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Grazing incidence X-ray fluorescence of periodic structures - a comparison between X-ray standing waves and geometrical optics calculations

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The data interpretation in the recently published paper with the above title is critised and it is shown that an alternative more physical model based on diffraction in periodic structures can explain the data better and more consistently.

In [1] the authors present a systematic study of the measured grazing incidence X-ray fluorescence (GIXRF) by varying the angle of grazing incidence ϑ and the orientation angle φ between the beam trajectory and the side walls of h=10 nm thick and w=1 μm wide chromium (Cr) stripes, which are deposited in a highly regular manner of parallel stripes on top of a silicon surface, as shown in figure 1.

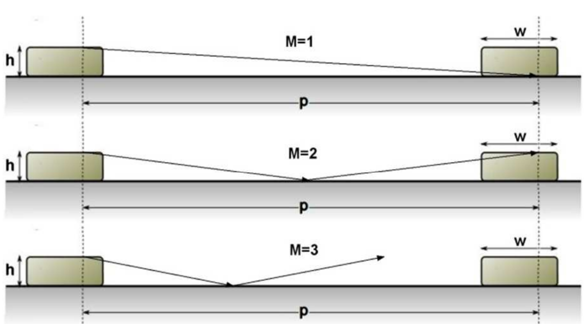

Figure 1: Possible paths for rays, which are refracted in the top surface of narrow chromium stripes on top of a flat silicon substrate. The number M is the number of bounces between two adjacent structures, which is given by $M = \frac{\tan\gamma}{h} p$, where γ is the refraction angle, i.e. it is the grazing inclination angle of the refracted beam with respected to the sample surface. The ratio p/w in the drawing is 6, as used in the experiment, while the height of the stripes is exaggerated compared to their width.

They treat the disturbance produced by the thin stripes with geometrical optics (GO) arguments, assuming that the beam trajectories will be effected only negligibly by diffraction. Oscillations in their GIXRF spectra, which are not present in pure X-ray standing wave (XSW) simulations could then be modelled in a combined XSW/GO approach. In this case one considers that part of the incident beam is following a deviated trajectory after it is refracted at very grazing angle of incidence in the top surface of the stripes. Then one can introduce the number of reflection processes or bounces M for the refracted beam between two adjacent stripes. The meaning of M is indicated in figure 1, in which the empty space, i.e. the stripe separation, p-w between two stripes is 5fold the stripe width w. This ratio corresponds to the properties of the test sample regardless of the orientation angle φ. The authors obtain a good interpretation of the position of all observed additional intensity maxima, when they restrict the number of bounces to the values M=3, M=6 and M=8.

The use of the latter sequence of numbers for M leads immediately to the first question, why no maximum is found for any other than the reported integer numbers and especially not for M=1 and M=2. As shown in figure 1 in order to hit a second stripe a beam being refracted in the top surface of a first stripe will hit the adjacent stripe for any real number for M between 0 and 2. M=1 and M=2 are special cases as the corresponding rays will successively hit further stripes. The second question is then for the meaning of the numbers. Any number larger than M=2, e.g. the reported M=3, lacks a physical justification as the reflecting interface for the second bounce is missing.

The authors of [1] also argue, that they can use geometrical optics (GO), i.e. ray-tracing, assuming that diffraction effects can be ignored. Now as far as diffraction is concerned, this will take place in any feature, which will confine the radiation beam laterally. Such confinement will introduce an angular spread $\Delta\vartheta$ into the transmitted/refracted beam, which order of magnitude can be estimated simply from the ratio λ/l, where λ is the wavelength of the light and l is the relevant feature dimension. The radiation beam is laterally confined mostly on two occasions, when it hits the side walls of the stripes of height h, undergoing relatively small refraction, and when it impinges at grazing incidence onto the top surfaces of the stripes, where it is subject to significant refraction as pointed out in [1]. At the operated grazing angles the incident beam will always see the structures in the projection with their nominal height of h=10 nm. On the other hand, when the stripes are perpendicular to the beam trajectory the apparent width of the stripes of w=1 μm is significantly shortened to $w' = w\sin\vartheta$. For the chosen angular range $\vartheta \leq 1°$ this leads to $w' \leq 17.5$ nm, i.e. the projected stripe width w' becomes similar to or even identical with the structure height h. Now the experiment was performed with a photon energy of 7 keV, which corresponds to a wavelength of $\lambda = 0.177$ nm. The related angular spread is thus of the order of $\Delta\vartheta \geq 0.01$ rad ($\geq 0.6°$). This is rather significant as it is of the same order of magnitude than the angle of grazing incidence ϑ. And it is significantly larger than the opening angle $2h/(p-w)$ of the cone of roughly 0.004 rad, in which a ray needs to be contained in order to impinge directly or after a single reflection process onto the side wall of the adjacent stripe. Thus due to this added angular spread only very little of the diffracted intensity will hit the next stripe. As a consequence the model of the authors is very questionable.

In an alternative model it is here proposed to interpret the data as being affected by the diffraction caused in the regular stripe structure with periodicity p [2]. Infact for the incident radiation the structure of size 6 mm x 6 mm presents itself as a regular stripe structure of 1000 stripes of periodicity p=6 μm. As such it will diffract the incident intensity into several diffraction orders. Here the diffraction orders being observed between the incident and the specularly reflected beam, i.e. the 0[th] diffraction order, will be called the internal orders and will be assigned a positive order number n. Then negative orders, or external orders, will be observed between the specularly reflected beam and the sample surface. In the present sign convention the grating equation is reading

$$n\lambda = p[\cos\vartheta - \cos\theta] \qquad (1),$$

where ϑ and θ are the angles of grazing incidence and of grazing diffraction. This equation holds when the grating ruling is perpendicular to the plane of incidence. In the experiment the related orientation angle φ was varied, and thus the effective grating constant is given by $p/\cos\varphi$. For the case $\cos\varphi \neq 1$ one speaks then also of the off-plane diffraction or of conical diffraction [3]. The latter indicates that the diffraction orders are not contained anymore in the plane of incidence but line up on an arc. The number of observable negative orders is limited as these orders can also progress in the substrate. The X-ray standing wavefield (XSW) above the substrate can in principle be calculated and it will require to sum to the plane incident wave all internal and external diffracted orders as plane waves with the corresponding different directions for their trajectories. It goes beyond the scope of this short note to discuss this in more detail.

Instead here a very special situation for the negative orders will be analysed. These orders can progress even tangentially with respect to the sample surface and ultimately they will enter into the substrate. These orders will only be observed when the angle of grazing incidence is larger than a minimum angle, at which the diffracted order will progress parallel to the sample surface, i.e. for $\cos\theta = 1$. The corresponding angle of grazing incidence is found as

$$\vartheta = \arccos\left[-n\frac{\lambda}{p}\sin\varphi + 1\right] \qquad (2),$$

which has some similarity with equation (3) presented by the authors in [1]. It is more convenient to use the series expansion of the cosine function in (1) as the involved angles are very small. This leads then for the same condition to

$$\vartheta = \sqrt{-2n\frac{\lambda}{p}\sin\varphi} \qquad (3).$$

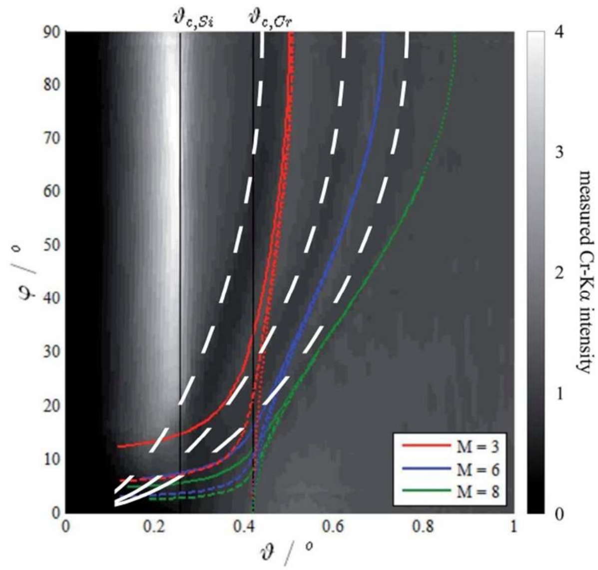

Figure 2: Figure 7 from [1]: Contour plot of the measured Cr-K_α fluorescence intensity. The simulations for local maxima as discussed in [1] are coloured red for M=3, dark blue for M=6 and green for M=8. The dashed white curves present the calculations for local minima according to equation (3) for n=-1, -2 and -3 (from left to right).

In crossing this angle by increasing the angle of grazing incidence a negative order changes from sample internal progression to sample external progression. When a negative diffraction order progresses externally almost parallel to the sample surface it will also excite the chromium fluorescence from the stripes. This possibility will be suppressed, when this order cannot progress anymore externally. As a consequence one would expect to observe a related small local intensity minimum in the fluorescence yield. And indeed this is observed, when the result according to (3) for the first three negative diffraction orders -1, -2 and -3 (dashed white lines) is superimposed over the measured spectra from [1] in figure 2.

Infact all three lines follow rather closely the measured minima. Neither the critical angle for the substrate nor for the chromium stripes have any effect on the diffraction, and thus the dashed white lines also predict minima below the respective critical angles.

The only relevant parameter in the presented treatment is the stripe pattern periodicity p. Neither the ratio between the periodicity and the stripe width nor the stripe height and the shape of the side walls will have any effect on the proposed minima position. These latter parameters, which determine the grating structure factor [2], will then have to be deduced from the intensity distributions into the different orders.

Notes and references

[a] Elettra – Sincrotrone Trieste S.c.p.A., S.S. 14 km 163.5 in Area Science Park, I-34149 Basovizza (TS), Italy.

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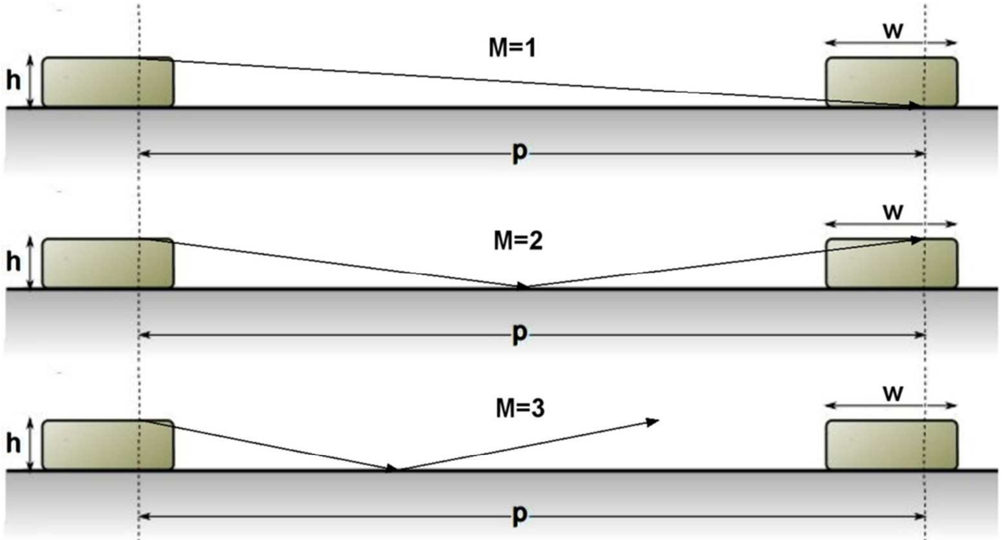

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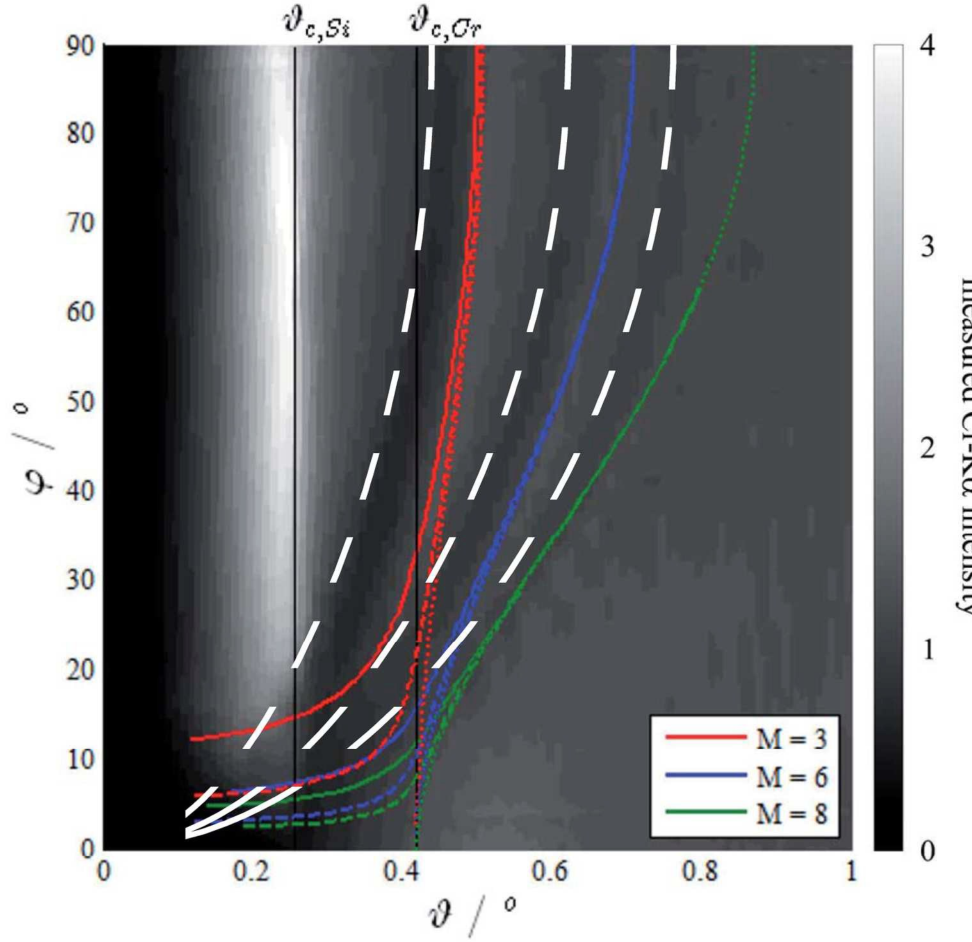

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