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Polymorphic phase transition in liquid and supercritical carbon dioxide

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Thermal density fluctuations of supercritical CO_2 have been studied using small-angle neutron scattering (SANS), whose amplitude (susceptibility) and correlation length show the expected maximum at the Widom line. At low pressure, the susceptibility is in excellent agreement with the values determined from the mass density. Surprisingly, droplets form between 20 and 60 bar above the Widom line, which we identified as the Frenkel boundary line separating liquid-like and gas-like states of supercritical fluids [1]. This observation is in contrast to the classical picture of supercritical fluids, where there are "no discernible differences between liquid-like and gas-like states" [2]. The droplets initially form spheres with a radius of ≈ 45 Å and transform into rods and globules at higher pressure, representing a liquid-liquid (polymorphic) phase transition with same composition but different density, the last one determining the order parameter [3]. The polymorphism in CO_2 is a new observation that will stimulate discussions about gas-like and liquid-like states in SC fluids, in particular as CO_2 represents a "simple" van der Waals fluid, unlike water, the most studied fluid showing polymorphism in its supercooled state.

[1] V. Pipich, D. Schwahn, Phys. Rev. Lett. 120 (2018) 14570b1.

- [2] C. Cockrell, V.V. Brazhkin, K. Trachenko, Physics Reports 941 (2021) 1-27.
- [3] V. Pipich, D. Schwahn, Scientific reports (2020) 10:11861.

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