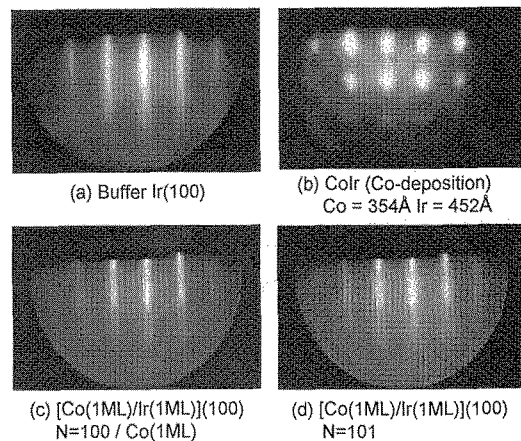

(a) Buffer Ir(100)   (b) CoIr (Co-deposition) Co = 354Å Ir = 452Å

(c) [Co(1ML)/Ir(1ML)](100) N=100 / Co(1ML)   (d) [Co(1ML)/Ir(1ML)](100) N=101

Fig. 2. RHEED patterns during the growth of [Co(n ML)/Ir(n ML)] multilayered films. Patterns for the (100) Ir buffer layer (a), co-deposited (100) CoIr film of Co 354 Å and Ir 452 Å (b), [Co(1 ML)/Ir(1 ML)](100) N= 100/Co(1ML) (c), and [Co(1 ML)/Ir(1 ML)] (100) N=101 (d).

between multilayer and co-deposition alloy films, in both cases, these films keep growing with same crystal orientation to that of the buffer layers. It is noted that good epitaxial growth could not be obtained when n was larger than 5 for (100) growth.

Fig. 3 shows X-ray diffraction patterns of [Co(n ML)/Ir(n ML)](100) multilayered films. The uppermost of the patterns shows that of a Co-Ir co-deposited alloy prepared with the same procedure as the multilayers cases for the reference. Closed circles express the fundamental reflection corresponding to the mean lattice constant in the films along the growth direction. Clear (100) superlattice peak was found in the $n = 1$ film. These lines means the formation of L1$_0$ structure in the film. A long range order parameter, S, is defined as $S = (r_A - F_A)/(1 - F_A)$. Here, r_A, and F_A are probability of A atoms on the A site and concentration ratio of A atom in a alloy. Diffraction intensities of ordered alloys would be expressed as follows:

$$\frac{I_S}{I_f} = S^2 \left(\frac{I_S}{I_f}\right)^* \qquad (1)$$

where I_S and I_f are diffraction intensities of a satellite peak and a fundamental peak.[4,15] Parentheses with asterisk in the right side of the equation express the calculated intensities for the ideally ordered alloy. The S value was deduced to be 0.26 in the present case. In this case, F_A is 0.5 and r_A is estimated to be 0.63 which must be compared with the value of 0.5 for the random alloy and 1 for the ideally ordered alloy. This suggests that the L1$_0$ structure was not perfectly established. The solubility of Co into Ir is not negligible because they form homogeneous solutions over the entire composition
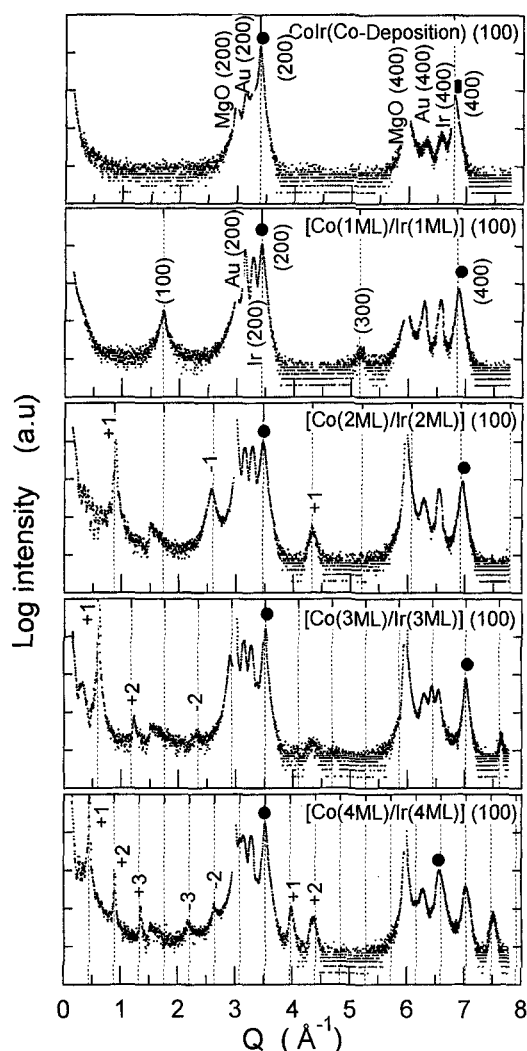
Fig. 3. Log intensity of X ray diffraction patterns in [Co(n ML)/Ir(n ML)](100) multilayered films. n denotes an each layer thickness in a bilayer. The upper most graph shows the pattern for a Co-Ir co-deposited alloy prepared with the same procedure. Closed circles in the figure depict the principal diffraction corresponding to the averaged lattice parameter along diffraction vector. Superscripts of indices express the order of satellite peaks from the principal diffraction. Dashed lines are guides for positions at Q=2π m/λ (m=1,2,3...). Broaden peaks around Q=1.5 originated in a sample holder for X-ray diffraction study.

range. Therefore, non-negligible intermixing at the interface is anticipated. This will be discussed later in connection with magnetization. The diffractions for other layer thickness, n = 2, 3, 4, satellite peaks due to the superlattice structure were observed in expected position.

Magnetization curves were recorded at room temperatures and also at 100 K for [Co(n ML)/Ir(n ML)](100) multilayered films and shown in Fig, 4 for n = 1 to 4 samples. That of a co-deposition alloy with almost equal atomic composition is also shown in the

figure. Magnetization of co-deposition film and n = 1 sample increase with decreasing temperature, while samples with $n \geq 2$ has small temperature dependence.
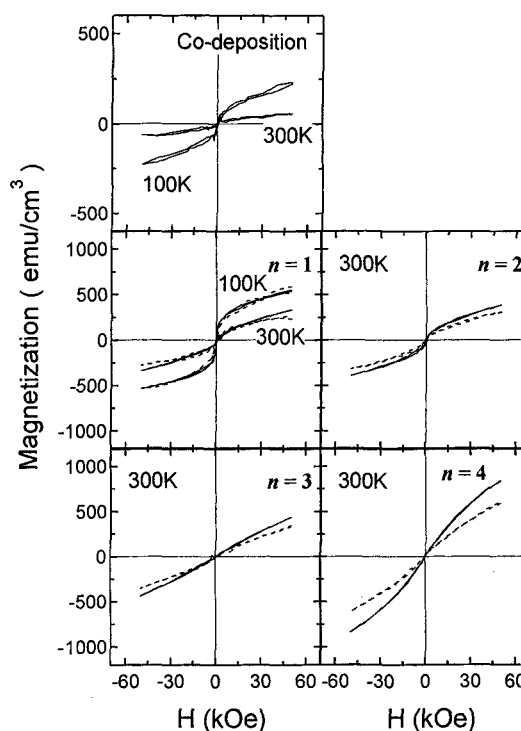

Fig.4   Magnetization curves for [Co(n ML)/Ir(n ML)](100) multilayered films with n = 1, 2, 3 and 4. Solid and dashed lines display in-plane and perpendicular magnetization curves. Sample temperatures are 100 K and 300 K.

Those for n = 1 and 2 are composed of small ferromagnetic part and linear part against an applied field. The magnitude of room temperature magnetization per unit volume of Co in the films are around 500 emu/cm^3 and smaller than 1000 emu/cm^3 for the multilayer where Co layer thickness were 7 Å ($n \sim$ 4).[7] When n = 3, magnetization linearly increase with the increase of magnetic field and did not change so much even at 100 K. This is different from cases for n = 1, in which the magnetization increased remarkably at low temperatures compared with room temperature value. When n =4, the magnetization reached at 800 emu/cm^3 at 50 k Oe and did not saturate at this field.

4. DISCUSSION

From the structural analysis, the artificial L1$_0$ structure was found to be partly realized in the [Co(1 ML)/Ir(1 ML)](100) sample. The earlier trial to grow the artificial structure was done for the Fe/Au system by Takanashi et al.[7] The long range order parameter of 0.26 obtained here is smaller than that for Fe/Au, 0.7. This is probably due to the difference of the solubility of component elements. In the case of Co/Ir, the solubility of Co to Ir is not negligible, while Fe/Au is known as an immiscible system. Inter-diffusion at the interface can be estimated by the decrease of magnetization about 1000

emu/cm^3 for Co layers of 7 Å. Assuming the moment reduction occurs only at the interfaces between Co and Ir layers, we roughly evaluated the thickness of the "dead" layer to be 0.5 ML, namely the interface Co layer has half magnitude of bulk. It is noted that, in the case of (111) samples, the superlattice peaks in $n = 1$ were seen in the X-ray diffraction pattern, however, was weaker than the case of (100) films.[13] The fact implies that the inter diffusion of Co and Ir is pronounced on the (111) growth.

The magnetism of the (100) sample with $n = 1$ is not clear at room temperature. The magnetization at 50 kOe was 40 % of that in the $n = 4$ sample and did not saturate. The magnetization increased and showed typical ferromagnetic characteristics as measurement temperatures were lowered. These facts suggest that the magnetic state of the $n = 1$ sample resembles to that of alloys whose transition temperature are distributed between room temperature and 100 K. On the other hand, temperature dependence of the magnetization process is not significant and Curie temperature of $n = 2$ sample is estimated to be much higher than room temperature. The amplitude of the magnetization is about half of films with Co layer thickness larger than n $= 3$ then the sample is considered to be almost in anti-ferromagnetic mixed with small portion of ferromagnetic part. In the case of $n = 3$ film, MH curves at room temperature(see Fig. 4) was found to be nearly a straight line and the same as at 100 K. The result supports a strong AF coupling exists and the Ir layer thickness coincides with the previous data for a [Co(7 Å)/Ir(6 Å)](100) multilayer.[7] As the n increases to 4, the magnetization curve changes slightly into ferromagnetic like with smaller saturation field. In this film, ferromagnetic interaction mixed with the strong AF coupling seen in the $n = 3$ case.

Comparing the magnetization process for the present system with those for multilayers[7], we can estimate the amplitude of AF coupling constant(J_{AF}). Providing the same amplitude of saturation magnetization of 1000 emu/cm^3 as the case of (111) growth[13], the saturation magnetic field(H_s) was estimated to be 130 ± 20 kOe for $n = 3$ sample by extrapolating the magnetization curve. Using the relation, $J_{AF}=M_s t_{Co}H_s/4$, J_{AF} is deduced to be 1.7 ± 0.25 erg/cm^2. This amplitude is slightly smaller than that for multilayers, 2 erg/ cm^2. It may be due to the difference in the saturation magnetization between the present case with $t_{Co} = 5.3$ Å and the multilayer case with $t_{Co} = 7$ Å. Taking into account this difference, the magnitudes agree with that of previous work and the strong AF coupling in the Co/Ir(100) system is reconfirmed in the present work.

5. CONCLUSION

We have succeeded to grow Co/Ir(100) atomically controlled superlattices via MBE technique. Although Co and Ir form homogeneous alloy all over the composition range at equilibrium state, we obtained L1$_0$ type Co/Ir(100) partly ordered artificial alloys with $S = 0.26$ for $n = 1$ and also highly ordered epitaxial superlattices for $n \geq 2$. Growth mode was layer-by-layer for superlattices grown on (100) orientation and 3-dimensional island growth for epitaxial alloy films. The magnetism of [Co(n ML)/Ir(n ML)](100) for $n = 1$

looks like that of the alloys and shows apparently small moments. The films for $n = 2$ shows ferromagnetic MH curves. The films thicker than $n = 3$, show strong AF coupling.

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