Surface microtopography

How flat is a cleaved crystal surface? Some instruments now available will give you a picture of it, complete with all its faults, down to single-atom steps.

Russell D. Young

It is difficult to imagine a scientist or engineer who has not become painfully aware of the importance of metal surfaces in today's technological world. Indeed, most of the work in surface science has been initiated to explain puzzling surface problems encountered in other fields of science and engineering. An important aspect of this problem has involved the determination of the detailed topography of metal surfaces.

In the last several years there has been considerable pressure to improve the resolution of instruments that measure surface topography. Surface scientists want to know the density of single- or multiple-atom steps on their carefully prepared single-crystal surfaces. Elaborate polishing, cleaning and smoothing techniques cannot replace a detailed knowledge of the actual surface topography.

In the field of thin-film devices, manufacturers have progressively reduced the size of their electronic elements to the point where one can anticipate devices employing single layers of atoms or molecules. But even today, with films in the 10–100-Å range, the device industry experiences an unacceptably high element failure rate due to surface imperfections, such as stacking faults, voids, multiple atom steps, and so on. Challenges have also appeared in the field of metallography where the details of cleavage steps, the nature of the surface contour near grain boundaries and the modification of surface topography with etching and corrosion have placed new demands on high-resolution topographic mapping.

My discussion here will be limited to metallic surfaces and to instruments that give a pictorial representation of the surface. These devices are the field-ion microscope, the transmission electron microscope, the scanning electron microscope, the optical microscope, the traditional contacting stylus instrument, and a new instrument called the "topografiner." These are all familiar techniques except the last; the topografiner, currently being developed at the National Bureau of Standards, uses a noncontacting field-emission probe to measure surface topography. Figure 1 is a map of a 180 line-per-mm diffraction-grating replica obtained with this device.

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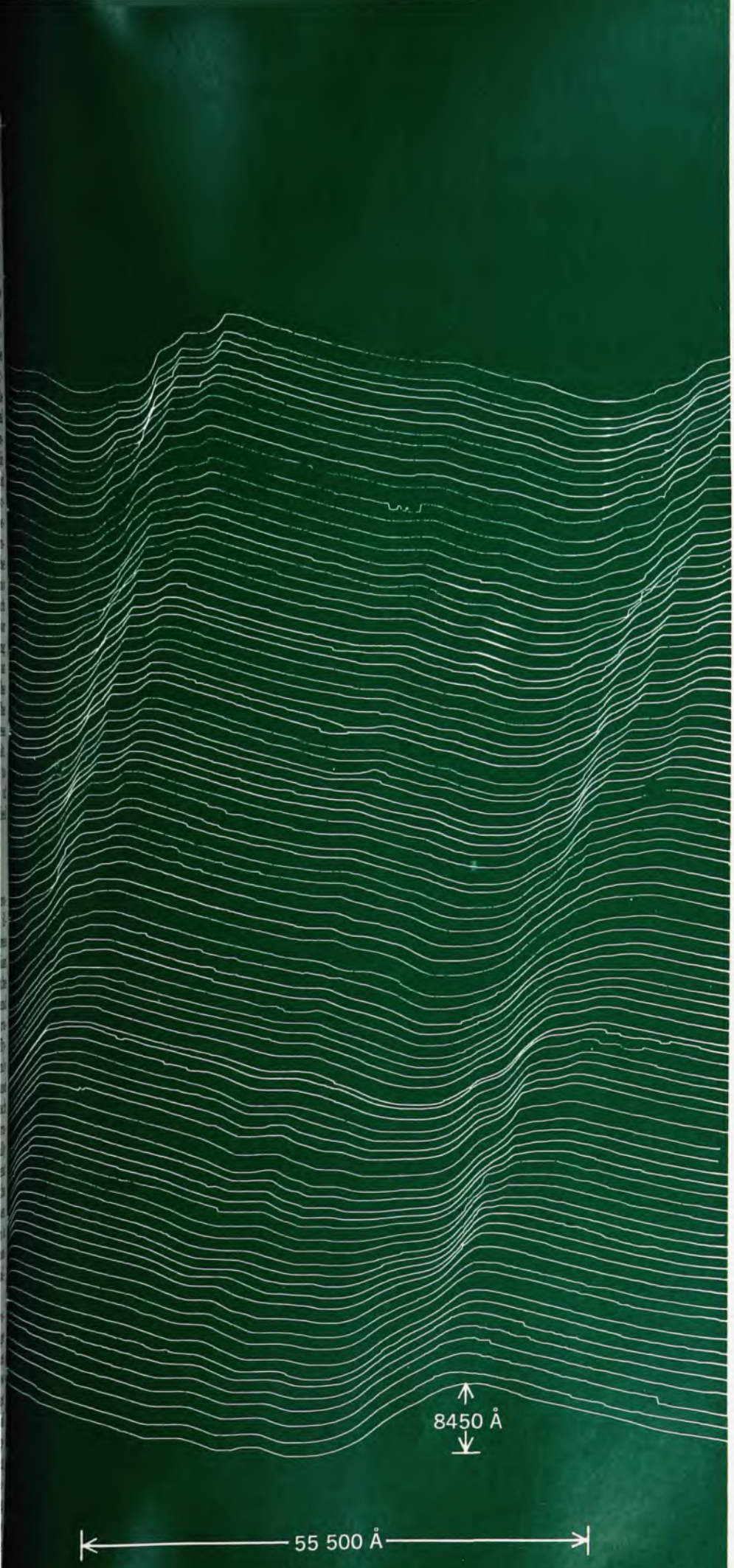
The field-ion microscope developed by Erwin W. Müller[1] is in a class by itself, enabling as it does the simultaneous preparation of a variety of atomically clean single-crystal planes. In conjunction with field electron emission, the field-ion microscope has been used successfully to study the diffusion of single adsorbed atoms on single-crystal planes,[2] chemisorption,[2] resonant tunneling of electrons through electronic energy levels of adsorbed atoms and molecules,[3] measurement of work function,[4] condensation[5] and other surface-science studies. However, like most instruments, it has limitations. The single-crystal planes are typically no more than 100 Å in diameter, which makes many experiments difficult or impossible. The presence of strong electrostatic fields perturbs processes at the surface and thus strongly limits the type of adsorbate and substrate. The inherent electron tunneling process often makes interpretation difficult. For the present purposes I would like to focus attention on characterization of larger surfaces than can be studied with the field-ion microscope.

Transmission electron microscope

The transmission electron microscope, like the optical microscope, illuminates the whole specimen area simultaneously and employs Gaussian optics in generating its image. With the exception of these two microscopes (and the field-ion microscope) the instruments I will discuss here are of the flying-spot variety; they illuminate only one spot on the specimen at a time and form their images sequentially. Each type has special advantages. Instruments with Gaussian optics generally have higher resolution than equivalent flying-spot instruments and spread the illumination over the whole specimen surface rather than concentrating it in a high power-density spot. Flying-spot instruments permit point-by-point analysis of surface properties.

In the transmission electron microscope[6] the electrons that form the image must pass through the specimen, thus limiting the specimen thickness to a few thousand angstroms. To study surface topography one must make a replica of the surface—for example, a carbon replica may be made by vacuum depositing a 100–1000-Å carbon film on the specimen surface, then carefully removing the film and mounting it for use with the microscope.[7] The image obtained from such a replica contains a representation

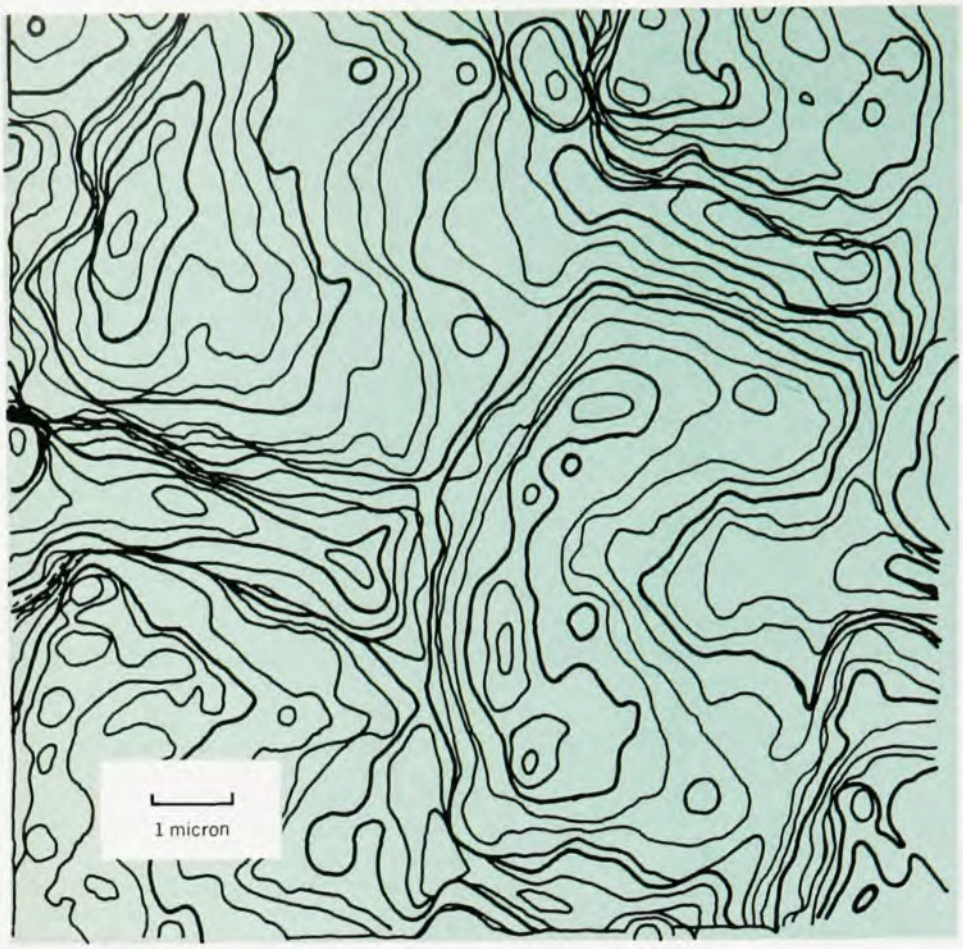


Topographic map of a 180-line-per-mm diffraction-grating replica, obtained with the "topografiner," a noncontacting field-emission probe developed at NBS. Figure 1

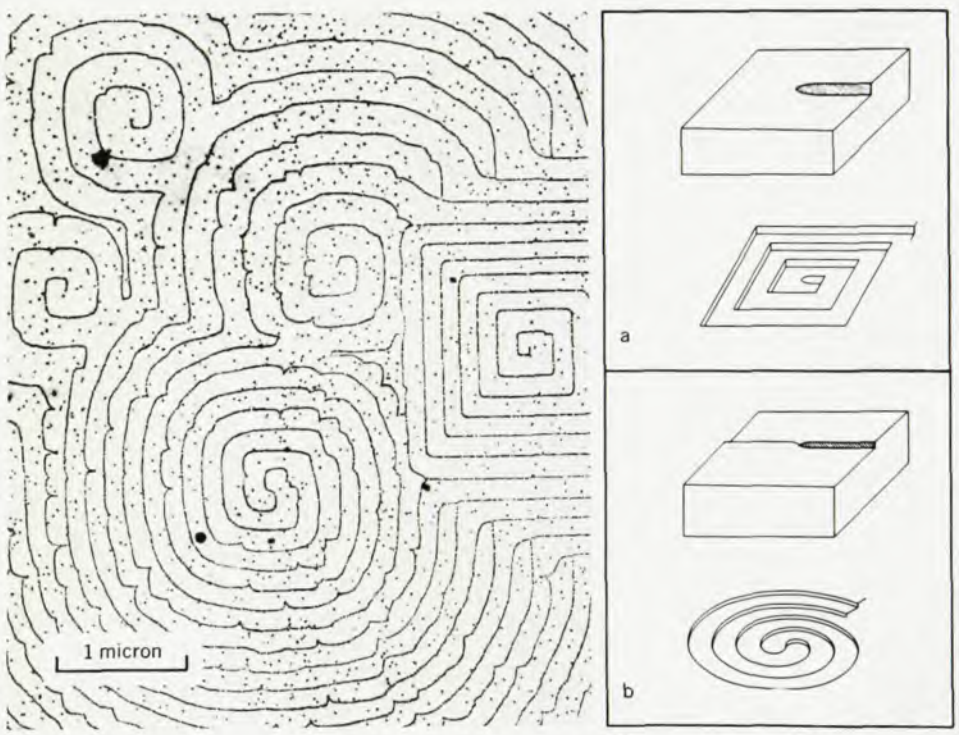
of the surface topography, but it is difficult to interpret. To obtain quantitative information about the specimen one must take two micrographs, the specimen being tilted through an angle, say 8 deg, between the two exposures. These micrographs may then be examined in a stereoscopic viewer to obtain a good qualitative representation of the surface profile. For quantitative work (photogrammetry) surface profiles can be measured and contour maps drawn with the aid of a measuring stereoscope, called a "stereometer." This procedure, however, is tedious and usually performed only with reluctance. Consider, for example, the number of datum points needed to produce a carefully measured contour map such as that of developing dental enamel[7] shown in figure 2. In this example the contour spacing is 1500 Å—close to the present limit of elevation resolution with the stereo technique. It appears unlikely that this resolution can be improved much beyond 100 Å, because of inherent limitations of the replicating technique and the measurement of tilt angle.

In 1958 George A. Bassett[8] developed a very specialized method for observing single or multiple-atom steps that are at least 30 Å apart with the transmission electron microscope. He discovered that if a monolayer or so of gold was evaporated onto a cleaved sodium-chloride crystal, the gold atoms formed nuclei, which collected along the edges of steps on the crystal surface. When a carbon replica was subsequently deposited, the gold nuclei were incorporated into the carbon replica and appeared as step decorations in the microscope image. Thus the technique was named the "gold decoration" technique. Without the gold decorations the steps do not provide sufficient contrast to be seen. Bassett noted that when slip steps crossed cleavage steps in the decorated patterns they were deflected by an amount that gave the total height of the cleavage steps. He then counted the number of decorated steps and showed that most of these must be single-atom steps, 2.8 Å high. A simple and convincing demonstration of the power of this technique was given by Heinz Bethge[9] who evaporated sodium-chloride crystals in vacuum to obtain broad single and double atom steps, as shown in figure 3.

Bethge has extended the technique to silver and copper crystals formed from molten droplets that were subsequently evaporated just below the melting tem-



Contour map of developing dental enamel, made with a pair of transmission electron microscope photographs examined under a measuring stereoscope; contour lines are drawn point-by-point from these measurements. Contour spacing is 1500 Å. (From reference 7.)
Figure 2



Atomic steps on a cleaved and evaporated sodium-chloride crystal, revealed by a transmission electron microscope by the gold-decoration–carbon-replication technique. Screw dislocations can lead to round or square spirals; single steps forming the edges of round spirals (b) combine in pairs to form the double steps at the edges of square spirals (a). (From reference 9.)
Figure 3

perature.[10] An examination of figure 4 shows several single-crystal regions larger than 1000 Å. J. G. Allpress and John V. Sanders have prepared larger silver planes by thermally etching single-crystal silver films in air.[11] Figure 5 shows a (111) silver surface that is traversed by a narrow twin with its surface etched to a (100) plane. Single-crystal areas many thousand angstroms in size are present with only a few slip steps. Smaller regions larger than 1000 Å are present without slip steps. It is very probable that particular planes of other metals can be similarly prepared.

The trend in surface science, as I mentioned earlier, is to single-crystal surfaces—or what their owners *believe* to be single-crystal surfaces. Techniques such as low-energy electron diffraction (LEED), secondary emission, Auger emission and appearance-potential studies probe the surface with electron beams. By careful electron optical design these beams could be made smaller than the single-crystal surfaces of copper and silver that are presently available and could thus be used to carry out studies of "idealized" surfaces. It is important to note that the gold decoration technique, including specimen preparation, gold deposition and carbon replication, can be carried out in ultra-high vacuum—that is, with pressures lower than 10^{-9} torr.

Scanning electron microscope

The scanning electron microscope is a flying-spot type of instrument with resolution intermediate between that of the transmission electron microscope and the optical microscope. I exclude here the high-resolution transmission type of scanning electron microscope developed by Albert Crewe because the reflection mode must be used in the direct study of metallic surfaces.

In the typical scanning electron microscope a beam from an electron gun is focussed to a spot about 100 Å diameter on the specimen. Secondary electrons, scattered electrons, photons or x rays from the specimen can be detected, amplified, and the resulting signal used to modulate the brightness of a cathode-ray tube display system. The electron beam is scanned across the specimen as a raster synchronous with the cathode-ray-tube scan. As the beam only strikes one spot of the surface at a time it is possible to use x rays and photons to obtain chemical information at *each* point on the surface.

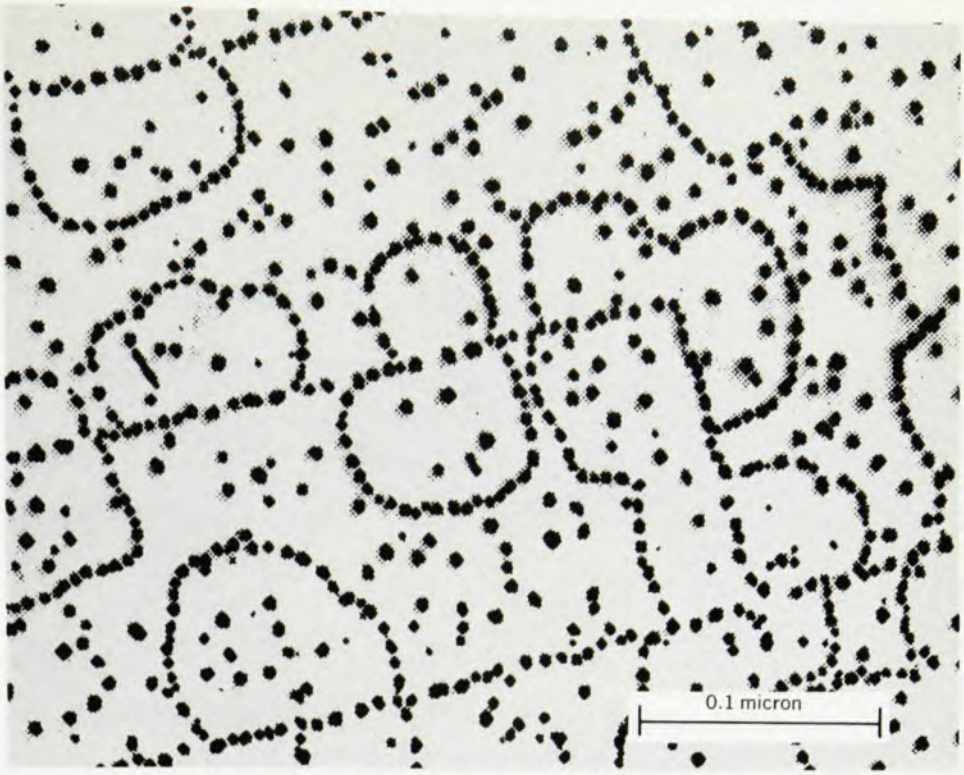
When used in the reflection mode the scanning electron microscope has a resolution determined by the way in which the electron beam is scattered by the specimen. The penetration of the incident electron beam into the specimen is called the "bloom" and is shown in figure 6.[12] Secondary electrons travel only short distances in a solid; so the

secondaries that reach the detector originate from a 50–100 Å volume near the surface. Decreasing the size of the electron beam below 50 Å has little effect on the resolution, which is usually limited to 100 Å. Some limited experiments can be carried out while observing the surface, but at present the deposition on the specimen of organic materials from the vacuum pumps severely limits the type and quality of these *in situ* experiments.

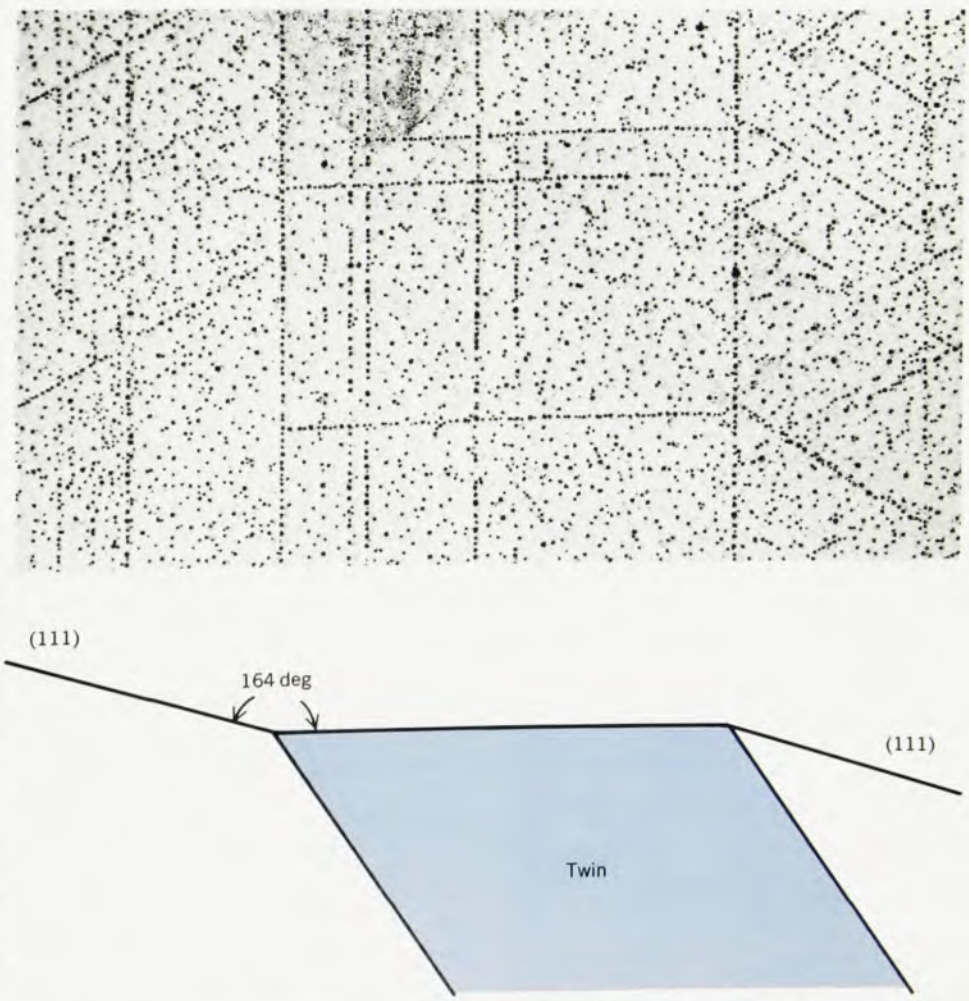
Contrast in the scanning electron microscope is affected by many factors. The number of secondary electrons depends strongly on the tilt of the region being probed and even more strongly on the presence of sharp edges. Electrical fields at the specimen surface, arising from the potential of adjacent electrodes, enhance emission at protrusions. The efficiency of secondary-electron production depends on the particular element present where the beam strikes the surface, the crystallographic surfaces exposed, the work function and the presence of adsorbed layers. The contrast gradient depends on the gain of the amplifier. For all these reasons one can be very badly misled in interpreting edges and bright and dark areas as topographic information. Stereoscopic pairs are essential, even for good qualitative interpretation of micrographs. As in the case of the transmission electron microscope, the scanning microscope is presently limited to about a thousand angstroms in elevation resolution with photogrammetry and stereo pairs. Thus there is no hope of seeing single-atom steps. Nonetheless, it is probably the most convenient and the most frequently used tool to examine the topography of surface-science specimens. Figure 7, a micrograph of a lightly electropolished copper-nickel-steel sandwich section, illustrates the ability of the scanning electron microscope to reveal surface texture.

Interference microscope

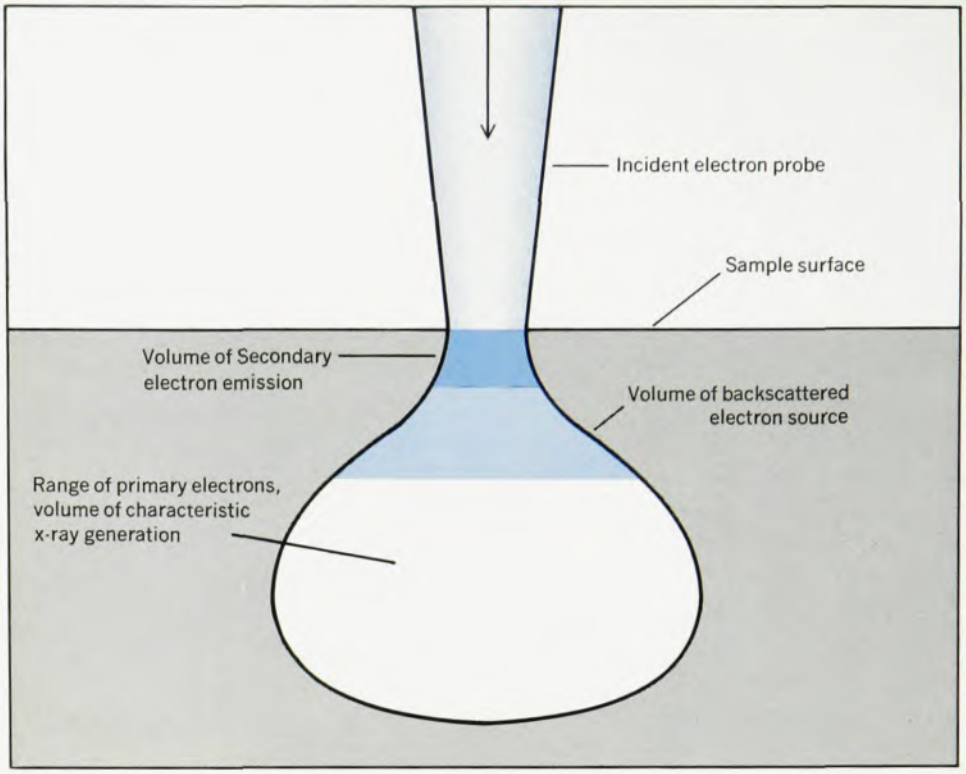
Among the various types of optical microscopes the interference microscope is the one most frequently used to obtain quantitative information about surface topography. A review of interference microscopes has been given recently by M. Francon.[13] Although the multiple-beam interference microscope developed by Samuel Tolansky[14] is much less popular than the two-beam type, I will limit my discussion to this instrument because of its extreme sensitivity, elegance and low cost. The simplest demonstration of the interference microscope is the familiar Newton's rings experiment where a flat glass plate is placed on a convex lens and viewed through a low-power microscope. Coherent light beams, reflected from the lens surface and from a point just above on the flat, interfere to form a series of bright and



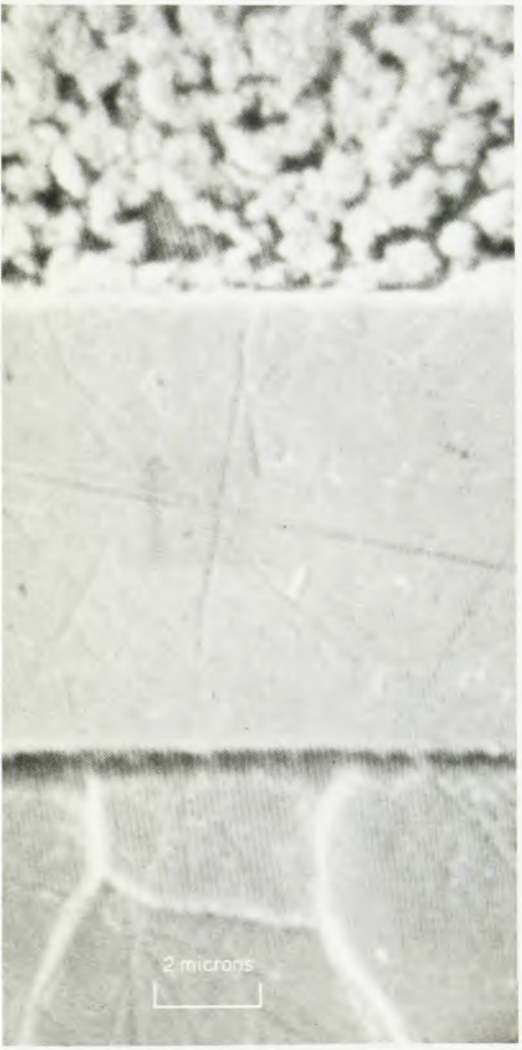
Evaporation structure and slip lines on a single crystal of silver, shown in a transmission electron microscope image after gold decoration and carbon replication. Note several single-crystal planes larger than 1000 Å. (From reference 10.)
Figure 4



Slip steps on a (111) silver surface traversed by a (100) plane. Note the large single-crystal areas free of slip steps. (From reference 11.)
Figure 5



"Bloom," or penetration of electron beam (in a scanning electron microscope) into the specimen. Secondary electrons originate in a 50–100 Å volume near the surface; backscattered electrons and x rays come from larger, deeper regions. Secondary electrons give the best resolution. (From reference 12.)
Figure 6



Copper-nickel-steel sandwich, shown in a scanning-electron micrograph. The sandwich was sectioned, highly polished, and lightly etched with nitric acid. The copper surface (above) was roughened by the etch; grain boundaries were exposed on the stainless-steel surface (below), and the nickel (center) retained the fine polishing scratches. (From David Ballard, NBS.)
Figure 7

dark rings. Starting from the point of contact between the flat and the lens a ring occurs each time the surfaces are separated by a multiple of half the light wavelength.

In the multiple-beam interferometer (figure 8) the lower surface of the flat glass plate is coated with a layer of silver, which may be up to 97% reflecting. If the convex lens in the Newton's rings experiment is replaced by the metallic surface to be studied, then the light reflects between the surface and the silver film a great many times before passing back through the silver film. These multiple reflections cause the fringes to sharpen dramatically. For a highly reflecting surface and a 97%-reflecting silver film the fringe width may be as small as 1% of the fringe separation.

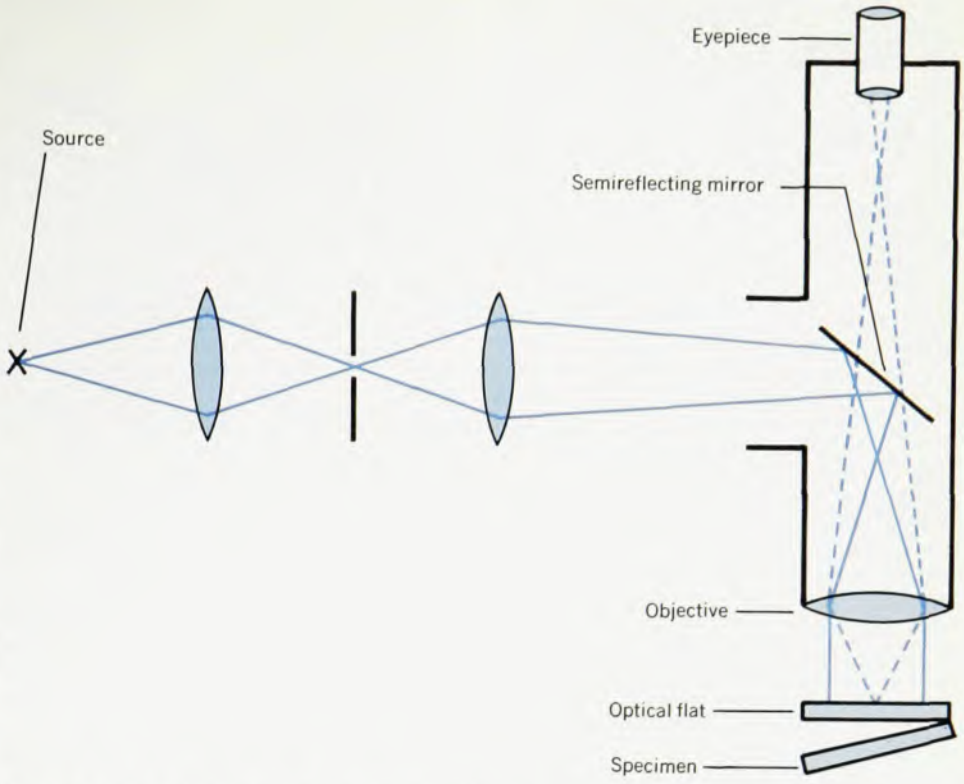
For extremely sharp fringes the resolution in the plane of the surface is only about 5 wavelengths because the multiply reflecting beam is slightly displaced sideways at each of the 50–100 reflections. On the other hand the elevation sensitivity is so good that steps in the surface topography as small as 5 Å can be resolved. Even greater resolution can be attained with the interference-contrast technique, where the flat is arranged so that it is parallel to the surface. Under these conditions a single dark fringe can be made to cover the whole field of view, and variations in light intensity across the fringe make it practical to detect steps of 1.5 Å, considerably less than the single-atom step height. An entirely different technique developed by James Dyson[15] combines the interference microscope with polarimetry to measure steps with a precision smaller than 1 Å.

The power of the Tolansky technique has been recognized and exploited by those who study crystal growth and imperfections but has been much neglected by those who work in surface science. Tolansky himself has studied the nature of several surfaces of single-crystal metals using the techniques just discussed; figure 9 shows, for example, the fringe pattern resulting from a slip band in a tin single crystal. The surface profile along any direction on the surface can be obtained by drawing an imaginary line on the micrograph in the region of interest. As one moves along the line the surface rises (or falls) about 2500 Å (depending on the wavelength of the illumination) for each fringe encountered. By careful interpolation between fringes it is possible to plot the profile with considerable accuracy. Tolansky has discussed[14] the details of such measurements, including methods for distinguishing between a hill and a valley, an upgrade or a downgrade, and so on. The presence of small steps on the surface can be detected by holding a straightedge along a sharp fringe line and noting where the line is suddenly

displaced sideways by a small amount. The ratio of the displacement to the distance between fringes is then multiplied by half the wavelength to give the step height. Tolansky reports that 5-Å steps have been repeatedly resolved on smooth surfaces. This technique is frequently used to measure the thickness of evaporated thin films.

Figure 10 shows the effect of electropolishing on the surface of a nickel single crystal. Electropolishing is frequently used in preparing surface-science specimens, so examination of these surfaces with an interference microscope should be profitable.

Figure 11 shows the cleavage structure of a bismuth single crystal. The corrugated zig-zag structure consists of fairly flat surfaces meeting at an angle. The angle is very highly magnified, because the slope of the fringe line depends on the magnification of the microscope in the horizontal direction and the fringe count in the vertical direction. For example, at a microscope magnification of 100 ×, a 45-deg fringe angle in a micrograph represents an angular magnification of 280 ×, and we see that the angle between the flat surfaces in figure 11 is considerably less than one degree. There is also a lesson to be learned here regarding the belief that "a cleaved surface is an ideal surface."



Multiple-beam interference microscope. Monochromatic light from the source illuminates a slit focused at the back focal point of the objective. Parallel light is transmitted through the silver film on the lower surface of the optical flat to the specimen surface. After repeated reflection a fraction of this light returns through the objective to the eyepiece. The resulting fringes can be pictured as arising from the intersection of the surface and a set of parallel lines spaced a half wavelength apart. (From reference 14.)
Figure 8

Contacting-stylus instruments

The recognized engineering standard for surface-profile measurement is the stylus instrument[16] known by various trade names such as "Tallystep," "Micro-topographer," "Dektak," and so on. These instruments employ a sensor head similar to a high-fidelity-phonograph pick up. A fine diamond stylus is coupled to a transducer whose amplified output is fed to a chart recorder. The transducer is moved parallel to the surface so that the stylus contacts the surface very lightly, sometimes with a force as small as 5 milligrams. The transducer may also be arranged to scan a series of closely spaced profiles that can be displayed on an X–Y recorder to obtain a pictorial representation of the surface. Stylus instruments range in elevation sensitivity from 25 to 250 Å with a horizontal resolution between 1 and 10 microns. This type of instrument will reveal considerable detail when used to examine the surfaces typically employed in surface-science studies, but unfortunately the stylus compresses a shallow trench as it slides across the surface, obliterating some of the original surface structure.

The topografiner

A group at the National Bureau of Standards is developing a noncontacting field-emission probe to measure surface topography with elevation resolution in the 1-Å range and horizontal resolution in the 100-Å range.[17] This is my own program, so you should be forewarned that unwarranted optimism may creep in when I compare this instrument with the others.

The basic principle of the topografiner is shown in figure 12. All motion in the instrument except that for coarse adjustments is generated by applying voltages to piezoelectric ceramic elements, which increase in length in proportion to the applied voltage. A sharply pointed emitter is scanned above the specimen surface by two piezoelectric drivers, X and Y. A constant current is passed through the field-emission point, establishing a fixed field at the emitter surface. Because the specimen (or anode) is nearby, once the emitter-to-specimen distance is fixed the voltage between the emitter and specimen is prescribed by Laplace's equation. In actual operation the emitter-to-specimen voltage is measured and fed to a servo system that applies the proper voltage to the Z-piezo driver to keep the emitter-to-specimen voltage constant, thus keeping the emitter-to-specimen spacing constant. The piezo voltage then corresponds directly to the emitter position and can be used to read out the profile.

Because field-emitted electrons travel from the tungsten emitter to the

Typical Resolutions of Surface-microtopographical Instruments

Instrument	Approx. vertical resolution (Ångstroms)	Approx. horizontal resolution (Ångstroms)	Topographic resolution (horiz. × vertical) (square Ångstroms)
Transmission electron microscope	1500	50	75 000
Scanning electron microscope	1500	100	150 000
Optical interference	5	25 000	125 000
Stylus instrument	25	10 000	250 000
Topografiner	30	4 000	120 000

Slip band on a crystal of cast tin, revealed by the multiple-beam interferometer. The soft tin surface was unpolished. Local fringe broadening in several areas is due to surface imperfections. *(From reference 14.)*
Figure 9

Electropolished nickel single crystal photographed by multiple-beam interferometry. Note the deep etch pits and surface roughening produced by the electropolishing process. (From reference 14.)
Figure 10

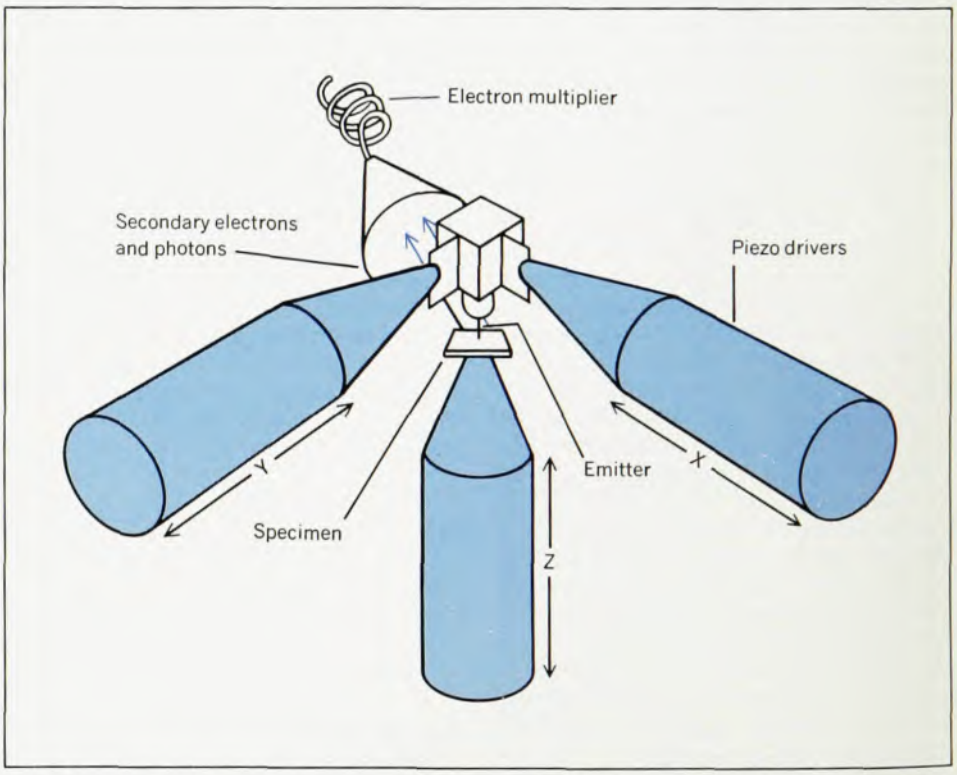
Bismuth single crystal. This interferometric micrograph of a cleavage face shows fairly flat surfaces meeting at an angle; the angle is highly magnified by the interference process and in this example is much less than one degree. (From reference 14.)
Figure 11

specimen it is necessary to provide a vacuum in the vicinity of the emitter and the specimen. At present we need a vacuum of about 10^{-9} torr, but new emitter materials should reduce this requirement.

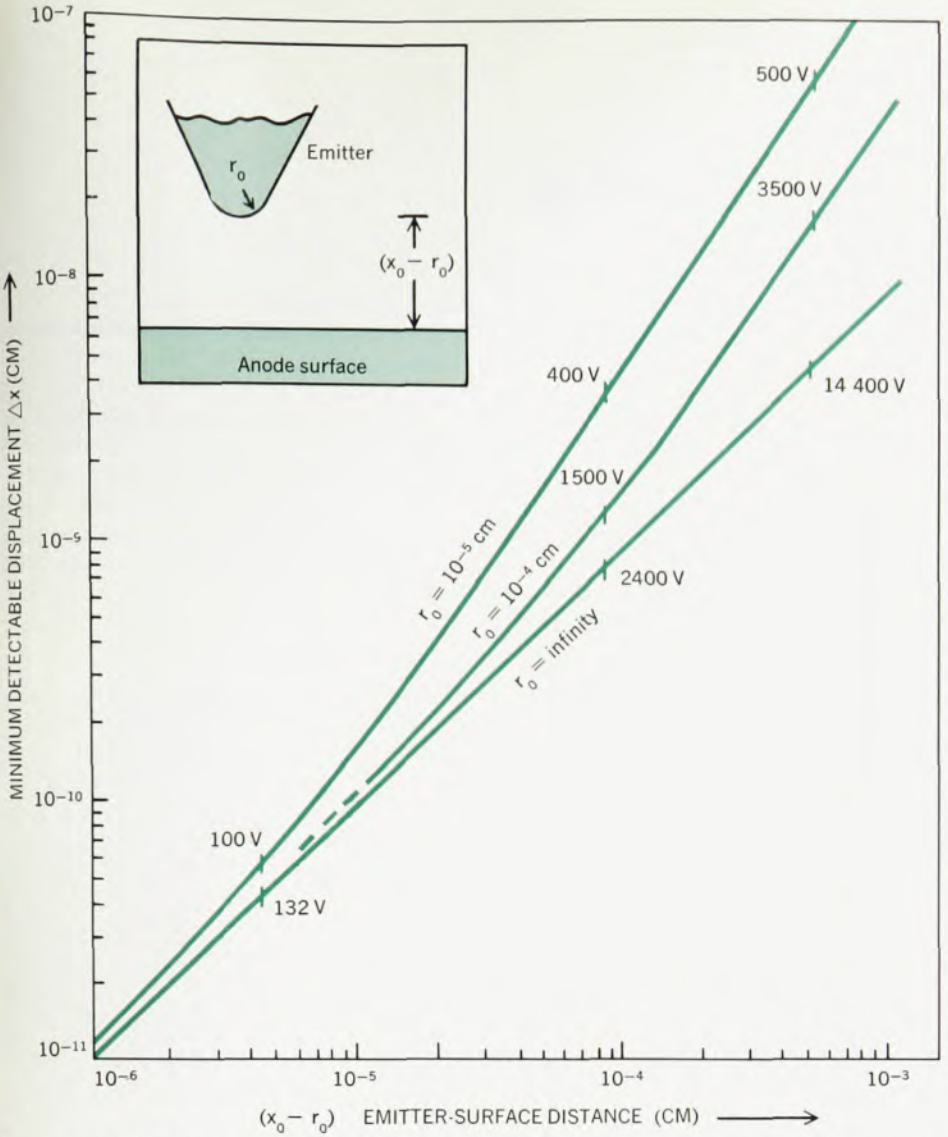
The instrument can also be used in an alternative mode. The secondary electrons and photons generated when the field-emitted electrons strike the specimen can be detected with an electron multiplier, amplified, and used to modulate the brightness of a cathode-ray-tube beam. This is expected to result in a secondary-electron or photon-emission picture of the surface similar to a scanning-electron-microscope picture. It can also be combined with other surface-science techniques, notably the appearance-potential technique, to determine the chemical nature of the surface under the probe at any point on the surface.

The minimum detectable change in emitter–specimen spacing determines the elevation sensitivity of the instrument. The theoretical sensitivity[17] is shown in figure 13. Thus, if no electrical noise or mechanical vibration were present a 1000 Å (10^{-5} cm) emitter operating about 500 Å (100 V) above the surface could be used to detect a change in surface elevation of less than 10^{-2} Å. Electrical noise and mechanical vibration limit the present instrument to 3 Å or about one atom-step elevation sensitivity. At present our typical vertical resolution is 30 Å. We are trying to improve signal-to-noise, and resolution should ultimately be limited by the thermal vibrations of the emitter and specimen.

The horizontal resolution of the topografiner is limited by the emitter–anode spacing and the emitter radius. The emitter can easily be kept very close to the surface while scanning, so the ultimate resolution in the plane of the surface is expected to be about twice the smallest achievable emitter radius, that is about 200 Å. The instrument also has a zoom feature in that the emitter can be withdrawn from the surface by increasing the emitter-specimen voltage as shown in figure 13. When the emitter is far above the surface the horizontal resolution is about equal to



The topografiner, shown very schematically. X and Y piezo drivers scan the emitter parallel to the specimen surface and slightly above it. Emitter-to-specimen voltage is determined by the constant current passing between them, and by their spacing. The Z piezo is controlled by a servo system that holds this voltage, and hence the emitter-specimen spacing, constant. Thus the Z-piezo voltage gives the surface profile directly.
Figure 12



Theoretical sensitivity of the topografiner, plotted as the minimum detectable change in emitter-specimen spacing. The servo loop gain is assumed sufficient to hold the emitter-specimen voltage constant to one part in 10^5. (From reference 17.)
Figure 13

the emitter-specimen spacing. Under these conditions the specimen surface can be rapidly scanned at low resolution to survey the topography before zooming in to study a portion of the surface in detail.

We are currently engaged in a feasibility study to determine the capabilities, design parameters and limitations of the basic instrument. We chose a gold-plated nickel replica of a 180-line-per-millimeter diffraction grating to work on at first, because its surface is already fairly well characterized. A topographic map of the replica surface made with this instrument is shown in figure 1.

When the emitter in the topografiner is brought to within 10 or 20 Å of the surface, electrons tunnel directly from the emitter to the anode in the same way that electrons tunnel through oxide layers in thin film devices. John Lamb and R. Jaklevic[18] have shown that if there are molecules present in the oxide-filled region between the metal electrodes, then electrons lose energy corresponding to the excitation of molecular spectra in these molecules. They have obtained beautiful, sharp spectra for a variety of molecules. The advantage of the topografiner results from the fact that the electrons are tunneling in vacuum and can be used to excite vibrational spectra of atoms and molecules adsorbed on well characterized surfaces. This technique should be useful in deciding whether or not molecules dissociate when adsorbing on particular types of surfaces as well as other questions in surface chemistry.

The usefulness of an instrument for measuring surface topography depends on its ability to measure simultaneously the horizontal and vertical separation between two points. The topografiner's horizontal and vertical resolutions are shown in the table (page 47) in comparison with the four other types of instrument that I have discussed, and it compares (perhaps surprisingly) favorably with them. Note that all instruments appear to have a topographic resolution of about 100 000 Å². The resolutions listed in the table are meant to represent the performance of the different instruments under typical working conditions, not necessarily the utmost that can be wrung from the devices with special care.

The uniqueness of the topografiner lies in its ability to combine high-resolution topographic mapping with concurrent secondary-electron-emission pictures of the surface along with chemical analysis. This capability led to the name of the instrument, which is taken from the Greek word *topographein*—"to describe a place."

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