ABSTRACT

In the present era of technology, materials science plays a key role in the development of several advanced technologies related to different sectors such as health, computing, defence, agriculture etc. Interestingly, the magnetic materials belong to a class of materials which has a wide range of applications. The properties of magnetic materials strongly depends upon the crystal structure, spin order and interaction, which can be investigated using diffraction techniques such as X-ray and neutron diffraction. Among the different magnetic materials, the Heusler alloys have become the most promising candidate in the field of modern research due to their possible applications in spintronics, thermoelectric, shape memory alloys, etc. More recently, the magnetocaloric effect (MCE) has been discovered in Heusler alloys and it is observed that these alloys exhibit high magnetic entropy change, magnetic transition appears at ambient temperature, which follow second order phase transition (SOPT) nature, and fulfil the requirements and hence attracted scientific society towards the development of magnetic refrigerant operant. For any material to be used in these applications, well controlled structural and magnetic properties are essentially required, which is found to be one of the most prime features of Heusler alloys and can easily be achieved by partial substitution of an element with other transition metal. However, a great deal of discussion is still required to understand that how this partial substitution of transition metal influences the structural as well as magnetic properties of Heusler alloys. Besides this, a correlation between MCE and the critical behaviour observed at phase transition in Heusler alloys needs also to be established.

In the present thesis, structural and magnetic properties of two families of Heusler alloys Co_2CrAl such as $Co_2Cr_{1-x}Ti_xAl$ ($x = 0, 0.25, 0.5$), $Co_2Cr_{1-x}Mn_xAl$ ($x = 0.25, 0.5, 0.75$) have been investigated. The Co_2CrAl has been chosen as prototype sample because it has Curie temperature (T_C) near the room temperature and posses a high magnetic moment, which are favorable for practical applications. Further, we have investigated the physical properties to see the effect of Ti and Mn substitution at Cr site and this provides extra degree of freedom by means of which their magnetic properties can be tuned. We show a SOPT from paramagnetic to ferromagnetic (PM-FM), where T_C of Co_2CrAl increases with both Ti and Mn substitution. The neutron powder diffraction measurements were carried out to calculate degree of structure ordering, and a correlation between structural and magnetic analysis was established for Co_2CrAl alloy. The structural ordering and hence magnetic moment increases with Mn substitution. All the alloys in these two families exhibit SOPT type nature across the T_C. The magnetic entropy change, and hence relative cooling power increases with substitution, which confirms the potential of these materials in multi-stage magnetic refrigeration. Moreover the critical behavior

analysis of $Co_2Cr_{0.75}Ti_{0.25}Al$ and $Co_2Cr_{0.25}Mn_{0.75}Al$ also confirms the presence of SOPT and indicates the existence of long-range FM interactions in these materials. Further, the structural and magnetic properties has been investigated across the T_C of Co_2FeZ (Z=Al, Ga) Heusler alloys, which are found to be in agreement with our previous experimental results. A structural transition from cubic to orthorhombic phase across T_C has been observed and thorough analysis of this transition has been carried out for Co_2FeZ Heusler alloys. The analysis of X-ray absorption spectra gives a complementary information to study local antisite ordering in Co_2FeAl alloy.

We have also studied the structural, transport, electronic, and magnetic properties of Co_2FeGa Heusler alloy nanoparticles, which have been synthesized via two different chemical approaches i.e. sol-gel and co-precipitation methods. The Co_2FeGa nanoparticles were synthesized with controlled size and their structural and magnetic properties have been discussed in detail. We found that the particle size is correlated to the domain structure, and size dependent multi-domain to single-domain transition has been demonstrated for the first time for any set of Heusler alloy nanoparticles.

Overall, in this thesis the structural and magnetic properties of Co-based Heusler alloys have been studied to understand the effect of doping and detailed analysis of critical behavior have been explored to understand magnetic interactions in these alloys along with a small section on the basic study of nanoparticles as well. By performing magnetic susceptibility, isothermal magnetization, and neutron diffraction measurements in large temperature range across the phase transition, we present detailed analysis of change in magnetic entropy, critical behavior as well as relative cooling power and established the correlation between atomic ordering, nature of transition and complex magnetic interactions present in these cobalt based Heusler alloys.