

Direct observation of surface-trapped diffracted waves

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(Received 9 June 1989)

We have made the first direct observation of a diffracted x-ray beam that occurs only at an interface in the grazing-angle diffraction geometry. The beam, which is unable to propagate into the bulk of either component of the interface, was detected at the surface of a Ge crystal. A roughened area etched into the surface permitted phase matching to a beam in the vacuum. The diffracted beam that is observed to escape in this configuration shows a wave-vector dependence that cannot be qualitatively explained by purely kinematic models of scattering from rough surfaces.

X-ray diffraction has become an important means to study the structure of surfaces. The technique of determining physical roughening by examining the truncation rods has been widely used.^{1,2} Diffraction at grazing angles from two-dimensional overlayers has also been extensively developed.³⁻⁵ As currently used, these two techniques are limited to regions of phase space where kinematic models of diffraction are valid. The extension of these techniques to more complicated systems which include adsorbates with low scattering amplitudes or diffraction at an interface will require a complete dynamical treatment of the diffraction problem including the substrate interaction with the overlayer. One way of studying the problem is to investigate dynamical diffraction in the grazing-angle geometry in the presence of roughened surfaces. Alexandrov *et al.*⁶ have dealt with the problem for amorphous surfaces, and previous experiments by Golovin and Imamov⁷ observed some dynamical effects, but with limited angular resolution.

Previous experiments by Cowan *et al.*⁸ and Durbin and Gog⁹ have observed all the diffracted beams in the grazing-angle geometry which escape the crystal. The dynamical theory of grazing-angle diffraction (GAD) for x rays^{10,11} predicts the existence of a tantalizing condition where the diffracted beam exists at the interface between two media but has no propagating solution in either medium. Such a condition has been labeled a superficial wave after a somewhat analogous condition at optical wavelengths described by Fano.¹² The condition is not merely a mathematical curiosity, because the theory predicts that it is possible to couple an incident wave to this beam. It is not merely a physical curiosity either, because the x-ray field which is produced by this diffracted wave should be an important component in grazing-angle x-ray standing waves (GAXSW), which could be used to determine registration of atomic overlayers on surfaces with high accuracy.^{13,14} Fluorescence measurements have indirectly confirmed its existence.⁸ The remaining problem has been to observe this last beam directly. A direct observation of the superficial beam is important be-

cause real surfaces and interfaces are not perfectly abrupt, and the highly localized beam samples the actual interface down to nanometer-depth resolution.^{14,15}

We report an x-ray-diffraction experiment in the grazing-angle geometry which observes the superficial diffracted beam directly. The experiment used highly collimated 8-keV radiation from a synchrotron light source, and monitored the specularly reflected and reflected-diffracted beams which escaped from a germanium crystal. The orientation of the crystal with respect to the incident beam allowed the diffracted beam to have a wave vector at the surface of the crystal of magnitude greater than it would have externally, which prevented it from escaping. The crystal was specially prepared with a quadrant etched into part of its surface where the superficial beam thereby produced was permitted to escape.

The geometry of GAD is shown in Fig. 1 using a notation which has been described previously.¹⁴ Diffraction occurs from crystal planes normal to the surface. For an incident x-ray beam penetrating the crystal, only the component of the wave vector parallel to the surface is conserved.

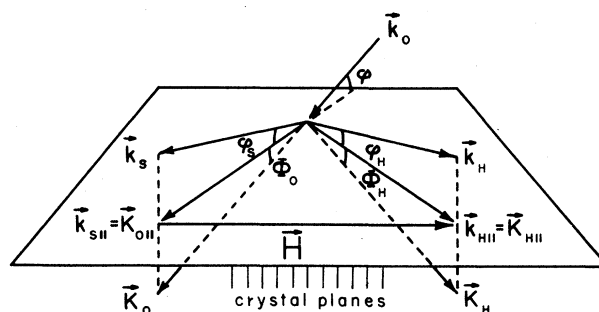


FIG. 1. Diagram showing the internally propagating and externally reflected beams possible in the GAD geometry, and their relationship.

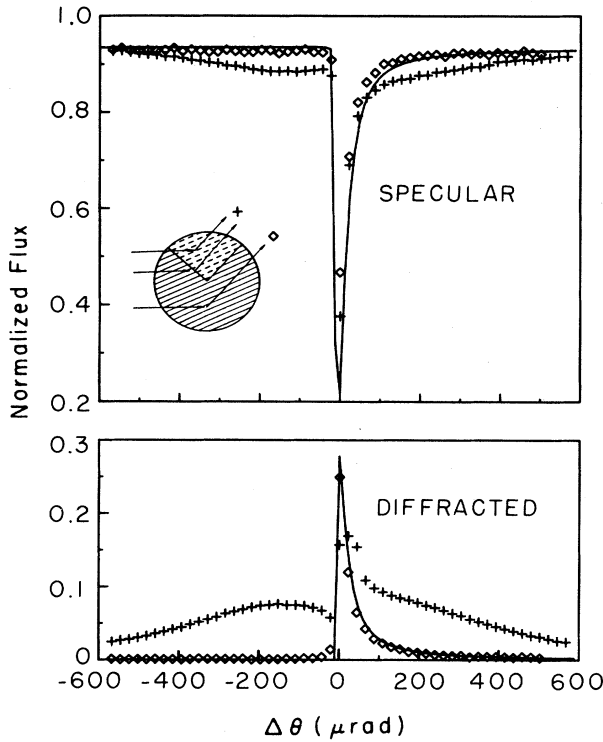


FIG. 3. Normalized flux of the specular beams (upper) and reflected-diffracted beams (lower) from the (111) surface of Ge at 8 keV. Fluxes are shown for $\varphi = 4.0$ mrad from the smooth surface (\diamond) and the etched surface (+) as a function of the deviation $\Delta\theta = \theta - \theta_{220}$. The solid lines are calculated fluxes for the diffracted and specular beams from a smooth surface. The diagram shows the orientation of the diffracting planes with respect to the step and the portion of the crystal surface which has been etched.

The specular flux from the smooth crystal surface (\diamond , upper) shows an appropriate dip at the diffraction condition because intensity from the specular beam is being redirected into an escaping diffracted beam.

From the etched surface, the superficial beam is visible as the diffracted beam (+, lower) which escapes for $\Delta\theta < \Delta\theta_c$. The angular dependence of the escaping superficial wave is at first glance unusual, since it does not just fill in asymmetry of the diffracted peak as observed from the smooth surface. It is interesting to note that added flux is also observed as a symmetrical shoulder on the diffracted beam in the region $\Delta\theta > 0$ where it was always permitted to escape. The Fourier components of surface roughness which supply crystal momentum q will also supply values $-q$ which will move allowed points on the grazing-angle dispersion surface¹⁴ into conditions of more favorable emission.

The specular flux which is observed from the etched surface (+, upper) shows an appropriate subtraction of flux at those angles where the diffracted beam is augmented. It might be argued that the specular beam flux observed from the flat surface should show a similar diminution of flux due to scattering into the superficial

diffracted beam, even if the superficial beam is only allowed to propagate along the crystal interface. However, the superficial beam has two channels of attenuation: absorption at the surface and rescattering into the specular beam which does escape. The latter process is proportional to the real part of susceptibility χ_{220} , while the former process is proportional to the much smaller imaginary part of χ_{220} . Thus the high reflectivity observed for $\Delta\theta < \Delta\theta_c$ from the smooth surface is not inconsistent with a strong diffracted-wave field at the interface.

The magnitude of momentum q_{\min} which needs to be taken up by the lattice in order to match the wave vector of the superficial wave at the interface to the wave vector in the vacuum is given by

$$\frac{q_{\min}}{k} = -\frac{\varphi^2}{2} + \Delta\theta \sin(2\theta_B).$$

The lattice is free to take up any momentum greater than q_{\min} that the roughness spectrum of the surface will allow. Consequently the flux of diffracted radiation in the region $\Delta\theta < \Delta\theta_c$ is proportional not to the spectral intensity of roughness but the integral of that spectral intensity from the highest q values down to q_{\min} .

We take as representative the momentum spectrum supplied by the crystal through an ideal step. The spectrum obtained by Fourier transforming the step is q^{-2} . We expect the roughness spectrum of the etched portion of the crystal to follow a similar power law with a different exponent. The scattering of the superficial wave into an escaping diffracted beam is assumed to be proportional to the spectral amplitude. The wave vectors and fields inside the crystal were calculated using dynamical diffraction theory. The wave vector due to each spectral component of surface roughness was coupled kinematically to the internal dynamical solutions and the resultant wave vector was used to match internal and external fields at the boundary of the crystal. A cutoff has been imposed on the maximum roughness wavelength to which the diffraction process should be sensitive: the coherence length of the x-ray beam. We estimate this length to be $3 \mu\text{m}$. The overall flux of the diffracted beam should be given by

$$F(\theta, \varphi) = A \int_{q_{\min}}^k dq \frac{e^{-q_c/q}}{q^2} f(q, \theta, \varphi),$$

where $q_c = 2\pi/L_{\text{coh}}$ is a low-frequency cutoff defined by the coherence length L_{coh} of the radiation used to sample the surface roughness, A is a constant, and $f(q, \theta, \varphi)$ is the flux due to each spectral component q for a given diffraction angle θ and angle of incidence φ .

Figure 4 shows a comparison between the observed flux diffracted from the etched surface or in the vicinity of the step (+) and the calculated flux diffracted from a surface with a q^{-2} spectrum of roughness (solid line). The calculated flux also includes a central unscattered component. The calculated flux has been normalized to be comparable to the integrated observed flux. The horizontal axis of Fig. 4 has also been calibrated in terms of q_{\min} .

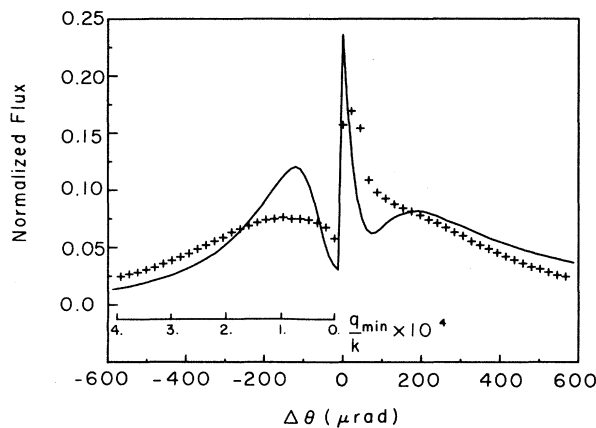


FIG. 4. A comparison of the diffracted flux observed (+) with the flux calculated (solid line) from a surface with a q^{-2} roughness spectrum (i.e., a step) with the cutoff described in the text. Both fluxes are plotted vs the deviation from θ_{220} described in Fig. 3. The separate horizontal axis labeled by the wave vector q_{\min} is the minimum tangential component which the surface roughness must supply in order for the superficial diffracted beam to escape.

The data show qualitative agreement with the calculated flux. The unexpected decrease of the superficial wave flux as $q_{\min} \rightarrow 0$ is seen to be due to the limit of coherence length of the x-ray beam, which cannot sample longer Fourier components of surface roughness. The mixed dynamic-kinematic model incorporated scattering from components of q only along the directions $\mathbf{K}_{0\parallel}$ and $\mathbf{K}_{H\parallel}$. Including components along other directions would

broaden the angular distribution of the scattered superficial beam.

While the mixed dynamic-kinematic model shows a qualitative agreement with the observed diffracted flux from the superficial wave, normalization of the calculated diffracted flux to the calculated specular flux gives a value for the diffracted beam which is an order of magnitude too small. Purely kinematic theories commonly applied to diffraction problems away from the diffraction peak^{1,2} are inadequate to explain this type of geometry since the amplitude of the enhancement is of the same order as the allowed diffraction peak itself. That and the proximity of the enhancement to the main peak will require a theory of roughness which takes multiple scattering into account (i.e., a dynamical theory).

In summary, we have observed directly a superficial diffracted x-ray beam, analogous to a surface plasmon, which exists only at the interface of a periodic medium. The wave is observed by means of a roughening of the surface, which supplies the additional missing momentum needed to match the wave vector of the diffracted beam trapped along the surface to one which can propagate in vacuum. The observation of an escaping superficial beam is in fact a highly sensitive measure of surface roughness on an atomic scale, whose wavelength is sampled to down to the coherence length of the radiation.

The authors would like to thank Dr. T. Vorburger and Mr. A. Hartman for sensitive stylus measurements of the sample surface. The experiment was conducted at the Cornell High Energy Synchrotron Source, which is supported by NSF Grant No. DMR-84-12465.

¹S. R. Andrews and R. A. Cowley, *J. Phys. C* **18**, 6427 (1985).

²I. K. Robinson, *Phys. Rev. B* **33**, 3830 (1986).

³W. C. Mara, P. Eisenberger and A. Y. Cho, *J. Appl. Phys.* **50**, 6927 (1979).

⁴P. Eisenberger and W. C. Mara, *Phys. Rev. Lett.* **46**, 1084 (1981).

⁵W. C. Mara, P. H. Fuoss, and P. E. Eisenberger, *Phys. Rev. Lett.* **49**, 1169 (1982).

⁶P. A. Alexandrov, A. M. Afanasiev, M. K. Melkonyan, and S. A. Stepanov, *Phys. Status Solidi A* **81**, 47 (1984).

⁷A. L. Golovin and R. M. Imamov, *Phys. Status Solidi A* **80**, K63 (1983).

⁸P. L. Cowan, S. Brennan, T. Jach, M. J. Bedzyk, and G. Materlik, *Phys. Rev. Lett.* **57**, 2399 (1986).

⁹S. M. Durbin and T. Gog, *Acta Cryst. A* **45**, 132 (1989).

¹⁰A. V. Andreev, *Usp. Fiz. Nauk* **145**, 113 (1985) [*Sov. Phys.—Usp.* **28**, 70 (1985)].

¹¹P. L. Cowan, *Phys. Rev. B* **32**, 5437 (1985).

¹²U. Fano, *J. Opt. Soc. Am.* **31**, 213 (1941).

¹³O. Sakata and H. Hashizume, Report of the Research Laboratory on Engineering Materials, Tokyo Institute of Technology (Japan) No. 12, 1987 (unpublished).

¹⁴T. Jach, P. L. Cowan, Q. Shen, and M. J. Bedzyk, *Phys. Rev. B* **39**, 5739 (1989).

¹⁵N. Bernhard, E. Burkel, G. Gompfer, H. Metzger, J. Peisl, H. Wagner, and G. Wallner, *Z. Phys. B* **69**, 304 (1987).

¹⁶B. M. Batterman and H. Cole, *Rev. Mod. Phys.* **36**, 681 (1964).