Surface states and spin density wave periodicity in Cr(110) films

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Abstract. Using angle-resolved photoemission, we have mapped the dispersion relations and Fermi contours for surface-localized electron states onto clean and hydrogen-covered Cr(110) surfaces. In particular, we have probed the relationship between hydrogen adsorption and the evolution of the spin density wave (SDW) periodicity in chromium thin films observed previously. We find qualitatively similar surface band dispersion relations to those on W(110) and Mo(110), although with a narrower bandwidth, broader spectral features, and a smaller impact from the spin–orbit interaction. We compare our results to existing first-principles calculations and find a significant disagreement for a surface band that produces a prominent surface Fermi contour. Upon hydrogen adsorption, the Fermi contour for a particular surface band becomes well nested at a wave vector that stabilizes a commensurate SDW. We suggest that a competition between commensurate two-dimensional (2D) and incommensurate 3D Fermi surface nesting plays an important role in the SDW energetics in thin Cr(110) films.
Electronic surface states on nominally clean surfaces continue to be of significant interest because of the many surface properties they impact [1]. This is particularly true on surfaces that exhibit ordered magnetic structures since the surface states can contribute to the generalized susceptibility and thereby help to stabilize a particular spin periodicity. An excellent example is offered by chromium surfaces since surface states might alter the structure and periodicity of the bulk spin density wave (SDW) in the vicinity of the surface. Early photoemission experiments and associated calculations, for example, suggested that the Cr(100) surface might have a net magnetic moment, despite bulk chromium being antiferromagnetic [2]–[6]. This result was at odds with previous spin-resolved photoemission results [7] and with more recent scanning tunneling microscopy experiments [8, 9]. These observe the nominally expected layered antiferromagnetic structure that is intimately connected to spin-polarized surface-localized electron states. More complicated phenomena can occur on Cr(110) due to the oblique orientation of the bulk SDW relative to the surface plane [10]–[14]. We have recently shown that hydrogen-induced changes in electronic structure at the surface of a Cr(110) film are directly correlated to the commensurability of the SDW within the film [15].

In this paper, we examine in greater detail the surface states and surface Fermi contours on clean and hydrogen-covered Cr(110) films grown on W(110) using angle-resolved photoemission (ARP). We find surface bands with dispersion relations that are qualitatively similar to those characterized previously on W(110) and Mo(110) [16]–[19], although their photoemission intensity on Cr(110) is significantly lower and their widths are larger. The correlation between SDW periodicity and hydrogen-induced changes is associated with the evolution of a particular surface state-nesting vector and thus is related to singular behavior of the generalized spin susceptibility in the vicinity of the surface. Similar to Mo(110) and W(110) [16, 18, 20], the Fermi contour of this band undergoes a topological transition upon hydrogen adsorption. We compare our measured dispersion relations to existing calculations [21, 22] and find that some bands are well-described by theory, while others are not.
2. Experimental procedures

Experiments were performed at the Electronic Structure Factory end station at beamline 7.0.1 at the Advanced Light Source at Lawrence Berkeley National Laboratory. Our procedures are very similar to those described previously [14, 18, 21]. We include results using soft x-ray synchrotron radiation and He-I UV radiation provided by a Specs UVS 300 noble gas resonance lamp. High quality epitaxial Cr films were grown on W(110) by room-temperature thermal evaporation followed by rapid annealing to ∼800 K. Our previous results have shown that film thickness plays a key role in determining SDW periodicity [14]. In most of the cases, films were deposited with a wedge-shaped thickness profile (1.3 < d < 15 nm) over a distance (5 mm) much larger than the 50 µm soft x-ray photon beam so that film thickness could be selected as desired. After preparation, the films were transferred in vacuum onto a liquid-He-cooled sample goniometer (20 < T < 300 K) for ARP measurements. The hydrogen was supplied by exposing the cooled crystal to 99.995% purity H₂ at a partial pressure of 5 × 10⁻⁹ Torr, during which time ARP data were continuously collected. A total exposure of 2 × 10⁻⁶ Torr-sec provided saturation coverage. Films displayed both surface state bands and strong quantization of bulk states having significant perpendicular dispersion, ensuring cleanliness and atomic flatness of the Cr-vacuum and the Cr-W interfaces. The base pressure of our vacuum system was typically 2 × 10⁻¹¹ Torr, though the pressure increased by about an order of magnitude during chromium evaporation and during operation of the resonance lamp.

The incident light polarization lay in a horizontal plane formed by the light path and electron analyzer (Gammadata R4000) axis, which were separated by 60°. This analyzer measures in parallel an angular range as much as 30° in the horizontal plane and an energy window dependent on the analyzer pass energy. Our sample holder had three precision angular degrees of freedom: the polar angle θ of the surface normal relative to the energy analyzer, azimuthal angle φ about the normal, and tilt of the normal out of the horizontal plane (β). All of these angles driven with stepper-motors and are under computer control. The precision is typically 0.1°. By varying hv, β, and θ at fixed φ, and converting to momentum coordinates by standard methods, we could sample arbitrary cuts through energy-momentum space.

3. Results and discussion

3.1. Surface state dispersion relations

First, we show how bands measured along a particular azimuth direction (figure 1) can simultaneously probe not only the relevant surface and bulk bands but also the SDW commensurability. Figures 1(a) and (b) exhibit ARP band maps of the clean and hydrogen-covered Cr(110) surface collected at a photon energy of 128 eV along the Γ → S line of the surface Brillouin zone (SBZ). As discussed previously [14], angular scans at this photon energy probe along a spherical surface through the electron and hole octahedra of the bulk chromium Fermi surface. As indicated in figure 2, the Γ → S line projects states from a plane passing through the flat faces of the Γ-centered electron octahedral segment of the bulk chromium Fermi surface [14, 22–25]. Along (100)-directions, these flat faces are well-nested to the faces of H-centered hole octahedral segments, and this nesting drives the incommensurate bulk chromium SDW [26]–[28]. The bulk band labeled B that disperses upward from the zone.
Figure 1. Band maps of a 5 nm thick Cr(110) film, collected from the (a) clean and (b) hydrogen-covered surfaces at $T = 30$ K. The photon energy ($128$ eV) was set to sample approximately the intense bulk band $B$ that forms the electron octahedron in the bulk Fermi surfaces. Note the weak backfolding of this band on Cr(110) as it approaches the Fermi level, indicating incommensurate (commensurate) SDW formation for the clean (hydrogen-covered) surface. Surface-localized bands, labeled as described in the text, appear at larger wave vector in the projected band gap. The fine-grain hexagonal structure, most evident near $k_{\parallel} \approx 0$, is an artifact of our channel plate detector.

The center $\bar{\Gamma}$ point in figure 1 forms the electron octahedron as it crosses the Fermi level $E_F$. Weak backfolded features observed beyond this crossing point indicate the presence of an SDW and associated band gap, as discussed previously and further below [14, 15]. These maps were collected from a film of thickness of 5 nm and at a temperature of 30 K. This places the SDW in the incommensurate (double back-folding, IC/SDW) and commensurate (single back-folding, C/SDW) phases for the clean and hydrogen-covered surfaces, respectively.

Outside the electron octahedron, i.e. beyond the Fermi level crossing discussed above, there is a large projected bulk band gap that supports well-defined surface states on the group VI-B surfaces [16]–[19]. For example, band maps along the $\bar{\Gamma} \rightarrow \bar{S}$ line of clean and hydrogen-covered W(110) surface are presented in figures 3(a) and (b). These were collected at the He-I photon energy of 21.2 eV, where several of the surface states and resonances have high photoemission intensity. Surface states ($S_1$, $S_2$ and $A$) and a surface resonance (SR), as labeled previously [18], are easily visible. The SR disperses downward from the Fermi level away from the zone center and meets the pair of states $S_1$ and $S_2$ close to the edge of the projected band gap. These two are practically degenerate and are close to the bulk band $B$ on the clean surface, but split and move away from the zone center as hydrogen is added to the surface, as shown in figure 3(b). These two bands are derived from a single band that has been split by the spin–orbit interaction [19, 29]. The more highly dispersive surface state $A$ is split off
Figure 2. Upper panel: the calculated bulk chromium Fermi surface segments, full on the left and sectioned on the right, in the bulk body-centered cubic Brillouin zone [13]. Nesting between the flat faces of the Γ-centered electron (red) and H-centered hole (green) octahedra leads to the incommensurate SDW ground state at wave vectors given by \( Q_F \) that are slightly different from the wave vector of the bulk H point. Lower panel: the calculated bulk Fermi surface [25] projected on to the bcc (110) SBZ. The flat faces of the bulk electron octahedral segments project near the \( \bar{\Gamma} \rightarrow \bar{S} \) line of the SBZ. The unshaded regions represent a large projected band gap populated by surface-localized states.

from the bulk ellipsoidal Fermi pocket (yellow in figure 2) [18, 19]. This state disappears upon hydrogen adsorption, as does the SR near the zone center.

Figures 1(a), (b), 3(c) and (d) indicate that analogous bands are also present on Cr(110), though with substantially weaker intensity and larger line width than on W(110). We have checked our assignments at other photon energies and we find that these are indeed two-dimensional (2D) states. The \( S_1 + S_2 \) pair is not measurably split on Cr(110) due to the much weaker spin–orbit interaction for a 3d compared to a 5d metal. One qualitative difference between the Cr(110) and W(110) surface electronic structure is a feature close to the Fermi level near the center of the SBZ observed on the latter but not the former. This is shown explicitly in figures 3(a) and (c), where a narrow band very close to the zone center oscillates about \( E_F \) on Cr(110), but is not observed on W(110). This feature is also a SR and is labeled SR′.

The evolution of the surface states and the SDW commensurability with hydrogen dose is illustrated in movie 1, composed of band maps collected from Cr(110) at \( h \nu = 128 \text{ eV} \) for hydrogen coverages from clean (figure 1(a)) to saturated (figure 1(b)). This movie illustrates the rapid disappearance of state A, the coupling between the evolution of \( S_1 + S_2 \), and the change in SDW commensurability, all induced by hydrogen exposure. In the absence of an SDW, the surface bands A and \( S_1 + S_2 \) lie in the projected band gap of body-centered cubic (bcc) chromium and would therefore be true surface states. In reality, the projected band gap does not exist in the presence of the commensurate or incommensurate backfolding of the bulk Brillouin zone.
Figure 3. Band maps of the (a) clean and (b) hydrogen-covered W(110) substrate, and (c) clean and (d) hydrogen-covered 10 nm Cr(110) film, collected at a photon energy of 21.2 eV and $T = 30$ K. Various surface and bulk bands are indicated, as described in the text. The chromium band widths are narrower and the features are broader than for W(110). Note, however, the overall similarity between these two, except for the presence of an extra SR, SR', near the Fermi level at the zone center on Cr(110).

and SBZ. For this reason, the $S_1 + S_2$ pair and the state $A$ are more precisely called weakly coupled SRs. The SDW-induced coupling between surface and bulk states is probably weak, and we will continue to present our results in the full bcc Brillouin zone.

The line widths of the surface states on Cr(110), particularly of $S_1 + S_2$, are systematically larger in energy and momentum than the corresponding states on W(110), and, most unusually,
Movie 1. Available from stacks.iop.org/NJP/10/023003/mmedia. Movie of band maps collected from Cr(110) as a function of hydrogen dose at a photon energy of 128 eV, as in figure 1. On the clean surface, state $A$ is visible near the $\bar{S}$ point, but this disappears quickly upon H-adsorption. The $S_1 + S_2$ pair is nearly degenerate with the intense bulk band edges on the clean surface, but moves to higher $k_\parallel$ into the projected band gap upon hydrogen adsorption. Note also the simultaneous evolution of the bulk band from double (IC) to single (C) backfolding upon hydrogen adsorption, as discussed previously [15]. The diffuse structures for $k_\parallel < -1 \text{Å}^{-1}$ reflects bands sampled in the second SZB. The 3D bulk states are not equivalent to those near $k_\parallel = 0$ due to variation of the perpendicular momentum, which makes the observed bands notably broader, too [14]. The 2D surface state is equivalent to that near $k_\parallel = 0$ due to reciprocal lattice symmetry and experiences the same evolution upon hydrogen exposure.

are also much broader than the bulk Cr states from which they are derived. We seek an exotic, possibly magnetic source of surface state broadening because the usual sources of broadening can be ruled out as follows.

Firstly, we considered whether there is a reduced crystallinity of the Cr(110) film than of W(110), which might follow from their large lattice mismatch and consequent high density of dislocations in the film. However, we have shown previously that our annealed chromium films have large and flat terraces that are conformal with the underlying W(110) substrate for thickness greater than $\sim 1.3 \text{nm}$ [14]. Those results suggest a similar step density and thus a similar crystalline quality. The bulk Cr states are quite sharp indicating good crystalline order within the films.

Secondly, we investigated whether there is a reduced Cr(110) surface order. Low energy electron diffraction beams from Cr(110) are nearly as sharp as those of the W(110) substrate, suggesting good surface quality. Surface contamination could also cause broader than expected photoemission features, though we have found no evidence for contaminants using core level spectroscopy. Absorbed hydrogen (i.e. into the bulk of our film) is a possible source of surface disorder since unlike tungsten, stable bulk chromium hydrides are known to exist. However, formation of such bulk hydrides is unlikely considering the low sample temperature ($\sim 20 \text{K}$) during H exposure and furthermore, the fact that the hydrogen coverage on Cr saturates at a similar coverage as on W. Another possibility is that the adsorbed hydrogen atoms are intrinsically more disordered on Cr(110) than on W(110). Quantum well states observed in other regions of $k$-space remain sharp after hydrogen exposure, suggesting high surface quality with and without hydrogen.

Finally, we considered the electron–phonon coupling. The observed broadening does not behave like the electron–phonon coupling observed in related systems, where the effect is much smaller and is temperature-dependent [30, 31]. We conclude that the conventional electron–phonon coupling cannot explain our results.

Possibly the excess broadening is due to electron–magnon scattering. The magnon modes in bulk chromium are complex and are not entirely understood [26], so it is difficult to devise a simple model that might be applied to our results. Moreover, it is not clear why magnons would couple so much more strongly to the $S_1 + S_2$ surface state than to the bulk state $\beta$ from which it splits. Possibly the magnetic excitations in our films are very different from those in
3.2. Comparison of experimental and calculated dispersion relations

Two recent first-principles calculations of the electronic structure of clean Cr(110) slabs can distinguish surface-localized states long the $\bar{\Gamma} \rightarrow \bar{S}$ direction [21, 22]. In one of these, Schiller et al [22], include more chromium layers and distinguish the surface bands more completely, so we compare our results to that calculation. The calculation assumed a commensurate SDW phase, which corresponds to a simple cubic bulk unit cell, though results are plotted in the bcc SBZ. Our dispersion relations for the clean Cr(110) surface have been collected both for thin films, which exist in the commensurate SDW phase, and for thicker films, where the SDW is incommensurate [14, 15]. We observe a little difference between the resulting dispersion relations for these two phases.

In Figure 4(a), we present a summary of our measured dispersion relations for clean Cr(110) along the $\bar{\Gamma} \rightarrow \bar{S}$ line from band maps like those in figure 1. Also included are calculated surface band dispersions [22] (red lines), and a projection of the bulk band structure of Papaconstantopoulos (gray) [25]. While the surface calculation predicts the dispersion
relations of band $A$ and the feature SR’ near the zone center at the Fermi level fairly well, the calculation does not correctly place SR or $S_1 + S_2$ on clean Cr(110). The connectivity of these bands is complicated and warrants some discussion. On clean W(110) (figure 3(a)), these two seem to merge below the Fermi level and could be counted as a single band that disperses from the continuum into the band gap. On clean Cr(110) in figure 3(c), the band maps are less clear, but SR is also apparently connected to $S_1 + S_2$, and the latter is measured to be essentially degenerate with the bulk band edge $B$. In contrast, the calculation for clean Cr(110) predicts bands that are similar to SR and $S_1 + S_2$, though located much too far out in the SBZ relative to the measurement. Upon hydrogen adsorption, the connectivity between SR and $S_1 + S_2$ is altered: the SR band disappears, leaving a shifted $S_1 + S_2$ on Cr(110), as in figures 1(b) and 3(d), and movie 1, or $S_1$ and $S_2$ on W(110), as in figures 1(b) and 3(b). These disperse monotonically upward from the zone center through the projected band gap to the Fermi level. It is perhaps illuminating that the surface band calculations that were used to support a ferromagnetic Cr(001) surface were also apparently in error [2]–[6].

Figure 4(b) presents our measured dispersion relations for the hydrogen-covered Cr(110) surface, again on a projection of the bulk band structure from Papaconstantopoulos [25]. As discussed above, surface state $A$ and the SR are quenched by hydrogen exposure, while $S_1 + S_2$ have moved into the projected band gap, extending from an intense bulk band located well below $E_F$ near the zone center. The SR’ resonance also survives hydrogen coverage, moving about 50 meV to higher binding energy. To our knowledge, the electron states for this surface have not been calculated.

3.3. Surface Fermi contour topology

Our previous work on this system suggested that the interplay between film thickness, hydrogen coverage, and SDW commensurability was driven by a competition between bulk (3D) and surface (2D) energetics [14, 15]. We now discuss this interplay in more detail.

Nesting of the bulk chromium Fermi surface, specifically, between the flat faces of the $\Gamma$-centered electron and H-centered hole octahedra shown in figure 2, provides the well-known driving force for an incommensurate bulk SDW. This nesting is slightly incommensurate because the hole octahedron is slightly larger than the electron octahedron, so these are connected by a vector that is $\sim 4\%$ different from the distance between the $\Gamma$ and H points. We previously suggested [14, 15] various candidate surface, thin film, or interface effects that might favor a commensurate ground state, one of which was nesting of surface Fermi contours. In this mechanism, the surface Fermi contours are nested by a vector exactly equal to the distance between the $\Gamma$ and H points in the bulk Brillouin zone, or equivalently, between the $\bar{\Gamma}$ and $\bar{N}$ or $\bar{S}$ and $\bar{S}$ points parallel to the $\bar{\Sigma}$-axis of the SBZ (see figure 2). Since the bulk C phase projects on to the surface as a $c(2 \times 2)$ spin structure [14, 15], we seek a $c(2 \times 2)$ nesting of the surface state Fermi contours. Various SBZs are shown schematically in figure 5, and the relation to the bulk Brillouin zone is given in figure 2.

The bands $A$, SR, SR’ and $S_1 + S_2$ all are located near $E_F$ and each forms a separate surface Fermi contour. Figures 5(a) and (b) show ARP Fermi level intensity maps of clean and hydrogen-covered Cr(110). These were collected over a range of photon energies to allow us to interpolate approximately planar sections through the $\Gamma$-H-P-N plane of the bulk Brillouin zone, and over a large range of angles so that several SBZs are sampled. Lighter colors in these maps indicate bands crossing at $E_F$, and thus correspond to segments of the 3D Fermi surface for
Figure 5. Photoemission intensity maps of (a) clean and (b) hydrogen-covered Cr(110) collected at the Fermi level in an approximately planar slice centered on the $\Gamma$-H-P-N plane of the bulk Brillouin zone, and over a large range of reciprocal space so that several SBZs are sampled. Reciprocal lattice symmetry is clearly visible so that the regions inside the red rectangles, which correspond to two SBZs, could be extracted and displayed in figure 6 for further image analysis using periodic boundary conditions. These images were collected from a 10 nm thick film at $T = 30$ K grown on W(110), though the surface bands and Fermi contours are insensitive to film thickness. The rhomb-shaped features at $\bar{\Gamma}$, $\bar{\bar{H}}$, and $\bar{\bar{N}}$ correspond to spherical slices through the bulk Fermi surface octahedra. For clarity of presentation, the symmetry labels and labels of the various Fermi contours are also given in figure 6.

bulk states or 2D Fermi contour for surface states and resonances. As explained elsewhere [14], the intense rhomb-shaped contours at $\bar{\Gamma}$, $\bar{\bar{H}}$ and $\bar{\bar{N}}$ (defined in figure 6(a)) correspond to the bulk electron and hole octahedra and can be used to determine the incommensurate Fermi surface nesting vector that drives the SDW transition, shown as $Q_F$ in figure 2.

For the autocorrelation analysis presented below, we have extracted rectangular sections indicated in figures 5(a) and (b) and presented these in figures 6(a) and (b), respectively. We have labeled this rectangular segment as a ‘$(1/2 \times 1)$’ SBZ since it corresponds to an artificially halved real space cell. Also visible in these figures are surface contours that have been labeled, in figure 6, by their association with the bands in figures 1 and 3. The elliptical structure associated with band $A$, for example, is visible near $\bar{\bar{S}}$ on the clean surface but disappears upon hydrogen exposure. Other closed elliptical structures associated with SR near the zone center $\bar{\Gamma}$ point and near the zone boundary $\bar{\bar{N}}$ point as well as triangular structures associated with SR’ near the zone center have very low intensity at this photon energy but can be observed at the He I resonance energy (not shown). Of greater interest is the contour associated with $S_1 + S_2$, since
Figure 6. Rectangular (1/2 × 1) zones of the Fermi maps for (a) clean and (b) hydrogen-covered Cr(110) indicated in (a) and (b), respectively. These rectangular sections encompass precisely two (110) SBZs and maintain the full SBZ symmetry, but they are more easily analyzed than the distorted hexagonal (1×1) SBZ. The curved green lines are guides to the eye that help indicate the evolution of the various surface Fermi contours upon hydrogen adsorption. Elliptical hole pockets centered at $\bar{\Gamma}$ and $\bar{S}$ are quenched upon hydrogen adsorption.
Figure 6. (continued) exposure. The surface band $S_1 + S_2$ is essentially degenerate with the edge of the electron octahedron on the clean surface, but moves into the projected bulk band gap upon hydrogen exposure and undergoes a topological transition to form an elliptical orbit around the $\bar{S}$ point, as in (b). Panel (c) is the difference between the images in (a) and (b). This difference emphasizes the surface-related features, particularly the $S_1 + S_2$ contours orbiting the $\bar{S}$ points. Nesting vectors $Q_A$ through $Q_E$ are discussed in the text. Panel (d) shows the autocorrelation of (c) and identifies various nesting vectors shown in both panels. Accidental mirror symmetry of the $S_1 + S_2$ contour upon hydrogen saturation leads to nearly perfect nesting of contours across the $\Delta$ and $\Sigma$ lines, with pronounced peaks at the $\bar{N}$ points in each symmetry azimuth. This nesting favors a $c(2 \times 2)$ surface periodicity, which corresponds to the commensurate bulk SDW phase when truncated at the (110) surface.

Movie 2. Available from stacks.iop.org/NJP/10/023003/mmedia. Movies of the Fermi map of Cr(110) as a function of hydrogen dose, as in figures 5 and 6. Again, on the clean surface, state $A$ is weakly visible near the $\bar{S}$ point, but this disappears quickly upon adsorption. The $S_1 + S_2$ pair is nearly degenerate with the intense bulk band edges on the clean surface, but moves into the projected band gap upon hydrogen adsorption. Backfolding of the bulk band is not visible since these images were collected at the Fermi level.

its evolution with hydrogen adsorption was shown to be associated with the thin film SDW phase diagram [15]. The contour associated with $S_1 + S_2$ is essentially degenerate with the bulk electron rhomb $B$ on the clean surface, but emerges into the projected band gap upon hydrogen adsorption and is seen as closed elliptical segments surrounding the $\bar{S}$ points. Between these two extremes, the contour has undergone a topological transition, similar to observations on Mo(110)+H and W(110)+H [16, 18, 19]. Like the $S_1 + S_2$ band that forms them, this contour has low photoemission intensity and fairly large $k$-space width. Movie 2 offers a series of Fermi maps as a function of hydrogen exposure, which helps to clarify the evolution of the $S_1 + S_2$ Fermi contour. $S_1 + S_2$ can be more easily seen in the difference map shown in figure 6(c), where we have scaled the clean and hydrogen-covered maps in figures 6(a) and (b) to have the same intensity in the electron rhomb, and then taken the difference. This ‘Fermi-surface crud test’ renders the elliptical $S_1 + S_2$ contours readily visible.

The Fermi maps in figures 5 and 6 were collected at $E_F$, so no SDW-backfolded features are visible. Figure 1 provides the corresponding band maps along the $\bar{\Gamma} \rightarrow \bar{S}$ line and indicate the change in SDW commensurability. On the clean surface, two backfolded bands are clearly visible in figure 1(a), corresponding to an incommensurate SDW state [14]. $S_1 + S_2$ has not yet split appreciably from the bulk band in figures 1(a), 5(a) and 6(a). After hydrogen adsorption, a single backfolded band is visible in figure 1(b), corresponding to the commensurate structure [14], and the $S_1 + S_2$ surface band has moved well into the band gap in figures 1(b), 3(d), 5(b), 6(b), and, most visibly, the two movies and figure 6(c). We showed previously that the system evolves continuously between these limits, with the movement of the surface band into the gap and the reduction to zero of the SDW incommensurability occurring.
in concert over the same hydrogen coverage range \cite{15}. In figure 1(b), the surface band disperses smoothly through the backfolded band with no apparent perturbation in the crossing region.

3.4. Nesting and SDW commensurability

These data certainly indicate some connection between the $S_1 + S_2$ surface band and SDW periodicity, but is that connection driven by Fermi surface nesting? Nesting of the corresponding $S_1 + S_2$ bands on W(110)+H and Mo(110)+H produce anomalies in the surface phonon dispersion relations \cite{32}–\cite{35}. Analogous Fermi surface nesting vectors on Cr(110), indicated by $Q_A$ and $Q_B$ in figure 6(c), might favor a particular spin or lattice periodicity, and this could influence the unusual competition between volume and surface energetics that drive the SDW phase behavior in Cr(110) films \cite{14, 15}. However, these nesting vectors are not related to the C- or IC-phase periodicity and therefore do not play a direct role in the SDW energetics. Phonon dispersions have not been measured on Cr(110), though the results in figure 6 suggest that such measurements would be very interesting.

While the analogy with Mo(110) and W(110) nesting and phonon anomalies is apparently not relevant to the SDW phase behavior on Cr(110), i.e. there exists another nesting. The symmetry elements of the $\overline{S}$ point include only the identity and 2-fold rotation; there are no mirror planes, since the $\overline{\Gamma} \rightarrow \overline{S} \rightarrow \overline{\Gamma}$ line is not perpendicular to the $\overline{H} \rightarrow \overline{S} \rightarrow \overline{H}$ line (as would be the case in a hexagonal SBZ, for example). However, the $S_1 + S_2$ Fermi contour in figure 6(c) is a slightly distorted circle, and very nearly possesses ‘accidental’ mirror symmetries about the two orthogonal lines connecting $\overline{S}$ points across the $\Delta$ and $\overline{\Sigma}$ lines in the SBZ. To the extent that this accidental mirror symmetry is perfect, the elliptical contours would be perfectly nested to their images across the $\Delta$ and $\overline{\Sigma}$ lines in the SBZ. To the extent that this accidental mirror symmetry is perfect, the elliptical contours would be perfectly nested to their images across the $\Delta$ and $\overline{\Sigma}$ lines in the SBZ, as indicated by the vectors $Q_C$ and $Q_D$ in figure 6(c). A given contour is mapped on to its neighbor upon translation by $Q_C$ or $Q_D$, in a kind of ‘giant’ nesting in 2D. We emphasize that this nesting is accidental; it is not required by the symmetry of the $(1 \times 1)$ SBZ, as would be the case, for example, in a hexagonal SBZ. Even more peculiar is that this nesting will not lead to backfolded bands since the nested bands very nearly overlay one another—unlike the 3D nesting between bulk electron and hole octahedral. The nesting vector $Q_E$ in figure 6(c), being the same as a reciprocal lattice vector, gives an example of ‘nesting’ that simply corresponds to reciprocal lattice symmetry of the $(1 \times 1)$ SBZ and would exist even if the Fermi contour did not exhibit accidental mirror symmetry. Corresponding elliptical orbits on Mo(110)+H and W(110) +H \cite{18, 19} are more elongated along the $\overline{\Gamma} \rightarrow \overline{S} \rightarrow \overline{\Gamma}$ line and do not possess this accidental symmetry.

At lower photon energy, the SR’ near the zone center is also affected by hydrogen adsorption and appears to be coupled to similar structures at the $\overline{H}$ and $\overline{N}$ points that might also favor a $c(2 \times 2)/C$-phase. To determine whether these structures result from simple SDW backfolding or indeed contribute to stabilizing the C-phase will require a separate study.

These various nesting vectors can be illustrated more precisely by autocorrelation functions of the Fermi maps, as has become popular in ARPES and STM studies of cuprate superconductors \cite{36}–\cite{38}. The difference map in figure 6(c) effectively removes much of the bulk intensity so that such an autocorrelation function will emphasize nesting of surface Fermi contours. Moreover, as shown in figure 5, these rectangular sections were selected to comprise exactly two $(1 \times 1)$ SBZs while also having boundaries that are aligned with the axes of the image, unlike those of the distorted hexagonal $(1 \times 1)$ SBZ. This means that the autocorrelation

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function of these images can be easily formed using standard image processing routines with periodic boundary conditions, all the while maintaining reciprocal lattice symmetry. The resulting autocorrelation image is shown in figure 6(d), along with SBZ boundaries for the $(1 \times 1)$ and $c(2 \times 2)/C$-phase and the various nesting vectors in figure 6(c). The strong nesting across the $\Delta$ and $\Sigma$ lines in the SBZ ($Q_C$ and $Q_D$) is manifested by the intense features located at the $N$ points of the $(1 \times 1)$ SBZs. These are equivalent to $\Gamma$ points of the $c(2 \times 2)/C$-phase SBZ, which in turn means that these nesting vectors favor the $c(2 \times 2)/C$-phase structure. Features associated with $Q_A$ and $Q_B$ are also observed in figure 6(d), though these are weaker since they couple only a small fraction of the elliptical contours. In contrast, the accidental nesting vectors $Q_C$ and $Q_D$ necessarily couple the entire contours and therefore lead to much stronger peaks in the autocorrelation map.

We propose that nesting of the $S_1 + S_2$ Fermi contour provides a surface-related driving force for the $c(2 \times 2)/C$-phase SDW, and that this 2D nesting can overcome the 3D nesting that favors the IC/SDW. However, we have previously shown that the C-phase SDW is also observed on clean Cr(110) films at low thickness [14]. Adding hydrogen to the surface simply shifts the C-or IC-phase line to lower film thickness [15]. Apparently, hydrogen is not a necessary ingredient to produce a C-phase ground state in Cr(110) films. Can 2D Fermi surface nesting explain the existence of the C-phase SDW in thin and clean Cr(110) films? The $S_1 + S_2$ Fermi contour on clean Cr(110) is essentially a band edge SR, degenerate with the edge of the bulk electron octahedron, still visible in movie 2. Although this resonance does not support the complete nesting degeneracy of the $S_1 + S_2$ Fermi contour, there is still significant nesting. The primary difference is that the surface state lies deep in the projected band gap on the H-covered surface and is therefore much more surface localized than on the clean surface. The impact on the 2D generalized susceptibility will be correspondingly larger upon hydrogen adsorption. This argument is identical to the reasoning that has been given to explain why the aforementioned surface phonon anomalies on W(110) and Mo(110) are observed only after hydrogen adsorption. It seems quite possible that the interplay between 2D and 3D Fermi surface nesting, and how this is modified by hydrogen adsorption, can qualitatively explain most of the unusual SDW phase behaviors in this system.

4. Conclusions

Our results confirm and further elucidate the previously-reported connection between the surface hydrogen coverage and the SDW phase adopted by Cr(110) films. They are consistent with the notion that the complex SDW phase diagram results from an interplay between interface and volume effects, specifically, between the surface and bulk Fermi surface nesting. The outer surface of the film plays a significant and possibly dominant role in the energetics that favors the C-phase, and our results therefore also explain the role of film thickness since the surface nesting should become a relatively more important interaction in thinner films. We cannot rule out other contributing interfacial mechanisms favoring the C-phase, e.g. the role of states localized at the Cr–W interface and surface spin anisotropy. Our primary conclusion in this regard is that Fermi contour nesting plays an important role through an accidental symmetry upon hydrogen adsorption that favors the commensurate phase.
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