Structure, thermodynamics, and relaxations in amorphous phase-change materials

Shuai Wei*
Department of Chemistry, Aarhus University, Denmark
*Email: shuai.wei@chem.au.dk

Amorphous phase-change materials (PCMs) (e.g. Ge-Sb-Te) belong to a family of functional materials for novel electronic and photonic applications. PCMs can be rapidly and reversibly switched between amorphous and crystalline phases. With large optical and electric property contrast between the two phases, they can be used to encode data for non-volatile memory, photonic, and neuromorphic computing devices. However, due to their disordered structures and poor glass forming ability, the amorphous phases are not well characterized, and a better understanding of structure-property relations is an urgency for the relevance of applications.

In this work, we demonstrate structure, thermodynamics, and relaxation behaviors of amorphous PCMs over 14 orders magnitude in timescale and a thousand degree of temperature. In amorphous solid, we show how thermal annealing and pressure may induce and diminish local distortions of defective octahedral structures (Peierls-like distortion), which drastically alter materials properties (e.g. stability, crystallization, compressibility, electronic density of states). Above the glass transition temperature, materials enter a metastable supercooled liquid state, where thermodynamic anomalies are uncovered and associated with dynamic crossovers and switching behaviors. We show the evidence of a liquid-liquid transition in the metastable state with femtosecond diffractions using X-ray free electron lasers (XFEL). At higher temperature above the melting point, the density fluctuations of liquid state exhibit ultrafast structural relaxations. We demonstrate an experimental strategy to directly observe atomic-scale relaxations using femtosecond X-ray photon correlation spectroscopy by harnessing the coherent X-rays of XFEL. The fast atomistic dynamics is underlying the low viscosity and high fragility behaviour of PCM liquids. We will also briefly discuss the Boson peak in low-temperature heat capacity and development of new PCM alloys. A better understanding of amorphous PCMs may lead to the tailoring of materials properties for novel applications in photonic, and neuromorphic computing devices.

References

[1] S. Wei, P. Lucas, and C. A. Angell, *Phase-Change Materials: The View from the Liquid Phase and the Metallicity Parameter*, MRS Bulletin **44**, 691 (2019).
[2] T. Fujita et al., *Pressure-Induced Reversal of Peierls-like Distortions Elicits the Polyamorphic Transition in GeTe and GeSe*, Nat Commun **14**, 1 (2023).
[3] Y. Cheng et al., *Highly Tunable β-Relaxation Enables the Tailoring of Crystallization in Phase-Change Materials*, Nature Communications **13**, 7352 (2022).
[4] P. Zalden et al., *Femtosecond X-Ray Diffraction Reveals a Liquid–Liquid Phase Transition in Phase-Change Materials*, Science **364**, 1062 (2019).
[5] S. Wei, P. Lucas, and C. A. Angell, *Phase Change Alloy Viscosities down to Tg Using Adam-Gibbs-Equation Fittings to Excess Entropy Data: A Fragile-to-Strong Transition*, J. Appl. Phys. **118**, 034903 (2015).
[6] S. Wei, *Anomalous Liquids on a New Landscape: From Water to Phase-Change Materials*, Journal of Non-Crystalline Solids: X **14**, 100094 (2022).

Shuai Wei is tenure-track Assistant Professor in Materials Chemistry and Centre of Integrated Materials Research at Aarhus University, Aarhus, Denmark. He is a VILLUM Young Investigator and a former AIAS Associated Fellow. His research interests are novel amorphous materials for innovative technologies, including phase-change materials (e.g. Ge-Sb-Te) for next generation non-volatile memory applications and 3D printed amorphous metals for structural applications.

Group homepage: https://chem.au.dk/AmorphousMatLab