

## Supporting Information

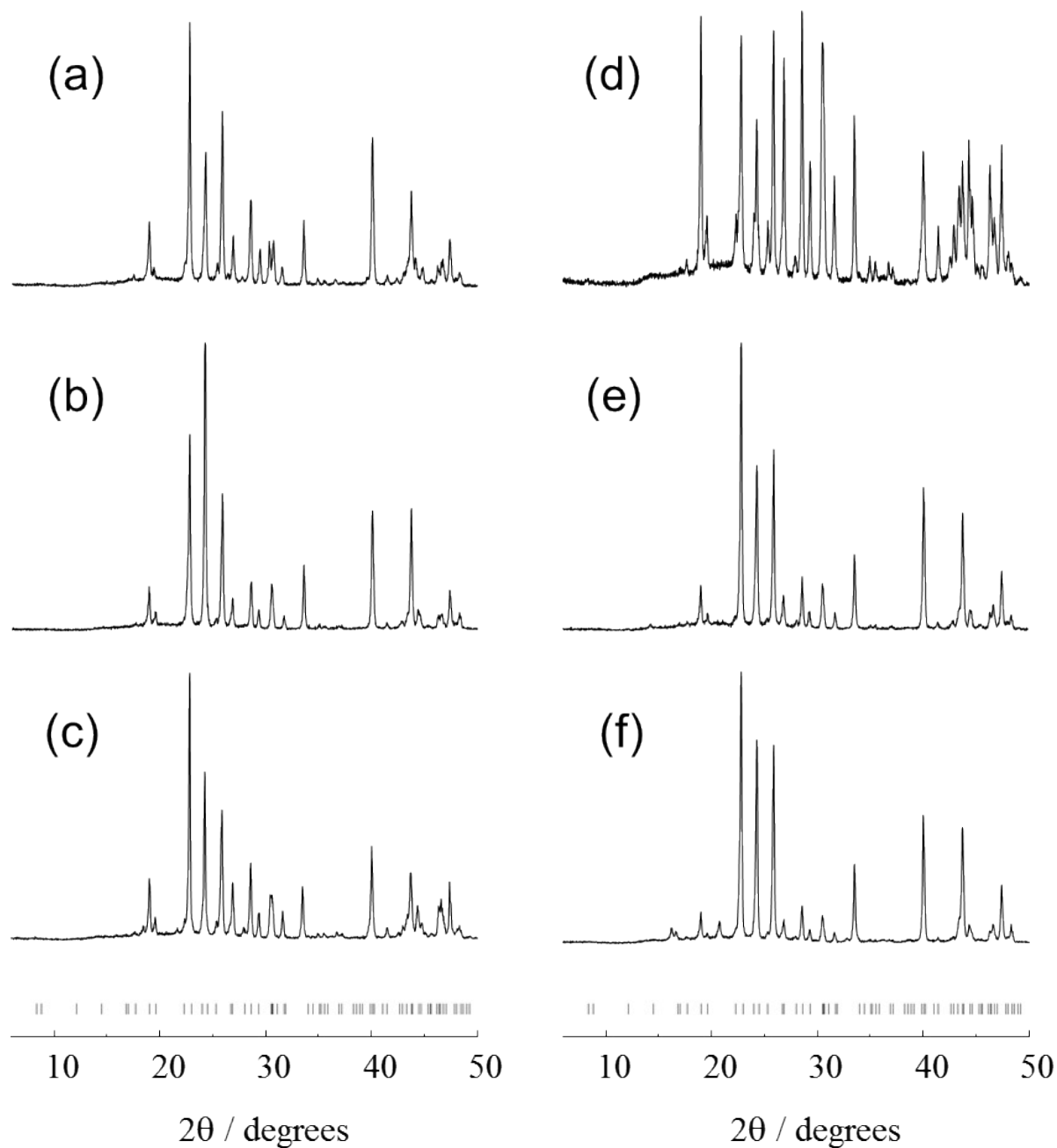
# Effect of Temperature and Large Guest Molecules on the C–H Symmetric Stretching Vibrational Frequencies of Methane in Structure H and I Clathrate Hydrates

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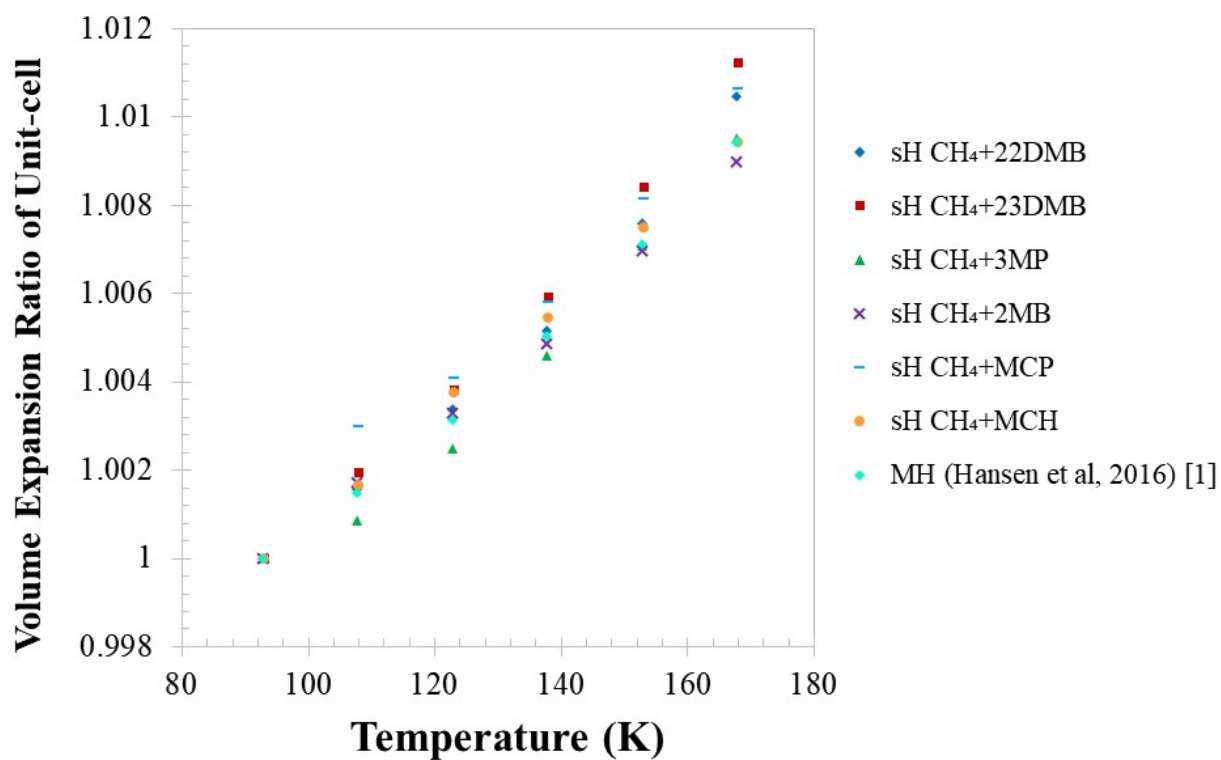
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**Fig. S1.** X-ray diffraction patterns of the structure H hydrates at 93 K: (a) CH<sub>4</sub> + 2,2-dimethylbutane hydrate; (b) CH<sub>4</sub> + 2,3-dimethylbutane hydrate; (c) CH<sub>4</sub> + 3-methylpentane hydrate; (d) CH<sub>4</sub> + 2-methylbutane hydrate; (e) CH<sub>4</sub> + methylcyclopentane hydrate; (f) CH<sub>4</sub> + methylcyclohexane hydrate. The curves in each pattern represent the observed intensities. The line marks below each pattern show the calculated peak positions for hexagonal ice.



**Fig. S2.** Temperature effect on volume expansion ratio of unit-cell for six types sH hydrates and sI CH<sub>4</sub> hydrate (MH).<sup>1</sup>

## Reference

- [1] Hansen, T. C.; Falenty, A.; Kuhs, W. F. Lattice Constants and Expansivities of Gas Hydrates from 10 K up to the Stability Limit. *J. Chem. Phys.* **2016**, *145*, 7066–7070.