

## 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 Brillouin 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,  $\text{NH}_3 \cdot \text{H}_2\text{O}$ ), dihydrate (ADH,  $\text{NH}_3 \cdot 2\text{H}_2\text{O}$ ), and trihydrate (ATH,  $\text{NH}_3 \cdot 3\text{H}_2\text{O}$ ). 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  $\text{NH}_3$  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%  $\text{NH}_3$  in the mantle of ice giants, AHH should thus be the dominant form of  $\text{NH}_3$  coexisting with  $\text{H}_2\text{O}$ -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  $\sim 18$  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