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Nonequilibrium Structural Phase Transformation of Interfacial Water Assemblies Observed by Ultrafast Electron Crystallography DING-SHYUE YANG, AHMED H. ZEWAİL, California Institute of Technology — Interfacial water, when compared with bulk phases, exhibits unique properties in various systems. By using ultrafast electron crystallography with atomic-scale spatiotemporal resolution, we report studies of structures and dynamics of interfacial ice assemblies on different surfaces. Structurally, ordered ice may be formed not only due to energetics of the water-surface interaction (commonly addressed as hydrophobicity and hydrophilicity), but also as a result of the surface morphology of the underlying substrate. The ultrafast dynamics also reveals new properties of a hydrogen-bond network. Following the photoexcitation of the substrate by a femtosecond infrared pulse, the interfacial ice structure undergoes, in few tens of picoseconds, nonequilibrium phase transformation identified through the observation of a structural isosbestic point in diffraction. This noncontinuous lattice expansion, from the unperturbed state to a highly expanded, nonthermal one, is caused by the structural vibration in the substrate and energy transfer across the interface. In addition, the cooperativeness of the hydrogen-bond network prevents a substantial evaporation lost of water molecules during repeated extreme expansion and recovery of the lattice. We provide the time scales involved and discuss the nature and implications of ice-substrate structural dynamics.

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