Iron is a critical and interesting element in fundamental research of magnetism because it is located between antiferromagnetic Mn and ferromagnetic Co on the Bethe–Slater curve. It has a high magnetic moment of 2.18 \( \mu_B/\text{atom} \) and Ni (\( \sim 0.57 \mu_B/\text{atom} \)). Iron exhibits different crystalline structures, e.g., bcc, fcc and hcp structures. Experimental results show that epitaxial growth between an ultrathin film and a substrate could create and control different metastable structures, e.g., bcc-Fe/Ag(001), fcc-Fe/Au(001), fcc–bcc Fe/Cu(001) and fcc–bcc Fe/Cu3Au(001). In those cases, ultrathin Fe films were deposited on fcc substrates at a rotation of 45° in the \( ab \) plane to match the epitaxial condition. These systems exhibit a thickness-driven perpendicular to in-plane spin reorientation transition (SRT). In Fe/Cu3Au(001) and Fe/Cu(001) systems, the SRT transition region is almost the same as the fcc to bcc(011) crystalline transition. Thus the SRT in those systems is likely the result of structural transformation. In the Fe/Ag(001) case, the SRT transition region appears at 6–7 monolayers (ML) at room temperature with a stable bcc(001) structure. In this case, SRT is caused by the competition between the surface anisotropy and the shape anisotropy. For the surface anisotropy, the interfacial condition between the thin film and the substrate also plays an important role. The interfacial conditions, e.g., surface reconstruction and roughness, are crucial considerations for SRT studies.

The lattice constant of bcc-Fe is 2.87 Å, as shown in Fig. 1(a). The bcc-NiAl(001) substrate with a lattice constant of 2.89 Å is also suitable for epitaxial growth. Ultrathin Fe films on NiAl(001) are weakly strained because of low lattice mismatch between bcc-Fe and NiAl(001) of around 1%, as shown in Fig. 1(b). Moreover, the NiAl(001) surface reveals a special reconstructed surface of \( \sqrt{2} \times \sqrt{2} \) R45° after annealing treatment exceeding 800 K. The top layer of NiAl(001) is determined by the annealing temperature, e.g., Al termination at an annealing temperature of 800 to 1400 K and Ni termination at an annealing temperature exceeding 1400 K.

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**Fig. 1.** (a) Crystalline structure of bcc-Fe (\( a = b = c = 2.87 \) Å). (b) Illustrations of epitaxially grown Fe on bcc-NiAl(001). The ocean blue circles indicate atoms on the top surface of unreconstructed NiAl(001). The red circles represent the cubic cells of bcc-Fe.
modulation and a lock-in amplifier were utilized on MOKE to investigate the hysteresis loops of the ultrathin films.

The thickness calibration and growth mode were measured using MEED oscillations. Figure 2 shows that the growth mode of Fe on NiAl(001) proceeds via a layer-by-layer model up to 7 ML. MEED intensity reached the first maximum peak after 1/3 of the layer-by-layer oscillation period. This might be because of the vacancy sites on the surface of NiAl(001). After sputtering and annealing, the LEED patterns revealed not only the p(1×1) spots but also the (c\sqrt{2} \times 3\sqrt{2})R45° superstructure on a clean NiAl(001) surface, as shown in Fig. 3(a). Such a reconstructed surface is not fully covered by aluminum and still has 1/3 vacancy sites on the top layer. After Fe was deposited on NiAl(001), it still revealed only p(1×1) spots, as seen in Figs. 3(b) and 3(c). No obvious difference appears as the Fe thickness increases. This result also indicates that the in-plane lattice constant of Fe films is almost the same as that of NiAl(001) substrate (2.89 Å). The interlayer distance of ultrathin films was obtained by calculating the kinetic approximation of LEED-I(E).\(^{14,15}\) The interlayer distance maintains almost the same value (1.43–1.44 Å in the Fe thickness range of 2.4–15 ML). These results imply that the crystalline structure of an ultrathin Fe film on NiAl(001) is close to bcc without significant expansion or compression.

The saturated magnetic signal of the films in the polar and the longitudinal (along the [110] direction) MOKE measurement is plotted as a function of the thickness in Fig. 4(a). The coercive field in polar (circle) and longitudinal (triangular and square) geometries. (c) Hysteresis loop of 3.0 ML Fe on NiAl(001) in polar geometry. (d) Hysteresis loop of 6.4 ML Fe on NiAl(001) in longitudinal geometry.
magnetic behavior. Furthermore, to investigate the temperature effect on the SRT behavior, we performed MOKE measurements at 110 K, according to which the SRT transition region was found not to vary significantly from the results at room temperature.

The easy axis of a magnetic thin film is determined by the effective magnetic anisotropy.\(^\text{[10]}\) The magnetic anisotropy usually consists of a series of terms of different physical origin. Examples include magneto-crystalline anisotropy, shape anisotropy, magneto-elastic anisotropy, and surface anisotropy. To assay this system, we use a phenomenological model\(^\text{[16]}\) to describe the thickness-driven SRT behavior. In this model the effective anisotropy \(K_{\text{eff}}\) is given by

\[
K_{\text{eff}} = \frac{K_a}{I} + K_s = \frac{K_a}{I} + (K_{\text{me}} + K_{\text{shape}} + K_{\text{crystal}}),
\]

(1)

where \(K_a\) is the surface anisotropy, which combines contributions from the thin film/substrate interface and the vacuum/thin film interface, and \(K_s\) is the volume contribution of anisotropy, which includes \(K_{\text{me}}, K_{\text{shape}},\) and \(K_{\text{crystal}}. K_{\text{me}}\) is the magneto-elastic anisotropy, which is related to the strain in the crystalline structure of thin films. The strain originates from the mismatch between the ultrathin film and the substrate. \(K_{\text{crystal}}\) is the magneto-crystalline anisotropy, which is related to the spin–orbit coupling in various crystalline structures. \(K_{\text{shape}}\) is the shape anisotropy, which is related to the demagnetization field induced by the geometrical shape of the sample. To clarify the reason for the SRT, we assayed several kinds of magnetic anisotropy in order. First, according to the LEED and LEED-\(I(E)\) results, the crystalline structure of Fe/NiAl(001) maintains a bcc structure from a thickness between 2.4 and 15.0 ML. The \(c/a\) ratio of bcc-Fe is in a small variation region and very close to 1. Therefore, the SRT is not induced by the structural transition or the strain in the crystalline structure. According to previous studies, the bulk magneto-crystalline anisotropy of bcc-Fe is small and approximately 3.62 \(\mu\text{eV/atom} = 4.8 \times 10^{-2} \text{MJ/m}^3\).\(^\text{[17]}\) The magneto-elastic anisotropy and the bulk magneto-crystalline anisotropy are very small compared with the shape anisotropy and can be neglected. Consequently, the shape anisotropy and the surface anisotropy are the principal factors causing magnetic anisotropy and, as evaluated, the competition between them controls the easy axis in this system. The shape anisotropy of bcc-Fe is \(-132 \mu\text{eV/atom}\) from \(M_{\text{Fe}} = 2.18 \mu\text{B/atom}\)\(^\text{[18]}\) and \(K_{\text{shape}} = -2\pi M^2\). The SRT appears in the transition region at which the effective anisotropy starts to change sign from positive to negative. Accordingly, the following formula can be obtained:

\[
K_s = -\frac{t_s}{2} \times K_v \simeq -\frac{t_s}{2} \times K_{\text{shape}}.
\]

(2)

Based on Eq. (2), the surface anisotropy in the Fe/NiAl(001) system is deduced to be 267.3 \(\mu\text{eV/atom}\). For the purpose of comparison, the surface anisotropies, derived from the critical thickness according to Eq. (2), of Fe/Ag(001) and Fe/Au(001) are listed in Table I, together with a theoretical value of free-standing Fe film. It is clear that the Fe/NiAl(001) system has a smaller surface anisotropy than the Fe/Ag(001) system. Because the surface anisotropy is related to the interface between the thin film and the substrate, a possible explanation for this result is surface reconstruction of NiAl(001). The NiAl(001) surface is not fully covered by Al and still has 1/3 vacancy sites. This partial covering might reduce the strength of the surface anisotropy. Thus, the SRT transition region is apparently reduced compared with that of the Fe/Ag(001) system.

In addition to the SRT phenomenon, the system exhibits soft perpendicular magnetization. In the thickness range of 2.4–3.7 ML, the coercive field of bcc-Fe with a perpendicular easy axis is very small \((H_c \approx 1 \text{ Oe})\), as already shown in Fig. 4(d). Such soft magnetic behavior is similar to that of Permalloy (NiFe), but the latter is observed only in the in-plane magnetization. The AES characterization shows no significant NiFe alloy formation at the interface. According to LEED-\(I(E)\) measurement, the interlayer distance of 2.7 ML Fe is approximately 1.44 Å, which is far below the interlayer distance of NiFe \((1.77 \text{ Å})\),\(^\text{[20]}\) indicating that Fe–Ni alloy formation at the Fe/NiAl interface is negligible. Consequently, both LEED-\(I(E)\) and AES measurements can exclude the possibility of Fe–Ni alloy formation. Therefore, such soft perpendicular magnetization is most likely due to the intrinsic properties of the Fe ultrathin films on the NiAl(001) surface.

In summary, ultrathin bcc-Fe on NiAl(001) exhibits soft perpendicular magnetic behavior in the low thickness range. The coercive field is approximately 1 Oe and obviously smaller than that of other similar systems, e.g., Fe/Ag(001) and Fe/Au(001). Furthermore, the SRT critical thickness in this system is around 4 ML. In the transition region, the ultrathin film sustains abcc structure with low strain. According to our calculation based on a phenomenological model, the surface anisotropy of the Fe/NiAl(001) system is 267.3 \(\mu\text{eV/atom}\). We suppose that such a small surface anisotropy in the Fe/NiAl(001) system could be attributed to the \(c\sqrt{2} / \sqrt{3} \times 45^\circ\)R45° superstructure of NiAl(001).

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<table>
<thead>
<tr>
<th>System</th>
<th>SRT region (ML)</th>
<th>(H_c) (Oe)</th>
<th>(K_s) ((\mu\text{eV/atom}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Free-standing bcc Fe</td>
<td></td>
<td></td>
<td>594.4(^\text{[14,15]})</td>
</tr>
<tr>
<td>bcc-Fe/Ag(001)</td>
<td>6.0–7.0(^\text{2)})</td>
<td>60–80 (20(^\text{3)})</td>
<td>429.0 ± 33.0</td>
</tr>
<tr>
<td>bcc-Fe/Au(001)</td>
<td>2.0–2.3(^\text{2)})</td>
<td>5–10 (20(^\text{3)})</td>
<td>141.9 ± 9.9</td>
</tr>
<tr>
<td>bcc-Fe/NiAl(001)</td>
<td>3.7–4.4</td>
<td>1 ± 0.5 (20(^\text{3)})</td>
<td>267.3 ± 23.1</td>
</tr>
</tbody>
</table>


