	Experiment title: Time-resolved evolution of SRO/MRO/LRO of Fe-based bulk metallic glasses with Invar effect	**Experiment number**: HC 4361
Beamline: ID15A	**Date of experiment**: from: 02/06/2021 to: 07/06/2021	**Date of report**: 09/08/2021
Shifts: 15	**Local contact(s)**: Gavin Vaughan	*Received at ESRF:*

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Report:

Invar effect, an anomalosly low coefficient of thermal expansion (CTE) below the Curie temperature characteristic to $Fe_{65}Ni_{35}$ ordered fcc solid solution, was observed also in the case of ferromagnetic bulk metallic glasses (BMGs). While its origin is still a matter of debate, existing explanations for crystalline materials are built on the fcc lattice. Even though BMGs lack any long-range order they universally show the Invar effect.

The volume transformation at the Curie temperature is fully reversible but it is completely unknown whether it is also accompanied by a structural rearrangement. The key point is to investigate whether local atomic rearrangement is taking place at the Curie temperature and if so, characterize it and relate it to the macroscopically observed Invar effect.

During this beamtime we performed Small-angle X-ray scattering (SAXS) and Wide-angle X-ray scattering (WAXS) to cover the full size range of any potential clustering (0.1 nm to 25 nm). Through analysis in real-space (derived from WAXS) it is possible to identify the variation in local arrangement and its change at the Curie temperature, while the SAXS profile gives information about the correlation of cluster arrangements on the large scale.

We measured a total of 5 BMGs $(Fe_{71.2}B_{24}Y_{4.8})_{96}Nb_4$, $(Fe_{73.2}B_{22}Y_{4.8})_{95}Mo_5$ and $(Fe_{0.71}X_{0.05}B_{0.24})_{96}Nb_4$ with X = Ho, Er, Tm) and two crystalline samples (pure Fe and $Fe_{64}Ni_{36}$ = Invar36). The BMGs and Invar36 was measured as thin disk (< 1mm thickness) while Fe was measured in the shape of a wire. All samples were

thermally cycled in a Linkam heating stage with the sequence 20°C → 400°C → 20°C → 550°C or 600°C → 20°C at 5 K/min for both SAXS and WAXS. For SAXS the MAXIPIX detector was used, while the WAXS was recorded with the PILATUS detector. SAXS was performed with a beam energy of 90keV and WAXS was done at a beam energy of 87keV.

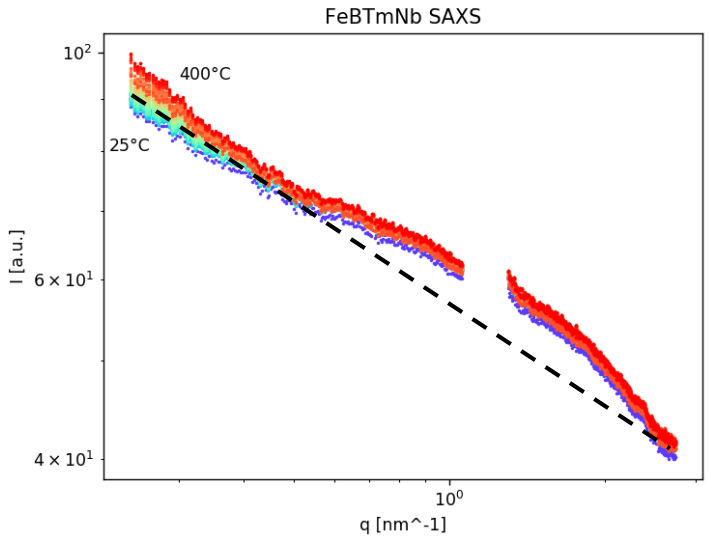

Figure 1: SAXS profile of FeBTmNb at various temperatures ranging from 25°C (blue) to 400°C (red).

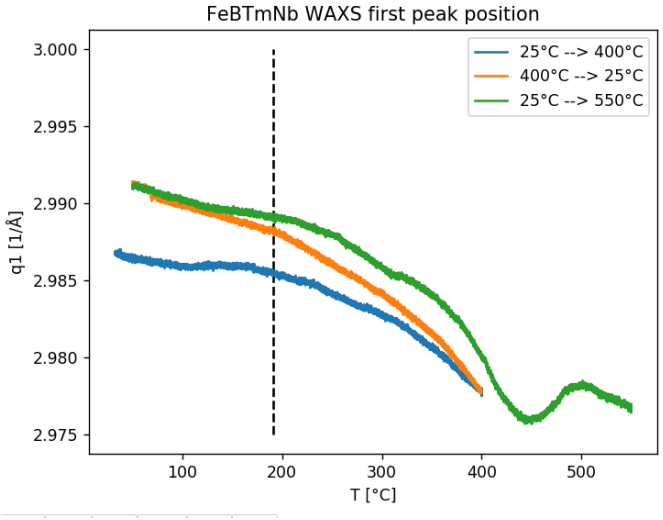

Figure 2: First peak position in the WAXS profile of FeBTmNb. The dashed line marks the Curie temperature.

Figure 1 shows the SAXS profile of the FeBTmNb alloy at various temperatures. It shows a temperature dependence at low q and a mostly temperature independent peak at $q \approx 1\ \mathrm{nm}^{-1}$. The thermal expansion of the alloy can be measured by XRD through the shift of the first (and also higher order) diffraction peaks. Figure 2 shows the position of the first diffraction peak of the FeBTmNb alloy. The slope is proportional to the CTE. It can be clearly seen that at the Curie temperatre (dashed line), the CTE suddenly increases. Moreover, as the heating cycle progresses, the peak position increases which corresponds to the reduction of free volume in the sample.

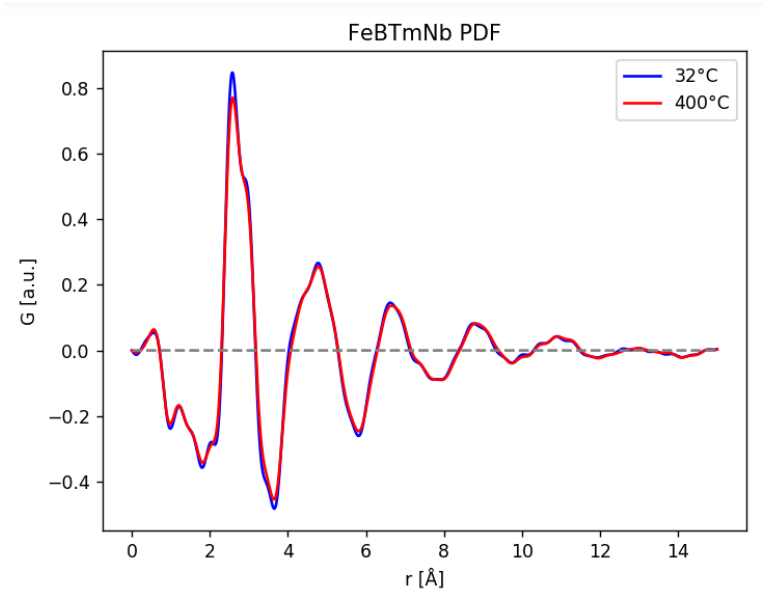

Figure 3: PDF of FeBTmNb below Curie temperature (blue) and above Curie temperature (red).

The pair-distribution function (PDF) of the FeBTmNb alloy can be calculated from the WAXS diffraction profile. It is shown in Figure 3 for a representative temperature each, below and above the Curie temperature of the alloy. The most pronounced effect of the volume transition is seen in the first atomi shell at around $r = 3$ Å, but also higher order shells are affected.

The experimental data is currently under analysis and a scientific publication is currently in preparation.