Interfacial alloying and interfacial coupling in Cr/Fe(001)

M. Freyss, D. Stoeffler, and H. Dreyssé

Institut de Physique et de Chimie des Matériaux de Strasbourg, 23, rue du Loess, 67037 Strasbourg Cedex, France
(Received 27 March 1997; revised manuscript received 22 May 1997)

The magnetic order of Cr layers on Fe(001) is studied taking into account interfacial alloying and possible interdiffusion. The interfacial alloy is modeled by either a one-monolayer or a two-monolayer ordered compound whose concentration is varied. The spin-polarized electronic structure is determined self-consistently by solving a d-band tight-binding Hamiltonian. We determine the concentration for which the phase of the layer-by-layer antiferromagnetic structure of Cr changes in the Cr film on Fe(001). We find that in the case of two interfacial mixed layers, a π phase shift occurs at a Cr concentration between 33% and 50% when three monolayers (ML) of Cr are deposited. The occurrence of this π phase shift changes to a concentration between 11% and 25% for a more important coverage of Cr (namely 11 ML). When only one mixed layer is considered, the phase of the antiferromagnetic stacking of Cr changes at a concentration between 25% and 33% for 3 ML of Cr and between 33% and 50% for 11 ML of Cr. A simulation of the variation of the magnetization during Cr growth shows that the more Cr and Fe are interdiffused at the interface, the more important is the decrease of the magnetization. We compare our results to the many experimental data available. [S0163-1829(97)10234-X]

I. INTRODUCTION

The exchange coupling between Fe layers through a Cr spacer has been widely studied since the discovery of its oscillating behavior as a function of the Cr thickness. Even if the origin of the oscillation is now well understood, and explained in connection with the Fermi surface topology,[1,2] some discrepancies remain between experimental observations and theoretical results. For instance, even though the two-monolayer (ML) period of the oscillation observed experimentally[3,4] is in accordance with numerous calculations, its phase is exactly opposite to that theoretically predicted:[5,6] for an odd number of Cr layers, an antiferromagnetic (AFM) coupling is observed whereas calculations predict a ferromagnetic (FM) coupling, and inversely for an even number of Cr layers. The same phase shift is observed by Unguris *et al.*[3] in the layered antiferromagnetic structure of Cr on Fe(001): due to the strong antiferromagnetic interfacial coupling between Fe and Cr and the layered antiferromagnetic structure of Cr, one expects for an even (resp. odd) number of Cr layers deposited on Fe(001) the magnetic moments of the Cr overlayer to be positive (resp. negative). The experimental results show just the opposite. This discrepancy can be accounted for by the abrupt interface considered in theoretical studies. In experiments, such perfect interfaces do not exist due to roughness or interdiffusion. In particular, Venus and Heinrich[7] have recently shown by angular resolved Auger electron studies (ARAES) the existence of a strong interdiffusion of Cr atoms at the Cr/Fe(001) interface: at a growth temperature of 246 °C the Cr atoms penetrate into the second atomic layer (counting from the surface) and the concentration of Cr is found to decrease from the surface to the substrate. On the other hand, no interdiffusion is predicted at the Fe/Cr(001) interface.[8] Heinrich *et al.* thus suggest that the phase shift observed in the oscillating exchange coupling in Fe/Cr/Fe(001) systems is the result of interface alloying at the Cr/Fe(001) interface.

More recently, other evidence for alloying at the Cr/Fe(001) interface was found by Davies *et al.*[9] and Pfandzelter *et al.*[10] By means of scanning electron microscopy (STM), Davies *et al.* measured the concentration of Cr on the surface layer during its growth. They showed that after the deposition of 1 ML of Cr, there only remains 10% of Cr on the surface layer, indicating that most of the Cr atoms have penetrated into the Fe substrate. Furthermore, it is only after the deposition of 2–3 ML of Cr that the surface layer contains more Cr than Fe. The same trend was found by Pfandzelter *et al.*[10] by means of proton- and electron-induced Auger-electron spectroscopy. The Cr concentration profile they measured is, from the surface layer to the bulk: 45%, 55%, and 0% for 1 ML of Cr deposited, and 70%, 100%, and 30% for 2 ML of Cr deposited. These results are different from those of Venus *et al.*[7] who show a gradual decrease of the Cr concentration from the surface to the bulk.

More quantitative studies were made by Bayreuther and co-workers: they measured with an alternating gradient magnetometer (AGM) the variation of the total magnetization during the growth of Cr on Fe(001) substrates with different degrees of roughness. With a flat Fe surface, they observed a strong decrease in the magnetization of approximately $5\mu_B$ per interface atom.[11] However, such a big decrease has not been reproduced until now. The more important the roughness is, the smaller the decrease in the magnetization is:[12,13] on a highly stepped Fe(001) substrate, no variation at all of the total magnetization occurs during the Cr growth.

In order to understand the experimental results, we study the magnetism of thin Cr layers deposited on a semi-infinite Fe(001) substrate with either a two-monolayer or a one-monolayer ordered alloy at the interface to simulate interdiffusion. Interface alloying is expected to have an important influence on the magnetic structure of the deposited film, as has already been shown theoretically for Rh and Ru on Ag(001) by Turek *et al.*[14] In their study, they considered a completely disordered alloy. Here, we calculate the magnetic
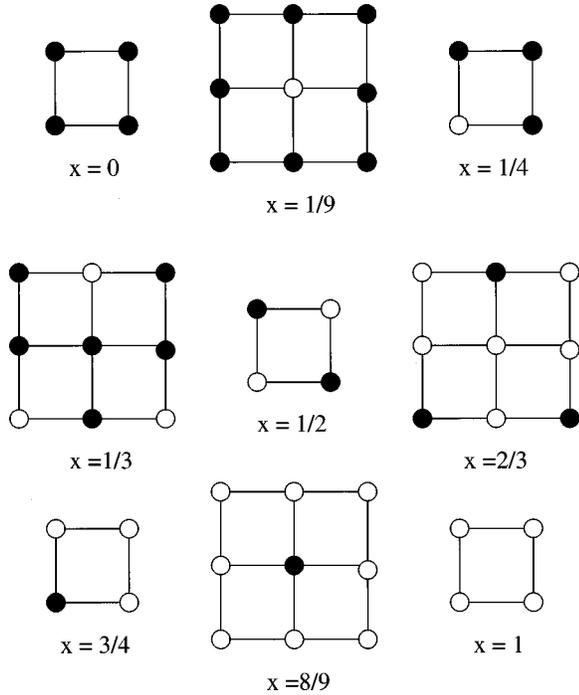
FIG. 1. Unit cells within the first interface mixed layer (counting from the Fe substrate). The white circles represent Cr atoms, the black ones Fe atoms. The unit cells of the second mixed layer is obtained by exchanging Fe and Cr atoms.

structure as a function of the concentration of an interfacial ordered alloy in the following situations:

$$Cr_n/Cr_{1-x}Fe_x/Cr_xFe_{1-x}/Fe(001) \quad (\text{case I}),$$

$$Cr_n/Cr_xFe_{1-x}/Fe(001) \quad (\text{case II}),$$

with $x = 0, \frac{1}{9}, \frac{1}{4}, \frac{1}{3}, \frac{1}{2}, \frac{2}{3}, \frac{3}{4}, \frac{8}{9}, 1$.

The coverage of Cr is varied from $n = 0$ to 2 ML. In some cases, the calculation is made up to $n = 10$ ML. The symmetry of the unit cell within the mixed interface layers is shown on Fig. 1 for the different concentrations considered. This unit cell is repeated by translation within the layer.

The aim of our study is to discuss the magnetic structure of the Cr film as a function of the alloy concentration x, and to determine for which concentration the experimentally observed phase in the layer-by-layer AFM structure of Cr can be obtained. The case of two interfacial mixed layers (case I) corresponds to an integer number of Cr layers deposited, namely $(n + 1)$, and can thus easily be compared to experimental results. The situation is different with one interfacial mixed layer (case II): as x increases, the quantity of Cr deposited varies from n monolayers (for $x = 0$) up to $(n + 1)$ monolayers (for $x = 1$).

D. Stoeffler et al.[15] already have considered the borderline case $x = 1$ in situation I. Such a case corresponds to the exchange of a complete monolayer of Cr and Fe at the interface. It was shown that such an exchange induces a phase shift in the AFM stacking of Cr: For an even number of Cr layers deposited the surface layer displays positive magnetic moments, as experimentally observed. The simulation of the variation of the total magnetization during Cr growth with

such an interface could reproduce very well the strong decrease observed by Turtur and Bayreuther.[11] Here, we consider a more general situation by introducing mixed Fe-Cr layers at the interface with different concentration and performing a systematical study. We also make a simulation of the variation of the total magnetization during Cr growth for various concentrations in case I.

In the following of the paper, we present in Sec. II the formalism used to compute the magnetic order. Then, in Sec. III we report the results obtained with two interfacial mixed layers (case I) and with one interfacial mixed layer (case II).

II. FORMALISM

The magnetic moments are calculated self-consistently by solving a d-band tight-binding Hamiltonian, and by means of the real-space recursive method. Only collinear magnetic orders are assumed. The advantage of the recursive method is that it does not require a symmetry in the direction perpendicular to the layers and enables thus to study systems with a low degree of symmetry.

In the basis of atomic orbitals $|im\sigma\rangle$ (i stands for the atomic site, m for the orbital symmetry, and $\sigma = \pm 1$ for the spin), the Hamiltonian is given by

$$H = \sum_{ii'mm'\sigma\sigma'} |im\sigma\rangle\langle i'm'\sigma'| \delta_{\sigma\sigma'} [\delta_{ii'}\delta_{mm'}\epsilon^o_{i\sigma m}$$

$$+ t^{im}_{i'm'}(1 - \delta_{ii'})] + \sum_{im\sigma} -\frac{1}{2}\sigma I_i M_i |im\sigma\rangle\langle im\sigma|. \quad (1)$$

The first term of H is the band term. $\epsilon^o_{i\sigma m}$ is the center of the $m\sigma$ band on the site i and $t^{im}_{i'm'}$ the intersite spin-independent hopping integrals. The second term of H is the exchange term which accounts for magnetism. I_i is the effective exchange integral and M_i the local magnetic moment on the site i. The parameters $t^{im}_{i'm'}$ and I_i were chosen to reproduce ab initio calculations with the FLAPW (full potential augmented plane waves) method. They have already been used satisfactorily in previous studies.[5,16]

Self-consistency is obtained by requiring that the band occupation on each site is equal to the bulk value N^o_i (local neutrality approximation), and that the exchange splitting $\epsilon^-_i - \epsilon^+_i$ is equal to $I_i M_i$:

$$N^+_i + N^-_i = N^o_i, \quad (2)$$

$$\epsilon^-_i - \epsilon^+_i = I_i M_i = I_i(N^+_i - N^-_i). \quad (3)$$

The energy levels ϵ^-_i and ϵ^+_i are thus determined so that both equations are verified: Equations (2) and (3) are solved for a given site i, keeping the energy levels of all other sites fixed, and this process is repeated for all sites until the self-consistency relations are satisfied. The band occupation is obtained by integration of the density of states up to the Fermi level. The density of states is calculated by means of the real-space recursive method with eight levels of the continuous fraction and using the Beer-Pettifor terminator.

The calculation of the magnetic moments is self-consistently made on all inequivalent atoms of the Cr film, on all inequivalent atoms of the mixed layers, and on the Fe

atoms on 10 layers below the interface. The moments of the rest of the semi-infinite Fe substrate are frozen to the bulk value.

The results are then discussed in term of the relative energy between the different magnetic solutions obtained for a given atomic configuration. The absence of repulsive term in the Hamiltonian can be justified by considering it independent of the magnetism. Its contribution to the total energy thus disappears when calculating the relative energy between the different solutions. Energy calculations with such a model has already proven to be satisfactory in a previous study of the Fe/Cr interlayer magnetic coupling.[17]

It is also to be noted that our study will only give a qualitative understanding of the effect of interdiffusion since the interfacial alloy is approximated by an ordered compound. A more precise treatment of the alloy would require the use of the coherent potential approximation (CPA) in the model.

III. RESULTS

For a given value of x, the number of magnetic configurations obtained by our self-consistent calculation can be very important when one or no pure Cr layer is considered ($n=0$ or $n=1$). These different magnetic configurations are obtained by choosing a different initial magnetic configuration in the self-consistent process. On the other hand, the number of configurations obtained with two pure Cr layers ($n=2$) or more reduces to one or two, whatever the value of x is. In that case, the solutions always present one of the two possible antiferromagnetic layer-by-layer structure for the pure Cr film. In the following, first the case with two mixed layers is discussed, then the case with one mixed layer.

A. $Cr_n/Cr_{1-x}Fe_x/Cr_xFe_{1-x}/Fe(001)$

As a example of multiple solutions obtained for a given value of x, Fig. 2 shows the solutions for $x=1/2$ and $n=0, 1$, and 2. In the presence of the mixed layers, the antiferromagnetic coupling between Cr and Fe first neighbors on the one hand, and Cr first neighbors pairs on the other hand cannot always be satisfied and magnetic frustrations are induced. It is probably due to such frustrations that a very large number of magnetic configurations can be obtained for a given value of x.

With $n=0$ [(Fig. 2(a)], in the ground state situation the Cr atoms of the lower mixed layer are frustrated with Fe and have an almost vanishing magnitude which stabilizes this configuration relatively to the second solution obtained. Indeed, in the second solution, the Cr atoms of the surface layer are frustrated with the Fe atoms of the plane below. And as the frustrated Cr atoms are at the surface, they have an enhanced magnetic moment. Thus the magnetic frustration cost more energy than in the case of small frustrated Cr atoms below the surface. The magnetic structure at the interface differs a lot when more Cr is added on top as shown in Figs. 2(b) and 2(c).

With one pure Cr monolayer [$n=1$, Fig. 2(b)], it is the metastable interfacial configuration obtained with $n=0$ that is displayed in the ground state and the Cr overlayer is ferromagnetic with negative moments. In this situation, the frustrations occur between the Fe atoms and the Cr atoms of the lower and upper mixed layers, that is below the surface.

In the two metastable solutions, the Cr overlayer displays a $c(2\times2)$ magnetic order, which induces frustations with the Fe atoms of the layer right below. Again, the origin of the unstability of these solutions can be attributed to the localization at the surface of the frustrations.

With two pure Cr layers [$n=2$, Fig. 2(c)], we find the two possible layer-by-layer antiferromagnetic structure of the pure Cr film. In the ground state, no Cr pair is frustated as the Cr atoms are antiferromagnetically coupled from one plane to the other. The only frustrations are between the Fe and the Cr atoms of the two mixed layers. The surface layer displays positive magnetic moments. On the other hand, in the metastable solution, there are more frustrations: between the Cr atoms of the first pure Cr layer and their Fe neighbors, and between the Cr atoms of the lower and the upper mixed layers. In this case, the surface layer displays negative magnetic moments, meaning that the phase of the AFM stacking of the pure Cr film is opposed to that of the ground state situation.

Figure 3 shows the results concerning the phase of the antiferromagnetic stacking of Cr as a function of the alloy concentration with 3 ML and 11 ML of Cr deposited ($n=2$ and $n=10$). As previously said, the phase of the stacking can be charaterized by the sign of the magnetic moments on the surface layer. Accordingly, Fig. 3 shows the average value of the surface layer magnetic moments as a function of the interfacial alloy concentration x. A clear trend can be noticed: With 3 ML of Cr, negative surface layer moments are obtained when x is smaller than 1/2. The same sign is also obtained with a perfect interface (i.e., no interdiffusion). On the other hand, positive moments are obtained when x is larger or equal to 1/2, that is to say when there are more Cr than Fe atoms on the first mixed layer (counting from the substrate), and more Fe than Cr atoms on the second mixed layer. This latter situation displays the phase experimentally observed in the AFM stacking of Cr. The magnetic structure of Cr on Fe(001) can thus be accounted for by a partial exchange of a monolayer of Cr and Fe at the interface. We can also see on Fig. 3 that the value of the surface layer magnetization is almost constant as a function of the alloy concentration x.

It is to be noted that when $x=1/9$, that is when the alloying is very weak and gradual from the Fe substrate to the Cr film, only one solution could be found. When other configurations with the other phase in the AFM stacking of Cr were introduced as the initial configuration in the self-consistent process, the phase would shift during the calculation, and the calculation would converge to the unique solution. This observation can be related to a result obtained by Stoeffler[18,16] which showed that with a perfect interface ($x=0$), no frustated configuration could be obtained in the AFM stacking of Cr when the thickness of the film was smaller than 6 ML. This is apparently also the case when the alloying is weak and the Cr thickness is small.

The phase shift in the magnetic structure of Cr between $x=1/3$ and $x=1/2$ occurs in the following way: For $x=1/3$ the Fe atoms in the first mixed layer, in larger amount than Cr, impose negative moments on Cr in the second mixed layer due to the strong AFM coupling between Fe and Cr. Then, those negative Cr moments impose positive moments on the pure Cr layer above, inducing thus no phase change in the magnetic structure of the Cr film compared to the case
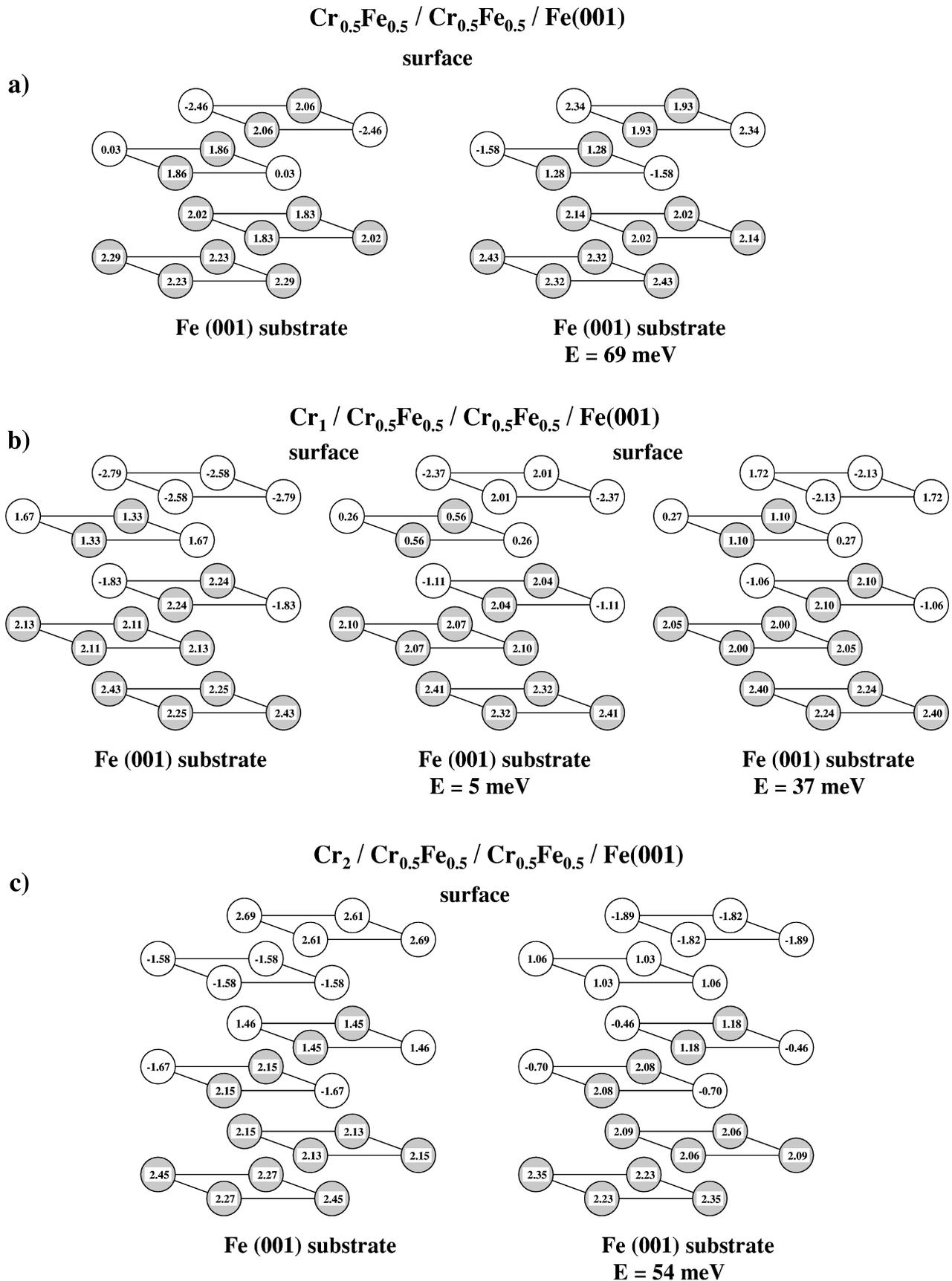
$Cr_{0.5}Fe_{0.5}$ / $Cr_{0.5}Fe_{0.5}$ / Fe(001)

surface

a)

Fe (001) substrate

Fe (001) substrate
E = 69 meV

Cr_1 / $Cr_{0.5}Fe_{0.5}$ / $Cr_{0.5}Fe_{0.5}$ / Fe(001)

b)

surface   **surface**

Fe (001) substrate

Fe (001) substrate
E = 5 meV

Fe (001) substrate
E = 37 meV

Cr_2 / $Cr_{0.5}Fe_{0.5}$ / $Cr_{0.5}Fe_{0.5}$ / Fe(001)

c)

surface

Fe (001) substrate

Fe (001) substrate
E = 54 meV

FIG. 2. Ground state and metastable solutions for $Cr_n/Cr_{1/2}Fe_{1/2}/Cr_{1/2}Fe_{1/2}/Fe(001)$ with (a) $n=0$, (b) $n=1$, and (c) $n=2$. The energy indicated is the interfacial energy relative to the ground state. Only the unit cell of each layer is shown, as well as only two layers of the semi-infinite Fe substrate.

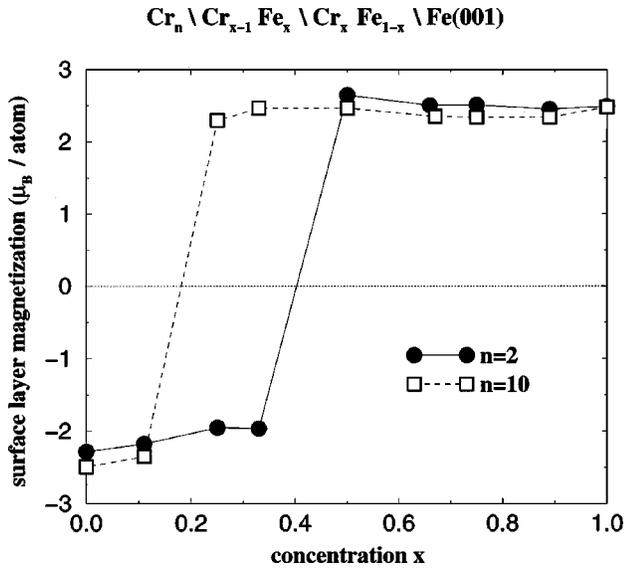$Cr_n \setminus Cr_{x-1} Fe_x \setminus Cr_x Fe_{1-x} \setminus Fe(001)$

FIG. 3. Value of the surface layer average magnetic moment as a function of the concentration x when 3 Cr layers (filled circles) and 11 Cr layers (squares) are deposited and with two interfacial mixed layers. The lines are guides for the eyes. The sign of the surface layer magnetic moments characterizes the phase of the antiferromagnetic layer-by-layer structure of Cr.

with a perfect interface. For $x=1/2$ [see Fig. 2(c)], the Fe atoms of the second mixed layer impose negative moments on the first pure Cr layer, inducing thus the π phase shift in the layer-by-layer AFM structure of Cr.

Calculations have also been perfomed with a bigger thickness of Cr, 11 ML, to verify if the Cr magnetic stacking is not changing as the thickness increases. Surprisingly, we have obtained that the phase shift occurs for a smaller concentration x compared to the situation with 3 ML of Cr. The phase shift now occurs between $x=1/9$ and $x=1/4$ as can be seen on Fig. 3. In this case, a small interdiffusion is enough to reverse the magnetization of the layers of the Cr film.

As a consequence, in Fe/Cr/Fe(001) trilayers, if interdiffusion is assumed on both interface in the range of concentration $x \geq 1/4$, we find that the coupling between Fe layers is the same as in the case of perfect interfaces (ferromagnetic for an odd number of Cr layers, and antiferromagnetic for an even one), as shown on Fig. 4. And if only one interface is interdiffused, namely the Cr/Fe(001) interface as it is assumed by experimentalists, the coupling between the Fe layers is reversed, and the coupling experimentally observed is obtained: ferromagnetic for an even number of Cr layers, and antiferromagnetic for an odd one.

The fact that the phase shift occurs for a smaller concentration with 11 ML of Cr than with 3 ML also means that for the concentrations $x=1/4$ and $x=1/3$ a change occurs in the magnetic structure of the alloy layers as the Cr coverage increases. This change for $x=1/4$ occurs at a coverage of 2 to 5 ML of Cr. In this range of coverage, the surface Cr moments are always negative. The alternating sign of the surface layer moments as a function of the coverage starts only after 5 ML of Cr deposited. As a consequence, as Fe and Cr couple antiferromagnetically, if the surface of Cr has negative moments, when covering it with Fe, Fe would then be ferromagnetically coupled to the lower Fe layer. Such a
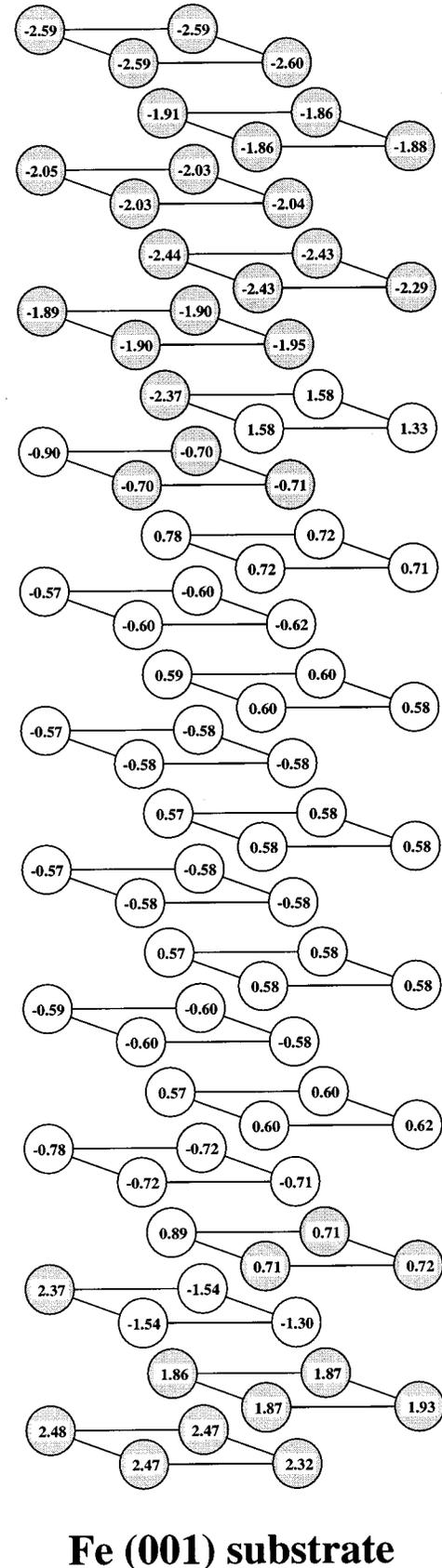
Fe (001) substrate

FIG. 4. Ground state for a $Fe_5/Cr_{12}/Fe(001)$ trilayer with two interdiffused interfaces. Only the unit cell of each layer is shown, as well as only two layers of the semi-infinite Fe substrate.
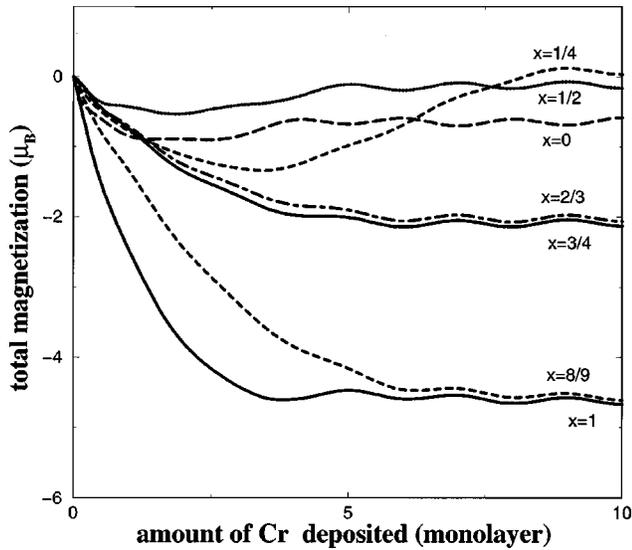
FIG. 5. Simulation of the variation of the total magnetization during Cr growth for different interfacial alloy concentrations x.

result could explain the ferromagnetic interlayer coupling between Fe layers observed up to 4–5 ML of Cr in the trilayers Fe/Cr/Fe(001).[3]

Figure 5 shows the simulation of the variation of the total magnetization during Cr growth with various concentrations. The model used is presented in the Appendix. Let us recall that we have supposed an important non-layer-by-layer growth mode. A trend can be noticed on the figure: in the first stage of the growth (coverage smaller than 3 ML), the magnetization decreases whatever the value of x is. For larger coverage, the magnetization continues to decrease up to an asymptotic value when x is larger than 1/2. And the more x is large the more the decrease in the magnetization is large: almost $5\mu_B$ when x equals 8/9 and 1. The decrease of the magnetization for these concentrations is related to the strong reduction of the Fe moments of the upper mixed layer. Indeed, in this range of concentrations, those Fe moments are of the order of $1\mu_B$ or much less. On the other hand, when x is smaller or equal to 1/2, the magnetization increases for coverage larger than 3 ML and then stabilizes to an asympotic value. The increase is especially important for x equal to 1/4. This can be explained by the fact that $x=1/4$ is the concentration for which changes occur in the magnetic structure within the mixed layers with increasing Cr thickness, as previously mentionned. The consequence is that the magnetization increases when the coverage of Cr is larger than 3 ML. It can also be noticed that the curves for $x=2/3$ and $x=3/4$ on the one hand, and the curves for $x=8/9$ and $x=1$ on the other hand tend to the same asympotic values. This can be attributed to the fact that for these concentrations, the values of the average Fe moments of the upper mixed layer are almost the same, namely about $0.75\mu_B$ for both $x=2/3$ and 3/4, and about $-0.5\mu_B$ for both $x=8/9$ and $x=1$.

This simulation can be compared to the experimental results of Bayreuther and co-workers. During the deposition of Cr on a flat Fe surface, they show a very important decrease of the magnetization of approximately $5\mu_B$.[11] These data are in accordance with our results in the case of an alloy con-

centration x larger than 3/4. As the Fe surface gets rougher, they find a decrease of the magnetization less important:[12,13] only $1.4\mu_B$ with an intermediate step density, and even zero μ_B with a highly stepped surface. Even if the structure of the interface of their samples is not precisely known, those latter results would be consistent with alloy concentrations x of around 2/3–3/4 and 1/4–1/2, respectively. By our electronic structure calculations we can thus give an estimation of the chemical structure at the interface of the samples.

B. $Cr_n/Cr_xFe_{1-x}/Fe(001)$

We have also calculated the magnetic order of Cr films on Fe(001) with only one interfacial mixed layer: $Cr_n/Cr_xFe_{1-x}/Fe(001)$ (case II) with n equal to 0, 1, 2 and in some cases 10 ML.

With $n=0$, that is with a mixed layer on top of the pure Fe substrate, all Cr moments are antiferromagnetically coupled to the Fe moments of the substrate and of the mixed layer. There is no magnetic frustation in the ground state. The values of the surface layer moments are almost constant as a function of x, especially the Cr ones. The values of the surface Cr moments are of the order of $3.33\pm0.05\mu_B$ and the value of the surface Fe moments are of the order of $2.65\pm0.11\mu_B$.

With one pure Cr layer added on top of the mixed layer ($n=1$), the ground state changes according to the value of x. First, for x in the range of 8/9 to 2/3 included, the Cr moments of the mixed layer are all always antiferromagnetically coupled to the Fe atoms, and the Cr moments of the pure surface layer are all positive, that is antiferromagnetically coupled to the Cr moments of the mixed layer below. The number of Fe-Cr first-neighbor bindings being for these concentrations smaller than the number of Cr-Cr first-neighbor bindings, the layer-by-layer antiferromagnetic structure of Cr prevails on the Fe-Cr antiferromagnetic coupling. As a consequence of the magnetic frustrations between Fe and Cr, the Fe magnetic moments on the mixed layer are reduced and smaller than $1\mu_B$. When x is larger or equal to 1/2, the number of Fe-Cr first-neighbor bindings is no longer smaller than the Cr-Cr ones. A different configuration thus arises for the ground state. Except for $x=1/9$, the ground state displays the following features: in the mixed layer, the Cr moments are antiferromagnetically coupled to Fe, as in the previous cases. The difference is in the pure Cr layer, which does not display an in-plane ferromagnetic order anymore: there are Cr moments with different signs within the pure layer, forming complex magnetic configurations. The ground state for $x=1/9$ is particular as the Cr moment of the mixed layer is positive, that is ferromagnetically coupled to the Fe substrate, contrary to the configuration found for all other concentrations. The pure Cr layer also displays a complex magnetic structure with moments of different signs.

With $n=2$ (two pure Cr layers), the situation is more simple: only solutions corresponding to a layer-by-layer antiferromagnetic structure of the pure Cr film could be found. The phase of the Cr magnetic stacking in the ground state, given in Fig. 6, shows the variation of the surface layer magnetization as a function of x with 2 and 10 pure Cr layers deposited. With 2 pure Cr layers, we see that the antiferromagnetic stacking of the Cr film changes when x is larger
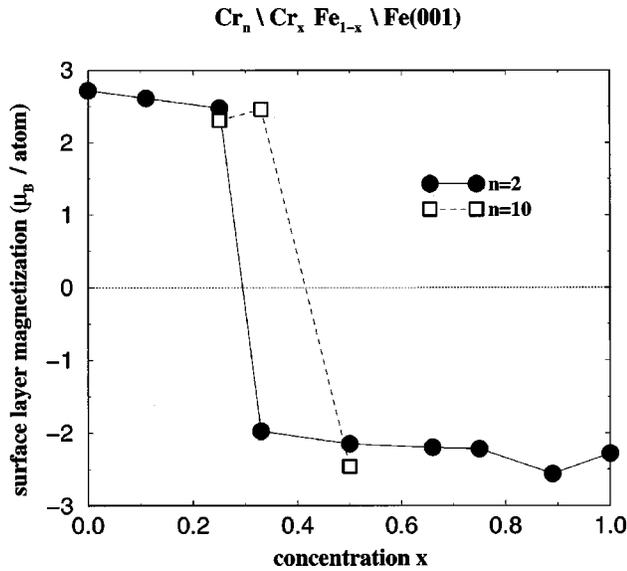
$Cr_n \setminus Cr_x\, Fe_{1-x} \setminus Fe(001)$

FIG. 6. Value of the surface layer average magnetic moment as a function of the concentration x when $n = 2$ ML (filled circles) and $n = 10$ ML (squares), and with one interfacial mixed layer. The lines are guides for the eyes. The sign of the surface layer magnetic moments characterizes the phase of the antiferromagnetic layer-by-layer structure of Cr.

than 1/4. This is quite surprising because it would have been expected that the transition occurs rather around $x = 1/2$, that is to say that the first pure Cr layer would have been expected to couple antiferromagnetically to the Fe atoms of the mixed layer as long as the Fe atoms are in majority in the mixed layer. This is actually not the case as shown in Fig. 7, which represents the ground state obtained for $x = 1/4$ and 1/3, i.e., the concentrations between which the transition occurs. We see that for $x = 1/4$, the first pure Cr layer is antiferromagnetically coupled to the Fe substrate. On the other hand, for $x = 1/3$ the first pure Cr layer is coupled ferromagnetically to Fe, leading to slightly smaller Cr moments on this layer.

On Fig. 7, one can also see that the value of the magnetic moments of both Fe and Cr of the mixed layer vary with x: For Fe, they are of order of $1.5-1.7\mu_B$ when $x = 1/3$ and 1.9 μ_B when $x = 1/4$. For Cr, they are of order of $-1.2\mu_B$ when $x = 1/3$ and $0.9\mu_B$ when $x = 1/4$. This result is in contradiction with the results obtained by Coehoorn:[19] by means of the augmented spherical waves (ASW) method, he computed the magnetic moments in superlattices of the type 5 ML Fe/1 ML Fe_xCr_{1-x}/5 ML Cr/1 ML $Fe_{1-x}Cr_x$ with $x = 0$, 1/4, 1/2, 3/4, and 7/8, the alloy layer being modeled as an ordered compound as well. The value of the moments were found approximately constant whatever the value of x, and of order of $2.0\mu_B$ for Fe and $-0.5\mu_B$ for Cr.

We have also considered the cases $x = 1/3$, 1/4, and 1/2 with 10 layers of pure Cr ($n = 10$) in order to see if, as in the
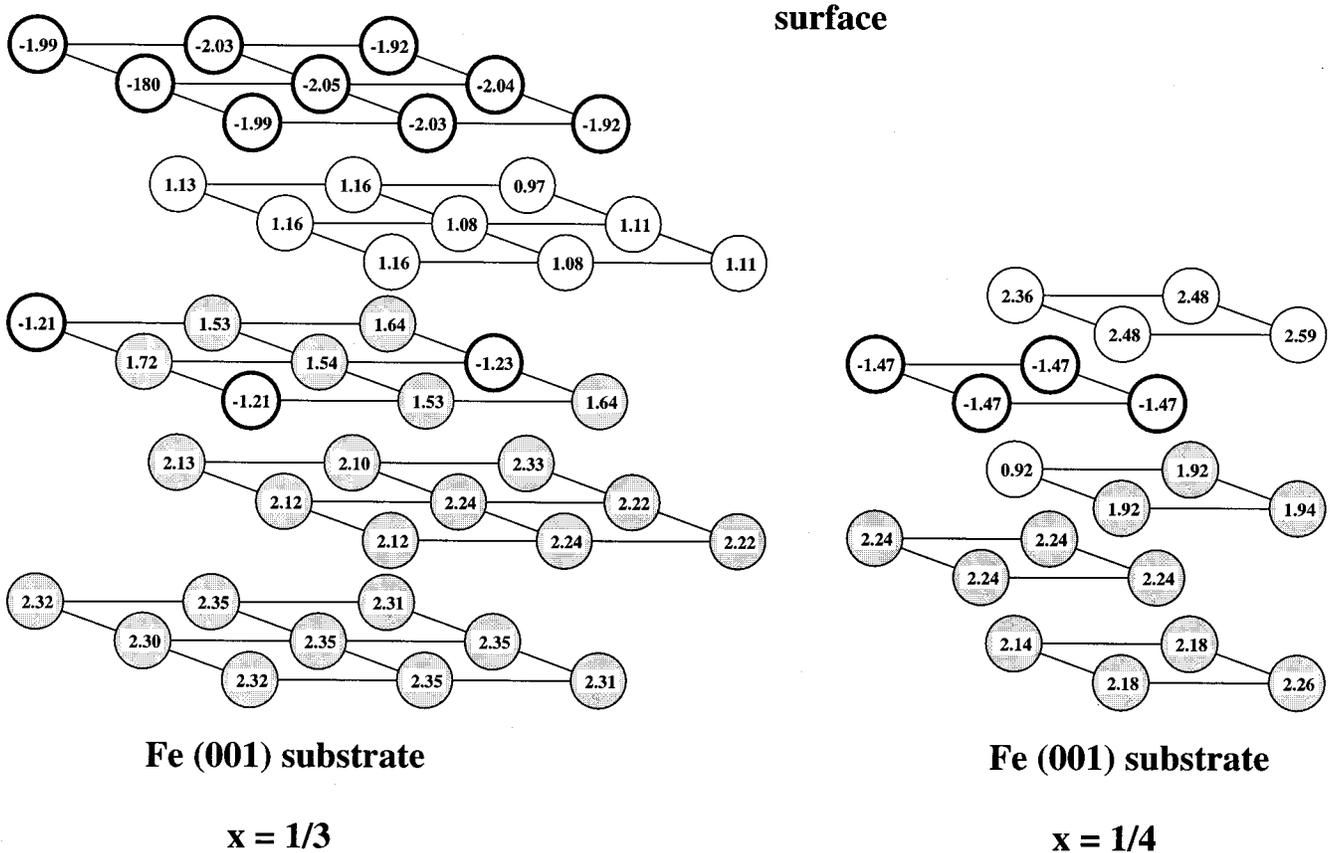
surface

Fe (001) substrate

x = 1/3

Fe (001) substrate

x = 1/4

FIG. 7. Ground state for $Cr_2/Cr_{1/3}Fe_{2/3}/Fe(001)$ and $Cr_2/Cr_{1/4}Fe_{3/4}/Fe(001)$. The white circles represent Cr atoms, the grey ones Fe atoms. A distinction is made between positive and negative Cr magnetic moments by means of the linewidth of the circles. In the case $x = 1/3$, the vertical scale has been expanded. Only the unit cell of each layer is shown, as well as only two layers of the semi-infinite Fe substrate.

previous case with two interfacial mixed layers, the transition occurs for a different concentration when the coverage of Cr increases. It is indeed the case as can be seen on Fig. 6: The antiferromagnetic structure of Cr changes when x is larger than 1/3, that is for a slightly larger value compared to the case with three monolayers of Cr deposited. Our results concerning the reversal of the sign of the magnetic moments in the pure Cr film are in good agreement with those of Coehoorn:[19] he found the reversal in the sign of the Cr magnetic moments in his 5 ML thick pure film for x between 0 and 1/4, which is close to our value of x obtained with a 2 ML thick pure Cr film (x between 1/4 and 1/3).

IV. CONCLUSION

We have calculated the magnetic moments of Cr films on a Fe(001) substrate with mixed layers at the interface and shown how the concentration of these mixed layers affects the magnetic order of the Cr film. We have shown that interdiffusion could account for experimental results: with two interfacial mixed layers, the exchange of one quarter of a monolayer of Fe and Cr is enough to reverse the layer-by-layer antiferromagnetic structure of a 10 ML thick Cr film.

Which concentration profile accurately describes the Cr growth on Fe(001) is highly dependent on the growth conditions. This is reflected by the various concentration profiles experimentally observed by the different groups[7,9,10] and by the various results obtained by Bayreuther *et al.*[11–13] Our simulation of the variation of the magnetization during Cr growth can give an estimation of the degree of interfacial interdiffusion.

In our calculations, the magnetic order is restricted to a collinear order. It is highly probable that a noncollinear order is more stable, at least near the interface, in order to minimize the effect of the magnetic frustrations induced by the interdiffusion. Unfortunately, calculations of the magnetic order with noncollinear moments in nonperfect systems still require too large a computing time.[20]

APPENDIX: SIMULATION OF THE VARIATION OF MAGNETIZATION DURING GROWTH

The variation of magnetization during Cr growth is simulated using a simple macroscopic model of growth. At time t, the variation of magnetization $M(t)$ is given by

$$M(t) = \sum_{n=1}^{\infty} \Omega_n (\theta_n(t) - \theta_{n+1}(t)),$$

where n is the index of the Cr layers deposited. $\theta_n(t)$ is the Cr coverage on the nth layer at time t [see Fig. 8(a)]. Ω_n is the difference of magnetization between n Cr layers deposited on Fe(001) (with two mixed layers at the interface) and the pure Fe(001) surface:
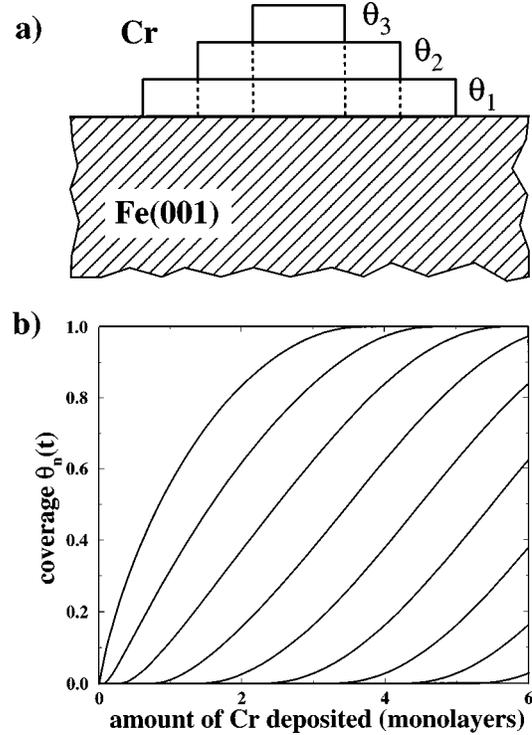

FIG. 8. (a) Macroscopic growth model of Cr on the Fe(001) substrate. θ_n is the Cr coverage on the nth layer. (b) Cr coverage $\theta_n(t)$ on the surface as a function of the amount of Cr deposited $\Sigma_n \theta_n(t)$ (in monolayers). The different curves correspond to the filling of the successive layers.

$$\Omega_n = \sum_{i=-9}^{n} M_i^t (Cr_{n-1}/Cr_{1-x}Fe_x/Cr_xFe_{1-x}/Fe(001)) - \sum_{i=-9}^{n} M_i^t (Fe(001)),$$

where M_i^t is the average magnetic moment of the ith layer. Nine layers of Fe below the interface are taken into account in the calculation of the difference of magnetization.

The most important thing is the simulation of $\theta_n(t)$, that is the growth mode of Cr. Here, we model $\theta_n(t)$ in the following way:

$$\theta_n(t) = \begin{cases} 0 & \text{for } t \leq (n-1)\delta \\ \dfrac{1 - \cos[\pi(\alpha t - (n-1)\delta)]}{2} & \text{otherwise.} \end{cases}$$

When $\theta_n(t)$ is equal to 0, no Cr is present on layer n, and when $\theta_n(t)$ is equal to 1, the nth layer is complete. In our theoretical study, the notion of time is arbitrary. A more relevant quantity which is equivalent to the time is the amount of matter deposited. It is defined as $\Sigma_n \theta_n(t)$. In the model, the only parameters to choose are δ and α. The parameter δ is the ''time'' between the beginning of two successive layers. When δ is 1, a layer starts once the previous one is complete, corresponding thus to a perfect layer-by-

layer growth. When δ is inferior to 1, the growth mode corresponds to the formation of islands. In our simulation, instead of setting δ, we set an equivalent quantity, $\theta_1(\delta)$, which is the amount of Cr deposited on the first layer when the second layer starts. We choose $\theta_1(\delta) = 0.06$ and $\alpha = 0.06$. Figure 8(b) shows the behavior of $\theta_n(t)$ in the simulation. With the parameters chosen, the seventh layer starts when the first one is completed.

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