# Resonant magnetic x-ray scattering from ultrathin Ho-metal films down to a few atomic layers

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**Abstract**

The magnetic structure of ultrathin Ho metal films grown on W(110) was studied by resonant magnetic x-ray scattering at the $L_3$ and $M_5$ resonances. For Ho films down to 14 monolayer (ML) thickness, a bulk-like helical antiferromagnetic structure is observed. For a 10-ML thick film, an altered magnetic structure and enhanced layer spacing is found.

*Keywords*: Holmium; Magnetic structure; Resonant magnetic x-ray scattering; Ultrathin films; $M_5$; $L_3$

# 1 Introduction

With the advent of highly intense synchrotron-radiation sources, resonant magnetic x-ray scattering (RMXS) has emerged as a powerful tool for the investigation of magnetic structures in solids [1]. By the choice of the photon energy, an element-selective measurement can be performed, which is not achievable by neutron scattering. In the development of RMXS, Ho metal has served as an important reference sample [2–4] because (i) the open $4f$ shell in Ho carries the strongest atomic moment among all elements, (ii) Ho is antiferromagnetic over a wide temperature range with the magnetic scattering signal well separated from that of charge scattering, and (iii) the Ho $L_3$ resonance at 8074 eV corresponds to a wavelength of 1.54 Å well suited for hard x-ray scattering. The helical antiferromagnetic structure of Ho consists of ferromagnetically ordered moments in the basal planes of the *hcp* lattice with

each plane rotated by a certain angle with respect to the neighboring plane, thus forming a helix along the crystallographic $c$ axis [5]. The helix period changes with temperature and amounts to $\approx 10$ monolayers (ML) at 40 K.

The investigation of thin Ho-metal films is interesting from two points of view. First, photoemission studies of lanthanide metals, for the sake of surface cleanness, are performed almost exclusively on epitaxial films grown in situ in ultrahigh vacuum (UHV) on a refractory-metal surface [6–9]. In order to understand the relation between magnetic order and electronic structure, the magnetic properties of thin films, which may be altered relative to those of bulk samples by substrate-induced strain [10], shape anisotropy [11], or finite-size effects [12], have to be characterized. Second, since the indirect exchange responsible for the magnetic structures of the lanthanide metals is of long-range nature [13], changes can be expected for thin films [14] and interesting effects should occur especially if the film thickness is of the order of the magnetic period.

In this contribution, we report on a study of the magnetic structure of ultrathin Ho metal films down to 10 ML thickness.

## 2 Experimental

RMXS experiments at the Ho $L_3$ edge were performed at the ID10A (Troïka 1) beamline of the ESRF and at the W1.1 beamline of HASYLAB. The measurements at the $M_5$ resonance were carried out at the U49/1-SGM beamline of BESSY II. In all cases, the same substrate crystal and the same evaporators were used, however, in two different UHV setups adapted for the two different energy ranges. For hard x-ray diffraction, a small vacuum chamber based on a commercial CF100 double cross and equipped with Be windows for the incoming and outgoing beam was used, which can be mounted on common diffractometers. For the soft x-ray scattering experiments at BESSY II, an UHV chamber directly attached to the beamline was equipped with a Si-diode detector on a rotatable arm that could be moved in vacuum. In both setups, thin Ho metal films were grown in situ on a W(110) substrate kept at room temperature. 99.99% pure metal was evaporated from a Ta crucible heated by electron bombardment. Films below 40-ML thickness were annealed to $\approx 700$ K for 5 minutes, thicker films were grown in two steps with intermediate annealing to 650 K after deposition of 20 ML and annealing to 850 K of the final film. The procedure leads to well-ordered single-crystalline films as checked by x-ray diffraction [15]. X-

ray scattering experiments were performed in specular geometry; for the hard x-ray magnetic scattering a graphite(006) crystal was used for polarization analysis in order to suppress the charge-scattering contribution to the reflected intensity [4]. The soft x-ray data were measured without polarization analysis. The scattering geometry was vertical for the hard x-rays and horizontal for the soft x-rays with horizontal polarization of the incident beam in both cases.

# 3 Results

Diffraction data from a 113-ML Ho film are shown in the left panel of Fig. 1 taken without and with polarization analysis, respectively. The length of the scattering vector, $L$, is given in units of the Ho reciprocal lattice parameter $c^*$. Without polarization analysis, the data are dominated by the charge-scattering signal from the Ho(002) Bragg peak at $L = 2c^*$ (upper curve). Because of the small number of layers in the film, the Bragg peak has a broad Laue shape with side maxima [16]. The increasing background towards higher $L$ values is caused by the crystal-truncation rod scattering from the W substrate [17]. With the polarization analyzer mounted (lower curve), the charge-scattering Bragg peak is still dominating because of an imperfect analysis, but now the two magnetic satellites at $L = (2 \pm 0.2)\, c^*$, corresponding to a 10-layer period of the magnetic structure, are clearly visible. This magnetic period agrees well with bulk Ho and an investigation of the temperature dependence of the magnetic structure reveals a strong similarity between the present film and bulk single crystals. As shown on the right panel of Fig. 1, the length of the magnetic modulation wave vector $\tau$ and the temperature dependence of the $(002 - \tau)$-satellite intensity compare well with the respective bulk data from Ref. 5.

As films of 113-ML thickness obviously still behave bulk like, it is interesting to study even thinner films. Data from the thinnest film investigated at the $L_3$ edge are presented in Fig. 2. The upper panel displays data taken with polarization analysis (solid circles), and with the analyzer turned by 90° in order to measure the charge-scattering signal (open circles), scaled to the same Bragg-peak height. In the lower panel, the difference between the two curves is shown on a linear scale. The magnetic satellite occurs at the same scattering vector as for the thick film, but the intensity from the charge-scattering background is already too strong to allow a more detailed investigation. Easier accessible, even for thin films, is the layer spacing as determined from the Bragg-peak position. In Fig. 3 the double-layer spacing, $c$,

of Ho films of different thicknesses is plotted versus temperature and compared to bulk data. All curves show the same characteristic minimum around the Néel temperature, $T_N$, of 131.4 K caused by magnetoelastic effects. Down to 23-ML film thickness, the layer spacing agrees with the bulk data within a maximum deviation of $\approx 0.1$ % around $T_N$. But for the 11-ML film, the layer spacing is significantly expanded. Since the change occurs in a thickness range, which is of the order of the bulk magnetic period, it is tempting to anticipate a relation to the magnetic properties of this films.

A direct investigation of the magnetic structure, even for these thin films, is possible by making use of the strong enhancement of the magnetic-scattering cross section at the Ho $M_5$ resonance at 1351.4 eV [3, 18], which amounts to about 6 orders of magnitude. At this energy, the wavelength of 9.2 Å is too large to study the crystalline structures, but it is well adapted to the long-period magnetic-structure of Ho metal. The upper panel of Fig. 4 shows specular x-ray scattering data from a 14-ML thick Ho film at 40 K (filled symbols) and at 180 K (open symbols) well above $T_N$ in the paramagnetic phase, recorded at the resonance maximum. The more pronounced oscillations in the high-temperature data are caused by interference of x-rays scattered at the film-substrate interface and at the film surface (Kiessig fringes) [19]. The additional intensity observed in the low-temperature data is of purely magnetic origin. This contribution, obtained as the difference between the two upper curves, is plotted in the lower panel. The difference yields a $(000 + \tau)$-magnetic satellite which has the same broad shape with Laue oscillations as the Bragg peak. Even though the main maximum is not clearly visible because of total-reflection and footprint effects at small angles, its position is fully determined by the two side maxima. For 14 ML Ho, $\tau = (0.2 \pm 0.04)\ c^*$, i.e. again the bulk value. The data from a 10-ML thick film (lower curve), however, look different. The main peak is considerably shifted to lower scattering vectors, and the fit analysis, assuming a uniform helical structure across the whole film, yields $\tau = (0.08 \pm 0.03)\ c^*$, corresponding to a helix period of about 25 ML.

# 4 Discussion

Epitaxial Ho films on W(110) show a bulk-like magnetic structure down to a thickness of 14 ML. Only for films as thin as the period length of the magnetic structure, noticeable changes occur. The different structure found for these films can be de-

scribed in terms of a helix with longer period, but different magnetic structures with, e.g., a turn angle that changes across the film cannot be ruled out. Since even a 14-ML film is thin on the length scale of the indirect-exchange interaction responsible for the helix, the persistence of a bulk-like structure in these thin films is quite surprising. If the observed change of the magnetic structure around 10 ML is indeed a finite-size effect, related to the bulk magnetic period length, cannot be answered yet, but a detailed systematic investigation of the temperature-dependent magnetic structures of films with different thicknesses will provide further insight. The strong enhancement of the magnetic-scattering cross section at the $M_5$ resonance, which involves an excitation directly into the narrow $4f$ states, provides a strong experimental contrast between magnetic and charge scattering and should make investigations of even thinner films possible.

The technique of soft-x-ray resonant magnetic scattering is applicable to other structures of sufficient extension, like multilayers and other nanostructures, and is not limited to the lanthanide $M_5$ resonances. Scattering at the Fe-$L_3$ threshold has been successfully applied to monitor, e.g., magnetic closure domains in monocrystalline FePd alloys [20], and can be readily used to study magnetic structures in ultrathin transition-metal films as well.

Since in-situ grown films can be investigated without a protective cap layer, the results can be readily related to studies by surface-sensitive techniques like photemission. The influence of cap layers can be inferred from a recent neutron-scattering investigation of a 46-Å ($\approx$ 16-ML) thick Ho film sandwiched between Y layers, where a magnetic structure quite different from that of the present uncapped films was observed, with a shorter magnetic period than in the bulk and with the helix extending into the Y metal [21].

# 5 Conclusions

In this contribution, the magnetic structure of thin epitaxial Ho metal films on W(110) has been studied and the potential of resonant soft-x-ray magnetic scattering for the investigation of ultrathin magnetic films has been demonstrated. The strong resonant enhancement at the Ho $M_5$ threshold allows the investigation of films of only a few monolayers thickness. For Ho films with a thickness comparable to the bulk-helix period, a deviation from a bulk-like magnetic structure is observed. The observation of a bulk-like magnetic structure for a film of 14-ML thickness is

quite surprising, because the indirect-exchange interaction is far reaching and the coupling for layers near surface and interface should be different from that for bulk layers [14]. A detailed investigation of the influence of temperature and thickness on the magnetic structure in the thickness range around 10 ML, which was not possible using hard x-rays, was shown to be feasible in the soft x-ray region.

# 6 Acknowledgments

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# Figure captions

**Fig. 1:** Left: Specular reflectivity in the region of the (002) Bragg peak of a 113-ML Ho film on W(110), recorded at the Ho-$L_3$ resonance without polarization analysis (upper curve) and using a graphite(006) polarization analyzer to suppress the charge-scattering signal (lower curve). Right: Magnetic modulation wave vector ($\tau$, upper panel) and magnetic-satellite intensity (lower panel) of the film (solid circles) compared to data from a bulk single crystal (solid lines from Ref. 5). The satellite intensities have been normalized to equal values at 80 K.

**Fig. 2:** Upper panel: Specular reflectivity in the region of the (002) Bragg peak of a 33-ML Ho film on W(110), recorded at the Ho-$L_3$ resonance with the charge-scattering signal suppressed (solid circles) and unsuppressed (open circles), normalized to the same Bragg-peak height. Lower panel: Difference between the two curves in the upper panel on a linear scale

**Fig. 3:** $c$-axis parameter (double-layer spacing) of various films of Ho/W(110) as compared to bulk data from Ref. 5.

**Fig. 4:** Upper panel: Reflectivity of a 14-ML Ho film on W(110) recorded at the Ho-$M_5$ resonance in the paramagnetic (open circles) and in the helical phase (solid circles). Lower panel: Difference between the curves in the upper panel and respective data for a 10-ML thick film. The solid lines represent the result of a fit analysis assuming a homogeneous helical structure in the film.

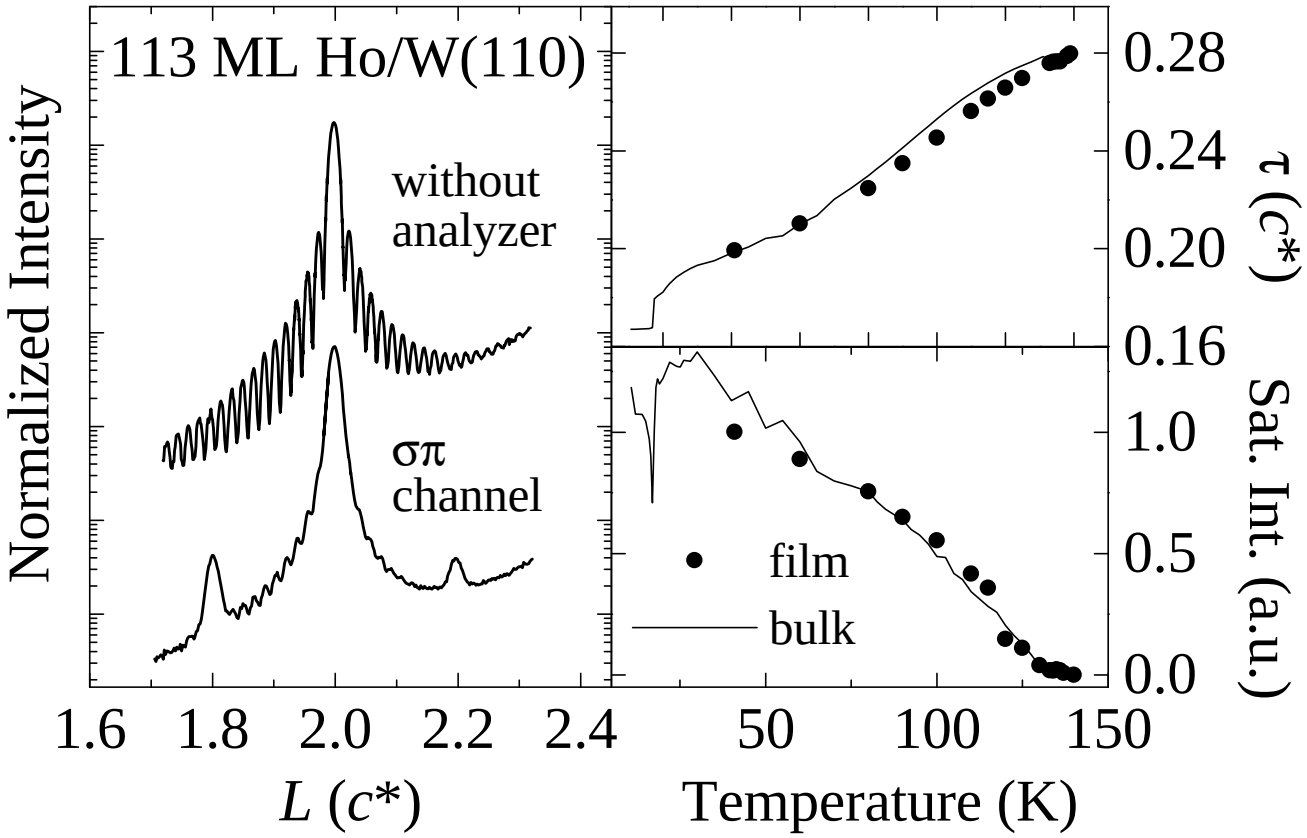
113 ML Ho/W(110)
without analyzer
σπ channel
Normalized Intensity
L (c*)
1.6
1.8
2.0
2.2
2.4
film
bulk
Temperature (K)
50
100
150
0.28
0.24
0.20
0.16
τ (c*)
1.0
0.5
0.0
Sat. Int. (a.u.)

Fig. 1

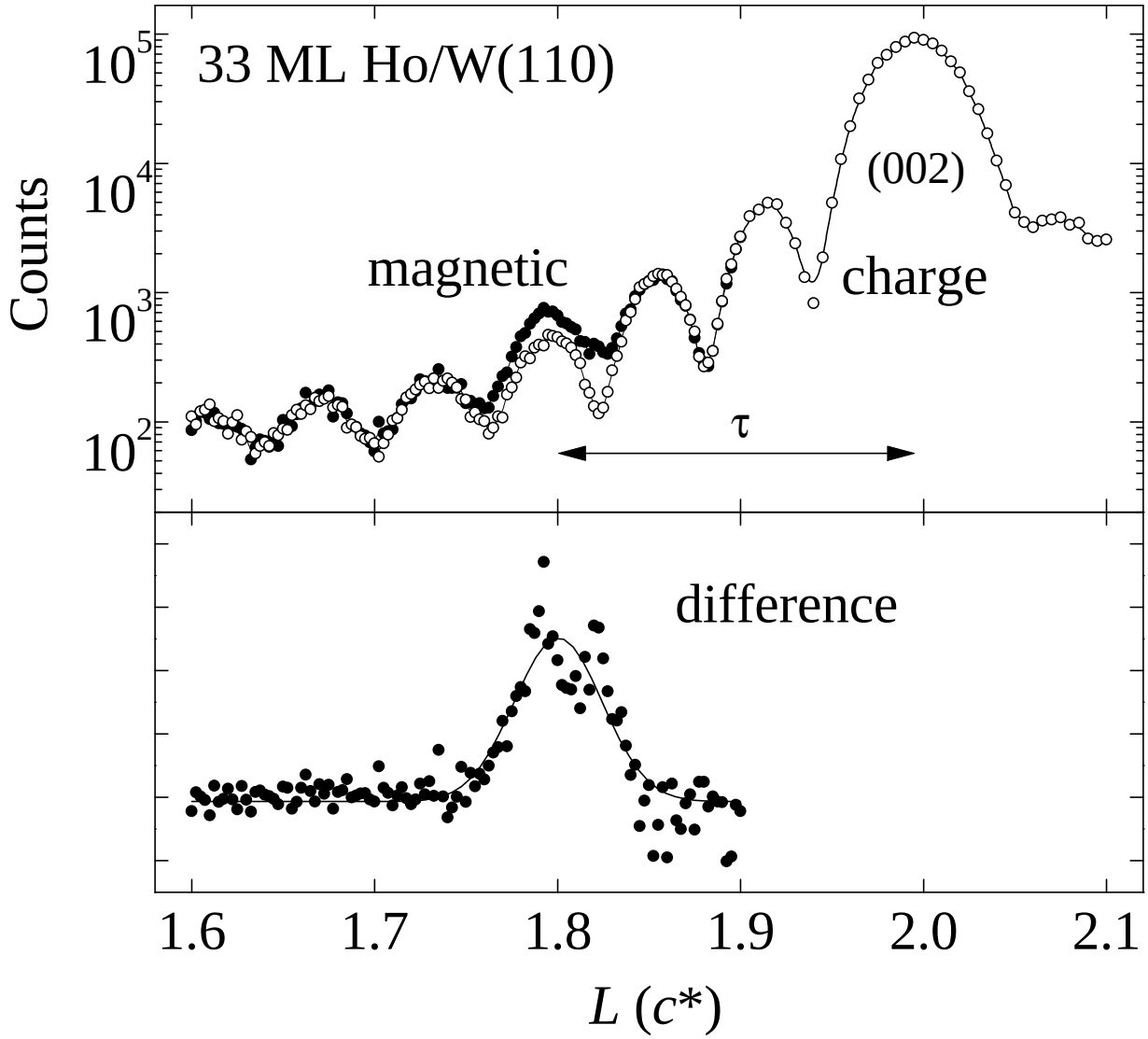
33 ML Ho/W(110)
Counts
$10^5$
$10^4$
$10^3$
$10^2$
(002)
magnetic
charge
τ
difference
1.6
1.7
1.8
1.9
2.0
2.1
$L$ ($c$*)

Fig. 2

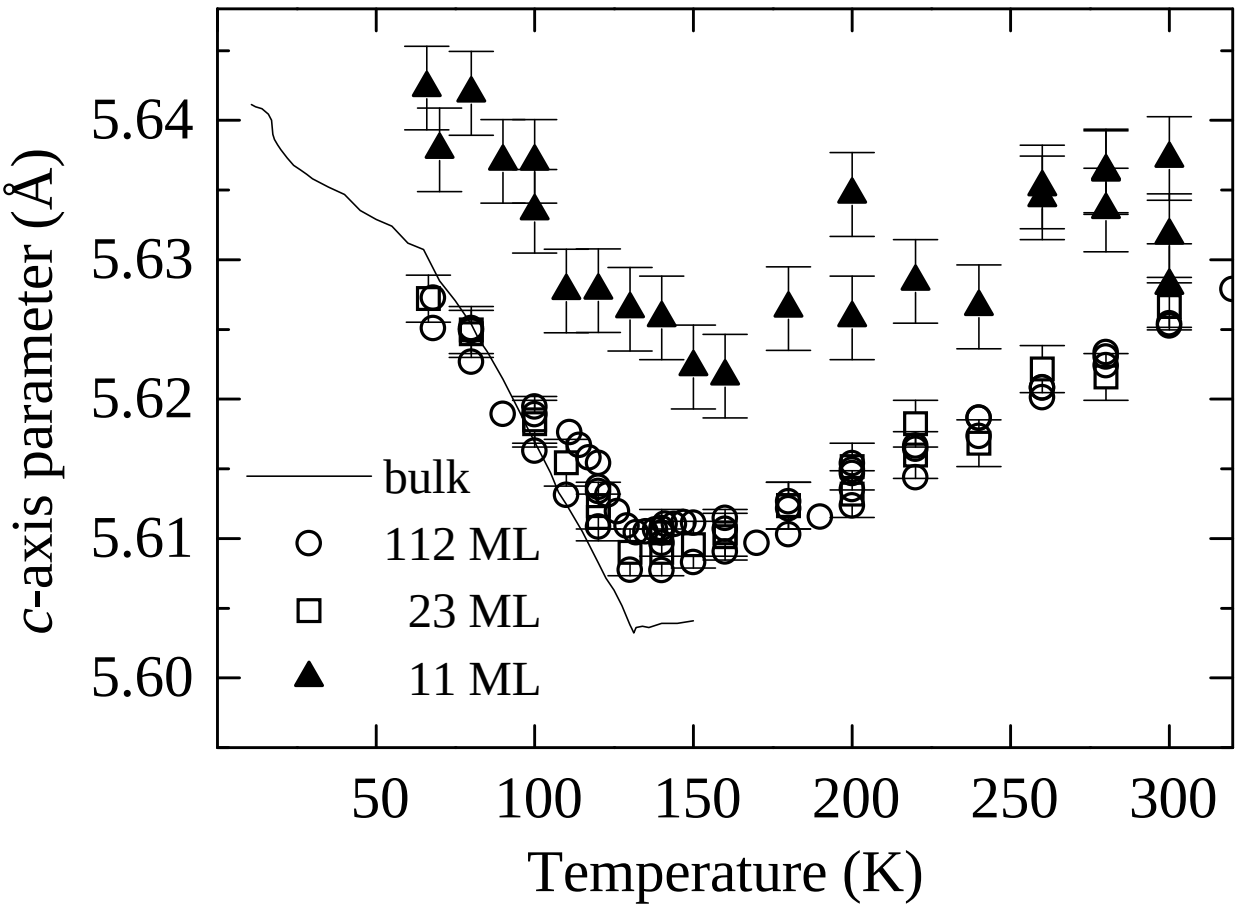
c-axis parameter (Å)
5.64
5.63
5.62
5.61
5.60
bulk
112 ML
23 ML
11 ML
50
100
150
200
250
300
Temperature (K)

Fig. 3

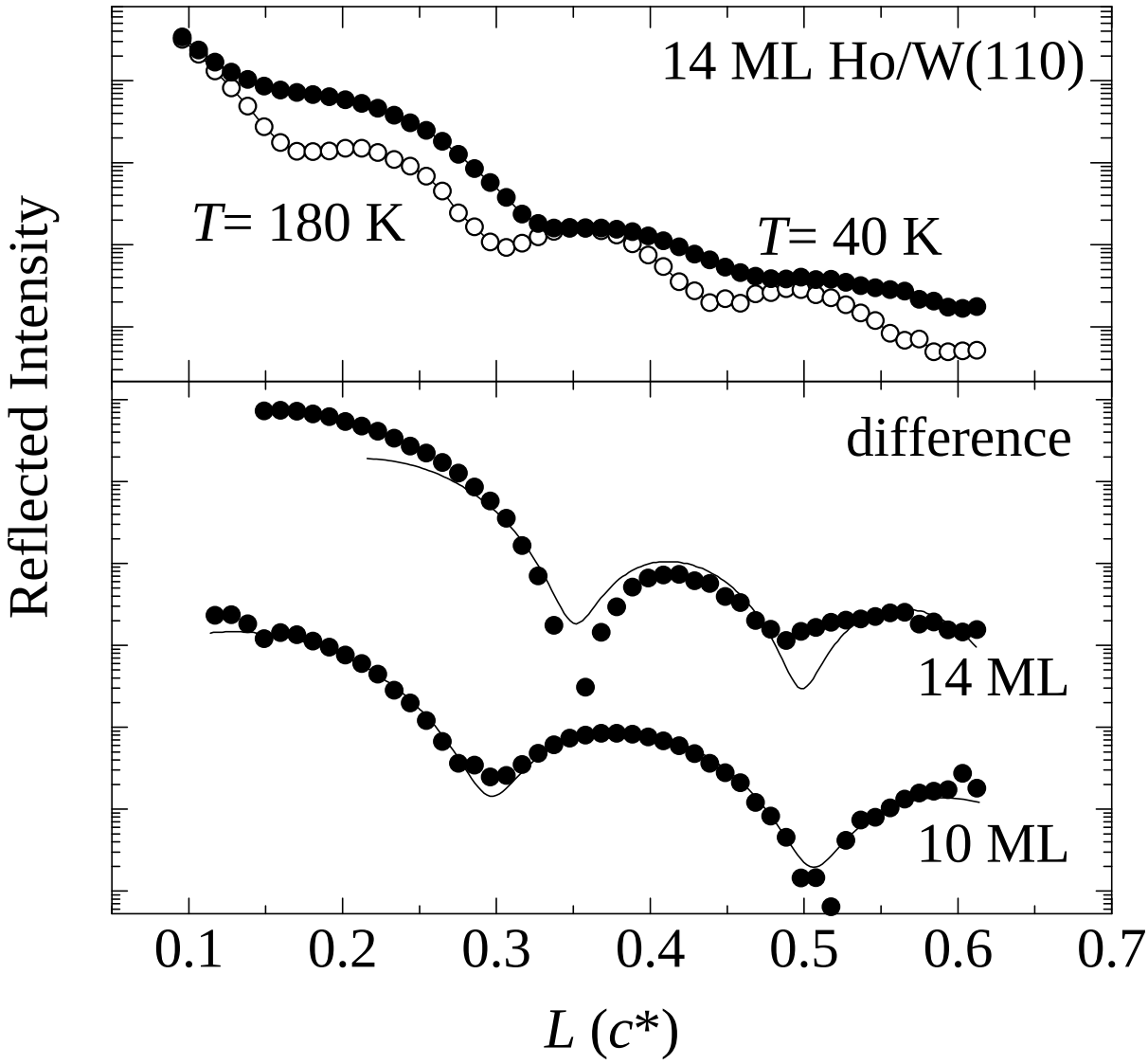
14 ML Ho/W(110)
T= 180 K
T= 40 K
difference
14 ML
10 ML
Reflected Intensity
0.1
0.2
0.3
0.4
0.5
0.6
0.7
L (c*)

Fig. 4