Suppression of the face-centered-cubic-hexagonal-close-packed stacking fault in Co/Cu(111) ultrathin films by pulsed laser deposition

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The thermal deposition of Co onto Cu(111) results in three-dimensional island growth and a face-centered-cubic-hexagonal-close-packed stacking fault, which hinders a complete antiferromagnetic coupling in Co/Cu(111) superlattices. We report that Co/Cu(111) films can be grown with good layer-by-layer morphology and significantly less stacking faults by pulsed laser deposition. We show that a complete antiferromagnetic coupling can be achieved in the pulsed laser deposited Co/Cu trilayer. © *1999 American Institute of Physics.* [S0003-6951(99)02003-3]

For technical reasons many current applications of magnetic thin films, e.g., sensors for disk drive and magnetic random access memory involve the deposition of face-centered-cubic (fcc) films with (111) texture.[1,2] Unfortunately, it is extremely difficult to obtain good layer-by-layer growth in these films because they are susceptible to a fcc-hexagonal-close-packed (hcp) stacking fault which forms at very little excess energy. Such a problem has been highlighted in the Co/Cu(111) superlattices grown by molecular beam epitaxy (MBE), as the expected oscillatory coupling has been rarely observed.[3–5] The fcc-hcp stacking fault in the Co layers causes the formation of a twinned fcc structure in the Cu spacer layers, leaving trenches in between which leads to a direct magnetic coupling between adjacent Co layers.[6–8] Recently some major progress has been made by Camarera *et al.* who have succeeded in improving the growth of Co/Cu(111) films by introducing Pb as a surfactant.[6–8] The Pb-assisted layer-by-layer growth of the Co films on Cu(111) has in turn strongly suppressed the formation of the fcc-hcp stacking fault and has further led to a complete antiferromagnetic (AF) coupling in a Co/Cu/Co sandwich structure prepared in a similar way. In this letter, we introduce a very different, yet more general approach to suppress the fcc-hcp stacking fault in the Co/Cu(111) system. Instead of using surfactants, we grow Co films on Cu(111) in a layer-by-layer mode by means of an ultrafast deposition method, i.e., pulsed laser deposition (PLD). This generates an instantaneous (pulsed) deposition rate about 6 orders of magnitude larger than that of the conventional MBE method while the average deposition rate is about equal. We have recently successfully applied the same PLD method to improve the growth and structure of Fe/Cu(111) and Fe/Cu(100) ultrathin films.[9,10] Here we will show that the fcc-hcp stacking fault has been suppressed in the PLD grown Co/Cu(111) films and a complete AF coupling is achieved in a PLD grown Co/Cu/Co trilayer.

The experiments were performed in an ultrahigh vacuum (UHV) multichamber system with a base pressure of 7×10^{-9} Pa which was equipped with Auger electron spectroscopy (AES), low energy electron diffraction (LEED), scanning tunneling microscopy (STM), and magneto-optical Kerr effect (MOKE) measurement facilities. Prior to the evaporation, the Cu(111) substrate was cleaned by cycles of Ar^+ sputtering and annealed at 750 K until clean Auger spectra and sharp LEED spots were obtained. The substrate was then kept at 230 K and placed 100 mm away from a Co target (4 N purity) for PLD. The output of an excimer laser with KrF (248 nm wavelength, 34 ns pulse length, typical pulse energy 270–300 mJ, and repetition rate 5 Hz) was focused onto the Co target to deposit Co onto the Cu(111) substrate. The average deposition rate was about 0.2 ML/min. After deposition the sample was transferred under UHV from the MBE chamber to the STM chamber for topographic characterization and then to an analysis chamber for LEED and AES. The magnetic characterization was performed *in situ* in both polar and longitudinal geometries. For comparison, films on the same Cu(111) substrate at the same temperature were also prepared by thermal deposition from a Co wire (5 N purity) heated by *e*-beam bombardment with a rate of about 0.2 ML/min.

For the TD Co/Cu(111) films, in the early stage of growth (<2 ML), the films are dominated by fcc structure with only a small density of stacking faults. At higher thickness, stacking faults become increasingly important and the films gradually transform to hcp structure.[11] In fact, a 5 ML Co film is almost fully hexagonally close packed,[11] which is usually thought to be the reason causing the twin structure when further growing Cu layers on top.[6,7] In contrast to the case of the TD films, the formation of the fcc-hcp stacking fault is strongly suppressed in the PLD grown Co/Cu(111) films. Figure 1 shows the current–voltage ($I-V$)-LEED curves of (10) and (01) beams for the clean Cu(111) substrate, the TD films and the PLD films. The (10) and (01) beams were defined from the LEED pattern shown as an inset in the upper left corner. The fcc and hcp stacking can be distinguished from the $I-V$ curves of these two beams since the intensities of (10) and (01) beams as a function of energy are inequivalent for a threefold symmetry like fcc (111) face and are equivalent for the sixfold symmetry of the hcp structure. At the bottom of the picture, $I-V$ curves of

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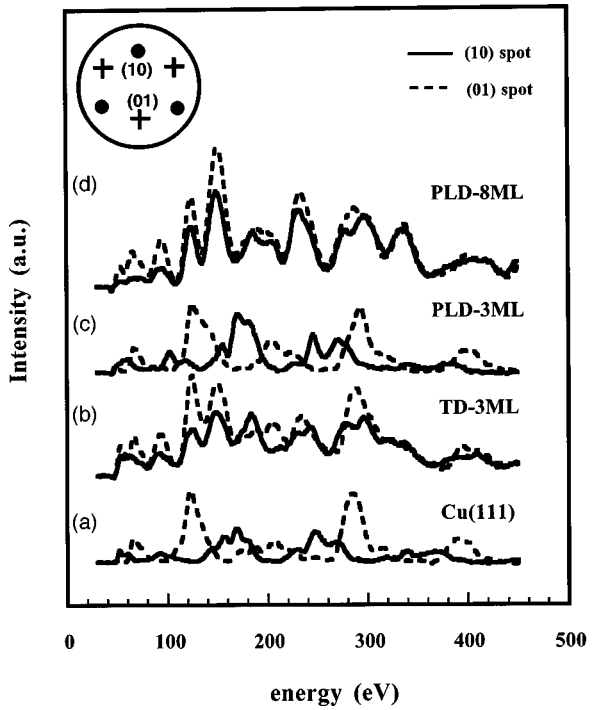

FIG. 1. LEED intensity vs energy ($I-V$) curves for the (10) and (01) beams of (a) clean Cu(111); (b) 3 ML TD-Co/Cu(111) films; (c) 3 ML PLD-Co/Cu(111) films; (d) 8 ML PLD-Co/Cu(111). The similarity of curves of (a) and (c) indicates hcp structure is suppressed in PLD films.

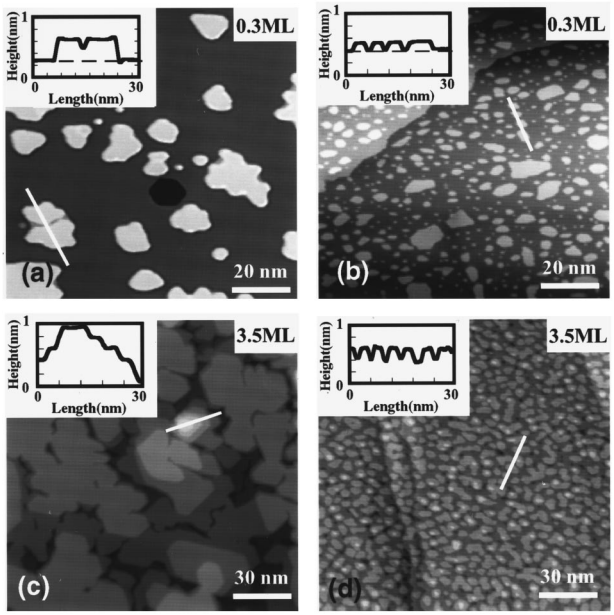

FIG. 2. STM topography images of Co/Cu(111) films prepared by thermal deposition (a), (c) and pulsed laser deposition (b), (d). The height scale of the line scans are shown in the insert. The islands of TD films are already double-layer high in the beginning state of film growth (a), where the PLD films consist of one monolayer high islands only (b). At higher thickness the TD films show a pronounced 3D growth (c), while the pulsed laser deposited films continue to grow layer-by-layer (d).

clean Cu(111) display threefold symmetry with inequivalent (10) and (01) diffracted beams, i.e., the intensity evolution with electron energy of each spot is different. Both TD- and PLD-Co films with thickness less than 2 ML have similar $I-V$ curves, indistinguishable from those of the clean substrate (not shown here). But when the thickness of the Co films is greater than 2 ML, sixfold symmetry in the LEED pattern is observed in TD-Co films, as shown by the $I-V$ curves for the 3 ML TD Co/Cu(111) film: now the two sets of $I-V$ peak positions with the electron energy of (10) and (01) beams are nearly identical. The somewhat different intensities of these two beams could be caused by the residual fraction of fcc stacking. While for the 3 ML PLD-Co film, the $I-V$ curves for the (10) and (01) beams display the exact threefold symmetry similar to those of clean Cu(111) substrate, the sixfold symmetry only becomes notable when the thickness reaches about 6 ML or above. Obviously, the hcp structure is suppressed in thin PLD-Co films. Our preliminary full dynamical $I-V$ LEED calculations yield the same results.[12]

Accompanying the suppression of stacking faults in the PLD films, a good layer-by-layer growth has also been achieved. Figure 2 shows a side-by-side comparison of the morphology of submonolayer (0.3 ML) and higher coverage (3.5 ML) Co/Cu(111) films prepared by TD (left column) and PLD (right column). A line scan across an island of the 0.3 ML TD film is displayed in the upper-left corner of the image. The TD films have a typical multilayer growth: the line profile of the 0.3 ML Co films shows that the height of the central island is about 2 ML high with a 1 ML-high brim in between. In the middle part of the image, a single-layer deep hole in the substrate is visible, which is thought to be a Cu vacancy island.[13] The substrate is still not completely wetted at 3.5 ML and there are five layers exposed on the surface as seen by the line scan through the island. For the PLD films, the morphology is close to layer-by-layer growth. The height of the island is 1 ML as seen from the line scan through the islands of the 0.3 ML films (upper-left corner) and much smaller than that of TD films. At 3.5 ML, the third layer is nearly filled while the fourth layer islands begin to grow. We have observed that the layer-by-layer growth continues up to about 8 ML.

The mechanism of the improvement towards layer-by-layer growth in ultrathin metallic films by PLD has already been discussed in our previous work.[13] Here we conjecture that in the Co/Cu(111) system, the suppression of the fcc-hcp stacking faults is a direct consequence of the improved growth. This can be understood in the following way. While in the initial stages of growth the Co atoms tend to occupy the normal fcc continuation sites,[14,15] the hcp phase stands for the natural phase of Co at room temperature and is thus energetically more favorable as compared to the fcc phase. Therefore, it is not surprising that there exists a certain critical thickness above which the Co films start to transform from fcc into hcp structure. Because of the different growth mode, the TD and the PLD Co films will reach this critical thickness at different nominal thickness. For example, assuming the critical thickness to be around 6 ML, the PLD films will transform into hcp structure at the nominal thickness of 6 ML because of the layer-by-layer growth. For the TD films, due to the island morphology the local thickness of the islands already exceeds the critical thickness (6 ML) just at a nominal thickness of 3 ML. This explains why the TD films transform into hcp structure at a lower nominal thickness compared to the PLD films.

The PLD technique also leads to an improvement in

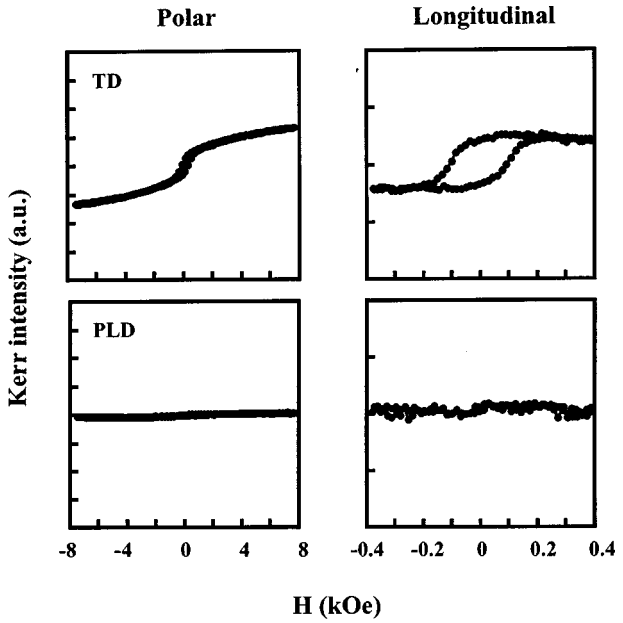

FIG. 3. Polar and longitudinal Kerr hysteresis loops for 4 ML Co/4 ML Cu/4 ML Co trilayer grown by TD (upper panel) and PLD (bottom panel) on Cu(111) measured at 230 K. The PLD film hardly exhibit hysteresis loops in both geometries, indicating that the two Co layers are completely antiferromagnetically aligned. No longitudinal signal can be measured for the PLD film even under the maximum field of 8 kOe (not shown here).

magnetic properties, e.g., the coupling between adjacent Co layers across Cu spacer layer. A complete AF coupling is observed in the PLD Co/Cu/Co trilayer on Cu(111). Figure 3 shows the polar and longitudinal MOKE loops measured at 230 K for 4 ML Co/4 ML Cu/4 ML Co trilayer grown at 230 K by TD and PLD, respectively. Note that in both TD and PLD trilayers, the Cu spacer layer was thermally evaporated from a crucible. Here we assign the thicknesses of Co and Cu layers the same as Ref. 7, where 4 ML Cu should yield maximum AF coupling. When grown by TD, the films show partial ferromagnetic coupling as indicated by the large remanence both in the polar and longitudinal loops. By contrast, the signal is close to zero in the PLD-grown 4 ML Co/4 ML Cu/4 ML Co trilayer, which means that the adjacent Co layers are aligned antiparallel and a complete AF coupling is achieved. (The fields available to use are not sufficient to break up the AF coupling.)

A calculation made by Gradmann and Elmers[16] pointed out that there is a critical width for the spacer below which complete ferromagnetic alignment remains stable in spite of AF coupling. It was suggested that the areal fraction of ferromagnetically coupling channels or holes must be less than some critical value in order to obtain AF order. Thus the absence of AF coupling in the TD Co/Cu(111) superlattices is understandable due to its hindrance of the coalescence of the islands. While in the PLD films, layer-by-layer growth makes the spacer layer coalesce at large scale and exceed the critical width so that the AF coupling can be obtained. It is important to note here that even though similar magnetic results have been observed in the Pb-surfactant-assisted Co/Cu superlattices,[6–8] the PLD technique is a more general method to be used in other magnetic superlattices.

In summary, we have shown that layer-by-layer growth of Co/Cu(111) can be obtained by the PLD technique. The formation of hcp stacking is greatly suppressed in the PLD films. The fact that the improved layer-by-layer growth and suppression of fcc-hcp stacking faults in the PLD films leads to a complete AF coupling in a PLD Co/Cu/Co trilayer makes the PLD a general and convenient technique for thin film fabrication.

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[1] S. S. P. Parkin, R. Bhadra, and K. P. Roche, Phys. Rev. Lett. **66**, 2152 (1991).
[2] M. T. Kief and W. F. Egelhoff, Jr., J. Appl. Phys. **73**, 6195 (1993).
[3] W. F. Egelhoff, Jr. and M. T. Kief, Phys. Rev. B **45**, 7795 (1992).
[4] M. T. Johnson, R. Coehoorn, J. J. de Vries, N. W. E. McGee, J. ann de Stegge, and P. H. Bloemen, Phys. Rev. Lett. **69**, 969 (1992).
[5] S. S. P. Parkin, R. F. Marks, R. F. C. Farrow, G. R. Harp, Q. H. Lam, and R. J. Savoy, Phys. Rev. B **46**, 9262 (1992).
[6] J. Camarero, L. Spendeler, G. Schmidt, K. Heinz, J. J. de Miguel, and R. Miranda, Phys. Rev. Lett. **73**, 2448 (1994).
[7] J. Camarero, T. Graf, J. J. de Miguel, R. Miranda, W. Kuch, M. Zharnikov, A. Dittschar, C. M. Schneider, and J. Kirschner, Phys. Rev. Lett. **76**, 4428 (1996).
[8] W. Kuch, A. Dittschar, M.-T. Lin, M. Salvietti, M. Zharnikov, C. M. Schneider, J. Kirschner, J. Camarero, J. J. de Miguel, and R. Miranda, J. Magn. Magn. Mater. **170**, L13 (1997).
[9] H. Jenniches, M. Klaua, H. Höche, and J. Kirschner, Appl. Phys. Lett. **69**, 3339 (1996); J. Shen, P. Ohresser, Ch. V. Mohan, M. Klaua, J. Barthel, and J. Kirschner, Phys. Rev. Lett. **80**, 1980 (1998).
[10] J. Shen, H. Jenniches, Ch. V. Mohan, J. Barthel, M. Klaua, P. Ohresser, and J. Kirschner, Europhys. Lett. **49**, 349 (1998).
[11] Ch. Rath, J. E. Prieto, S. Müller, R. Miranda, and K. Heinz, Phys. Rev. B **55**, 10791 (1997).
[12] A. Zangwill, Mater. Res. Soc. Symp. Proc. **280**, 121 (1993).
[13] M. Ø. Pederson, I. A. Bönicke, E. Laegsgaard, I. Stensgaard, A. Ruban, J. K. Nørskov, and F. Besenbacher, Surf. Sci. **387**, 86 (1997).
[14] B. P. Tonner, Z. L. Han, and J. Zhang, Phys. Rev. B **47**, 9723 (1993).
[15] B. Piveteau, D. Spanjaard, and M. C. Desjonquères, Phys. Rev. B **46**, 7121 (1992).
[16] U. Gradmann and H. J. Elmers, J. Magn. Magn. Mater. **137**, 44 (1994).