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**Methane Clathrate at Gas-Water Interface Studied by Sum Frequency Vibrational Spectroscopy** RONGDA LIANG, HUIJIE XU, SHUMEI SUN, CHUANSHAN TIAN, Fudan Univ — Methane clathrate (hydrate) is a rich resource of naturally occurring crystalline substance comprising a guest methane molecule embedded in a water cage at low temperature and high pressure. Despite its great importance in energy, climate effect and gas/oil transportation, the initial formation process of gas clathrate remains elusive, particularly in experiment. In this work, using sum-frequency vibrational spectroscopy (SFVS), we reported our experimental observation of methane clathrate formed at water-gas interface, where both water and methane molecules are abundant in the hetero-interfacial region, in a large range of temperature and pressure. The surface vibrational spectrum of gas clathrate was obtained for the first time. The resultant SF spectrum reveals that clathrate-like water network develops at interface much earlier than the thermodynamic phase transition point of the bulk clathrate. Our results provide molecular-level understandings of structure and formation process of gas hydrate at the interface, which has important bearing on inhibition of blockage in gas (oil) pipelines, carbon dioxide sequestration, chemical energy storage and so on.

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