Reply to the Comment on "Grazing Incidence X-ray Fluorescence of periodic structures – a comparison between X-ray Standing Waves and Geometrical Optics calculations"

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Abtract: We respond to the comment by W. Jark and D. Eichert on our earlier article concerning geometrical optics based data interpretation of grazing incidence X-ray fluorescence experiments. The potential of the alternative, diffraction based model proposed in the comment is recognized. However, weak points of this method, especially the one concerning its inapplicability to non-periodic structures, are also presented. We reply to the questions raised by the comment giving a more detailed explanation of the parameterization that was used to depict characteristic spectral features. Finally a simple experimental test that can be run to validate both methods is proposed.

The authors would like to thank W. Jark and D. Eichert for their valuable comment [1] on our original paper [2]. We believe that their proposed approach has a high potential. This complementary methodology can be expected to have relevant advantages for periodic structures.

However, the proposed diffraction-based model cannot be applied to non-periodic structures such as deposited nano- and microparticles [3,4] or dried liquid droplets [5,6]. Such unarranged systems are very common types of samples in Grazing Incidence X-ray Fluorescence (GIXRF) investigations. Besides, the diffraction based model would hardly apply to interpret grazing emission X-ray fluorescence measurements [7]. In these cases, the geometrical optics approach still provides a match between theory and experiment.

The reader should note that understanding of the GIXRF angular intensity profiles from non-periodic patterns was our main motivation to develop the geometrical optics approach [8]. The numerical model was first tested on simple geometrical systems, e.g., periodic cuboidal islands. During these tests we encountered intriguing intensity modulations whose intensities and positions were very sensitive to the period and height of the model structures. These intensity variations were completely unexpected and were not confirmed by any other theoretical models presently used for GIXRF assessments [7].

The main motive to the work presented in our article was to experimentally verify the existence of such spectral features. And indeed, these features were observed.

When analysing simulated data, we noticed that the modulation maxima positions could be parameterized by M – the number of bounces the refracted beam would process between two adjacent stripes if there was a continuous Cr layer. With some modification of the imaginary part of the Cr refractive index we found that such a parameterization could also reproduce the modulation positions in the real data (see Figure 1).

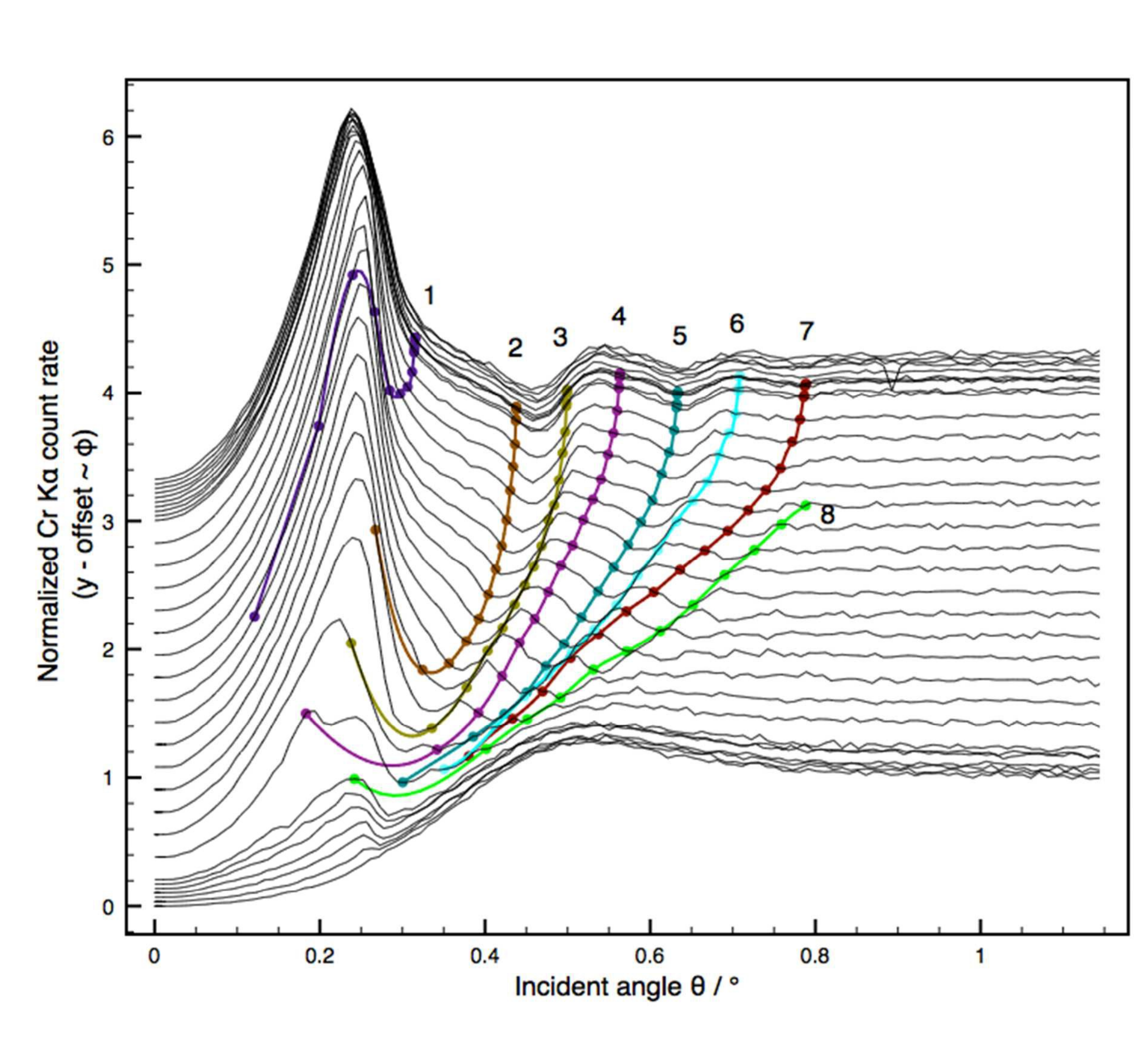

Fig. 1 Measured GIXRF profiles superposed with lines corresponding to M = 1, ..., 8. The figure presents the same experimental data as presented in Figure 2 in the Comment [1].

The authors of the comment pose two particular questions: (1) why no maximum is found for any integer numbers other than M = 3, M = 6, and M = 8 in particular for the numbers "between 0 and 2"; and (2) what is the physical meaning of M as, in case of our particular sample, for any number larger than M = 2 the reflecting interface for the second bounce is missing.

Beginning with the first question, the integer numbers M are in fact markers of the increased probability of interference effects. As is well known, interferences can be either constructive or destructive. In Figure 1, the measured GIXRF profiles are superposed with lines corresponding to M = 1, ..., 8. These lines fit to positions of either local maxima or minima in the angular intensity profiles. In particular, modulations for M = 1 and M = 2 could explain the asymmetry of the particle like peak as they both fit into the broad structure at the high angular part of the peak.

The answer to the second question seems challenging. The authors of the comment are right that for any number higher than M = 2 the reflecting interface for the second bounce is missing. Still, in the simulated spectra the modulations for higher M values are clearly seen but no physical explanation could be found so far for these higher M contributions.

We were considering taking diffraction effects in periodic systems into account, but we decided to concentrate on the geometrical optics modelling approach due to our long term interest in non-periodic structures and to probe first the goodness of this approach with well characterized periodic structure samples. This first test was found to be successful since, as stated by the authors of the comment, our method leads to a good interpretation of the position of all experimentally observed spectral features.

However, it would be interesting to find a link between geometrical optics driven interferences and diffraction. It might be indeed possible that some particular diffraction effects can be rendered with the geometrical optics calculations.

Finally, the validation of both geometrical optics and diffraction approaches is to be conducted by appropriate experiments. A simple experiment comparing GIXRF angular profiles of two samples with striped structures differing only in thickness could be sufficient. The authors of the comment claim that the calculated diffraction pattern should not depend on the height of the structure. The geometrical optics method states the opposite. In this respect, future experiments are expected to guide us towards the determination of the applicability ranges of both approaches.

Acknowledgments

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