

Structural relaxation on a bulk metallic glass forming Zr-Ti-Cu-Ni-Be liquid

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The microscopic and macroscopic dynamics of a bulk metallic glass forming liquid $\text{Zr}_{46.8}\text{Ti}_{8.2}\text{Cu}_{7.5}\text{Ni}_{10}\text{Be}_{27.5}$ (Vit4) were studied in a containerless processing environment employing electrostatic levitation (ESL). Together with quasielastic neutron scattering (QNS), this allows accurate determination of dynamical properties of the chemically highly reactive liquid over a temperature range more than 200 K in the equilibrium melt. In the QNS experiments, a microscopic mean structural relaxation time was obtained in the wavenumber range of $0.3\text{--}2.6\text{ \AA}^{-1}$. The viscosity of the freely suspended melt was measured using the oscillating droplet method.

The structural relaxation observed by QNS on Vit4 is dominated by self motion of the Ni/Ti/Cu at small wavenumbers. In contrast, at the wavenumbers close to the structural factor maximum of the liquid, it is largely contributed by the collective Zr motion. Despite of their different natures, the derived relaxation times of both, as well as the mean transport coefficient D of the Ni/Ti/Cu, are proportional to each other within in the measurement uncertainties over the entire investigated temperature range. Moreover, the relaxation times obtained by QNS are also proportional to the macroscopic viscosities at similar temperatures within the experimental error.

The proportionality between the different microscopic relaxation times and the macroscopic viscosity indicates that the relaxation processes on different time and length scales exhibit similar temperature dependencies. In addition, this leads to $D\eta \approx \text{constant}$, which is in contrast to the Stokes-Einstein (SE) relation. Similar behaviour has also been found in the binary $\text{Zr}_{64}\text{Ni}_{36}$ liquid. This observation is in line with the mode coupling theory (MCT) prediction. At temperatures close to the critical temperature T_c , MCT expects the temperature dependence of the relaxation times and viscosity follows scaling laws $\tau(T) \propto D^{-1} \propto \eta \propto (T - T_c)^\gamma$, resulting in asymptotically $D\eta = \text{constant}$. The MCT prediction is based on highly correlated atomic dynamics. It seems that even in the equilibrium melt of Vit4, more than 200 K above its T_c , the structural relaxation is still dominated by such kind of correlated motions, in the case of both mass transport and viscous flow.