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ESRF

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

http://193.49.43.2:8080/smis/servlet/UserUtils?start

Reports supporting requests for additional beam time

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Rehybridization of atomic orbitals under pressure: A high-pressure Compton-scattering study on Silicon (2)	**Experiment number**: HE-2132
Beamline:	**Date of experiment**: from: 19.06.2006 to: 26.06.2006	**Date of report**:
Shifts:	**Local contact(s):** **Thomas Buslaps**	*Received at ESRF:*

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Report:

Aim of this experiment was to show the feasibility of high pressure Compton scattering experiments along with the study of the rehybridization of Silicon atomic orbitals under pressure using a diamond anvil cell sample environment. On the basis of the preliminary Silicon high pressure Compton scattering experiment conducted at beam line ID15a of the European Synchrotron Radiation Facility (see J.S. Tse, D.D. Klug, D.T. Jiang, C. Sternemann, M. Volmer, S. Huotari, N. Hiraoka, V. Honkimäki, and K. Hämäläinen, Appl. Phys. Lett. **87**, 101905 (2005)) we decided to change the sample system from Silicon to Germanium. The latter of course has very similar physical properties and undergoes the same phase transitions as found in the case of Silicon (H. Katzke, U. Bismayer, and P. Toledano, Phys. Rev. B **73**, 134105 (2006)) so that an analogue rehybridization of atomic orbitals can be expected. The main reason for this decision was that a much stronger contrast of the Compton scattering signal between gasket and pure sample can be achieved. Additionally, the Germanium K-edge at 11103 eV can be used much better as indication for a proper subtraction of the gasket background due to the fact that the K-edge is well separated from the quasi-elastic line within the Compton spectrum in contrast to the K-edge of Silicon at 1839 keV.

The experiment was accomplished at beam line ID15a of ESRF. The incident radiation was monochromatized by fixed-exit Laue monochromators to 81 keV and the incident

beam was focused to 7 µm vertically and 50 µm horizontally by Aluminium compound x-ray refractive lenses. This way the focus of the incident radiation was restricted to the small sample volume within the diamond anvil cell. The scattered radiation was collimated by an adjustable lead slit of 0.3 mm width placed very close to the sample position to reduce parasitic scattering from the sample environment. The radiation was detected by means of a liquid nitrogen cooled Germanium multielement detector at a scattering angle of 154° . Overall a momentum space resolution of 0.45 a.u. was achieved with this setup. High purity polycrystalline Germanium powder was loaded into a Mao-Bell modification of a Merrill- Basset diamond anvil cell. Ruby chips were placed on top of the sample as pressure calibrants and during the experiment the pressure was measured ex-situ by the standard ruby fluorescence method. The alignment of the sample with respect to the incident beam was accomplished by measuring the transmitted intensitiy by a pin-diode. A more precise alignment was obtained in addition by taking diffraction patterns using a MAR 345 image plate detector. At each pressure Compton scattering spectra of the sample (together with the background signal from the gasket) and of the gasket itself were measured. Several single spectra were taken with accumulation times of 1800 s. During the experiment Compton profiles of Germanium were taken at ambient pressure, 1.2 GPa and 5.2 GPa. Due to problems with the diamond anvil cell, unfortunately, it was not possible to further increase the pressure as planned for the last 1.5 days of the experiment. Therefore, we decided to change the sample system again to measure Compton profiles of different clathrates we planned to use as backup samples in case something unexpected would happen with the high pressure setup. Measurements of these kind of samples will support our earlier studies on similar systems conducted at beam line ID15b of ESRF utilizing the high resolution Compton spectrometer.

Silicon clathrates are materials buildt up of Silicon nano-cages which are endohedrally intercalated with guest atoms (see e.g. A. San-Miguel and P. Toulemonde, High pressure research **25**, 159 (2005)). Due to the fact that only small portions of the powder samples were available, the focusing at this beam line was indispensible to conduct such kind of measurements so that the very same experimental setup was used to efficiently exploit the last 1.5 days of the beam time. The studied clathrates were Rb_6Si_{46}, Ba_8Si_{46} and Cs_8Sn_{44}. Moreover, Tin, Silicon and Si136 (a synthesized clathrate with almost empty Silicon nano-cages) Compton profiles have been measured as a reference for the analysis of the clathrate's spectra.

The high pressure Compton profiles will be analyzed as described by Tse et al. (Appl. Phys. Lett. **87**, 101905 (2005)) and compared with calculations using all electron first-principles local density method (CRYSTAL98) taking into account electron correlation. For the Compton profiles of the clathrates the valence electron contributions of the spectra will be extracted in a manner that guest-host interaction in these materials can be studied and will be compared with results we obtained on the systems Na_8Si_{46} and K_8Si_{46}. This comparison will be supported also by corrsponding calculations and accomplished with the focus on charge transfer from the guest atoms to the Silicon host lattice.