Experimental report

09/06/2017

Proposal:	**6-05-976**	**Council:**	4/2016
Title:	Structure of GeS2 Glass by NeutronDiffraction with Isotope Substitution		
Research area:	Materials		

This proposal is a resubmission of 6-05-957

Main proposer:	**Philip Stephen SALMON**
Experimental team:	Michela BUSCEMI Anita ZEIDLER
Local contacts:	Henry FISCHER

Samples: GeS2 with 70Ge and 73Ge isotopes

Instrument	Requested days	Allocated days	From	To
D4	5	5	24/06/2016	29/06/2016

Abstract:

The method of neutron diffraction with isotope substitution will be used to measure the full set of partial structure factors for the prototypical chalcogenide glass GeS2, which is used in the fabrication of optoelectronic and non-volatile memory devices. The results will be used to differentiate between the various models that have been proposed for GeS2 glass, a necessary first step in identifying its structure-function relationships. The results will also be used in the search for suitable protocols for simulating the structure of GeS2 glass by using first-principles molecular dynamics simulations, and will provide a firm basis for interpreting the structural transformations that occur to GeS2 glass under high pressure and temperature conditions.

Atomic scale structure of GeS_2 glass

Exp. No.	6-05-976
Dates:	24/06/2016 to 28/06/2016
Experimental Team:	Anita Zeidler (University of Bath)
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Local Contact:	Henry Fischer
Instrument:	D4C

GeS_2 is one of a family of network glass-forming systems with the MX_2 stoichiometry (M = Si, Ge; X = O, S, Se) that provide a backbone matrix for materials with multiple applications. GeS_2 is used, for example, in the fabrication of optoelectronic devices on account of its high nonlinearity and photosensitivity [1-3], and as the basis of solid state electrolytes in non-volatile memory devices [4-7]. In order to understand the properties of these glasses, it is first necessary to establish the atomic-scale structure. Accordingly, there have been many experimental [8-15] and theoretical [16-20] investigations of GeS_2 glass, which has long been regarded as an important test case for examining contrasting models for topological disorder, especially as the crystal structures form both two-dimensional (2D) and three-dimensional (3D) networks under ambient conditions [21]. The structure of the glass has not, however, been established. In particular, first-principles molecular dynamics simulations of GeS_2 glass using different approaches lead to different conclusions as to the existence or not of a significant fraction of homopolar (or "wrong") Ge-Ge and S-S bonds [16-20]. Thus, there is a need for unambiguous experimental information on the partial pair distribution functions, $g_{\alpha\beta}(r)$, for GeS_2 glass.

We decided to use the method of Ge isotope substitution to obtain unambiguous information on the partial structure factor level for GeS_2. We prepared 4 samples of GeS_2 which were identical in every respect apart from their isotopic enrichment in Ge. Specifically, we prepared $^{70}GeS_2$, $^{73}GeS_2$, $^{Nat}GeS_2$, and a sample containing a 50/50 mixture of the two isotopes, $^{Mix}GeS_2$. Using the D4c instrument with an incident wavelength of 0.4989(1) Å, diffraction patterns were then measured of the samples in their container, the empty container, the empty instrument, and a vanadium rod for normalisation purposes. The results will be used to inform molecular dynamics simulations.

During the experiment the polishing of the Tanzboden lead to a misalignment of the detectors, compromising the reproducibility of the background scattering and hence the accuracy of the data obtained to be able to make a full analysis. However, an example of a first order difference function where the sulphur-sulphur correlation is removed by taking the difference between $F_{70GeS2}(q)$ and $F_{73GeS2}(q)$ is shown in Fig. 1.

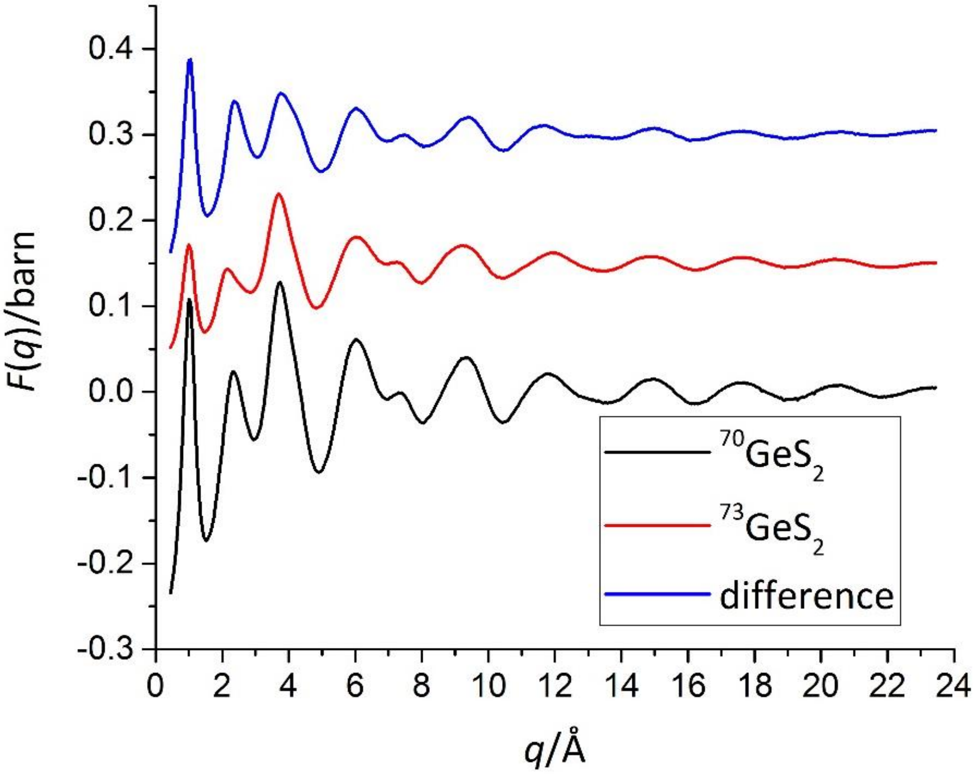

Fig. 1: *The total structure factors for two of the samples and their difference function.*

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