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Melting dynamics of ices by time-resolved light scattering

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How does a crystal melt? How long does it take for melt nuclei to grow? The complete unfolding of the melting process, from the first nucleation events to the final achievement of a bulk liquid structure, is not known to date, as for the timescales and for the time-evolution of the heterogeneities involved.
The melting mechanisms have been addressed by several computational and experimental works, covering a sub-nanosecond time window with sample sizes of tens of nanometers, thus suitable to determine the onset of the process, but unable to unveil the following dynamics [1]. On the other hand, macroscopic observations of phase transitions, with millisecond or longer time resolution, account for processes occurring at surfaces and time-limited by thermal contact with the environment.
Here we fill the gap between these two extremes investigating the melting of ice in the entire mesoscopic regime. A bulk ice Ih or ice VI sample in a sapphire anvil cell is homogeneously heated by a picosecond infrared pulse, which delivers all the energy necessary for complete melting [2]. The lattice is therefore homogeneously heated from within, above its melting temperature [3]. The evolution of melt/ice interfaces thereafter is monitored by Mie-scattering with nanosecond resolution, for all the time needed to the sample to re-equilibrate, which may take milliseconds.
We observe the evolution of the liquid domains: growth and coalescence, and then refreezing due the heat removal by the cold environment. The growth of liquid domains over distances of microns takes hundreds of nanoseconds and is orders of magnitude slower than H-bond vibrational dynamics.
Water ice is the archetypal H- bonded and this investigation is fundamental as computation cannot access this length and time scales. Further investigations will focus on the phase transitions of gas hydrates, making use of light scattering and moving on to transient spectroscopic methods, able to distinguish among different chemical moieties.
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