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Coherent fcc stacking in epitaxial Co/Cu superlattices

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A series of epitaxial Co/Cu superlattices has been grown on GaAs(110) substrates by molecular-beam epitaxy. Detailed analysis of x-ray diffuse scattering scans along Co($10\bar{1}l$) reveals metastable fcc stacking of Co, with stacking coherence extending across Co layers at thicknesses below 20 Å, while coherent interfaces are obtained at Co thickness up to 40 Å. We discuss possible mechanisms for stabilizing the stacking sequence.

Molecular-beam epitaxy (MBE) techniques have led recently to an upsurge of activity in experimental studies of artificially structured magnetic materials.[1] Under appropriate conditions such growth methods permit the fabrication of single-crystal films in which exceedingly thin magnetic layers can be produced. A related area of interest stimulated by MBE deposition capabilities concerns the growth and properties of coherent *metastable* structures. These are materials which are stabilized epitaxially in phases that have no counterpart in the bulk under ambient thermodynamic conditions. As a relatively new area of exploration, metastable phases are particularly interesting in the context of cooperative magnetic phenomena[2] and the tailoring of magnetic anisotropy by means of epitaxy. In all cases, controlled sample preparation is the key to making reliable measurements which serve as experimental tests of interesting theoretical predictions on low-dimensional magnetic phenomena.[3,4]

Cobalt and copper are interesting candidates for superlattice studies because both elements occur as close-packed crystals in the bulk, with a 2.0% lattice mismatch within close-packed planes (the nearest-neighbor distances of Co and Cu are 2.51 and 2.56 Å, respectively). In addition, the very limited mutual solubility seen in the bulk phase diagram of Co and Cu favors the formation of abrupt interfaces. Here we report our observation of Co/Cu superlattices in which the Co layers are stacked predominantly in the metastable fcc sequence. We distinguish stacking coherence and interfacial coherence and report their dependence on Co layer thickness.

Samples of Co/Cu superlattices were prepared in a Vacuum Generators VG-80 MBE system on annealed GaAs(110) substrates. A growth orientation along close-packed planes was established by the deposition of a buffer layer consisting of 20-Å Co followed by 200-Å Cu. The substrate temperature was 50 °C. We note that there is not a particularly good lattice match between Cu(111) and GaAs(110), which has an in-plane unit mesh of dimensions 5.65×7.99 Å. In fact, our reflection high-energy electron diffraction (RHEED) observations indicate that direct growth of Cu on GaAs(110) tends to produce a disordered film; thus the 20-Å Co layer [which probably grows as (110) bcc (Refs. 2 and 5) Co] is essential as a structural bridge between (110)GaAs and (111)Cu. RHEED observations show that the Cu buffer layer acquires (111) orientation at a thickness of approximately 50 Å, with additional sharpening of RHEED features after growth to 200 Å.[6]

After growing the buffer layer, sequential layers of Co and Cu were deposited. The rate of deposition was 0.5 ± 0.05 Å/sec for Cu (from a Knudsen cell at 1260 °C) and 0.3 ± 0.1 Å/sec for Co (from an electron beam hearth). Co layer thicknesses were established by manually closing the Co shutter at a fixed reading of a quartz-crystal film thickness monitor; thus we are able to control Co layer thicknesses regardless of drifts in the Co deposition rate. Six samples were prepared with Co layer thicknesses of 5, 10, 15, 20, 30, and 40 Å, accurate to ±10%. In each sample the Co layers were alternated with Cu layers of thickness 25 ± 3 Å. The total superlattice thickness was approximately 1500 Å in all cases. Finally, a protective 20-Å Au cap was grown on the top Cu layer of the superlattices. RHEED patterns observed after the growth of the final Cu layer indicated that the films consist of well-oriented close-packed planes.

We note that one cannot distinguish (111) fcc growth from (0001) hcp growth with the RHEED technique. Information complementary to that contained in the RHEED patterns is provided by the map of the in-plane x-ray scattering intensity shown in Fig. 1. As described previously[7] this type of x-ray scan is performed in transmission and in this case the absorption of GaAs necessitates thinning the substrates to approximately 100 μm in order to allow penetration of Mo $K\alpha$ x rays. The x-ray contours of Fig. 1 confirm the orientation and epitaxially ordered nature of the superlattices indicated by the RHEED patterns. The map also determines the epitaxial relationship of the superlattice with respect to the GaAs substrate (inset). Both (111) fcc growth and (0001) hcp growth consist of the stacking of close-packed planes with sixfold rotational symmetry. Thus there are close similarities in the diffraction patterns of lattice-matched fcc and hcp crystals growing in a close-packed orientation. Specifically, the fcc ($2\bar{2}0$) and hcp ($11\bar{2}0$) reciprocal-lattice points will coincide. Thus the peak labeled $h(11\bar{2}0)$ and $c(2\bar{2}0)$ in Fig. 1 arises from a combination of fcc and hcp scattering. We note that in contrast to the Co/Au system,[5] this peak is not split. No evidence of splitting occurs over the entire range of Co thicknesses which we have investigated; we conclude that Co and Cu layers are strained in such a way as to maintain in-plane coherence at Co thicknesses of up to at least 40 Å. As

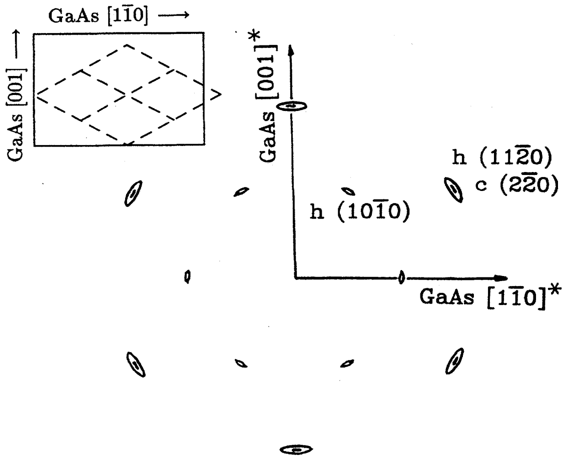

FIG. 1. In-plane scattering intensity of a (40-Å Co)/(25-Å Cu) superlattice. The two intensity contours are drawn at 130 and 65 counts/sec. The background is approximately 60 counts/sec for these scans. GaAs peaks are not included in the figure; the GaAs axes indicate the orientation of the superlattice with respect to the substrate. Inset: epitaxial relationship (in direct space) between the unit cells of the metal superlattice (dashed lines) and the GaAs substrate (solid lines).

shown in Fig. 2, the position of this in-plane peak gradually shifts from that of Cu to that of Co with increasing Co layer thickness, presumably in response to a minimization of elastic strain energy.

The peak in Fig. 1 labeled $h(10\bar{1}0)$ is due to the presence of hcp stacking. There is no fcc reflection at this reciprocal-lattice point; in fcc notation it can be designated $\frac{2}{3}(2\bar{1}\bar{1})$. Whereas we expect a significant contribution to the structure factor from hcp Co, the $h(10\bar{1}0)$ reflection is actually much weaker than one would estimate from a model containing purely hcp Co and fcc Cu layers. We therefore decided to analyze the stacking structure in detail as a function of Co layer thickness.

Whether a close-packed structure stacks according to the fcc $(ABC\ldots)$ sequence or the hcp $(AB\ldots)$ sequence is revealed by x-ray scans along $(10\bar{1}l)$, or by similar scans parallel to c^* subject to the condition that $h-k\neq 3m$, where m is an integer. Diffraction intensities where $h-k=3m$ are unaffected by the hcp-fcc transition.[8] Fcc periodicity is revealed by peaks which occur at $\pm 2\pi/3d_{111}$ Å^{-1} along $(10\bar{1}l)$, while hcp periodicity is revealed by peaks at $\pm 2\pi/2d_{0001}$ Å^{-1} as well as at $(10\bar{1}0)$.

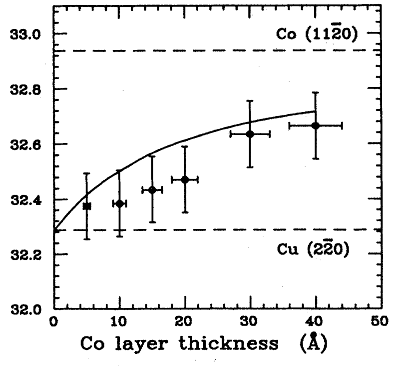

FIG. 2. Position of the $h(11\bar{2}0)/c(2\bar{2}0)$ peak vs Co thickness. Dashed lines indicate the position of this peak for bulk hcp Co (above) and bulk fcc Cu (below). The weighted average (determined by the Co- to Cu-layer ratio) of the bulk peak positions is indicated by the solid line.

In order to interpret the x-ray data quantitatively, it is necessary to employ a model which gives x-ray intensities for varying distributions of stacking sequences of close-packed planes. Models treating *random* distributions of stacking faults in hcp lattices predict a broadening of hcp peaks without the appearance of fcc peaks. It is only recently that models such as that of Sebastian and Krishna (SK) (Ref. 9) have treated the insertion of *correlated* fcc regions into an hcp matrix according to a more detailed prescription. In their model they consider the hcp to fcc transition to occur by a random insertion (nucleation) of faults in an hcp crystal, followed by growth at these sites into fcc domains by the occurrence of stacking faults at every alternate set of planes. This arrangement of stacking faults transforms hcp to fcc stacking symmetry.

In Fig. 3 we have plotted the x-ray intensity measured along $(10\bar{1}l)$ together with the profile predicted by the SK model. The model includes three parameters: γ, the probability of random growth faults in the parent hcp phase; α, the probability of nucleation of fcc growth sites; and β, the probability of continued growth at these nucleation sites. The fits shown in Fig. 3 return values of $\gamma,\alpha,\beta = 0.20$, 0.94, 0.07, and 0.30, 0.80, 0.05 for the superlattices with 10-Å and 40-Å Co layers, respectively. In order to obtain the fraction of hcp and fcc material corresponding to a given set of SK model parameters, we have performed simulations of superlattice growth according to the rules given by the model. We estimate that the values of the three parameters correspond to superlattices with 10% and 17% untransformed hcp material in the *whole* sample for the case of 10-Å and 40-Å Co layers, respectively. When these values are compared to the total quantity of Co present in each sample (which is a function of individual Co layer thickness) we find that the fraction of Co occurring as unfaulted hcp remains constant at approximately 35% across the entire range of Co thicknesses studied. That is, Co layers occur with predominantly fcc stacking symmetry. However, the distinction between fcc and hcp stacking becomes somewhat obscure in the limit of Co layers two or three monolayers thick. One would expect that the fraction of hcp Co would increase with Co thickness over the range which we have studied. In contrast, the persistence of a 35% hcp fraction, regardless of Co thickness, is reminiscent of a stable stacking polytype, which is discussed below.

In addition to revealing the stacking form of the Co layers, the $(10\bar{1}l)$ scans provide a measure of the stacking coherence length within the superlattice. In Fig. 4 we have plotted $2\pi/\Delta_q$ vs Co layer thickness, where Δ_q is the measured full width at half maximum of the fcc peak in the $(10\bar{1}l)$ scan; here $2\pi/\Delta_q$ provides an estimate of the distance over which atomic planes are coherently stacked according to the fcc sequence. An interesting finding here

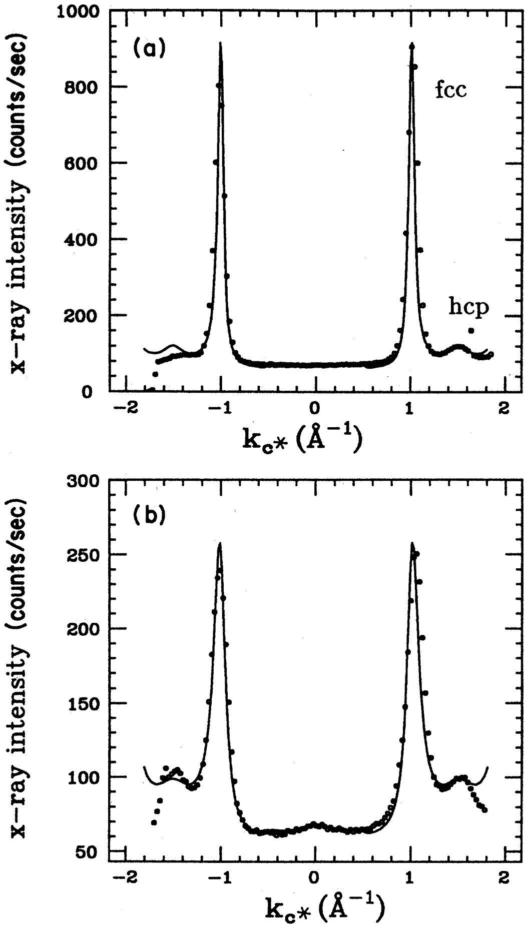

FIG. 3. X-ray scattering intensities along (10$\bar{1}l$). Fcc reciprocal-lattice points are located at $\sim \pm 1$ Å^{-1} and hcp points occur at $\sim \pm 1.5$ Å^{-1}. The data are given by points and the solid line is the fit obtained with the model of Sebastian and Krishna (Ref. 9). (a) Sample with 10-Å Co layers. (b) Sample with 40-Å Co layers.

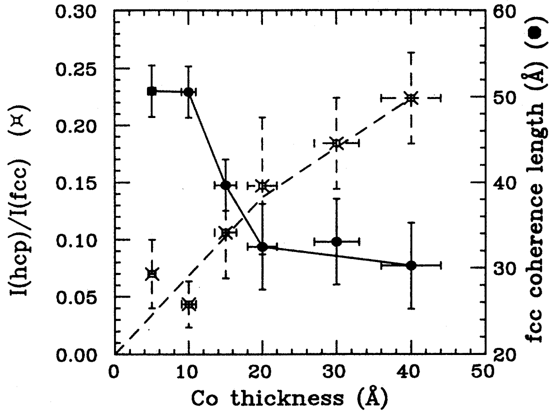

FIG. 4. Intensity ratios of the fcc and hcp peaks in the (10$\bar{1}l$) scans and coherence lengths estimated from the widths of the fcc peaks, both plotted as a function of Co layer thickness.

is that the fcc stacking coherence increases dramatically for Co layer thickness $\lesssim 20$ Å. Note that for the 10-Å Co superlattice the stacking order is maintained across neighboring Co layers. In other words, not only is there excellent coherence of Co and Cu lattices within the growth plane, as shown by Fig. 1, a high degree of stacking order extends in the growth direction as well. In addition, Fig. 4 includes a plot of the ratio of hcp to fcc peak intensities as a function of Co layer thickness.[10]

We turn now to discuss the origin of fcc stacking of Co in these superlattices. The stabilization of fcc Co by epitaxial growth on substrates such as (001) Cu is well known[11] and can be understood in terms of the cubic symmetry of the substrate plane. It is less clear why fcc Co would be favored in the stacking of *close-packed* atomic planes, since both cubic and hexagonal phases of Co possess the same symmetry within such planes.

One of the earliest ideas relevant to this problem is based on the change in *d*-band occupation which takes place across a transition series in the Periodic Table. The bcc-hcp-fcc sequence of bulk Fe, Co, and Ni has been accounted for this way.[12] In the same context, one might argue that elastic strains, which are considerable in the Co/Cu superlattice ($1 \sim 1.3\%$), might favor the fcc phase of Co. We recall also that marginally stable elements, of which hcp Co is an example, might be destabilized by charge transfer across the interface of two dissimilar metals.

These mechanisms are quite intricate and first-principles one-electron calculations are notoriously unreliable for the magnetic transition metals. The behavior has not been worked out in detail for any specific superlattice systems. In general, however, one would expect an ***abrupt*** transition to fcc from hcp from models such as that of Ref. 12 which determine phase stability and do not consider the coexistence of stacking structures. On the other hand, models based on oscillatory interlayer potentials exhibit sequences of long-range polytypes in (bulk) close-packed structures.[13] A similar approach has also been taken in a recent model appropriate to bimetallic superlattices.[14] As the modulation period in a superlattice is reduced (to $\lesssim 10$ monolayers) the overlap of Friedel-like potentials of stacking faults close to the interfaces can lead to a situation whereby a fault occurs on every alternate atomic plane converting hcp to fcc or vice versa. Our findings are consistent with such a model and indicate an admixture of fcc and hcp stacking within Co layers.

Finally, we mention some of our recent results showing striking differences in the magnetic anisotropy between Co$^{\mathrm{hcp}}$/Au superlattices[5] and the Co$^{\mathrm{fcc}}$/Cu superlattices described above. The findings presented here suggest an interesting correlation between the detailed structure and the magnetic behavior. These aspects will be discussed in a forthcoming paper.[15]

In summary, we have demonstrated the growth of epitaxial Co/Cu superlattices in which the Co atoms are predominantly arranged with cubic, rather than hcp symmetry. This transition in Co stacking occurs through the influence of the Cu layers which form a single coherent structure with the Co layers.

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