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Sample-angle feedback for diffraction anomalous fine-structure spectroscopy

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Diffraction anomalous fine-structure (DAFS) experiments measure Bragg peak intensities as continuous functions of photon energy near a core-level excitation. Measuring the integrated intensity at each energy makes the experiments prohibitively slow; however, in many cases DAFS can be collected quickly by measuring only the peak intensity at the center of the rocking curve. A piezoelectric-actuator-driven stage has been designed and tested as part of a sample-angle feedback circuit for locking onto the maximum of the rocking curve while the energy is scanned. Although software peak-tracking requires only a simple calculation of diffractometer angles, it is found that the additional hardware feedback dramatically improves the reproducibility of the data.

Keywords: diffraction anomalous fine structure; XAFS; anomalous scattering; sample-angle feedback.

1. Introduction

Diffraction anomalous fine-structure (DAFS) experiments measure Bragg peak intensities as continuous functions of photon energy in the vicinity of a core-level excitation (Sorensen *et al.*, 1994). The oscillations in the anomalous-scattering amplitude, $\Delta f(E)$, observed above the absorption-edge energy are due to solid-state effects and can be used to obtain information about the local environment of the resonant atoms. The diffraction condition is used to measure selectively the fine structure from a subset of the resonant scatterers based on their long-range order. For example, the DAFS measured at reciprocal lattice points unique to one component or phase of an inhomogeneous material contains local structural information about the resonant atoms in that phase alone. In addition, the DAFS amplitudes from ordered structures with inequivalent resonant sites in the unit cell are linear combinations of the fine structure from the individual sites, and can be combined to isolate local structural information from a specific site. The DAFS amplitudes, $\chi(\mathbf{Q}, E)$, isolated from the measured intensity signal can by analyzed and interpreted using standard X-ray absorption fine-structure (XAFS) methods (Cross, 1996).

The integrated intensity of a Bragg peak, in the weak scattering limit, is proportional to the squared magnitude of the unit-cell structure factor. Measuring a complete θ-rocking curve at every energy point in the DAFS scan, however, makes DAFS experiments prohibitively slow for most synchrotron users. For thin film samples with regular rocking curves it has been observed that the peak intensity, $I(E, \theta_B)$, is proportional to the integrated intensity, $\int I(E, \theta)\, d\theta$, after correcting for fluorescence background, over the entire energy range of a DAFS experiment (Stragier *et al.*, 1992). Under these conditions, undistorted $\chi(\mathbf{Q}, E)$ can be obtained by measuring the peak reflectivity as a function of energy. The position of the Bragg peak can be calculated easily as a function of energy and given as an instruction to the diffractometer motors while the incident energy is scanned. This software peak-tracking method of measuring DAFS significantly decreases the amount of time necessary to collect a complete spectrum; however, it is crucial that each measurement be taken at the same point on the rocking curve. Any drift away from the peak $\theta_B(E)$ will appear as a decrease in the intensity. Systematic errors, such as energy tracking in the monochromator (Kim *et al.*, 1991) or temperature-induced variations in the lattice parameters, will distort the fine structure and background functions and cause poor reproducibility in the data. In addition, round-off errors in calculating the monochromator or diffractometer motor positions can introduce systematic point-to-point noise that is difficult to distinguish from stochastic noise.

One way to ensure accurate tracking of the Bragg peak is by electronic feedback on the detector intensity. This method is commonly used to maintain maximum intensity in scanning monochromators for synchrotron beamlines and in many other applications (Cowan *et al.*, 1983). In this work, a piezoelectric-actuator-driven sample stage was designed and tested to drive the sample angle as part of a feedback circuit for improving the reproducibility of DAFS data collected with software peak-tracking. Fig. 1(*a*) shows a sketch of the flexured sample stage and Fig. 1(*b*) shows the experimental arrangement for Bragg peak tracking with feedback. A small modulation was added at

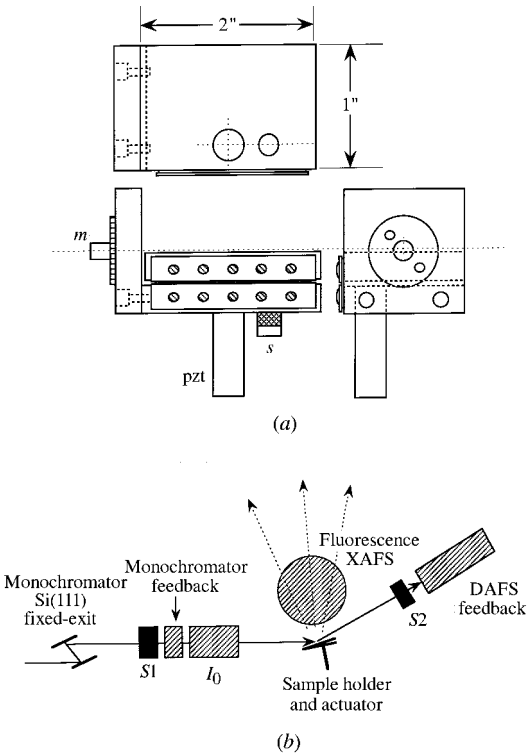

Figure 1
(*a*) Sketch of the sample stage showing the piezoelectric actuator (pzt), restoring spring (*s*) and goniometer mounting post (*m*). (*b*) Experimental arrangement for a DAFS experiment with Bragg peak-tracking and sample-angle feedback locking. The Bragg angle is calculated by the diffractometer control software at each energy, and the lock-in circuit corrects any small errors in the sample angle position.

the actuator to dither the sample angle around θ_B, producing a synchronous signal in the detector proportional to the derivative of the rocking curve. The detector output was fed to the input of a lock-in amplifier and the DC output of the lock-in (proportional to the deviation from the peak of the rocking curve) was fed back to the actuator, keeping the sample aligned at the peak of the rocking curve during the DAFS scans. The angular positions for the θ and 2θ diffractometer motors were calculated using the DAFS capabilities of the *LabVIEW*-based *NSLS-DAC* beamline control and data-acquisition package (Furenlid *et al.*, 1997) currently used for XAFS spectroscopy on several beamlines at the National Synchrotron Light Source. The sample-angle feedback was used to correct for observed systematic errors in the angle position of the order 0.002–0.01°. Errors of this magnitude are 5–25% of the sample rocking curve FWHM of ~0.04°, making feedback correction essential.

2. Experiment

The sample stage shown in Fig. 1(*a*) was made from two 2.5 × 5 cm aluminium plates joined along one of the long edges by a weak flexure of stainless-steel shim stock. A Burleigh PZL-060-00 piezoelectric actuator attached to the bottom plate pushes against the top plate to change the sample angle. The actuator has 40 μm extension and was mounted 1 cm from the flexure, giving a maximum of 0.23 degrees of angular displacement. A spring-loaded bolt provides the restoring force. The bottom plate was attached along one of the short edges to a 2.5 cm-square aluminium block and a standard Huber 1003 specimen holder was attached to the back of the plate to provide a stainless steel post for mounting the stage on the goniometer head.

Fig. 2 shows the Fe *K*-edge DAFS from the (001) reflection of an Fe/Si multilayer; the inset in Fig. 2(*a*) shows the reflectivity of the multilayer in the vicinity of the (001) peak, at 7412 eV. Four sequential DAFS scans are overplotted in Fig. 2(*a*), and an expanded view of the near-edge region is shown in Fig. 2(*b*) for clarity. The first two scans (lower intensity curves) were collected using software peak-tracking only. The second two scans, taken immediately following the previous data, were collected with both software peak-tracking and hardware sample-angle feedback, under otherwise identical experimental conditions. Careful examination of the expanded view in Fig. 2(*b*) is required to see any difference between the two scans with hardware feedback, and the systematic glitches observed in the data with software feedback only have been significantly reduced.

All of the data were collected at NSLS beamline X23B using the *NSLS-DAC* software in DAFS mode for the software peak-tracking. The sample was nominally (40 Å Fe/14 Å Si) × 25 layers with a 30 Å Fe buffer layer and a 30 Å Ge cap, grown on an Si(111) substrate by ion-beam-sputter deposition (Chaiken *et al.*, 1996). The scans were collected in the energy range −300 to 750 eV with respect to the Fe *K*-edge. The monochromator energy was calibrated to 7112 eV at the first peak in the energy derivative of the fluorescence XAFS, and the sample and detector angles were aligned at 7412 eV before starting the series. The data in Fig. 2 show a typical level of reproducibility for DAFS experiments with software peak-tracking, with perhaps slightly worse point-to-point noise due to a diffractometer motor control error in a beta version of the DAFS mode.

The sample stage was driven at 35 Hz to avoid conflict with the X23B monochromator at 77 Hz. The Burleigh PZL-060-00 piezoelectric actuator was driven using an EG&G Princeton Applied Research 5210 lock-in amplifier into a Burleigh PZ-70 amplifier on the 0–500 V range setting and running the output of the Burleigh through a 4:1 voltage divider. The amplitude of the oscillator was adjusted to give a ~3% drop in the detector output at the peak of the rocking curve. The incident beam size was defined by slits at *S*1 set to ±0.75 mm horizontal by ±0.15 mm vertical to give an approximately 1.5 cm footprint in the middle of the scan range. A 5 cm gas ionization chamber flowing dry N_2 was used to monitor the incident beam flux, around 10^9 photons s^{-1} at 7412 eV for this slit setting. A 15 cm ionization chamber flowing a mixture of Ar and N_2 gases was used at the DAFS feedback detector. Diffracted beam slits *S*2 located 15 cm from the sample, to block fluorescence background into the DAFS detector, were set to ±1 mm vertical and ±0.75 mm horizontal. The large vertical aperture accepted the entire rocking curve at the maximum actuator displacement, and allowed for any drift in 2θ stage position during multiple scans. Both the I_0 and I_{DAFS} detectors were held at 1000 V DC between 1 cm plates, and the photocurrent was amplified using Keithley model 428 current

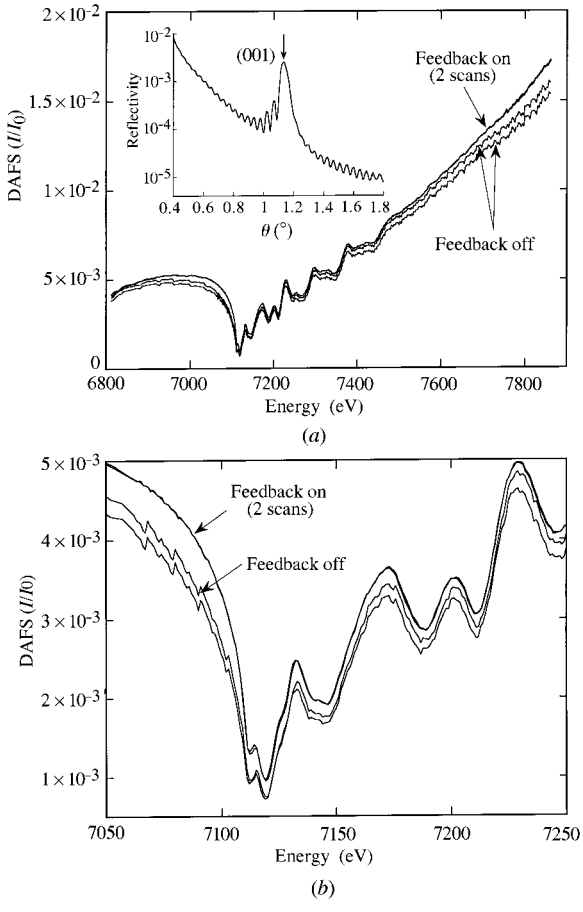

Figure 2
Fe *K*-edge DAFS collected with sample-angle feedback on the (001) peak of an Fe/Si multilayer. The inset shows the sample reflectivity measured at 7142 eV. Four consecutive scans are overplotted in (*a*) and (*b*), two with software peak-tracking only (lower intensity) and two with additional sample-angle feedback. The bottom figure is an expanded view in the near-edge region. The scans taken without feedback have systematic high-frequency glitches as well as a slow drift away from maximum intensity. The scans taken with feedback on are almost indistinguishable and approximately 5% higher in intensity.

amplifiers at 10^7 and 10^9 gain, respectively, with the time-constant for the DAFS feedback amplifier set to 3 ms.

3. Discussion

The DAFS scans shown in Fig. 2 were collected in approximately 23 min each, which is comparable with the time to collect an XAFS spectrum over the same energy range. Note that the background fluorescence needs to be measured separately from the DAFS in the peak-tracking mode; however, this is easily performed by turning the hardware feedback off and running the identical scan with a fixed offset in θ, a built-in feature of *NSLS-DAC*. In addition, it is wise to measure the complete rocking curve at a few energies to confirm the uniformity of $I(E, \theta_B)/\int I, \theta \, d\theta$, and to determine the scale of the background signal. While initial set-up time for DAFS experiments is naturally a little longer than for XAFS (because of the diffractometer alignment), this work has shown that the data collection times can be comparable. In conclusion, the sample-angle feedback described in this work is a simple and effective solution to many of the systematic problems encountered in DAFS experiments.

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