Competing Magnetic Interactions in Rare Earth Nanocomposite and Amorphous Systems

Personnel: R. W. McCallum, M. J. Kramer, and K.W. Dennis

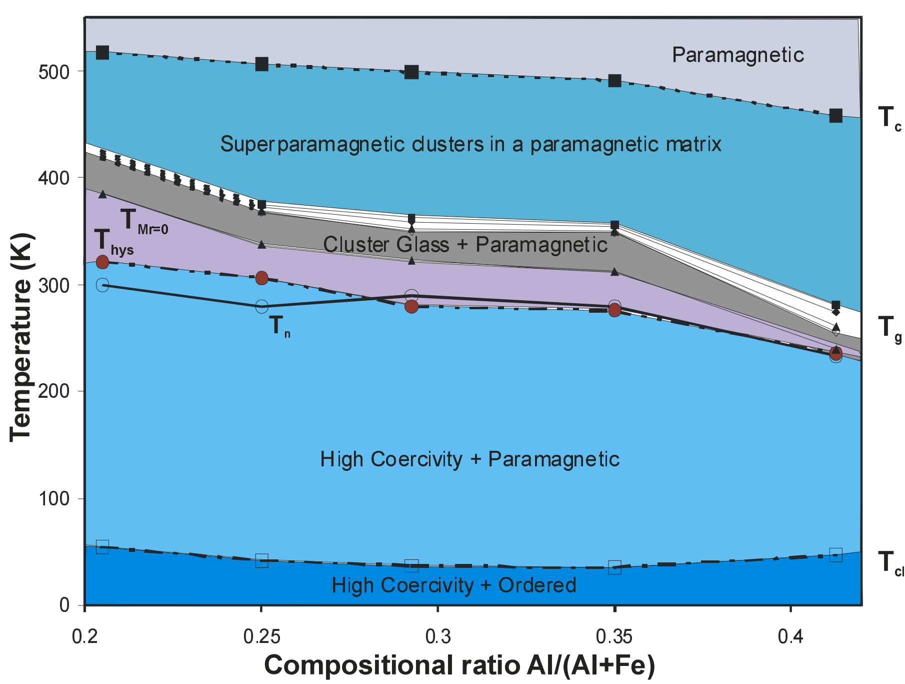
Abstract:

This project is directed at understanding the role of disorder on magnetic and electronic properties with primary emphasis on rare-earth alloys and compounds. In order to perform these studies in a systematic manner it is necessary to develop an understanding of the factors controlling the introduction of site-specific defects in crystalline materials, microstructural development in nanocrystalline materials, and short-range order in disordered alloys. By controlling these factors, length and energy scales may be systematically varied so that competing interactions may be studied. The measurement of magnetic and transport properties enhances the understanding of the both extrinsic and intrinsic properties of these materials and provides a means to characterize the local structure of the materials.

Recent Results:

In nanocrystalline and amorphous magnets, the magnetic properties are critically dependent on the ratio of the magnetic exchange length L_{ex} to the characteristic structural size D. L_{ex}, determined from the size below which the magnetostatic energy associated with uniform magnetization of a volume of material, is less than the increase in exchange energy required to demagnetize that volume. For a nanocrystalline material D is the grain size, while for an amorphous material it is determined by fluctuations in the composition or short-range order within the glass. Thus, for amorphous materials in particular, control of both composition and processing conditions is critically important.

Magnetic studies of the bulk metallic glasses (BMG) with compositions in the region $Nd_{60}Fe_{30-x}Al_{10+x}$ have been conducted using DC magnetization from 4.2 to 873 K and fields up to 5 Telsa. AC measurements have been made over five orders of magnitude in frequency. The magnetic properties of the material are strongly dependent on temperature and reflect a material that magnetically is highly inhomogeneous. The magnetic phase transitions as a function of composition are summarized in the fig-



Magnetic phases as a function of composition and temperature for $Nd_{60}Fe_{30-x}Al_{10+x}$ melt spun ribbons.

ure. As the material is cooled, superparamagnetic clusters form within a paramagnetic matrix. This region is bounded by a frequency dependent spin glass freezing of the clusters. Measurements of the thermomagnetic remnant magnetization indicate that there is a region with essentially zero coercivity that extends about 50K below the spin glass transition. The onset of measurable hysteresis either in M vs. H curves or by comparing field cooled and zero field cooled M vs. T measurements occurs about 50K below the disappearance of the thermoremnant magnetization. This is coincident with the antiferromagnetic ordering of a fraction of the matrix. Below the antiferromagnetic ordering the coercivity of the ordered fraction of the sample increases rapidly and greatly exceeds 5T at low temperature. Ordering of the remaining paramagnetic matrix occurs at about 50K. Structural studies of the BMG have been made using transmission electron microscopy with transmission electron microscopy/electron energy loss spectroscopy (TEM/EELS) and synchrotron radiation techniques. The Nd-Fe-Al alloy consists of clusters of approximate 1.2 nm diameter in an amorphous matrix. The cluster size is about 60% of the unit cell dimension of the expected primary solidification phase. The magnetic measurements indicate there are both ferromagnetic clusters with T_c ~ 500K and antiferromagnetic clusters with T_n ~250 K. Given the observed cluster size, the bulk values of the magnetic anisotropy are inconsistent with the observed coercivities. The large values of the coercivity require that the anisotropy be significantly enhanced by interfacial effects, indicating that surface anisotropies play a dominant role in these materials. This enhancement of the anisotropy has been observed in isolated nanoparticles by other researchers.

Significance:

The magnetic properties of Nd-Fe-Al BMGs reflect the role of different length scales in magnetism. The ordering temperature of clusters within the BMG appears to be determined by short-range order on the scale of a few nearest neighbor distances. As a result, different clusters order at distinctly different temperatures. The magnetic exchange length in the glass is larger than this characteristic structural length. As a result, the clusters are interacting so that they undergo a spin glass transition rather than a superparamagnetic blocking. Given the characteristic cluster sizes, the temperature dependence of the magnetic properties can not be explained in terms of normal volume anisotropy but must include interfacial energies. Exchange coupling between antiferromagnetic and ferromagnetic clusters may provide a large contribution to this energy.

Future Work:

In order to investigate the association of the clusters in the BMG with subcritical nuclei of the expected solidification phases, other members of the class of glass-forming alloys of the form RE-TM-X where RE is a rare earth element, TM a transition metal and X can be Al, Au, Ag, Cu, Ga, and Si will be investigated. For substitutions of other Pr or Sm for Nd, the magnetic properties of the equilibrium phases are distinctly different from those of the Nd, which, if our hypothesis is correct, should significantly vary the magnetic properties of the BMG. Preliminary measurements of $Pr_{60}Fe_{30}Al_{10}$ indicate that the temperature dependence of the coercivity is considerably different than that of the corresponding Nd BMG. Detailed measurements of the composition dependence of the magnetic properties of the Pr BMG and the associated equilibrium phases will be carried out. Cluster formation in the liquid and the glass for both Nd and Pr BMGs will be studied with the aid of pair distribution function analysis utilizing high-energy synchrotron radiation.

Interactions:

Strong interactions exist with the other programs in the Magnetism Focus Area. Close collaborations with L. H. Lewis of Brookhaven National Laboratory (BNL) and D. J. Branagan, Idaho National Environmental and Engineering Laboratory (INEEL) are supported by the CSP Nanostructured Magnetic Materials. Ongoing international collaborations with Argentina and Vietnam have been enhanced by funding from The National Science Foundation (NSF) International Programs Office.