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**Als-Nielsen, Jens Aage**

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# AN EXAMPLE OF PROGRESS AND FUTURE PERSPECTIVES IN X-RAY SYNCHROTRON DIFFRACTION STUDIES

Jens Als-Nielsen
Physics Department

Abstract. The development ir diffraction studies of 2-dimensional mono-atomic films physisorbed on graphite over the past decade is described. Synchrotron X-ray diffraction studies represent a break-through in this field and future possibilities are mentioned.

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CONTENTS

## INTRODUCTION

The present report describes one out of numerous examples of scientific progress based upon the rapid development of synchrotron radiation as an X-ray source. The progress can be seen as a series of jumps which could be continued by the construction of dedicated synchrotron sources such as the ESRF. We are convinced that analogous case stories will be abundant around the dedicated synchrotron sources now under construction in USA and Japan. The question is whether Europe will or will not take part in this fascinating development. And now to one specific case story.

In recent years it has been realized that structures of matter, being for example crystallographic structures or magnetic structures, and associated phase transitions are intimately related to the dimensionality. It turns out that knowledge of fundamental structures and phase transitions in the 2- or 4-dimensional world are of great importance for the understanding of similar phenomena in our ordinary 3-dimensional world. The present description is concerned with examples from the two-dimensional universe, which apart from its fundamental features aluded to above may also be of more direct importance in context with our understanding of entities such as the biological membrane or catalytic surfaces for chemical reactions.

The experiments to be considered here involve a substrate supporting a two-dimensional, mono-atomic film. Ideally, the lateral interaction between the substrate and the film should be negligible or at least small compared to the intra-atomic forces between atoms in the film, and graphite is in this non-trivial respect a good substrate. The structure of the two-dimensional film can in principle be studied by diffraction but the number of diffracting atoms in the film is of course much smaller than that from a 3-dimensional crystal. However, a substantial exposed film area can be obtained in certain commercial graphite products such as grafoil. They consist of a huge number of hexagonal graphite layers with cavities here and there in which the physisorbed film can be formed. The diffraction pattern from the films may thus be thought of as a two-dimensional powder pattern. Ideally one would like to have a large number of cavities N each having a large lateral extent L, so that the two-dimensional character is not being masked by uncontrolled boundary effects. Unfortunately, it turns out that in practice one cannot obtain large N and L simultaneously so it is quite clear that a large L substrate requires a strong radiation source and/or a large scattering cross section. In low energy electron diffraction the scattering cross section . so large that a considerable fraction of the incident beam is scattered by a single film and one has indeed obtained LEED patterns from films on the face of single crystal substrates. However, the resolution is relatively poor, the intensity of the Bragg peaks is severely influenced by multiple scattering and the required high vacuum makes large parts of the phase diagram inaccessible. We shall here consider two other scattering probes, the neutron and the X-ray photon, where these fundamental difficulties do not exist but where the source strength become the limiting factor.

## HISTORY

Figure 1 displays the development up to 1981. Published data are here replotted with a common abscissa and intensities scaled to yield the same area under the diffraction peak. The first diffraction pattern was obtained by Kjems, Passell et al (ref. 1) at Brookhaven National Laboratory in 1974 using neutron scattering, top part of fig. 1. The adsorbed film consisted of nitrogen molecules, which have a large neutron scattering cross section, physisorbed on grafoil with a film area of several hundred $m^2$ in the beam but with a typical cavity length L of only about 100 Å. The signal is small compared to the background from the substrate but with the empty cell count rate subtracted a diffraction peak from the nitrogen film is clearly observable.

This pioneering work led to a number of subsequent neutron scattering studies (ref. 2) including inelastic spectra both in USA (mainly at Brookhaven) and in Europe (mainly at Risø).

In 1978, Horn, Birgeneau and co-workers demonstrated that it was possible to obtain accurate diffraction line profiles from a graphite product with a considerably larger value of L using X-rays from a 12 kW rotating anode, see middle part of fig. 1. The sample was Krypton films physisorbed on a graphite product called ZYX and the exposed film area is about 50 $cm^2$. The large value of L is reflected in a sharper Bragg peak and the data indicated L ≈ 450 Å which was consistent with estimates from the rounding of specific heat data. Although the finite-size limitation seemed to be a substrate property it was nevertheless decided to carry out an experiment at the Stanford Synchrotron Radiation Laboratory (SSRL) using the high resolution 3-axis spectrometer developed there by Moncton, Brown et al (ref. 4). The resulting line profile is shown in the bottom part of fig. 1, yielding a value of L around 2000 Å.

Without going into any detail concerning the Krypton experiment we cite from the conclusion of ref.4: "...use of a synchrotron source has enabled us to improve our resolution by a factor of 20 and elucidate physics which was entirely unexpected."

In a subsequent study on the melting of 2-dimensional solid Argon in HASYLAB the substrate was characterized by a Krypton line-profile and results almost identical both with regard to intensity and L were found (ref. 5).

However, since Ar is a weaker X-ray scatterer than Kr, the intensity at the HASYLAB set up is marginal for studying the melting transition (ref. 6), see right hand part of fig. 2.

The experimental set up was optimized to the best compromise between intensity and resolution, but as the X-ray source was unfocussed radiation from a bending magnet (ref. 6), there appeared to be the possibility for a substantial improvement by using the 7-pole wiggled, focussed beam line at SSRL. The left part of fig. 2 shows recent, unpublished results, ref. 7. Again an order-of-magnitude improvement is apparent and a detailed study of the melting of Argon films is indeed feasible at the SSRL facility.

## CONCLUSIONS

The development of a particular field within 2-dimensional physics during the last decade is primarily due to improved radiation sources. We have described the steps from neutron scattering to the rotating X-ray anode, further to synchrotron radiation from a bending magnet, and finally to wiggled, focussed synchrotron radiation. For each jump an order-of-magnitude improvement was obtained with entirely unexpected physics emerging. The last jump to the wiggled focussed X-ray beam is not possible in Europe presently but hopefully such a beam line will be available in HASYLAB some time in 1984. We see no reason at all why the development necessarily should stop here and it certainly will not in the U.S.A. Clearly it would be most valuable to study lighter atoms than Ar with equivalent precision. More fascinating, it seems now within reach to use X-ray diffraction from just one single film on a single crystal substrate, and there can be little doubt that unexpected interesting physical phenomena will be discovered. However, this particular field of research may indeed have passed its pioneering stage when the ESRF is ready to operate in about 7 years from now. Our conclusion is that synchrotron radiation has already opened up new perspectives in a variety of areas in different sciences, and the present description is just one out of numerous examples. Each time the source characteristics have improved substantially new physics has emerged and there is every reason to believe that the science around the ESRF source will follow this pattern.

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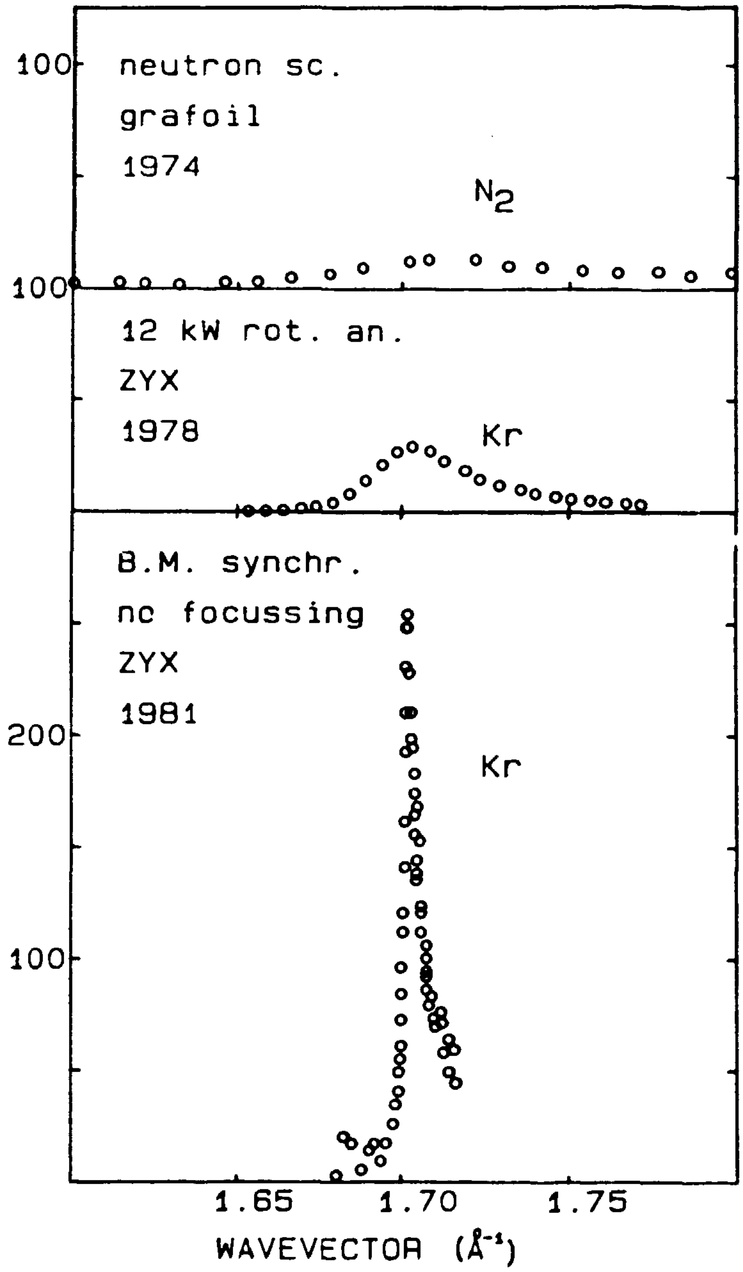


Figure 1. Diffraction profiles of the (1,0) reflection of a registered √3×√3 mono-atomic film on graphite. The data from ref. 1, 3 and 4 have been scaled so all peaks appear with the same area.

Top: Neutron data of $N_2$ on grafoil, ref. 1.

Middle: Rotating anode X-ray data of Kr on ZYX graphite, ref. 3.

Bottom: Same sample as middle panel, but using synchrotron radiation from a bending magnet without focussing devices, ref. 4 and 5.

Ar on ZYX graphite
Synchrotron data

7-pole Wiggler
2 mrad mirror

Bending Magnet
No mirror

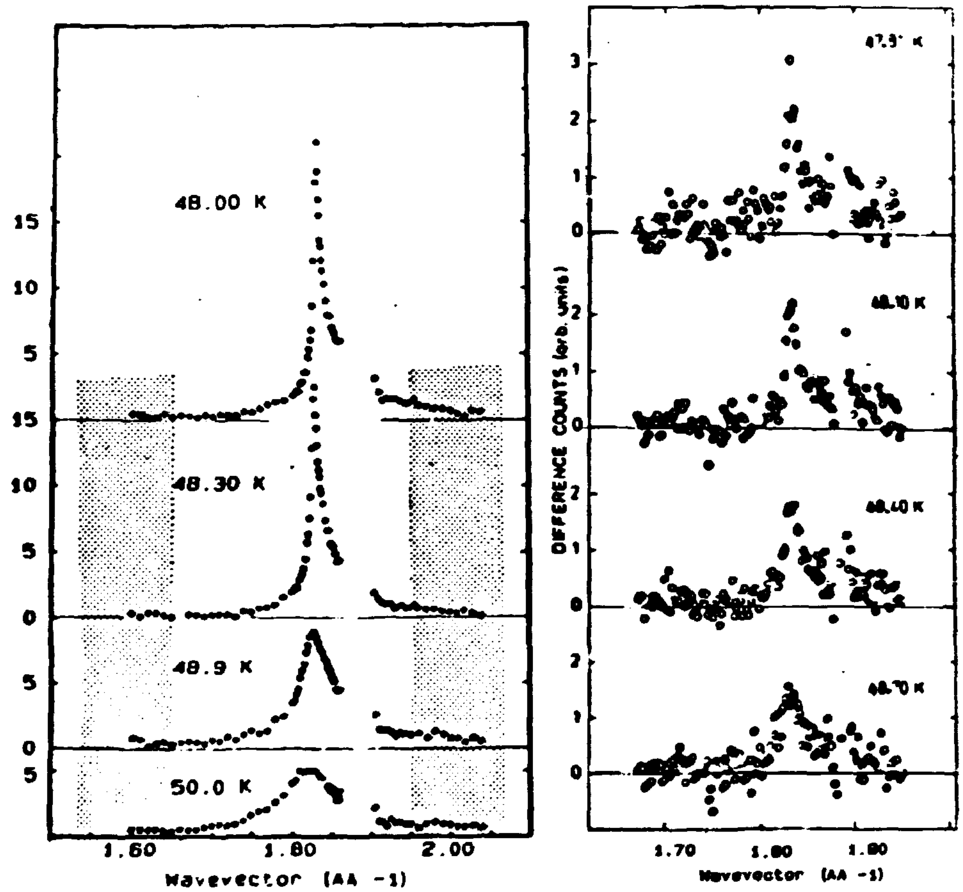


Figure 2. Melting of 2-dimensional Ar on ZYX graphite. The solid phase is incommensurate with the underlying graphite substrate lattice.

Right panel: Synchrotron source equivalent to that for bottom part of fig. 1, ref. 6.

Left panel: 7-pole wiggler, 2 mrad focussed synchrotron radiation, ref. 7.

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Title and author(s)

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Jens Als-Nielsen
Physics Department, Risø National Laboratory,
DK-4000 Roskilde, Denmark

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Department or group
Physics

Group's own registration number(s)

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Abstract

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