SYNOPSIS

This thesis describes the studies of some Heusler alloys, having Fe as one of the constituents, from measurements of bulk and microscopic magnetic properties. Magnetic systems like Heusler alloys $\text{Ru}_2\text{FeGe}$, $\text{Rh}_2\text{FeGe}$, $\text{Rh}_2\text{FeSi}$ and their related compounds $\text{RuFeSi}$ and $\text{RuFeB}$ were synthesised and studied for the first time. Experimental techniques of X-ray diffraction, AC susceptibility, DC magnetisation, Mössbauer spectroscopy, Zero velocity thermal scanning, X-ray and physical density measurements have been used in these investigations.

Heusler alloys are a class of intermetallic compounds crystallising in $L_{21}(C_{1b})$ structure, and are represented by the general formula $X_2YZ(XYZ)$, where $X$ and $Y$ are transition metals like Cu, Mn, Co, Ni, Ru, Rh, Pd..., and $Z$ atom is any member of the large number of the sp elements. In the $L_{21}$ structure $X$ atom occupy $(A,C)$ sites, $Y$ and $Z$ atom occupy $B$ and $D$ site respectively. A large number of these alloys can be formed which exhibit a high degree of atomic order.

The category of alloys in which the $Y$ atoms are moment bearing e.g. $\text{Cu}_2\text{MnAl}$ are interesting. These magnetic atoms, in an ordered $L_{21}$ phase, are extremely well separated occurring as third nearest neighbours to each other, thus leading to an absence of any direct overlap of the d-orbitals of these magnetic atoms. These magnetic atoms though 25% of the total number can still be effectively treated as dilute magnetic impurities in an otherwise nonmagnetic host. This tremendous simplification as compared to
the situation in pure metals make this class of Heusler alloys ideally suited for the study of their magnetism, which is essentially mediated by the conduction electrons. The conduction electron polarisation (CEP) arising from these dilute magnetic impurities can be effectively probed by hyperfine fields at non-magnetic probe atoms.

Considerable efforts have been made both recently and in the past to understand the conduction electron polarisation effects in metallic systems by measuring the magnitude and sign of the net spin polarisation i.e. the hyperfine fields at the non magnetic probe ions in the Heusler alloys. The systematics of hyperfine fields (HFs) itself is interesting in these type of Heusler alloys as the valence state of the non-magnetic atom severely modifies the CEP in the localised region around it.

Extensive data on the hyperfine fields at non-magnetic probes replacing Sn (at the D site) now exists in literature. Some systematic trends in HF have emerged from these investigations. It is observed that for low valence sp elements the HF is seen to be negative, while for high valencies the HF is seen to be positive. For impurities with valency 4 \((s^2p^2)\) like Sn, Ge the HF is seen to lie at the sign crossover. This behaviour is similar to that observed for HF at dilute sp-impurities in conventional ferromagnets Fe, Co, Ni. The ease with which a non-magnetic sp element can be replaced by another together with the simplicity of their crystallographic and magnetic structure
make these alloys stringent test-bed for several CEP theories.

The Blandin and Campbell (BC) model for the HF could account for the systematic trends in the HF mentioned above. Recently making use of the HF at sp elements in ordered and disordered Heusler alloys a set of new parameters were obtained in the BC model which could qualitatively explain most of the HF data on non-magnetic atoms at various sites. This parametrization also predicted HF to be nearly identical, at least the sign of the hyperfine fields (HF), in isostructural, and iso-electronic Heusler alloys.

In the present investigations the existing series of Ru and Rh based alloys have been extended by a set of new iso-electronic Heusler compounds in which the constituent Sn has been replaced by the semimetal Ge and Si. The hyperfine fields at $^{57}$Fe and $^{119}$Sn as a dilute impurity in the new Heusler alloys are studied.

During our investigations of the HF in the above mentioned alloys we found as a result of progressively replacement of the sp element, Sn by Ge, and Si, a unique class of magnetic systems containing Fe, viz., $\text{Ru}_x\text{Fe}_y\text{Si}$, RuFeB, ($X + Y < 3$). The results obtained so far defy conventional description of these systems. From bulk measurement in the ordered state it is found that the systems have small magnetic moments, do not show saturation in magnetic field upto 10 kOe and have large magnetic ordering temperature $T_m$. However over a large temperature range below $T_m$, 
no appreciable hyperfine field (HF) at Fe nuclei by Mössbauer Effect is observed. A large field at Fe is seen to build rapidly over a narrow temperature range below a characteristic temperature $T^* \ (\sim 0.1 \ to \ 0.2 \ T_m)$ and is not accompanied by any change in the bulk magnetism. This evolution of magnetic moment of Fe far below the ordering temperature $T_m$ is unique among any known magnetic systems. These systems are presumably the first known examples which seem to lie between the two extreme in the theory of magnetism.

The thesis is divided into five chapters.

A brief introduction to the current status in magnetism with particular reference to local and itinerant behaviour and also general remarks regarding to a unified picture of spin fluctuations are presented in chapter 1. Also a brief introduction to the subject of hyperfine fields is given in this chapter. These form the necessary prerequisite background to understand, and interpret the magnetic behaviour of the studied compounds.

Chapter 2 deals with the experimental techniques used in the investigations, like AC susceptibility, DC magnetisation, Mössbauer spectroscopy, Zero velocity thermal scanning, X-ray and physical density. This chapter also deals with some of the instrumentation developed, like Mössbauer spectrometer, and its associated electronics and cryostats. A brief description on the data handling and analysis programmes used for Mössbauer experiments will be also given in this chapter.
Chapter 3 describes in detail the magnetic studies of Heusler alloys Ru$_2$FeGe, Rh$_2$FeGe, Rh$_2$FeSi.

In chapter 4 the hyperfine field investigations, AC susceptibility DC magnetization, (4.2 K to 1000 K), zero velocity thermal scanning, X-ray diffraction, Metallgraphic results and Arrots plots of the systems Ru$_x$Fe$_y$Si and Ru$_x$Fe$_y$B, are presented. Mixed ferrite systems Cu$_{1-x}$Cd$_x$Fe$_{2}$O$_4$ and Cu$_{1-x}$Zn$_x$Fe$_{2}$O$_4$ have been investigated using $^{57}$Fe Mossbauer spectroscopy over a temperature range 80°K to 600°K. The addition of the diamagnetic Cd and Zn into the CuFe$_2$O$_4$ system weakens the magnetic interaction between Fe-Fe pairs giving rise to a distribution both in the hyperfine magnetic field and the Neel temperature due to formation of clusters. It was seen that the spectra could be fitted very well when such distributions were incorporated in the fitting procedure and there was no need to invoke any relaxation phenomenon as is usually done for such systems. Our measurements thus confirm the validity of a static hyperfine field distribution model in these mixed ferrite systems.

A brief summary of all the important results and conclusions drawn from them are present in the final chapter.

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