**Movie S1.** The [linked movie](https://youtu.be/a8AJy9It8dA) shows the molecular interaction of the methane bubble with water and crystalline hydrate at the crystal/liquid interface at the beginning of the dissociation simulations at Tb=288 K. It provides a closer look into the crystal/liquid interface at the beginning of the dissociation (t ~ 0.3 ms). The water molecules (cyan spheres) that form the cages at the interface are continuously exchanged with the free water molecules from the liquid phase due to the breaking and reforming of the hydrogen bonds. Although this allows the trapped methane (red spheres) to escape, the cage keeps its structural integrity by instant enclathration of another free methane molecule. 

**Movie S2.** The [linked movie](https://youtu.be/Sp7kViDXqts) shows the dissociation of the methane hydrate motifs at the crystal/liquid interface for the time interval between 3.7 and 4.0 μs (the middle of Region 2 in Fig. 3 of the manuscript) at Tb=288 K. The results show that the hydrate cages at the crystal/liquid interface undergo a continuous cycle of dissociation and reformation before they fully dissociate. Red and cyan spheres represent methane and water molecules, respectively.

**Movie S3.** The [linked movie](https://youtu.be/YrC-PNRl_9Q) illustrates the formation and growth of the methane bubbles within the solid hydrate initiated by the ephemeral secondary dissociation path. Molecular trajectories are provided for five different xy-slices along the z-direction at a boundary temperature of 288 K. Methane and water are shown by red and cyan colors, respectively. 

**Movie S4.** The [linked movie](https://youtu.be/7zJK70aBruc) provides a detailed look at the molecular events leading to the formation of gas bubbles within the solid hydrate at the third xy-plane. The hydrate’s rigid movements cause the bubble to disappear and reappear intermittently. Methane and water are shown in red and cyan colors, respectively.