Nanocrystallization of $25K_2O$-$25Nb_2O_5$-$50GeO_2$ glass studied by SAXS/ASAXS

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In recent years, there is an intensive search for transparent nanocrystallized glasses, because such materials have a high potential for optical applications. Very recently, it was found that a glass with the composition $25K_2O$-$25Nb_2O_5$-$50GeO_2$ ($KNbGeO_5$) shows prominent nanocrystallization and that nanocrystallized glasses show an unusual Vickers hardness [1].

The glass transition temperature of the $KNbGeO_5$ glass is T_g = 622 °C. Crystallized glasses obtained by heat treatments at 620-720 °C for 1 h keep good transparency. The only nanocrystalline phase in the heat-treated glass samples was identified to be $K_{3.8}Nb_5Ge_3O_{20.4}$ by X-ray diffraction (XRD). The glass density increases gradually with increasing heat treatment temperature.

According to the XRD patterns, the quenched base glass is entirely amorphous. TEM images of the crystallized glass obtained by a heat treatment at 720 °C for 1 h show a large number of nearly spherical nanocrystals about 20-30 nm in size. The aim of the present SAXS / ASAXS study, performed on the same glass samples as reported in [1], is a more detailed investigation of the nucleation and crystallization behaviour of the $KNbGeO_5$ glass. In particular, the ASAXS measurements at the K-edges of Ge and Nb were performed in order to get information about potential size dependence of the scattering contrast of the nanocrystals. Such information can be very important for the verification of certain predictions made in a newly developed approach to nucleation theory [2].

SAXS and ASAXS experiments were performed using the JUSIFA instrument at HASYLAB. Two samples, one as prepared (sample 3) and a sample annealed at 720 °C for 1 h (sample 1), were measured. The samples were thinned and polished to a thickness of about 90 µm. The SAXS curves of both samples in Fig. 1 show pronounced shoulders at Q-values of about 0.4 nm^{-1}. The differentiell volume size distributions were calculated (Fig. 2) by means of a non-linear least-square fitting program using a log-normal size distribution of spheres. The averaged sizes are 3.3 nm (as quenched) and 3.8 nm for the annealed sample. Therefore, SAXS reveals particles contained in the quenched sample already, for which XRD yielded amorphous behaviour.

To clarify the nature of the scattering effect at small values of Q, we performed ASAXS at different energies below the K-edge of the element Ge. The curves in Fig. 3 show an energy dependent shape. That will allow to analyse the element specific composition fluctuation and to distinguish surface caused scattering effects from volume scattering.

References

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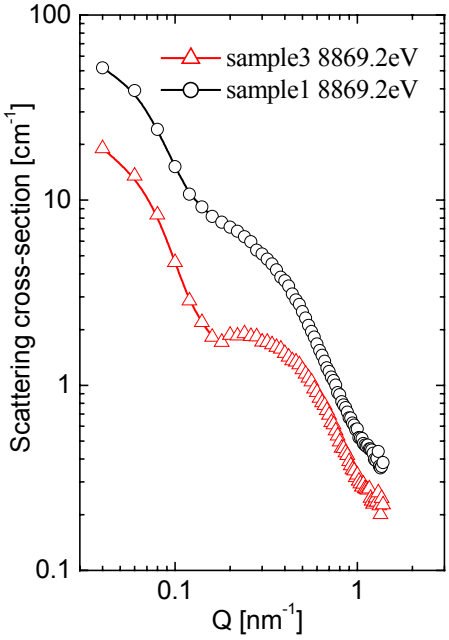

Figure 1: SAXS curves of both samples measured at 8869.2 eV far from all X-ray absorption edges of elements containing in the samples.

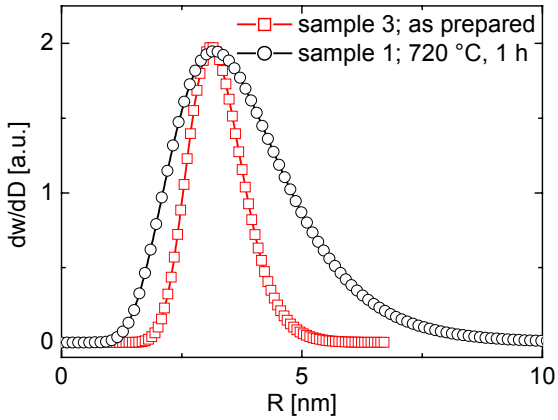

Figure 2: Differential volume size distributions derived from the SAXS curves in Fig. 1.

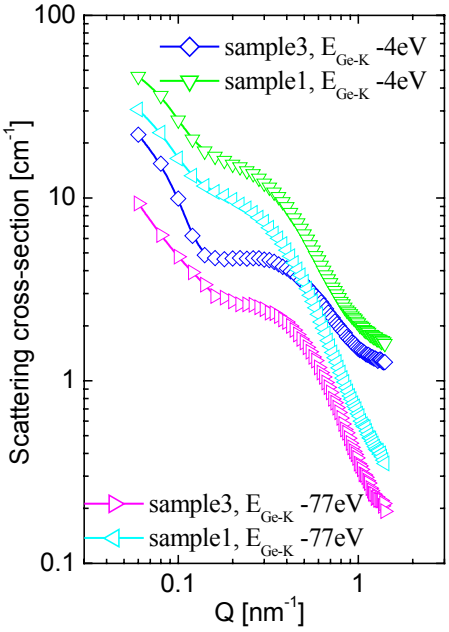

Figure 3: ASAXS contrast variation near the Ge-K edge.