Bent silicon crystal in the Laue geometry to resolve x-ray fluorescence for x-ray absorption spectroscopy

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A highly strained, curved silicon crystal in the Laue geometry has been used as a large-area x-ray fluorescence analyzer for x-ray absorption spectroscopy. The analyzer is able to resolve the $L\alpha$ fluorescence lines for neighboring actinide elements. A large gain in the signal to background ratio has been demonstrated for small quantities of Np in the presence of U, with the U fluorescence peak approaching 1000 times the magnitude of the off-peak background. © *2003 American Institute of Physics*. [DOI: 10.1063/1.1618014]

INTRODUCTION

Measuring x-ray fluorescence from a sample is a common method of obtaining x-ray absorption spectroscopy (XAS) data from dilute samples and other sample configurations for which the transmission absorption measurement is impractical. The primary limitation to using XAS for many interesting environmental and biological problems is the presence of a high level of background radiation from a variety of sources, such as elements with lower energy absorption edges, radioactive elements, or scattered radiation from other beam interactions with the sample. Several methods have been developed to address these limitations, but each has its own strengths and weaknesses. A highly bent silicon x-ray analyzer in the Laue geometry has some notable advantages compared to other detection methods. This paper discusses experiments that demonstrate how a bent Laue analyzer (BLA) may be used to resolve the L fluorescence x rays of neighboring actinide elements, permitting measurements that would be either difficult or impossible using other techniques.

ANALYZER DESIGN

The design concept of the BLA has been discussed previously.[1,2] The essential property of such an analyzer is that bending the crystal solves the mismatch between the narrow acceptance angle of a perfect crystal and the large divergence of fluorescence from the sample.[1] The shape needed to preserve the correct angle of incidence between the fluorescence x rays and the diffraction planes over a wide angle is a logarithmic spiral.[3,4] This is represented in polar coordinates by

$$r(\theta)=\rho_0\cos(\chi-\theta_B)\exp[\tan(\chi-\theta_B)\theta], \tag{1}$$

where ρ_0 is the bending radius of the crystal at $\theta=0$, χ is the asymmetry angle between the reflecting planes and the surface normal, and θ_B is the Bragg angle of the reflection planes for the x-ray wavelength of interest. Except for the case in which χ is equal to θ_B and the logarithmic spiral becomes a circular section, the correct form is difficult to produce accurately with dynamical bending.[1] Therefore, we have used a fixed bender form, a computer-numerical-control (CNC)-milled aluminum block, over which the silicon wafer has been bent.

In addition to matching the Bragg condition over a large angle, bending the crystal increases the bandwidth, or rocking curve width, making the analyzer nearly an ideal adjustable bandpass filter, with the exceptions of only absorption and a lowered reflection efficiency for extreme bending. For a highly strained silicon wafer (in the kinematical limit), the expected resolution of the BLA for a reflection can be written as

$$\Delta\theta(T)=\frac{T}{\rho}\left[\tan(\chi\pm\theta_B)+\frac{1}{2}(1+\nu)\sin 2\chi \mp\tan\theta_B(\cos^2\chi-\nu\sin^2\chi)\right], \tag{2}$$

where T is the thickness of the wafer and ν is the Poisson ratio.[5] This formula clearly breaks down as ρ approaches infinity (i.e., a flat crystal) and the bandwidth, $\Delta\theta$, approaches the intrinsic linewidth of a perfect crystal. The energy resolution is

$$\frac{\Delta E}{E}=\Delta\theta\cot(\theta_B). \tag{3}$$

Since the BLA is nonfocusing, a large area detector is required. The diffracted x-rays are scattered by $2\theta_B$ relative to the x rays incident upon the analyzer. For an analyzer of any reasonable size, this results in overlap of the diffracted x rays with the x rays coming directly from the sample. Therefore, Soller slits must be placed between the analyzer and the detector to absorb radiation from the sample source point. A key to the BLA's ultimate success lies in the correct implementation of the Soller slits.

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