On the Relation of Viscous Flow and Atomic Diffusion in Glass-Forming Metallic Liquids

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The dynamics in fluids is governed by two intimately related properties: viscosity and atomic diffusion. While the first describes macroscopic transport of momentum by collective motion of the particles, the latter describes single particle diffusive transport. A common relation, that is often taken for granted in order to calculate required diffusion coefficients of atoms or molecules in a liquid from the viscosity, or vice versa, is the Stokes-Einstein relation. This relation was derived in order to study the diffusive motion of a mesoscopic sphere in a viscous medium. However, when the diffusing objects are of atomistic size, deviations of the diffusion coefficient and the viscosity from the Stokes-Einstein behaviour can be observed.

From an experimental point of view, a direct proof of the Stokes-Einstein relation is still very difficult due to the lack of reliable experimental data of the transport coefficients. The accurate measurement of diffusion data, using long capillary methods, is subject to large errors due to additional transport of mass by buoyancy driven flow effects. Pollution of the sample from chemical reactions with the container walls complicates the measurement of both properties. In order to check the Stokes-Einstein relation measurements need to be carried out over a sufficiently large temperature range. This includes high temperatures favouring convection and chemical reactivity. With the development of advanced containerless processing techniques, such as electrostatic levitation (ESL), we are now in a position to master these challenges.

Using ESL, we carried out quasielastic neutron scattering (QNS) experiments and determined accurate self-diffusion coefficients, in liquid Zr-Ni and bulk glass-forming Zr-Ti-Ni-Cu-Be alloys. These data were measured over a broad temperature range from 1000 K to 1700 K. Viscosity of these liquids were also measured via the oscillating drop technique in ESL over a broad temperature range: With these results for a dense glass forming system, the relation of viscous flow and diffusion of mass could be checked in unequalled detail [1].

Our data show that the product of diffusion and viscosity is constant for the entire temperature range contradicting the Stokes-Einstein relation. According to Mode-Coupling-Theory (MCT), the dynamics in a liquid is strongly coupled when the particle density is large. This leads to a freezing of the atomic motion when MCT's critical temperature T_c is approached upon cooling. In this case, the diffusion coefficient and the inverse of the viscosity are proportional to the same scaling law. Our results have been obtained at temperatures well above T_c. Apparently possible deviations from the MCT scaling behaviour are similar with respect to temperature for both mass transport coefficients.

[1] J. Brillo, A. I. Pommrich, A. Meyer, Phys. Rev. Lett. **107**, 165902 (2011).