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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 $Ge_1Sb_2Te_4$, $Ge_2Sb_2Te_5$, $Ag_4In_3Sb_{67}Te_{26}$(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 3GPa for liquid PCMs, which permits extracting viscosity from microscopic structural relaxations usually accessible to simulations and scattering techniques.

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