Polarimetry of Specular and Non-Multiple-Scattered Electromagnetic Radiation from Selectively Roughened Si Surfaces

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We show by application to Si that polarimetric measurements on radiation reflected nonspecularly from rough surfaces can distinguish between geometric-optics (facet or tangent-plane) and Rayleigh-Fano (random diffraction grating) scattering models, thus providing new details about surface roughness. A class of macroscopically rough but microscopically smooth surfaces is found that gives accurate specular ellipsometric data while being unsuitable for reflectance measurements.

Much recent theoretical[1-7] and experimental[7-14] work has been done on the reflection of electromagnetic radiation from rough surfaces, not only to gain a better understanding of scattering but also for application to solar-selective surfaces[15] and to the ellipsometry of nonideal surfaces[12] and chemical-vapor-deposited film growth[16] where minor roughness scattering makes reflectance measurements unreliable. Also, roughness is important in vacuum-ultraviolet and x-ray spectroscopy where short-wavelength synchrotron radiation is used, and has been suggested to be a contributing factor in enhancing Raman scattering from adsorbates on surfaces.[17] To date, nonspecular (scattered) radiation from rough surfaces has been extensively studied in the optical range by intensity measurements,[8-10] and specular radiation by fixed-wavelength ellipsometry.[11-14] If we presuppose that one of various possible models apply, the former method yields topographical information, but it cannot discriminate among models because of an unlimited number of adjustable parameters (e.g., statistical distributions of local slopes in physical-optics approximations[3,4] or Fourier roughness coefficients in random-diffraction-grating or Rayleigh-Fano theories[5]). Fixed-wavelength specular ellipsometric measurements on rough surfaces are generally interpreted by assuming a surface Garnett film[6] although these data are also insufficient to determine its properties completely.

Here we combine single-wavelength polarimetric measurements on radiation scattered from selectively roughened Si single-crystal samples with spectroscopic ellipsometric measurements from 1.5 to 5.8 eV to show that scattered-light polarimetry can distinguish between scattering models and thus determine new details about roughness present on a surface. Surprisingly, even though polarimetry data are intensity independent and therefore free of the adjustable-parameter ambiguities of intensity methods, to our knowledge no attempt has been made to measure the properties of the polarized component of scattered radiation from rough surfaces, although Azzam and Bashara have investigated ellipsometrically the intense diffracted orders from gratings[7] and Heitmann and Permien have compared intensity ratios to Rayleigh-Fano predictions for silver films.[10]

Measurements were made on selectively roughened surfaces prepared by etching Syton-polished $\langle 111\rangle$ surfaces of 860-$\Omega\cdot$cm n-type single-crystal Si wafers. Silicon was chosen because $\langle 111\rangle$-selective ($NaOH$-H_2O) and chemical polish (CP-4) etches are available.[18] These etchants preserve the perfection of the underlying lattice and thereby eliminate ambiguities arising from intrinsic optical-property differences or depolarizing subsurface multiple reflections[19] that result from distressed overlayers left by mechanical abrasion. Further, ϵ_2 is small enough at 6328 Å to be neglected in model computations of polarization dependence but large enough to suppress unwanted back reflections in the sample. Finally, the 20-Å natural oxide of Si is not a significant factor in the measurements, and data can be taken under ambient conditions.

Polarimetry and intensity data for scattered radiation were obtained using a Gaertner model L119 manual null ellipsometer with a 2-mW He-Ne laser source. The light was incident at 70° and was chopped at 210 Hz to facilitate stray-light rejection and the determination of null using phase-sensitive detection. Intensity was measured to within 20% over 6 orders of magnitude with the aid of neutral-density filters. For polarimetry measurements, accuracy was of the order of 0.1° for the specular beam and decreased as a result of decreasing intensity off specular. Measurements were terminated when uncertainties reached ± 5°. Spectroscopic ellipsometric data were obtained with the rotating-analyzer instrument described previously.[20]

Although data were taken on other surfaces as well, we discuss here in detail only the results obtained on three representative ⟨111⟩ surfaces: an original Syton-polished surface of high optical quality, an etched CP-4 surface of apparent high quality but with a well-developed orange-peel finish, and a saw-cut macroscopically rough back surface etched in NaOH sufficiently long to remove microscopic damage. The latter surface showed a roughness-induced 30% reflectance loss in an $f/7$ 6328-Å reflectometer and thus was completely unsuitable for reflectance measurements.

Figure 1 shows the relative intensity of scattered light as a function of observation angle for these three surfaces. To avoid possible ambiguities in interpretation that might arise from local variations in roughness over the surfaces, these intensity data were obtained under the same conditions as the polarimetry data given in Fig. 2. The Syton and CP-4 surfaces are both macroscopically smooth showing an intensity drop of about 6 orders of magnitude by 4° scattering angle. By contrast, the scattering intensity for the macroscopically rough NaOH surface decreased only 2 orders of magnitude by 4°. Beckmann[3] showed in the geometric-optics approximation that the decrease of intensity for a normally distributed random rough surface would be quadratic on a logarithmic plot; our samples show a linear decrease which may be interpreted as exponential, rather than Gaussian, statistics. A quadratic remnant near 2° can be seen on the Syton, and weakly on the NaOH, surface data, showing the presence of two types of statistical roughness on these surfaces. This remnant is not equivalent to the much broader structure seen on very rough metal surfaces by Beaglehole and Hunderi,[9]

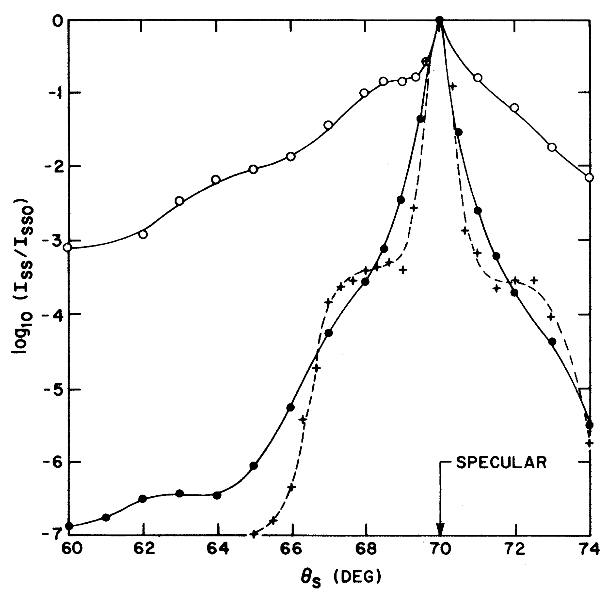

FIG. 1. Scattered light intensity versus observation angle for s-polarized light ($A = P = 90°$) for Si surfaces: crosses, Syton polished; unfilled circles, NaOH etched; and filled circles, CP-4 etched. Specular reflection occurs at 70°. All data are normalized to the specular intensity.

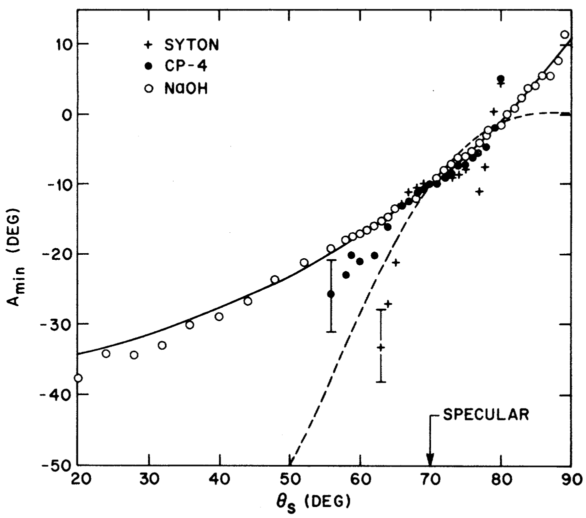

FIG. 2. Analyzer null azimuth versus observation angle for surfaces of Fig. 1 for incident 70° light linearly polarized at $P = -45°$. Geometric-optics (solid line) and Rayleigh-Fano (dashed line) model calculations are also shown.

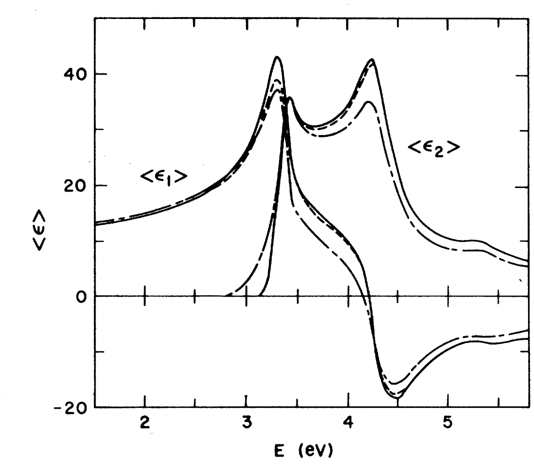

FIG. 3. Pseudo-dielectric-function data $\langle\epsilon\rangle = \langle\epsilon_1\rangle + i\langle\epsilon_2\rangle$ for surfaces of Fig. 1 from specular-component ellipsometry: solid line, Syton and NaOH; long-dashed line, CP-4. Also shown (short-dashed line) is a spectrum obtained by etching a CP-4 surface for 1 min in NaOH.

which arises from angular prefactors to the exponential statistical factor.

We consider next the polarimetric data for these surfaces shown in Fig. 2, taken at an illumination angle $\theta_i = 70°$ and a variable scattering angle θ_s, both defined relative to the average surface normal. Over the ranges shown the scattered radiation was almost completely plane polarized, showing that multiple-scattering effects were negligible.[4,19] Thus we show only the azimuthal angle for extinction for incident radiation linearly polarized at $P = -45°$ with respect to the plane of incidence. Also shown are model calculations for the microscopically smooth geometric-optics (facet or tangent plane) and microscopically rough Rayleigh-Fano (random diffraction grating) approximations, for which, respectively,

$$\tan(A_{\min}{}^{GO}) = \frac{[\epsilon \cos\bar{\theta} - n_\perp(\bar{\theta})][n_\perp(\bar{\theta}) + \cos\bar{\theta}]}{[\epsilon \cos\bar{\theta} + n_\perp(\bar{\theta})][n_\perp(\bar{\theta}) - \cos\bar{\theta}]} \cot P, \tag{1a}$$

$$\tan(A_{\min}{}^{RF}) = \frac{[n_\perp(\theta_s) + \cos\theta_s][n_\perp(\theta_i) + \cos\theta_i][\epsilon \sin\theta_s \sin\theta_i - n_\perp(\theta_s) n_\perp(\theta_i)]}{[n_\perp(\theta_s) + \epsilon \cos\theta_s][n_\perp(\theta_i) + \epsilon \cos\theta_i]} \tan\theta_i \cot\theta_s \cot P, \tag{1b}$$

where $n_\perp(\theta) = (\epsilon - \sin^2\theta)^{1/2}$, $\bar{\theta} = (\theta_i + \theta_s)/2$, and ϵ is presumed to be real. Equation (1b) has not been given previously but may be derived easily from intermediate results given in Ref. 5. For Si at 6328 Å, $\epsilon = 14.87 + i0.22 \simeq 14.87$.

The CP-4 and Syton surfaces show characteristics of both models, while the NaOH surface clearly follows the microscopically smooth tangent-plane model, but not the Rayleigh-Fano theory, over a wide range of scattering angles. This implies that the apparently rough NaOH surface actually consists of relatively flat, locally smooth facets and thus despite appearances could yield good specular ellipsometric data.

The results of such spectroscopic specular ellipsometric measurements are shown in Fig. 3. We found that the spectra of the pseudo dielectric function, $\langle\epsilon\rangle$ (data analyzed ignoring possible surface films), for the Syton and apparently rough NaOH sufaces were identical to within 2% of the maximum ϵ_2 value, while the CP-4 spectrum was reduced. The reason for the *a priori* surprising Syton-NaOH agreement is, of course, clear from Fig. 2: The preferential $\langle 111\rangle$ NaOH etch cuts planar facets on top of the originally rough $\langle 111\rangle$ Si surface and the high f number of ellipsometer selects only radiation specularly reflected from these facets. The CP-4 data are characteristic of coverage by a dielectric film of effective thickness of the order of 15 Å, i.e., a rough Garnett film.[6] To test the facet-etching model further, we treated a similar CP-4 surface in NaOH-H_2O. After 1 min, considerable microscopic smoothing is already evident as seen from the data shown in Fig. 3. After 3 min, the original Syton data were recovered, showing that the face was microscopically smoothened (though macroscopically roughed) by the preferential $\langle 111\rangle$ action of the NaOH-H_2O etch. Finally, stylus measurements showed rms microscopic roughness over 10–50-μm widths of about 20, >100, and 10 Å for the Syton, CP-4, and NaOH surfaces, respectively, in good qualitative agreement with the above conclusions.

The identification of a type of macroscopically rough but microscopically smooth surface suggests that accurate ellipsometric data may be taken on surfaces cleaned by the standard ultrahigh-vacuum technique of Ar bombarding and annealing, a method that typically yields macroscopically rough surfaces, provided that a low-index surface orientation is chosen to allow facets to form during annealing. Published fixed-wavelength data on Si are inconclusive,[11,13] but preliminary spectroscopic ellipsometric results on Ar-bombarded and annealed clean $\langle 111\rangle$ Ge surfaces show that this behavior is indeed obtained.[21] Finally, the possibility of progressive microscopic smoothing by selective etching should provide convenient systems for the study of rough-surface models by both polarimetry of nonspecular radiation and spectroscopic ellipsometry of the specular component as shown here. Further work in progress and full details will be published elsewhere.

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Application of Coherent-Potential Approximation to Disordered Muffin-Tin Alloys

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I report implementation of the coherent-potential approximation for obtaining the electronic density of states and component charge densities in disordered muffin-tin alloys. Illustrative results for Cu_xNi_{1-x} are presented. The extent to which the self-consistency in treating disorder influences the electronic spectrum is considered.

It has become clear in the recent years that, in order to obtain a realistic description of the electronic spectrum of disordered transition- and noble-metal alloys, the atomic potentials must be treated within the framework of the muffin-tin model, as is usually done for the corresponding perfect crystals, and that the simple one- and two-band tight-binding model Hamiltonians are not adequate in these cases.[1] In this connection, two of the most commonly used approximations have been the coherent-potential approximation (CPA) and the average t-matrix approximation (ATA). Of the two, the CPA treats the disorder self-consistently and is to be preferred. The attractiveness of the ATA derives from its relative simplicity in application to realistic models.[2,3] In spite of the significant progress made with regard to the application of these two approximations to the muffin-tin Hamiltonian, difficulties have persisted with each of the schemes. The CPA formalism is well developed,[1,4] but its practical implementation to the muffin-tin Hamiltonian has not been possible because of the difficult and repeated Brillouin-zone integrations necessary to solve the CPA self-consistency equation.[5] By contrast, the difficulties with the ATA are formal in nature: Although the currently used ATA expression for the average density of states $\langle\rho(E)\rangle$ gives reasonable results in all instances studied so far, the corresponding expressions for the component densities $\langle\rho_{A(B)}(E)\rangle$ (i.e., the electronic charge densities associated

