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High-pressure phase stability and elasticity of ammonia hydrate

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Abstract

Phase stability and elasticity of ammonia hydrate have been studied using Raman spectroscopy and Brillion scattering in diamond-anvil cells up to 53 GPa at 300 K. Here we have established the high-pressure phase diagram of ammonia hydrate in three different compositions, including ammonia monohydrate (AMH, $NH_3 \cdot H_2O$), dihydrate (ADH, $NH_3 \cdot 2H_2O$), and trihydrate (ATH, $NH_3 \cdot 3H_2O$). In contrast to previous experimental results, our Raman and Brillouin measurements at 300 K have shown that all three ammonia hydrates start to dehydrate at 2.1–2.2 GPa. Dehydration of the ammonia hydrate leads to the formation of single-crystal ice-VII and an increase in the concentration of NH_3 in the residual liquid. The residual liquid finally turns into solid ammonia hemihydrate phase II (AHH-II) at 4–4.6 GPa, leading to a 28% jump in the compressional-wave velocity (V_P). Considering a 10–15 vol% NH_3 in the mantle of ice giants, AHH should thus be the dominant form of NH_3 coexisting with H_2O-ice in the ice giants. Further Brillouin measurements provide crucial constraints on the V_P of AHH and the single-crystal elasticity of ice-VII at high pressures and 300 K. V_P of AHH increases smoothly with pressure. No anomalous change in V_P of AHH was identified up to 39 GPa, although a solid to solid phase transition was noted to occur at ~18 GPa by Raman measurements. In addition, the elasticity of single-crystal ice-VII, which was the dehydration product of ammonia hydrate, has been determined up to 53 GPa at 300 K. The deviation of C_{12} from C_{44} observed at 11.4 and 14.6 GPa could be caused by the hydrogen bond symmetrizations or the ordering of dipole of single-crystal ice-VII. An abnormal softening in the elastic moduli C_{11}, C_{12}, and the adiabatic moduli K_S together with stiffening in C_{44} was observed between 42 and 53 GPa, which should be caused by the transition from ice-VII to its pre-transitional state. Of particular interest is the dramatic increase in the anisotropy of ice-VII with increasing pressure. Combining the sound velocity of AHH and ice-VII, we have modeled the V_P of ice giants with a volume ratio of 20% AHH and 80% ice-VII in the mantle. The obtained high-pressure phase diagram and elastic properties of ammonia hydrate could contribute to understanding the structure of the mantle in the ice giants and satellites.

Keywords: Ammonia hydrate, AHH, single-crystal ice-VII, elasticity, phase transition, ice giants