X-ray Emission Spectroscopy at High-Pressure High-Temperature Conditions

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The spin-state of iron in Earth-forming iron bearing phases is important for understanding processes in the deep Earth. Many high-pressure studies of the spin state of iron in iron-bearing oxides have been conducted in recent decades, some involving x-ray emission spectroscopy (XES) [1-4], whereas others employed Mössbauer spectroscopy [3, 5,6]. However, only a few studies have been performed at high-temperature conditions [7, 8] due to the technical difficulties.

Here we report our recent success in further developing the capability to perform XES at high-pressure and high-temperature conditions, using a combination of a diamond anvil cell and double-sided laser-heating. The experiments were conducted at the 13-IDD GeoSoilEnviro Center for Advanced Radiation Sources (GSECARS), at the advanced Photon Source (APS), Argonne National Laboratory (ANL). This beamline is designed high-pressure high-temperature diamond anvil cell x-ray diffraction (XRD) experiments. Due to the low energy used in XES experiments, a Be gasket with a cBN-epoxy insert was used and the signal was collected through the gasket.

XES spectra of the $K_{\beta 1,3}$ fluorescence lines were collected at different *P-T* conditions using a curved silicon 440 analyzer, with a nominal spherical diameter of ~1 m in a Rowland circle spectrometer geometry. A Pilatus 100k Si detector was used in a fixed position. Calibration of the analyzer angle was performed using a wire of Fe, assuming that the main peak is at 7058 eV, corresponding to an angle of 66.18°. The Fe atoms were excited using an x-ray energy of 10.75 keV. Typical spectra were collected in the range of 7018-7078 eV with steps of approximately 0.3 eV. At each energy, the data was collected multiple times (up to 10), each time for up to 10 seconds, during both increase and decrease of analyzer energy. For collection of XRD data, the excitation energy was changed to 25 keV.

Python software was developed to control the experiment parameters, automatically select the rectangular region of interest (ROI) on the detector corresponding to the maximum signal for each analyzer angle, and preview the integrated signal as it is being collected. Both the raw images and the integrated spectra are saved. Additional Python software was developed for complete analysis of the data, including reintegration of the raw images with custom-shaped ROIs and an option for background subtraction from a secondary ROI.

An example of XES data collected at pressures above 1 Mbar, and temperatures up to 2500 K will be shown.

References:

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