Exchange anisotropy as a probe of antiferromagnetism in expanded face-centered-tetragonal Mn(001) layers

Citation for published version (APA):

Kohlhepp, J. T., Wieldraaijer, H., & Jonge, de, W. J. M. (2006). Exchange anisotropy as a probe of antiferromagnetism in expanded face-centered-tetragonal Mn(001) layers. *Applied Physics Letters*, 89(3), 032507-1/3. [032507]. <https://doi.org/10.1063/1.2222342>

DOI:

[10.1063/1.2222342](https://doi.org/10.1063/1.2222342)

Document status and date:

Published: 01/01/2006

Document Version:

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
- The final author version and the galley proof are versions of the publication after peer review.
- The final published version features the final layout of the paper including the volume, issue and page numbers.

[Link to publication](#)

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license above, please follow below link for the End User Agreement:

www.tue.nl/taverne

Take down policy

If you believe that this document breaches copyright please contact us at:

openaccess@tue.nl

providing details and we will investigate your claim.

Exchange anisotropy as a probe of antiferromagnetism in expanded face-centered-tetragonal Mn(001) layers

J. T. Kohlhepp,^{a)} H. Wieldraaijer, and W. J. M. de Jonge

Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands and Center for NanoMaterials (cNM), Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

(Received 20 March 2006; accepted 13 June 2006; published online 19 July 2006)

Manganese (Mn) grows coherent and with an expanded metastable face-centered-tetragonal (e-fct) structure on ultrathin fct Co(001)/Cu(001) template layers. From the temperature dependence of the observed unidirectional Mn/Co interface exchange anisotropy, an antiferromagnetic state with a blocking temperature around 410 K is found for a 21 monolayer thick e-fct Mn(001) film. The temperature dependent coercivity enhancement of the Co films, which is induced by the proximity of the antiferromagnetic Mn layer, suggests a Néel temperature above 410 K for this Mn phase.

© 2006 American Institute of Physics. [DOI: 10.1063/1.2222342]

In the last two decades numerous research studies on magnetic exchange interactions observed between two magnetically ordered thin layers were reported.^{1,2} This tremendous interest not only stems from the intriguing fundamental physics but also from the importance of magnetic interactions for the functionality of modern so-called spintronic devices, which exploit the spin of an electron in addition to its electrical charge.³ Especially in giant magnetoresistance or tunnel magnetoresistance devices very often antiferromagnets (AFMs) exchange coupled to ferromagnets (FMs) are key components. Here, the function of an AFM is mainly to pin the magnetization of an adjacent FM to a certain direction to insure a desired switching behavior and ergo functionality. Another application could be envisioned in the field of future next generation high density recording media by exploiting the coercive field enhancements and magnetic anisotropy changes, which are typically observed in AFM/FM systems, to ensure the magnetic stabilization of nanometer-size FM particles by surrounding them with an AFM shell.⁴

Despite this enormous potential for present and future technological applications, a fundamental understanding of the exact mechanisms, responsible for the so-called exchange anisotropy and several associated phenomena observed,^{1,2} is still not achieved. Mainly triggered by the direct application perspectives, the majority of the reported studies deals with polycrystalline samples. These structures, however, suffer from different sorts of imperfections, such as interdiffusion, uncontrolled interface roughness, grain size and phase distributions, random magnetic anisotropies, etc. Single-crystalline systems with simple lattices offer well-defined and tunable interfaces, spin orientations, and anisotropies, allowing systematic studies to unveil in more detail the exchange mechanisms involved, but are only rarely reported.^{5,6}

In this letter, we introduce a well-defined single-crystalline AFM/FM model system which is based on tetragonally distorted Mn(001) thin films, epitaxially stabilized on fct Co(001). We present structural evidence for a metastable Mn phase, namely, *expanded* face-centered-tetragonal (e-fct), a slightly distorted fcc(γ) Mn phase with an axial

ratio $c/a > 1$. Recent theoretical calculations have shown that, in addition to a contracted metastable phase ($c/a = 0.945$), such a metastable structure with $c/a = 1.048$ should indeed exist.⁷ By exploiting the exchange interaction at an AFM/FM interface, it is shown that e-fct Mn(001) is in an AFM state with a Néel temperature (T_N) well above room temperature.

The Mn layers were deposited on thin magnetic fct Co(001) template films, which were grown on Cu(001) single crystal substrates in a VG-Semicon V80M molecular beam epitaxy system. This multichamber system is equipped with a range of surface characterization techniques, such as scanning tunneling microscopy (STM), low energy electron diffraction (LEED), x-ray photoelectron spectroscopy (XPS), and Auger electron spectroscopy (AES). During the experiments it was noticed that the growth quality of the Mn is very sensitive to impurities. By using high purity Co (99.99%) and Mn (99.999%) and applying extensive outgassing procedures, deposition pressures below 5×10^{-11} mbar could be reached. In this way impurity inclusions in the films and at the interfaces were minimized. The Cu(001) crystals were treated with several Ar sputter and anneal cycles to ensure clean and atomically flat surfaces. During deposition the substrate temperature was maintained at 50 °C. The Mn films reported here were grown at a rate of 1.5 monolayer (ML)/min as deduced from an accurately calibrated quartz microbalance. After *in situ* characterization the Co/Mn bilayers were covered with 20 Å Cu and 20 Å Al to avoid the oxidation of the magnetic layers in the *ex situ* magnetization measurements. These capping layers had no measurable influence on the structural properties of the Mn, at least not for the thicknesses we used for the *ex situ* experiments.

LEED diffraction patterns and LEED $I(V)$ curves of the 00-spot were measured to identify the crystal structure and to determine the lattice constants. Complementary x-ray photoelectron diffraction (XPD) and x-ray diffraction (XRD) experiments were used to confirm these results independently. The magnetic properties of the Co/Mn bilayers were determined with the longitudinal magneto-optical Kerr effect (MOKE) at room temperature, and with a superconducting quantum interference device (SQUID) magnetometer working in the temperature range from 5 to 400 K. While with

^{a)} Author to whom correspondence should be addressed; electronic mail: j.t.kohlhepp@tue.nl

FIG. 1. (Color online) (a) LEED 00-spot $I(V)$ curves of 10 ML Co(001)/Cu(001) films, uncovered and covered with 2.9 and 16.7 ML Mn(001), respectively. Depending on the Mn cover thickness two different series of Bragg peaks are observed as indicated by the two sets of maxima labeled (1) and (2). (b) Vertical interlayer distances deduced from such LEED $I(V)$ measurements vs the Mn overlayer thickness, either grown on 10 ML (squares) or 20 ML (circles) Co(001). The right axis shows the ratio of the out-of-plane to the in-plane lattice constant (c/a) assuming coherent growth.

MOKE it was possible to rotate the external magnetic field along all desired crystal directions, in the SQUID, due to the limited sample space and the shape of the available Cu crystal, the field was restricted to the in-plane $[100]$ Cu direction.

We have tested the growth of Mn on Co(001) template thicknesses of 10 and 20 ML. The thickness of 10 ML is in the region where Co grows close to layer by layer on Cu(001) and adopts the Cu(001) in-plane lattice constant (pseudomorphic growth). For this thickness the Co(001) surface is dislocation-free and on average only two atomic levels are exposed, as seen by STM.⁸ For the second thickness, 20 ML, the Co starts to relax slightly, which is accompanied by the appearance of isolated dislocation lines and extra roughening of the surface (now exposing four atomic layers on average).⁸ However, the growth of Mn remains unaffected by these tiny but measurable changes of the Co structure and morphology. Initially, Mn fills the grooves on the Co surface and forms a very flat surface for the subsequent layer-by-layer growth mode of Mn, which is observed up to a thickness of 10–15 ML (this thickness being slightly higher for the thinner and flatter Co films). Beyond this, a somewhat roughened and disordered surface evolves. The LEED diffraction patterns of Mn overlayers is dominated by the existence of sharp $p(1 \times 1)$ spots at the positions of the Co substrate spots, demonstrating a coherent (pseudomorphic) growth with the Co in-plane interatomic spacing. However, in particular, for the growth on thin Co films, indications of several different surface reconstructions are observed. For example, between 2 and 4 ML a weak $c(12 \times 8)$ and above 10–15 ML a weak $p(18 \times 1)$ superstructure is visible. We interpret these reconstructions as being confined to the surface and not reflecting a major change in the bulk Mn structure. Such surface reconstructions are commonly found in the growth of metastable materials such as, e.g., fcc Fe(001) on Cu(001).⁹

From LEED patterns we can determine the in-plane crystal symmetry and interatomic spacing, but no information about the lattice structure parallel to the surface normal is accessible. However, from the energy dependence of the intensity of the mirror beam (00-spot), the vertical interlayer distance $d_{\perp}$ can be estimated by a simple kinematical

FIG. 2. (Color online) Room temperature longitudinal MOKE hysteresis loops of 21 ML Mn/10 ML Co/Cu(001) measured with an applied external field H along different in-plane directions, both for as grown and from $T = 435$ K field-cooled magnetization states. In the right cartoon the field-cooling direction with respect to the crystal axes is depicted. Note that the MOKE laser spot did not stay exactly at the same sample position during rotation of the sample.

calculation.¹⁰ In Fig. 1(a) LEED $I(V)$ curves for uncovered films and those covered with 2.9 ML Mn(001) and 16.7 ML Mn(001) capped 10 ML Co(001)/Cu(001) are plotted. Depending on the overlayer thickness, two different series of Bragg maxima are identified, and in a certain Mn thickness range (up to roughly 3.5 ML) a coexistence of both peak series is observed. We have performed $I(V)$ experiments for many different Mn overlayer thicknesses on both 10 and 20 ML thick Co films. The results of these experiments are shown in Fig. 1(b) and summarized as follows. While Bragg series (1) is clearly originating from diffraction at Co lattice planes, series (2) is unambiguously coming from the Mn lattice. In the coexistence regime electrons diffracted at the Co underlayer lattice are still detected through the Mn overlayer. This interpretation is supported by the observation that peaks of series (1) of Mn capped Co samples are distinctly more attenuated at lower order, which reflects the energy dependence of the inelastic mean free path of electrons originating from an underlayer (here Co) through an overlayer (here Mn).¹¹ Series (1) is associated with $c = 2d_{\perp} = 3.46$ Å. Taking into account an ideal coherent growth of Co on Cu(001), this results in an axial ratio of $c/a = 0.957$, in excellent agreement with earlier independent studies using other methods.¹² For Mn [series (2)] we get $c = 2d_{\perp} = 3.77$ Å, or alternatively an axial ratio of $c/a = 1.043$, thus fct growth with an expanded c axis (e-fct). This c/a ratio is independent of both the Mn and the Co thicknesses used, indicating a metastable e-fct phase. Such a phase with $c/a = 1.048$ was actually predicted recently.⁷ Complementary XPD and XRD experiments resulting in $c/a = 1.055$ have shown that the kinematic LEED probably slightly underestimates c ,¹³ nevertheless all experiments unambiguously show that a metastable e-fct Mn phase indeed exists and that it can be stabilized on a suitable substrate such as, in our case, Co(001) coherently grown on Cu(001).

The specific layered structure which we used to stabilize e-fct Mn(001) on top of Co(001) offers also a unique possibility to trace the so far unknown magnetic characteristics of

FIG. 3. (Color online) Temperature dependence of the coercive (circles) and exchange bias (triangles) fields of 21 ML Mn/10 ML Co/Cu(001) as measured by SQUID magnetometry. For comparison, the coercive fields of Cu covered 10 ML Co/Cu(001) are shown (stars).

e-fct Mn. Thin fct Co(001) films are known to be ferromagnetic with T_C 's above 600 K and well-defined magnetic anisotropies for thicknesses above 3–4 ML.^{14,15} These properties not only permit the detection of a potential AFM order in the Mn via the direct interface exchange coupling but also make the determination of temperature dependent AFM properties possible. An AFM order of e-fct Mn is indeed demonstrated by the MOKE magnetization loops depicted in Fig. 2. The as grown state, measured along three in-plane high-symmetry directions [Figs. 2(a)–2(c)], is characterized by multicomponent loops, reflecting the magnetic domain state of the as grown Co (frozen in by the Mn overgrowth) in the sample region probed by the laser spot. In all directions loop shifts caused by the pinning of the Co domains, which have magnetization directions along the $\langle 110 \rangle$ easy axes, are observed. *This is a clear sign of AFM/FM exchange interaction and thus AFM order in e-fct Mn.* A homogeneous pinning direction is achieved by heating the sample above the blocking temperature (T_B) of the AFM/FM system with a subsequent cooldown in a magnetic field. We choose the $[-100]$ direction because it is also the field direction available in our SQUID. The room temperature loops of the field-cooled sample are plotted in Figs. 2(d)–2(f). They show that a well-defined exchange biased loop is observed when the field is applied along this direction a hard-axis loop is found.

The temperature dependence of the characteristic parameters of the exchange interaction, the coercive (H_C) and exchange bias shift (H_E) fields, are plotted in Fig. 3 for the same film as in Fig. 2. We utilized a 21 ML thick Mn layer because we have shown earlier, that this thickness is sufficient to approximate the “bulk” properties of e-fct Mn in which we are mainly interested in the present study.¹³ Thus, finite size effects will not play a role for this sample. Over the entire accessible temperature range, both distinct loop shifts and enhanced coercivities are detected. However, around 400 K a collapse of H_E is observed. By extrapolation of H_E to zero, a blocking temperature around 410 K can be estimated. This value is consistent with $T=435$ K being suf-

ficient in defining a homogeneous pinning direction (see Fig. 2).

However, T_B depends on several parameters, for example, the ratio between the magnetic AFM anisotropy and the exchange coupling strength, and is therefore not a completely intrinsic material property. A much more natural parameter describing an AFM is T_N , which in principle can also be determined from FM/AFM exchange coupling experiments. An AFM loses its ability to pin an FM at T_B ; however, the AFM/FM interaction does not vanish. As long as an AFM order exists, magnetic interface interactions will also remain, which will manifest themselves in enhanced H_C 's because the interface exchange interaction induces irreversible magnetization changes at the interface of the AFM. By inspecting Fig. 3 we notice that H_C decreases strongly up to RT and then levels off to a value which is an order larger than H_C found in Cu covered Co. Thus, H_C does not follow the collapse observed in H_E , indicating that T_N does not coincide with T_B and is certainly higher than 410 K. Unfortunately, the exact value of T_N was not accessible in our experiments.

Nevertheless we can conclude that in the present study e-fct Mn is an AFM with a reasonably high T_N exceeding RT. This has to be compared with α -Mn, the thermodynamically stable phase of Mn, where AFM order is only observed below 100 K. From an extrapolation of the magnetic properties of high-temperature quenched Mn rich $X_y\text{Mn}_{1-y}$ ($X=\text{Cu, Fe, Ni, Pd, } \dots$) bulk crystals to $y=0$, it can be inferred that fcc or fct Mn should have a T_N between 500 and 540 K.¹⁶ This value would be consistent with our results. More dedicated experiments should clarify this in the near future.

The importance of the fct Mn/Co rests in its striking simplicity. Both AFM and FM are single crystalline with a simple structure, are made of single elements, are coherently grown, and possess a well-defined interface. These properties make comparative theoretical and experimental studies attractive and feasible, which open opportunities to gain deeper insights into the still incompletely understood exchange anisotropy phenomenon.

- ¹J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999).
- ²A. E. Berkowitz and K. Takano, J. Magn. Magn. Mater. **200**, 552 (1999).
- ³S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
- ⁴V. Skumryev, S. Stoyanov, Y. Zhang, G. Hadjipanayis, D. Givord, and J. Nogués, Nature (London) **423**, 850 (2003).
- ⁵R. Jungblut, R. Coehoorn, M. T. Johnson, J. van de Stegge, and A. Reinders, J. Appl. Phys. **75**, 6659 (1994).
- ⁶F. Offi, W. Kuch, and J. Kirschner, Phys. Rev. B **66**, 064419 (2002).
- ⁷J. Hafner and D. Spišák, Phys. Rev. B **72**, 144420 (2005).
- ⁸J. T. Kohlhepp and W. J. M. de Jonge (unpublished).
- ⁹M. Wuttig, B. Feldmann, J. Thomassen, F. May, H. Zillgen, A. Brodde, H. Hannemann, and H. Neddermeyer, Surf. Sci. **291**, 14 (1993).
- ¹⁰J. P. Pendry, Low Energy Electron Diffraction (Academic, London, 1974).
- ¹¹M. P. Seah and W. A. Dench, Surf. Interface Anal. **1**, 2 (1979).
- ¹²H. Wieldraaijer, W. J. M. de Jonge, and J. T. Kohlhepp, Phys. Rev. B **72**, 155409 (2005).
- ¹³J. T. Kohlhepp and W. J. M. de Jonge, Phys. Rev. Lett. **96**, 237201 (2006).
- ¹⁴C. M. Schneider, P. Bressler, P. Schuster, J. Kirschner, J. J. de Miguel, and R. Miranda, Phys. Rev. Lett. **64**, 1059 (1990).
- ¹⁵P. Krams, F. Lauks, R. L. Stamps, B. Hillebrands, and G. Güntherodt, Phys. Rev. Lett. **69**, 3674 (1992).
- ¹⁶Y. Endoh and Y. Ishikawa, J. Phys. Soc. Jpn. **30**, 1614 (1971).