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Structure and magnetism in ultra-thin Cr layers on W(110): How well do experiment and theory agree?

B. Santos[1], J. de la Figuera[1,4], J. M. Puerta[2], J. I. Cerda[2], R. Stumpf[3], K. F. McCarty[3]

1 Dpto. de Física de la Materia Condensada and Centro de Microanálisis de Materiales, Universidad Autónoma de Madrid, Madrid 28049, Spain
2 Instituto de Ciencia de Materiales, CSIC, Madrid 28049, Spain
3 Sandia National Laboratories, Livermore, California 94550, USA
4 Instituto de Química-Física "Rocasolano", CSIC, Madrid 28006, Spain

Introduction

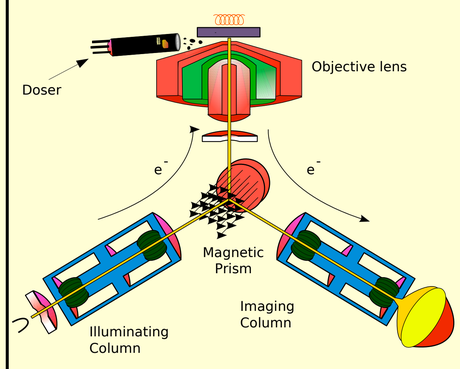
Low energy electron diffraction [1] is one of the basic tools for determining surface structures with high accuracy, with known error limits [2]. Although big unit cells are still a challenge, LEED has provided for a long time the contraction of out-of-plane distances on the non-reconstructed surfaces of transition metals.

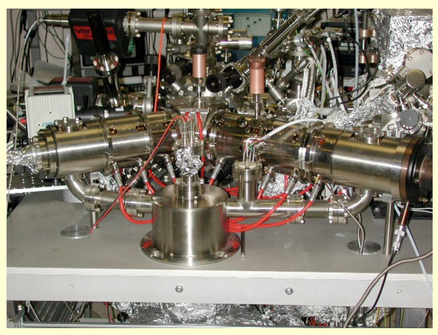
Ab-initio calculations based on Density Functional Theory are also mature. The determination of the equilibrium distance on transition metal surfaces has been performed for quite some time. A systematic disagreement between ab-initio predictions and LEED experimental determinations was reported in transition metals[3]. Since then, recent determinations of transition metal contractions have removed several of the discrepancies[4,5], so far mostly from the experimental side.

Cr is a transition metal that has been the focus of magnetic studies. It is an itinerant antiferromagnet in the bulk, and presents an spin density wave. Its popularity has increased due to its role in Fe/Cr multilayers and giant magnetoresistance devices. Nevertheless, ultra-thin Cr film studies are scarce [6]. We do not know of any multiple scattering LEED determination of ultra-thin Cr/W(110) films.

In this work, we present ab-initio calculations of Cr(110), W(110) and ultra-thin films of Cr/W(110) together with LEED experimental determinations of the same structures. The experimental determinations have been done in flat areas whenever possible.

Experimental details and Data acquisition



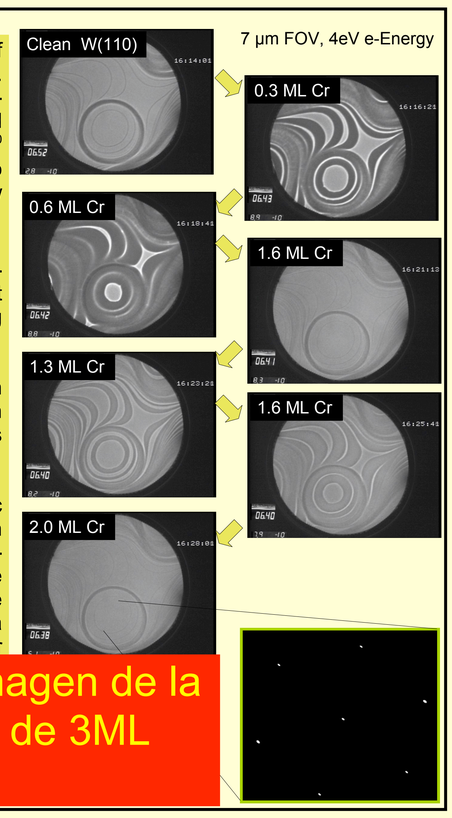


A Low Energy Electron Microscope was used to characterize the growth of Cr on W(110), and to acquire the LEED Intensity-vs-Voltage (IV) curves[5]. The films were grown in the LEEM UHV chamber from a electron-bombardment doser, with typical flux of 1ML/7 min for the ultrathin films, and up to 1ML/min for thick films. The background pressure remained in the 10^{-10} torr during the experiments. The W(110) crystal was cleaned by exposure to O_2 followed by repeated flashes to 2200ºC. Terraces larger that a few microns could be routinely found on the surface.

The growth was followed in real time by LEEM. The first 2ML growth layer-by-layer. The third layer grows until it is nearly complete, at which point it starts to dewet giving rise to tall 3D islands sourrounded by a 2ML wetting layer.

To obtain a Cr(110) surface, thick Cr films (~100ML Cr) were grown on W(110) at room temperature and flash annealed to 600°C while observing in LEEM. The evolution of the surface morphology thick films will be the focus of forthcoming work.

The LEED patterns were in all cases 1x1 indicating in-plane pseudomorphic growth of Cr on W(110) in ultra-thin films. In Cr thick films the 1x1 pattern corresponds to a different, bulk terminated, in-place lattice spacing. The IV-curves are acquired with the LEEM instrument by changing the power of the imaging lenses in order to image the backfocal plane of the objective. The use of LEEM as a LEED diffractometer provides several advantages over a conventional LEEM system, among them the ability to acquire the specular beam in normal incidence. In this work, we used an illum… limits the electron beam diameter on the sample to ~… gather the diffracted electrons from a single terrace … sample was cooled to room temperature before measurin…



Alguna imagen de la superficie de 3ML Cr?

Ab-initio calculations

Bulk W(110) and Cr(110)

The bulk W and Cr lattice parameters was calculated using both LDA and GGA, using the Vienna VASP code[7].

Only GGA finds an antiferromagnetic ground state for Cr (m=1.08 μ_b/atom), LDA result is always non-magnetic. Spin-polarized GGA (sp-GGA) reproduces the experimental results better.(see table)

	Exp	Sp-GGA	N-sp-GGa	LDA
W	3.165	3.169	3.169	3.127
Cr	2.885	2.854	2.837	2.778

Thin films of Cr on W(110)

We use sp-GGA and non-sp-GGA with a slab with 8 W layers and the correct number of Cr layers on top. The calculated relaxations are in % (see table). Note that the non-spin-polarized relaxations are always much larger. By comparison, the relaxation of Cr lattice matched to the in-plane W(110) lattice, is -15.3% (sp-GGA) and -21.8% (non-sp-GGA).

	Top layer	W-Cr	Cr2-Cr3	Cr1-Cr2
sp substrate	-3.8			
n-sp substrate	-3.8			
sp 1ML	+0.04	-8.55		
non-sp 1ML	1.46	-15.4		
sp 2ML	-0.16	-6.82	-18.3	
non-sp 2ML	-0.27	-6.28	-30.1	
sp 3ML	-0.16	-7.37	-15.6	-18.1
non-sp 3ML	-0.07	-10.2	-16.9	-30.6

We also calculated by sp-GGA the magnetic moments per atom for Cr and the top W layer. The top Cr layer has an enhanced magnetic moment of 2.9μ_b. Intermediate Cr layers are close to the strained bulk value (2 μ_b) except for the Cr atoms directly on top of the W(110) surface (1.7μ_b).The top W layer picks up about 0.3μ_b from contact with Cr.

The enhanced moments of strained and surface Cr compared can be explained by increased effective volume per atom.

	Top W	Cr1	Cr2	Cr3	Cr4
1ML	0.31	2.68			
2ML	0.26	1.76	2.84		
3ML	0.25	1.64	1.95	2.89	
4ML	0.24	1.56	1.88	2.05	2.91

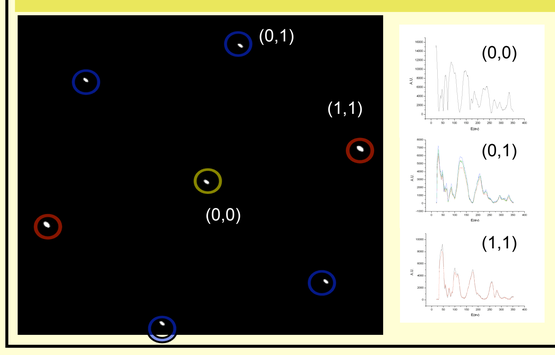
Cr(110) and W(110) surface calculations

All calculations were performed using 8 layers slabs for W(110) and 9 for Cr(110) (the minimum required so the center of the slab shows in-plane antiferromagnetic coupling with magnetic moments like in bulk Cr).

The relaxations found are -3.88%,+0.65% and 0.02% for W(110) and -2.09%,+0.24%,-0.01% for Cr(110), respectively for the topmost layer, second and third layer.

LEED calculations

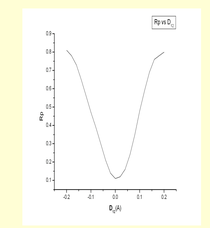
The (00), 4x(01) and 2x(11) beams in the range of 75-350eV, giving a total energy range ΔE of 980eV, were acquired with the LEEM instrument. Equivalent beams were averaged (the Pendry factor Rp between equivalent beams was max. 0.10).
Fully dynamical IV-curve calculations were performed with a modified version of the van Hove-Tong package.

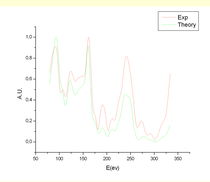


The surface was modelled by stacking the necessary number of Cr layers over 5 layers of W bulk, using the renormalized forward scattering (RFS) approach.

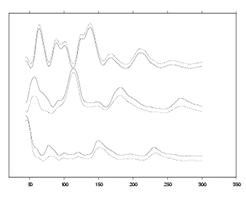
We explored the parameter space comprised of the topmost three interlayer spacings, d_{1-3}, by calculating the IV curves over fine 3D-grids. The interlayer spacings were swept over wide ranges.

All simulations were performed for a temperature of T = 300 K. The experiment–theory agreement was quantified via Pendry's R-factor, while the error bars for each parameter were obtained from its variance: $\Delta Rp = Rp_{min} \times \sqrt{(8E_i/\Delta E)}$, where E_i gives the optical (inner) potential. Correlations between the structural parameters were taken into account for the error-limits estimation. We note that all structural parameters derived in this work represent well-defined minima in their respective R-factor plots. Non-structural parameters such as the muffin-tin radius, the optical potential or the Debye temperatures at the surface planes were varied.





W(110)

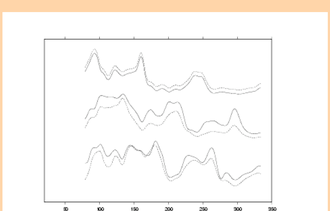


Rp=0.13±0.03
Structural parameter in this case are:
Top W layer is realaxed:
-2.8±1.2%
next to last layer:
0.2±1.0%

We compare our calculations with other authors and our theoretical calculations

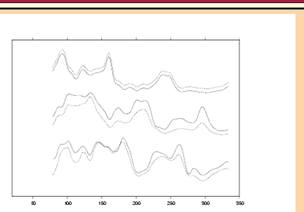
	Δd12	Δd23
Arnold et al.[1]	-3.1±0.6	0.0±0.9
Teeter[2]	-3.0±1.3	0.0±0.9
Disagree[3]	-3.0±1.3	0.2±1.3
Rayos x[4]	-2.7±0.5	0.0±0.3
This job (theory)	-3.8	
This job (experiments)	-2.8±1.0	0.2±1.0

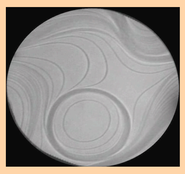
Cr(110)



Rp=0.10±0.02
Structural parameters:
First Cr layer is relaxated
-2.2±1.36 %

1ml Cr/W

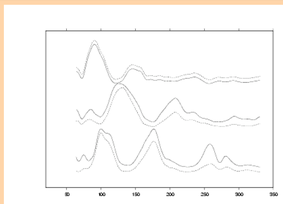




Rp=0.10±0.02
Structural parameter in this case are:
First Cr layer is relaxated
-8.0 ± 0.7%
Top W layer is expanded
+0.31%

	Theory	Experiment
1st ML	-8.5%	-8.0 %

2ml Cr/W

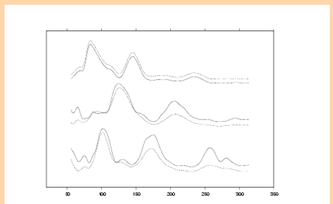


The second Cr layer fits better than others calculus
In this case
Rp=0.08±0.02

Structural parameter in this case are:
First Cr layer is relaxated
-17.0 ± 1.2%
Second Cr layer is relaxated
-7.4 ± 1.29 %
Top W layer is expanded
+0.44%

	Theory	Experiment
1st ML	-18.3%	-17.0%
2nd ML	-6.8%	-7.4 %

75% 3ml Cr/W



The third Cr layer is not completed only (75%ML)
In this case
Rp=0.11±0.03

Structural parameter are:
First Cr layer is relaxated
-16.0±1.3%
Second Cr layer is relaxated
-13.6±1.4 %
Third layer is relaxed
-7.0 ± 4.4%
Top W layer is also relaxed
-1,9%

	Theory	Experiment
1st ML	-18.1%	-16.0%
2nd ML	-15.6%	-13.6%
3rd ML	-7.3%	-7.0%

Conclusions

We have followed the growth of ultra-thin films of Cr on W(110) by LEEM.

We have measured the LEED IV curves, with the LEEM instrument, of selected areas of clean W(110), clean Cr(110) [obtained by growing a thick Cr film on W(110)] and Cr films 1, 2 and 3ML thick on W(110).

We have performed ab-initio calculations on the same surfaces and films, both spin-polarized and non-spin-polarized using VASP.

The ab-initio results and LEED experimental determinations for the clean W(110) surface are in agreement, and they reproduce recent experimental work[4].

For the Cr films, the ab-initio calculations are quite different depending on whether the calculation is spin-polarized with antiferromagnetic ordering or not, with last-layer out-of-plane contractions which are nearly double for the non-spin-polarized calculations.

Our experimental results are in good agreement in all cases with the ab-initio predictions with spin-polarization, implying that Cr ultra-thin films are antiferromagnetically ordered. We are not aware of any direct experimental confirmation of this result.

References

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