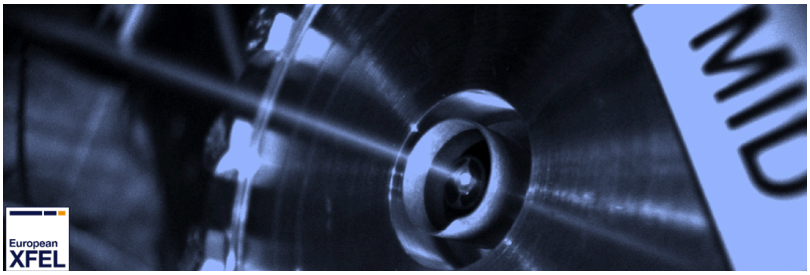

Contribution ID: **25** Type: **Talk**

Glass formation in phase-change materials: The impact of a liquid-liquid transition

Thursday 19 January 2023 09:15 (20 minutes)

Glasses are formed when liquids are cooled fast enough and far enough [1]. Both criteria must be fulfilled to avoid crystallization during cooling, because thermodynamically, the crystalline state is always favorable. Therefore, thermodynamics alone cannot explain glass formation and the limiting cooling parameters are determined by the kinetic properties of a supercooled liquid. That is why glass formation is often referred to as the "freezing-in"of a disordered liquid.

The underlying physical property of the supercooled liquid is the atomic diffusivity, whose temperature dependence varies greatly between different types materials. Pure fcc metals like copper crystallize even during rapid quenching due to a high diffusivity over a wide temperature range of supercooling that is only accessible in molecular dynamics simulations [2]. Several oxides, such as SiO2, are good glass formers and have a strongly temperature-dependent diffusivity, which enables glass formation even during slow cooling [3]. An intermediate behavior is observed in phase-change materials (PCMs), which are used in non-volatile electronic and optical memory devices [4]. The diffusivity of a common phase-change material, AgIn:(Sb2Te) in the moderate supercooling regime is high, but reduces rapidly below a certain temperature [5]–[7]. This behavior has previously been explained based on empirical models and is commonly referred to as liquid-liquid transition.

By using femtosecond X-ray diffraction, we have observed a structural change around this transition in the PCMs AgIn:(Sb2Te) and Ge15Sb85 [8]. The structural information indicates that a Peierls-like atomic distortion is formed below the transition. A structural order parameter is found to correlate with the activation energy of diffusivity, which establishes for three PCMs and related materials a relationship between kinetics and atomic structure. Ab-initio molecular dynamics simulations provide further insight into the mechanism and reveal the increase of covalency in the low-temperature state.

However, the magnitude of the distortion could not be quantified experimentally, due to the low momentum transfer of the X-ray scattering experiment. High photon energy FEL radiation provides an ideal means of investigating the disordered structure of these supercooled liquids by means of pair distribution function analysis. These states of matter cannot be prepared repetitively and require a single shot structure-determination. Liquid-liquid transitions were reported also for several other supercooled liquids and many of them exist only on the sub-nanosecond timescale before crystallization set in. Furthermore, the high photon energy will bring more elements in reach for anomalous scattering techniques, which is beneficial for the investigation of multicomponent materials.

[1] I. Gutzow and J. Schmelzer, The Vitreous State. Springer, 1995.

[2] H.-J. Lee, Y. Qi, A. Strachan, T. Cagin, W. A. Goddard, and W. L. Johnson, "Molecular Dynamics Simulations of Supercooled Liquid Metals and Glasses,"MRS Proceedings, vol. 644, p. L2.3, Mar. 2000, doi: 10.1557/PROC-644-L2.3.

[3] R. H. Doremus, "Viscosity of silica,"J Appl Phys, vol. 92, no. 12, pp. 7619–7629, Dec. 2002, doi: 10.1063/1.1515132.

[4] M. Wuttig and N. Yamada, "Phase-change materials for rewriteable data storage,"Nat Mater, vol. 6, no. 11, pp. 824–832, 2007.

[5] J. Kalb, M. Wuttig, and F. Spaepen, "Calorimetric measurements of structural relaxation and glass transition temperatures in sputtered films of amorphous Te alloys used for phase change recording,"J Mater Res, vol. 22, no. 3, pp. 748–754, 2007, doi: 10.1557/JMR.2007.0103.

[6] M. Salinga et al., "Measurement of crystal growth velocity in a melt-quenched phase-change material.,"

Nat Commun, vol. 4, p. 2371, 2013, doi: 10.1038/ncomms3371.
[7] J. Orava, H. Weber, I. Kaban, and A. L. Greer, "Viscosity of liquid Ag–In–Sb–Te: Evidence of a fragile-to-strong crossover,"J Chem Phys, vol. 144, no. 19, p. 194503, 2016, doi: 10.1063/1.4949526.
[8] P. Zalden et al., "Femtosecond x-ray diffraction reveals a liquid–liquid phase transition in phase-change materials,"Science (1979), vol. 364, no. 6445, 2019, doi: 10.1126/science.aaw1773.

Primary author: ZALDEN, Peter (Eur.XFEL (European XFEL))

Presenter: ZALDEN, Peter (Eur.XFEL (European XFEL))

Session Classification: Dynamics of functional materials

Track Classification: Dynamics of functional materials