The thesis discusses the studies carried out by us on chiroptical properties of some interesting optically active lanthanide single crystals. It covers in all, the optical activity of three different single crystal systems, each with its distinct unit cell as well as Ln^{3+}-site structural characteristics. In general, attempts have been made to look into the transition mechanisms operating.

Chapter I starts with a brief early history of optical activity together with different areas of its application. A review of chiroptical studies carried out on lanthanide complexes so far, both in solids and in solution, has been presented. The dearth of spectra-structure correlation for lanthanides at the present time has been emphasized and the scope of the thesis outlined.

The second chapter deals with the relevant theoretical aspects required for analysis of our observations. Theories on one-photon lanthanide transitions have been discussed. The assumptions and limitations of Judd-Ofelt theory and the basic idea behind the 'dynamic' coupling model have been discussed. The basic principles of optical and magneto-optical activity have also been outlined. The chapter also discusses the possibility of observing two-photon (TP) optical activity.

Chapter III outlines our experimental approach. Section-A contains techniques adopted for crystal growth and orientation, as well
as experimental techniques for measuring CD and absorption spectra at different temperatures and under the same spectral resolution. The section ends with the procedure employed to process the raw data. Section B deals with the details of the theoretical model used by us to calculate the lanthanide optical activity for CF component-to-component transitions. The discussion is general and can be applied to any axially symmetric lanthanide complex having authentic chiral environment.

Chapter IV covers the experimental findings with Gd-DG single crystal. The relative values of the ratio of rotational strength (\( R_K \)) to the square root of dipole strength (\( D_K \)) are compared with the relative values of the gross magnetic dipole moments of the \( f \rightarrow f \) transitions. As expected, circular dichroism have been found to be relatively more prominent for transitions where \( \Delta L = |1| \cdot |2| \) compared with \( \Delta L \geq 2 \). J-mixing between close-lying states limits the usefulness of the \( \Delta J = 0 \), \( \pm 1 \) selection rule in this case.

Chapter V deals with the identification of \( ^{2}K_{15/2} \) and \( ^{2}K_{13/2} \) states of Er\(^{3+} \) in DG crystal. Both the states are close-lying with other states originating from different L-values. Therefore, J mixing being less, \( \Delta J = 0 \), \( \pm 1 \) selection rule is exploited for their spectral assignments.

Chapter VI reports the studies made on Eu\(^{3+} \)-doped K\(_3\)Nd\(_2\)(NO\(_3\))\(_9\) - the cubic anhydrous double nitrate system, Ln-DN (Ln=Pr, Nd).Low-resolution CF-unresolved CD and absorption spectra are discussed. Chiroptical properties of transitions of Eu\(^{3+} \) have been compared.
with those of the Nd\(^{3+}\) transitions. Attention has been drawn on the magnetic dipolar contribution to some of the Nd\(^{3+}\) transitions which are generally considered to be of electric dipole origin.

The seventh chapter (Chapter VII) deals with the low-temperature high-resolution CD and absorption spectra of the interesting chiral cubic crystal \(K_3Pr_2(\text{NO}_3)_9\). A model calculation of optical activity has been done to correlate the observed rotational strengths for CF component-to-component transitions. Agreement with experiment is only partial, presumably due to uncertainties in evaluation of theoretical parameters. The possibility of higher order contribution to circular dichroism has been indicated.

The last chapter (Chapter VIII) deals with yet another optically active lanthanide single crystal system - the Pyrogermanates, \(\text{Ln-PG}\). Room temperature absorption and CD spectra for the \(^8S_{7/2} \rightarrow ^6P_J\) transition in Gd-PG have been presented. A theoretical calculation with limited basis set has been attempted. The sign matches for CF component-to-component transition for intrinsically magnetic dipole-allowed transitions, \(^8S_{7/2} \rightarrow ^6P_{7/2}, ^6P_{5/2}\) but not for the \(^8S_{7/2} \rightarrow ^6P_{3/2} (J = 2)\) transition.