Abstract

Much effort has been devoted in recent years trying to understand systems where the frustrations in the microscopic magnetic exchange interactions are present. These frustrations can result from competing exchange or random anisotropy. Many experimental systems with competing exchange interaction show a double transition (reentrant) behavior: below a critical temperature, a well defined transition from paramagnetic (PM) to ferromagnetic (FM) state followed by one more transition at lower temperature (spin-glass (SG) like transition) into a magnetically frozen state. Amorphous (a-) $Fe_{100-c}Zr_c$ alloys (containing 8 to 12 at. % of Zr) system appears to be a classic case where all the above-mentioned features play a key role in determining the magnetic structure and has been a fertile ground for the exploration of novel magnetism. Various models have been proposed to explain this peculiar low temperature magnetic state. However, the exact nature of the low temperature transition and that of the reentrant state has eluded clear-cut understanding so far. The rapid variation observed in magnetic structure even for a small at. % change in Zr and presence of magnetic inhomogeneity has lead authors to study the temperature dependence of magnetic and transport properties by various substituting elements in a-$Fe_{90}Zr_{10}$ alloy in order to understand the reentrant behavior. However, substitution of Co, Ni, Si, B, V and Cr destroys the reentrant behavior. On the other hand, substitution of Mn decreases T_c almost linearly and reentrant behavior is observed upto 10 at. % of Mn. Moreover, the preliminary measurements of characterization of magnetic state have been observed to deviate one from the other and no transport studies are reported in detail. Therefore, a systematic composition (Mn) dependence of magnetic and electrical transport characterization is very much essential to understand the effect of Mn on a-FeZr alloy.

The present research study is concerned with understanding of nature of the temperature and field dependence of magnetic state and electrical transport properties in these mixed exchange interaction systems. The following studies have been carried out in detail:

- ☺ Spin quenching technique is used to prepare amorphous samples in ribbon form. X-ray diffraction and Scanning electron microscope studies have been used to confirm the amorphous nature. Energy dispersive X-ray analysis and differential thermal analysis have been used to study the Elemental analysis, homogeneity and stability of the samples.
- ☺ Temperature dependence of low-field ac susceptibility and dc magnetization measurements is used to identify the double transition behavior and nature of the magnetic phase transitions in the present investigated samples. The critical behavior of the magnetic phase transition has been investigated in detail.
- ☺ Magnetic isotherm and ac susceptibility (field and frequency dependence) measurements have been performed to study the

magnetization behavior and to identify the low temperature magnetic state in these reentrant magnetic alloys.

- ☺ Zero-field and in-field electrical resistivity measurements have been performed to i) identify the various scattering mechanisms that are responsible for total resistivity, ii) understand the effect of field on various scattering mechanisms and iii) quantify whether any interrelationship exists between magnetic and electrical properties.
- ☺ Longitudinal and transverse magnetoresistance measurements have been used to understand the nature of the magnetism and hence to determine various transport parameters.
- ☺ Systematic isothermal annealing studies have been carried out in order to investigate the thermal effects on magnetic and electrical properties.

A systematic investigation of amorphous $Fe_{100-c}Zr_c$ binary and $Fe_{90-x}Mn_xZr_{10}$ ternary alloys permitted us to draw a number of conclusions as follows: Amorphous nature is confirmed through detailed structural characterization. Existence of double transition (reentrant behavior) is observed for all the compositions. The magnetic parameters such as high field susceptibility, spin glass behavior, coercivity, local magnetic anisotropy, nonsaturation behavior, obtained from low temperature magnetization data are consistent with the presence of *mixed state*, which is discussed in terms of competing FM and antiferromagnetic (AFM) exchange interactions. A detailed analysis of the magnetization data reveals that spin wave excitations, single particle and local-spin-density-fluctuations (LSDF) contribute dominantly to the thermal demagnetization of spontaneous magnetization. The temperature dependence of magnetization behavior has been explained in terms of nearest neighbor Heisenberg model. However, the applied field ≥5kOe suppresses the LSDFs. Consistent with the prediction of *spin fluctuation* model, the spin fluctuation coefficients (which reflect the spin fluctuation contribution to resistivity in different temperature ranges) increase with increasing Mn concentration. A detailed study of magnetic order-disorder phase transition by various analyses shows that presence of disorder has no influence on the critical behavior. The magnetoresistance and electrical resistivity provide additional information about the magnetic properties of these alloys due to their close dependence on the magnetization. Development of spontaneous resistive anisotropy (SRA) close to the ferromagnetic ordering temperature confirms the emergence of polarized field at that temperature. Structural relaxations upon isothermal annealing show the development of nanocrystalline particles and have considerable changes in Curie temperature and the spin-glass transition temperature. All the investigated amorphous alloys are *weak itinerant* ferromagnets.

Part of this thesis work in a more comprehensive manner is presented in journal articles, which are listed in list of publications.