

nitrogen-halogen or better still hydrogen-halogen distances are considerably shorter than the sum of van der Waals radii.<sup>11,14,18</sup> The experimental hydrogen-halogen distances are 2.32 Å and 2.49 Å (1 bar, 300 K) while the van der Waals radii sum to 3.0 Å and 3.2 Å for NH<sub>4</sub>Cl and NH<sub>4</sub>Br, respectively. Cohesive energy calculations also suggest the existence of hydrogen bonding and place the effect to be approximately 3% of the total lattice energy or 4.4 and 4.0 kcal mole<sup>-1</sup> in the chloride and bromide respectively.<sup>19</sup> Deuteration often provides information about hydrogen bonding. In the presence of bonding, the smaller zero point energy of deuterium should lessen the amplitude of the hydrogen vibration, the repulsive potential, and the bond energy.<sup>20</sup> The lattice constant of ND<sub>4</sub>Cl at room temperature and the volume change associated with the lambda phase transition are smaller than for NH<sub>4</sub>Cl, consistent with weaker hydrogen bonding.<sup>21-23</sup> On the other hand at room temperature, the NH<sub>4</sub>Br lattice expands upon isotopic substitution,<sup>21</sup> Hydrogen bonding is also expected to increase the intensity, to broaden the peak and to decrease the frequency of the hydrogen stretching mode,<sup>24</sup> while the energies of the hydrogen deformation and the librational mode of the NH<sub>4</sub><sup>+</sup> ion should increase. Effects characteristic of hydrogen bonding were observed in a preliminary high pressure Raman study of NH<sub>4</sub>Cl.<sup>25</sup> On the other hand, hydrogen bonding is not supported by the near infrared studies (3000-7000 cm<sup>-1</sup>) on NH<sub>4</sub>Cl, where it is reported that the N-H bond strength increases (1.1% at 20 kbar) rather than weakening as the hydrogen-halogen distance decrease.<sup>26-27</sup>

An NH<sub>4</sub><sup>+</sup> ion has T<sub>d</sub> symmetry both in the isolated state and in the ordered phase IV. Four fundamental modes are allowed under the T<sub>d</sub> point group with the following irreducible representations: 1 A<sub>1</sub> + 1 E + 2 F<sub>2</sub>. ν<sub>1</sub>(A<sub>1</sub>) and ν<sub>3</sub>(F<sub>2</sub>) are the symmetric and the asymmetric hydrogen stretching modes, while ν<sub>2</sub>(E)