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The anomalous breakdown of the Stokes-Einstein relation in Ge-Sb-Te and Ag-In-Sb-Te alloys and its connection to fast crystallization in the supercooled liquid

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Phase-change materials such as Ge-Sb-Te and Ag-In-Sb-Te alloys can be reversibly switched between amorphous and crystalline states on a timescale of nanoseconds. These alloys possess unique features such as ultrafast switching, strong electrical/optical contrast, and a relatively stable amorphous state, which are interesting for non-volatile computer memory devices and neuromorphic computing applications. While significant progress has been made on the understanding of their crystalline phases and bonding natures, the liquid-state behavior (especially supercooled liquid state) has been less explored and its connection to crystallization kinetics needs to be better understood. Here we characterize the structural relaxation on the timescale of picoseconds and the self-diffusion coefficients of the Ge-Sb-Te and Ag-In-Sb-Te alloys in a high fluidity state above their melting points using quasi-elastic neutron scattering (QENS) on the time-of-flight (TOFTOF) spectroscopy. We find experimental evidence of an anomalous breakdown of the Stokes-Einstein relation in this high atomic mobility state. The breakdown shows the same features of the well-known anomalous liquid water . We also show that the origin of this breakdown is unlikely the dynamic heterogeneities that have been usually discussed in the deeply undercooled viscous liquid. Rather it may be related to a liquid-liquid transition (and possibly a liquid-liquid critical point) hidden below the melting temperature in the supercooled liquid state. The liquid-liquid transition in supercooled phase-change materials is also a semiconductor-metal transition and a fragile-strong transition. The anomalous liquid behavior may explain why this class of material has ultrafast crystallization kinetics at high temperatures, favorable for fast data read/write speed in memory devices, while its amorphous phase retains relatively stable at room temperature for long-time data retention.

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