Synopsis

The experimental investigation of some paramagnetic complexes shows, in addition to usual cooperative phenomena characterized by ordered arrangements of magnetic moments, a critical point phenomena in their paramagnetic phases which is associated with anomalous magnetic and thermodynamic behaviour and a change in their crystal structure. Efforts were made to get an understanding of the difficult problems of this newly observed phase transition as well as its origin and the properties of the cooperative state itself from the experimental and theoretical points of view. A theoretical model was attempted to represent the magnetic configuration o the low temperature phase of the transition.

In the introductory chapter we present the thermodynam aspects of phase transition with a particular emphasis on the shape of the specific heat anomaly, thermodynamic models and spin wave spectra of the magnetic systems, and a discussi of the characteristic features of weak ferromagnetism.

Chapter II is devoted to description of the specific heat, magnetic susceptibility and magnetic anisotropy apparat An effort is made to present a detailed description of the experimental set up of the low temperature specific heat apparatus which we constructed in our laboratory.

The experimental results comprising specific heat, principal magnetic susceptibility and magnetic anisotropy dat

of some cobalt-fluoro-complexes in the temperature region 200-300°K are given in chapter III. The specific heat data of the isomorphous zinc-fluoro-complexes in the same temperature rang which are used to isolate the irregular part of the specific heat of the cobalt complexes are also given. In addition to these results we present anisotropy data of nickel and manganese-fluoro-complexes.

In the final chapter we discuss the experimental results given in the preceding chapter as well as the results of the studies of optical absorption and X-ray data. A preliminary study of the magnetic and specific heat data suggests that there exist an anomalous region ($T_c \sim 250^\circ K$) in the paramagnetic phases of the cobalt complexes. It is shown from an analysis of optical absorption and X-ray data at both room and low temperatures that the cobalt-fluoro-complexes undergo a second order transition from uniaxial to monoclinic symmetry. We propose a theoretical model to explain the magneto-crystalline anisotropy of the monoclinic phase. The magnetic symmetry of the monoclinic phase is the same as that required for weak ferromagnetism to appear below the Neel point. On the basis of this model the experimental and theoretical magnetic anisotropies are correlated with a reasonable choice of crystal field parameters. On examining the detailed shape of specific heat singularities it appears that the approach to the transition point is logarithmic, similar to that of λ-transition in ^4He. The comparison of both magnetic (χ_1) and specific heat singularities at the same transition point suggests that there may

be a correlation between them. Finally we discuss the possible origin of the phase transition. The fact that unlike the complexes containing C_o^{++} ions having orbitally degenerate ground state the isomorphous nickel fluoro-complexes containing N_i^{++} ions having orbitally nondegenerate ground state do not undergo such transition suggest that the origin of the transition may be due to Jahn-Teller instability of the C_o^{++} ion. It is suggested that the transition may be attributed to cooperative Jahn-Teller effect.