



A new molecular level insight into how the hydrogen bond network is disordered in homogeneous ice melting

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“How a solid melts from inside?” is a fundamental question of physical chemistry, but not well understood [1]. Ice melting is usually induced at the interface between ice and a wall or other surfaces such as a dust particle, called as heterogeneous melting. On the other hand, homogeneous melting starts from inside the crystal due to just thermal fluctuations. Unlike heterogeneous melting, homogeneous melting rarely occurs unless ice is heated over 0° (super heated state) [2]. In crystalline ice, all of the water molecules share four hydrogen bonds (HBs) with their four nearest neighbours. The motion and structure are really restricted by the surrounding hydrogen bond network. The water molecules in ice are ordered and very stable, while that of liquid is disordered. Thus, ice melting is a disordering process, escaping from the global minimum on the energy landscape, gaining entropic stabilization. The past studies have shown that ice melting is the incidental accumulation of local defects [3,4,5]. However, we didn't believe that the restricted 3-dimensional HBs network is easily broken.

In this study, using molecular dynamics simulation, we have revealed that ice melting is not a random process, but instead proceeds through a specific pathway [6]. A very small trigger—i.e. the formation of the separated pair defect—plays a crucial role in inducing the disordering involved in the phase transition. The defect prevents the system going back to the global minimum and accelerates the transformation of HBs rearrangement. We were able to give a molecular-level description for “What is a trigger to form the initial embryo leading ice melting?”

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