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Polymorphic phase transition in liquid and supercritical carbon dioxide

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Thermal density fluctuations of supercritical (SC) CO₂ were explored using small-angle neutron scattering (SANS) whose amplitude (susceptibility) and correlation length show the expected maximum at the Widom line. The susceptibility is in excellent agreement with the evaluated values on basis of mass density measurements. A surprising observation is droplet formation above the gas-liquid line and between 20 and 60 bar above the Widom line, the corresponding borderline identified as the Frenkel line. The droplets start to form spheres of constant radius of about 45 Å and transform into rods and globules at higher pressure. The droplet formation represents a liquid-liquid (polymorphic) phase transition of same composition but different number density, whose difference defines its order parameter. Polymorphism in CO₂ is a new phenomena, it characterizes the gas-like to liquid-like transition in SC fluids and might be of particular interest for better understanding polymorphism, since CO₂ represents a “simple” van der Waals liquid in contrast to water, which is the most widely studied liquid showing polymorphism in its supercooled state.

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