Kinetics of decelerated melting

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Abstract

Melting presents one of the most prominent phenomena in condensed matter science. Its microscopic understanding, however, is still fragmented, ranging from simplistic theory to the observation of melting point depressions. Here, we combine a multi-method experimental approach with computational simulation to study the microscopic mechanism of melting between these two extremes. We exploit crystalline structures in which melting occurs into a metastable liquid close to its glass transition temperature. The associated sluggish dynamics real-time observation of homogeneous melting. In-depth information on the structural signature is obtained from various independent spectroscopic and scattering methods, revealing a step-wise nature of the transition before reaching the liquid state. A kinetic model is derived in which the first reaction step is promoted by local instability events, and the second is driven by diffusive mobility. Computational simulation provides further confirmation for the sequential reaction steps and for the details of the associated structural dynamics, reconciling concepts of surface and bulk melting. This successful quantitative modelling of the low temperature melting of zeolite crystals, reconciling homogeneous with heterogeneous processes, should serve as a platform for understanding the inherent instability of other zeolite structures, as well as the prolific and more complex nano-porous metal organic frameworks.

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