X-ray Absorption Spectroscopy experiments using Crystal Analyser Spectrometer

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Introduction

A crystal analyser spectrometer (CAS) is under development on CRG-FAME (ESRF, Grenoble), based on spherically bent silicon crystals developed for Resonant Inelastic X-ray Scattering experiments [1]. The main aims of this development on a X-ray Absorption Spectroscopy beamline are the possibility to perform i) XAS studies on elements diluted in strongly fluorescent matrix, ii) site-selective XAS studies in mixed-valence compounds [2] and iii) high-resolution XANES studies [3]. For this purpose, we choose to use spherically bent crystals with a 0.5m radius of curvature in the Rowland geometry.

Methods and Materials

Tests have been performed using a Si(444) bent crystal. Its intrinsec energy resolution has been measured with the Kα fluorescence peaks of metallic copper (Fig. 1). Assuming a lorentzien shape of the measuring peaks, the total resolution is the sum of the natural fluorescence width of the selected emission, the monochromator Darwin width and the analyser resolution. These values are gathered in Tab. 1. The estimated CAS resolution is then found around 1.5±0.1 eV.

This resolution is larger than the one needed for RIXS experiments, around 200meV [1] but much smaller than the resolution of an energy resolved solid Ge detector (around 200-300eV, depending on the shaping time). The fluorescence line delivered by a selected diluted element can then be precisely separated from the other emission lines (issued for example from the major elements).

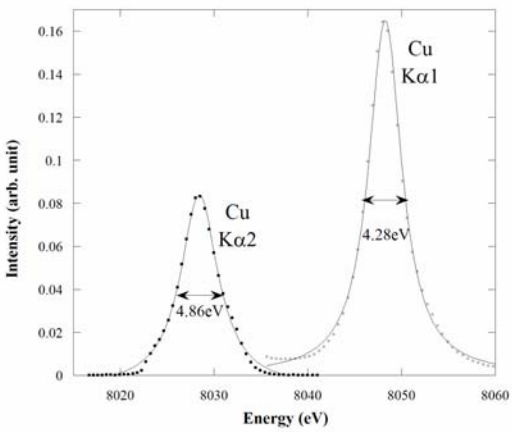

Fig. 1 Cu Kα Fluorescence experimental peaks

Fluorescence line	Cu Kα1	Cu Kα2
Fluorescence peak width (W_{peak})	4.28±0.05	4.86±0.05
Natural fluorescence width (W_{fluo})	2.28±0.05	2.78±0.05
Monochromator Darwin width (ω_D)	0.46	0.46
CAS resolution (W_{peak}-W_{fluo}-ω_D)	1.5±0.1	1.6±0.1

Tab. 1 Parameters involved in the CAS resolution estimation

Results

XAS acquisitions have been performed with this CAS on liquid reference compound ($CuSO_4$ aqueous solution, [Cu]=1%), in transmission mode or by selecting either the Cu Kα1 or Kα2 fluorescence lines (Fig. 2).

EXAFS oscillations are similar in the three measurements. The main differences are clearly visible in the pre-edge area. The energy resolution is clearly improved in the "high-resolution" fluorescence measurements in comparison with the transmission one. This improvement is also linked to the appearance of small differences in the edge features, depending of the probed transition, 1s to $2p_{3/2}$ (Kα1) or 1s to $2p_{1/2}$ (Kα2).

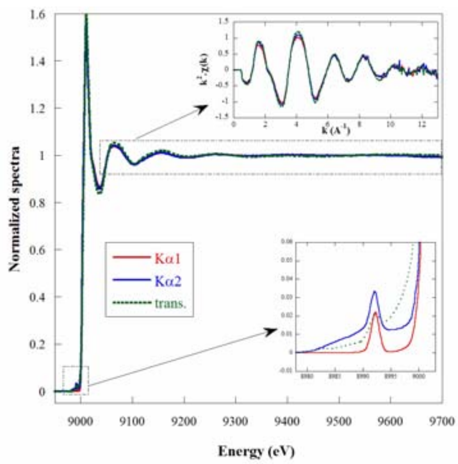

Fig. 2. XAS spectra measured in transmission and fluorescence modes. Insets: $k^2\chi(k)$ oscillations and pre-edge features. Counting rate: 100 000 (Kα1) and 50 000 (Kα2) counts/sec

Discussion

The quality of the obtained spectra, both in the EXAFS (good signal-to-noise ratio) and XANES (increase of the energy resolution compare to the transmission signal) areas is really correct and offers promising perspectives for the near future crystal analyser on CRG-FAME. This spectrometer will be really complementary of our 30-elements solid state Ge Canberra detector.

Acknowledgements

We thank Abhay Shukla (IMPMC, Paris) for the load of the silicon bent crystal.

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