High-resolution X-ray Scattering

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D. E. Moncton
Brookhaven National Laboratory
Upton, New York 11973

BNL--32304

and

DE83 004596

Bell Laboratories
Murray Hill, New Jersey 07974

G. S. Brown
Stanford Synchrotron Radiation Laboratory
Stanford, California 94305

Abstract

Beamline and spectrometer instrumentation for high-resolution x-ray scattering at the Stanford Synchrotron Radiation Laboratory is described. The combination of photon intensity at the sample of $\sim 10^{13}$ photons/sec/ 10 mm^2 and the momentum-transfer resolution of $\Delta Q \sim 10^{-4} \text{ \AA}^{-1}$ make this a unique facility. Examples are given of data obtained on two-dimensional phase transitions both in studies of thin (2 molecular layers) liquid-crystal films and rare-gas monolayers on pyrolytic-graphite substrates.

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I. Introduction

In a general x-ray scattering experiment, both x-ray momentum Q and energy E are exchanged with a sample material. Each of these quantities can be measured with a certain precision, ΔQ and ΔE , respectively, ultimately limited by the instrumental resolution. Considering both the instrumental approaches and the physics being pursued, a natural separation occurs between experiments emphasizing momentum resolution and those emphasizing energy resolution. Synchrotron x-ray sources permit, in principle, a dramatic increase in the resolution in each of these domains. Instrumentation allowing momentum resolution on the order of $\Delta Q \approx 10^{-4} \text{ \AA}$ has been under development at SSRL (Stanford Synchrotron Radiation Laboratory) since 1978. The culmination of this effort has been the establishment of a dedicated scattering beamline based on radiation from an 8-pole wiggler electromagnet.

The instrumentation comprising this beamline and the associated x-ray scattering spectrometer will be described in Section II. A discussion of intensity considerations is given in Section III, and the resolution of the instrument is described in Section IV. A brief illustration of the performance of the apparatus is provided in Section V in the context of some recent experiments. Finally among the conclusions drawn in the last section, some remarks are made about the possibilities for high-energy resolution x-ray scattering.

II. Instrumentation

For the past two years, a beamline shown schematically in Fig. 1

at the Stanford Synchrotron Radiation Laboratory (SSRL) has been dedicated to general x-ray scattering experiments. Radiation for this beamline (SSRL VII-2) comes from an 8-pole electromagnetic wiggler running generally at fields of 14-18 kG. Its critical energy is given by ϵ_c (keV) = 0.0665 B(kG) E_e^2 (GeV). Therefore, at typical running energies $E_e = 3.0$ GeV, a 18 kG field provides $\epsilon_c = 10.8$ keV. The performance of this device is described in detail in an article by Brown et al.¹ in these proceedings. As shown in Fig. 1, the remaining components of the beamline are optically similar to the first focussed beamline (II-3) constructed at SSRL.² In the present beamline, the radiation emitted by the wiggler is collimated by adjustable slits (S1) and then focussed by a 58 cm long mirror located in a UHV tank 13 m from the source. The mirror is fabricated from fused quartz in a cylindrical geometry. With a radius of 100 mm it is designed for operation at an incident angle of 7.7 mrad. The mirror surface is coated with platinum (~1000 Å) which gives a critical energy of 10.2 keV. Approximately 4.6 mrad of radiation is accepted in the horizontal plane with the slits fully opened. The mirror is mounted in a bending device, which induces a constant curvature (radius ≈ 1.6 km) in the direction along the beam path to produce a toroidal figure which is an excellent approximation to the ideal surface, namely, an ellipsoid of revolution.³ Thus, the radiation is effectively focussed in both the horizontal and vertical direction.

The converging radiation is incident on a second slit (S2), which together with slit S1, provides the principal vertical collimation for the incident beam. The S1-S2 collimating system establishes a physical reference for the incident beam which is independent of the x-ray source

position and angle of emittance. Thus the beam provided to the monochromator may vary in intensity as a result of source and steering drifts, but not in incident angle. These intensity drifts can then be renormalized with the use of an incident beam intensity monitor. In the absence of an effective collimating system, diffraction experiments continuously suffer from drifts in angle and in the energy provided by the monochromator. With such collimation, data reliability is considerably increased.

The monochromator for this beamline is a fixed-separation, double-crystal device⁴ operating in a helium atmosphere. The first crystal is 3.18 cm thick and water-cooled to prevent thermal distortion, whereas the second crystal is thinner (0.64 cm) and requires no cooling. This arrangement is found to have adequate thermal stability in the high intensity (100 watt) focussed wiggler beam. To date all diffraction experiments have been done with a fixed energy of approximately 7.1 keV, but scanning is routinely done with identical monochromators in spectroscopy experiments. A range of Bragg angles from 30° down to 10° is appropriate for the use of the (111) reflection of silicon or germanium since it corresponds to the region from the Be window absorption cutoff of ~4 keV to 12 keV, somewhat above the mirror cutoff.

The final component of beamline instrumentation shown schematically in Fig. 1 is the x-ray spectrometer. This device is a Huber 5020 six-circle goniometer permitting access to the full three-dimensional reciprocal space of the sample using the θ , χ , and ϕ circles. The scattering angle is defined by the 2θ circle which carries a two-circle goniometer for angular and spectral analysis of the

scattered beam. All of the motions are driven by stepping-motor/gear reducer systems capable of making angular steps of 5×10^{-4} degrees. The absolute angular setting accuracy of each of these circles is generally about 5×10^{-3} degrees but the reproducibility and relative setting accuracy for motions less than 0.1 is considerably better than the minimum step size mentioned above. Care has been taken to insure that the operation of this spectrometer in the unconventional vertical-diffraction-plane attitude does not seriously compromise the precision of the instrument. We also note that x-ray counting, both in the monitor detector and in the scattered photon detector, is done with standard NaI scintillation counters.

III. Intensity Considerations

Having described the beamline instrumentation, we now proceed to discuss the performance of the instrument, in terms of photon intensity and momentum resolution. The intensity of wiggler radiation produced in beamline VII-2 (per 100% bandwidth) at a typical diffraction energy of 7.1 keV is 2.3×10^{16} photons $\text{sec}^{-1} \text{ mrad}^{-1}$, assuming SPEAR operating at $E_e = 3.0$ GeV and $I = 50$ mA. To determine the flux incident on the sample, it is necessary to take into account the effective energy bandwidth of the monochromator and the horizontal angular acceptance of the mirror. For Ge(111) the bandwidth is $\Delta E/E = 3.2 \times 10^{-4}$, while for Si(111) the corresponding number is 1.3×10^{-4} . The horizontal acceptance of the mirror can be varied up to 4.6 mrad using slits. Thus, under the most favorable conditions, the incident intensity is theoretically 3.3×10^{13} photons/sec. In practice one must allow for the transmission of the Be windows and the short section of helium-filled line, as well as the

reflectivity of both the mirror surface and the monochromating crystals. None of these represent large corrections, so that the available flux is in excess of 10^{13} photons/sec. Ideally the size of the focussed beam would be equal to the source size, about 1 mm high $\times$ 4 mm wide. However, mirror figure errors in combination with the intrinsic aberrations of the ideal ellipsoid⁵ produce a "bow-tie" image of the electron beam. In practice, however, over 50% of the intensity is passed by a slit 2 $\times$ 4 mm at the sample position.

It is instructive to compare these intensities with those available on standard fixed-target x-ray sources. For a 1 kwatt copper tube operating with a 50 kV, 20 ma electron beam, the characteristic Cu K α radiation at 8 keV is produced with about 0.1% efficiency and radiated into a 2π solid angle from a source typically 1 mm² in size. Since the natural bandwidth of the Cu K α line (~ 3 eV) is similar to that produced by the synchrotron monochromator using Ge crystals, we need only compare angular and spatial brightness. For the synchrotron we find $dI/d\Omega dA = 0.9 \times 10^{13}$ photons sec⁻¹ μ steradian⁻¹ mm⁻² where as for the fixed-target x-ray tube $dI/d\Omega dA = 1.2 \times 10^7$, about six orders of magnitude less. Typically the fixed-target experiments can collect up to 100 μ steradians with suitable optics, but this choice reduces the angular resolution. Also, higher intensity rotating-anode tubes can provide up to 50 times higher electron beam currents, so that the gain associated with synchrotron radiation may be reduced to a factor of about 200 for experiments requiring only modest angular resolution performed on 50 kW rotating anodes. Nevertheless, as we shall illustrate shortly, the resolution is critical to many questions of the structural physics of phase transitions, so brightness is often the proper comparison to make. Before

discussing actual experimental results, however, a brief discussion of resolution is appropriate.

IV. Resolution

In general, the resolution of an x-ray diffraction instrument is the precision with which one can define the x-ray momentum transfer, $\vec{Q} = \vec{k}_i - \vec{k}_f$, where $\vec{k}_i$ and $\vec{k}_f$ are the incident and final photon wave vectors. Since the momentum transfer $\vec{Q}$ is a vector quantity, there are three spatial dimensions associated with the resolution function $R(\Delta\vec{Q})$, two in the diffraction plane defined by $\vec{k}_i$ and $\vec{k}_f$ and one perpendicular to it. The resolution function falls off monotonically in all three directions and its integral is unity. It is often convenient to think of the resolution volume as the volume enclosed by the 50% contour of the resolution function. In principle it is possible to both calculate and measure this function, but in practice difficulties arise for both. For calculations, one must have detailed functional forms for all the angular, spatial, and spectral characteristics of the real radiation including the effects due to finite electron beam size and divergence as well as the single electron radiation functions. Furthermore, one needs to parameterize the focussing mirror, the double crystal monochromator and the single crystal analyzer crystals completely in all relevant dimensions. Finally, an appropriate convolution of all these functions must be done. So far, no sophisticated attempt has combined all of these ingredients but substantial progress is being made.⁶

It is useful to compare this problem to the similar one in neutron scattering. In that case the problem has been effectively solved⁷ by

using gaussian functions to approximate crystal reflectivities and collimator acceptance functions. For neutrons considerable simplification results from the diffuse, uncollimated source characteristics, the lack of reflecting optics, and the use of mosaic rather than perfect crystals. We have, therefore, relied heavily on measurements of the resolution in our data analysis thus far. The measurement is not trivial either. Since, by definition, the resolution is a property of the instrument, a "sample" with a delta-function cross-section $S(Q) = \delta(Q-Q_0)$ must be chosen for resolution measurements. However, every real material has a nonvanishing scattering power and thus a scattering function of finite width. To make measurements in which the sample diffraction width is small enough to be ignored, the sample's scattering power must be significantly lower than the monochromator. Fortunately we have been carrying out a set of experiments on liquid crystal films^{8,9} which in certain phases have essentially perfect crystalline character. Since they are composed of complex organic molecules, their Bragg reflections have considerably smaller structure factors than silicon. We believe that diffraction from these systems provides a true measure of the resolution.

In general the most important coordinate in which to ascertain the resolution is parallel to the momentum transfer Q (the "longitudinal" resolution), as shown by a scan through a Bragg peak at $Q = 1.4498 \text{ \AA}^{-1}$ in Fig. 2. A number of important points are illustrated by this figure. First, the size of the longitudinal resolution is shown to be on the order of $\Delta Q = 10^{-4} \text{ \AA}^{-1}$ for this configuration using Si(111) crystals throughout. In combination with the increased intensity of synchrotron sources, the dramatic improvement in resolution over

conventional set-ups (usually $\sim 10^{-2} \text{ \AA}^{-1}$) is responsible for the new physics which is emerging from these instruments. The second point illustrated in Fig. 2 is the trade-off between intensity and resolution permitted by adjusting the vertical and horizontal collimation using slits S1. Finally, we note the appearance of asymmetry in the resolution at large horizontal acceptances. This asymmetry is characteristic of the ellipsoidal mirror geometry. The focussing characteristics of these mirrors are complicated and they have been studied in some detail by ray-tracing techniques.³ For the scattering experiments of interest here, the principal complication is the coupling in which the horizontal divergence drives an additional asymmetric vertical divergence in the converging beam. This divergence exceeds the natural vertical divergence of the source ($\sim 0.2 \text{ mrad}$) for horizontal acceptances in excess of about 2 mrad . Of course many experiments involve measurements of scattering profiles which have a considerably broader intrinsic width, and in these cases the asymmetry presents no problem. However, for careful measurements of sharp profiles, one must sacrifice at least a factor of 2 in intensity to achieve a symmetric instrumental resolution.

The other components of the resolution have not been studied in as much detail, but a few remarks about them are appropriate. The resolution perpendicular to the momentum transfer in the scattering plane, the "transverse" resolution, is related to the longitudinal resolution roughly by a factor of $\tan\theta$, where θ is half of the scattering angle. Thus at low scattering angles the transverse resolution is considerably narrower in reciprocal lattice units than the longitudinal resolution, whereas the reverse is true at high scattering

angles. Finally, the out-of-plane resolution is not determined by the vertical divergence of the synchrotron beam, but rather by the horizontal divergence together with the horizontal acceptance of the analyzer system. In practice this latter contribution usually dominates, typically having a value of $\alpha_h = 20$ mrad. The resulting resolution ΔQ is given by $k_1 \alpha_h = 7.0 \times 10^{-2} \text{ \AA}^{-1}$. This component of the resolution is essentially independent of the scattering angle, unlike the in-plane resolution components.

The scattering-angle, or momentum-transfer dependence of the resolution is an important aspect of the instrument's performance. We examine it in some detail for the longitudinal resolution as shown in Fig. 3 for a few different beam collimations. Unlike data shown in Fig. 2, these measurements were made with strongly scattering samples with broad intrinsic diffraction widths. They are not, therefore, true measurements of the resolution, but only serve to illustrate the strong dependence of the resolution on the momentum-transfer. Comparison of the widths from the data of Fig. 2 with those given in Fig. 3 for $Q = 1.45 \text{ \AA}^{-1}$ shows this effect. The main feature of the resolution shown in Fig. 3 is the deep minimum which occurs at $Q = 2\pi/d$, where d is the monochromator d -spacing. For Si(111) $2\pi/d = 2.0 \text{ \AA}^{-1}$. The minimum occurs at this point because each successive Bragg scattering event experienced by the beam, including that from the sample, occurs at the same absolute scattering angle but in the opposite sense. Thus the correlation between photon energy and direction is preserved throughout the scattering process. In this so-called dispersionless configuration, the resolution is a function of only the intrinsic widths of the monochromating crystals. As the scattering angle is changed, the finite

vertical divergence of the incident beam begins to determine the resolution, eventually dominating the width with a contribution which increases linearly in Q . As the vertical divergence is reduced, the magnitude of this contribution is decreased, as shown in the figure. It is interesting to note that the design of the beamline with mirror preceding the monochromator is essential to achieving the nondispersive minimum in the resolution.

V. Experimental Results

The experimental activity on Beamline VII-2 at SSRL involves a rapidly-growing community of some 30 scientists from over 10 institutions. In the 1-1/2 years since the first operation of this beamline, a wide variety of experiments have been carried out, generally having common needs for intensity and resolution. The experiments range over a number of areas: (1) problems in liquid crystal physics, involving studies of melting in thin films;⁹ (2) surface physics studies of reconstruction of semiconductor surfaces;¹⁰ (3) studies of the phases of adsorbates on graphite^{11,12} and copper substrates;¹³ and (4) studies of incommensurate charge-density wave structures and phase transitions.¹⁴ It is not possible to review all the work in this article, so two examples have been chosen of work with which the present authors are particularly familiar. Both experiments are aimed at the general question of the nature of the crystallization process in two-dimensional (2D) matter. This topic has been discussed in a previous review article on two-dimensional melting.¹⁵ Here we will review the highlights.

A. Liquid Crystal Thin Films

The first studies of 2D crystallization exploited the ability

of some liquid crystals to form freely suspended films. These can be made as thin as two molecular layers (50 Å) with an area of 1 cm². This technique was developed at Harvard¹⁶ and has been exploited in synchrotron x-ray scattering experiments at SSRL.⁹ As shown in Fig. 4(a), the film is drawn across an open hole in a glass cover slide. In the crystalline phase, the molecules order with their long axes perpendicular to the film plane and form a simple close-packed hexagonal lattice. Optical reflectivity measurements can easily measure the thickness of the film, which is found to be uniform over the entire sample area.

In Fig. 4(b) a schematic view of the reciprocal space representing this 2D hexagonal lattice is shown. Bragg rods are observed by scanning the x-ray momentum transfer Q . Figure 4(c) shows data obtained⁸ on a material called 40.8 [N-(4-n-butyloxybenzylidene)-4'-n-octylaniline] using a conventional (50 kW) rotating anode x-ray generator, and Fig. 4(d) shows data on another material, $\overline{14S5}$ (4-n-pentybenzenethio-4'-n-tetradecyloxybenzoate) taken using synchrotron radiation.⁹ We note that the data of Fig. 4(c) took 24 hours to take whereas the data in 4(d) was obtained in a total of 15 minutes. In both cases, the samples are in the 2D crystalline state, where structure factor singularities should theoretically¹⁷ be of the form $S(Q) = |Q-G|^{-2+\eta}$. This form is quite different from the delta-function peaks $S(Q) = \delta(Q-G)$ routinely observed in three-dimensional systems. Simply speaking, the fluctuations in a two-dimensional crystal have a fundamentally stronger effect than in three dimensions and they effectively destroy long-range order. The two-dimensional solid does rigorously exist, however, with properties such as a finite shear modulus, which differentiate it from a liquid.

As long as the exponent η , which depends only on the two elastic moduli of the 2D solid, is less than a critical value η^* which is always between $1/4$ and $1/3$, the solid structure is stable.¹⁸ The solid lines in the figures are fits to the data obtained by convoluting the resolution function (dashed line) with $S(Q) = |Q-G|^{-2+\eta}$, where $\eta = 0.15$ in the data of Fig. 4(c) and 0.13 for 4(d). Detailed studies of the melting behavior of multilayer films from 2 to 12 layers have recently been carried out, further demonstrating the importance of high resolution x-ray scattering for extracting the physics of these systems.

B. Physisorbed Rare Gas Monolayers

To date substantial progress on the 2D melting problem has been made by Heiney et al.¹² using the system consisting of slightly more than one monolayer of xenon atoms, as shown schematically in Fig. 5(a). The xenon Lennard-Jones diameter is larger than the separation of the relevant minima in the substrate potential of the graphite and hence the natural lattice spacing of the adsorbate is incommensurate with the substrate. In Fig. 5(b) we show a series of scans of x-ray intensity versus the radial momentum transfer through the Bragg rods [see Fig. 4(b)]. The disorder of the graphite substrate causes an azimuthal-averaging of the scattering, rendering it effectively cylindrical. In addition, substrate tilt disorder (i.e. in the orientations of the graphite planes) leads to an asymmetric lineshape with excess high- Q scattering. This disorder is, however, well understood and does not prohibit extracting the intrinsic lineshapes as the data of Fig. 5(b) demonstrate. In the experiment, the melting transition occurs at $T_M = 152$ K. Two scans below T_M shown in the upper portion of Fig. 5(b) are well described by

lineshapes based on convoluting the scattering function $S(Q) = |Q-G|^{-2+\eta}$ with the resolution function and performing the appropriate azimuthal and tilt average. In contrast to the liquid crystal case described above, the resolution width here is dominated by the coherence size (~ 2000 Å) of the substrate domains. The data in this 2D crystal phase can be fit within experimental error with the exponent η increasing with temperature to a value at melting in the range predicted by theory.

Data taken in the liquid phase between 152 K and 160 K show a continuous evolution of the correlation length over an entire decade limited only by the finite size of the substrate domains. As shown in Fig. 5(c), the temperature dependence of the correlation length is well described by the theoretical form (solid line) predicted by the current (2D) melting theory.¹⁸ Of course, the most notable observation is that the correlation length grows increasingly rapidly as the melting temperature is approached from the fluid side to a length scale far in excess of those seen in any 3D liquid. These data thus provide very convincing evidence for a transition quite different in character from 3D melting.

VI. Conclusions

Synchrotron radiation has characteristics uniquely suited to new research areas in structural physics by enabling unprecedented intensities and momentum transfer resolution. However, as we mentioned briefly at the outset, the potential exists for extending the impact of synchrotron radiation into areas concerned with the dynamics of condensed matter systems. Preliminary studies have been carried out¹⁹ toward the aim of achieving inelastic x-ray scattering with energy

transfer resolution in the 1 to 10 meV range. The consensus is that back-reflection²⁰ Bragg scattering from high index reflections in perfect-crystal silicon can provide $\Delta E/E \sim 10^{-6}$ to 10^{-7} at photon energies around 15 keV. A spherically bent crystal¹⁹ with a radius on the order of 2 meters would collect a large solid of radiation in order to provide adequate intensities with such a small energy bandwidth. The same techniques could be used for both a monochromator and analyzer thus permitting a spectrometer to study the sample's inelastic structure factor $S(Q, \omega)$. Once this becomes possible, an entirely new class of experiments will be possible looking at low frequency collective electronic as well as lattice-dynamical excitations.

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Figure Captions

- Figure 1. A schematic illustration of the SSRL Beamline VII-2 instrumentation.
- Figure 2. Measurements of the longitudinal resolution shown as a function of the collimation determined by slits S1 (Fig. 1). Percentage figures are given for the relative losses in intensity suffered with improved collimation.
- Figure 3. The momentum transfer dependence of the longitudinal resolution is shown for different collimations.
- Figure 4. a) Schematic representation of the liquid crystal film drawing technique with inset showing an enlarged section of a 2-layer crystalline film. b) The reciprocal space appropriate to a simple 2D hexagonal lattice showing the different directions of the x-ray scans. c) X-ray data obtained on a 4-layer film using a rotating anode x-ray generator with a diffraction apparatus having a resolution shown by the dashed line (width $\approx 0.04 \text{ \AA}^{-1}$). The solid line is a fit of the power-law lineshape described in the text with an exponent $\eta = 0.15$. The inset shows the temperature dependence of η with the two curves representing upper and lower limits of the uncertainty in the fitting procedures (after ref. 8). d) X-ray data obtained on a 2-layer film at the Stanford Synchrotron Radiation Laboratory using the improved resolution available with synchrotron beams (width $\approx 6 \times 10^{-4} \text{ \AA}^{-1}$). The solid line is a fit to the power-law lineshape with $\eta = 0.13$ (after ref. 9).

Figure 5. a) An illustration of a monolayer of xenon adsorbed on the (001) surface of graphite. The solid lines for $T = 135.0$ K and $T = 151.3$ K are fits to a lineshape as discussed in the text. The solid lines for $T = 152.0$ K and 160.0 K are fits to a powder and tilt-averaged Lorentzian (after ref. 12).

c) Inverse correlation length, κ , determined from a Lorentzian lineshape analysis. The solid line is a fit of κ vs. T to the theoretical prediction (see ref. 18).

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FIGURE 1

FIGURE 2

FIGURE 3

(a)

RECIPROCAL SPACE

(b)

FIGURE 4

(c)

(d)

XENON ADSORBED
ON GRAPHITE (001) SURFACE

FIGURE 5