Formation of crystallographic texture in rf sputter-deposited Cr thin films

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Although it is well known that Cr underlayers sputter deposited on glass or NiP/Al substrates have either the (002) or (110) textures, the mechanism of the formation of the crystallographic textures is not clear. A model is proposed for the formation of the crystallographic texture of sputter-deposited Cr thin films. A systematic set of experiments has been carried out to test the model. It was found that the (110) texture, which is usually found in Cr thin films deposited on substrates without preheating, can form at elevated temperatures (250 °C) when deposited at low Ar pressure or by applying substrate bias. The initial stage of the texture formation was also investigated by using very thin Cr films. It was found that the (002) texture can be initiated directly on the substrate surface, while the (110) texture appears not to form directly on substrate surface, but rather as a result of film growth. The proposed model is consistent with the experimental results. © *1994 American Institute of Physics.*

I. INTRODUCTION

Cr thin films are commonly used as underlayers for thin-film magnetic media. It has been reported that Co alloy films grow epitaxially on the Cr underlayers.[1–4] As a result, the crystallographic texture and the grain size of the Co alloy films, which strongly influence the magnetic properties, are determined primarily by the Cr underlayer. Understanding how the crystallographic texture forms is important to better design the sputtering process and control the crystallographic texture and other microstructural characteristics of the Cr film, and hence control these same features of the subsequently deposited magnetic film.

It is well known that sputtered Cr thin films have either the (110) or (002) crystallographic texture.[5–8] A growth mechanism based on the selection of grains with faster growth habits is proposed to explain the fact that the Cr grains with the (110) orientation tend to dominate the surface at large thicknesses;[6,9] however, this mechanism alone is not sufficient to understand the dependence of the crystallographic texture on process conditions. For example, an important question is why the (002) texture forms only at elevated substrate temperatures. On the other hand, while the dependence of the Cr layer texture on individual sputtering parameters, such as substrate temperature, Ar pressure, and substrate bias, has been studied by various groups,[4–8] the results to date do not give a complete picture of the relationships between process conditions and crystallographic texture. This is because the film growth process is controlled by the combined effects of all the sputtering parameters, which depend strongly on the sputtering system being used. A systematic investigation is necessary to obtain a better understanding of the dependence of the Cr texture on the various sputtering parameters.

In this work a model for the formation of the crystallographic texture is proposed based on the minimization of surface energy. In conjunction with this model, a systematic set of experiments has been carried out. The effects of substrate temperature, forward sputtering power, Ar pressure, and substrate bias are discussed in terms of the proposed model.

II. MODEL FOR THE FORMATION OF THE CRYSTALLOGRAPHIC TEXTURE OF Cr

Two mechanisms of the formation of texture are proposed in this model. In the first mechanism (mechanism I), the crystallographic texture originates from the preferential orientation of islands, which are nucleated on the substrate surface before a continuous film forms. In the second mechanism (mechanism II), the crystallographic texture develops by the faster growth of grains with certain orientation after the film becomes a continuous film. Both of these processes are based on the minimization of surface energy at the time when the crystallographic texture develops.

A. Formation of texture during nucleation of islands

It is commonly observed that islands are nucleated before a continuous film forms. If an island is in its thermodynamic equilibrium state, its shape and orientation is determined by the minimization of its total surface (including interfacial) energy. This has been demonstrated theoretically by Winterbottom[10] and Cahn and Taylor.[11] There are three kinds of surfaces: the free surface of the Cr thin film S_1, the free surface of the substrate S_2, and the interface between the Cr thin film and the substrate S_{12}, as illustrated in Fig. 1(A). The free surface S_1 is likely to be composed of crystallographic planes (facets), such as the {110} and {002}. Among them, the {110} planes have the lowest surface energy because, for bcc Cr, the atoms of the {110} plane are the closest packed.[12] In our model, it is assumed that the S_1 is composed of the {110} and {002} planes.

The equilibrium shape of the islands depends on the relative values of the specific surface or interfacial free en-

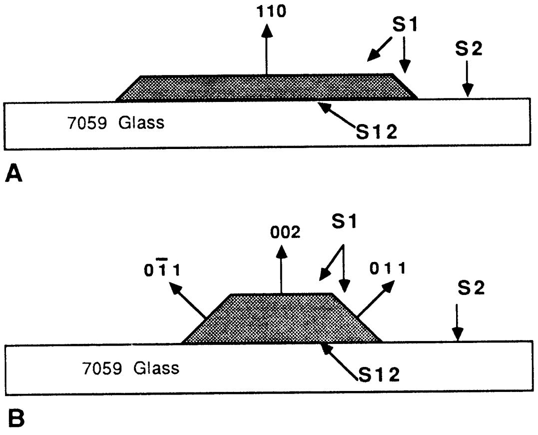

FIG. 1. Schematic of (A) a flat and (B) an equiaxed island.

ergies γ_2, γ_{12}, $\gamma_{(110)}$, and $\gamma_{(002)}$. Given these values of γ, the equilibrium shape can be predicted by Winterbottom's construction.[10] The larger the γ_{12}/γ_2 ratio, the smaller the area associated with S_{12}. When γ_{12}/γ_2 is smaller, the ratio of the height of the island to the area of the interface S_{12} is small; in other words, the island tends to spread out. If γ_{12}/γ_2 is larger, the ratio of the height of the island to its length increases, causing the island to be more equiaxed, as illustrated in Fig. 1(B). The orientation of the islands is further decided by the relative values of $\gamma_{(110)}$ and $\gamma_{(002)}$. Since the (110) planes have the lowest surface energy, they tend to cover the most area of the free surface S_1. When islands have a flat shape as shown in Fig. 1(A), the (110) plane is on the top because the area of the top is larger than the total area of the sides. When the islands have the equiaxed shape shown in Fig. 1(B), a (002) plane is on the top and the (110) planes are on the sides, since this configuration maximizes the {110} surface area, and thus minimizes the total surface energy.

When the sputtering conditions allow many islands to obtain surface energy equilibrium before they impinge on each other, the islands have a preferential orientation. After the islands impinge on each other and finally become a continuous film, the preferential orientation of islands becomes the crystallographic texture of the film. For bcc Cr, depending on substrate material, the preferential orientation of islands could be either the (002) or the (110), corresponding to equiaxed and flat islands, respectively. On the other hand, experimental results show that when Cr is deposited on a glass substrate, either the (002) or the (110) texture can be obtained depending on the sputtering conditions.[5–8] As we will see later in this section, mechanism II always leads to the development of the (110) texture. Therefore, if the (002) texture is to form, it must be formed during the nucleation of islands by mechanism I.

When the sputtering conditions do not allow the individual islands to reach surface energy equilibrium before they impinge on each other, the islands do not have a strong preferential orientation. A crystallographic texture may develop after the film becomes continuous by the faster growth of grains with a certain orientation.

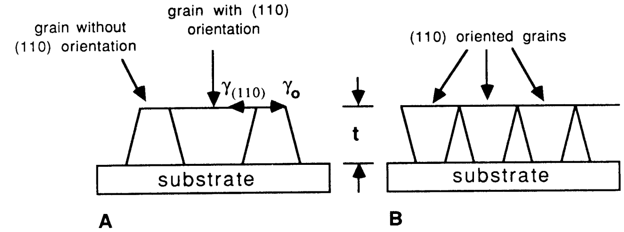

FIG. 2. Schematic to show the faster grain growth and the effect of impingement island size: (A) films with large impingement island size, (B) film with small impingement island size.

B. Formation of texture by faster growth of grains with certain orientation

When the islands impinge on each other, a polycrystalline film forms. Assuming that the grains originally have random orientation, some grains will grow faster than others during the deposition;[9] namely those favored by the reduction in surface energy. The grains with an orientation which allows the grains have a plane with lower free energy on the surface of the film will extend at the expense of other grains. This is illustrated in Fig. 2 by comparing the state when the film has a thickness t to that when it has a thickness $2t$. In the temperature region of our sputtering experiments, the extension of grains only happens on the surface because the mobility of grain boundaries throughout the film is negligible. As one orientation is more favorable than the others, with regard to grain growth, a crystallographic texture develops during the deposition. For bcc Cr, the {110} planes have the lowest surface energy, hence, the (110) texture is the one which develops by this mechanism.

The rate of the (110) texture development depends on the impingement island size, which is the size of islands when they begin to impinge each other. The smaller the impingement island size is (hence the higher the number of grains per unit area), the easier the (110) texture develops. As illustrated in Fig. 2(B), the (110) plane covers the whole surface by the faster growth mechanism when the film is thicker than t for the film with smaller impingement island sizes. This is not the case for a film with larger impingement island size [see Fig. 2(A)].

Up until now our model has assumed that grains have random orientation. In fact, even if a (002) texture develops initially, as long as some grains have the (110) orientation, the (110) grains will "win out" as the film grows thicker. It has been reported that the Cr texture can change from the (002) to the (110) texture as the film becomes thicker.[6,9] To explain this, consider the film growth just after the film becomes continuous. Some grains have the (002) orientation while a few have the (110) orientation. The (002) texture, which has originated from the islands (mechanism I) will change to the (110) texture as the film grows thicker.

The model is summarized as follows.

(1) If the islands can reach their equilibrium shape before impingement, the islands prefer the (002) orientation, hence the (002) texture forms. We call this texture formation mechanism mechanism I.

TABLE I. Dependence of the nucleation and growth parameters on sputtering conditions. 1st: very strong; 2nd: not so strong; 3rd: weak; no: no effect.

Nucleation and growth parameters	Sputtering conditions			
	Preheating	Ar pressure	Sub. bias	Power
Sub. temperature	1st	3rd	2nd	2nd
Deposition rate	no	2nd	2nd	1st
Kin. energy of Cr	no	1st	2nd	2nd
Kin. energy of Ar	no	2nd	1st	3rd

(2) If the islands cannot obtain their equilibrium shape before they impinge on each other, the (110) texture will develop during the growth of the film. We call this mechanism mechanism II. The smaller the impingement island size, the more easily the (110) texture develops.

As discussed above, the crystallographic textures of thin films depend on the orientation, shape, and size of the islands at the initial stage of film formation. The film formation is considered to be a process of nucleation and then growth. This process is controlled by the deposition rate, the kinetic energy of the Cr atoms, the kinetic energy of the impinging Ar^+ ions, and the substrate temperature. These parameters can be adjusted by varying the sputtering conditions. So, by examining the dependence of the crystallographic texture of the thin film on the sputtering condition, we can test the model.

III. EXPERIMENTAL TEST OF THE MODEL: Cr THIN FILMS ON GLASS SUBSTRATES

Cr films were deposited at various sputtering powers, Ar pressures, and substrate bias on substrates at either room temperature or preheated to 250 °C. All the Cr thin films were rf sputter deposited on Corning 7059 glass substrates in a Leybold-Heraeus Z-400 sputtering system with a background pressure of 7.0×10^{-7} Torr. The rate of deposition was determined by step profilometry measurements and these rates were used to calculate the various thicknesses of the films. The films are 1000 Å thick except when specified otherwise. The crystallographic texture was studied by x-ray diffraction using Cu $K\alpha$ radiation.

Each nucleation and growth parameter depends on several sputtering parameters. For example, the substrate surface temperature T_s depends mostly on the preheating temperature, but also depends on substrate bias and forward sputtering power. When applying a substrate bias, the substrate surface is bombarded by accelerated Ar^+ ions, so the T_s increases. During the sputtering process, part of the atomic impact energy transforms into thermal energy on the surface of the substrate. Thus, by increasing the forward sputtering power, the substrate temperature also increases. The Ar pressure may also affect the substrate temperature as it affects the atomic impact energy. A qualitative order of magnitude of the dependence of the nucleation and growth parameters on sputtering conditions in our experimental range is summarized in Table I.

In the following, the dependence of crystallographic texture on each of the sputtering parameters is discussed in the light of our proposed model. At the end of this section, the initial stage of texture formation is also discussed to further test the model.

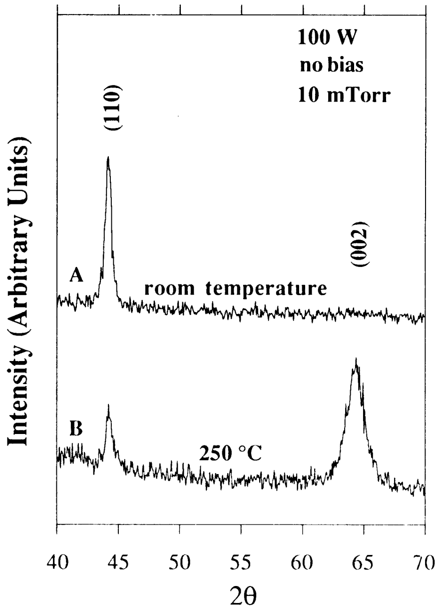

FIG. 3. The x-ray spectra of two 1000 Å films: (A) no preheating, and (B) preheated to 250 °C.

A. Preheating substrate temperature

The Cr films deposited at room temperature all show the (110) crystallographic texture; however, films deposited at 250 °C have textures which depend on the specific values of sputtering power, Ar pressure, and the substrate bias.

The x-ray diffraction spectra of two samples, deposited with and without preheating, respectively, are shown in Fig. 3. Both samples were deposited with 100 W sputtering power and 10 mTorr Ar pressure and without applying substrate bias. The one deposited without preheating shows the (110) texture while the one deposited with substrate preheating to 250 °C is dominated by the (002) texture. This is consistent with the results of previous studies.[4-6]

The dependence of the crystallographic texture on the substrate temperature can be well explained by our model. The nucleation of islands depends on the mobility of the adatoms on the substrate surface and the kinetic energy of bombarding particles. By increasing the substrate temperature, the mobility of the adatoms increases, promoting surface diffusion. The size of the critical nucleus, which is the minimal stable nucleus, increases with increasing temperature. Hence, the rate of formation of stable nuclei decreases rapidly with increasing temperature and the film preserves its islandlike character for larger thickness as the temperature is increased.[13] Thus, at high temperature, more time (and more matter) is needed for the formation of a continuous film; hence, elevated substrate temperature promotes large, equilibrium islands, which have (002) planes parallel to the film plane, and the (002) texture thus forms by mechanism I. This explains why the (002) texture develops when films are deposited at elevated temperatures. On the other hand, when

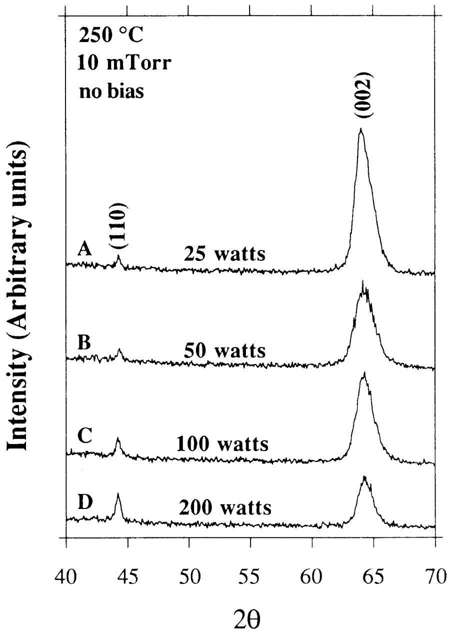

FIG. 4. The x-ray spectra of Cr thin films deposited with various sputtering powers: (A) 25 W, (B) 50 W, (C) 100 W, and (D) 200 W.

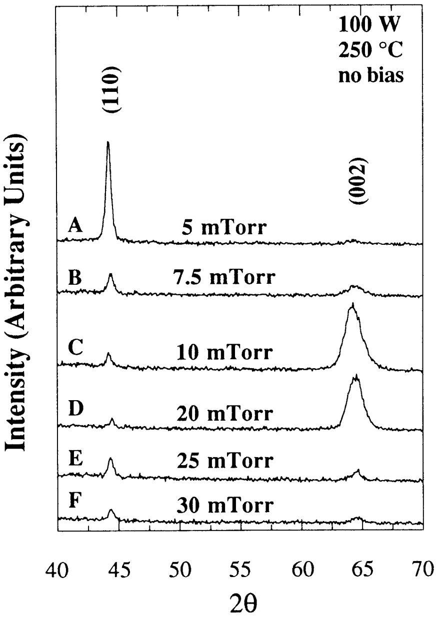

FIG. 5. The x-ray spectra of Cr thin films deposited at various Ar pressures: (A) 5 mTorr, (B) 7.5 mTorr, (C) 10 mTorr, (D) 20 mTorr, (E) 25 mTorr, and (F) 30 mTorr.

deposited at low substrate temperatures, the atoms are "quenched" onto the substrate surface and the concentration of nucleation centers is high; hence, the islands are small and might not reach their equilibrium shape and/or orientation. The islands thus have a random initial orientation. After the islands impinge each other, the (110) texture will develop by mechanism II.

B. Sputtering power

The x-ray diffraction spectra for Cr films deposited with different sputtering powers are shown in Fig. 4. The Ar pressure was 10 mTorr and the substrate was preheated to 250 °C for all the films. No substrate bias was applied during deposition. The (002) texture dominates in all of the four samples, but the ratio of the intensities of the (002) and (110) peaks, $I_{(002)}/I_{(110)}$, decreases as sputtering power increases. This can be understood by considering the dependence of deposition rate on sputtering power. The most significant change on the nucleation and growth parameters accompanying the change in the sputtering power is the deposition rate, which is approximately proportional to the sputtering power. At lower deposition rate, the islands have more time to reach their equilibrium state. This produces a strong (002) texture. Also, at lower deposition rate, the concentration of nucleated islands is lower and therefore they have more time to grow. Hence, the impingement island size is large. As discussed in the previous subsection, both of these factors will help to maintain the (002) texture. Thus, films deposited at low sputter power at elevated temperatures have stronger (002) texture than those deposited at higher sputter powers.

C. Ar pressure

The x-ray diffraction spectra of Cr films deposited at different Ar pressures are shown in Fig. 5. All of these films were deposited at 100 W sputtering power and the substrate was preheated to 250 °C, without applying substrate bias. It can be seen that from 5 to 10 mTorr the intensity of the (110) peaks decreases and the intensity of the (002) peaks increases. In the region from 10 to 20 mTorr the (002) texture dominants. With further increase in the Ar pressure both the (110) and (002) peaks become weak, implying that these samples have a more random distribution of orientations.

The dependence of crystallographic texture on Ar pressure can be understood based on the influence of Ar pressure on the kinetic energy of the deposited Cr atoms. The average kinetic energy of the deposited Cr atoms decreases with increasing Ar pressure, due to collisions between the sputtered atoms and the Ar molecules. At 5 mTorr Ar pressure, the mean free path of the sputtered atoms is approximately 2 cm. The distance between the target and the substrate is about 2.5 cm. Hence, on average, a sputtered atom collides with an Ar atom one time while it travels from target to the substrate. For atoms sputtered in 10 mTorr Ar gas the average number of collisions is twice of that of the atoms sputtered in 5 mTorr Ar gas; thus, the energy of sputtered atoms when they reach the surface is much less for those in the 10 mTorr Ar pressure.

The kinetic energy of the deposited Cr atoms acts in two aspects. First, because the particles with high energy may create defects (locally breaking the chemical bond of the substrate material), the sites of impingement will become sites of preferential nucleation.[14] Hence, the concentration of nucleation centers is increased by bombardment of Cr atoms with higher kinetic energy. On the other hand, part of the

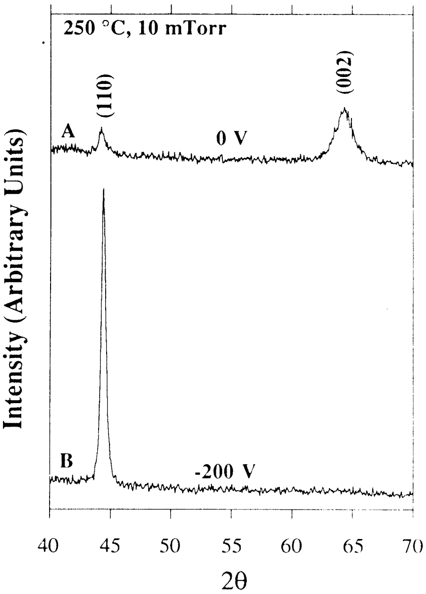

FIG. 6. The x-ray spectra of Cr thin films deposited (A) without applying substrate bias and (B) with −200 V substrate bias.

kinetic energy will transform into thermal energy on the surface of the substrate. Hence, Cr atoms with higher kinetic energy have a larger mobility, which promotes the attainment of equilibrium for the islands. When the film is deposited with low Ar pressure (5 mTorr), at which the kinetic energy of the bombarding particles (in this case, the Cr atoms) is very high, the impingement island size is small. Based on our model, this promotes the development of the (110) texture by mechanism II, after the film became continuous. When the film is deposited with medium Ar pressure (10–20 mTorr), the concentration of islands is not so high, and also the mobility of the adatoms (Cr atoms) is high enough for the islands to reach their equilibrium state before they impinge on each other, causing the (002) texture to form by mechanism I. When the film is deposited with high Ar pressure (25 and 30 mTorr), the mobility of the adatoms is too small for the islands to reach equilibrium, so they have a random distribution of orientations. The low atomic mobility also prevents the (110) texture from developing by mechanism II because surface diffusion is required in that mechanism; so, the films deposited at high Ar pressure do not have a strong texture.

D. Substrate bias

When a substrate bias is applied, the Ar$^+$ ions, which are accelerated by the substrate bias voltage, also bombard the substrate. Two Cr films were deposited with and without substrate bias at the same deposition rate. The preheated substrate temperature was 250 °C and Ar pressure was 10 mTorr. The film deposited without applying substrate bias was deposited with 100 W sputtering power. For the one deposited with −200 V substrate bias, the sputtering power was 150 W. This power was chosen to keep the same overall deposition rate as the process without substrate bias. The x-ray diffraction spectra of these two samples are shown in Fig. 6. The (002) texture is dominant in the film deposited without applying substrate bias; however, the (110) texture is strong for the film deposited with a −200 V substrate bias. The substrate bias increases the kinetic energy of the bombarding particles (in this case, the Ar$^+$ ions), producing the (110) texture. This is consistent with our understanding of the texture dependence on Ar pressure, as discussed above. It is noted that applying substrate bias also raises the substrate temperature; however, as we have shown, a higher substrate temperature promotes the (002) texture. Hence, the formation of the (110) texture in this case is not due to the heating effect of applying substrate bias, but due to the large number of nucleation sites created by the Ar$^+$ bombardment.

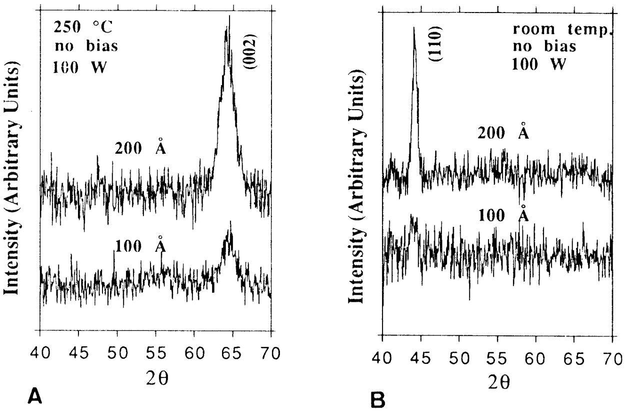

FIG. 7. The x-ray spectra of very thin Cr films: (A) substrate was preheated to 250 °C, no applied substrate bias, (B) substrate was not preheated, no applied substrate bias.

E. Initial stage of texture formation

In the model, the (002) texture originates from the island stage by mechanism I, hence, the (002) texture forms at the very beginning of the deposition, while the (110) texture develops after the islands impinge.

The initial stage of texture formation was investigated by using very thin Cr films. Two sets of samples were studied. Each set contains two samples which were deposited under the same conditions but are either 100 or 200 Å thick. They were all sputtered with 100 W forward power at 10 mTorr Ar pressure. Other sputtering parameters are specified in Fig. 7. The x-ray diffraction spectra of the first set of samples, which were deposited at 250 °C without applying substrate bias, are shown in Fig. 7(A). They both show the (002) peak and the intensities of the peaks are nearly proportional to the film thickness. This implies that the (002) texture is present from the initial stages (below 100 Å). However, as shown by Fig. 7(B), when the Cr film is deposited at room temperature, the (110) texture is very strong for the 200 Å film but it is not significant for the 100 Å Cr film. This shows that the (110) texture is formed during the later growth of the film, not at the beginning of the deposition.

IV. SUMMARY

In this article a model for the formation of crystallographic texture of sputter-deposited Cr thin film has been proposed. This model is based on the minimization of surface and interfacial energies. A set of systematic experiments has been carried to test this model. Consistent with the reported experimental results, when deposited at room temperature the Cr films on glass always have the (110) texture. Our new experimental findings include: (1) When deposited at elevated temperatures, the Cr films can have either the (002) texture or the (110) texture, depending on the sputtering power, Ar pressure, and/or substrate bias; the (110) texture is promoted by the bombardment energy of either deposited Cr atoms or Ar^+ ions; (2) the (002) texture can be initiated directly on the substrate surface, while the (110) texture appears not to form directly on substrate surface, but rather as a result of film growth.

These experimental results can be explained by our model, and hence serve to support the model. The model could be further tested by observing the shape of the initial islands directly with techniques such as atomic force microscopy and scanning tunneling microscopy. This is included in our plans for future research.

ACKNOWLEDGMENTS

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[1] J. Daval and D. Randet, IEEE Trans. Magn. **MAG-6**, 768 (1970).
[2] M. Ishikawa, N. Tani, T. Yamada, Y. Ota, K. Nakamura, and A. Itoh, IEEE Trans. Magn. **MAG-22**, 573 (1986).
[3] T. Ohno, Y. Shiroishi, S. Hishiyama, H. Suzuki, and Y. Matsuda, IEEE Trans. Magn. **MAG-23**, 2809 (1987).
[4] K. Hono, B. Wong, and D. E. Laughlin, J. Appl. Phys. **68**, 4734 (1990).
[5] H. J. Lee, J. Appl. Phys. **57**, 4037 (1985).
[6] S. L. Duan, J. O. Artman, B. Wong, and D. E. Laughlin, J. Appl. Phys. **67**, 4913 (1990).
[7] D. P. Ravipati, W. G. Haines, and J. L. Dockendorf, J. Vac. Sci. Technol. A **5**, 1968 (1987).
[8] J. Pressesky, S. Y. Lee, S. L. Duan, and D. Williams, J. Appl. Phys. **69**, 5163 (1991).
[9] M. A. Parker, K. E. Johnson, C. Hwang, and A. Bermea, IEEE Trans. Magn. **MAG-27**, 4730 (1991).
[10] W. L. Winterbottom, Acta Metall. **15**, 303 (1967).
[11] J. W. Cahn and J. Taylor, in Phase Transformations '87, 1988, p. 545.
[12] D. A. Porter and K. E. Easterling, *Phase Transformations in Metals and Alloys* (Van Nostrand Reinhold, New York, 1981), p. 112.
[13] L. Eckertova, *Physics of Thin Films* (Plenum, New York, 1986), p. 115.
[14] L. Eckertova, *Physics of Thin Films* (Plenum, New York, 1986), p. 130.
[15] T. W. Hickmott, Appl. Phys. Lett. **15**, 232 (1969).
[16] D. D. Maria, L. M. Ephrath, and D. R. Young, J. Appl. Phys. **50**, 4015 (1979).