Speckle in coherent x-ray reflectivity from Si(111) wafers

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We report the observation of x-ray speckle in the reflected beam from Si(111) wafers illuminated at grazing incidence. An intense coherent 8-keV x-ray beam was prepared using a wiggler source and multilayer monochromator optics. We demonstrate that the speckle patterns are specific to the region of the sample that is illuminated. From the trade-off between surface sensitivity and signal as a function of perpendicular momentum transfer, we infer that the speckle is due to the surface morphology on a micrometer length scale. We document and explain the evolution of the speckle patterns from nearly specular at low q_z to highly structured at larger q_z. [S0163-1829(97)08536-6]

When a beam of coherent electromagnetic radiation traverses a region that introduces random phase differences across the wavefront, whether by transmission through an inhomogeneous medium or by reflection from a rough surface, the resulting intensity distribution, observed far from this region, will have a granular nature commonly referred to as ''speckle.'' The detailed speckle pattern in the far field can be constructed using Huygen's principle and gives rise to a Fraunhofer diffraction pattern. This pattern contains detailed information about the structure of the region. In the case of reflection from a rough surface, the random phase changes are caused by differing heights across the surface and thus its speckle pattern contains information about its morphology. Prior to the development of insertion devices such as wigglers and undulators at second- and third-generation synchrotron radiation sources, experimental techniques that make use of speckle were limited to optical wavelengths using lasers. The large brilliance of these new sources translates directly into coherent flux and has led to the recent success of coherent x-ray diffraction (CXD) experiments.[1–7] These have shown the utility of x-ray speckle to study both static structure[1,4–6] and dynamics[2,3,7] of condensed matter systems on length scales inaccessible to visible light.

Highly polished surfaces are widely used as ideal dielectric boundaries in optics, not only in the visible part of the spectrum, but also for x rays. For example, to reflect ideally, the surface of a mirror must be ''optically flat,'' meaning it must deviate from planar by significantly less than one wavelength of the light used. If this were not true, then its imperfections could be ''seen.'' For different reasons, for Si wafers to be useful for fabricating electronic devices, their surfaces should be microscopically flat within angstroms over the length scale of a typical device, namely micrometers. Wavelengths in the x-ray range must be used to be sensitive to these imperfections, in any case, and very high resolution is also required to reach the necessary lateral distances.

X-ray reflectivity is a well-known method of measuring surface roughness.[8,9] An ideal surface follows the Fresnel law with a specular reflectivity that falls off with momentum transfer, q, as $1/q^4$. The reflectivity of a rough surface drops more quickly and displays a diffuse component in addition to the specular part.[10] This nonspecular component is related in an average way to the roughness on the surface. If the sample were illuminated instead by a *coherent* x-ray source, then the nonspecular diffuse component would exhibit fine structure instead. This fine structure is a speckle pattern that we will show to be related to the specific morphology of the illuminated portion of the surface. At grazing incidence the footprint on the sample is highly elongated along the beam direction, so the CXD experiment will yield specific information about the surface on a micrometer length scale, but with a field of view extending several hundred micrometers.

In this paper we report the observation of fine structure in the reflectivity from several Si(111) wafers illuminated by an intense coherent x-ray beam. We demonstrate that these features are a specific function of the illuminated region by observing them change as the beam is scanned across the wafer surface. We attribute this ''speckle'' to the surface morphology from its unique dependence on momentum transfer.

A CXD experiment requires a spatially and temporally coherent beam of sufficient flux to gather data in a reasonable time. As is well known, the transverse (spatial) and longitudinal (temporal) coherence lengths from an incoherent source are related to the source size and frequency band-

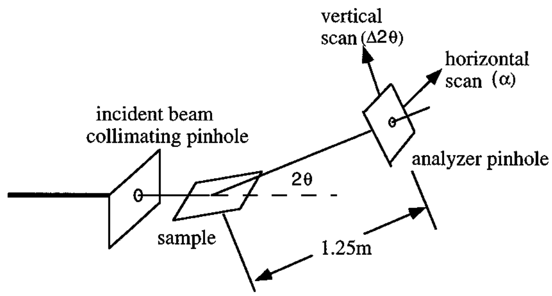

FIG. 1. Sketch of the experimental setup used for the coherent x-ray reflectivity experiments described in the text.

width, respectively. A transversely coherent beam at the sample position can thus be obtained by placing an aperture, with a size smaller than the transverse coherence length, in the beam just before the sample. The parameters for the X25 beam line at the National Synchrotron Light Source (NSLS) yield a transverse coherence length of 55 μm at the sample in the vertical direction and roughly 5 μm in the horizontal direction.[2] In order to insure a spatially coherent incident beam, a pinhole of nominal diameter 5 μm was used.

The restriction on the temporal coherence in a CXD experiment is that the maximum optical path length difference (PLD) among the scattered rays does not exceed the longitudinal coherence length, $\xi_{\rm long}=\lambda E/\Delta E$, determined by the energy bandwidth, $\Delta E/E$, of the source and optics. The PLD between two scattering regions in the specular configuration depends on their surface height difference and on their lateral separation. We estimate a maximum PLD of ≈ 80 Å.[15] Significantly, the PLD does not depend on the penetration of the x rays into the bulk as it does for ''bulk'' CXD experiments. This has allowed us to use multilayer optics with $\Delta E/E \approx 1.5\%$ giving $\xi_{\rm long}\approx 100$ Å. Although longer $\xi_{\rm long}$ values are possible with crystal optics, the use of multilayers has the advantage of 100 times more flux (8×10^6 photons/s through our 5 μm pinhole) while providing adequate longitudinal coherence to observe speckle.

Figure 1 illustrates the experimental setup. The beam incident on the sample was defined by a 5 μm aperture mounted on an x-y translation stage capable of submicrometer resolution. The distance from aperture to the sample was 100 mm. An analyzer aperture, used to scan the reflected beam, was mounted on the two-theta arm of a four-circle diffractometer (main axis horizontal) at a distance of 1.25 m from the sample via a second x-y translation stage. Vertical and horizontal analyzer scans of the 5 μm direct beam without a sample yielded the expected Fraunhofer-like diffraction patterns, verifying the lateral coherence.

Figure 2 shows typical speckle patterns seen in the reflected beam from a Si(111) wafer with a perpendicular momentum transfer, q_z, of 0.1 Å^{-1} using a 20 μm analyzer aperture. Repeated scans made without moving the sample reproduced this result, but moving the beam to illuminate a different region of the sample resulted in dramatic changes in the number and positions of the peaks, as illustrated. Moving the illuminated region by *less* than the beam footprint [compare Fig. 2(b) with 2(a)] resulted in small changes, mainly of relative intensity, while translating the beam to a completely different area changed the speckle pattern completely [Fig. 2(c)]. This demonstrates that the speckle patterns are indeed sensitive to the *specific* arrangement of scattering regions within the illuminated area of the sample. If this same scan had been performed with an incoherent incident beam, it would have resulted in only a smoothly varying diffuse scattering profile. This profile could in principle be constructed by averaging the independent speckle intensity distributions from many scattering domains.

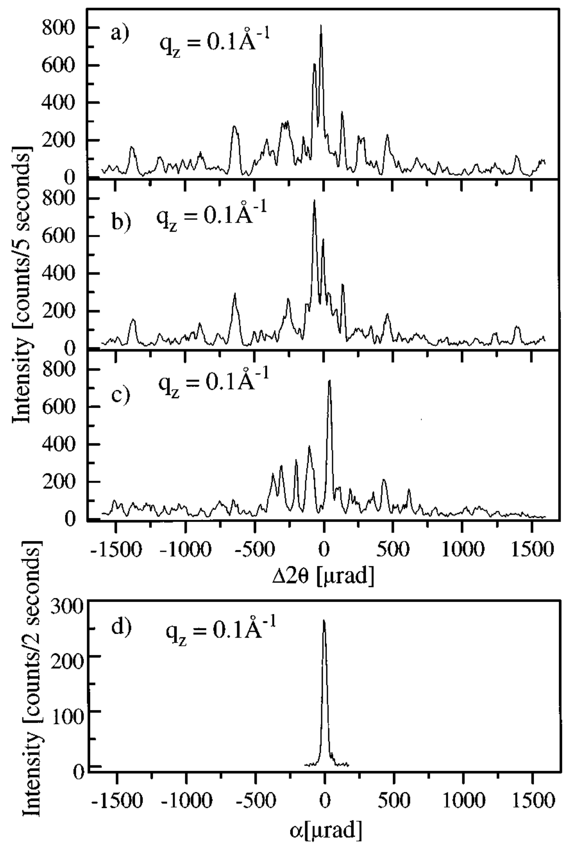

FIG. 2. Scans of the analyzer aperture in the $\Delta 2\theta$ direction (a,b,c) showing ''speckle.'' The beam was moved along the beam direction by 20% of its footprint size between scans (a) and (b). (c) Result of a large movement of the beam to a different area of the sample. (d) Scan of the aperture perpendicular to the $\Delta 2\theta$ direction.

While the scans in the 2θ direction revealed a rich speckle structure, scans of the analyzer aperture in the horizontal direction showed little structure beyond a single peak whose width corresponded to the Fraunhofer diffraction from the 5 μm incident beam pinhole alone. Figure 2(d) shows a scan across the most intense peak in Fig. 2(c) in the direction perpendicular to 2θ. The explanation for this apparently one-dimensional behavior lies in the extreme asymmetry of the incident beam footprint on the sample due to its small angle of incidence.[6] If the spatial extent of the features that give rise to the speckle pattern is greater than the width, yet smaller than the length, of the beam footprint on the sample, then the resulting speckle pattern will be the mutual interference function of a linear array, as observed.

Speckle patterns arise when there is interference between scattered beams that have been somehow phase shifted in their reflection from different regions of the sample. If we consider that the surface will have a spatially varying height

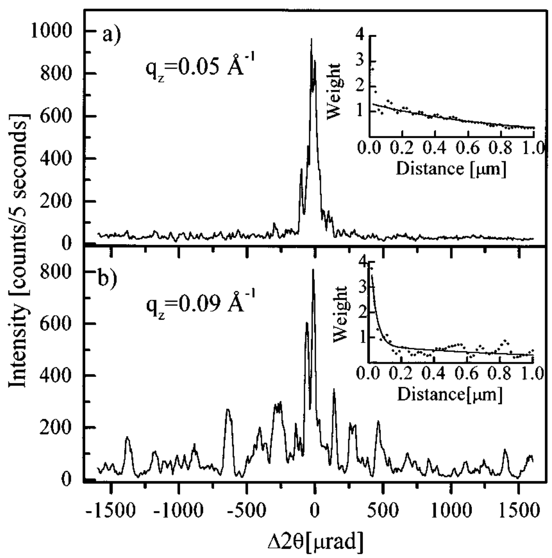

FIG. 3. Scans in the $\Delta 2\theta$ direction of the same silicon wafer at (a) $q_z = 0.05$ Å^{-1} and (b) $q_z = 0.09$ Å^{-1} demonstrating the increased sensitivity to roughness at larger perpendicular momentum transfers. Fourier transforms of the intensity distributions (inset) have been fit with exponential functions of decay length 0.8 μm in (a) and with 0.035 μm and 1.24 μm in (b).

function, $h(x)$, we find that the relative phase of the reflected wave at a given q_z is just equal to $q_z h(x)$. Assuming that all parts of the surface reflect with the same amplitude, the scattered amplitude in the one-dimensional case will be proportional to

$$A(q_\parallel) = \int_{-d/2}^{d/2} \exp(iq_z h(x))\exp(iq_\parallel x)dx,$$

where q_z and $q_\parallel$ are the components of momentum transfer perpendicular and parallel to the surface and d is the length of the illuminated area of the sample. In the case when $q_z h(x) \ll \pi$, the first exponential can be expanded to first order:

$$A(q_\parallel) \approx \sin(q_\parallel d/2)/q_\parallel + iq_z \int_{-d/2}^{d/2} h(x)\exp(iq_\parallel x)dx.$$

In this approximation the scattering amplitude splits into ''specular'' and ''diffuse'' components just as in the situation of classical x-ray reflectivity.[10] Here the distinction between the two components is less clear because they both have peaks of the same width in $q_\parallel$. If Δh represents the maximum height excursion across the illuminated region then $q_z \Delta h \ll \pi$ is the condition for the specular term to dominate, since the phase differences will be too small to cause destructive interference. The intensity is then just a modified version of the direct beam's Fraunhofer pattern. On the other hand when $q_z \Delta h \cong \pi$, destructive interference starts to occur between scattering regions leading to richly structured speckle patterns.

To test this model, the evolution of the speckle pattern was therefore studied experimentally as a function of q_z by changing the angle of incidence, as shown in Fig. 3. The reflected beam with $q_z = 0.05$ Å^{-1} displays nearly specular behavior with only a few speckles present, while at $q_z = 0.09$ Å^{-1} it shows strong fine structure [Fig. 3(b)]. This supports our prediction of increased sensitivity to surface roughness at larger q_z.

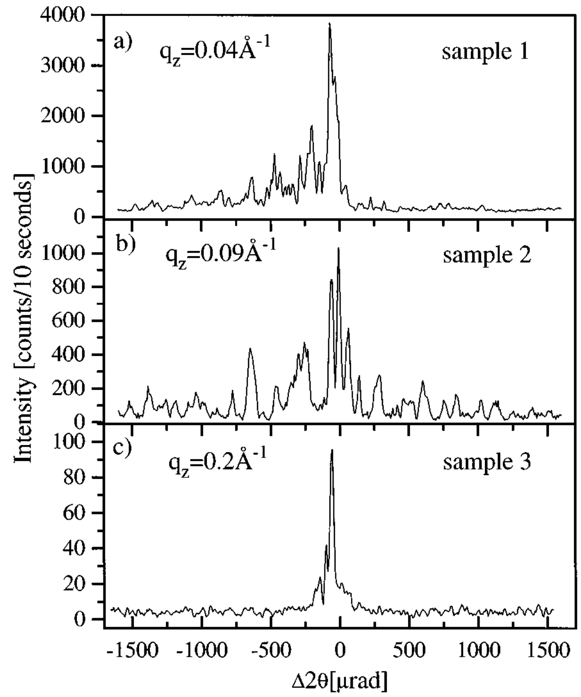

FIG. 4. $\Delta 2\theta$ scans of three different Si (111) wafers. Each shows a different threshold q_z value for the onset of speckle, indicating that each has a different roughness.

The above argument predicts that a sample of roughness $\sigma_{\rm rms}$ (on the length scale of the beam footprint) will have a characteristic threshold $q_z = \pi/\sigma$ beyond which pronounced speckle will appear. We therefore examined Si(111) wafers from different sources with very different roughness values, as measured by stylus profilometry. The surface in Fig. 4(a) has a well-developed speckle pattern already at $q_z = 0.04$ Å^{-1} indicating its vertical roughness is on the order of 80 Å. The sample in Fig. 4(b) that exhibits speckle at $q_z = 0.09$ Å^{-1} would have vertical roughness of about 35 Å and the sample in Fig. 4(c) that remains specular to $q_z = 0.2$ Å^{-1} has roughness less than 10 Å. The rms roughness values determined from the profilometer data for the three samples of Figs. 4(b) and 4(c) were 65, 26, and 11 Å, respectively, in good agreement. Roughness on finer vertical length scales can be probed by observing speckle further out in q_z, but at a cost of total intensity. The present arrangement allows us to follow the reflectivity curve out to about 0.2 Å^{-1} before we run out of signal. Third generation undulator radiation sources with their large increase in brilliance should greatly increase the range of q_z that can be explored using this technique.

The speckle pattern also contains information about the lateral correlations on the surface. The widths of the individual speckles in a pattern each correspond to the width of the central maximum of the Fraunhofer diffraction pattern of the source aperture. They therefore do not reveal any additional information about the surface. However the *overall* width of the speckle distribution is inversely related to the average lateral size of the features causing the speckle pattern. This can be seen most easily by Fourier transformation of the intensity distribution to obtain a real-space correlation function, as shown in the inset in Fig. 3. The Fourier transform of the data of Fig. 3(a) displays a broad component whose decay length approaches the size of the beam. The Fourier transform in Fig. 3(b) shows an additional narrow component due to the correlations between the scattering regions across the surface. The width of this latter component is 0.035 μm measured across the beam, corresponding to a characteristic domain size of about 3 μm along the surface of the wafer.

In summary, we have observed coherent diffraction effects, in the form of ''speckle,'' in reflection from Si(111) wafers illuminated by a transversely coherent beam at small angles. The favorable requirements for temporal coherence in this geometry have allowed the use of broadband radiation provided by multilayer optics for a considerable advantage in flux. We demonstrated that the intensity in the speckle patterns is sensitive to the specific morphology within the illuminated region. The speckle patterns were observed to change from being nearly specular at small q_z values to exhibiting profound structure at larger q_z due to a progressive increase in surface sensitivity. An estimation of the roughness of the surface was obtained by realizing that strong interference occurs when the phase between different scattering regions approaches π.

At the present stage of development, we are limited to a field of view of a few hundred micrometers, but this will become much smaller with the less-grazing angles at larger q_z. This technique therefore can provide specific morphological information about surfaces on technologically relevant length scales nondestructively. Progress is currently being made on the reconstruction of the surface morphology directly from the speckle pattern,[11] which could render this technique a valuable imaging tool,[12] complimenting other techniques such as LEEM.[13] The biggest future potential is the possibility of studying surface fluctuations using x-ray photon correlation spectroscopy (XPCS).[2–4,7] Fluctuations on length scales already accessible by this method are known to be important during crystal growth[13] and these could be probed directly in the time domain. With the increase in coherent flux expected from undulator radiation at third-generation sources, this technique will be able to probe roughness on atomic length scales, and allow XPCS of surfaces in thermal equilibrium.[14,15]

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