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Title

Rattling the cage: Micro- to mesoscopic structure in liquids as simple as argon and as complicated as water

Source

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Abstract

The water molecule has the convenient property that its molecular polarizability tensor is nearly isotropic while its dipole moment is large. As a result, the low-frequency anisotropic Raman spectrum of liquid water is mostly collision-induced and therefore reports primarily translational motions while the far-infrared (terahertz) and dielectric spectrum is dominated by rotational modes. Atomic and globular-molecular liquids have a zero dipole moment as well as an isotropic polarizability tensor. These spectrum-simplifying properties were exploited in a study of a number of liquids and solutions using ultrafast optical Kerr-effect (OKE) spectroscopy combined with dielectric relaxation spectroscopy (DRS), terahertz time-domain spectroscopy (THz-TDS), and terahertz field-induced second-harmonic generation (TFISH) spectroscopy. For room-temperature ionic liquids (RTILs), liquid water, aqueous salt solutions, noble gas liquids, and globular-molecular liquids it was found that, in each case, surprising structure and/or inhomogeneity is observed, ranging from mesoscopic clustering in RTILs to stretched-exponential dynamics in the noble gas liquids. For aqueous electrolyte solutions it is shown that the viscosity, normally described by the Jones-Dole expression, can be explained in terms of a jamming transition, a concept borrowed from soft condensed matter studies of glass transitions in colloidal suspensions. [All rights reserved Elsevier]. (114 References).