- Fig. 4. Shift of the librational frequency (cm<sup>-1</sup>) of NH<sub>4</sub>Cl with increasing pressure at 296 K. The values obtained from one-phonon spectra are indicated by the light circles (0) while those obtained from twophonon spectra are shown by the dark circles (0). The "order-disorder" phase transition occurs at 9-10 kbar.
- Fig. 5. Shift of the internal mode frequencies of the  $NH_4^+$  ion in  $NH_4Cl$ . . . and  $NH_4Br$  with change in nitrogen-halogen distance,  $d_{N-X}$ , at 296 K. For the symmetric, hydrogen-stretching mode,  $v_1$ , only the wavenumber observed at 1 bar is indicated.
- Fig. 6. Variation of the librational frequency (cm<sup>-1</sup>) of NH<sub>4</sub>Cl and NH<sub>4</sub>Br with lattice constant a<sub>0</sub>. In NH<sub>4</sub>Cl, values from both one-phonon (light circles, 0) and two-phonon (dark circles, 0) spectra are shown, while those in NH<sub>4</sub>Br were all obtained from two-phonon excitation process.
  Fig. 7. Pressure induced frequency variation of the combination bands in
- NH<sub>4</sub>Cl and NH<sub>4</sub>Br at 296 K. Curves designated as A, B and C represent combination bands:  $\nu_2 + \nu_6$ ,  $\nu_4 + \nu_6$  and  $\nu_4 - \nu_6$  respectively. Fig. 8. Peaks centered around  $\nu_3$  (3150 cm<sup>-1</sup> at 1 atm) in NH<sub>4</sub>Cl are shown at 5.9 and 22.7 kbar pressure for unpolarized light. Polarization studies at atmospheric pressure show the presence of five peaks with  $\alpha_{xz}$  and one with  $\alpha_{xx}(\nu_1)$  polarization. These peaks could not be

resolved in the high pressure spectra.

Fig. 9.

Effect of isothermal (296 K) and isobaric (1 atm) "disorder-order" phase transition on the relative Raman intensity of  $\nu_4$ (TO) and  $\nu_4$ (LO) component in NH<sub>A</sub>C1.

1.