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Breakdown of the Stokes-Einstein Relation Above the Melting Temperature in Liquid Phase-Change Materials

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The dynamic properties of liquid phase-change materials (PCMs), such as viscosity η and atomic self-diffusion coefficients D , play an essential role in ultrafast phase switching behavior of novel non-volatile phase-change memory applications. To connect η to D , the Stokes-Einstein relation (SER) is commonly assumed to be valid at high temperatures near or above the melting temperature T_m and is often employed for assessing liquid fragility (or crystal growth velocity) of technologically important PCMs. However, using quasi-elastic neutron scattering (QENS), we provide experimental evidence for a breakdown of the SER even at temperatures above T_m in the high-atomic-mobility state of well-known PCMs $\text{Ge}_1\text{Sb}_2\text{Te}_4$, $\text{Ge}_2\text{Sb}_2\text{Te}_5$, $\text{Ag}_4\text{In}_3\text{Sb}_{67}\text{Te}_{26}$ (AIST), and GeTe . This implies that although the viscosity may have strongly increased during cooling, the diffusivity can remain high due to the early decoupling, being a favorable for fast phase switching behavior of the high-fluidity PCM. We discuss the origin of the observation and propose the possible connection to a metal-semiconductor and fragile-strong transition hidden below T_m . In addition, the infinite-frequency shear modulus is experimentally determined ranging from 2 to 3 GPa for liquid PCMs, which permits extracting viscosity from microscopic structural relaxations usually accessible to simulations and scattering techniques.

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