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Optical reflectance anisotropy of the growth of Fe monolayers on W(110)

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Abstract

We report measurements of the optical anisotropy of Fe layers grown on the W(110) surface using reflection anisotropy spectroscopy (RAS). As the first monolayer of Fe is deposited onto W(110), the resonance-like RAS profile of the clean surface is reduced in intensity. We find evidence for the surface state on W(110) surviving as an interface state following Fe deposition. We observe an anisotropic optical response from Fe layers grown on top of the first two monolayers, where a broad peak at 3 eV dominates the RAS response. The results are simulated in terms of a layered Fresnel reflection model incorporating either a strained Fe overlayer or an Fe overlayer whose dielectric properties are approximated by a simple Lorentzian oscillator. Both approaches are found to produce simulated RA spectra that are in good agreement with experiment. The former approach provides evidence that RAS can detect anisotropy in strained overlayers and that 7 ML films have bulk-like electronic and optical properties.

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

1. Introduction

The W(110) surface is widely used as a substrate for the epitaxial growth of thin metal films. The high surface energy of W favours an initial layer-by-layer growth mode and there is an absence of alloying and inter-diffusion at the film/substrate interface. The growth of Fe on W(110) has been the focus of a number of studies concerned with determining the atomic structure and the magnetic properties of the Fe films. It has been found that at room temperature, the first monolayer (ML) of Fe grows with the same lattice parameter as the W substrate [1–3], termed ‘pseudomorphic growth’. Due to the smaller atomic size and lattice constant of Fe, as compared to W, the pseudomorphic ML is geometrically strained with respect to the W crystal surface. With further Fe deposition, islands nucleate and grow to form the second monolayer and misfit dislocations are observed in these islands as a result of a lowering of the strain energy [4, 5]. Strained monolayers and thin films are interesting from both a fundamental and a technological viewpoint as coatings with potentially unique physical and chemical properties.

A detailed insight into the growth of Fe on W(110) has come from the results of scanning tunneling microscopy (STM) studies [4–8]. The STM study by Bethge *et al* [4] found that for Fe coverage (θ) < 0.6 ML, discrete islands of 1 ML height and of no preferred growth direction partially cover the surface. Island coalescence, initially inhibited due to the lattice misfit, starts to occur above 0.6 ML and, apart from some open channels between neighbouring islands, the first ML is complete before the second layer is initiated. With further coverage, monolayer-height islands are observed on top of the first ML. These islands are anisotropic in shape, elongated in the [001] direction, with aspect ratios between 1:2 and 1:4 [4]. The third layer proceeds with a larger number of islands nucleating relative to the previous layers. The anisotropic growth appears to persist at higher coverage, with 10–30 ML thick films exhibiting an anisotropic morphology along the [001] direction [4].

In the work reported here, we investigate the optical properties of the Fe/W(110) interface using reflection anisotropy spectroscopy (RAS). RAS [9–11] is a versatile probe belonging to the epioptics family of techniques designed

for the study of surfaces and interfaces [12]. When RAS is applied to a cubic substrate, surface sensitivity is achieved by the cancellation of contributions from the bulk crystal, and the reflectance anisotropy (RA) spectrum arises from anisotropy originating at the surface. The RAS response of clean W(110) has been investigated previously in a combined experimental and theoretical study [13]. Transitions between electronic states were identified that give rise to the RAS response of the clean surface. Carroll *et al* [14] have reported RAS results from 1 ML height Fe stripes of widths up to 9 nm, formed on a vicinal W(110) substrate. The damping of the RAS signal from the vicinal W(110) surface following sub-ML to 1 ML Fe coverage was noted and the Fe nanostripes did not produce a strong RAS signature [14]. Here, we report RAS results from higher Fe coverages on the singular W(110) surface and simulate the results in terms of a layered Fresnel reflection model. We find evidence for the surface state on W(110) surviving as an interface state following Fe deposition, and we observe an anisotropic optical response from Fe layers grown on top of the first two monolayers.

2. Experimental details

The experiments were performed in an ultra-high vacuum (UHV) environment. The W(110) crystal was cleaned by repeated cycles of heating to 1600 K in an O₂ atmosphere of pressure 1×10^{-6} mbar, followed by a return to UHV and flashing several times to >2300 K. Surface order was confirmed by low-energy electron diffraction (LEED) and cleanliness monitored using x-ray photoelectron spectroscopy (XPS). With the W(110) substrate at room temperature, Fe was deposited using an Omicron EFM evaporator loaded with an Fe rod (purity 99.998%). We use the definition of Fe coverage $\theta = 1$ ML as the number of atoms in the pseudomorphic Fe layer, which is the same as the number of atoms in a W(110) surface layer, 1.42×10^{19} atoms m⁻². Coverage was determined from a comparison of LEED data with the characteristic LEED patterns and spot intensities observed at specific coverages by Gradmann and Waller [1] and comparison of Auger L₃MM peaks with those of Nahm and Gomer [15].

RAS [9–11] probes as a function of energy the linear optical response of a surface by measuring Δr , the difference in reflectance of normal incidence linear-polarized light for orthogonal linear polarizations, normalized to the mean reflectance r . For W(110) the difference between the two orthogonal surface directions [001] and $[1\bar{1}0]$ is measured. The complex reflectance anisotropy is given by

$$\frac{\Delta r}{r} = \frac{2(r_x - r_y)}{r_x + r_y} = \frac{2(r_{[001]} - r_{[1\bar{1}0]})}{r_{[001]} + r_{[1\bar{1}0]}} \quad (1)$$

where r_x represents the complex Fresnel reflection amplitude for x polarization. The real part of the complex RA is measured using an RA spectrometer based upon the Aspnes design [9] and utilizing a Xe photon source and photoelastic modulator [16]. Optical access into the UHV chamber for the external RAS instrument is provided by a low-strain window. Experimental artefacts are removed from the spectra using

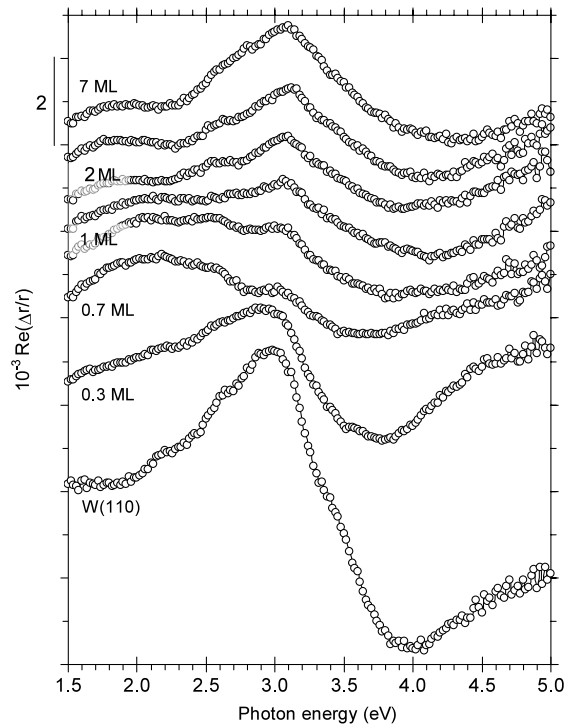


Figure 1. RA spectra of the clean W(110) surface and increasing Fe deposition onto W(110) up to 7 ML. Successive spectra are offset on the vertical axis for clarity.

a correction function obtained by measuring spectra with the sample in two orthogonal positions. RA spectra were recorded with the sample at room temperature and *in situ*, i.e. sequentially during the continuous deposition of Fe. The time taken to record a spectrum from 1.5 to 5.0 eV was ~ 3 min and the coverage quoted refers to the amount deposited at the mid-point (3.25 eV) of the spectrum. The difference in Fe coverage between the start and end of a spectrum is ~ 0.3 ML.

3. Results and discussion

3.1. Clean W(110)

The RA spectrum of the clean W(110) surface at room temperature is shown in figure 1. The RA spectrum is dominated by a strong resonance-like feature centred at 3.4 eV, with positive and negative peaks at approximately 3 and 4 eV, respectively. The RA response of W(110) has been measured previously and interpreted with the aid of surface electronic structure calculations performed within a joint density of states (JDOS) approach [13]. The RAS lineshape is thought to arise as a result of transitions between occupied surface states with p character and unoccupied d states. In the topmost surface layer there exist surface states with p_x and p_y character, and it is the difference in the contributions from $p_x \rightarrow d$ and $p_y \rightarrow d$ transitions that gives rise to the resonance-like RAS lineshape [13]. A subsequent calculation of the band structure and surface dielectric function of W(110) by Ammi *et al* [17] also found that the peaks at 3 and 4 eV have their origins in $p \rightarrow d$ transitions. Martin *et al* [13] found that the contributions to the RAS profile derive from

transitions involving the topmost surface layer, whereas Ammi *et al* [17] found that the 4 eV peak is associated with transitions originating from sub-surface layers. At higher energy, there are indications of a smaller contribution to the RAS at ~ 5 eV [13, 17]; however, this region is subject to greater uncertainty in experimental measurement due to relatively low reflectivity and greater sensitivity to experimental artefacts such as window strain.

3.2. Fe/W(110)

The evolution of the RAS of W(110) upon the deposition of Fe and the subsequent growth of islands and monolayers is shown in figure 1. Spectra were recorded *in situ* during continuous Fe deposition. For deposition up to the first ML (figure 1), the resonance of the W(110) substrate is reduced in intensity in a similar way to that found for the exposure of the clean surface to oxygen [13] and similar to RAS results of the deposition of Ag on W(110) [18]. One notable difference for Fe/W is that at $\theta \sim 0.7$ ML (figure 1) an increase in RAS intensity over the range 1.5–2.6 eV is observed. In this coverage regime, 1 ML height Fe islands partially cover the W substrate [4]. The morphology of the Fe islands is consistent with a reduction in the resonance-like RAS profile of clean W(110), and the increase in RAS observed between 1.5 and 2.6 eV must be associated with the Fe island/W interface. Scanning tunneling spectroscopy (STS) results [7] have found that the pseudomorphic Fe islands of 1 and 2 ML height exhibit an empty state peak in the STS data, just above E_F . This result is in agreement with band structure calculations that have found an increased density of states just above E_F for a pseudomorphic Fe ML on W(110) [19]. The STS empty state was attributed as being a stress-induced feature [7]. We speculate that the RAS intensity below 2.6 eV for $\theta \sim 0.7$ ML (figure 1) could be related to transitions involving this empty state.

As coverage increases, the islands develop into a complete monolayer and the RAS intensity below 2.6 eV is reduced. The spectrum of figure 1 for ~ 1 ML is very similar in profile to the W(110) RAS spectrum observed following exposure to 0.5 ML oxygen [13]. The small peak observed at 2.6 eV for 1 ML Fe (figure 1) is also present in the O/W(110) data [13]. The origin of this small peak could then either be due to a residual W signal following quenching, or be instrumental in origin as this energy coincides with a strong emission line from the Xe lamp. It can be concluded that for $\theta \sim 1$ ML Fe, there are no new states specific to Fe that are present in the RAS results. A similar conclusion was reached for a vicinal W(110) substrate partially covered in aligned 1 ML height Fe nanostripes [14].

For $2 \text{ ML} \leq \theta \leq 7 \text{ ML}$, the growth of a broad RAS peak at 3.1 eV is observed (figure 1). This peak dominates the spectrum in this coverage regime. While the peak is similar in energy to the 3 eV RAS peak characteristic of clean W(110), the broad 3.1 eV Fe peak has a more symmetrical shape compared to the resonance-like feature of the clean surface. The fact that the 3.1 eV RAS peak is positive means that from equation (1) the reflection coefficient $r_{[001]} > r_{[1\bar{1}0]}$. The Fe layers proceed via the growth of anisotropic islands elongated

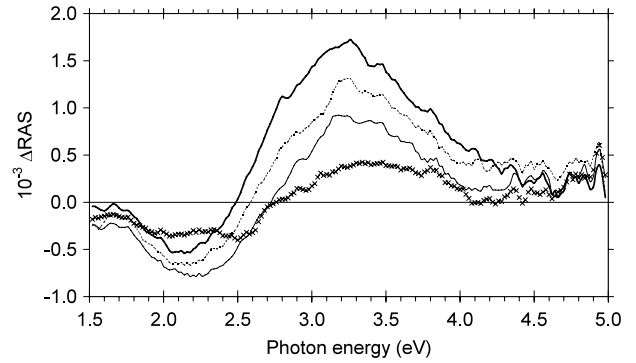


Figure 2. Difference RA spectra obtained by subtracting the spectrum of 1 ML Fe/W(110) from the spectra for: 1.5 ML (\times), 5 ML (thin line), 6 ML (dashed line) and 7 ML (thick line).

in the [001] direction [4] and so the RAS results show that the reflection of polarized light along the long axis of the islands, $r_{[001]}$, is greater than that orthogonal to them.

The changes in the optical anisotropy with increasing Fe deposition beyond the 1 ML Fe/W interface can be plotted as difference spectra. Figure 2 shows four difference spectra (Δ RAS) obtained by subtracting the 1 ML Fe/W RAS spectrum (figure 1) from RAS spectra obtained from higher coverages up to a maximum of 7 ML. The 1 ML spectrum is chosen as a common reference spectrum for subtraction since this represents the point at which the optical anisotropy of the W(110) surface is partially quenched, and the subtraction of this spectrum from higher coverage spectra shows the changes in optical anisotropy due to the growth of Fe on top of the 1 ML Fe/W interface. The difference spectra have been smoothed by means of a first neighbour average to reduce noise. Although the spectra were recorded *in situ* during continuous Fe deposition, and so are not of a static coverage, the subtraction method still serves to highlight the growth of the ~ 3 eV peak and the extent to which it broadens with increasing Fe deposition. In addition to the main change in RA at 3 eV, changes in optical anisotropy at 2.2 eV are revealed whereas no significant change is observed in the RA response above 4.5 eV with increasing Fe deposition (figure 2).

3.3. Simulating the RAS data

While the RAS response of the W(110) surface has been calculated [13], there are currently no first principles calculations of the optical response of the Fe/W(110) interface. To gain further insight into our RAS results, we compare the experimental data to simulations that are based upon a consideration of the reflection of light at near-normal incidence from a simplified model system. We then evaluate the simulated data and discuss the assumptions and limitations involved in this approach.

To simulate the RAS of clean W(110) we use the three-phase model developed from Fresnel theory by McIntyre and Aspnes [20] that is often used to simulate RA spectra of clean single-crystal surfaces [11]. For the Fe/W(110) results we use an extension of the three-phase model that includes an overlayer phase [21], which represents the Fe layers on

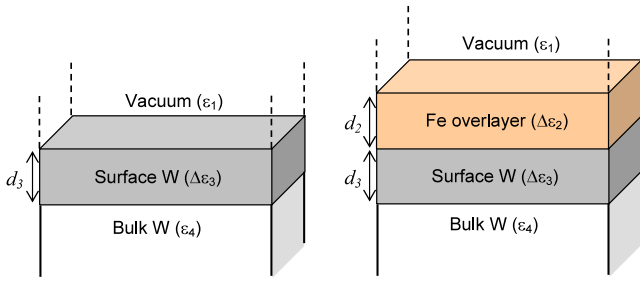


Figure 3. The three- and four-phase model systems used in the simulations.

top of the W substrate. The three- and four-phase models simplify the system to discrete layered phases, each with its own complex dielectric function ϵ . A vacuum phase ($\epsilon_1 = 1$) and an isotropic W bulk (ϵ_4) sandwich the surface W phase, and any overlayer phase, as shown in figure 3. The interfacial phases are biaxially anisotropic with $\Delta\epsilon = \epsilon^x - \epsilon^y$, where x and y are orthogonal in-plane directions aligned along x and y polarizations. The interfacial phases have effective thickness $d \ll \lambda$, the wavelength of light, in order to satisfy the thin film limit [20]. The reflection coefficients and $\Delta r/r$ for normal incidence reflection from the model system are then determined. In the thin film limit the contributions of the anisotropic phases 2 and 3 are additive and the RAS response of the four-phase system is given by [21]

$$\frac{\Delta r}{r} = \frac{4\pi i}{\lambda(\epsilon_4 - 1)} [d_2 \Delta\epsilon_2 + d_3 \Delta\epsilon_3]. \quad (2)$$

Equation (2) reduces to the well-known expression for a three-phase system with the choice of $d_2 \Delta\epsilon_2 = 0$. The real part of equation (2), using $\epsilon = \epsilon' - i\epsilon''$, is used to simulate the experimental RAS results.

We first simulate the RAS of the clean W(110) surface using a three-phase model (figure 3) where the W surface

phase, $\Delta\epsilon_3$, is based upon a Lorentzian oscillator of energy ω_t , strength S and width γ occurring for one or both of the RAS polarizations. For x polarization the oscillator is described by [22]

$$\epsilon_3^x = 1 + \frac{S/\pi}{\omega_t - \omega + i\gamma/2} \quad (3)$$

and similarly for an oscillator occurring for y polarization. The dielectric properties of W (ϵ_4) are obtained from tabulated data [23] and the usual approach of setting the surface phase effective thickness $d_3 = 1$ nm is followed. A Lorentzian oscillator (equation (3)) for x and y polarization is used to simulate the $p_x \rightarrow d$ and $p_y \rightarrow d$ transitions, respectively, of the clean surface, which give rise to the resonance-like lineshape [13]. Lorentzian oscillators are usually used in RAS simulations to represent transitions involving electron excitation from occupied surface states [11]. The RA spectrum resulting from this simulation is shown in figure 4(a) and the parameters used in the simulation are listed in table 1. The simulation produces a spectrum that is in good agreement with experiment. The energies of the transitions used in the simulation (2.7 and 3.4 eV) are close to the energies of the peaks found in the difference between the calculated surface JDOS for the two polarizations, as a result of transitions from p_x (2.4 eV) and p_y (3 eV) surface states to the d band [13]. The calculated energies in the JDOS method are expected to be slightly lower in energy than experiment due to self-energy effects [13].

For the 1 ML Fe/W(110) surface, only a quenching of the W(110) signal was observed experimentally (figure 1), resulting in a spectrum that is very similar to that observed following exposure of W(110) to 0.5 ML of oxygen [13]. Thus a simulation using the three-phase model, without an Fe overlayer phase, should be sufficient. We find that this is indeed the case: the RAS of 1 ML Fe/W(110) is well simulated using the three-phase model with $d_3 = 1$ nm and one transition for y polarization. The simulation is shown in

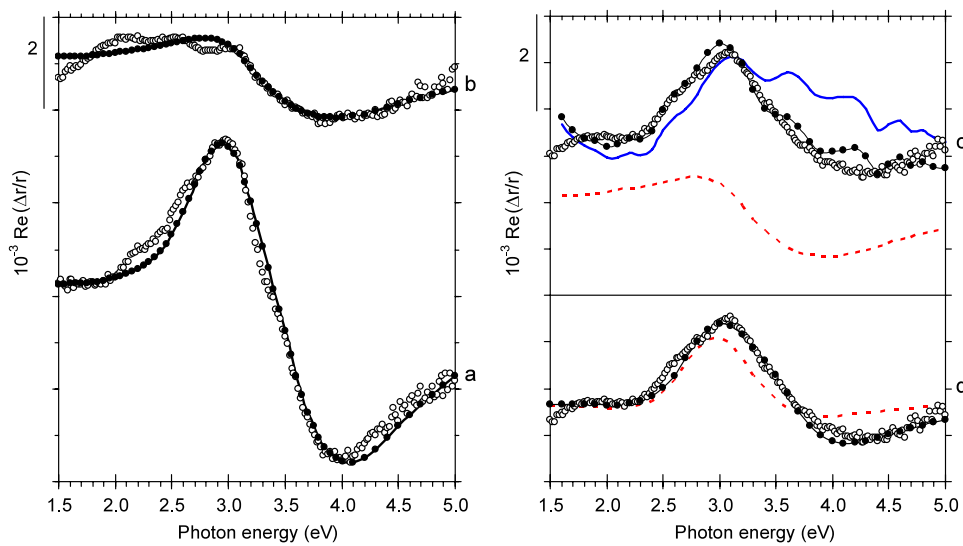


Figure 4. Comparison of simulated (●) and experimental (○) RA spectra of (a) the clean W(110) surface, (b) 1 ML Fe/W(110), (c) 7 ML Fe/W(110), where the solid (blue) and dashed (red) lines show the contributions from the first and second terms of equation (2), respectively. (d) Shows an alternative simulation of 7 ML Fe/W(110)—see the text. The simulated spectra are offset on the vertical axis to overlap the experimental spectra.

Table 1. Values used to simulate the RAS of W(110), Fe/W(110) and Ag/W(110). Lorentzian and derivative contributions refer to equations (3) and (4), respectively.

Structure	Polarization	Lorentzian			Derivative	
		ω_l (eV)	S	γ (eV)	ΔE (eV)	$\Delta\Gamma$ (eV)
W(110) (figure 4 (a))	y	3.4	5.0	1.0	—	—
	x	2.7	2.0	1.0	—	—
Fe/W(110)						
1 ML (figure 4(b))	y	3.3	1.2	0.9	—	—
7 ML (figure 4(c))	y	3.3	1.2	0.9	-0.2	-0.2
7 ML (figure 4(d))	y	3.5	1.7	1.0	—	—
	x	2.7	1.4	1.0	—	—
Ag/W(110)						
5 ML (figure 5)	y	3.5	1.7	0.9	0.03	-0.01

figure 4(b) and the oscillator values are listed in table 1. The values used relative to those of the clean surface simulation are consistent with the partial quenching of the W(110) RAS signal. Calculations for W(110) have found that the main contribution to $\Delta\epsilon_3$ comes from the $p_y \rightarrow d$ transitions [13], and this contribution persists in our simulation for 1 ML Fe/W, reduced in strength from that of the clean surface. The relatively smaller contribution from the p_x states for the clean surface now becomes negligible. Our 1 ML Fe/W RAS results show a reduced and slightly red-shifted W signal as it appears to change from a surface to an interface state (table 1). We note that the *ex situ* RAS results of Fleischer *et al* [24] for Au-capped Fe on vicinal W(110) have been interpreted in terms of a dominant W surface signal being reduced in intensity as the clean W surface develops into an Fe/W interface. These authors concluded that the surface state present on the vicinal W surface changes into an interface state upon the creation of both Au/W and Fe/W interfaces. Our results are in agreement with this interpretation.

To simulate higher Fe coverage, we begin by using the four-phase model (figure 3) with the same parameters as the 1 ML Fe/W interface simulation used for the $d_3\Delta\epsilon_3$ term in equation (2). Since the additional Fe forms islands, then layers, on top of the interface and no alloying or inter-diffusion occurs we assume that the surface (now interface) state $p_y \rightarrow d$ transitions are unaffected and no changes to the oscillator parameters of the 1 ML Fe/W interface signal are expected. Before we simulate an anisotropic overlayer, we must first consider the case of an Fe overlayer which is isotropic. The addition of an isotropic overlayer could affect the RAS signal due to optical absorption in this phase. As equation (2) only describes surface and overlayer phases that are anisotropic, a full calculation of the normal incidence reflectance for each polarization, R_x and R_y , from the four-phase structure is necessary, from a consideration of the reflection from each interface [25]. The real part of the RAS is then obtained from $2\Delta r/r \approx \Delta R/R$. For the Fe overlayer ϵ_2 we use the tabulated dielectric data for Fe [26]. We find that when $d_2 = d_3 = 1$ nm the isotropic Fe layer has a negligible effect on the simulated spectrum. As the Fe overlayer is increased in thickness above 1 nm, the simulated signal is reduced in RAS intensity with a small shift to lower energy of the quenched resonance-like profile of figure 4(b). The new Fe peak observed experimentally at 3.1 eV (figure 1) is not

reproduced by the presence of an isotropic Fe overlayer. Thus we can conclude that the 3.1 eV RAS peak (figure 1) is due to optical anisotropy originating in the Fe overlayer. From figure 2 it appears that a small amount of optical anisotropy at ~ 3 eV is present at $\theta \sim 1.5$ ML which becomes dominant for $\theta > 2$ ML (figure 1).

We now simulate an anisotropic Fe overlayer using equation (2), employing two different methods of approximating $\Delta\epsilon_2$. In the first method, the overlayer represents a uniaxially strained Fe interfacial region whose dielectric properties are related to those of bulk Fe. This approach is taken since it is known that the first few monolayers of Fe on W(110) exhibit a significant and anisotropic strain [27]. RAS results of several semiconductor [28, 29] and noble metal [30–35] surfaces have been successfully simulated by basing the dielectric properties of the surface on the energy derivative of the dielectric function of the bulk crystal, $d\epsilon/dE$. These surfaces are strained, which leads to small anisotropic shifts in energy and linewidth of bulk transitions at critical points that are thought to dominate the RAS response at those energies. The strained Fe layers at the interface with W are assumed to have a similarly perturbed electronic structure. We assume differences in energies ΔE and linewidths $\Delta\Gamma$ of interband transitions between x and y polarizations near a critical point. For this case we can write [36]

$$\Delta\epsilon_2 = (\Delta E - i\Delta\Gamma) \frac{d\epsilon}{dE}. \quad (4)$$

Photoemission results have found evidence that for 3 ML Fe/W(110) [37] and for 5 ML Ni/W(110) [38] the thin overlayers exhibit electronic structure that is very similar to that of the corresponding bulk material. A bulk-like electronic structure of Ag, when grown on W(110), is implied from the RAS results for 5 ML Ag films [18]. These observations help to support our use of equation (4), where ϵ_2 is related to the dielectric properties of bulk Fe. A weakness in our use of equation (4) is that we do not know from which critical point the transitions originate. Inspection of the band structure of Fe [39–41] shows that there could potentially be contributions from many interband transitions, not necessarily all at critical points. Nevertheless, we proceed with the assumption that equation (4) is valid and we use the four-phase model to simulate the highest coverage studied, the 7 ML Fe/W(110) results. The thickness of the surface and overlayer phase in the model are essentially scaling factors and fixing $d_2 = d_3 = 1$

nm gives an equal weighting to the two phases via equation (2). The parameters ω_t , S and γ of the surface are fixed to those of the 1 ML Fe/W interface, leaving only ΔE and $\Delta\Gamma$ of the Fe overlayer as variables (table 1). With this approach it is possible to produce a simulated spectrum that reproduces the broad anisotropic peak at 3 eV, in good agreement with experiment as shown in figure 4(c). The good agreement of the simulation with experiment suggests the formation of a bulk-like electronic structure for $\theta = 7$ ML. As $\Delta E = \Delta\Gamma$ in the simulation (table 1) the energy difference and broadening effects have equal weight. We note that the 1 ML Fe/W RAS results do not require such an Fe energy-derivative overlayer, although the monolayer is strained. It is assumed that 1 ML Fe is too thin to contribute a distinct Fe signature to the total optical anisotropic response.

The second method of simulating the anisotropic Fe overlayer bases $\Delta\epsilon_2$ on a Lorentzian oscillator, as used to simulate the RAS of the clean W surface. Optical conductivity ($\propto \epsilon''E$) data of Fe derived from experimental [26] and calculated [41] data show a large and broad (~ 1 eV FWHM) peak at 2.7 eV. The same peak is observed at 2.4 eV at low (~ 4 K) temperature [42]. In general, peaks in ϵ'' , and so optical conductivity, arise from interband transitions at critical points. The calculated density of states (DOS) and band structure of Fe show that there are two large peaks in the DOS either side of E_F , separated by ~ 2.5 eV [41], offering many possible transitions to contribute to this conductivity peak, particularly transitions between the relatively flat bands near the P critical point [39, 41]. We represent such transitions in our simulation with an oscillator of energy 2.7 eV. With the y transition of $\Delta\epsilon_3$ fixed to the values of the 1 ML Fe/W simulation, the overlayer $\Delta\epsilon_2$ requires a transition in x , rather than in y , to match the sign of the experimental RAS profile. The resulting simulation using the values for x listed in table 1 and with the y transition fixed to the values of the 1 ML Fe/W simulation is shown by the dashed line in figure 4(d). The simulation does not produce a good fit to the data above 2.8 eV. The simulation can be improved by allowing the parameters of the y interface transition to vary. A blue-shift and increase in oscillator strength for y , as listed in table 1, produce a good fit, as shown by the filled circles in figure 4(d). Simulating the 7 ML Fe overlayer with a contribution from a modified W y transition indicates that the Fe/W interface state is altered with increasing Fe deposition. This is in contrast to the energy-derivative simulation (figure 4(c)), where this transition remained the same. While the new x transition is of the same energy as the x transition used to simulate the RAS of the clean surface (table 1), it is unlikely that it is the same transition since this transition is lost following 1 ML Fe deposition (table 1). It is more likely that the overlayer x transition used in the 7 ML simulation is a new state related to the Fe overlayer, and related to the 2.7 eV peak in the optical conductivity of Fe [26, 41].

The simple optical models shown in figure 3 are capable of producing simulated RAS data that are in good agreement with experiment; however, there are assumptions and limitations in these models. The four-phase model simplifies the system into discrete phases; there is no account of surface or interface roughness and the assumption is made that the 7 ML Fe

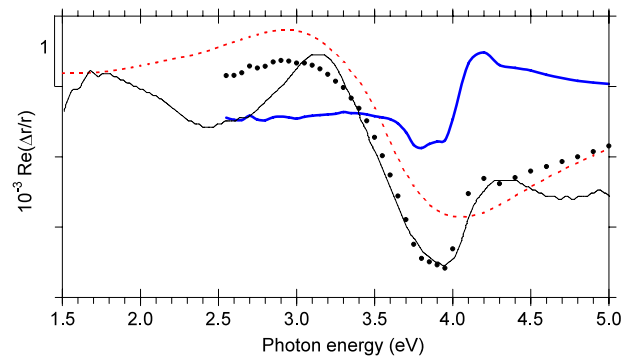


Figure 5. Comparison of simulated (●) and experimental (—) RA spectra of 5 ML Ag/W(110). The experimental data are from [18]. The thick solid (blue) and dashed (red) lines show the contributions from the first and second terms of equation (2), respectively. The simulated spectra are offset on the vertical axis to overlap the experimental spectra.

overlayer can be simulated using the dielectric function data of bulk Fe. The latter has some support from other studies where similar thickness overlayers have been found to have electronic properties similar to those of the bulk material [18, 37, 38], although the optical properties may show differences. No account is made of any strain in the W surface induced by the pseudomorphic Fe overlayer; however, the good fit obtained for 1 ML Fe/W by quenching the W(110) signal suggests that any contribution of this effect to the RAS signal must be small. The RAS response of a strained W surface has been explored by Fleischer *et al* [24]. First principles calculations of the optical anisotropy of the Fe/W(110) interface, that include the effects of the pseudomorphic strained Fe monolayers, are desirable, and we hope that the experimental results presented here will stimulate new theoretical work on the electronic and optical properties of this system.

Finally, we note that the two methods of approximating $\Delta\epsilon_2$ in the higher coverage simulations give equally good results (figures 4(c) and (d)), and it is not obvious which method is to be preferred—both having reasonable physical grounds. In order to evaluate further the use of $\Delta\epsilon_2 \propto d\epsilon/dE$ in the four-phase model we now apply it to other relevant data. Sun *et al* [18] have reported RAS results of the growth of Ag on W(110) up to 5 ML thickness. The majority of the Ag atoms in the first ML are commensurate with the bcc W(110) surface [43, 44] and there is strain in the commensurate Ag layer, similar to the Fe/W(110) system. We use the four-phase model with $\Delta\epsilon_2 \propto d\epsilon/dE$ —that for 7 ML Fe/W(110) produced a simulated RAS in good agreement with experiment (figure 4(c))—to simulate the 5 ML Ag/W(110) results of Sun *et al* [18]. The overlayer in figure 3 now represents a uniaxially strained Ag film and equation (4) is used for the overlayer with the dielectric function data of Ag measured from 2.5 eV onwards by Stahrenberg *et al* [45]. The values used in the simulation are listed in table 1 and the resulting spectrum is shown, with a comparison to the experimental results, in figure 5. Although the simulated peak at 3 eV is much broader around its maximum than the experimental data, the simulated and experimental profiles show good agreement

above 3 eV (figure 5). The oscillator values used for ϵ_3 (the W phase) in the 5 ML Ag/W simulation are similar to those for the 7 ML Fe/W simulation (table 1), suggesting the presence of a similar W interface state in the two systems. With Ag, the use of the energy derivative of ϵ (equation (4)) is perhaps more straightforward than for Fe. It is known that interband transitions at the L critical point dominate the RAS response of the clean Ag(110) surface between 3 and 5 eV, and RAS of Ag(110) has been simulated successfully in terms of the energy-derivative model [34]. The solid line in figure 5 shows the contribution from equation (4) which models the transitions at the L point, and this contribution has a very similar profile to that of the RAS data. We conclude that the model with the approximation $\Delta\epsilon_2 \propto d\epsilon/dE$ is capable of simulating, to good agreement with experiment, strained Ag and Fe overlayers on W(110).

4. Conclusions

We have investigated the optical reflectance anisotropy of Fe/W(110) up to 7 ML Fe thickness. As the first monolayer of Fe is deposited onto W(110), the resonance-like RAS profile of the clean surface is reduced in intensity. We find evidence for the surface state on W(110) surviving as an interface state following Fe deposition, in agreement with previous results using a vicinal W(110) surface [24]. We observe an anisotropic optical response from Fe layers grown on top of the first two monolayers, where a broad peak at 3 eV dominates the RAS response. The results are simulated in terms of a layered Fresnel reflection model incorporating either (i) a strained Fe overlayer with an optical response proportional to the energy derivative of the bulk Fe dielectric function, or (ii) an Fe overlayer whose dielectric properties are approximated by a simple Lorentzian oscillator. Both approaches are found to produce simulated RA spectra that are in good agreement with experiment. The former approach provides evidence that RAS can detect anisotropy in strained overlayers and that 7 ML films have bulk-like electronic and optical properties.

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