## Controlling Spin Density Wave Phases in Thin Cr(110) Films

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Using angle-resolved photoemission, we have mapped the spin density wave phase diagram of Cr(110) thin films grown on W(110) as a function of film thickness, temperature, and hydrogen surface coverage. We find commensurate and incommensurate phases that are separated by nearly continuous transitions. Surface localized electron states play a key role in the observed thin film phase behaviors. Our results indicate a delicate balance between spin density wave stabilization energy and surface and interface energetics that can be used to control the competition between commens urate and incommensurate phases.

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Chromium, the prototypical itinerant spin density wave (SDW) antiferromagnet [1, 2], displays rich and fascinating physics when confined to thin film structures. The richness derives from the fact that many low energy degrees of freedom – SDW phase and amplitude, incommensurability, propagation direction, and spin orientation – are dramatically impacted by strain, alloying, defects, doping, and interfacial exchange interactions. Previous studies were largely concerned with the latter effect due to technological interest in spintronic devices, and were confined to (100)-oriented films in which quantization of the SDW across the film thickness affects the SDW phase diagram [6–14].

Cr(110)-oriented films bonded to non-magnetic substrates have been less often studied but are potentially very interesting since interlayer magnetic exchange and SDW quantization [15–17] are not relevant. Weaker interactions can then dominate phase behaviors and the potential for external control is enhanced. Previously, we reported a temperature- and thickness-dependent phase transition between commensurate (C) and incommensurate (IC) phases in Cr(110) thin films, and argued that the transition was governed by a balance between interface and volume energetics; the detailed mechanism, however, was unclear[18].

In this Letter, we unambiguously show that the controlled modification of the surface electronic structure can drive such a transition. We show that hydrogeninduced changes to the surface electronic states of the outer Cr surface play an important role in determining the thin film SDW phase behavior, probably through changes to the magnetic susceptibility. Equivalent control could as well be achieved in a field-effect type device thereby suggesting an unusual level of control of a magnetic state with electric field.

Probing the SDW in Cr films less than 10 nm thick is difficult using conventional neutron [6, 7, 15] and x-ray scattering [19] due to the weak scattering signals available. We have recently shown that ARP is exquisitely sensitive to SDW periodicity while also probing the band structure and Fermi surface nesting that are key aspects of the underlying energetics [18]. Moreover, the use of a wedge-shaped thickness profile and real-time data collection as a function of hydrogen dose allows systematic variation of non-thermal as well as thermal parameters so that the full  $(T, d, \theta_{\rm H})$  phase diagram can be mapped.

Experiments were performed at the Electronic Structure Factory on beamline 7.0.1 at the Advanced Light Source as described previously [18, 20–22]. Cr was grown epitaxially on W(110) by room-temperature evaporation followed by rapid annealing to  $\sim 800$  K. The film thickness d was varied from 1.3 to 15 nm over a distance (5 mm) much larger than the 50  $\mu$ m photon beam used in ARP. These wedge-profiled films were transferred in vacuo onto a cooled goniometer for ARP measurements. Hydrogen was adsorbed up to 2 L saturation coverage at T=20-40 K and at pressure  $\sim 5 \times 10^{-9}$  T. Films displayed both strong surface state bands and quantization of those bulk states having significant perpendicular dispersion, ensuring cleanliness and atomic flatness of the interfaces. The base pressure was  $2x10^{-11}$  T, which increased to  $2x10^{-10}$ T during Cr evaporation.

Nesting along (001) directions between flat faces of  $\Gamma$ centered electron and H-centered hole octahedral Fermi surfaces, shown in Fig. 1(a) [3, 4], stabilizes the SDW in Cr [1, 2, 5]. Fig. 1(b) displays a momentum (k) distribution map at the Fermi level  $(E_{\rm F})$  in the  $\Gamma$ -H-P-N plane of the second bulk BCC Brillouin zone (BZ). These data were interpolated from a much larger data set in which the photoemission angles, the photon energy  $h\nu$ (from 120-200 eV), and the electron binding energy Ewere systematically varied, and represent truly planar cuts through the electron and hole octahedra and the Hcentered electron knob shown in Fig. 1(a). The Cr Fermi surface topology stabilizes IC SDWs because the associated backfolding vectors, illustrated in Fig. 1(b), are not commensurate with the lattice. The larger nesting vector is  $Q_{\rm F} \equiv \frac{2\pi}{a}(1+\delta_{\rm F})$ , where  $\delta_{\rm F}$  is the incommensurability defined by the Fermi surface dimensions. We determined  $\delta_{\rm F} = .05 \pm .005$ , in agreement with the incommensurabil-



FIG. 1: (a) The arrow indicates the nesting between the  $\Gamma$ -centered electron and H-centered hole octahedral Fermi surfaces stabilizes the SDW in Cr [1–5]. (b) Cr ARP intensity map at the Fermi level ( $E_{\rm F}$ ) in the  $\Gamma$ -H-P-N plane of the bulk BCC Brillouin zone, showing the nested flat faces from panel (a).

ity of bulk Cr [19]. As shown below, the actual SDW incommensurability,  $\delta_{\text{SDW}}$ , can depart significantly from  $\delta_{\text{F}}$  over large ranges of d and  $\theta_{\text{H}}$ .

ARP bandmaps, acquired at  $h\nu = 128$  eV along the dashed line in Fig. 1(b) (the surface  $\overline{\Gamma} \cdot \overline{S}$  line) show the E-k relationship of the direct and backfolded bands, whose splitting (multiplied by a geometric factor) gives  $\delta_{\text{SDW}}[18]$ . Figs. 2(a-b) show such ARP band maps for film thickness d = 3 nm and d = 6 nm, respectively, at T = 30 K. Panel (a) shows a single backfolded band characteristic of the C-phase  $(0 = \delta_{\rm SDW} \neq \delta_{\rm F})$ , which is unexpected for bulk Cr of high crystalline quality, while panel (b) exhibits two backfolded bands indicative of an IC-phase (0 <  $\delta_{\text{SDW}} = \delta_{\text{F}}$ ). Similar to other incommensurate systems [23, 24], only the two lowest order folded bands (out of the infinitely many present) exhibit measurable photoemission intensity. These data indicate that the IC phase is more stable in the thick-film limit, but some other energetic interaction stabilizes the C-phase in thinner films.

Fig. 2(c) shows the dramatic changes observed at d = 6 nm after saturating the Cr surface with a monolayer of hydrogen. Hydrogen adsorption removes some surface states and resonances and forces the emergence of a surface state from the band edge of the electron octahedron [25]. Such behavior is very similar to previous results on W(110) and Mo(110) [20, 21]. This charge rearrangement is accompanied by a transition from IC to C backfolding, which can be reversed by gently annealing the sample to remove H. These results indicate that hydrogen adsorbed on the outer surface extends the regime of C phase stability to thicker films.

The relationship between the changes in surface electronic structure and SDW commensurability can be seen more dramatically with images composed of momentum distribution curves (MDCs), shown in Fig. 3. Each MDC is a constant-energy slice from the backfolded region in a band map as in Fig. 2. An intensity map composed of such MDCs shows the evolution of the backfolding as a function of a non-thermal parameter, either d or  $\theta_{\rm H}$ . Fig. 3(a) shows the smooth evolution of the splitting of the backfolded bands, and thus of  $\delta_{\rm SDW}$ , as a function of



FIG. 2: ARP bandmaps at  $h\nu = 128$  eV, T=30K along the  $\overline{\text{S}}-\overline{\Gamma}-\overline{\text{S}}$  line in the surface BZ for film thickness (a) d = 3 nm and (b) d = 6 nm showing single (double) backfolding representing commensurate (incommensurate) phases, resp. (c) shows the commensurate sample at d = 6 nm after adsorption of a monolayer of H on the Cr surface.

d for T = 120 K and  $\theta_{\rm H} = 0.2$  L. As discussed above, in the thick films the observed splitting corresponds to the bulk incommensurability, but this evolves smoothly to zero as the film thickness is reduced. The thickness where the incommensurability goes to zero is a point on a phase line, as discussed further below.

Fig. 3(b) shows the evolution with H coverage for T= 45 K and d = 6 nm for the MDC at ~110 eV below  $E_{\rm F}$  cutting through the surface state, the SDW backfolded bands, and the bulk electron pocket band. Fig. 3(c) shows the same data after subtracting a constant bulk contribution from each slice, so that the hydrogendependence of the states is more visible. As in Fig. 2(b), the clean surface exhibits an IC-SDW, and this evolves smoothly to the C-phase as hydrogen is adsorbed. Most importantly, a simultaneous evolution in surface electronic structure is also readily apparent in Fig. 3(b). A sharp surface state (SS) that is nearly degenerate with the edge of the bulk electron octahedron on the clean surface moves into a large projected bulk band gap as hydrogen is adsorbed. The SS broadens significantly during the IC $\rightarrow$ C transition. This evolution of surface electronic structure occurs over precisely the same coverage range where the incommensurability changes most rapidly.

The images in Figs. 3(a-c) graphically demonstrate the coupled dependencies of SDW commensurability on sur-

face electronic structure, film thickness, and temperature. The underlying phase behaviors are given in Fig. 3(d), which plots the temperature  $T_c$  at which the C-IC transition occurs as a function of d at three different hydrogen coverages  $\theta_{\rm H}$ . The  $\theta_{\rm H}=0$  plane corresponds to the  $\delta_{\rm SDW}(T, d)$  results published recently [18] and clearly show the (large d, low T) and (small d, high T) preferences for the IC- and C-phases, respectively. Adsorption of hydrogen stabilizes the C-phase and shifts the phase line approximately rigidly to larger thickness.

The above discussion assumed that the backfolded bands reflect the true bulk SDW incommensurability, in spite of the well-known surface sensitivity of ARP. Several observations support this assumption. First, a scan of the photon energy proves that the primary and backfolded states are bulk states. Second, these bands are very similar to the bulk bands of the isolectronic Mo(110)and W(110) surfaces we have studied [20-22, 26, 27]. Third,  $\delta_{\text{SDW}}$  converges on the known bulk incommensurability. Fourth, we have recently prepared  $Cr_{1-x}V_x$  alloys, and the composition-dependence of the incommensurability in thick films is similar to that in bulk alloys [25]. Fifth, we do not see evidence for SDW-related gaps of the surface states, so there is no distinct surface magnetic periodicity [25]. Finally, we have reproduced the results at many equivalent bulk BZs from photon energy 21.2 through 500 eV and at various angles. This spans several BZs and a broad range of photoemission sampling depth. To interpret all of these results in terms of an exotic surface magnetic phase would be very difficult.

Two arguments also support that we probe bulk states. First, any incommensurate  $\langle 100 \rangle$ -oriented SDW intersects a (110) interface with a spin structure that is itself incommensurate [17, 18]. That will make the coupling between surface and bulk spin structures very stiff since any change in spin structure near the surface will require many unfavorable local exchange interactions. Lastly we argue that ARP probes electrons from the outermost one or two layers including bulk states provided that their wavefunctions have sufficient overlap with the surface, which is almost always the case for ordinary metals.

The expected energetic stabilization of the IC phase due to gap opening is small (<1 meV per atom [28]) and can be easily balanced by one or more surface, interface, or thin-film effects favoring the C phase. These factors include interface exchange coupling, strain, evolution of the nesting vector, defects, quantum well (QW) state formation, and interfacial electronic effects. Of these, interface exchange coupling is unlikely since W and H are non-magnetic. The next four factors are potentially thickness-dependent and should be considered as possible stabilizing mechanisms for the C phase. We have already discussed in detail why the thickness dependence cannot be explained by these so we focus on the H-dependence here. H adsorption is unlikely to strain the film, nor do we observe a change in the bulk nesting vectors. QW



FIG. 3: Momentum distribution in the SDW-backfolded region in Fig. 2. These show the smooth evolution of the SDW incommensurability with (a) film thickness d at constant  $(T, \theta_{\rm H})$  and (b) hydrogen coverage  $\theta_{\rm H}$  at constant (T, d). (c) same as (b) with bulk contribution subtracted. Where the two backfolded bands merge to form a single band is a point on a C-IC phase diagram, and these points are collected to produce the phase lines in panel (d).

states are expected to be unaffected by H coverage. Surface disorder induced by H can be ruled out because at the highest coverages we expect a defect-free, saturated surface.

The clear connection between surface electronic structure and  $\delta_{\text{SDW}}$  proves that the electronic structure at the free Cr surface plays a major if not determining role in the C-IC energetics. To forge a precise understanding will require a detailed first principles calculation, though very likely there must be a significant interaction between surface electronic and spin degrees of freedom. (A recent paper suggests that electron-magnon coupling might be strong on this surface, though we are unable to reproduce those results in detail and the observed coupling was to the bulk electron octahedron in any case [29].)

One possible mechanism that can favor the C-phase is that the magnetic susceptibility function  $\chi(q)$  at the surface or interface can be singular at a commensurate q value. While we cannot probe the interface electronic structure to test this, our experiments directly test this notion at the surface. In Fig. 4 we show the k- and  $\theta_{\rm H}$ -dependence of the surface Fermi contours. The clean Fermi surface contours at  $\overline{S}$  have a tilted elliptical shape with no apparent nesting vector favoring the C phase. With H coverage, however, the surface states at  $\overline{S}$  disappear and a second surface state (shown in Figs. 2-3) splits off from the bulk bands, eventually forming the race-track shaped contours around  $\overline{S}$  shown in Fig. 4. These are oriented along the [001] direction and therefore have considerable nesting at the C-SDW wavevector. This may provide sufficient screening to the second and deeper layers to favor the C phase over the IC phase.

Another possible contribution to C-phase stability is surface magnetic anisotropy. The C-phase SDW produces a c(2x2) magnetic superstructure at a (110) interface, wherein the spin density alternates between neighboring atoms. By contrast, the (110) plane samples all



FIG. 4: Momentum distribution at  $E_{\rm F}$  for (a) clean and (b) hydrogen-covered Cr(110). The commensurate nesting vector C connects poorly-nested and well-nested surface Fermi contours (dotted lines) in (a,b) respectively.

spin densities in the SDW in the IC phase. Therefore, the C-phase can naturally minimize the surface magnetoanisotropy energy, while the IC phase cannot. However, other influences of the interfacial electronic structure, such as spin-orbit coupling at the Cr-W interface [27], cannot be ruled out. Nor can we comment on the possible role of the spin flip transition at T=123K in bulk Cr.

A key feature of our results is the ability to control the magnetic structure of a thin film with an external perturbation. This is a prime goal of many spintronic technologies, though of course hydrogen adsorption on a clean surface is not a very suitable control mechanism in real devices. The evolution of the electronic structure implies significant charge rearrangement in the surface layers, however, so a better way to think about achieving useful control might be in terms of a field-effect device. The notion of controlling a thin film magnetic phase *via* the impact of an electric field on the surface magnetic anisotropy, operating through an electron-spin interaction, appears to be new and worth pursuing.

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