Strain engineering of magnetic anisotropy in thin ferromagnetic films

2009 J. Phys.: Condens. Matter 21 314012

(http://iopscience.iop.org/0953-8984/21/31/314012)

J. Phys.: Condens. Matter **21** (2009) 314012 (6pp) doi:10.1088/0953-8984/21/31/314012

Strain engineering of magnetic anisotropy in thin ferromagnetic films

R Zdyb[1,2], A Pavlovska[1] and E Bauer[1]

[1] Department of Physics, Arizona State University, Tempe, AZ, 85287-1504, USA
[2] Institute of Physics, Maria Curie-Sklodowska University, Plac M Curie-Sklodowskiej 1, 20-031 Lublin, Poland

Received 20 December 2008
Published 7 July 2009
Online at stacks.iop.org/JPhysCM/21/314012

Abstract
Magnetic properties of iron films grown on Au layers with different thicknesses on a W(110) surface are studied with spin polarized low energy electron microscopy. The iron thickness for the onset of ferromagnetic order depends approximately linearly on the thickness of underlying gold film. The easy axis direction also depends upon the Au thickness. It is parallel to the tungsten [1$\bar{1}$0] direction at the onset of magnetization for one and two monolayers of gold. For thicker gold films the easy axis is parallel to the [001] direction. The direction of the easy axis and the onset of ferromagnetic order are discussed in terms of magnetic anisotropies, interaction between the iron overlayer, gold and tungsten substrate, Fe film strain and morphology.

1. Introduction

The magnetization direction in ultrathin ferromagnetic films has been the subject of numerous studies because of both fundamental interest in low dimensional magnetism and its importance in practical applications. It was already suggested many years ago that misfit strain on rigid substrates induces magnetic anisotropy different from that of the bulk that can persist for many monolayers, as evident in the system Fe on W(110) [1]. In this paper we show that suitable interfacial layers can eliminate this effect so that the ferromagnetic layer has from the very beginning the magnetic anisotropy of the bulk which makes the [001] direction the easy axis in Fe films. An example is provided by Fe on W(110) with a Au interfacial layer. Two aspects are important for achieving this goal. (1) Strain relaxation via the interfacial layer. The Young modulus is 411 GPa for W and 78 GPa for Au, the shear modulus is 161 GPa for W and 27 GPa for Au, while the corresponding moduli for Fe are 211 and 82 GPa [2]. Therefore an interfacial Au layer absorbs much of the strain exerted by the W substrate on the Fe layer. (2) Misfit minimization for the easy axis in the bulk ferromagnet. If the Fe(110) layer is oriented with the [001] direction parallel to the [1$\bar{1}$0] direction of the Au interfacial layer, then the distances between the atomic rows perpendicular to these directions are 2.884 and 2.866 for Au and Fe, respectively. This gives a row misfit of only 0.6% while the misfit in the [1$\bar{1}$0] direction of Fe is 23.2%. These two factors together remove the driving force for the [1$\bar{1}$0] easy axis.

The Fe/W(110) system has been studied extensively for several decades. Its morphology and structural properties were determined in the late 1980s and 1990s [3–5]. It was found that the Fe layer is pseudomorphic (ps) up to 1.2–1.8 ML depending on deposition temperature. The lattice misfit of 10.4% between Fe and W causes at about 1.2 ML the formation of a one-dimensional lattice of misfit dislocations along the [001] direction, which accommodate the huge strain in the layer. Then, at about 2 ML a two-dimensional (2D) lattice of misfit dislocation appears in the Fe layer. STM studies reveal that the 2D lattice is well ordered and creates regular patterns in microscopy images [1, 5]. In the 2D lattice of misfit dislocations additional rows of Fe atoms are incorporated along the ⟨111⟩ directions. The existence of the lattice distortions was also observed in low energy electron diffraction (LEED) experiments as additional spots [3]. The distortions were observed even in films as thick as 11 ML [1]. The periodicities associated with the distortions were reported to be 33.4 Å and 37.1 Å [5] or 35.84 Å and 50.76 Å [1] in the [001] and [1$\bar{1}$0] directions, respectively.

The large lattice misfit of 10.4% between the ps Fe monolayer and the W substrate induces significant stress in the iron film. Optical bending method measurements show in the monolayer thickness range an anisotropic stress of 65 and 44 GPa in the [1$\bar{1}$0] and [001] directions, respectively, and above 4 ML an isotropic 13 GPa stress [6]. Recent measurements give in a 13 ML thick Fe film an isotropic in-plane strain of +1.2% and normal to the film a strain of +0.22% [1].

The easy axis of magnetization in ultrathin Fe films was found to be an in-plane one and parallel to the $[1\bar{1}0]$ direction [7, 8]. The authors of [7, 8] took the surface anisotropy to be responsible for the direction of the easy axis. The idea of surface anisotropy as a driving force for the $[1\bar{1}0]$ direction of magnetization was recently supported by an experiment in which the influence of gold, silver and oxygen layers on the spin reorientation transition was investigated [9]. Both silver and gold overlayers caused a decrease of the Fe thickness at which the spin reorientation transition takes place. The authors concluded that the overlayer 'eliminates' Fe surface anisotropy that makes the Fe film bulk-like with the easy axis parallel to the [001] direction.

However, it is also well known that magnetoelastic anisotropy has a significant influence on the direction of the easy axis in ultrathin ferromagnetic films [6 and references therein]. The strain tensor elements together with magnetoelastic coupling coefficients B_1 and B_2 define the magnetoelastic energy density which influences the direction of the easy axis. In the case of Fe/W(110) the strain values found in a 13 ML thick Fe film give a magnetoelastic anisotropy energy density of -0.076 MJ m^{-3} which indicates the in-plane $[1\bar{1}0]$ direction as the easy axis [1].

The system Fe/Au(111) has been studied less. Considering the surface energies of Fe and Au, 2.939 and 1.626 J m^{-2}, respectively, it is very surprising that Fe forms pseudomorphic monolayers on Au(111) at all, as previously reported [10, 11]. At about 3 ML a transition to the iron bcc phase has been reported and the growth becomes rougher. In the bcc phase the Fe layer grows with Fe[001] parallel to Au$[1\bar{1}0]$ and Fe$[1\bar{1}0]$ parallel to the Au$[11\bar{2}]$ direction.

The magnetization direction of Fe in the bcc phase grown on a thick layer or on a Au(111) single-crystal substrate is parallel to the Fe[001] axis like in bulk Fe. Below 2, 3 ML Fe a weak tendency to out-of-plane anisotropy was observed [11–14] and in the submonolayer thickness range the coexistence of in-plane and out-of-plane anisotropy [11].

The direction of the easy axis in ultrathin Fe films in the bcc phase on a thick (111)-oriented Au layer and on a Au(111) single crystal can be attributed to the magnetoelastic anisotropy caused by the lattice misfit [15]. While in the Fe$[1\bar{1}0]$ direction, which is parallel to the Au$[11\bar{2}]$ direction, the strain is about 23%, in the Fe[001] direction it is 0.6%. Because of the small misfit in the [001] direction the Fe lattice locks in this direction into the Au lattice and builds up considerable strain with increasing thickness. In the $[1\bar{1}0]$ direction the Fe lattice is floating on the Au(111) surface because of the large lattice mismatch. This suggests that the easy axis in Fe layers on W(110) may be changed from $[1\bar{1}0]$ to [001] by inserting a thin strain-changing Au layer between the W substrate and the Fe layer. In this paper we report results of a study of the influence of the thickness of the underlying gold Au film on the easy axis direction of the ultrathin Fe film.

2. Experimental details

The experiments were performed using a spin polarized low energy electron microscope (SPLEEM) with a base pressure in the high 10^{-11} mbar range. The SPLEEM is a conventional LEEM instrument equipped in addition with a source of spin polarized electrons and a spin polarization manipulator which allows orientation of the electron beam polarization vector in any desired direction with respect to the sample crystallographic directions. The details of the SPLEEM instrument can be found elsewhere [16, 17].

Magnetic images are obtained by subtracting two images recorded with opposite electron beam polarization, $I_+ - I_-$, where I_+ and I_- represent the reflected intensities of oppositely polarized beams. Subtraction eliminates the non-magnetic contrast and leaves only the features which are associated with the sample magnetization. The difference divided by the sum of the two images results in the so-called asymmetry image: $A = (I_+ - I_-)/(I_+ - I_-)$.

The W(110) surface was cleaned by standard procedures by heating to 1350 K at an oxygen pressure of 10^{-7} mbar followed by flashing to about 2000 K. Iron and gold were deposited from resistively heated crucibles at rates of 0.17 and 0.1 ML min^{-1}, respectively. During deposition the pressure stayed in the low 10^{-10} mbar range. The first two monolayers of Au were grown at 600 K. The remaining number of monolayers were deposited close to room temperature. This procedure ensures quasi-monolayer by monolayer growth of Au which can be seen in the LEEM images as a change of contrast from monolayer to monolayer due to quantum size effects [18]. Fe was deposited on the top of Au layer at room temperature. The Fe film thickness was calibrated via the time needed to complete the first pseudomorphic monolayer on the bare W(110) surface at about 650 K in a separate experiment. Fe thickness accuracy was determined as ± 0.05 ML. All SPLEEM images were recorded at room temperature.

3. Results and discussion

Figure 1 shows SPLEEM images taken just after the onset of ferromagnetic order in ultrathin Fe films grown on W(110) covered with 1, 2, 3 and 4 ML Au, recorded in two in-plane directions perpendicular to each other. In all cases out-of-plane images show no ferromagnetic contrast.

In the SPLEEM images taken just after the onset of ferromagnetic order the easy axis is parallel to the tungsten $[1\bar{1}0]$ direction on the bare substrate and on 1 and 2 ML of gold (figures 1(a) and (c)), while at larger gold thickness the easy axis is parallel to the tungsten [001] direction (figures 1(f) and (h)). In all cases except for on 2 ML Au, the easy axis remains in the initial crystallographic direction in the Fe thickness range investigated, of about 6 ML of Fe. On 2 ML Au the magnetization changes with thickness in a complicated manner associated with structural changes which will be reported elsewhere [19].

The onset of ferromagnetic order depends on the thickness of the underlying gold film. Figure 2 shows asymmetry versus Fe thickness curves for iron grown on 1, 2, 3 and 4 ML thick gold films on the W(110) surface. In the case of 1 ML Au the onset occurs at 1.48 ML. This is within the limits of accuracy the same thickness as for Fe grown on the bare W(110) surface which is 1.5 ML [20, 21]. With increasing gold thickness

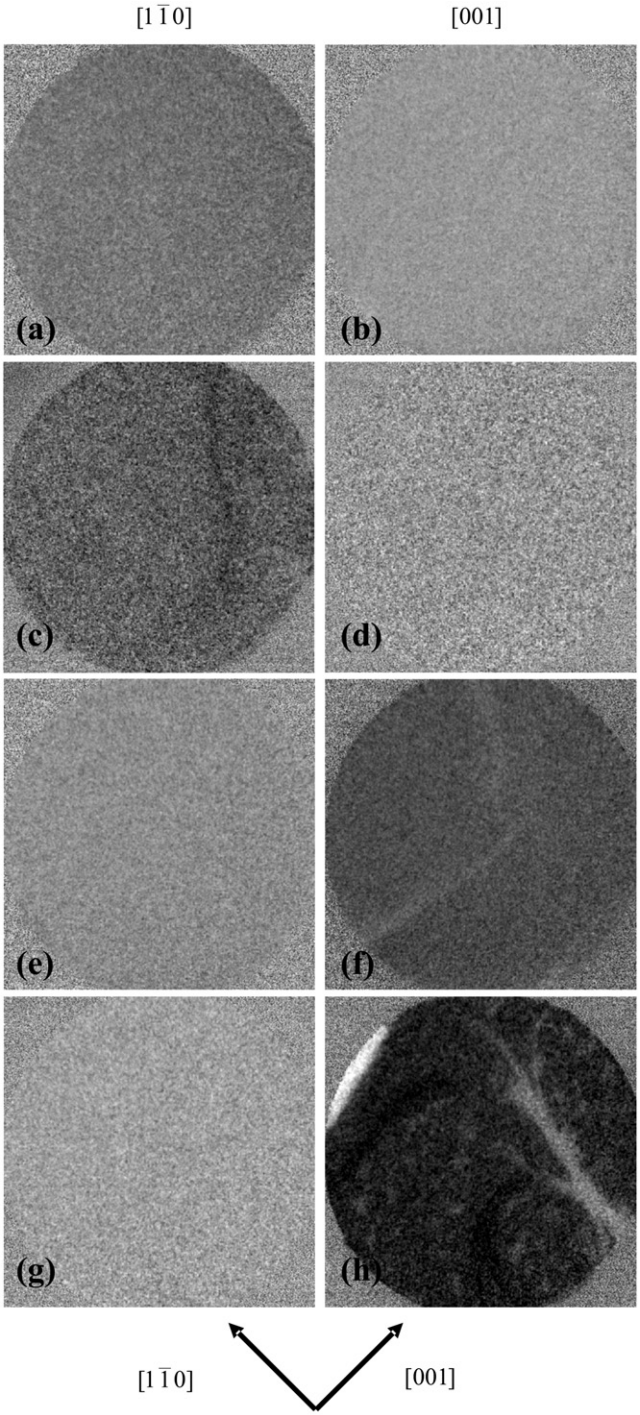

Figure 1. SPLEEM images of x Fe monolayers on n Au monolayers on W(110). (a) and (b): $x = 1.72, n = 1$, (c) and (d): $x = 1.60, n = 2$, (e) and (f): $x = 1.89, n = 3$, (g) and (h): $x = 2.04, n = 4$. Images in the left/right column were recorded with the incident electron polarization vector parallel to the [1$\bar{1}$0]/[001] direction. The dark regions in (a), (c), (f) and (h) indicate magnetic contrast while the gray regions in (b), (d), (e) and (g) images indicate absence of magnetic contrast.

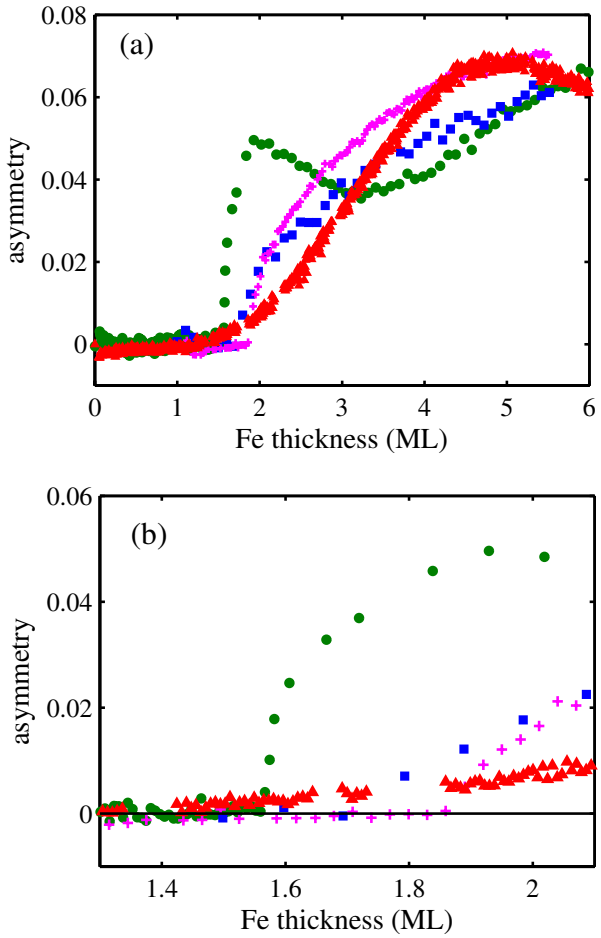

Figure 2. (a) Asymmetry versus Fe thickness on n Au monolayers. Red triangles: $n = 1$, green circles: $n = 2$, blue squares: $n = 3$, pink crosses: $n = 4$. (b) Magnified Fe thickness region at which onset of magnetization occurs.

(This figure is in colour only in the electronic version)

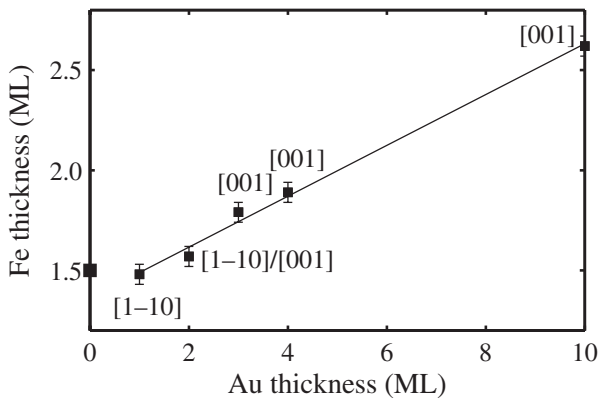

Figure 3. Fe thickness at which ferromagnetic order appears versus Au thickness. The solid line is least-squares fit. The easy axis direction is indicated for the Au thicknesses studied.

the onset of magnetization shifts towards larger Fe thickness, reaching 2.62 ML Fe on 10 ML Au. As shown in figure 3, the onset thickness of the magnetization increases linearly with increasing gold thickness.

It is well known that Fe atoms strongly interact with a tungsten surface and the iron 3d bands hybridize with the tungsten 5d bands. On the other hand, the 5d W bands do not interact much with the Au 5d bands [22], and similarly

the Au 5d bands are well separated from the Fe 3d bands [9], so direct d–d interactions are minimal. Thus, inserting a Au layer between the Fe film and W substrate should considerably decrease the interaction of Fe atoms with the tungsten surface. However, as observed in the experiment, the presence of 1 ML of gold on the W(110) surface does not change the direction of the easy axis and the thickness of the onset of magnetization. Both remain the same as for the Fe grown on the bare W(110) surface. Clear changes require three or more gold monolayers.

1 ML Au on W(110) can be considered as a distorted Au(111) plane with the atomic distances in the W[001] direction increased to fit the W periodicity and with the atomic distances in the W[1$\bar{1}$0] direction compressed in order to partially compensate for the reduced packing density [23]. The resulting lattice misfit between Fe and the Au monolayer is 10.4% and 13.7% in the [001] and [1$\bar{1}$0] directions, respectively, instead of 0.6% and 23.2% for bulk Au(111). In addition, the electronic density of states of the Au monolayer is completely different from that of the (111) surface of bulk Au and reflects strong interaction with the W substrate [22]. The apparent coupling with the tungsten substrate together with the almost unchanged lattice misfit between Fe and the Au monolayer-covered W substrate are responsible for the fact that the Au monolayer does not change the easy axis direction induced by the substrate.

As mentioned above, in order to suppress the influence of the W substrate on the magnetic anisotropy sufficiently, so that the easy axis of the Fe film acquires the direction of bulk Fe from the very beginning, 3 ML Au are necessary. LEED makes this understandable. At three or more monolayers LEED gives within the limits of error bulk interatomic distances in the (111) plane. The Au[11$\bar{2}$] direction is oriented approximately parallel to W[1$\bar{1}$0] and the Au[1$\bar{1}$0] direction correspondingly approximately parallel to W[001]. No misfit dislocations could be seen upon Fe deposition but the (00) spot is elongated in the [1$\bar{1}$0] direction. This suggests that the Fe layer consists of structures elongated in the [001] direction, such as the flat roof structures reported previously on the basis of detailed high resolution LEED studies of the growth of Fe on W(110) [24], with possible distortions along the [1$\bar{1}$0] direction which are not ordered well enough to give diffraction spots. LEED also shows that the Fe layer grows pseudomorphically on the Au layer. Therefore, the Fe lattice has to be expanded by more then 23% in the Fe [1$\bar{1}$0] direction and 0.6% in the [001] direction. The Fe lattice cannot accommodate such a large lattice misfit in the [1$\bar{1}$0] direction, so it has to be either floating with the bulk-like lattice constant in this direction or form misfit dislocations. On the other hand, the lattice mismatch of 0.6% causes the outcome that the Fe layer can be accommodated by strain in the [001] direction. The strain couples through the magnetoelastic coefficients B_1 and B_2 to the magnetization and gives rise to the magnetoelastic anisotropy. Thus, there are magnetoelastic and in-plane shape anisotropy contributions which force the magnetization easy axis to be parallel to the [001] direction.

The observed changes of the easy magnetization axis in the iron films with the thickness of the interfacial Au layer clearly indicate a competition between different

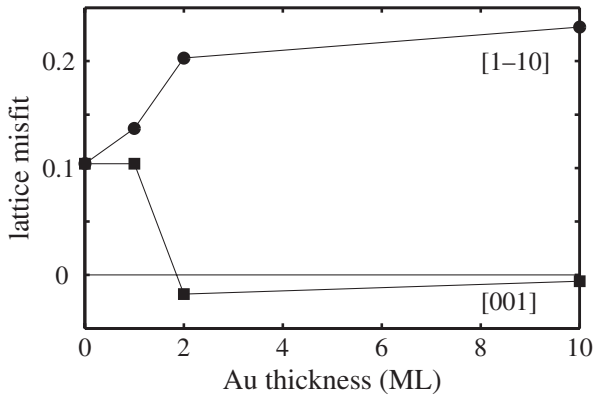

Figure 4. Lattice misfit between the Fe film and n ML Au/W(110) substrate versus number n of Au monolayers for two in-plane directions.

anisotropies. Several anisotropies may be competing: magnetoelastic anisotropy, magnetic surface anisotropy, magnetic step anisotropy, shape anisotropy and magnetocrystalline anisotropy.

Magnetoelastic anisotropy has been reported to be responsible for the in-plane [1$\bar{1}$0] easy axis in Fe films on W(110) [1, 25 and references therein]. The same direction was also found in our experiments for Fe films on the bare substrate, on 1 ML Au, and just after the appearance of ferromagnetic order on 2 ML Au. Considering the observed rotation of the easy axis from the magnetoelastic anisotropy point of view the strain anisotropy in the Fe film must change with increasing Au coverage. Comparing the lattice misfit between Fe and underlying n ML Au/W(110) substrate, where n denotes the number of Au monolayers, two regions can be distinguished in figure 4. The Fe layer experiences tensile strain in both the [1$\bar{1}$0] and [001] directions below 2 ML Au while above that thickness the strain becomes compressive in the [001] direction. In this direction the misfit is small (0.6%) so the layer can lock in with the Au film with little strain. In the [1$\bar{1}$0] direction the large misfit (23.2%) cannot be accommodated so the Fe layer rapidly relaxes toward the atomic distances in the bulk with increasing thickness. The magnetoelastic anisotropy associated with the strain in the [001] direction could explain the observed changes of the direction of the easy axis. The more complicated behavior of the direction of the easy axis on 2 ML Au represents a transition stage and will be discussed elsewhere [19].

Surface anisotropy is another factor that can determine the direction of the easy axis [7, 8]. The observed rotation from the [1$\bar{1}$0] direction to the [001] direction means in this case that the surface anisotropy changes. A possible reason for the change is a floating Au layer on the top of Fe film. As shown in [26], one monolayer of Au on the top of a Fe film grown below 500 K induces a [001] easy axis. However, our LEED studies did not show any extra spots characteristic for the Au overlayer [26]. In addition, already on 3 ML Au the [001] direction was the easy axis in the Fe films. If one Au ML had floated on top of the Fe film a behavior similar to that observed for 2 ML Au would be expected. However, starting with 3 ML Au we

do not see any particular differences between the properties of Fe films with increasing Au coverage except for as regards the Fe thickness at which ferromagnetic order appears. Thus, it appears unlikely that surface anisotropy is responsible for the change of the direction of the easy axis although its influence cannot be excluded without complementary surface composition-sensitive studies.

A third, easy axis-determining factor could be magnetic step anisotropy which could be connected also with shape anisotropy. Like for Fe layers grown directly on W(110), also for Fe layers grown on all Au layers the (00) spot is strongly elongated in the $[1\bar{1}0]$ direction. This indicates that the Fe layer has flat ridges along the [001] direction with a high density of steps along the [001] direction as observed in the previous studies [24, 27 and references therein]. The magnetic anisotropy associated with the steps is of the same order of magnitude as surface anisotropies and supports an easy axis in the [001] direction [28]. A [001] easy axis is also supported by the shape anisotropy of the [001] ridges similar to the quasi-one-dimensional structures observed in other elongated Fe structures [29]. However, similar ridge structures form also in Fe films grown directly on W(110) and on 1 and 2 ML Au so these anisotropies cannot be the main cause for the $[1\bar{1}0]$ to [001] direction. Finally, magnetocrystalline anisotropy, which induces a [001] easy axis, can contribute to the SRT but is also too weak compared to the magnetoelastic anisotropy to be the major driving force as seen from the comparison of Fe films on W(110) with $[1\bar{1}0]$ easy axis with Fe on n ML Au ($n \geqslant 2$) on W(110) with the [001] easy axis.

Figure 3 shows that the Fe thickness t_{on} at which ferromagnetic order appears increases with increasing thickness of the underlying gold film from about 1.5 ML to about 2.6 ML. It is known that Fe atoms strongly hybridize with the W(110) surface. This shifts the minority bands below the Fermi level which results in a low magnetic moment of about 2.5 μ_B of the iron atoms [30]. On the other hand, the weak hybridization with Au results in a larger value of the magnetic moment (2.8–3μ_B) [31]. One would therefore expect that the Au layer between Fe film and W substrate, which decouples them and prevents strong hybridization, would reduce the Fe thickness at which magnetic order is observed. As already discussed, 1 ML Au strongly interacts with the W(110) surface; thus it has little influence on t_{on}. The increase of t_{on} at larger Au thicknesses can be attributed to the surface morphology of the growing film. While the growth is quasi-ML-by-ML, usually three layer levels are present at the same time and at room temperature the ML islands in the various levels are small, providing many steps. The atomic roughness increases somewhat with Au thickness, providing an increasing number of steps. A rougher surface requires also more iron atoms for the formation of a continuous layer. Both effects lead to a lower coordination number of the Fe atoms which decreases the Curie temperature of the Fe film. In addition, monatomic steps in a continuous ferromagnetic layer weaken spin–spin interactions between the atoms in the neighboring terraces and reduce the effective Curie temperature [32]. These factors qualitatively explain the observed increase of the Fe thickness at which ferromagnetic order appears with increasing Au coverage.

In summary, we have investigated the evolution of ferromagnetic order in Fe films grown on Au layers with different thickness on W(110). Both the direction of the easy axis and the Fe thickness at which the onset of ferromagnetic order occur depend upon the thickness of the underlying gold film. A comparison of the various magnetic anisotropies that determine the easy axis strongly suggests that magnetoelastic anisotropy is responsible for the observed changes of the magnetization direction with Au film thickness. However, the influence of a surface anisotropy change due to a floating gold layer on top of Fe film cannot be completely excluded. The observed increase of the Fe thickness of the onset of ferromagnetic order with increasing Au thickness can be attributed to the increasing substrate roughness and to finite size effects associated with the increasing number of surface steps.

Acknowledgments

The work was supported by the Polish Ministry of Science and Higher Education under Grant No. N202 159 32/4302 and by the National Science Foundation under Grant No. DMR-9818296.

References

[1] Popescu R, Meyerheim H L, Sander D, Kirschner J, Steadman P, Robach O and Ferrer S 2003 *Phys. Rev.* B **68** 155421
[2] *Landolt–Börnstein* vol II/5 p 236 ; Webelements
[3] Gradmann U and Waller G 1982 *Surf. Sci.* **116** 539
[4] Berlowitz P J, He J-W and Goodman D W 1990 *Surf. Sci.* **231** 315
[5] Bethge H, Heuer D, Jensen Ch, Reshöft K and Köhler U 1995 *Surf. Sci.* **331–333** 878
[6] Sander D 1999 *Rep. Prog. Phys.* **62** 809
[7] Gradmann U, Korecki J and Waller G 1986 *Appl. Phys.* A **39** 101
[8] Elmers H J and Gradmann U 1990 *Appl. Phys.* A **51** 255
[9] Baek I-G, Lee H G, Kim H-J and Vescovo E 2003 *Phys. Rev.* B **67** 075401
[10] Stroscio J A, Pierce D T, Dragoset R A and First P N 1992 *J. Vac. Sci. Technol.* A **10** 1981
[11] Ohresser P, Brookes N B, Padovani S, Scheurer F and Bulou H 2001 *Phys. Rev.* B **64** 104429
[12] Chappert C, Bruno P, Bartenlian B, Beauvillain P, Bounouh A, Megy R and Veillet P 1995 *J. Magn. Magn. Mater.* **148** 165
[13] Xu J, Howson M A, Hucknall P, Hickey B J, Venkataraman R, Hammond C, Walker M J and Greig D 1997 *J. Appl. Phys.* **81** 3908
[14] Toulemonde O, Petrov V, Nait Abdi A and Bucher J P 2004 *J. Appl. Phys.* **95** 6565
[15] Zdyb R and Bauer E 2003 *Phys. Rev.* B **67** 134420
[16] Duden T and Bauer E 1995 *Rev. Sci. Instrum.* **66** 2861
[17] Bauer E 2005 *Magnetic Microscopy of Nanostructures* ed H Hopster and H P Oepen (Berlin: Springer) p 111
Bauer E 2005 *Modern Techniques for Characterizing Magnetic Materials* ed Y Zhu (Boston, MA: Kluwer) p 361
[18] Tringides M C, Jałochowski M and Bauer E 2007 *Phys. Today* **60** 50
[19] Zdyb R, Mentes T O and Bauer E 2009 in preparation
[20] Elmers H J, Hauschild J, Fritzsche H, Liu G and Gradmann U 1995 *Phys. Rev. Lett.* **75** 2031

[21] Wagner K, Weber N, Elmers H J and Gradmann U 1997 *J. Magn. Magn. Mater.* **167** 21
[22] Knoppe H and Bauer E 1993 *Phys. Rev.* B **48** 5621
[23] Bauer E, Poppa H, Todd G and Davis P R 1977 *J. Appl. Phys.* **48** 3773
[24] Albrecht M, Fritzsche H and Gradmann U 1993 *Surf. Sci.* **294** 1
[25] Sander D 2004 *J. Phys.: Condens. Matter* **16** R603
[26] Elmers H J and Gradmann U 1994 *Surf. Sci.* **304** 201
[27] Albrecht M, Gradmann U, Furubayashi T and Harrison W A 1992 *Europhys. Lett.* **20** 65
[28] Albrecht M, Furubayashi T, Przybylski M, Korecki J and Gradmann U 1992 *J. Magn. Magn. Mater.* **113** 207
[29] Zdyb R, Pavlovska A, Jałochowski M and Bauer E 2006 *Surf. Sci.* **600** 1586
[30] Spišák D and Hafner J 2004 *Phys. Rev.* B **70** 014430
[31] Wang J-T, Li Z-Q, Sun Q and Kawazoe Y 1998 *J. Magn. Magn. Mater.* **183** 42
[32] Zdyb R and Bauer E 2008 *Phys. Rev. Lett.* **100** 155704