butions from the attractive hydrogen-halogen coupling (hydrogen bonding effects) which would be important due to the large ionic polarizability of the halide ions.[55] Attractive forces also may arise from the van der Waals interaction between the NH_4^+ and the halide ion, from dynamic polarization effects introduced during vibration,[56] and as a result of the dipole induced on the halide ion by the hydrogens when the NH_4^+ ions are antiparallel.[17] The last attractive potential is believed to be quite important in "disordered" NH_4Cl V and NH_4Br II.[35]

The potential for the librational motion of the NH_4^+ ion is usually developed in terms of the spherical harmonics.[17] In the disordered phase, the leading nonspherical term is the fourth-order term; and an approximate solution which relates the barrier height (V_o) to the first-order librational frequency is given by:[57]

$$h\nu_6 = (8\hbar^2 V_o/I) - (5\hbar^2/2I) \qquad (2)$$

Here, I is the moment of inertia, and ν_6 is the librational frequency. In this approximation, the barrier height should vary inversely as the fifth power of the lattice constant, a_o. A potential for octapole-octapole interaction which varies inversely as the seventh power of lattice constant is usually added to the above expression for the potential when all the NH_4^+ ions are parallel to each other in the ordered phase. When the barrier height (V_o) for "disordered" NH_4Cl V is evaluated from the one-phonon librational frequency (Fig. 4) on the assumption of a cubic potential (Eq. 2) and a constant moment of inertia, V_o is observed to be proportional to $a_o^{-6.7}$ rather than the expected a_o^{-5}. The barrier height for ordered NH_4Cl IV shows a weaker dependence on the lattice constant with V_o now proportional to $a_o^{-2.5}$. Although the calculations for phase IV is based on librational frequency obtained from two-phonon excitation, it is felt that this is not the cause for the weak dependence on lattice constant in the ordered phase. The pressure dependence