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Fine-tuning of the spin-density-wave state in Cr/V heterostructures via hydrogen uptake

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Abstract

We show that the magnetic state in rather thick Cr films can be finely tuned via hydrogen uptake into adjacent vanadium layers at rather low hydrogen pressures. By changing the hydrogen concentration and, hence, the electronic structure in the V layers, it is possible to affect the global properties of spin-density waves (SDWs) in Cr layers, including the SDW period and the Néel temperature. We provide direct experimental evidence that hydrogen uptake into V layers can be used to switch between incommensurate and commensurate SDW states in a reproducible way.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Spin-density-wave antiferromagnetism in thin Cr films and Cr-based heterostructures has attracted considerable attention during recent years [1–3]. Bulk Cr is an itinerant antiferromagnet and its low temperature behavior is associated with the existence of incommensurate spin-density waves (I-SDWs) below the Néel temperature T_N of about 311 K [4]. The I-SDW consists of a sinusoidal modulation of the amplitude of the antiferromagnetically ordered Cr magnetic moments with a wavevector determined by the nesting properties of the Cr Fermi surface and incommensurate with the bcc Cr lattice spacing. The I-SDW is accompanied by a charge-density wave and a strain wave (SW) corresponding to periodic modulations of the charge density and the lattice spacing, respectively,

which are also incommensurate with the bcc lattice periodicity of Cr. Elastic strains and chemical doping with elements increasing the electron concentration, for example, Fe or Mn, may cause the SDW to become commensurate (C-SDW) with the Cr lattice periodicity [4, 5].

Compared to the case for bulk crystals, Cr magnetism in thin films and heterostructures is conditioned by a number of additional factors. In Cr-based heterostructures, the SDW is confined and therefore the spontaneous formation of the I-SDW due to the Fermi surface nesting competes with interfacial effects, with proximity effects of adjacent layers, with additional epitaxial strains due to lattice misfits, and with dimensionality effects. Dimensionality effects are clearly seen in thin Cr layers, where the Cr layer thickness determines the I-SDW period and the number of periods per layer; in Cr layers thinner than one SDW period only a C-SDW phase can be observed [6–10]. Interfacial effects were mainly reported for Cr/ferromagnetic-metal systems, like Cr/Fe, Cr/Ni and

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Cr/Co, where the interface disorder and exchange coupling are responsible for the global SDW behavior in Cr layers [1–3, 11]. Additional epitaxial strains and lattice misfits reported for Cr/paramagnet systems (Cr/Cu [11], Cr/Pd [11], Cr/Ag [12]) tend to suppress the I-SDW phase in favor of the C-SDW one and, sometimes, to determine the SDW propagation direction. Proximity effects are usually manifested in a suppression or an enhancement of the Cr magnetic moment within interface regions causing pinning of the I-SDW nodes or antinodes, respectively, at the interfaces [9].

Cr/V is a unique system where the SDW state in Cr is conditioned by the hybridization between the very similar Fermi surfaces of Cr and V. In particular, the proximity effect of vanadium causes the occurrence of a magnetically dead layer near Cr/V interfaces [13–15] and strongly affects the global SDW properties, including the SDW polarization and propagation direction, the SDW period, and the Néel temperature [16, 17]. In the present paper, we provide direct experimental evidence for a novel method of fine-tuning the spin-density-wave state in Cr/V heterostructures via hydrogen uptake. Although surface electron states in Cr are known to be modified on hydrogen-covered Cr surfaces [18], hydrogen does not penetrate into Cr layers because Cr does not form hydrides under equilibrium conditions at small and medium hydrogen pressures. However, vanadium is a specific material easily absorbing hydrogen in a reversible way from the surrounding atmosphere and it is possible to affect the SDW state in Cr indirectly via hydrogen uptake into adjacent V layers by changing the Cr–V interface hybridization properties.

2. Experimental details

The present investigations were done on a [Cr(500 Å)/V(14 Å)]₄ multilayer grown with an UHV sputtering system on MgO(100) substrate. The sample was covered with a 30 Å thick catalytically active Pd layer in order to protect the sample from oxidation and to enhance the hydrogen uptake. Structural characterization done with high resolution x-ray reflectometry and wide-angle diffraction has revealed an excellent layer structure and a high degree of structural coherence [14]. The SDW behavior and its modification under hydrogen uploading have been determined by combined scattering experiments with neutrons and synchrotron x-rays. The x-ray experiments were performed at the magnetic scattering beamline ID20 of the ESRF (Grenoble, France) [19]. The incident photon beam was 99.8% linearly σ polarized in the scattering plane; the beam energy was selected to be 12 keV. The neutron scattering experiments were performed on the triple-axis spectrometer UNIDAS (Forschungszentrum Jülich, Germany) and the four-circle diffractometer D10 (Institut Laue-Langevin, Grenoble, France). The background radiation was sufficiently filtered out by using PG analyzer crystals fixed to zero energy transfer. The x-ray and neutron measurements were performed within the temperature range from 30 to 320 K for different hydrogen pressures up to 1 bar. To perform hydrogen uptake, the sample was exposed at room temperature within a closed cycle refrigerator with an external hydrogen reservoir via a small capillary. After filling the reservoir and, thus, the sample

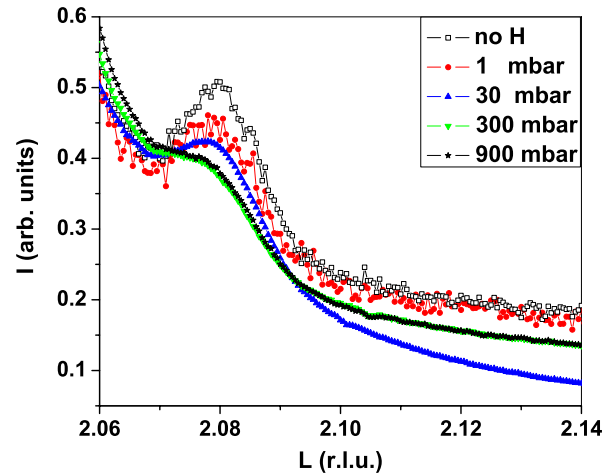


Figure 1. Synchrotron x-ray scans taken at temperature $T = 200$ K through the I-SW satellite position in the L direction around the Cr(002) fundamental Bragg position at different hydrogen pressures.

space with the desired hydrogen pressure, the sample was cooled down to a low temperature of about 30 K. The sample was exposed to hydrogen gas in the synchrotron experiments and to deuterium gas in the neutron experiments. This was done because of the large coherent neutron scattering length of deuterium and to avoid the incoherent neutron scattering background from hydrogen. Otherwise this choice should not make any difference to the Cr SDW results. Neutron and x-ray experimental procedures, corresponding scattering cross sections and selection rules can be found in our previous publication on Cr/V multilayers [14].

3. Results

The scattering results obtained with neutrons and synchrotron radiation on the SDW state in the sample and the full magnetic phase diagram without hydrogen have been reported elsewhere [14]. At low temperatures, there exists an I-SDW propagating in the out-of-plane direction, with Cr atomic spins being also out-of-plane oriented, corresponding to a longitudinal I-SDW. At higher temperatures of about 220–240 K, the I-SDW gradually switches to the commensurate phase with out-of-plane spins, which remains stable from 240 K up to temperatures well above the room temperature. Hydrogen uptake into vanadium layers affects the SDW state, especially at temperatures close to the Néel temperature for the I-SDW (240 K), by suppressing the incommensurate SDW phase. Figure 1 shows scans taken with synchrotron x-rays in the L direction through the SW satellite position around the Cr(002) fundamental Bragg position at temperature $T = 200$ K for different hydrogen pressures from 0 to 900 mbar. The second SW satellite $L = 1.92$ is not shown because it was significantly contaminated by a V(002) Bragg peak. The SW satellite position provides information on the SDW period, whereas the satellite intensity can be used as an order parameter to characterize the phase transition from the I-SDW to the C-SDW. As is evident from the figure, hydrogen uptake decreases the satellite intensity and shifts its position toward

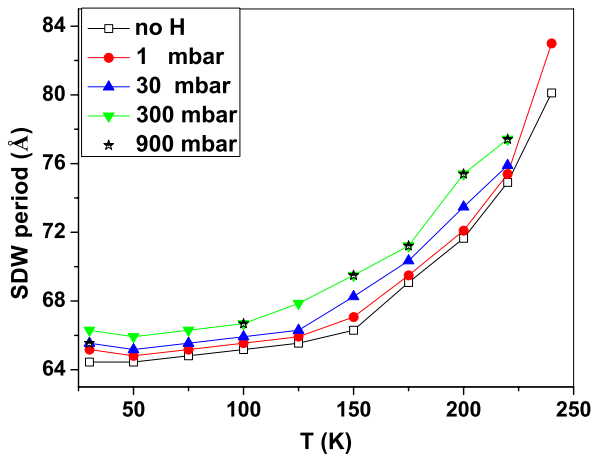


Figure 2. Influence of the hydrogen pressure on the temperature dependence of the SDW period as retrieved from synchrotron x-ray scattering experiments.

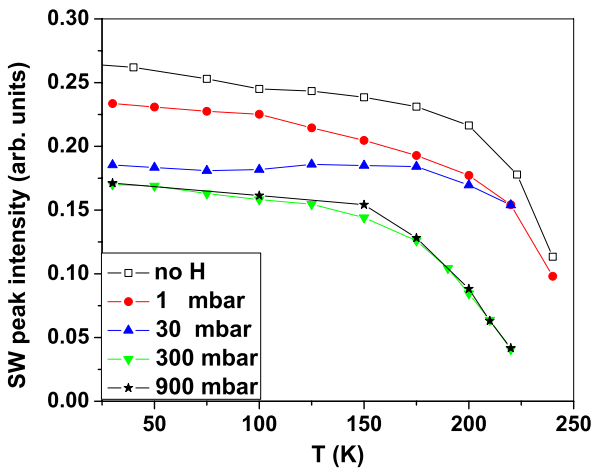


Figure 3. Influence of the hydrogen pressure on the temperature dependence of the SW peak intensity as retrieved from scattering experiments with synchrotron radiation. This quantity may be considered as an order parameter for characterizing the transition from an I-SDW to a C-SDW state.

the fundamental Cr(002) Bragg peak, which implies a decrease of the population of the I-SDW state and an increase of the I-SDW period. These measurements also show a clearly defined saturation effect: with further increase of the hydrogen pressure beyond 300 mbar the SDW state is not affected any more. Similar behavior but with different strength was detected over the entire temperature range from 30 K to the Néel temperature for the I-SDW. Figures 2 and 3 summarize the information on the influence of hydrogen uptake on the SDW period and the SW satellite intensity as deduced from our synchrotron x-ray scattering scans. In addition to the above mentioned effects, both diagrams also show that hydrogen loading affects the Néel temperature for the I-SDW phase by decreasing it from $T = 240$ K for the hydrogen-free state to $T = 220$ K for the hydrogen-saturated state.

The nature of the hydrogen-driven phase transition becomes clear from neutron scattering results. In the case

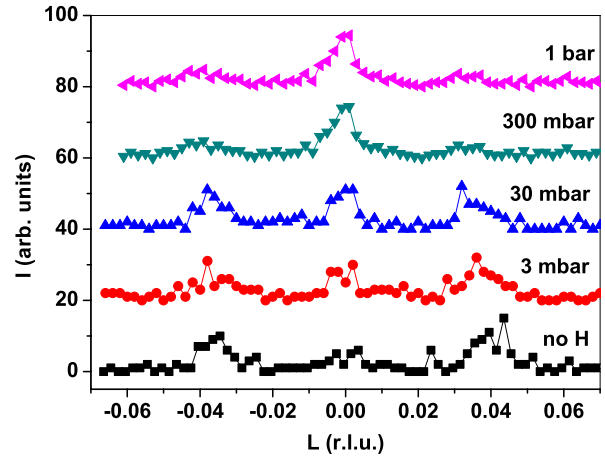


Figure 4. Neutron scattering scans taken in the L direction through the Cr(010) position at temperature $T = 220$ K. The satellites observed in the hydrogen-free state at about $L \sim \pm 0.04$ r.l.u. correspond to satellite peaks from the I-SDW. The peak at $L = 0$ is due to magnetic neutron scattering from a C-SDW or antiferromagnetic order. The scans show a smooth transition from the incommensurate to the commensurate SDW phase driven by the hydrogen uptake into vanadium layers.

of the out-of-plane longitudinal I-SDW, the antiferromagnetic order is manifested by SDW satellite peaks around the forbidden Cr(010) position in the L direction. Switching from the I-SDW state to the C-SDW state should be seen by the appearance of the corresponding peak at the antiferromagnetic Cr(010) peak position. Figure 4 shows neutron scattering scans taken at temperature $T = 220$ K in the L direction through the Cr(010) peak position. As is evident from the scan, in the hydrogen-free state the I-SDW dominates, with only a negligible amount of the commensurate SDW phase being observable. Hydrogen uptake changes the ratio between the incommensurate and commensurate phases and, finally, at a high enough hydrogen pressure the I-SDW phase disappears completely. The effect of the hydrogen-driven transition between these two phases is completely reproducible; after warming up the sample and repeating the hydrogen loading procedure, the SDW evolution with hydrogen uptake is completely reproducible.

4. Discussion

Summing up the above experimental results, we note the following hallmarks of the hydrogen-induced transition in Cr/V heterostructures. The hydrogen uptake into adjacent vanadium layers changes globally the SDW behavior in rather thick Cr layers. In particular, (i) it suppresses the I-SDW phase and promotes the C-SDW state; (ii) it increases the SDW period of the I-SDW phase; (iii) it causes a decrease of the Néel temperature for the I-SDW phase; (iv) it shows a saturation effect at a hydrogen pressure of about 100–300 mbar. Whereas at low temperatures far from the Néel temperature the hydrogen uptake causes just a little suppression of the I-SDW only, at higher temperatures the I-SDWs and C-SDWs start to coexist. When the temperature is

close enough to the Néel temperature for the I-SDW phase, it is possible to perform a smooth and reproducible transition from the I-SDW to C-SDW state with hydrogen uptake.

The observed hydrogen uptake effects are likely to be of electronic nature and not related to structural and strain effects. Epitaxial strains and interface effects, generally considered as governing factors in thin-film Cr systems [1], play a minor role in the observed phenomena. Hydrogen loading produces some additional strain in vanadium layers but it does not significantly affect the chromium layers. In V-based multilayers, like Fe/V, Mo/V, Cr/V, hydrogen is known to concentrate in the center of vanadium layers without causing any structural changes in adjacent layers. Usually a hydrogen depletion layer of about 2–3 ML in thickness is found near the interfaces [20]. During our synchrotron x-ray scattering measurements, we have traced the intensity and width of the fundamental Cr(002) Bragg peak and detected no change with hydrogen pressure, which excludes any significant additional strain caused by hydrogen in Cr. Similar observations were recently reported for other Cr/V multilayers [21]. Although we observed a good layer structure in the hydrogen-free state, some minor Cr/V interface imperfections are likely to be present, and hydrogenation may cause some additional degradation of the interface structure. However, interface effects can be easily smoothed out over the 50 Å magnetically dead regions near the Cr/V interfaces [14]. More likely, the observed effects are due to changes in the vanadium electronic structure upon hydrogen uptake. Hydrogen uptake increases the electron concentration in vanadium layers and shifts the V Fermi level due to the H 1s–V 3d hybridization [22]. This Fermi surface change upon hydrogen uptake may also modify the Cr–V 3d interface hybridization.

In fact, all the features reported here are very similar to those observed in bulk Cr crystals under chemical doping with elements which increase the electron concentration like Mn [5, 23, 24]. In contrast to chemical doping, hydrogen tuning shows a saturation effect, in our case at about 100–300 mbar, above which no further change in the Cr magnetic state is observed. This saturation effect is connected with the hydrogen solubility limit reported for thin vanadium films [25] and some V-based multilayers, including Cr/V superlattices [21]. Although hydrogen is localized only in the inner part of vanadium layers, the effect of the hydrogenation goes far beyond changes in the electronic structure of vanadium layers and Cr–V interface regions and includes a global change in the entire Cr layer of 500 Å in thickness. Thus the SDW state in the Cr layer is remotely controlled by the hydrogen in adjacent V layers. This is similar to the remote control that has been recently observed in Fe/V superlattices, where small amounts of hydrogen in the V layers have a dramatic effect on the magnetic moment of Fe [26]. It should be mentioned that an alternative model has been put forward that explains the incommensurate–commensurate transition upon hydrogen loading through a triggering effect, which allows the Cr spin structure to enter the antiferromagnetic ground state which otherwise is separated by an energy barrier [27].

We expect hydrogen loading experiments to open new ways to tune the SDW state in Cr-based heterostructures, and,

in particular, to trigger smooth and reproducible switching between incommensurate and commensurate phases. This property may also be useful in the design of new spin-valve devices.

5. Conclusion

In summary, we investigated the influence of hydrogen uptake on the spin-density-wave magnetism in Cr/V heterostructures. We show that hydrogen uptake into vanadium layers suppresses the incommensurate SDW phase and enhances the commensurate one in chromium layers. When the temperature is close enough to the Néel temperature for the incommensurate SDW phase, hydrogen loading makes it possible to switch reversibly between incommensurate and commensurate SDW phases in Cr.

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References

- [1] Zabel H 1999 *J. Phys.: Condens. Matter* **11** 9303
- [2] Pierce D T, Unguris J, Celotta R J and Stiles M D 1999 *J. Magn. Magn. Mater.* **200** 290
- [3] Fishman R S 2001 *J. Phys.: Condens. Matter* **13** R235
- [4] Fawcett E 1988 *Rev. Mod. Phys.* **60** 209
- [5] Fawcett E, Alberts H L, Galkin V Yu, Noakes D R and Yakhmi J V 1994 *Rev. Mod. Phys.* **66** 25
- [6] Fullerton E E, Riggs K T, Sowers C H, Bader S D and Berger A 1995 *Phys. Rev. Lett.* **75** 330
- [7] Fullerton E E, Bader S D and Robertson J L 1996 *Phys. Rev. Lett.* **77** 1382
- [8] Fullerton E E, Robertson J L, Prinsloo A R E, Alberts H L and Bader S D 2003 *Phys. Rev. Lett.* **91** 237201
- [9] Mibu K, Takeda M, Suzuki J, Nakanishi A, Kobayashi T, Endoh Y and Shinjo T 2002 *Phys. Rev. Lett.* **89** 287202
- [10] Jiko N, Mibu K and Takeda M 2005 *Phys. Rev. B* **71** 014414
- [11] Bödeker P, Schreyer A and Zabel H 1999 *Phys. Rev. B* **59** 9408
- [12] Demuynck S, Meersschaet J, Dekoster J, Swinnen B, Moons R, Vantomme A, Cottenier S and Rots M 1998 *Phys. Rev. Lett.* **81** 2564
- [13] Almokhtar M, Mibu K, Nakanishi A, Kobayashi T and Shinjo T 2000 *J. Phys.: Condens. Matter* **12** 9247
- [14] Kravtsov E, Brucas R, Hjörvarsson B, Hoser A, Liebig A, McIntyre G J, Milyaev M A, Nefedov A, Paolasini L, Radu F, Remhof A, Ustinov V V, Yakhov F and Zabel H 2007 *Phys. Rev. B* **76** 024421
- [15] Amitouche F, Issolah A, Bouarab S, Vega A and Demangeat C 2009 *Surf. Sci.* **603** 117
- [16] Kravtsov E, Nefedov A, Radu F, Remhof A, Zabel H, Brucas R, Hjörvarsson B, Hoser A and Wilkins S B 2004 *Phys. Rev. B* **70** 54425
- [17] Kravtsov E, Nefedov A, Zabel H, Brucas R, Hjörvarsson B, Hoser A and McIntyre G J 2005 *J. Phys.: Condens. Matter* **17** 3143
- [18] Rotenberg E, Krupin O and Kevan S D 2008 *New J. Phys.* **10** 023003

- [19] Paolasini L, Detlefs C, Mazzoli C, Wilkins S, Deen P P, Bombardi A, Kernavanois N, de Bergevin F, Yakhou F, Valade J P, Breslavetz I, Fondacaro A, Pepellin G and Bernard P 2007 *J. Synchrotron Radiat.* **14** 301
- [20] Hjörvarsson B, Rydén J, Karlsson E, Birch J and Sundgren J E 1991 *Phys. Rev. B* **43** 6440
- [21] Eriksson A K, Liebig A, Ólafsson S and Hjörvarsson B 2007 *J. Alloys Compounds* **446/447** 526
- [22] Andersson P H, Fast L, Nordström L, Johansson B and Eriksson O 1998 *Phys. Rev. B* **58** 5230
- [23] Yakhmi J V, Gopalakrishan I K and Iyer R M 1984 *J. Phys. F: Met. Phys.* **14** 923
- [24] Alberts H L and Lourens J A J 1987 *J. Phys. F: Met. Phys.* **17** 727
- [25] Bloch J, Hjörvarsson B, Olsson S and Brukas R 2007 *Phys. Rev. B* **75** 165418
- [26] Remhof A, Nowak G, Zabel H, Björck M, Pärnaste M, Hjörvarsson B and Uzdin V 2007 *Eur. Phys. Lett.* **79** 37003
- [27] Uzdin V M, Zabel H, Remhof A and Hjörvarsson B 2009 submitted