Crystallization of Undercooled Quantum Liquids: para-H₂, ortho-D₂, and their mixtures with Ne

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We have extended the experimental production of liquid microjets (filaments) in vacuum, first realized by Faubel et al. [1], to studying the crystallization of undercooled quantum liquids: para-hydrogen (pH₂), ortho-deuterium (oD₂), and their mixtures with Ne. These highly collimated filaments, less than 10 microns in diameter, are an ideal medium to produce undercooled liquid samples and to investigate the homogeneous solidification process, free from wall effects [2]. The filaments exit from cryogenic capillary nozzles into vacuum, to cool down fast by surface evaporation, although with a temperature gradient across the jet due to their finite size radius and thermal conductivity. The filaments are monitored by laser shadowgraphy, and analyzed by means of high performance Raman spectroscopy [3], revealing their structure and temperature. The high spatial resolution of Raman spectroscopy allows observing in situ the structural changes of the liquid microjets, with a time resolution of ~10 ns. The filaments of pure pH₂ can be cooled down to 9 K (normal melting point at 13.8 K), before eventually solidifying at a crystal growth rate of ~33 cm/s [4]. Filaments of diluted mixtures of oD₂ and pH₂ show a significant slowdown in the crystallization kinetics with respect to the pure substances [5], which is more pronounced in their mixtures with small amounts of Ne impurity. In the case of the pH₂/oD₂ mixtures, the observed slowdown can be interpreted in terms of a different effective size, caused by a purely mass-induced difference in zero-point quantum delocalization. This effect is confirmed in the mixtures with Ne, with a much larger quantum effective size ratio.

References

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