Suppression of silicide formation in Fe films grown on Si(001)

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Thin ferromagnetic Fe films were grown at both room and low temperatures (150 K) on (2×1) and on Au-passivated Si(001). In the case of Fe grown on (2×1) Si(001), the magnetization sets in at an Fe coverage of 3.6 monolayers (ML) at room temperature and of 2.3 ML at 150 K, indicating a reduced magnetization due to silicide formation. To suppress Fe–Si reactions we used a 2 ML thick Au buffer layer deposited in two steps. We performed a reconstruction by the deposition of about 0.5 ML of Au at ~1000 K followed by the growth of another 1.5 ML at room temperature or at 150 K. Room temperature growth results in rough Au films and no earlier onset of magnetization in comparison to the case of unbuffered Fe films. However, a strong out-of-plane magnetization is measured at the onset coverage. By contrast, at 150 K, smooth Au buffer films are observed, acting as protective layers against Fe and Si intermixing. Growth on these buffer layers results in an early onset of the out-of-plane magnetization at 1.4 ML of Fe and a larger Kerr signal. Both these findings point at a significant reduction of silicide formation. A 2.3 ML thick Fe film grown on Au passivated Si(001) shows a reversible magnetic behavior upon annealing to 280 K and cooling back to 150 K, indicating a thermally stable system. © *2000 American Institute of Physics.*
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I. INTRODUCTION

The widespread use of Si in semiconductor technology together with new trends in microelectronics raises the question whether Si is able to cope with the challenges posed by the emerging field of spin electronics. In this respect, the investigation of thin metallic films grown on semiconductors and their interfaces is of special interest. Therefore, studies of the reaction of Fe grown on Si(001)[1,2] have been done, special attention being paid to the investigation of direct gap[3] semiconducting iron disilicide[4–6] for its possible use as near infrared light sources and detectors. In the case of Fe grown at room temperature (RT) on Si(001) by molecular beam epitaxy, it was found[7] that the Fe/Si interface is far from being sharp, and an amorphous phase with the stoichiometry close to Fe_3Si starts to form after ~1 monolayer (ML) of Fe is deposited. The reaction slows down above 5 ML,[7] to end abruptly at 10 ML,[8] with the beginning of body-centered-cubic (bcc) Fe formation. A long-range in-plane ferromagnetic order in the silicide films was shown[8,9] to set in after the deposition of 4 ML of Fe at RT, with a reduced magnetic moment[8] in comparison to bcc Fe.

The amorphous silicide at the interface,[7,8] strongly related to the existence of a Schottky barrier at the Fe/Si interface, will act as a strong scatterer for electrons passing through the interface, potentially reducing the spin information. To avoid this, a thin, stable and spin-preserving passivation layer of the semiconductor surface is highly desirable in order to reduce intermixing at the Fe–semiconductor interface. On the other hand, S, Se, or B passivation used in previous studies[10,11] give rise to a rougher surface and it is not clearly established to which extent these can prevent silicide formation. A recent investigation of Fe films grown on S-passivated Ge (Ref. 12) shows no reduction in the magnetization onset coverage and brings evidence for S migration to the surface during deposition. The segregation of the passivation agent on top of the film may reduce the passivation and contaminate the Fe film. An alternative passivation was achieved by the growth of ~10 ML thick $CoSi_2$ film on Si(001) prior to the Fe deposition[13] reducing Si migration and favoring the growth of a good epitaxial bcc Fe film.

In view of recent studies,[14,15] Au seems to be appropriate at least for the relatively large attenuation length ($\lambda > 230$ Å) for ballistic electron transport in Au grown on Si(001).[15] Besides, the possibility of suppressing the spin–flip scattering for tunneling electrons through an Fe/Au multilayer base fabricated onto an *n*-type Si(001) collector,[14] makes Au the favorite candidate for our purpose. For these reasons, the thermodynamically stable Au-induced reconstruction of Si(001) surface[16,17] has been used to reduce silicide formation at the Fe/Si interface.

II. EXPERIMENT

The Si(001) substrates were cut from commercially available single crystal wafers, mounted on nonmagnetic sample holders and inserted into the vacuum chamber via a load lock system. The base pressure in the preparation chamber was below 1.0×10^{-10} mbar. The samples were first degassed by direct heating up to ~850 K, and flashed afterwards at increasing current values to remove the native oxide layer. During both degassing and flashing, the pressure was kept below 5.0×10^{-9} mbar. The clean crystals showed no traces of contamination in the Auger spectra, and very sharp (2×1) low energy electron diffraction (LEED) patterns. The crystals were considered clean only when the dimer rows of

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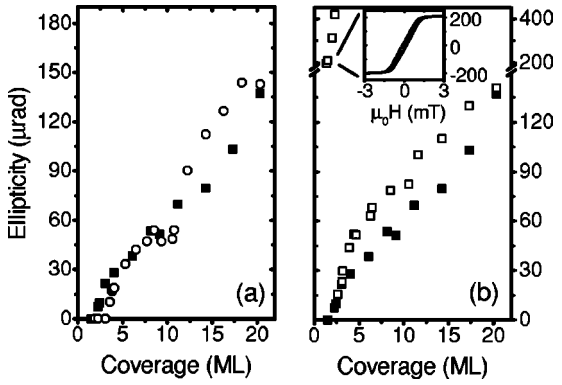

FIG. 1. MOKE ellipticity in longitudinal geometry at saturation vs Fe coverage: (a) films grown on (2×1) Si(001) at room temperature (open circles) and 150 K (solid squares); (b) films grown at 150 K on Au-covered Si(001) (open squares; the points above 200 μrad correspond to the out-of-plane magnetization) and on (2×1) Si(001) [solid squares — same data as in (a)]. The inset in (b) shows the out-of-plane hysteresis loop at the magnetization onset (1.4 ML of Fe) in the film grown at 150 K on Au-covered Si(001).

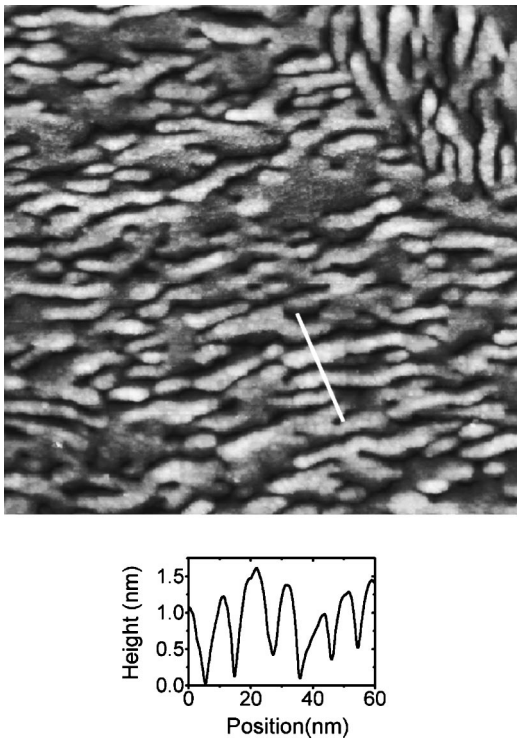

FIG. 2. A 200×200 nm^2 STM topograph of a 20.4 ML thick Fe film grown at room temperature on Si(001). The image is taken at a bias voltage of 0.8 V and a tunneling current of 0.1 nA. A line profile taken along the white line shown on the STM image is plotted underneath.

the (2×1) reconstruction were clearly visible in scanning tunneling microscope (STM) topographs. Henceforth, it will be understood that all of the STM scans were performed *in situ* at RT, regardless of the deposition temperature. The films were grown at both RT and low temperature (LT≅150 K) on (2×1) Si(001) single crystal substrates by molecular beam epitaxy at a base pressure below 1.0×10^{-10} mbar. The Au-induced reconstruction of Si(001)[16,17] was performed by depositing less than 1 ML of Au at temperatures around 1000 K. The ML Au and Fe coverages are defined in terms of atomic densities of face-centered cubic (fcc) Au(001) and bcc Fe(001) layers, respectively. The quality of the reconstruction was checked *in situ* with both LEED and STM. Different overall topographs were obtained according to the temperature employed for performing the reconstruction. The magnetization of the Fe films was probed *in situ* at the growth temperature, by the magneto-optic Kerr effect (MOKE) in the longitudinal geometry. For some very thin films showing perpendicular magnetization, also polar MOKE loops were taken. As a light source we used a commercial multimode laser diode with the wavelength of 670 nm. The angle of incidence was approximately 50°.

III. RESULTS AND DISCUSSION

A. Fe grown on (2×1) Si(001)

It has been shown that the deposition of several monolayers of Fe on Si(001) at RT results in the formation of an amorphous ferromagnetic iron silicide,[8] with an in-plane oriented magnetic moment, whose composition is close to Fe_3Si.[7] No LEED pattern was detected even for submonolayer coverage, in agreement with previous works.[2–8] The in-plane onset of magnetization at RT occurred after no less than 3.6 ML of Fe were deposited, as we found by MOKE. By increasing the amount of deposited Fe, a smoothly increasing MOKE signal is detected [the open circles in Fig. 1(a): MOKE ellipticity at saturation], except for Fe coverages between 9 and 12 ML. Recent spin-integrated and spin-resolved photoemission studies[8] showed that the silicide formation ends abruptly at 10 ML where we observed a kink in the MOKE signal. We relate this kink to changes in magnetic moment, structure and/or chemical modifications at the growth front. A rough film is obtained by RT growth and the typical silicide morphology consisting of elongated islands is preserved at least up to 20.4 ML of Fe, as shown in the 200×200 nm^2 STM scan in Fig. 2. We correlate the late onset of magnetization observed in RT grown films to the weak ferromagnetism of the amorphous silicide film and to an intermixed interface.

To reduce the thermally activated silicide formation, we reduced the growth temperature to 150 K. This induces an earlier onset of in-plane magnetization at 2.3 ML as shown in Fig. 1(a) (solid squares). We did not observe a perpendicular orientation of the magnetic moment, as reported for Fe grown at 100 K on Si(001) (Ref. 9) probably because the growth temperature in our experiments was higher. The early onset of magnetization we observed at 150 K might be due to the low growth temperature, or to the low measuring temperature. Since a 2.3 ML thick Fe film grown at RT shows no magnetic signal at both RT and 150 K, we come to the conclusion that the low growth temperature reducing intermixing is responsible for the early onset of magnetization. Additionally, the kink at about 10 ML of Fe in the MOKE ellipticity at saturation versus coverage in Fig. 1(a) is less pronounced than in the RT growth case. We associate this finding with a reduced intermixing according to a smooth change in the photoemission spectra with Fe thickness at 100 K, as has been shown by Kläsges *et al.*[8] However, the early onset of magnetization is not accompanied by a significantly

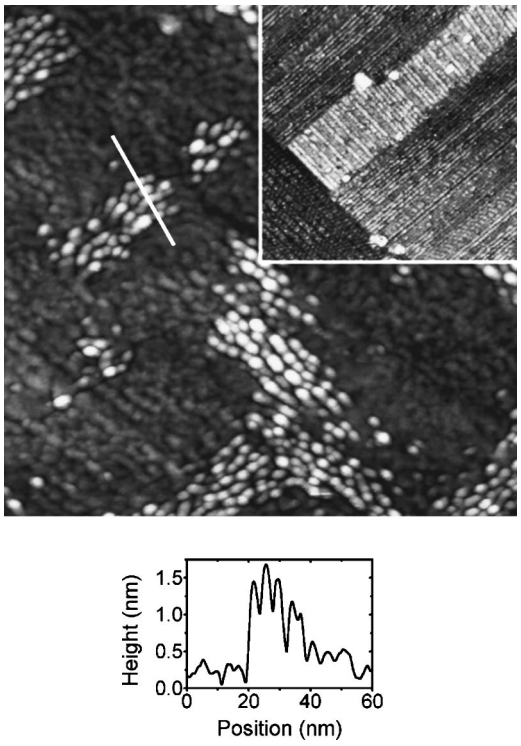

FIG. 3. A 200×200 nm^2 STM topograph of a 20.4 ML thick Fe film grown at 150 K on Au-covered Si(001) and then annealed to room temperature. The image is taken at a bias voltage of 1.6 V and a tunneling current of 1.0 nA. A line profile taken across a group of silicide grains of typical height is plotted underneath. It was performed along the white line shown on the STM image. The inset shows a 100×100 nm^2 STM topograph of a reconstructed surface obtained by the deposition of less than 1 ML of Au at about 1000 K. The scanning parameters were 1.6 V bias voltage and 0.1 nA tunneling current. The roughness of the reconstructed surface is similar to the one of (2×1) Si(001).

larger Kerr signal in comparison to the RT growth. This points at only a minor reduction of intermixing at 150 K.

B. Fe grown on Au-covered Si(001)

In an attempt to hinder silicide formation, we proceeded with passivating the (2×1) Si(001) by depositing less than 1 ML of Au at 1000 K. A STM topograph of such Au-induced reconstructed surface is shown in the inset of Fig. 3. The surface atoms rearrange in long stripes running along the [110] and [1$\bar{1}$0] directions. These stripes were shown to lie behind the occurrence of different structures which coexist on the surface.[18,19] About 1.5 ML of Au was further added at RT or LT as a buffer layer to further inhibit the diffusion of Si into the Fe layer. We emphasize here that our findings on the magnetic behavior of the Fe films grown on Au-covered Si(001) should not be related to the previous reports on the magnetic properties of Fe grown on Au(001),[20–22] for the very complex structure of the reconstructed surface induces a different growth mode and orientation of the Au buffer layer.

The onset of magnetization in the Fe film grown on Au-covered Si(001) at RT takes place at about the same coverage (3.9 ML) as in the case of films deposited at RT on (2×1) Si(001). However, at the onset coverage, an out-of-plane orientation of the magnetic moment is detected, which flips in-plane with adding about 1 ML of Fe. The tendency toward droplet formation due to high Au mobility at RT does not favor the growth of a smooth film, and consequently its role as a buffer layer is not fully achieved. Moreover, the lower surface free energy of Au with respect to that of Fe is responsible for the segregation of a single Au capping monolayer.[23] This leaves roughly only 0.5 ML of Au together with about 0.6 ML of Au (Ref. 18) involved in the reconstruction to limit the Fe–Si intermixing. This is not the main reason for the late magnetization onset at RT though, because increasing the thickness of the buffer layer does not bring about any significant change. The reason for the late onset of magnetization might lie in the roughness of the Au buffer layer.

However, growing both the buffer layer and the Fe film at LT on the Au-induced reconstructed Si(001) surface favors the onset of an out-of-plane magnetization as early as at 1.4 ML of Fe [the corresponding polar loop is shown in the inset of Fig. 1(b)], excluding the existence of a significant magnetically dead layer. The magnetization keeps its perpendicular orientation up to about 2.5 ML, where it flips into the plane when increasing the amount of deposited Fe. Again, we emphasize the existence of a strong out-of-plane magnetic moment in the very thin Fe film at temperatures higher than 100 K, reported as the prerequisite for the occurrence of a thermally unstable perpendicular magnetization on freshly prepared Au(001).[22] The plot of MOKE ellipticity at saturation measured at LT versus coverage [open squares in Fig. 1(b); the points above 200 μrad stand for the out-of-plane magnetization] provides us with another striking result: for in-plane magnetized films we observed strongly enhanced Kerr signals in comparison to films grown without buffer layers at the same temperature. From the onset of magnetization at 1.4 ML of Fe and from the enhanced magnetic signal at LT we conclude that silicide formation is successfully suppressed. A silicide film resulting after the deposition of 1.4 ML of Fe on (2×1) Si(001) at LT does not show any magnetic signal in MOKE. The suppression is further confirmed by the absence of any kink in the plot of ellipticity at saturation versus coverage around 10 ML in the case of Fe grown on buffered Si(001) at 150 K [the open squares in Fig. 1(b)].

Furthermore, a 2.3 ML Fe film grown at LT on Au-covered Si(001) appears to be thermally stable with respect to annealing up to 280 K, as may be concluded from the hysteresis loops in Fig. 4. A decrease in ellipticity from 418 μrad at LT (the solid line in Fig. 4) to 308 μrad is measured at about 280 K (the dotted line in Fig. 4). This is due to the reduction of the magnetization when the Curie temperature is approached. It retrieves its initial value upon cooling back to 150 K (the dashed line in Fig. 4), indicating no significant loss of magnetization due to intermixing. The overall square-like shape of the hysteresis loops and the out-of-plane orientation of the magnetic moment is preserved throughout the process. The increase in coercivity noticed after the cycle is completed might be caused by minor local silicide formation, hindering domain wall motion.

A STM topograph of a 20.4 ML thick Fe film grown at LT on Au-covered Si(001) and then annealed at RT (Fig. 3)

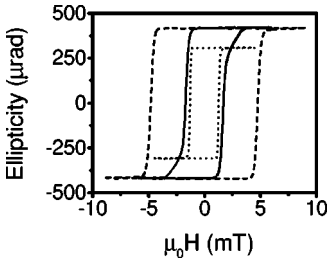

FIG. 4. The hysteresis loops measured by MOKE of an out-of-plane magnetized 2.3 ML thick Fe film grown on Au-covered Si(001) at 150 K (solid line), annealed to 280 K (dotted line) and cooled back to 150 K (dashed line).

shows groups of about 1.7 nm tall silicide grains (the white grains), lying on the top of an otherwise smooth film, which preserves the overall appearance of the underlying reconstructed surface (the inset of Fig. 3). One reason for the occurrence of these silicide grains, apparently oriented along the [110] and [110] directions, might reside in the existence of incomplete reconstructed patches acting as sources for Si diffusion into the Fe layer, triggered by warming up to RT. It is worth mentioning that such patches always appeared in our STM scans if the reconstruction was performed at temperatures lower than about 1000 K. Besides, no LEED pattern was detected throughout the thickness range investigated.

IV. CONCLUSIONS

The strong intermixing in Fe grown on Si(001) at RT takes place at least up to 10 ML and results in the formation of ferromagnetically ordered iron silicide,[7,8] whose magnetization at RT sets in after depositing 3.6 ML of Fe. The growth at 150 K yields an earlier onset of magnetization at 2.3 ML of Fe, partly as the result of a reduced reactivity of the Si(001) surface. However, from the evolution of Kerr signal at saturation with coverage it is deduced that this reduction is only minute. Less than 1 ML of Au deposited at high temperatures (around 1000 K) onto Si(001) induces a complex reconstruction[18,19] of the surface. The Au-induced reconstruction,[16,17] along with an additional ultrathin Au layer of 1.5 ML has been employed to limit the Si diffusion into the Fe film. With this buffer layer, the magnetization always sets in with the magnetic moment oriented out-of-plane, regardless of the deposition temperature, at Fe coverages of 3.9 and 1.4 ML at room temperature and 150 K, respectively. The early onset of magnetization at 1.4 ML in films grown at 150 K on Au-covered Si(001), along with the absence of any sharp feature at 10 ML in the plot of the largely enhanced Kerr signal at saturation versus coverage points at the successful hampering of silicide formation and the growth with virtually zero magnetically dead layers. A 2.3 ML thick Fe film grown at LT on Au-covered Si(001) is shown to be thermally stable upon annealing up to 280 K. This is confirmed by the preservation of the perpendicular orientation of magnetization and by the reversible behavior of the MOKE signal's magnitude upon completing the thermal cycle. These findings prove the possibility of growing stable and almost silicide free thin Fe films on Si(001) by simply employing a noble metal-passivation layer. Using a thin Au buffer layer alleviates the problems associated with the silicide formation and makes Si a good candidate for spin electronics.

[1] J. M. Gallego and R. Miranda, J. Appl. Phys. **69**, 1377 (1991).
[2] G. Crecelius, Appl. Surf. Sci. **65/66**, 683 (1993).
[3] M. C. Bost and J. E. Mahan, J. Appl. Phys. **58**, 2696 (1985).
[4] J. E. Mahan, K. M. Geib, G. Y. Robinson, R. G. Long, Y. Xinghua, G. Bai, M. A. Nicolet, and M. Nathan, Appl. Phys. Lett. **56**, 2126 (1990).
[5] J. Alvarez, J. J. Hinarejos, E. G. Michel, J. M. Gallego, A. L. Vazquez de Parga, J. de la Figuera, C. Ocal, and R. Miranda, Appl. Phys. Lett. **59**, 99 (1991).
[6] J. M. Gallego, J. M. Garcia, J. E. Ortega, A. L. Vazquez de Parga, J. de la Figuera, C. Ocal, and R. Miranda, Surf. Sci. **269/270**, 1016 (1992).
[7] J. M. Gallego, J. M. Garcia, J. Alvarez, and R. Miranda, Phys. Rev. B **46**, 13 339 (1992).
[8] R. Kläsges, C. Carbone, W. Eberhardt, C. Pampuch, O. Rader, T. Kachel, and W. Gudat, Phys. Rev. B **56**, 10 801 (1997).
[9] Z. H. Nazir, C.-K. Lo, and M. Hardiman, J. Magn. Magn. Mater. **156**, 435 (1996).
[10] E. Kaxiras, Phys. Rev. B **43**, 6824 (1991).
[11] W. Yajun, R. J. Hamers, and E. Kaxiras, Phys. Rev. Lett. **74**, 403 (1995).
[12] P. Ma, G. W. Anderson, and P. R. Norton, Surf. Sci. **420**, 134 (1999).
[13] P. Bertoncini, P. Wetzel, D. Berling, G. Gewinner, C. Ulhaq-Bouillet, and V. Pierron Bohnes, Phys. Rev. B **60**, 11 123 (1999).
[14] K. Mizushima, T. Kinno, K. Tanaka, and T. Yamauchi, Phys. Rev. B **58**, 4660 (1998).
[15] M. K. Weilmeier, W. H. Rippard, and R. A. Buhram, Phys. Rev. B **59**, R2521 (1999).
[16] K. Oura, Y. Makino, and T. Hanawa, Jpn. J. Appl. Phys. **15**, 736 (1976).
[17] K. Oura and T. Hanawa, Surf. Sci. **82**, 202 (1979).
[18] X. F. Lin, K. J. Wan, J. C. Glueckstein, and J. Nogami, Phys. Rev. B **47**, 3671 (1993).
[19] H. Minoda, K. Yagi, F.-J. Meyer zu Heringdorf, A. Meier, D. Kähler, and M. Horn von Hoegen, Phys. Rev. B **59**, 2363 (1999).
[20] S. D. Bader, E. R. Moog, and P. Grünberg, J. Magn. Magn. Mater. **53**, L295 (1986).
[21] W. Dürr, M. Taborelli, O. Paul, R. Germar, W. Gudat, D. Pescia, and M. Landolt, Phys. Rev. Lett. **62**, 206 (1989).
[22] C. Liu and S. D. Bader, J. Vac. Sci. Technol. A **8**, 2727 (1990).
[23] S. D. Bader and E. R. Moog, J. Appl. Phys. **61**, 3729 (1987).