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In-plane magnetic anisotropy of bcc Co on GaAs(001)

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Epitaxial growth of Co on GaAs(001) and its in-plane magnetic anisotropy are studied using reflection high-energy electron diffraction, a high-resolution transmission electron microscope, and the magneto-optical Kerr effect. In the initial and final stages of growth, Co exists in single-crystalline body-centered-cubic (bcc) and hexagonal-closed-packed (hcp) phases, respectively, while in the middle stage the coexistence of the bcc and hcp structures is observed. For the bcc Co thin films on GaAs(001), a fourfold in-plane magnetic anisotropy with easy axes along the ⟨100⟩ directions is realized and discussed. [S0163-1829(98)04915-7]

The $3d$ transition metals exist in a variety of crystallographic and magnetic phases. Thin-film growth of these materials on crystalline substrates allows the forces present at the interface to drive the film into specific crystalline structures. These structures may be in a thermodynamically stable phase, a known high-pressure or high-temperature phase, or even a phase not previously observed. They greatly increase the variety of magnetic materials by essentially making ''new'' materials from ''old'' elements.[1]

The epitaxial growth of Co films serves as a good example. It is known that the hexagonal-close-packed (hcp) and face-centered-cubic (fcc) structures are, respectively, stable and metastable phases of Co. The body-centered-cubic (bcc) structure, which does not occur in nature, was realized by Prinz with epitaxial growth on a GaAs(110) substrate.[2] However, it was later pointed out by Liu and Singh that bcc Co is not a true metastable phase but a force-induced phase.[3] The in-plane magnetic anisotropy of such a bcc Co thin film on GaAs(110) was further determined and a negative value for the cubic anisotropy constant K_1 was proposed.[2] If this were true, a fourfold in-plane magnetic anisotropy with easy axes along the ⟨110⟩ direction would then be expected in the bcc Co films on GaAs(001) substrates. In fact, a fourfold in-plane magnetic anisotropy with the easy axes along the ⟨100⟩ rather than the ⟨110⟩ direction was observed by Blundell *et al.*[4] Interestingly, it was later argued by Gu *et al.* that Co films grown on GaAs(001) were actually not body-centered cubic but two-domain hexagonal close packed by which the fourfold magnetic anisotropy along the ⟨100⟩ direction could be explained by such a microstructure.[5] Obviously, the epitaxial structure of Co on GaAs(001) and its magnetic anisotropy are still very controversial. In this work, we present a clear picture of the epitaxial growth of Co on GaAs(001), which clears up the previous controversy about the structure of Co thin films on GaAs(001). With the help of this clear picture, we prove that the bcc Co films on GaAs(001) show a fourfold in-plane magnetic anisotropy.

Co films were grown in a molecular-beam epitaxy (MBE) growth chamber connected with the VG-ESCALAB-5 electron spectrometer system. The Te-doped GaAs(001) single-crystal wafers were polished and treated by ordinary device cleaning process. The final substrate cleaning was performed using two different procedures. One is our routinely used method, i.e., argon ion bombardment followed by annealing.[6] The other is a chemical etching method, using $H_2SO_4H_2O_2$:H_2O=5:1:1 before loading into the MBE system and flashing to 580 °C in the ultrahigh vacuum chamber. We found in our experiment that the latter procedure provides better surface morphology for the clean GaAs(001) surfaces. For clean GaAs(001) substrates before epitaxy, 4×1 reconstruction patterns were observed by reflection high-energy electron diffraction (RHEED), while 4×1 with streaks reconstruction patterns (sometimes called 4×2 reconstruction) were seen by low-energy electron diffraction. The Auger spectra show that the surfaces are free of carbon and oxygen contamination. The background pressure in the growth chamber was 3×10^{-8} Pa. The 99.99% pure Co pieces were charged in an Al_2O_3 crucible of a Knusden cell. The deposition rate used in this experiment was fixed at 0.2 nm/min as measured by a quartz thickness monitor. The pressure during growth was lower than 1×10^{-7} Pa.

Figure 1(a) shows a typical RHEED pattern for a clean GaAs(001) surface, with an electron beam along the [110] direction. Following the evolution of the RHEED pattern as a function of Co thickness, we first find that a new pattern starts to appear at ~0.2 nm as the substrate spots fade away. This new diffraction pattern becomes completely dominant at ~0.8 nm. A typical diffraction pattern at this stage of Co growth is shown in Fig. 1(d). It is obvious from such a well-ordered pattern that the Co overlayer grows in a single-crystalline structure on the GaAs(001) substrate at this stage. The same pattern was also found by rotating the electron beam 90° to the [110] direction, which indicates that the Co thin film has a fourfold in-plane symmetry. Using the procedure proposed earlier,[6] we can easily calculate the corresponding diffraction pattern as shown in Fig. 1(g) if we assume that the Co epilayer exists in a bcc structure. From the good agreement between the experimental and calculated patterns [Figs. 1(d) and 1(g)], we can immediately tell that Co grows in a bcc structure at this stage. On the other hand,

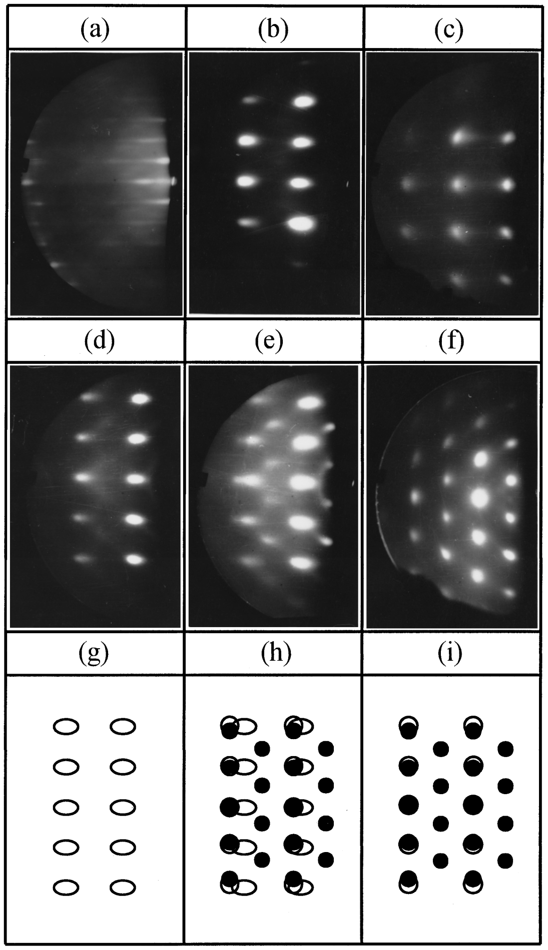

FIG. 1. RHEED patterns for different thin films grown on the GaAs(001) substrate, taken with the incident electron beam along the substrate [110] direction: (a) clean GaAs(001), (b) 5.6-nm bcc Fe, (c) 8.0-nm fcc Mn, (d) 1.5-nm bcc Co, (e) 3.5-nm bcc and hcp Co, (f) 15-nm hcp Co, and (g)–(i) calculated diffraction patterns corresponding to (d)–(f), respectively. ○ and ● correspond to the two hcp domains mentioned in the text.

a more straightforward way to determine the epitaxial structure is to try to compare the observed diffraction patterns with those of known structures. Here we present in Figs. 1(b) and 1(c) the diffraction patterns of the bcc Fe[7] and fcc Mn[6] that were epitaxially grown on GaAs(001). It is easy to see the different shapes of these two patterns: The fcc phase has a squarelike pattern and the bcc phase has a rectanglelike pattern. Therefore, from the close similarity between Figs. 1(b) and 1(d). It is confirmed again that the Co overlayer on GaAs(001) at the early stage of growth is indeed a bcc structure, with the epitaxial relationships (001)[001]Co//(001)[001]GaAs.

Increasing the Co thickness beyond ∼2 nm some other extra spots appear and are superposed on the bcc Co diffraction pattern. It might seem at first glance that polycrystalline growth is starting to develop at this stage, but this is actually a transition stage where a hcp structure grows on top of the bcc structure. Figure 1(e) shows a typical RHEED pattern in this transition stage. Again, the main feature of this experimentally observed pattern is predicted in the calculated result shown in Fig. 1(h), assuming the coexistence of bcc and hcp Co phases where the epitaxial relationships of hcp Co on GaAs(001) are those proposed earlier,[5] i.e., (1210)[0001]Co//(001)[110]GaAs and (1210)[0001]Co//(001)[110]GaAs.

When the Co thickness goes beyond ∼6 nm, the RHEED pattern is completely dominated by the hcp structure, as shown in Fig. 1(f). Here the sixfold symmetry can be clearly seen. This very-well-defined sixfold pattern gives the initial impression that we might have a single-crystal hcp Co structure with (1210)[0001]Co//(001)[110]GaAs. If this were true, then we would expect to see a different diffraction pattern after rotating the electron beam 90° to the [110] direction. Surprisingly, we see exactly the same pattern upon performing the experiment. This implies that the single-crystal-like sixfold diffraction pattern might actually come from the two perpendicularly oriented hcp Co domains proposed earlier,[5] with [0001]Co//[110]GaAs and [0001]Co//[110]GaAs together. We show the calculated diffraction pattern in Fig. 1(i) assuming such a two-domain structure. Strictly speaking, one might argue that the comparison to Fig. 1(f) is not fully satisfied since one does not see in experiment the expected two sets of split spots. On the other hand, as expected from Fig. 1(i), we do observe that the intensity of the second and fourth columns from right to left are much higher than what they should be. Presumably, the lack of spot splitting in the RHEED pattern is caused by the fact that the splitting is too small if the sizes of the hcp Co clusters are not large enough to produce sharper diffraction patterns. The strong evidence for such a two-domain hcp structure comes from the top-view diffraction pattern viewed using the transmission electron microscope. Figures 2(a) and 2(b) show such a diffraction pattern together with the calculated results assuming the two-domain hcp Co structure. It is immediately concluded that the last stage of Co growth is indeed in the two-domain hcp phase.

The controversy about the epitaxial growth of Co on GaAs(001) can now be understood. Blundell *et al.* declared, using their RHEED measurement, that Co grew in the bcc structure during the initial growth stage. However, they did not compare their patterns with any theoretical calculations or with the patterns of known structures such as bcc Fe. Their conclusion was therefore questioned by Gu *et al.*[5] Here we have unambiguously confirmed that Co grown on GaAs(001) is in the bcc phase during the early stage of growth. This clears up the previous controversy. We must point out that the TEM measurement happens to be insensitive for detecting the bcc Co phase. This is simply because the lattice parameter of bcc Co is almost exactly half of that of GaAs;[2] this prevents one from seeing the contribution of the bcc Co phase, which is overwhelmed by the strong diffraction spots of the GaAs substrate. However, we have done both *in situ* RHEED and *ex situ* TEM measurements in this work and it is clear that the two-domain hcp Co structure appears only after the growth of bcc Co on GaAs(001). Accordingly, the in-plane magnetic anisotropy of Co on GaAs(001) should be checked again with more careful structure characterization.

With the help of *in situ* RHEED monitoring, we have prepared bcc Co thin films on GaAs(001) by terminating growth just when we observe slight traces of hcp Co. The thickest bcc Co film we can get is about 2.0 nm. A 1.5-nm-

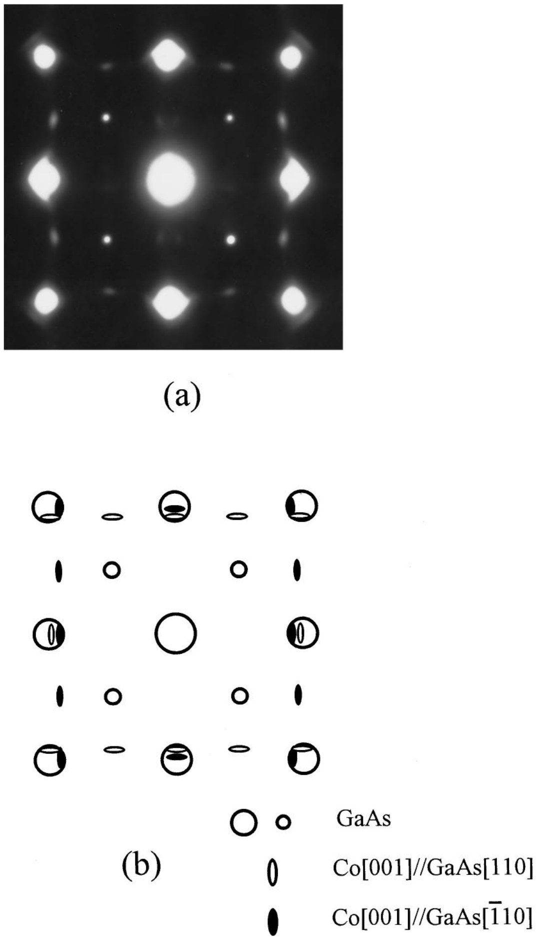

FIG. 2. In-plane TEM diffraction pattern (a) for 29.4-nm hcp Co on GaAs(001) and (b) calculated assuming a two-domain hcp Co structure.

thick Mn cap is then deposited on top of the bcc Co thin film before it is taken out for magnetic measurement. Samples capped with Cu have also been checked and no difference is found in the magnetic measurement. Figure 3 shows a series of hysteresis curves measured using the magneto-optical Kerr effect for a 1.5-nm Mn/1.7-nm bcc Co/GaAs(001) sample. Although the data look noisier than might be expected, fourfold symmetry with easy axes along the ⟨100⟩ directions can still be clearly seen. Blundell *et al.* achieved the same result when observing the bcc Co phase on GaAs(001) as well.[4] Presumably, ours is a real experimental demonstration for the fourfold in-plane magnetic anisotropy of bcc Co on GaAs(001) since we are very sure that no hcp contributions are mixed in, which could also give a similar fourfold anisotropy.[5] In fact, the same fourfold magnetic anisotropy for a 1.5-nm Mn/4-nm Co/GaAs(001) sample is also found in our work; however, we do know that this is a mixed bcc and hcp phase. By the way, the low signal-to-noise ratio in Fig. 3 is caused by the fact that the effective thickness of ferromagnetic Co film is actually less than 1.7 nm because of its reaction with the GaAs substrate, which leads part of the deposited Co film to a nonmagnetic or an antiferromagnetic interface phase. It is also noted that bcc Co thin films thicker than 10 nm on GaAs(001) were declared by some authors,[8,9] but no direct structural data supporting this argument were shown in their papers. On the other hand, both this and other work[4,5] prove with more careful structural characterizations that the bcc Co phase cannot be prepared thicker than 3 nm. In the following, we try to see what the foregoing magnetic result indicates about the bcc Co phase.

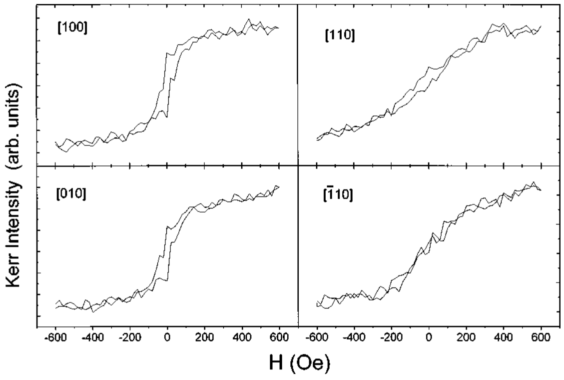

FIG. 3. In-plane magneto-optical Kerr effect measurements for the 1.5-nm Mn/1.7-nm bcc Co/GaAs(001) system relative to the [110] direction of a bcc Co thin film.

The in-plane magnetic anisotropy for bcc Co on GaAs(110) was shown by Prinz: [100] is the easy axis, [111] is the intermediate, and [110] is the hard axis.[2] This result can be explained by neither a positive nor a negative cubic anisotropy constant K_1. For a typical positive K_1, [100] should be the easy axis, [110] the intermediate, and [111] the hard axis. For a typical negative K_1, [111] should be the easy axis, [110] the intermediate, and [100] the hard axis. However, the cubic anisotropy constant K_1 for the bcc $Fe_{1-x}Co_x$ alloy is known to vary from positive to negative with increasing x. The transition happens at $x \sim 0.4$. With this fact in mind, Prinz would rather assume that bcc Co on GaAs(110) has a negative K_1 but a strong uniaxial anisotropy along the [100] direction. In this way, the magnetic anisotropy of bcc Co on GaAs(110) could be understood. However, the result realized here, i.e., the fourfold anisotropy with easy axes along the ⟨100⟩ direction, certainly does not agree with negative K_1. Therefore, the magnetic anisotropy of bcc Co on GaAs(110) and (001) cannot be understood using a unified model. We are now proposing two possible explanations for this. One might argue that the K_1 of the bcc Co phase is actually not negative but positive; this agrees well with direct experimental observations that the ⟨100⟩ direction is the easy axis in both our case of Co on GaAs(100) and Prinz's case of Co on GaAs(110). Meanwhile, the reversal of easy and hard axes of ⟨110⟩ and ⟨111⟩ in Prinz's case was presumably a minor effect caused by some interface effects at bcc Co/GaAs(110). Alternatively, one might argue that the interface effect plays an important role at bcc Co/GaAs(001), which could also produce a reversal of easy and hard axes of ⟨100⟩ and ⟨110⟩ in our case. In fact, such a reversal was indeed found for Fe on GaAs at reduced thickness (<5 nm).[10] Before more direct experimental evidence is obtained, we could say that the mechanism causing the fourfold in-plane magnetic anisotropy of Co on GaAs(001) is still an open question.

In summary, we have clarified in this experiment the controversy about the epitaxial growth of Co on GaAs(001). The in-plane magnetic anisotropy has been determined to be a fourfold symmetry with easy axes along the ⟨100⟩ direction. We believe that the interface effect plays an important role for this magnetic anisotropy, but the mechanism by which it does so is still an open question.

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