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LBL-36713
UC-401
Preprint

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

CHEMICAL SCIENCES DIVISION

Submitted to Review of Scientific Instruments

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January 1995

sw9526

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

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This work was supported by the U.S. Department of Energy, Materials Sciences Division, under Grant No. DE-FG02-92ER45468 through U.S. Department of Energy under Contract No. DE-AC03-76SF00098

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The SpectroMicroscopy Facility at the Advanced Light Source is based on a high brightness, high resolution beamline, and includes a collection of projects designed to exploit the unique characteristics of the soft x-ray beam. The beamline itself is comprised of a 5 meter long, 5 cm period undulator, a spherical-grating monochromator with water-cooled gratings, and adaptive optics for refocusing the monochromatic beam to spot sizes less than 50 microns. Two experimental stations use the "micro-focus" beam of 50 micron diameter, and a third station uses a "nano-focus" beam formed by a zone-plate lens which demagnifies the micro-focus spot. Experimental stations include an "ultraESCA" spectrometer for small-area spectroscopy and photoelectron diffraction, a scanning transmission x-ray microscope, and photoelectron microscopes.

1. INTRODUCTION

The SpectroMicroscopy Facility beamline (7.0) at the Advanced Light Source is comprised of a 5 meter, 5-cm-period undulator,[1] a >8,000-resolving power spherical grating monochromator covering the spectral range from 100 eV to 1300 eV,[2] and refocusing optics that deliver resolved fluxes of $>10^{12}$ photons/sec into a focal spot less than 50 μm diameter.[3] This combination of high brightness, high spectral resolution, and finely focused beam spots opens new opportunities in spectroscopy and microscopy experiments of photoelectrons and fluorescence. The first experiments described here, performed during the commissioning phase of the beamline, highlight the performance of these three factors.

The SpectroMicroscopy Facility (Fig. 1) was designed to supply up to four experimental endstations with a monochromatic X-ray beam, through the use of interchangeable mirrors and experimental chambers designed to allow for passage of the beam during idle periods (tandem chambers). A pair of interchangeable horizontal refocusing mirrors (b) switches the beam from the monochromator between two branchlines with either a 6-degree or a 4-degree deflection. A stigmatic, "micro-focus" spot is formed along each of these branchlines, with a difference in focal length of 1 meter to provide separation between experiments. The change in focal length can be compensated by an adaptive vertical refocusing mirror (a), so that a stigmatic focus is obtained on both branchlines. Experimental stations (c,d) are placed at each micro-focus location and are designed to allow the beam to transit the vacuum chamber so as to enter additional, tandem, experiments. On the 6° branch line, the micro-focus spot is the object point for additional demagnification in a pair of zone-plate microscopes (e), one of which works at atmospheric pressure in transmission, and the other of which is an ultra-high vacuum photoemission microscope.

The SpectroMicroscopy Facility has permanent endstations for the "ultraESCA" project, the scanning transmission x-ray microscope (STXM), and the scanning UHV

photoemission microscope (SPEM). The ultraESCA station is installed and operational; the STXM and SPEM are under construction. In addition, two interim experiments have been performed at the micro-focus points: soft x-ray fluorescence spectroscopy and photoemission microscopy. The fluorescence experiments were performed with a high-resolution grating soft x-ray spectrometer designed by Nordgren and co-workers of the USX group at the University of Uppsala, Sweden. This experiment alternates with PRISM (Paraxial Ray Imaging Spectro-Microscope), a parallel-imaging photoemission microscope designed by Tonner and co-workers at the University of Wisconsin-Milwaukee.

2. ULTRA-ESCA

The "ultraESCA" project is an attempt to make dramatic improvements over conventional x-ray photoelectron spectroscopy performance in several areas at once: high intensity, small area, high energy resolution and high angular resolution. The ultraESCA station is equipped with a 137 mm radius hemispherical photoelectron spectrometer with multi-channel detection. A variable-aperture lens capable of ±0.5° angular resolution will be used in conjunction with a precision sample goniometer for scanned-sample photoelectron diffraction experiments. A comparison of the sensitivity of ultraESCA has been made to that of a modern, conventional photoemission system using a monochromatized Al K_α laboratory source. As a figure of merit for sensitivity in an analytical use of x-ray photoemission spectroscopy, we use the number of photoelectron counts per second, per unit sample area, per equivalent energy bandwidth. Compared to the conventional system, we find an enhancement of $>2\times10^4$ at the ultraESCA station.[4]

One demonstration of the ultraESCA capabilities is a simple scanning photoemission microscopy experiment using the spatial resolution of the refocused x-ray spot. Figure 2 shows a raster scan image of Au 4*f* photoelectrons from a 360 μm

period gold grid atop a silicon wafer. Using the known line width of the wires (30 μm) and correcting for the horizontal incidence angle (60°), we determine the x-ray spot size to be ~25 μm × 25 μm. The vertical spot size is controlled by the imaging of the monochromator exit slit with near unit magnification by a variable-radius refocusing mirror. The size of the horizontal beam spot, refocused by a fixed-radius mirror, is ultimately limited by the size of the electron beam. Beamline apertures and spectrometer pass energy were adjusted to obtain in excess of 1 MHz count rates of the Au 4*f* peak. The image in Fig. 2 can also be acquired very rapidly with total electron yield. This imaging capability with high throughput is very useful in locating small samples, as in the Curium experiment (Section 3), and in rapidly performing 'chemical-shift' microscopy with very high overall energy resolution.

3. NANOGRAM ACTINIDE PHOTOEMISSION

The first ultraESCA experiment to be performed at the SpectroMicroscopy Facility was a proof-of-principle demonstration of the analysis of a minute quantity of a radioactive transuranic sample. In this experiment approximately 1.25 μg of the isotope curium-248 was deposited in the form of a curium oxide distributed uniformly over a spot 2.5 mm in diameter on a platinum disk. The total activity of the curium sample was less than 20 nanocuries, most of which arises from a trace curium-246 impurity in the curium-248 material. With a 50 μm focused x-ray beam, only approximately 5 nanograms of curium-248 are estimated to be illuminated by the photon beam. The quantity of interest in future experiments will be matched to our focused spot size.

Spectra were collected for photoelectrons emitted from the curium 4*f*, 4*d* and valence electron energy levels as well as the O 1s core level. The relative cross-sections of the curium 4*f* and 4*d* transitions, as a function of photon energy, were roughly determined using the Pt substrate as a reference. A representative curium 4*f* spectrum

is shown in Fig. 2. Previously, curium photoelectron spectra had been recorded with fixed-energy laboratory sources only.[5]

The results of this curium investigation have major implications for future studies of radioactive materials; minute quantities which can be handled safely without elaborate precautions are sufficient for detailed chemical analysis with ultraESCA capabilities. The scanned-sample imaging ultraESCA capability demonstrated in Fig. 2 will be used in the future to locate minute samples as well as to measure the distribution of chemical compounds across a sample. The technological impact of such measurements is expected to span nuclear solid-waste characterization and storage, non-proliferation of nuclear weapons, and fundamental research of the electronic properties of the heaviest elements.

4. SURFACE CORE-LEVEL PHOTOELECTRON DIFFRACTION

The atomic assignment of surface core-level shifts (SCLS) of clean reconstructed silicon surfaces remains controversial.[6,7] Because of the inherent structural information contained in photoelectron diffraction intensity variations, further progress in this area can be made with experimental measurements of the angular and/or photon energy dependence of the SCLS spectra. Because of the high resolution required and low occupation of the spectral components, these results typically require many days or weeks to acquire a sufficiently large data set.

With a high brightness, high-resolution source, data collection times can be greatly reduced, so that experiments can now potentially be performed within a single synchrotron shift. One example is the Si 2*p* SCLSs of the Si(100) 2×1 reconstructed surface. Fig. 4 shows a high-resolution Si 2*p* spectrum excited by 152 eV photons. The spectrum was collected in the angle-integrated mode of the spectrometer input lens with 3 eV pass energy (<50 meV resolution) and >8,000 bandpass of the monochromator. Visible is a low binding energy shoulder (*S*) resulting from a surface

Si species. One or more additional surface species (*S'*) also exist at higher binding energy. Quantification of these results using a 3-peak curve-fit of the data is shown in Fig. 4. Sufficient statistics for curve-fitting were obtained in only 10 seconds of data collection. Also, greater than 3 MHz at the Si $2p_{3/2}$ peak has been obtained by widening horizontal apertures without degrading the photon bandwidth., and operation down to 0.6 eV pass energy for higher spectrometer resolution exhibits only the expected linear decrease in count rate.

Fig. 5 shows the dependence of these three Si $2p$ SCLS intensities upon azimuth angle from a Si(100) 2×1 sample. The low and high binding energy surface components exhibit the 4-fold symmetry of the bulk with distinctly different oscillations than the bulk signal. The total acquisition time of the azimuth scan was less than 20 minutes. This data collection speed increases the feasibility of many more angle-scanned or energy-scanned component-resolved photoelectron diffraction experiments for the analysis of sub-monolayer species, and is especially important for surfaces that are reactive or degrade rapidly with time.

5. HIGH-RESOLUTION SOFT X-RAY FLUORESCENCE

An interim endstation installed on the 6° branchline is equipped with a high resolution grating soft x-ray spectrometer. The high brightness from the undulator increases the sensitivity to emission from sub-monolayer signals as well as allowing for higher resolution emission spectra. The narrow focusing capability allows moving the excitation spot closer to the spectrometer entrance slit while remaining matched to the acceptance of the spectrometer; a further gain in efficiency is realized by the larger net solid angle of emission seen by the entrance slit. High resolution, tunable photons allow selective excitation of transitions between electronic levels greatly enhancing the ability to interpret the emission spectra.

The spectrometer covers the large spectral range of 50-1000 eV with the use of multiple fixed gratings and a large two-dimensional detector.[8] The three chamber system consisting of analysis, prep and loadlock vessels is detailed elsewhere.[9] In addition to 6-axis sample manipulation, LN_2 cooling and resistive heating capabilities, the analysis chamber is most notably able to rotate 90° under UHV conditions which allows the polarization dependence of soft x-ray emission to be studied. Figure 6 illustrates the polarization dependence of diamond C K-emission excited on resonance with the core exciton, where fluorescence detection perpendicular to the plane of polarization of the incident photons shows a dramatic increase in the exciton emission. The magnitude of this resonant enhancement and resolution of detailed structure in the sideband for parallel emission illustrates at least an order of magnitude greater sensitivity in comparison to previous synchrotron measurements.[10] In addition to reduction of data acquisition times from hours to minutes due to increased flux, the ability to use a narrower bandpass enhances the resonant excitation condition. Polarization dependent studies in general also allow orbitals with different angular character to be probed. In addition to diamond samples, polarization-dependent and angularly-resolved experiments of selectively-excited x-ray emission from high T_c systems, fullerenes, and molecular ices have been performed at beamline 7.0.[9]

6. SUMMARY

Commissioning tests for the SpectroMicroscopy Facility at beamline 7.0 at the Advanced Light Source have shown that the beamline meets or exceeds all design specifications for spectral brightness, energy resolution and focused spot sizes. This performance of the beamline has allowed us to demonstrate dramatic improvements in sensitivity for photoemission and soft x-ray fluorescence spectroscopy experiments.

ACKNOWLEDGMENTS

This work was supported by the Department of Energy, Materials Science Division, under grant No. DE-FG02-92ER45468.

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CAPTIONS

Fig. 1. Present layout of the SpectroMicroscopy Facility including: (a) vertical and (b) horizontal refocusing optics, and (c) "ultraESCA" and (d) x-ray fluorescence micro-focus endstations on the 4° and 6° branchlines. Future nano-focus zone-plate microscopes will reside at location (e).

Fig. 2. Raster-scan ESCA image highlighting the sub-50 μm spot size achievable by the beamline refocusing optics. A gold grid with 360 μm periodicity and 30 μm width wires was imaged using Au 4*f* electrons excited by 400 eV photons.

Fig. 3. Curium 4*f* spectrum obtained with hν=1040 eV excitation. An estimated 3 nanograms of curium oxide was excited by the ≈50 μm diameter beam spot.

Fig. 4. High resolution Si 2*p* spectrum at hν=152 eV exhibiting surface core-level shifts (*S*, *S'*) associated with the Si(100) 2×1 surface reconstruction [inset, low energy electron diffraction pattern showing the 2×1 surface order].

Fig. 5. Component-resolved Si 2*p* amplitudes as a function of azimuth angle for θ =55° from normal emission. Data collection time for individual spectra was 10 seconds.

Fig. 6. Polarization dependence of diamond C K-emission excited on the core exciton [inset, photoabsorption spectra showing the selective excitation energy].

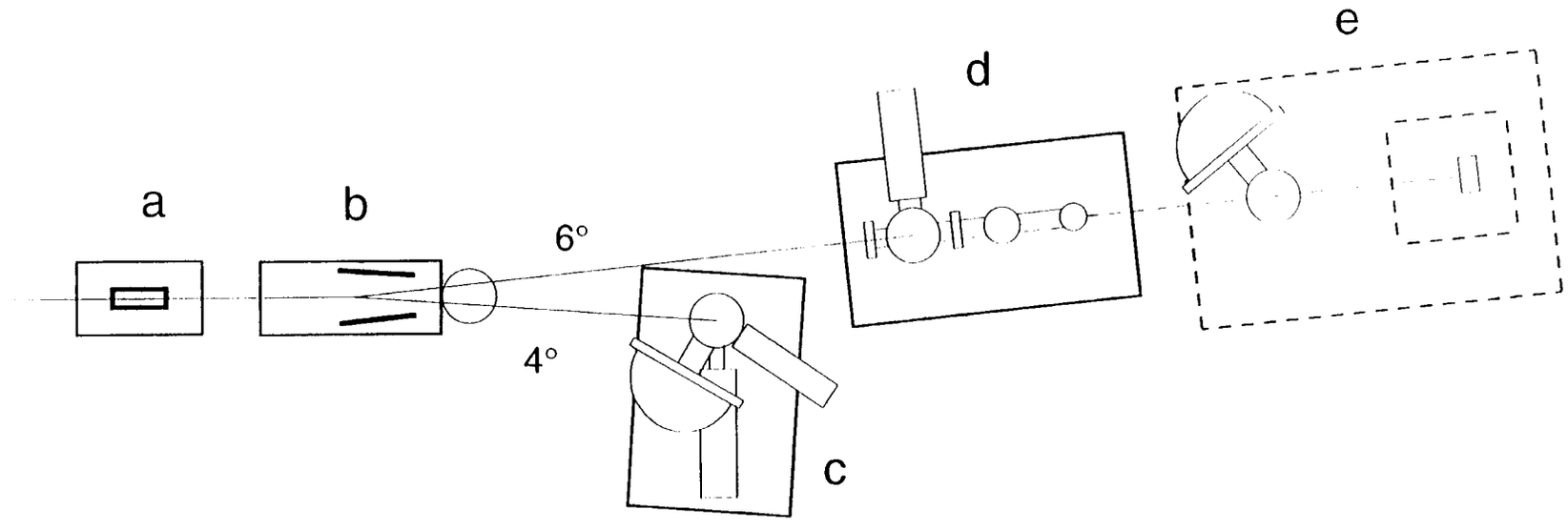

Figure 1

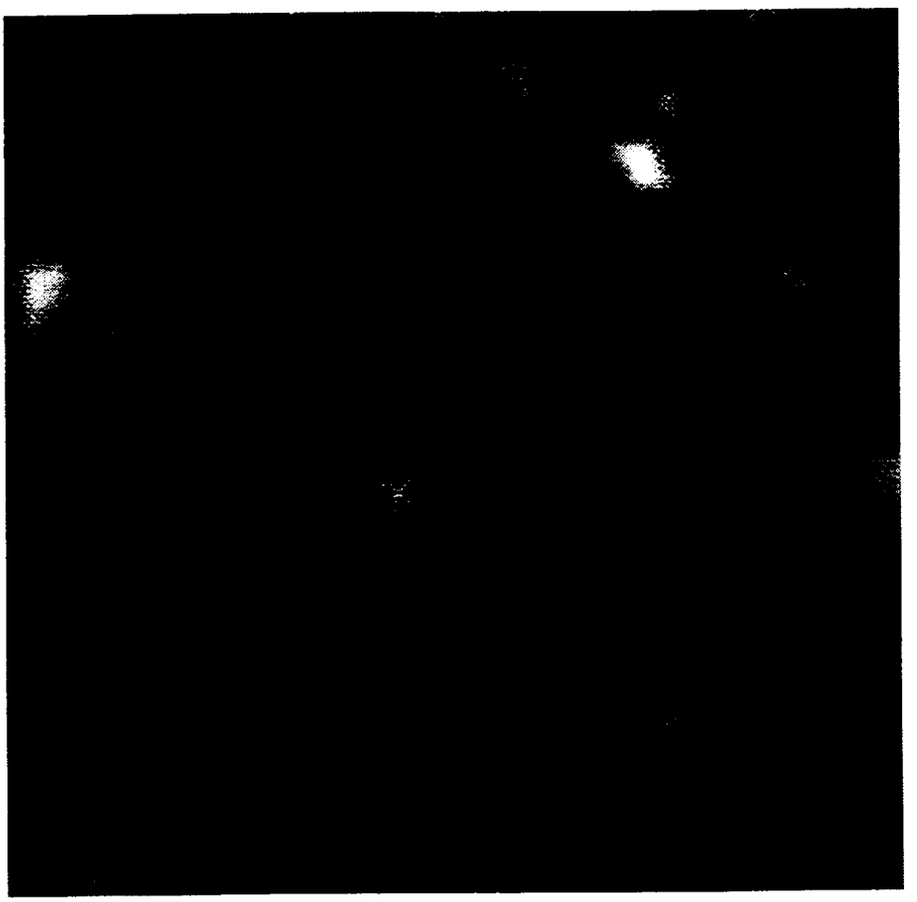

Figure 2

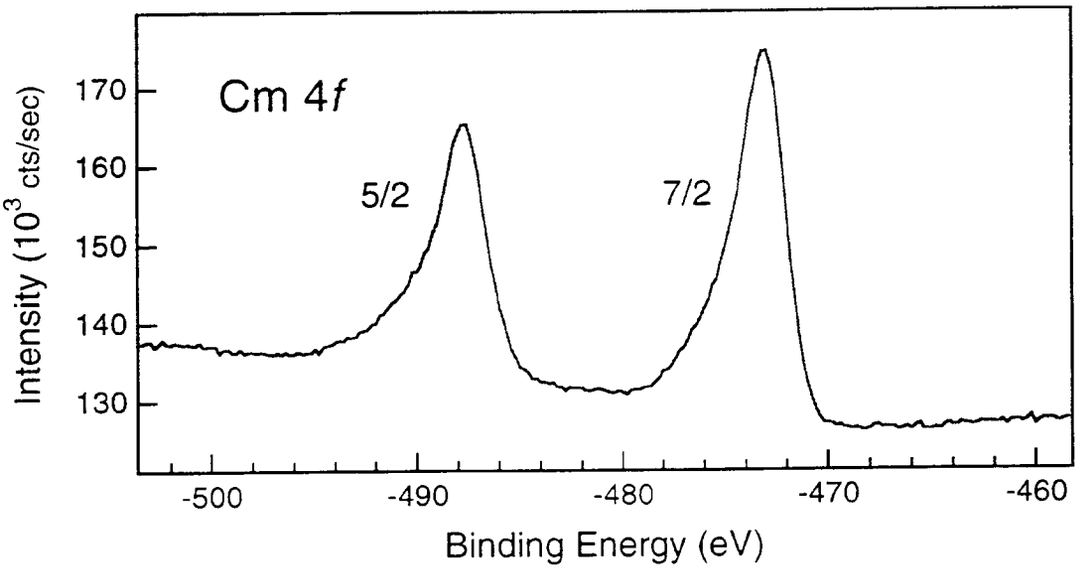
Cm 4f
5/2
7/2
Intensity (10^3 cts/sec)
170
160
150
140
130
-500
-490
-480
-470
-460
Binding Energy (eV)

Figure 3

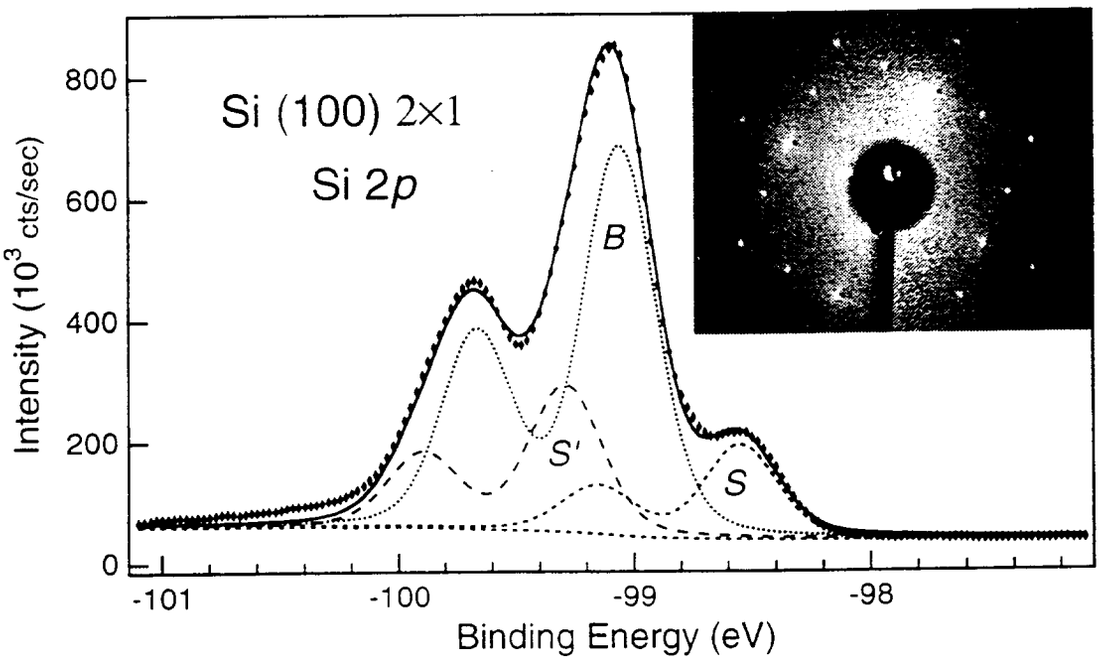
Si (100) 2×1
Si 2p
B
S'
S
Intensity (10^3 cts/sec)
800
600
400
200
0
-101
-100
-99
-98
Binding Energy (eV)

Figure 4

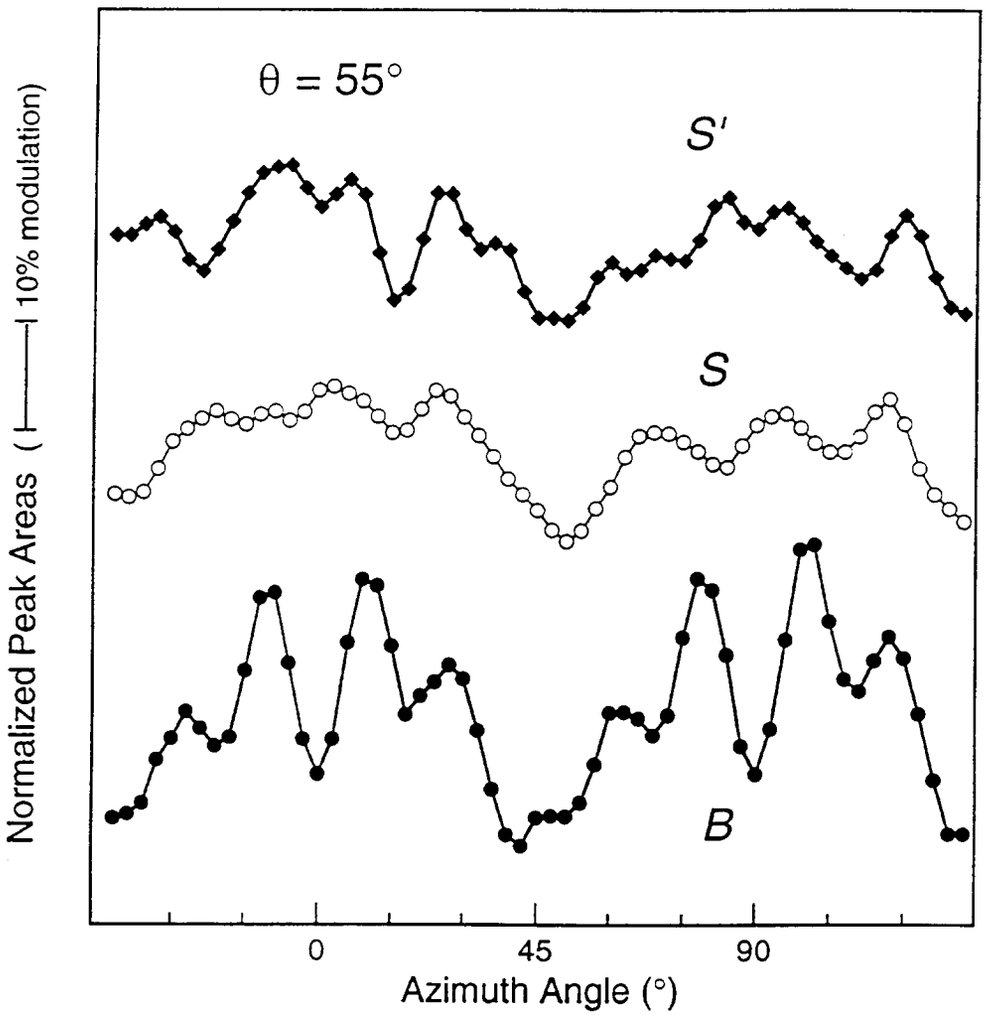
θ = 55°
S'
S
B
Normalized Peak Areas (10% modulation)
0
45
90
Azimuth Angle (°)

Figure 5

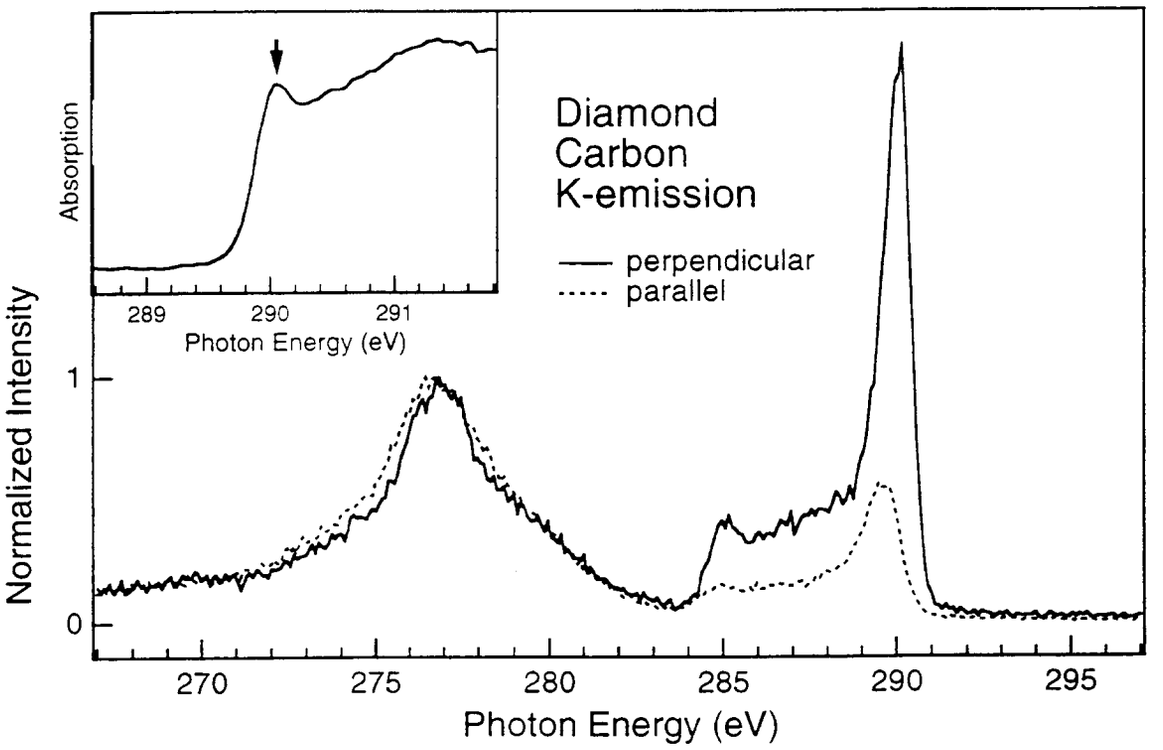
Absorption
289
290
291
Photon Energy (eV)
Diamond
Carbon
K-emission
perpendicular
parallel
Normalized Intensity
1
0
270
275
280
285
290
295
Photon Energy (eV)

Figure 6