Experimental report

01/12/2015

Proposal: **6-03-433**

Council: 4/2014

Title: Short-range order in binary glass forming Zr-based melts

Research area: Materials

This proposal is a continuation of 6-03-422

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Samples: Zr50Ni50
Zr50Ni50-60
Zr2Fe
Zr50Ni50-58
Cu64.5Zr35.5

Instrument	Requested days	Allocated days	From	To
D20	5	4	16/12/2014	20/12/2014

Abstract:

The short-range order in metallic liquids plays a fundamental role in governing the physical properties of the melt. Phenomena such as diffusion, viscosity, solidification and glass-formation are all linked to the interplay between melt structure and dynamics. Although pronounced short-range order is found in many bulk metallic glass-forming systems, the exact correlation between short-range order and glass-forming ability is still not well understood. Thus, we intend to study of the short-range order in glass-forming ZrNi, ZrCu and ZrFe melts. Using isotope substitution and neutron diffraction we intend to measure the partial static structure factors of electrostatically levitated Zr50Ni50 droplets on D20, as well as the temperature dependence of the static structure factors of the Cu65Zr35 and Zr2Fe liquids. These experiments are of fundamental importance in exploring the effect of chemical short-range order on atomic transport phenomena in Zr-bearing liquids. The results of these studies will be analyzed within the framework of mode-coupling theory, in order to establish a more detailed understanding of the structural origin of glass-forming ability.

The interplay between structure and dynamics in undercooled liquids is a key to understanding atomic transport processes, as well as solidification and glass-formation from the melt. Despite extensive study, the correlation between short-range order of the melt and glass-forming ability is still not well understood. In particular, definitive correlations between glass-forming ability, physical properties and short-range order of the melt require determination of partial structure factors in both the stable and metastable liquid states, as well as over a large compositional range.

In the proposal 6-03-433 in December 2014, we set out to continue our previous investigations mainly on the short-range order in the binary Zr-Ni system [1] (here $Zr_{50}Ni_{50}$) using the container-less processing technique of electrostatic levitation (ESL). During this experiment, we achieved the necessary scattering contrast through isotopic substitution of Ni, such that a determination of the Faber-Ziman partial structure factors is possible. Figure 1 shows the static structure factors S(Q) in the equilibrium liquid at 1560 K of $Zr_{50}Ni_{50}$, measured using our ESL furnace. Each S(Q) data set represents one measurement taken at this temperature with different Ni isotope – i.e. ^{nat}Ni, ^{60}Ni and ^{58}Ni.

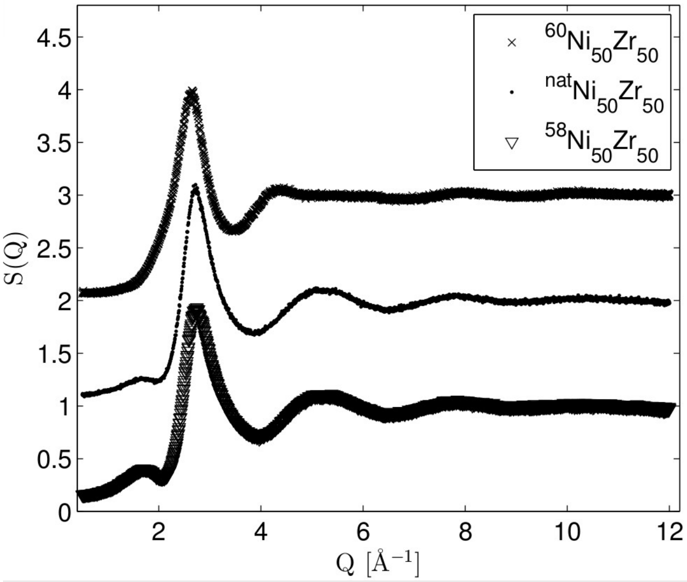

Figure 1: Static structure factors S(Q) of isotopically substituted samples of $Ni_{50}Zr_{50}$ measured on D20 at a temperature of 1560 K using the electrostatic levitation furnace. The top two curves have been shifted along the vertical axis by 1 and 2 units, respectively. The pre-peak apparent around 1.8 A^{-1} is a consequence of a chemical ordering in the melt.

We were able to carry out measurements at 5 different temperatures for all isotopic compositions of $Zr_{50}Ni_{50}$, over a temperature range of some 300 K, which encompasses both the equilibrium and undercooled liquid states.

Additional measurements were carried out on the composition $Cu_{64.5}Zr_{35.5}$, both in the equilibrium and undercooled liquid. Due to the weak scattering contrast available with Cu isotopes, these measurements will be combined with synchrotron X-ray diffraction measurements, allowing us to the determine the Bhatia-Thornton partial structure factors S_{NN} and S_{NC} with good precision [2]. Partial structure factors on this composition are particular useful, as $Cu_{64.5}Zr_{35.5}$ can be cast into bulk glassy samples, with a critical diameter of 2 mm. On the other hand, Zr-Ni is marginal glass-forming system, having a critical thickness limited to thin ribbons on the order of some tens of micrometers. Thus, by comparing the different partial structure factors in these systems, it is our aim to better understand the connection between short-range order and glass-forming ability.

As the proposed Zr_2Fe composition was not able to remain stable under levitation using ESL, we decided to investigate the composition $Hf_{35}Ni_{65}$ instead. The S(Q) taken in the equilibrium liquid at 1520 K is shown in Fig. 2. Despite the much larger absorption cross-section of Hf (104.1 barns), we were nevertheless able to achieve reasonable data quality within an experimental run of ~ 30 min/temperature. This experiment thus proves the feasibility of performing future measurements on the Hf-Ni system by employing isotopic substitution of Ni. As Hf is chemically similar to Zr, it is interesting to investigate whether a similar structure-dynamics interplay exist compared to Zr-Ni. Furthermore, the incoherent neutron scattering cross-section of 2.6 barns allows an investigation of the Hf self-dynamics, using quasielastic neutron scattering, which which we have already undertaken. This is not possible with e.g. Zr-Ni, due to the negligible incoherent cross-section of Zr.

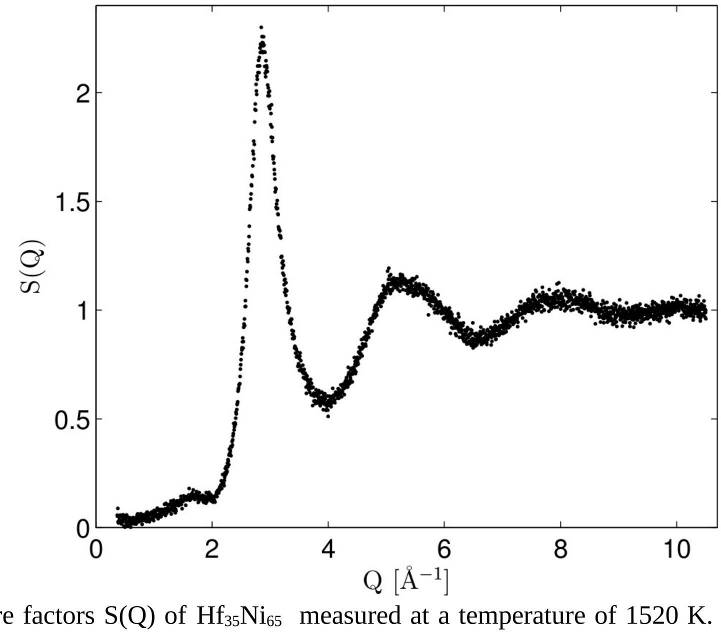

Figure 2: Static structure factors S(Q) of $Hf_{35}Ni_{65}$ measured at a temperature of 1520 K. Similar to ZrNi, a pre-preak arises due to a chemical ordering in this melt.

Altogether, the proposed scientific objectives were met with minimal experimental difficulties. The results obtained from this proposal will be integrated into our currently running studies to investigate the structure-dynamics relations in metallic melts using neutron and X-ray scattering combined with container-less measurements of thermophysical properties [3–5].

[1] T. Kordel, D. Holland-Moritz, F. Yang, J. Peters, T. Unruh, T. Hansen, and A. Meyer, Phys. Rev. B **83**, 104205 (2011).

[2] D. Holland-Moritz, F. Yang, T. Kordel, S. Klein, F. Kargl, J. Gegner, T. Hansen, J. Bednarcik, I. Kaban, O. Shuleshova, N. Mattern, and A. Meyer, EPL **100**, 56002 (2012).

[3] C. C. Yuan, F. Yang, F. Kargl, D. Holland-Moritz, G. G. Simeoni, and A. Meyer, Phys. Rev. B **91**, 214203 (2015).

[4] S. Klein, D. Holland-Moritz, D. M. Herlach, N. A. Mauro, and K. F. Kelton, EPL **102**, 36001 (2013).

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