Summary: Research programme- based on the work done at Saha Institute of Nuclear Physics - with relevant publications reference and citations –

Focus: " Physics of magnetism at low temperature and at high magnetic field – role of ordered moments in the absence of true long range order, new materials, instrumentation,"-

We have undertaken a systematic experimental studies on advanced magnetic materials of intermetallic alloys and oxides to investigate the nature of magnetism, namely, "what happens to the ordered moments in the absence of true long range order" for the samples prepared through structural, quenched, chemical process.

I) Cluster Glass: Clusters are formed due to short range ordering of atomic magnetic moments at some temperature near T_C . Below T_C, these clusters show spin glass like freezing behavior, in contrast to canonical spin glass systems in which isolated atomic moments freeze out .Due to the finite range FM ordering below T_C in each cluster, the system also shows features similar to those found in reentrant spin glass (RSG) systems. Therefore study of static and dynamic response of cluster glasses (CG) in contrast to canonical spin glasses (SG) or re-entrant spin glass is important. The authors have initiated a new approach, namely, the field-cooled coercivity, to observe the progressive freezing of the clusters below Curie temperature due to thermal activation for $La_{0.5}Sr_{0.5}CoO_3$. (Physical Review B **54** 9267 (1996) – citation: **132**). In a related work on $La_{0.875}Sr_{0.125}MnO_3$ system, the effect of particle size on the magnetic and transport properties has been correlated by us with the domain state (cluster), changes in the Mn-O-Mn bond angle, and Mn-O bond length in comparison with bulk sample. (Physical Review B **68**, 054432 (2003)- citation **38**). A similar cluster glass approach was shown for nanocrystalline spinel oxide CoFeZnO- (J. Mag. Magn. Mater. **248** 101 (2002)- citation **24**).

II) Spin compensation temperature in Ferrimagnets- Spinel oxide: The interest in the spinel oxides stems from the fact that spinel ferrite (nano) particle is an ideal small particle magnetic system whose crystal chemistry can be controlled unlike pure metal particles where the crystal chemistry is basically fixed. Spinel structure is conductive if the octahedral site is occupied by a low valence transition metal ion. Our approach was to focus on search for large magneto resistance in spinel oxides to investigate magneto resistance associated with order-disordered transition and also to study spin compensation temperatures. We have shown that $Co_{0.2}Zn_{0.8}Fe_{1.6}Ti_{0.4}O_4$ spinel ferrite is more conductive (J.applied Physics **93** 2780 (2003)). In a ferrimagnet there are two non-compensating sub-lattices of local dipoles called M_A and M_B with

M_A # M_B . For $T<T_C$, $M = M_A + M_B$. In a very few cases, $M_A = M_B$ at a temperature called the spin compensation temperature(T_K). Between T_C and T_K , the contribution of the other sub lattice dominates. In the classical theory of ferrimagnets Neel had predicted an unusual thermal variation of such spontaneous magnetization. In most magnetic materials, the magnetization increases monotonically with decreasing temperature. In order to search for spin compensation temperature, the theoretical prediction of the magnetic properties is difficult in view of the various types of exchange/super exchange interactions involved, particularly, in metal ion substituted oxides. Thus in real material the identification of T_K itself has been a challenging task. Even though Neel predicted theoretically T_K in 1948, the first experimental observation was reported in certain metal oxides in 1962 that too without much details and more recently in molecular based magnets (1999) were reported. Independently, during this period, we have reported (Solid State Communication **103** 269 (1997)& J.Magn.Magn.Materials **202** 359(1999)) the first experimental study for ferrimagnets ($Fe_2Mo_{1-x}Ti_xO_4$) where not only the spin compensation temperature due to two sub- lattice contributions for magnetizations are different under zero field-cooled condition and field cooled process, but also the magnetisation with temperature maintains symmetry with reference to the direction of the applied field

II) Inter metallic alloys -Spin Glass features in the long range ordered systems: The physical significance of the second harmonic components (2ω) in the reentrant spin glass state (RSG), as an evidence for the presence of long range order in RSG has been proposed for the well ordered NiFeAu alloys. (Solid State Communication 82 591 (1992) ,Citation-**22**). We have investigated dynamical susceptibility of metamagnetic materials with $ThCr_2Si_2$ structures (UCu_2Ge_2, CeFeAl) where there is no chemical or crystallographic disorder in search of spin glass like features in the ordered systems. (Physical Review B 46 6236 (1992), Citation-**23**; Physical Review B 50 1084 (1994)- citation **31**)

III) Instrumentation: We have proposed a new experimental technique called the “Enhancement of AC susceptibility” by applying second ac field with different frequency on ac susceptibility measured by the conventional method. This is some what similar to method used in tape recorder where a high frequency signal is mixed with audio signal to ensure greater “fluidity” in magnetic media. (Rev. of Sci. Instrum. **67** 789 (1996),ibid **68** 2834 (1997)).This method will help us to study domain nucleation, and domain wall motion and separate out the intra-domain contribution from the bulk magnetization. We have applied this method to a number of FM systems like Gd, reentrant spin glasses, and canted spin systems. The data have been interpreted with the help of simulation models based on dry friction like pinning of domain walls for systems having FM domain structure (J. Magn. Magn. Materials 224, 210 (2001))

IV) Small particle magnetism We have also shown the increase of magnetization below anti-ferromagnetic ordering, unconventional relaxation in AFM nano particle, enhancement of surface magnetization in $CoRh_2O_4$, lattice expansion and magnetism in $MnCr_2O_4$, a simple electrical conductivity scaling law of the spinel oxide $Mn_{0.5}Ru_{0.5}Co_2O_4$ and are considered to be the original work.(Physical Rev B **68** 134433 (2003) Physical Rev B **69** 054430 (2004) Phy.Rev B **72** 094405 (2005) Phy.Rev B **73** 144413 (2006)). Core-shell model has been proposed not only for the above spinel oxides but also for crystalline and amorphous $La_{0.67}Ca_{0.33}MnO_3$ nano particle system. (J.applied Physics **95** 113909 (2009)).

V) Recent work 2008-09: New observation in Inter metallic alloys are NTCR, NTE, GMR: The Negative Temperature Coefficient of Resistance (NTCR) is generally observable only in amorphous metals, highly disordered (structurally and/or chemically) systems, liquid metals and quasicrystals. Further, in general, the oxygen-based compounds that exhibit Negative Thermal Expansion (NTE), are semiconductors or insulators but there are also a very few metallic magnetic compounds which show NTE [e.g., Invar alloys.] . We have recently reported the unusual result of NTCR, in an ordered crystalline intermetallic compound, $GdPd_3B_{(1-x)}C_x$. This is an ordered and non-Kondo, AFM system. Our work suggests that the chemical disorder is not the causative factor for NTCR observed in this system. These ordered alloys also exhibit unusual isotropic negative thermal expansion (NTE) behavior which till now has been known in oxide systems (insulators) and some invar alloys. This work also establishes some synergy between isotropic NTE and PTCR in these intermetallic compounds. (Applied Phy. Letter **92** 261913 2008, Euro Phy. Letters **84** 47007 (2008). We have also observed negative giant magnetoresistance (GMR) in cubic $TbPd_3$(-30%), which is the highest among the RPd_3 series(Applied Phys. Letter **94** 172509 (2009).

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