Collective Dynamics and Localized Guest Vibrations of Methane and Xenon Hydrate

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Clathrate hydrates are a special class of inclusion compounds, in which small guest molecules or atoms are trapped in cages formed by an ice-like host lattice of water molecules. In recent years the clathrate hydrates have attracted considerable interest as large deposits of methane hydrate have been discovered on the oceanic sea floors.
In the case of the gas hydrates the guest-host interaction is weaker than the bond strength of the cage structure. Nevertheless, the hydrophobic interaction between the guest molecules or atoms and the cage forming water molecules is thought to be responsible for a coupling between the localized low frequency vibrations of the guest molecules and the acoustic host lattice modes.
We report the results of inelastic x-ray and neutron scattering experiments on methane and xenon hydrate. The neutron scattering experiments focused on the localized vibrations of the guest molecules or atoms inside the ice cages. We could extract the frequencies of the guest vibrations and confirm a coupling between the guest and host vibrations [1,2]. With the help of inelastic x-ray scattering experiments we could determine the powder averaged dispersion curve of methane and xenon hydrate. The results provided insight into the coupling mechanism, supplying experimental evidence for an avoided crossing between the localized guest modes and the acoustic host lattice phonons [2]. On the basis of lattice dynamical calculations the experimental findings of both the inelastic neutron and the inelastic x-ray scattering experiments could be reproduced, showing that the avoided crossing promotes an extensive mixing of the guest and host modes. It is concluded that the mixing of the modes leads to the coupling of the guest and host vibrations observed in the inelastic neutron scattering experiments and to the collective guest modes found in the inelastic x-ray spectra.

References

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