The continuing attractions of magnetism

S. N. Kaul
School of Physics, University of Hyderabad, Hyderabad 500 134, India

Here I discuss, in brief, some of the scientifically exciting and technologically important aspects of magnetism, identify the areas that hold great promise both from basic and applied points of view and suggest a future course of action which will ensure that the scientific and technological activity in the country keeps pace with the developments taking place in the rest of the world.

EVER since the discovery of lodestone (magnetite), magnetism has occupied the centre stage in the scientific and technological development of countries round the globe. Everlasting interest in the field of magnetism stems from a wide variety, and the richness, of phenomena it offers from the fundamental point of view and from the fact that most of these phenomena have been, and continue to be, exploited for technological purposes. In view of the major advances that have taken place in this field of condensed matter science particularly during the past three decades, the statement that magnetism is in a period of renaissance is no exaggeration. Several perplexing aspects of magnetism continue to intrigue scientists worldwide. Some of these aspects are briefly described under different headings and the current situation in India in these newly emerging areas of magnetism is also summarized. Several frontier areas are identified and a future plan for action is suggested.

Amorphous magnetism

By virtue of the fact that no two atomic sites are equivalent and no long-range atomic order exists in amorphous solids, crystal field varies from point to point and each magnetic ion has a preferred alignment for its moment along a *local* 'easy axis' determined by the *local* crystal field. As a consequence, the direct Heisenberg (isotropic) exchange interactions compete with the *local* uniaxial anisotropy to produce new types of magnetic order (under certain conditions) such as, speromagnetism, asperomagnetism and sperimagnetism, besides those (spin glass, mictomagnetism or cluster spin glass, ferromagnetism, ferrimagnetism and paramagnetism) prevalent in solids with regular crystal lattice. Owing to many attractive properties, e.g. low magnetic losses, high permeabilities, low coercivity, low magnetic anisotropy and high resistivities, amorphous ferromagnets have found applications[1] in pulse power sources, digitizers for computers, identification markers, pressure and torque sensors, and magneto-optic recording. Though the field of amorphous magnetism is about 30 years old, many basic questions remain unanswered. For instance,

(i) Why should magnetic properties of certain amorphous alloy systems be extremely sensitive[2] to small additions of impurity atoms?
(ii) Why a majority of amorphous ferromagnets prefer an *fcc*-like[3] nearest-neighbour coordination?
(iii) Why do the Curie temperature and saturation moment have far greater values[3] in several Co-based amorphous alloys than in *hcp* Co?
(iv) How can one justify the existence of spin glass order[4] in some Fe-rich amorphous alloys?
(v) Based on the knowledge of properties of Fe-, Co- and Ni-based amorphous alloys, can one predict the magnetic state[5] of amorphous Fe, Co and Ni?
(vi) What is the nature of magnetic excitations in spero-, aspero- and speri-magnets?

Work in this area is being actively pursued by the research groups at Central University Hyderabad (CUH), IITs, TIFR, IISc and Institute of Physics, Bhuvaneswar.

Magnetism in multilayers and sandwiches

Metallic multilayer structures (sandwiches and superlattices) *consisting* of magnetic layers (Fe, Co, Ni, NiFe) separated by nonmagnetic spacer layers (Cr, Ru, Cu, Au, Ag) exhibit unusually high values (typically 30%; the highest value of 100% reported[6] for Co/Cu multilayers) of saturation magnetoresistance[7] when the current flows in a direction parallel to the layers and the direction of magnetic field also lies within the plane of the layers. This effect has come to be known as giant magnetoresistance (GMR). GMR is extremely sensitive to interlayer moment alignment and this property can be exploited to read information from a magnetic disc. The GMR in some 3*d* transition metal/noble metal multilayers is accompanied[8] by a large magneto-thermopower (GMTEP) and a linear relationship exists[9] between GMTEP and the inverse of GMR. A number of theoretical models proposed[10] to understand GMR have enjoyed only a limited success so far. A microscopic theory of spin-dependent scattering is called for.

Research in this newly emerging area is still in its infancy in India with just two groups, one at University of Pune and the other at IIT, Bombay, engaged[11] in such pursuits.

Surface and low-dimensional magnetism

Low-dimensional magnetic systems including surfaces, interfaces and thin films, have attracted considerable attention during the past decade because the lowered symmetry and co-ordination number in the surface layer lead to a variety of exotic phenomena such as localized electronic surface states or surface resonance states, a narrowed *d* band, magnetic moment enhancement, large perpendicular or 'in-plane' magnetocrystalline surface anisotropy and complex magnetic ordering, and as such open up new vistas for practical applications.

The theoretical prediction that nonmagnetic 3*d* metals like Ti and V and 4*d* metals like Ru, Pd and Rh should possess ferromagnetism as monolayers still awaits experimental confirmation. Though the full-potential linearized augmented plane wave method[12], based on the local spin density functional theory, has successfully explained magnetic moment enhancement and in some cases, surface magnetic anisotropy quantitatively, there is still need for a microscopic theory which can adequately describe the role of surfaces in magnetic reversal processes and treat magnetic anisotropy, magneto-optic and thermal effects properly.

To my knowledge, only a single theory group at S. N. Bose Institute, Calcutta, is involved in this type of activity.

Magnetism in granular solids

Granular magnetic solids, consisting of single-domain ferromagnetic particles embedded in an immiscible insulating[13] (e.g. Fe–SiO_2, Ni–Al_2O_3) or metallic[14] (e.g. Co–Cu, Fe–Cu, Co–Ag, Fe–Ag, etc.) medium, have been extensively studied in the recent years. Isolated magnetic particles are found in materials with a metal volume fraction (x_v) less than the percolation volume threshold (x_p) of about $x_p = 55\%$. A host of single-domain characteristics and other nanostructure-induced enhanced properties have been discovered. These properties also include *isotropic negative* GMR which appears only in granular ferromagnetic systems[15] (in which noble metal forms the immiscible medium, e.g., Co–Ag, Co–Cu, Fe–Ag, Fe–Cu), and (Fe–Ni)–Ag with the volume fraction of ferromagnetic particles $x_v < x_p$. Within the region of $x_v < x_p$, GMR varies with x_v and reaches a maximum[16] value of 85% at 5 K and 25% at 300 K when $x_v \approx 25\%$. The theories proposed for GMR in multilayers are *specific* to them and hence cannot be applied to granular magnetic solids. This area of magnetism is full of promise as far as the technological applications are concerned and challenging from the fundamental point of view.

An important area such as this has escaped the national attention so far.

Magnetism in ultrafine particles

Ultrafine particles (UFP) (20–500 Å in size) of 3*d* transition metals (Cr, Fe, Co, Ni) and their alloys (e.g. Fe–Cr) exhibit unusual[17, 18] magnetic properties. For example, (i) bcc Cr particles, 20–70 nm in size, are ferromagnets[19] with $T_c \simeq 850$ K as contrasted with bulk Cr, which is an antiferromagnet with $T_N = 310$ K, and (ii) coercivity (H_c) of magnetic UFP attains its maximum value, which is far greater than that of bulk counterparts, when the particle size lies in the single-domain range. H_c values as high as 1250 Oe have been obtained[18] in $Fe_{95}Cr_5$ UFP with an average diameter of 210 Å (which is close to the single domain size in bulk Fe) and the temperature coefficient (TC) of H_c in $Fe_{100-x}Cr_x$ UFP with $x \leq 20$ is much smaller than that of bulk Fe in the temperature range 20–450°C. Higher coercivity and lower TC of H_c make these materials serious candidates for several technological applications. Magnetic excitations in these materials remain unexplored and so remain localization and percolation phenomena. The anomalous properties of UFP listed above have eluded a satisfactory explanation so far. Very recently, amorphous Fe–Ni–B and Fe–P–B UFP have been successfully prepared by the chemical reduction method[19]. Amorphous nature of these particles adds a new dimension to the already complex problem. Research in this frontier area is yet to start in India.

Magnetism of dilute systems, re-entrant phenomena and percolation

When a ferromagnet/antiferromagnet is magnetically *diluted* by *randomly* replacing magnetic atoms (ferromagnetic (FM)/antiferromagnetic (AF) bonds) by non-magnetic ones (AF/FM bonds), a critical concentration (x_c) of non-magnetic atoms (AF/FM bonds) or the *percolation threshold* is reached above which FM/AF state is unstable and the spin glass (SG) order sets in. For x just below x_c, the spin system exhibits a transition from paramagnetic (PM) to FM (or AF) state at a critical temperature T_C (or T_N) which is followed at a lower temperature T_{RE} by another transition from FM (or AF) to the re-entrant (RE) state. This type of behaviour has been observed in a large number of spin systems[20] regardless of whether they are crystalline or amorphous, insulating or metallic, ferromagnetic or antiferromagnetic. Randomly site- and bond-diluted magnetic systems provide a fertile testingground for percolation theories but no significant progress has been made in this direction. The basic questions that have not found any satisfactory answers so far are:

(i) What is the nature of the RE state?
(ii) Are the FM–RE and PM–SG transitions true phase transitions in the thermodynamic sense?
(iii) What is the exact structure of the infinite percolating cluster at and above x_c?

The research groups at CUH, TIFR, IISc, IITs, BARC and SINP, Calcutta are actively engaged in this area of magnetism.

Phase transitions and critical phenomena

The field of critical phenomena enjoys the rare distinction of being one of those areas of condensed matter physics in which experiments have preceded and strongly motivated theoretical understanding of the basic phenomena and the theoretical developments, in turn, have paved way for a number of novel experiments. As a consequence, considerable progress has been made in understanding critical phenomena in many magnetic systems, particularly those with annealed disorder. By comparison, the nature of phase transitions and the associated critical-point phenomena in quenched-random site- and bond-diluted spin systems with composition near the percolation threshold (i.e. in strongly quench-disordered systems) and in systems with weak or strong local random magnetic anisotropy have remained obscure.

Study of phase transitions and critical phenomena in new exotic materials such as multilayers and superlattices, thin-film and low-dimensional materials, ultrafine particles and granular magnets, is expected to unravel a wide spectrum of exciting phenomena, e.g., dimensionality crossover, uniaxial or random magnetic anisotropy crossover, long-range (dipolar, RKKY etc.) interaction effects, surface and finite-size effects, new fixed points and new universality classes.

Critical phenomena in spin systems with or without quenched disorder have been, and are being, extensively[21] studied by the research groups at CUH, IIT, Kanpur and IIT, Kharagpur, IISc, Bangalore and Indian Association for the Cultivation of Science, Calcutta.

The Invar and anti-Invar problem

The Invar behaviour is characterized by small but nearly constant thermal expansion over a wide temperature range around Curie (T_c) or Neel (T_N) temperature, large spontaneous volume magnetostriction, large high-field susceptibility, unusually large pressure dependence of magnetization and T_c or T_N, and appreciable variation of elastic constants, Young's and bulk moduli with temperature. This behaviour is not confined to the traditional *fcc* FeNi alloys alone but is more widespread; Invar properties are exhibited by materials with *bcc*, hexagonal or even amorphous structure as well as by rare earth (RE)-transition metal compounds with Laves phase structure (e.g., $\mathrm{RECO_2}$, $\mathrm{REMn_2}$) or compounds like the hard ferromagnet $\mathrm{Fe_{14}Nd_2B}$. Invar materials such as FeNi, FeNiCo ('Super-Invar') and FeCoCr ('Stainless Invar') are used in bi-metals, precision tools and instruments, laser sources, seismographic devices and shadow masks in high-resolution TV tubes. Temperature-independent elastic behaviour has been found in FeNiCr ('Elinvar') and FeNiCoCr ('Super-Elinvar') alloys. These materials find applications in time recording instruments, reeds and reed relays and delay lines. Antiferromagnetic FeCrMn Elinvar is used as hair spring and construction material for 'antimagnetic' wrist watches.

Just as the Invar behaviour is associated with a temperature-independent smaller than 'normal' thermal expansion in a magnetically *ordered* state ($T < T_c$ or T_N), Anti-Invar behaviour is characterized by a temperature-independent *larger* than 'normal' thermal expansion in magnetically *disordered* state ($T > T_c$ or T_N). While low-temperature moment-volume instabilities (MVI) play a crucial role in Invar materials, high-temperature MVI have a direct bearing on anti-Invar behaviour. Anti-Invar properties have recently[22] been found in a number of *fcc* binary and ternary alloys of 3*d* transition metals (e.g. in $\mathrm{Fe}_x\mathrm{Ni}_{80-x}$ $0 < x \leq 70$), for which anti-Invar effect vanishes as the Invar composition $x = 65$ at.% is approached, and in $\mathrm{Fe_{50}Ni}_x\mathrm{Mn}_{50-x}$ alloys).

Though the Invar effect was discovered nearly a century ago, its origin is still a challenging problem of solid state magnetism. Anti-Invar is a relatively new phenomenon and has not found any satisfactory explanation till now. Another poorly understood aspect of Invar behaviour is that the value of spin wave stiffness measured by inelastic neutron scattering experiments is considerably larger than that deduced from bulk magnetization measurements.

Invar behaviour in amorphous magnetic systems has formed a major part of the research activity for the group at CUH.

Areas of interest

In the years to come, R & D efforts should, in my opinion, concentrate largely on the areas of magnetism such as amorphous magnetism, magnetism in multilayers (sandwiches and superlattices) , surface and low-dimensional magnetism, magnetism in ultrafine particles and granular materials, dilute magnetism and re-entrant phenomena, percolation and critical phenomena, garnet bubble films, magneto-optic recording materials, permanent magnetic materials, magnetostrictive materials, Invar problem and itinerant-electron magnetism, valence fluctuation and heavy fermion systems, Kondo lattices and magnetic semiconductors.

Future plan

The following measures are called for if R & D activity in condensed matter science in our country has to catch up with that in the developed countries.

1. Existing research groups need to be strengthened in terms of both manpower and equipment; each group should have at least one major experimental facility, complete in all respects, and manpower trained to exploit full potential of that facility.
2. Duplication of major facilities among different groups must be avoided.
3. In order to achieve major breakthroughs in S & T with optimal use of manpower and equipment, research groups with similar/overlapping interests should work together with a common (global) goal in mind.
4. Advanced instrumentation and electronic repair centres need to be set up in different parts of the country for maintenance/repairs of imported equipment and for indigenously fabricating/developing sophisticated scientific instruments at substantially lower cost.
5. Apex bodies set up in various subjects by the funding agencies like DST, UGC, CSIR and DAE to recommend funding for, and monitor, research projects should draw membership from active research groups in the discipline concerned.
6. The agencies such as DST, CSIR and UGC should bring out national directories, wherein complete information about the groups engaged in applied research and the type of expertise they possess is compiled, and make them available in open market so that industries can have a free access to such material. This will help establish strong linkages between educational institutions, national laboratories and industries and thereby ensure full utilization of the scientific and technological manpower available in the country.

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