

Dynamics and Thermodynamics

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According to conventional wisdom, a supercritical fluid is one that doesn't exhibit distinct liquid- or gas-like states. This may need to be revised in light of measurements that show a sharp change in the speed of sound in supercritical argon when it crosses a well-defined line on its pressure versus temperature phase diagram. Our results have been recently reported in two papers appeared on *Physical Review Letters* and *Nature Physics*, in which we investigate how the dynamics of a dense liquid evolves in the supercritical phase.

The coherent dynamics of liquids, at wavelenghts comparable to the mean interparticle distances, involves frequencies in the THz range. In this regime most liquids display universal features, such as a sound velocity value exceeding the adiabatic one (positive dispersion), due to the presence of high frequency relaxation processes. On a qualitative ground, one might expect that on abandoning the liquid phase the positive dispersion should vanish. As a matter of fact, indeed, relaxation processes are likely to become inactive as soon as the dynamics loses its cooperative nature and the concept of instantaneous vibrations is no longer well defined. We have shown that a strongly supercritical fluid (oxygen at $T \approx T_c$ and $P > 102$ PC) exhibits dynamical properties that are commonly ascribed to ordinary liquids. Based on the comparison with the behavior of several other fluids as a function of their thermodynamic state, we conclude that the extrapolation -well above the critical point of the liquid/vapor coexistence line- marks the boundary between simple liquid dynamics and the collision dominated regime characterizing the gas phase. We suggest that the extension of the coexistence line beyond the critical point splits the P-T phase diagram of simple fluids into a gaslike and a liquidlike domain, respectively, which differ in the local structure, resembling the subcritical behaviors.