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# Misfit strain and magnetic anisotropies in ultrathin Co films hetero-epitaxially grown on Au/Cu/Si(1 1 1)

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## Abstract

The relation between misfit strain and uniaxial perpendicular magnetic anisotropy (PMA) has been studied in ultrathin Co single films hetero-epitaxially grown on Au/Cu/Si(1 1 1), by means of spin-wave Brillouin light scattering. The misfit strain in Co was intentionally controlled by the thickness of the Au-interlayer. The interface PMA with Cu-overlayers is proportional to the misfit strain, while the volume PMA is equal to a bulk value. We show Co-thickness dependences of PMA in the atomic-scale thickness region to be thinner than several monolayers (ML). The experimental values of the PMA are smaller than the calculated ones, while assuming constant volume and interface PMAs, when the misfit strain is large. In addition, the interface PMA is largely affected by ultrathin Au-overlayers, which indicate a significant deformation of the Co lattice near the upper interface after forming those overlayers. The interface PMA increases monotonically with increasing Au-overlayer thickness ranging from 1 to 3 ML. © 2002 Published by Elsevier Science B.V.

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## 1. Introduction

We have grown hetero-epitaxial hcp-(0001) planes of ultrathin Co films on a  $7 \times 7$ -Si(111) surface with ultrathin Cu- and additional Au-buffer layers by molecular beam epitaxy. For the use of this hybrid ferromagnetic-semiconductor structure for spin-related electronic or photonic devices, magnetic anisotropies should be examined

from a microscopic point of view [1]. In this paper, the relation between misfit strain and uniaxial perpendicular magnetic anisotropy (PMA) has been studied in ultrathin Co single films grown on Au/Cu/Si(111), by means of spin-wave Brillouin light scattering (BLS). Such PMA is especially important, since it can be applied for many magnetic devices including storage media due to its bistability and large strength [2]. In ultrathin magnetic films, effects of the interfaces on the PMA become significant. Therefore, we evaluate the interface PMA solely from the Co-thickness  $t_{\text{Co}}$  dependence of PMA. In addition to this, we

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focus on single magnetic film structures rather than superlattices, because the roles of underlayer and overlayer for the PMA can be separated. The effects of overlayers, which have a larger lattice constant than the Co layer, on the PMA have not been well-examined. Large matrix samples were prepared under identical ultra-high vacuum (UHV) conditions with different thicknesses of the Co layers and other underlayers or overlayers, which allowed us to investigate unambiguous dependences of the PMA on the thicknesses of those layers.

## 2. Experimental procedures

Details of the sample preparation were described elsewhere [3]. Ultrathin Co films were deposited on a  $7 \times 7$ -Si(111) surface with buffer layers, under UHV. After forming a 4 nm-thick Cu-buffer layer, an Au-stepped wedge was deposited with various thicknesses from 0 to 5 monolayers (ML) using a PC-controlled shutter. Then, the substrate was rotated by  $90^\circ$  and a Co wedge was deposited, which was followed by a Cu-overlayer. We made two types of Co wedge sample. One had a stepped wedge with the Co-thicknesses ranging from 4 to 10 ML (matrix samples A), and another one had a gradient-thickness wedge of Co ranging from 0 to 7 ML in order to examine the very thin region (matrix samples B). Additionally, one more set of matrix samples was prepared with a Co-stepped wedge and a stepped wedge of Au-overlayer to examine the effects of ultrathin overlayers (matrix samples C). This Au-overlayer wedge was covered with an additional Cu-overlayer to prevent oxidation of the Co. Magnetic anisotropy constants were determined from the magnetic-field dependence of the spin-wave BLS frequency by taking into account the effects of the magnetization direction, the magneto-static and exchange energies depending on  $t_{\text{Co}}$ . The interface PMA constant was determined from the  $t_{\text{Co}}$ -dependence of PMA.

## 3. Results and discussion

In our previous report [3], a lattice constant of Au on Cu(111) increased with increasing Au-

thickness and reached a bulk value at 5 ML. A lattice constant of Co was uniformly expanded due to its hetero-epitaxial growth on the Au, which was followed by a typical misfit-type lattice relaxation. The lattice mismatch of the Co/Au interface was up to 14% with 5 ML of the Au-interlayer. The PMA constant of Co in that hetero-epitaxial system is well-expressed by a sum of uniaxial perpendicular interface and volume PMA constants above 4 or 5 ML. The interface PMA with Cu-overlayers is proportional to the misfit strain, while the volume PMA is equal to that of the bulk value [4]. It is understood within a framework of magneto-elastic effect caused by the misfit strain  $\varepsilon$ . The effect of in-plane misfit strain on the anisotropy has been previously discussed based on a critical layer thickness  $t_C$  [5].  $\varepsilon$  is phenomenologically expressed by an equation as  $\varepsilon = (a/t_{\text{Co}}) + b$ , except for the initial few ML of Co, above  $t_C = 1$  ML. The constant  $a$  indicates an amplitude of  $t_{\text{Co}}$ -dependence of the misfit strain near the lower interface, and  $b$  is the thickness-independent uniform strain. The thickness-dependent variation of  $\varepsilon$ , which is expressed by this constant  $a$ , can contribute to the interface anisotropy through the magneto-elastic effect. Fig. 1 shows Au-interlayer thickness dependences of  $a$  and the PMA constants in our Cu/Co(x)/Au(y)/Cu(111)-matrix samples A. Here,  $a$  was determined from the  $t_{\text{Co}}$ -dependence of the in-plane lattice spacing, which was quantified by continual observations of RHEED patterns during the film growth. As can be seen, it is clearly shown that the first-order interface PMA constant  $K_{\text{u,I}}^{(1)}$  increases monotonically with increasing Au-interlayer thickness and it coincides well with the monotonic increase in  $a$ . It is shown that the value of  $b$  is  $<0.2\%$  for the system with the Au-interlayer. This agrees with the fact that the volume term  $K_{\text{u,V}}^{(1)}$  is almost a bulk value. On the other hand,  $K_{\text{u,I}}^{(1)}$  of Co/Cu is higher than that of Co/Au/Cu, although  $a$  is fairly close to 0. This is a clear evidence of the effect of modification of the electronic structure at the Co/Cu interface on  $K_{\text{u,I}}^{(1)}$ . On the contrary, the decrease in  $K_{\text{u,V}}^{(1)}$  in Co/Cu indicates the insufficient quality of the crystallinity as in the combination of Fcc (100) and (110) planes.

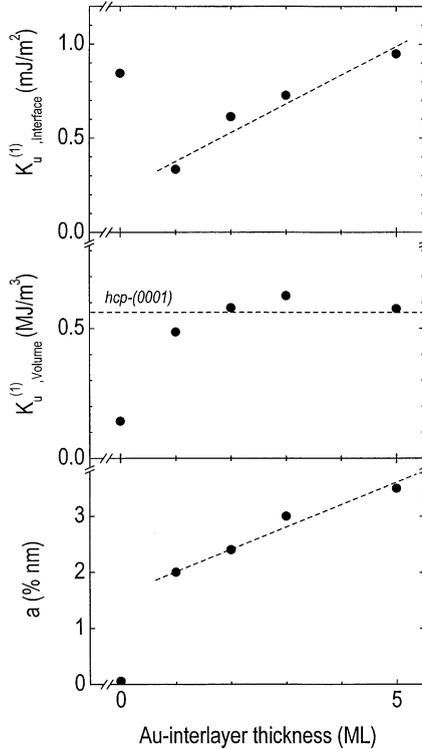


Fig. 1. The first-order interface PMA constant  $K_{u,I}^{(1)}$  and the volume PMA constant  $K_{u,V}^{(1)}$  as a function of Au-interlayer thickness. A strain parameter  $a$  which is determined from the  $1/t_{Co}$ -dependence of in-plane strain is also plotted (see text). Broken lines for  $K_{u,I}^{(1)}$  and  $a$  are guide for the eye. A broken line for  $K_{u,V}^{(1)}$  shows a value for well-characterized bulk-like HCP-Co(0001) films [6].

Next, we show the  $t_{Co}$ -dependence of the anisotropies in ultrathin film region thinner than 7 ML, with various Au-interlayer thicknesses. The result for matrix samples B is shown in Fig. 2, where the product of  $K_u^{(1)}t_{Co}$  is plotted as a function of  $t_{Co}$ . Here, solid lines are best-fitted calculations for matrix samples A used for the above experiment with the Co-thicknesses ranging from 4 to 10 ML. In matrix samples A, the experimental values can be well-expressed by the linear relation with the thickness-independent  $K_{u,I}^{(1)}$  and the bulk-like  $K_{u,V}^{(1)}$ , as  $K_u^{(1)}t_{Co} = K_{u,I}^{(1)} + K_{u,V}^{(1)}t_{Co}$ . From this linearity, we can determine the value of interface PMA from the intercept on the vertical axis. The gradient of this linearity corresponds to the volume PMA. Also, a broken

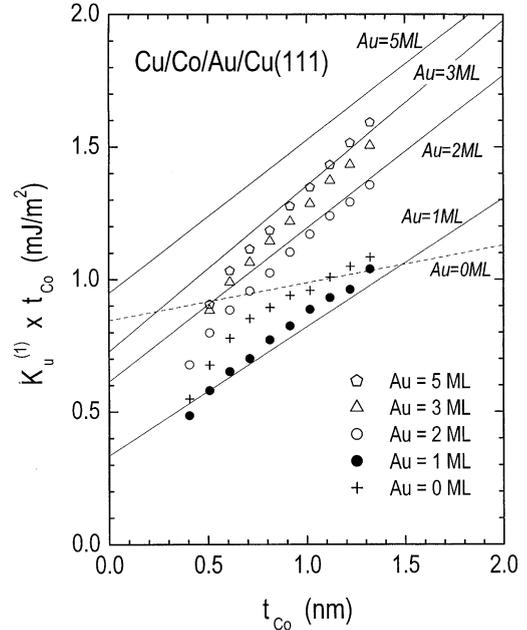


Fig. 2. Plot of  $K_u^{(1)}t_{Co}$  vs  $t_{Co}$  in the Co-thickness region thinner than 7 ML (1.4 nm) with various Au-interlayer thicknesses in matrix samples B. 1 ML of Co corresponds to 0.204 nm. Solid lines are least-squares fits obtained for other matrix samples A as used in Fig. 1, with  $1\text{ nm} \leq t_{Co} \leq 2\text{ nm}$  (see text). A broken line is also a least-squares fit for Co/Cu in the same samples A.

line is a result of the same calculation for the Co/Cu structure in the same matrix samples A. With only 1 ML of the Au-interlayer, experimental values in the new matrix samples B obey the linear relation obtained from the thicker Co films in matrix samples A. It indicates that both the constant  $K_{u,I}^{(1)}$  and the bulk-like  $K_{u,V}^{(1)}$  are maintained down to 2 ML of Co (0.4 nm). However, in this thinner film region, discrepancies are observed between experimental values and the linear relations for the thicker Au-interlayers. As can be seen, the discrepancy starts at 5 ML of Co (1.0 nm) with 2 ML of the Au-interlayer. The discrepancy is more apparent for the Co films with 3 or 5 ML of Au, where the misfit strain is larger. Our result shows degradations of the PMA in these thinner Co films with Au-interlayers above 2 ML, since the linear relation represents constant values of volume and interface anisotropy constants. Also, the discrepancy becomes significant in the Co/Cu

system, although the lattice constants of Co and Cu are fairly close. These phenomena cannot be explained by the misfit-strain-induced magneto-elastic effect as discussed above. We observe the effects of structural inhomogeneities, such as atomic-scale interface roughness, defects, and local strain, on the field dependence of the spin-wave BLS spectrum width [3]. Such structural inhomogeneities should play an important role in the degradation of the interface anisotropy in these ultrathin films. However, it should be noted that our direct observations of a narrow and sharp spin-wave BLS spectrum and its field dependence assured the existence of a well-defined ferromagnetic ordering in these ultrathin Co films even with 2 ML.

The PMA constants are also affected by an overlayer material like Au, which indicates significant deformation of the Co lattice structure after forming the overlayer with a larger lattice constant than Co. To understand this phenomenon, the Au-overlayer thickness dependence of the anisotropy is examined in Cu/Au(x)/Co(y)/Au(1 ML)/Cu(111) matrix samples C, as shown in Fig. 3. We observe a linear relation for each Au-capping layer, indicating the existence of a constant interface anisotropy such as  $K_u^{(1)}t_{Co} = K_{u,I}^{(1)} + K_{u,V}^{(1)}t_{Co}$ . As can be seen,  $K_{u,I}^{(1)}$  increases as a factor of 3.5 with increasing Au-capping layer thickness up to 3 ML and then saturates, while  $K_{u,V}^{(1)}$  is constant with a bulk value.  $K_{u,I}^{(1)}$  at one Co/Cu interface is  $\frac{0.84}{2} = 0.42 \text{ mJ/m}^2$ , as shown in Fig. 1. Using this value,  $K_{u,I}^{(Co/Au=1 \text{ ML})}$  with 1 ML of the Au-interlayer is also obtained as  $0.31 - 0.42 = -0.11 \text{ mJ/m}^2$  in Fig. 1, since the interface anisotropy constant is a sum of  $K_{u, \text{lower-interface}}^{(Co/Au=1 \text{ ML})}$  and  $K_{u, \text{upper-interface}}^{(Cu/Co)}$ . Therefore, we can determine the interface anisotropy constant for the upper interface in these matrix samples C. Here, the magneto-elastic effects for  $K_{u,I}^{(Co/Cu)}$  is negligible for the Co/Cu interface because of the near-zero value of  $a$ . It means that physical origins of  $K_{u,I}^{(Co/Cu)}$  are of the Neel-type interface anisotropy due to symmetry-breaking of the lattice and modifications of the electronic structure at the interface [2]. The experimental values of  $K_{u, \text{upper-interface}}^{(Au/Co)}$  for the upper interfaces with Au-overlayers are larger than  $K_{u, \text{upper-interface}}^{(Cu/Co)}$ . Therefore, the increase in the

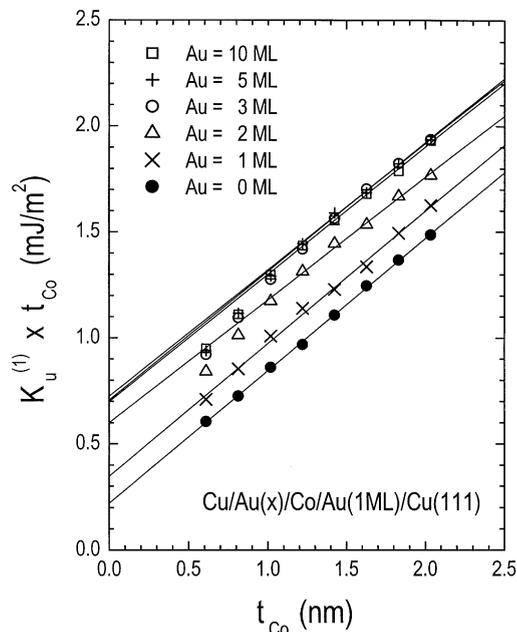


Fig. 3. Plot of  $K_u^{(1)}t_{Co}$  vs  $t_{Co}$  in matrix samples C of Cu/Au-overlayer/Co/Au(1 ML)/Cu(111) with various thicknesses of Au-overlayers. Solid lines are least-squares fits for  $t_{Co} \geq 1 \text{ nm}$ . The intercept on the vertical axis indicates a total interface PMA constant for both lower and upper interfaces. The gradient of the linearity corresponds to the volume PMA constant.

interface PMA with increasing Au-overlayer thickness can be attributed to the additional local strain and misfit dislocation of Co near the upper interface after forming the Au layers, although the good epitaxial growth was continuously observed in the Au-overlayer by means of RHEED. The lattice constant of the Au-overlayer changes from that of Co to that of bulk Au with only 1 ML of the deposited Au-overlayer. Therefore, the film coverage of 1 ML-thick Au-overlayer is enough to show such bulk-like lattice structure of Au. Our result demonstrates that such local strain and misfit dislocations occur in Co with atomic-scale thicknesses of the Au-overlayer of 1 ML and then saturate with 3 ML. The interface anisotropy measurement can clarify such an additional deformation of the lattice near the interface and its thickness dependence with sufficient sensitivity in magnetic ultrathin films after forming overlayers, as shown here.

#### 4. Conclusions

In summary, we study the relation between misfit strain and magnetic anisotropies in ultrathin Co single films hetero-epitaxially grown on Au/Cu/Si(111). The interface uniaxial perpendicular anisotropy with Cu-overlayers is proportional to the misfit strain, due to the magneto-elastic effect. We show the Co-thickness dependence of the anisotropies in quasi-monatomic film region down to a few ML. The experimental values of the anisotropy constant are smaller than the calculated ones, while assuming constant interface and volume anisotropies, when the misfit strain is larger. In addition, the interface anisotropy is markedly affected by Au-overlayers, which have larger lattice constants than Co. The interface anisotropy constant increases with increasing Au-capping layer thickness up to 3ML and then saturates.

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