ESRF

Experiment title:

EXAFS in molten fifth period metals

Experiment number:

HC359

Beamline: BM29-BL18

Date of Experiment: from: 14-Feb-1996 to: 16-Feb-1996

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Shifts: 9

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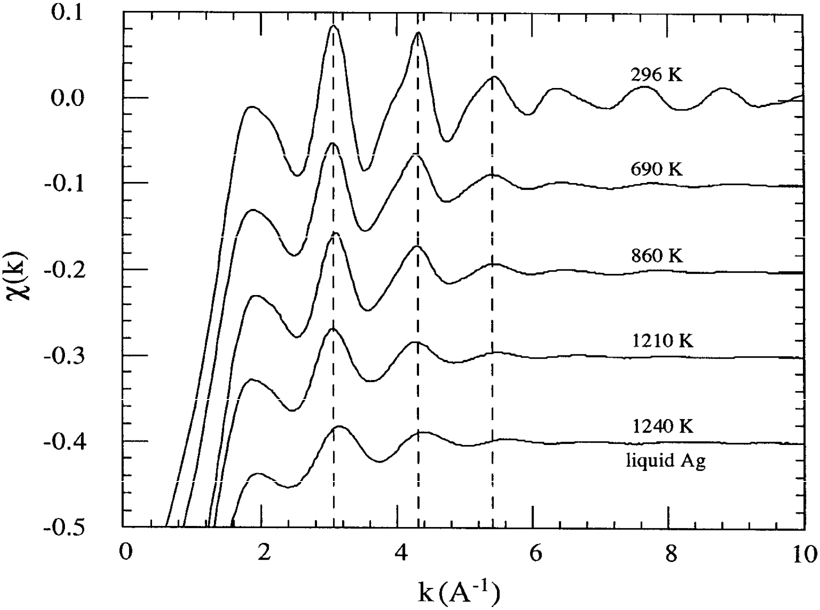
Report:

The first experiment concerning proposals HC359 has been performed during 9 shifts in 16 bunch mode in the first 1996 run. 3 shifts were used to install the high temperature oven and optimize the experimental setup for high energy operation. This included the installation and optimization of new photodiode detectors suitable to work in the 15-30 keV range.

The monochromator was equipped with a pair of flat Si(111) crystals that are able to provide flux up to 27.5 keV with excellent resolution. Typical beam size defined by entrance slits was 5×0.2 *mm.* Harmonic rejection was performed by crystal detuning. Due to the exceptional mechanical stability the operation with a fixed voltage on the piezo was sufficient to maintain a roughly constant detuning setting of about 50% through the scan (up to 2 keV in length).

The experimental setup and procedures were similar to those described in previous publications [1,2]. During the available beamtime we were able to perform a complete experiment on Ag at the Ag K-edge 25.51 keV. Solid Ag has been measured at several closely spaced temperatures between RT and the melting point T_m = 961.9 ‘C. Liquid Ag was measured up to 200 ‘C above T_m and up to 200 ^{0}C below T_m under supercooled conditions. The exceptional performances of the photo diode detectors allowed us to collect extremely low-noise spectra in all of the temperature range under study. This was achieved in spite of the absorption from windows, sample holder and intrinsic sample inhomogeneities.

The melting transition was unambiguously identified as a clear discontinuity in the absorption spectrum. The temperature readout resulted stable and accurate within ±10 “C. On cooling the temperature below T_m supercooling effects were clearly observable. The sharpness of the melting transition guarantees that the sample purity was maintained during the experiment. This was achieved thanks to the high vacuum conditions and reducing sample environment.



Some of the collected spectra are shown in the figure, the upper four spectra refer to crystalline Ag at increasing temperatures. The thermal effect that induces a reduction of the signal intensity in the high k region is quite evident (EXAFS Debye- Wailer). The bottom spectrum instead refers to liquid Ag just above T_m, basically at a temperature very close to the measurement of the hottest solid. The two spectra are remarkably different especially in the phase of the oscillation indicating remarkable changes in the average first neighbor distance and asymmetry.

The interpretation of these spectra will be performed along the lines described in recent publications [2,3,4,5,6] on the basis of a comparison between experimental and theoretical signals. In particular the quality of these data will allow us to retrieve reliable information on the average local environment, including asymmetries in the nearest neighbor distribution [2]. For the liquid sample reliable information on the radial distribution function $g(r)$ will be obtained up to about r=4-5 Å [2,3].

Present measurements provide a unique insight into the microscopic interactions in Ag. The possibility to determine accurate structural details and to follow the onset of the enharmonic behaviour in the solid, measuring directly the first neighbor distribution as a function of temperature, are characteristic of this spectroscopy. This physical observable is the one that directly reflects the asymmetry of the interatomic potential. The possibility to follow the melting transition and perform exactly the same data analysis on solid phase and liquid phase spectra is also a unique characteristic of XAS,

Present results prove the feasibility of these kind of EXAFS experiments on the high temperature setup available at BL18 and encourage the submission of further proposals along the same research line.

[1] A. Filipponi and A. Di Cicco, Nucl. Inst. & Methods for Phys. Res. B 93,302 (1994).

[2] A. Filipponi and A. Di Cicco, Phys. Rev. B **51, 12322 (1995).**

[3] A. Filipponi, J. Phys.: Condensed Matter 6,8415 (1994).

[4] A. Filipponi, A. Di Cicco, and C. R. Natoli, Phys. Rev. B 52,15122 (1995).

[5] A. Filipponi and A. Di Cicco, Phys. Rev. B 52,15135 (1995).

[6] A. Filipponi, J. Phys. Condensed Matter 7,9343 (1995).