Heterogeneous Molecular Dynamics in Quasi-liquid Layers of Ice Surface Using Molecular Dynamics Simulation

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Quasi-liquid layers (QLLs) exist on the surface of ice and have a remarkable influence on the distinct chemical and physical properties of ice. These layers vary significantly in heterogeneity, ranging from nanometers to millimeters. While the formation of partially ice-like structures has been suggested based on simulations [1,2], the molecular-level comprehension of this heterogeneity remains elusive. In this work, we explore the heterogeneity of molecular dynamics on QLLs using molecular dynamics simulations. To clarify the differences between QLLs from ice and liquid bulk, the molecular trajectories in QLL were compared to those of ice and liquid water molecules using machine learning-based analysis. Dynamics features extracted by the analysis showed distinct molecular dynamics in the bulk of the liquid and solid waters, whereas those of QLLs spanned over both the dynamics and their intermediate state. Our results suggest that the molecular dynamics of QLLs do not consist of a mixture of solid and liquid water molecules. However, molecules within the domains often switch their dynamical state. Despite the absence of any observable characteristic domain size, QLLs exhibit a strong dependence on temperature and crystal face regarding their long-range ordering. Rather than featuring static solid- and liquid-like regions, our findings suggest the existence of heterogeneous molecular dynamics in QLLs. This provides a molecular-level understanding of the surface properties of ice.