# Abstract

Chromate spinel oxides exhibit a plethora of novel physical properties. In this Ph.D. thesis, we have designed materials based on such selected chromate spinel oxides ($ACr_2O_4$), synthesized them and investigated different physical properties (mainly, magnetic, structural and electrical properties) by employing multiple experimental techniques. A common way of lifting geometric-magnetic-frustration (GMF) in $ACr_2O_4$ is through the global structural transition from the high-symmetry cubic phase, which is driven either by orbital Jahn-Teller (JT) effect (mediated by JT active ions, as seen in $FeCr_2O_4$, where $Fe^{2+}$ is JT active) or Spin-JT effect (mediated by strong spin-lattice coupling, as seen in $CdCr_2O_4$). $MnCr_2O_4$ and $CoCr_2O_4$ ($Mn^{2+}$, $Co^{2+}$ and $Cr^{3+}$ ions are JT inactive) show exceptions to the above general rule, as they undergo long-range ferrimagnetic (FIM) ordering within their corresponding cubic phases. Thus, origin of the long-range FIM ordering still remains intriguing in this two systems. We investigate this paradox and found that both $MnCr_2O_4$ and $CoCr_2O_4$ undergo hitherto undetected glassy magnetic ordering just above the FIM ordering. Through detailed neutron diffraction study, we elucidate that the observed glassy magnetic ordering is associated with the onset of weak spin-JT driven microscopic structural modifications, which along with *A*-site magnetic moment circumvent the GMF to an extent so that long-range FIM order could establish in this two systems. Based on this knowledge, we have engineered a $Mn_{1-x}Cd_xCr_2O_4$ series that enables to controllably tune the strength of spin JT effect and demonstrate its explicit dependency on GMF. Cd doping systematically tune different magnetic interactions in $MnCr_2O_4$ and due to the appearance of dominant antiferromagnetic interaction, the GMF becomes very high in the higher *x* compounds. Detailed dielectric, specific heat and temperature-dependent x-ray diffraction illustrate the commencement of strong spin-JT (associated with large GMF) induced global structural transition to relieve the GMF for the higher *x* compounds. On the other hand, we have also designed $Cu_{1-x}Cd_xCr_2O_4$ series in order to investigate the origin of ferroelectricity in $CuCr_2O_4$. Through detailed magnetic and structural studies, we elucidate the decoupling of the pyrocurrent peak temperatures (associated with the parent as well as doped compounds) with any magnetic and structural phase transitions, thus, illustrating the absence of ferroelectricity in $Cu_{1-x}Cd_xCr_2O_4$. The space charge origin of the observed pyrocurrent peaks is confirmed through heating rate dependent and 4-cycle pyrocurrent measurements. Besides, we have explored the observation of a high-temperature reversible pyrocurrent peak in $MgCr_2O_4$ and have elucidated its extrinsic origin.

**Key words**: JT effect, GMF, ferrimagnetism, ferroelectricity, structural transition