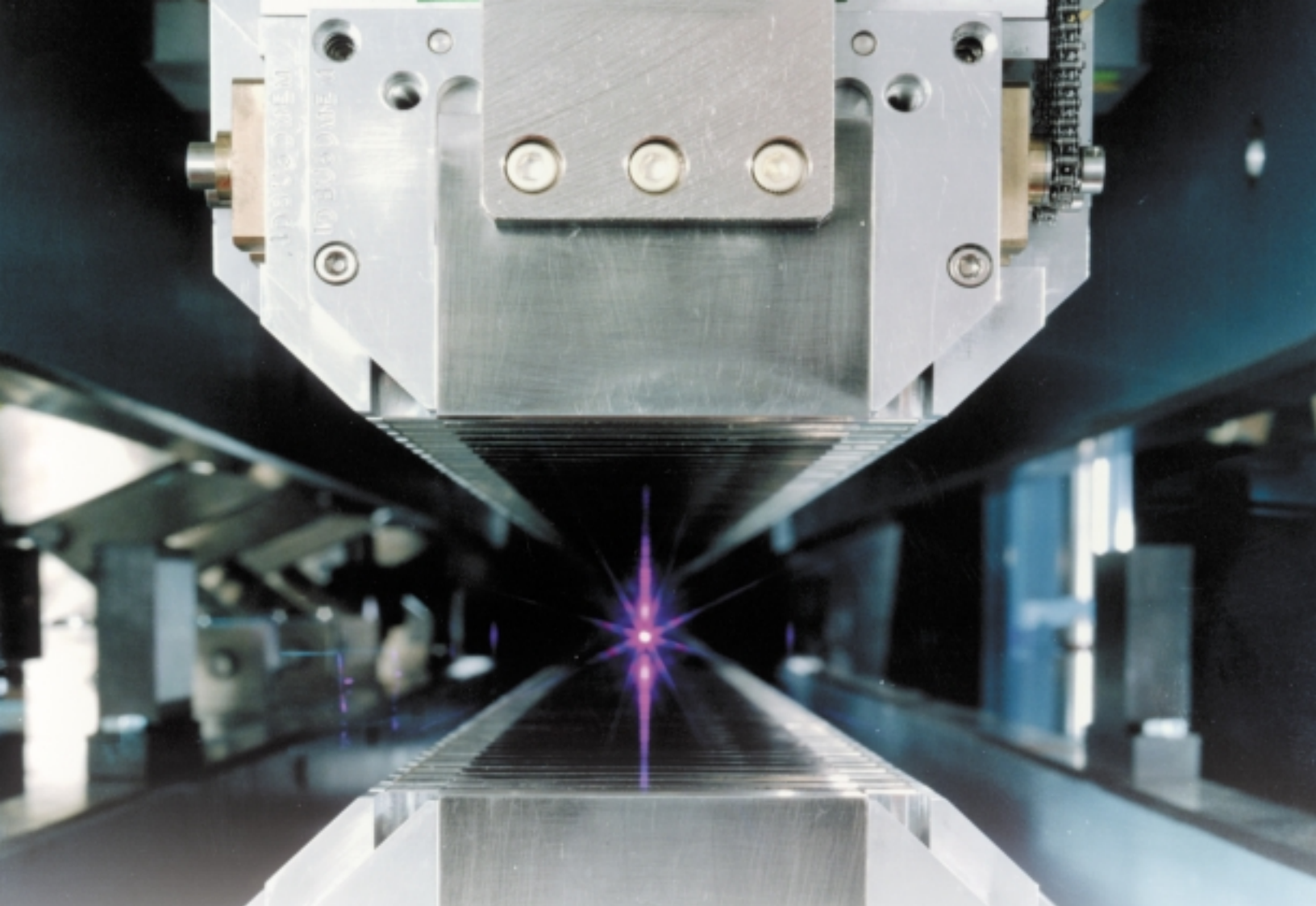

Actinide Photoemission Measurements at the Advanced Light Source

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The Advanced Light Source (ALS) at the Lawrence Berkeley National Laboratory is the nation's premier third-generation synchrotron light source for performing vacuum ultraviolet (VUV) and soft x-ray studies of materials. We have used Beamline 7.0.1 at the ALS to perform photoemission spectroscopy (PES) measurements of plutonium metal and compounds. The beamline operates over the energy range 80–1200 electron-volts (eV) using an undulator (shown in the top photo) with a 5-centimeter period and a spherical-grating monochromator. Spectral resolutions of up to 8000 ($E/\Delta E$) can be achieved with a light flux (per 0.01 percent bandwidth) of approximately 10^{13} photons per second (photons/s) for energies less than 200 eV, 10^{12} photons/s for intermediate energies, and 10^{11} photons/s for energies greater than 500 eV. Shown in Figure 1 is a schematic of the beamline from the undulator insertion device in the synchrotron ring to the end stations (or analysis chambers), where the light is used for materials and surface measurements. To select the photon energy, we use one of three gratings. The adjustable beam-defining apertures and the refocusing mirrors set the size of the photon spot at the sample surface.

The UltraESCA photoemission analysis chamber was built around a 137-millimeter spherical-capacitor electron-energy analyzer (Physical Electronics Omni IV), which allows high-resolution photoemission measurements, as well as photoelectron imaging through adjustment of the input lenses. The schematic in Figure 2 shows the location of the UltraESCA analysis chamber on the beamline, past the last refocusing mirror, relative to the other instruments on the beamline. This location takes advantage of the small focus of the photon beam, which may be as small as 50 micrometers in diameter at

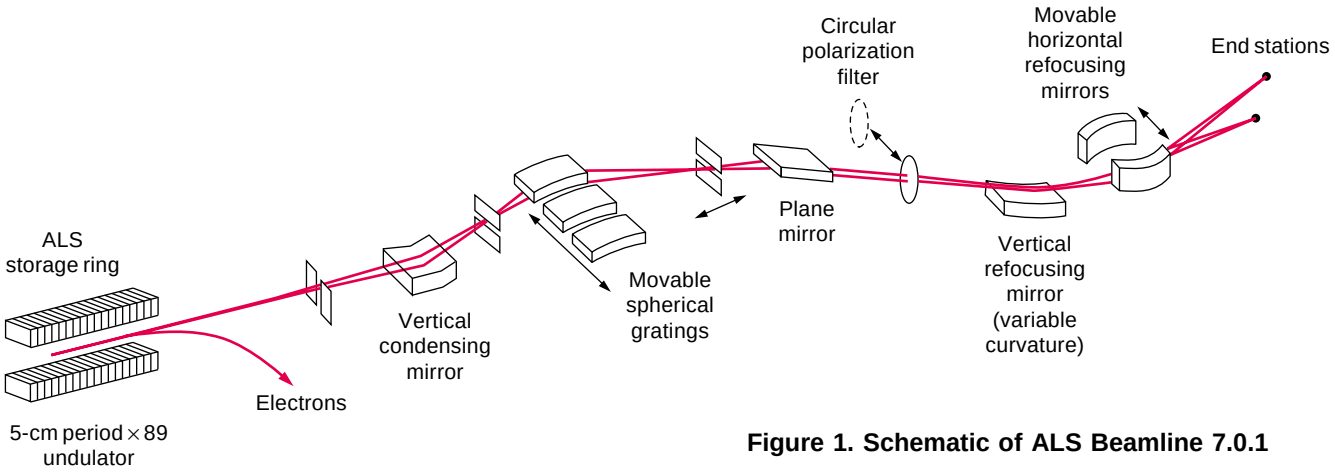

Figure 1. Schematic of ALS Beamline 7.0.1

the sample surface, while still maintaining the high flux. This feature was critical in our measurements, and it will also be so in future experiments because the samples we analyze are very small. In addition to dealing with limitations regarding the size of samples, we attempted to measure the photoemission spectrum from a single-crystallite surface, 500 × 1000 micrometers in size, which was surrounded by a matrix of other crystallites. The focused photon beam allowed us to measure exclusively from the surface of this single crystallite. Future work will include measurements for mapping the electronic structure bands of plutonium metal and x-ray photoelectron diffraction measurements to examine surface crystal structure. In addition, a dedicated actinide spectrometer is being assembled to allow final sample cleaning and preparation to take place within the analysis chamber. In this way, excessive sample transfer operations are eliminated and sample purity is ensured. We will be able to move this spectrometer system to other ALS beamlines and thus use other specific light characteristics (such as photon energy range or polarization) in our measurements.

To make these measurements at the ALS, we designed and built a special vacuum system with which to handle the samples. We called it the Sample-Handling and Integrated-Transfer System for Plutonium Intense-Light Experiments. The system is a quad-chamber, wholly enclosed ultrahigh-vacuum ion-pumped system with sample-transfer devices. It also includes a differentially pumped ion gun and sample-heating stage used for sample cleaning (by argon-ion sputtering) and annealing. This system was temporarily connected to the UltraESCA analysis chamber during our experiments so that the plutonium metal samples could be prepared and transferred under vacuum, with surfaces kept clean until the samples could be measured by photoemission. The initial sample preparation, cleaning, and analytical spectroscopy take place at Los Alamos. The samples are then placed under vacuum into transfer vessels, which are maintained at a pressure of 10^{-8} torr with a battery-powered ion pump system. These transfer vessels are then shipped to the ALS, where they are coupled to our sample-handling system. To maintain the cleanliness and integrity of the samples, we never allow them to see the atmosphere but transfer them under high vacuum. The sample-handling system then allows us to perform a final clean-and-anneal cycle before the samples are transferred into the UltraESCA analysis chamber for PES measurement. ■

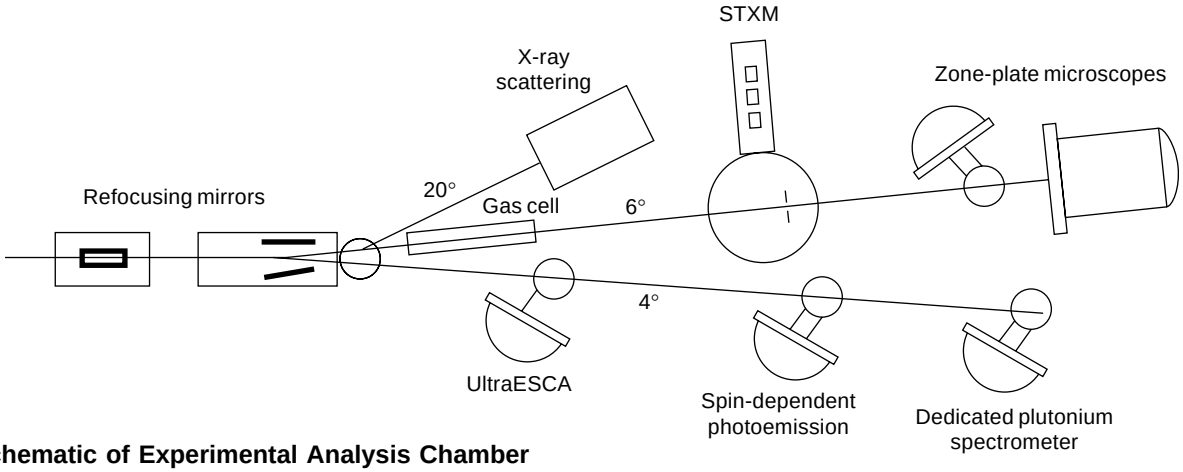

Figure 2. Schematic of Experimental Analysis Chamber