Exchange bias can be observed at an interface between ferromagnetic (FM) and antiferromagnetic (AF) materials under a field-cooled condition [1–3]. It is a result of interfacial magnetic coupling between FM and AF layers where the direction of magnetic anisotropy is shared by the setting. While the sample is cooled between the Néel temperature and Curie temperature with a presence of an applied magnetic field, the interfacial AF spins are coupled to the FM spins. During the FM magnetisation reversal, the AF layer spins do not follow the applied magnetic field, resulting in a horizontal shift in a magnetisation loop [4]. To increase the interfacial exchange bias, thin FM/AF layers with their crystalline matching are favourable. Thin layers can maximise the interfacial coupling and are more appropriate for device implementation [5]. The exchange bias has been used to form a magnetically pinned layer in devices such as magnetic sensors and read heads for hard disk drives.

There have been very few reports on exchange bias behaviour of Co-based Heusler alloys, which is a half-metallic ferromagnet [6] since there are several critical parameters to be controlled such as lattice mismatch, diffusion across the interface and impurities. The largest value of exchange bias at room temperature (RT) reported for Heusler alloy sample was 2520 Oe [7] with a Ni-based polycrystalline bulk (superparamagnetic Ni2Mn(Al,Si)) 250 Oe with a Co-based polycrystalline film (Co2FeAl0.5Si0.5/IrMn) [8]. These works are all based on the conventional exchange coupling induced at the FM/AF interfaces. However, according to Culbert et al [9], a weak AF Cr ultrathin layer attached onto a Heusler alloy layer has been theoretically predicted not to affect its half-metallicity but to introduce the exchange coupling due to its interfacial strain. Cr is selected because it has a good epitaxial relationship with a conventional Heusler alloy, e.g. Co2FeAl0.5Si0.5 (CFAS). Epitaxial CFAS/Cr/CFAS tri-layers have then been...
grown by Furubayashi et al [10], however they only show negligible exchange bias. This discrepancy between the theory and experiment may be caused because defects can be induced inevitably during the growth due to the lattice mismatch between the FM and AF layers (1.4%) [11, 12].

In this study, the uniaxial anisotropy is intentionally introduced in a Heusler alloy/Cr interface to enhance the exchange bias. The effect of crystalline strain on the exchange bias at the CFAS/Cr interface is investigated using repeated CFAS/Cr stacks by three times. Epitaxial growth and precise magnetic measurements with accurate in-place magnetic field control allow systematic study on the interfacial exchange bias and the induced lattice strain.

Samples with a stack structure of Cr (3)/Ag (30)/(CFAS (t CFAS)/Cr (t Cr))/Au (3) (nm), where CFAS is the FM layer and Cr is the AF layer, were grown on a single crystal MgO(001) substrate by ultrahigh vacuum (UHV) molecular beam epitaxy (MBE) at RT. The base pressure of the MBE system was $1.2 \times 10^{-8}$ Pa and the pressure during the deposition was between $4.5 \times 10^{-8}$ and $6.8 \times 10^{-7}$ Pa. The deposition rate of these layers was controlled by adjusting the electron-beam current and was maintained to be at $0.01 \text{nm s}^{-1}$. The MgO substrate was pre-cleaned by ultra-sonication in acetone, isopropyl alcohol (IPA) and deionised water for 5 min each, followed by pre-annealing at 600 °C for 1 h in the UHV chamber. The Cr/Ag seed layer was used to improve the crystallinity of CFAS as previously reported in similar films [13]. A series of samples were produced by varying the CFAS thickness to be $1 \text{nm} \leq t_{\text{CFAS}} \leq 4 \text{nm}$ and the Cr thickness to be $0.3 \text{nm} \leq t_{\text{Cr}} \leq 1.2 \text{nm}$. These thicknesses were much smaller than the previous study on a similar system as reported in reference [10] and with more CFAS/Cr interfaces by repeating these layers by three times. No annealing was performed during and after the deposition to avoid inter-layer-diffusion of the Cr layer.

The crystalline structures of the samples were analysed by x-ray diffraction (XRD) at the photon factory (PF) in the High Energy Acceleration Organization (KEK). ADE Model 10 vibrating sample magnetometer (VSM) was used to study magnetic properties of the samples, which were cut into a circular shape to remove any shape anisotropy, measured at RT and 193 K. Here, the Curie temperature of bulk CoFeAl, which has similar magnetic properties with CFAS has been reported to be 1170 K [14] and the Néel temperature of bulk Cr has been reported to be 311 K [15], both of which are above the measurement temperatures in this study. The exchange bias was measured by following the York Protocol [3] with the setting temperature of 498 K and the cooling field of 2 kOe. X-ray circular dichroism (XMCD) measurements were also performed at the PF in the KEK by applying a magnetic field of 120 kOe perpendicular to the film.

Reflection high-energy electron diffraction (RHEED) images for analysing surface structures were obtained during the MBE growth. Images were taken before and after the growth of each layer as respectively shown in figure 1. The RHEED pattern from the MgO substrate has relatively large

Figure 1. Schematic multilayered structure and the associated RHEED patterns taken (a) after annealing the MgO(001) substrate at 600 °C for 1 h and after the deposition of (b) Cr, (c) Ag, (d) CFAS and (e) Au layers at RT.

Figure 2. XRD pattern of the ($t_{\text{CFAS}} = 3 \text{nm}/t_{\text{Cr}} = 0.9 \text{nm}$) sample. The magnified image in the vicinity of 30–33° is also shown.
spots, indicating that the surface of the substrate possess long-range roughness after annealing. Such roughness can be removed by depositing the Cr and Ag buffer layers as confirmed by the streak RHEED patterns. The pattern after the Ag deposition at RT clearly shows a formation of the face-centred cubic (fcc) (001) surface observed, confirming the epitaxial relationship of MgO(001)[1 1 0]/Ag(001)[1 1 0] with 3.1% lattice mismatch as reported previously [16]. The CFAS deposition is found to make the surface to be slightly rough as shown by the increase in the width of the streaks. This is due to initial island-like growth of the CFAS layer as similarly shown by the increase in the width of the streaks. The pattern clearly shows strong spots from the MgO [1 1 0] axis and the smallest c-axis mismatch [19]. For the CFAS/Ag sample, the uniaxial anisotropy is found to be the maximum at almost 30° from the MgO[1 1 0] axis. The origin of the minor tilt is not clear at this stage but it may be induced by the interfacial strain. As the thickness of the Cr layer increases above 3 nm, the uniaxial anisotropy changes its orientation along the MgO[1 1 0] axis above five monolayers (~3 nm) of the CFAS deposition as previously reported. Figure 4(b) shows the corresponding uniaxial anisotropy at ~30° induced by the Cr layer is interfacial sensitive and can be relaxed along the MgO[1 1 0] axis above five monolayers (~3 nm) of the CFAS deposition as previously reported.

Figure 3. Magnetisation curves of the (a) \( t_{\text{CFAS}} = 2 \text{ nm} \) and the Cr layer was first optimised to induce the maximum change onto the magnetisation curves of the CFSA/Cr samples. Figure 3(a) shows the magnetisation curves with \( t_{\text{CFAS}} = 2 \text{ nm} \) and \( t_{\text{Cr}} = 0.9 \text{ nm} \) measured at RT and (b) \( t_{\text{CFAS}}/t_{\text{Cr}} = 0.9 \) with 1 nm \( \leq t_{\text{CFAS}} \leq 4 \text{ nm} \) along the MgO[1 1 0] axis.
Figure 4. (a) Coercivity polar plot for the four samples with $1 \, \text{nm} \leq t_{\text{CFAS}} \leq 4 \, \text{nm}$ with $t_{\text{Cr}} = 0.9 \, \text{nm}$. The corresponding remanence polar plot is also shown for (b) the sample with $t_{\text{CFAS}} = 2 \, \text{nm}$ and (c) the 2 nm thick single CFAS film grown on the MgO substrate. (d) Representative magnetisation curves for the $t_{\text{CFAS}} = 2 \, \text{nm}$ sample.

Figure 5. XMCD profiles for (a) Co and (b) Fe atoms in the sample with $t_{\text{CFAS}} = 2 \, \text{nm}$ and $t_{\text{CFAS}} = 0.9 \, \text{nm}$. Red and blue lines represent the magnetic field applications at $0^\circ$ and $55^\circ$ from the plane normal, respectively.

The remanence of the sample with $t_{\text{CFAS}} = 2 \, \text{nm}$ shows a loop shift of 18 Oe as shown in figure 4(d). This is five times larger than the previous reported value [9]. This proves that the exchange bias can be induced even by a weak AF layer by introducing additional interfacial strain in an epitaxial FM/AF structure.
In order to confirm the orbital and spin moments of the CFAS layer, XMCD measurements were also performed. As shown in figure 5(a), the circularly polarised x-ray was introduced at tilted angles of both 0° and 55° from the plane normal. The XMCD results show a spin moment ($m_{\text{spin}}$) of (0.847–0.849) $\mu_B$/atom and orbital moment ($m_{\text{orb}}$) of (0.078–0.085) $\mu_B$/atom for Co. These values are about 15% smaller as compared with the previously reported value in a similar Heusler alloy Co$_2$FeAl [20]. This reduction may be caused by possible atomic disorder at the CFAS/Cr interfaces as discussed above. 

For Fe, $m_{\text{spin}}$ and $m_{\text{orb}}$ are measured to be (1.095–1.161) $\mu_B$/atom and (0.154–0.1610) $\mu_B$/atom, respectively. These values agree very well with the previous report [20]. These results further confirm indirectly that the sample maintains its half-metallicity as theoretically predicted [21], indicating the increase in the coercivity and exchange bias is due to the strain-induced coupling at the CFAS/Cr interfaces. Therefore, the exchange coupling between the Heusler alloy films and antiferromagnetic Cr films can be controlled by tuning the lattice matching without sacrificing their half-metallicity.

In summary, we have measured a loop shift of 18 Oe at 193 K in the [Co$_2$FeAl$_{0.5}$Si$_{0.5}$ (2 nm)/Cr (0.9 nm)]$_3$ samples due to the lattice mismatch at the interface. This value is five times larger than the previously reported value [10]. The spin and orbital moments of the samples are found to be similar to those in the Co$_2$FeAl single layer. These findings agree with the theoretical prediction [9]. For device applications a larger loop shift is required but such a strained system can offer an effective method to increase the exchange bias. Such a system may reveal insights of interfacial exchange coupling without sacrificing the half-metallicity of the Heusler alloys and may increase the design flexibility for spintronic devices.

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