6027

Scanning tunneling microscopy study of the growth of Cr/Fe(001): Correlation with exchange coupling of magnetic layers

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Scanning tunneling microscopy (STM) and reflection high-energy electron diffraction (RHEED) were used to study the epitaxial growth of Cr on Fe(001) whiskers as a function of the Fe whisker temperature during growth. The STM images give real space views of the morphology of Cr growth, which can be correlated with the nature of the RHEED intensity oscillations. Layer by layer growth is found for Cr deposition on an Fe(001) surface at 300 °C, and very rough growth, limited by diffusion kinetics, is observed at lower temperatures. The variation in the interlayer exchange coupling in Fe/Cr/Fe sandwiches as a function of the thickness of the Cr interlayer, which has been found to depend strongly on the growth temperature of the Cr interlayer, can be explained by the thickness fluctuations determined from the STM measurements of Cr films grown at different temperatures.

I. INTRODUCTION

The magnetic properties of thin films are profoundly affected by the physical structure developed during growth. Controlling this physical structure provides a means to control a number of different properties, including domain structures, coercivity, and magnetic anisotropy.[1] Recently, it was established[2] that the magnetic exchange coupling of two ferromagnetic films of Fe through a nonmagnetic layer such as Cr depended strongly on the growth conditions of the Cr film, and therefore, one expects, on the structure of the Cr film. In this paper we report on our investigation of the growth of Cr on Fe using scanning tunneling microscopy (STM) and reflection high-energy electron diffraction (RHEED). The structure of the film is found to depend strongly on the temperature of the substrate during growth. We discuss how variations in the thin film structure directly explain variations in the observed magnetic properties. Superlattices of alternating thicknesses of a ferromagnetic and a nonferromagnetic metal have been shown to exhibit a "giant magnetoresistance,"[3,4] and hence nanostructures of this type form a promising new class of magnetic sensors.

II. EXCHANGE COUPLING IN Fe/Cr/Fe

The sensitivity of the exchange coupling to thin film growth was first observed in studies of the Fe/Cr/Fe(001) sandwiches.[2] The sandwich consists of a linearly increasing thickness of Cr, that is, a wedge, grown on an Fe single crystal, which was then covered by a thin Fe film of approximately ten layers. The Fe overlayer may couple to the Fe substrate, with its magnetization in the same direction, "ferromagnetic coupling," or in the opposite direction, "antiferromagnetic coupling." An image of the magnetization in the Fe overlayer shows how it is coupled to the magnetization of the Fe(001) whisker substrate, and hence how the coupling varies with the thickness of the Cr interlayer. The magnetization image is obtained by measuring the spin polarization of the secondary electrons in a scanning electron microscope, a technique called scanning electron microscopy with polarization analysis (SEMPA), described elsewhere.[2,5] Such an image of the magnetization in the Fe overlayer is shown in Fig. 1. The Cr interlayer increases in thickness from 0 to 40 layers over a distance of 0.4 mm from left to right across the image. At the thinnest part of the Cr layer on the left, the coupling is ferromagnetic (white in the image). With increasing Cr thickness, the coupling changes to antiferromagnetic (black in the image) and continues to oscillate with a period of 12 ± 1 layers change in Cr thickness.

There is a striking variation in the magnetization images of Fig. 1(b) and 1(c) for Fe/Cr/Fe(001) sandwiches, where the Cr was grown on the Fe whisker held at 200 and 350 °C, respectively, compared to growth at a substrate temperature of 30 °C in Fig. 1(a). In Fig. 1(c), after an initial region of ferromagnetic coupling, the exchange coupling reverses with each additional Cr layer, that is, it oscillates with a period of approximately two layers. Careful SEMPA measurements have shown that the period is actually 2.105 ± 0.005 layers, which leads to the "phase slip" evident as the wider white band at 24–25 layers in the magnetization image[2,6] of Fig. 1(c). Although the short period coupling is dominant, the long period coupling still exists in Fig. 1(c), and can be observed in a more detailed analysis of the magnetization image. At the intermediate growth temperature of 215 °C, some residual short period coupling is observed up to about 16 layers, but the long period coupling dominates over most of the image. These results stand in contrast to the coupling at the lowest growth temperature of 30 °C, where the long period oscillations of the coupling dominate the magnetization image. Clearly, the Cr growth temperature has a large effect on the exchange coupling.

III. GROWTH OF Cr/Fe(001)

The Fe(001) whisker offers an early perfect single crystal substrate.[7] The Fe whisker surface was cleaned by ion bombardment at 750 °C and then cooled to room temperature for the STM measurements. A 0.72×0.72 μm square STM image is shown in Fig. 2. There is a single atomic step visible in the image. One single atom high step was observed approximately every micrometer, corresponding to an align-

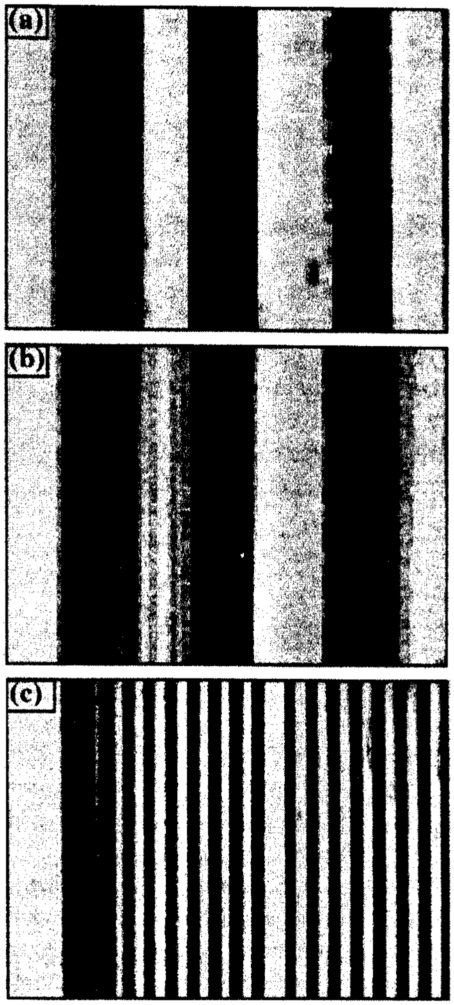

FIG. 1. Images of the magnetization in an Fe layer coupled through a Cr interlayer of varying thickness to an Fe(001) substrate. The Cr spacer layer, which increases in thickness from 0 to 40 layers from the left to the right of the images, was grown at Fe substrate temperatures of 30, 200, and 350 °C in (a)–(c), respectively. The magnetization of the Fe overlayer is parallel (ferromagnetically coupled) to the substrate in the white regions and antiparallel (antiferromagnetically coupled) in the black regions.

ment of the surface to better than 0.01°. This is much better alignment that is obtained in the conventional preparation of metal crystals and also avoids damage due to surface polishing.

The temperature dependence of Cr growth was studied by evaporating Cr on the Fe(100) whisker surface held at different temperatures and then cooling to room temperature for the STM measurements. With this method, a "snapshot" of the growth is obtained[8] at different stages, as we have illustrated in extensive studies of the homoepitaxial growth of Fe on Fe(001).[9] When a Cr atom reaches the Fe surface, it will diffuse until it collides with another atom to form a stable nucleus for island growth or until it reaches an existing island edge and is incorporated. Thus, there is a competition between nucleation and growth, depending on the deposition rate and on the surface diffusion, which is strongly temperature dependent.

RHEED was used to monitor the growth during the thin

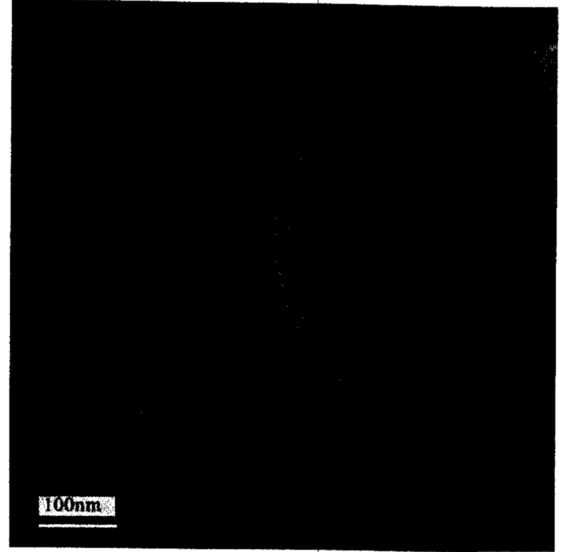

FIG. 2. A 720×720 nm STM image of the Fe(001) whisker surface showing a single atomic step. The higher level is indicated by the lighter gray.

film deposition at different substrate temperatures. Examples of RHEED intensity oscillations as a function of deposition time are shown in Fig. 3 for deposition at 100, 215, and 300 °C, for final film thicknesses of 4.5, 4.65, and 3.7 layers, respectively. The strong damping of the oscillations after two cycles is indicative of rough growth at 100 °C. The cusp-like nature of RHEED intensity oscillations that return to nearly their initial value indicate a very different kind of growth at 300 °C. As seen from the intensity oscillations, the deposition rate was between one and two layers per minute in the three cases.

After deposition was halted in the three film growths of Fig. 3, the sample was allowed to cool to room temperature, and STM images were acquired. The STM images for growth at 100, 215, and 300 are shown in Figs. 4(a)–4(c), respectively, at widely different magnifications. The growth at 100 °C shown in Fig. 4(a) is very rough with portions of five layers, layers three through seven, visible. The amount of each layer exposed is fit well by a Gaussian with σ, the

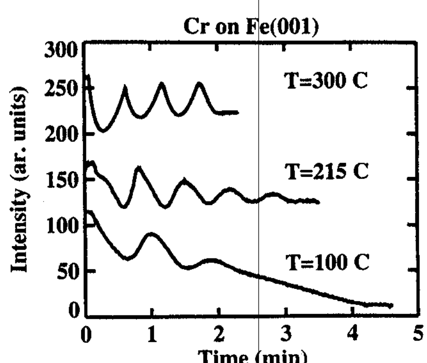

FIG. 3. RHEED (0,0) beam intensity oscillations measured during growth of Cr films on Fe at 100, 215, and 300 °C. The RHEED measurements were made with a 10 keV beam at the antiphase angle of incidence of 64 mrad. The growth was stopped after 4.5, 4.65, and 3.7 layers, respectively.

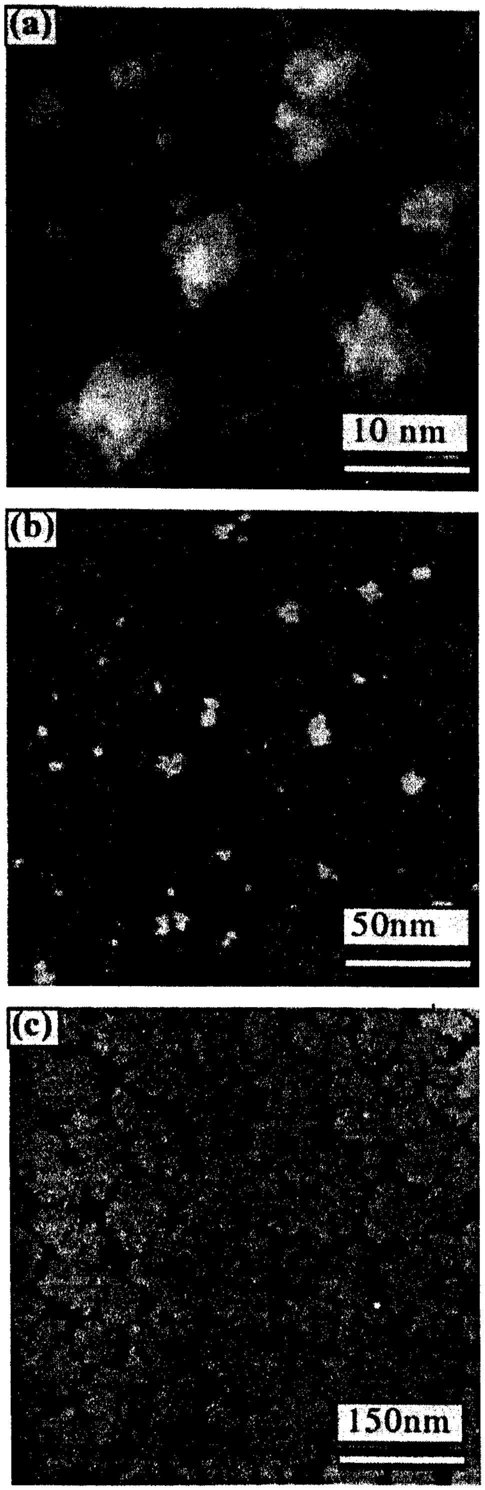

FIG. 4. The STM measurements of the Cr films, the growth of which was monitored by the RHEED oscillations of Fig. 3, after growth was stopped and the sample cooled to room temperature. The images are for Cr grown on Fe(001) at (a) 100 °C, (b) 215 °C, and (c) 300 °C.

rms roughness, equal to 0.77 layers (0.111 nm). At the intermediate temperature growth shown in Fig. 4(b), the islands are significantly larger, owing to the diffusion coefficient. The rms roughness is 0.47 layers (0.068 nm). In Fig. 4(b), the four gray levels correspond to layers three through six.

The growth of Cr on the Fe whisker held at 300 °C, Fig. 4(c), is distinctly different from the rough growth at the lower temperatures. At this temperature, the incident atoms diffuse readily and large islands are obtained that grow to complete one layer before the next layer begins. This case of true layer by layer growth has the characteristic RHEED intensity oscillations shown in the upper curve of Fig. 3. Another look at this layer by layer growth over a larger area is shown in Fig. 5. There is a step running diagonally across the image. The Cr grows out from the step, giving it the irregular contour, unlike the characteristically smooth contour of steps on the clean substrate as observed in Fig. 2. On the upper edge of the step, one observes a denuded region a few tens of nm wide due to the step edge acting as a sink for the diffusing atoms.

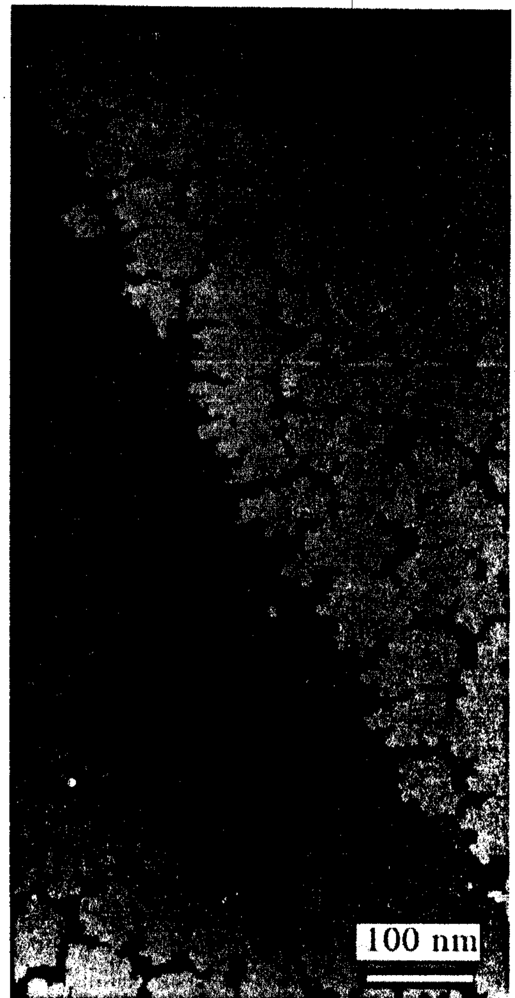

FIG. 5. The layer by layer growth of Cr on Fe(001) at 300 °C showing growth out from the step and a denuded region above the step edge.

IV. CORRELATION OF EXCHANGE COUPLING WITH GROWTH

The temperature dependence of the exchange coupling shown in Fig. 1 can be understood in terms of the STM measurements. The reversal of the Fe overlayer magnetization with each additional layer of the Cr spacer is not observed at temperatures when the growth temperature leads to a rough Cr film. The STM measurements tell us how many layers are present at the surface of a Cr film of some average thickness. Because of the large terraces of the Fe whisker substrate, Fig. 2, the measured roughness of a Cr film gives directly the fluctuation in the thickness of the film. The STM measurements show that on a very microscopic scale the fluctuations are not arbitrary but always occur in discrete increments of atomic layers as expected.

The Fe overlayer magnetization responds to two competing interactions. First, there is the *interlayer* exchange coupling through the Cr interlayer, which we have been discussing. Second, there is an *intralayer* exchange coupling that prevents the magnetization within the Fe overlayer from reversing abruptly, and instead causes it to change over characteristic distances of order 100 nm. The magnetization of the Fe overlayer cannot reverse over the length scales characteristic of the size of the islands formed during the Cr growth. Rather, at each average thickness, the interlayer exchange coupling to which the Fe overlayer responds is determined by adding the contribution to the coupling through regions of discrete thickness in the Cr growth front weighted by the area of each region. In the case of layer by layer growth, where only two layers are in the Cr growth front, the coupling reverses as the area weighted strength of the interaction of the additional layer becomes larger than that of the completed layer. In the case of rough growth with several layers in the growth front, the strength of the coupling at each layer thickness, corresponding to an integral number of layers, is summed, weighted by the relative area of that layer exposed. As we describe quantitatively elsewhere,[10] it is possible to start with a model that fits the magnetic coupling in the layer by layer growth case well, and, simply by adding the individual contributions to the coupling determined by the thickness fluctuations taken from the STM measurements, to reproduce the magnetization profiles corresponding to the magnetization images of Figs. 1(a) and 1(b). Because it is thickness fluctuations that are important, rather than interface roughness, layer by layer growth, even on a rough substrate, should allow observation of short period oscillations of the coupling. The difficulty encountered by others in observing short period oscillations of the exchange coupling through Cr grown on substrates other than Fe whiskers points to inhibition of layer by layer growth by rough substrates.

V. CONCLUSION

The STM measurements provide a quantitative picture of the Cr growth front at each temperature. The layer by layer growth at 300 °C leads to distinctive cusp-like RHEED intensity oscillations with undamped maxima, whereas the rough growth at lower temperatures leads to rapidly decaying rheed intensity oscillations. Wang *et al.*[11] suggested some time ago, before short period oscillations in the exchange coupling were observed experimentally, that thickness fluctuations would prevent the observation of the short period oscillations calculated in their theory. From our STM measurements we obtain a quantitative picture of the thickness fluctuations in the Cr film grown at different temperatures. We found that the same model for the interlayer exchange coupling, which fits the magnetization data for layer by layer growth also fits the magnetization data at lower temperatures when the thickness fluctuations are taken into account.

ACKNOWLEDGMENTS

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