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A Study of Antiferromagnetic Nanoparticle Systems in Magnetic Moment Distribution

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ABSTRACT: The aim of this study is to the structural, thermal and magnetic characterization of synthesized two lines ferrihydrite, six lines ferrihydrite and a composite of nickel oxide and two lines ferrihydrite nanoparticles are presented. Average crystallite sizes determined by x-ray diffraction line broadening are close to average particle sizes determined by transmission electron micrographs. This observation indicates that each particle is a crystallite. Transmission electron micrographs also show that particles are of arbitrary shapes and sizes. Both types of ferrihydrite nanoparticles are found to decompose to hematite on heating in air. All three investigated systems are antiferromagnetic and found to exhibit super paramagnetic behaviour due to finite size effect. Zero field cooled susceptibility is seen to peak at particular temperatures whereas field cooled susceptibility decreases monotonically with increasing temperature. Both curves are found to bifurcate at particular temperatures. Below this bifurcation temperature, the magnetization relaxes slowly and so hystereses in the M-B loops are observed. But above the bifurcation temperature any hysteresis in the M-B loops are not seen because the relaxation of magnetization is very fast. In this region, systems are said to be in super paramagnetic state. Deep analysis of magnetization data in super paramagnetic state on each system is presented. Magnetization as a function of applied magnetic field data on two and six lines ferrihydrite nanoparticles are fitted to the modified Lange in function. Estimated fit parameters are found to depend on range of applied magnetic field. That is, these fit parameters do not truly characterize the data. This analysis ignores distribution in particle magnetic moment caused by distribution in particle size and shape. Keeping this issue in mind, the magnetization data are fitted to the modified Lange in function considering a distribution in particle magnetic moment. This time, estimated fit parameters are found to be almost independent of range of applied magnetic field and so truly characterize the data.

The magnetization as a function of applied magnetic field at several temperatures in super paramagnetic region are analysed for both types of ferrihydrite nanoparticles. In this way, particle magnetic moment distribution present in each system is also estimated.

KEYWORDS: Antiferromagnetic Nanoparticle Systems, Magnetic Moment Distribution, Nickel Oxide, Ferrihydrite Nanoparticles.

I. INTRODUCTION

The surface effect has a significant impact on the magnetization of magnetic nanoparticles. Size and surface effects have a significant impact on the behaviour of magnetic nanoparticles. It is commonly known that these systems exhibit unusual behaviour. A phenomenon like superparamagnetism can only be observed in extremely small magnetic particles. Paramagnetism-like behaviour can be seen in this cluster of microscopic magnetic particles. Several studies have been done on the superparamagnetic properties of ferro and ferrimagnetic materials. The smallest details on the surface are in a state of disarray. Saturation magnetization diminishes with decreasing particle size as a result of this. Antiferro-magnetic particles, on the other hand, behave differently in the same circumstance. There is no magnetic moment in a bulk antiferromagnetic. It is possible to detect a magnetic moment in antiferromagnetic nanoparticles due to the disordered surface moments, but the core provides no magnetic moment. With decreasing particle size comes an increase in the magnetism of antiferromagnetic particles. Small magnetic particles can also be used in a variety of technological fields. Magnetic recording material is the primary use of these systems. To record a specific digital signal, the magnetic moments of particles are lined up in a specific direction and the signal is recorded digitally. With time, this direction of the magnetic moment of a particle must stay the same. The stored data will be lost if the same occurs over time. To boost storage density, recording media contain smaller particles. However, particles must be big enough to prevent the system from entering a superparamagnetic state. In a solution called Ferro fluid, iron and ironimagnetic nanoparticles are suspended in a non-magnetic fluid. Particles' dipolar interactions are greatly reduced when fluid is present. Additionally, magnetic nanoparticles play a critical role in the delivery of specific drugs. For this, magnetic

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nanoparticles that are biocompatible and loaded with pharmaceuticals are created. An external magnetic field is employed to focus these particles on the damaged location inside the body. In the future, cancer and tumour cells may be treated with this technology. Uniform distribution and accompanying adverse effects are avoided by using this strategy.

Hyperthermia can also be utilised to kill cancerous or tumorous cells, and is referred to as such. Tumor cells are more vulnerable to heat than normal cells, and they die at a temperature of 41 to 47 degrees Celsius. In order to obtain the necessary temperature, external alternating magnetic fields are used to generate heat from magnetic nanoparticles delivered to the tumour..

II. NANOMATERIALS

There are a variety of ways in which nanomaterials are discovered and utilised. Naturally, some of these substances can be found as well. Natural nanomaterials such as ferrihydrite and ferritin are the greatest examples. In contrast, the first one may be found in dirt, whereas the second one can be found in the blood of mammals. While some nanomaterials are synthesised in the field, the vast majority are created in the lab by a variety of chemical and physical techniques [7, 8]. Nanomaterials have piqued the interest of scientists and engineers from a wide range of fields in recent decades due to their unusual properties and potential usage in a variety of technological applications. Because of their size and surface effects, these materials behave differently than their bulk form. Energy bands are seen in bulk materials. In these materials, the number of atoms exceeds Avogadro's number. For practical purposes, the electronic energy levels in energy bands are so close together that these levels are regarded as being continuous. The electrical, optical, and magnetic properties of several bulk materials may now be explained using this theory. The movement of conduction electrons is limited if the bulk material's size are lowered to the nanoscale range. The de Broglie wavelength is now bigger than the nanomaterials' size in this case. There are now discrete electronic energy levels, and the band theory no longer applies. For the quantum size effect to be realised, the average distance between energy levels must be greater than the thermal energy. The surface atoms of any crystal are subjected to a variety of problems. However, compared to the total number of atoms in a bulk crystal, the number of atoms lying on the surface is insignificant. Because of this, bulk crystals are not affected by surface influences. However, if any dimension of the crystal is decreased to the nanometer range, a considerable fraction of the total atoms emerge on the surface. When defining the overall behaviour of the material, these surface atoms cannot be neglected.

PROPERTIES OF NANOMATERIALS

Reduction in size of crystalline solids alters its overall behavior. The variation in important features of solids due to reduction in size is summarized as follows.

1 Physical property

Interatomic forces hold atoms together in solids. In order to break through the interatomic force fields, these atoms must reach melting point first. Surface disturbances reduce the average strength of these internal forces. Because of this, nanoparticles have lower melting points than their bulk counterparts.

Germanium nanowires, on the other hand, have a melting point of only 650 degrees Celsius, while bulk germanium may reach temperatures of 9300 degrees C. Dislocations, another type of lattice defect, can also affect bulk materials. It is possible to eliminate this flaw by lowering the material's dimensions. As a result, nanoparticles are more durable than their bulk counterparts. Polycrystalline copper with an average grain size of 50 micrometres is five times as hard as 6 nanometer copper particles.

2 Chemical properties

Valance electrons of atoms participate in chemical reactions. We can increase number of surface atoms by reducing dimensions of a material. This increases the chemical reactivity. Because of same reason the catalytic activity of different catalysts is also enhanced.

3 Electrical properties

The quantum size effect governs electron conduction in nanomaterials. Thermal energy must be smaller than the typical distance between electronic energy levels in order to notice this effect. A nonlinear relationship exists between metal current and applied voltage in this scenario. It's actually a stair-like structure. Researchers have also looked into the quantum dot's I-V characteristics. In this, no current flows till the applied voltage reaches , where C is capacitance of

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the particle. The current through the system increases in steps at applied voltage and so on. This phenomenon is called Coulomb blockade.

4 Optical properties

The band gap of optically active materials decreases as a result of the quantum size effect. As a result, the absorption peak is located at shorter wavelengths of illumination. The crystal sizehas been shown to alter the colour of materials as well. Metallic nanoparticles' colour changes are mostly caused by surface plasmon resonance. When the incident radiation's wavelength grows larger than the nanocrystals', a resonance arises. Nanowires exhibit the same effect. However, two surface plasmon resonance modes can be found here. One is transverse, whereas the other is longitudinal. At the nanoscale, gold and silicon appear red, whereas their bulk counterparts are yellow and grey.

5 Mechanical properties

Mechanical strength of a material is lowered by presence of several crystal defects. But nanomaterials are usually free from such defects and so have high mechanical strength. For example, in bulk form, steel is much stronger than carbon. But carbon nanofibers are found to be much stronger than the steel.

III. SUPERPARAMAGNETISM

Ferroelectric crystals have a preferential crystallographic direction for the magnetization vector. It is referred to as a "easy axis" in the industry. The magnetization vector can sometimes be challenging to align in a specific direction. It's referred to as a "hard axis." An external magnetic field can be used to rotate the magnetization vector in the opposite direction of the easy axis. The magnetocrystalline anisotropy energy Ean is the energy associated with the direction of the magnetization vector with relation to the easy axis. There are a few additional preferred directions for the magnetization vector in crystals. Uniaxial anisotropy is the simplest form of magnetocrystalline anisotropy. In this case, the system's magnetization vector has only one preferred direction of orientation. With its hexagonal structure and magnetic properties, cobalt is an excellent example. c axis is easy and perpendicular c axis is hard for this system. The easy and hard axes are the same. The anisotropy energy of magnetocrystalline anisotropy is

$$E_{an} = -(K_2 \cos^2\theta + K_4 \cos^4\theta + K_6 \cos^6\theta + - - - - - -)V, \qquad (1.1)$$

where K2, K4, K6, - - - - are anisotropy constants and V is volume of the crystal. Here θ is angle between easy axis and particle magnetic moment vector. The first term is usually much stronger than rest of terms and so

$$E_{an} \approx -K_2 V \cos^2\theta. \qquad (1.2)$$

The anisotropy energy is directly proportional to the volume of the crystal, as seen in the above equation. When illustrated in Figure 1.1, for a particular crystal, the anisotropy energy Ean changes as the angle is varied. An energy barrier K2V exists between the two lowest-energy orientations at = 0 and 1800. Both external magnetic fields and increased temperature can be used to break through this barrier.

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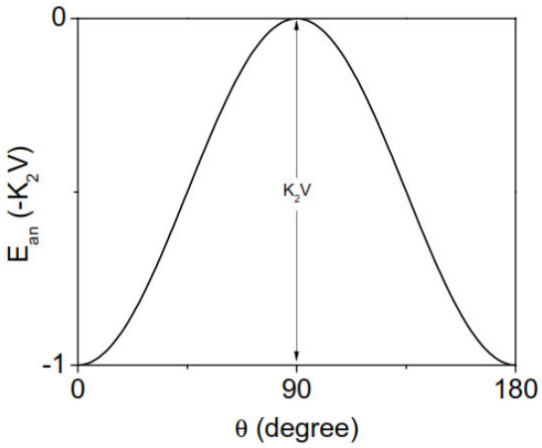

Fig. 1: Variation of magnetocrystalline anisotropy energy Ean as a function of angle θ for an uniaxial crystal.

Anisotropy energy can be lowered by decreasing the crystal's size. Magnetic anisotropy flips between two least-energy orientations if thermal energy, which is on the order of kBT, becomes significantly more than it. Small crystals act as paramagnetic ions when gathered in a mass. Superparamagnetism is the name given to this phenomenon. Particles or crystallites are the most common terms for these tiny crystals, which are often referred to as crystals.For uniaxial particles probability that the magnetization vector flips to the opposite orientation separated by an energy barrier K2V at temperature T is proportional to Frequency ν with which the magnetization vector flips between two orientations of minimum energy must be proportional to this probability. That is

$$\nu = \nu_0 e^{-\frac{K_2 V}{k_B T}}, \qquad (1.3)$$

where ν0 has a value between 108 and 1012 Hz [26]. This is known as Arrhenius law which describes dynamics of superparamagnetic particles. The relaxation time τ can be given by

$$\tau = \tau_0 e^{\frac{K_2 V}{k_B T}}. \qquad (1.4)$$

When V is included in the exponent, the transition to superparamagnetic state occurs at a certain temperature. When particles in an assembly are distributed in size, the relaxation time will only be on the order of experimental observation time for a small subset of the particle sizes. The magnetization process can only be helped by these specific particles. Changing the frequency of the supplied ac magnetic field can alter the experimental observation time required for ac magnetization measurements. Measurements of dc magnetization, on the other hand, take only a few hundred microseconds to complete.It occurs, for , when the energy barrier equals to approximately 25 kBT. The temperature TB at which this happens is given by and called as blocking temperature. Let us consider a collection of N non-interacting particles, each of magnetic moment and volume V at temperature T in an external applied magnetic field B~ . The energy of each such particle is

$$\epsilon = -\vec{\mu_p} \cdot \vec{B} + E_{an} = -\mu_p B \cos\alpha - K_2 V \cos^2\theta, \qquad (1.5)$$

where α is angle between Projection of along applied magnetic field direction is Its expectation value, using Boltzmann statistics, is

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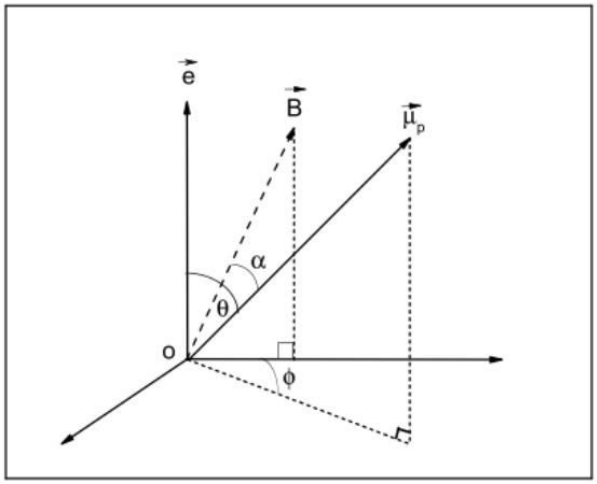

Fig. 2: Details of various angles used in calculation of magnetization.

IV. MAGNETIC MATERIALS

We live in a world full of magnetic materials. The development of magnetism is largely due to the tireless work of numerous individuals who have made significant contributions to our current understanding of this fascinating phenomenon. A long distance has been made in the field of magnetic materials since our ancestors first learned about lodestone. Many modern technology rely on the magnetic properties of materials like these. Magnetization, coercivity and remanence determine whether or not a magnetic material can be used in a specific application based on its magnetic properties. Different sorts of magnetic materials, such as oxides and alloys, can be used. Nearly every aspect of our daily lives involves the use of magnetic materials in some form. For example, AINiCo, hard ferrites, Sm-Co magnets and NdFeB magnets are used in a wide range of industrial applications. Electrical engineering and power electronics find many uses for soft magnetic materials with high saturation magnetization, higher permeability, and very low energy dissipation in the generation, transportation, and consumption of electrical energy. In addition to telecommunications and electronics, soft magnetic materials are used. Permanent magnets for loudspeakers, generators, video and audio tape storage, digital data storage in ferrite memory cores, drum hard discs, and floppy discs, to name a few uses for magnetic materials. As a result of recent breakthroughs in magnetism, several materials that could be used in advanced technologies, such as GMR read heads and MRAMS, have a bright future. Using both the electron spin and charge, diluted magnetic semiconductors (DMS) can be employed in spintronics and magnetoelectronics. DMS are a remarkable class of materials where magnetic ions are integrated into a semiconductor host lattice. As a result, magnetic materials have been and will continue to be an essential element of the technological revolution.

MAGNETISM IN MATERIALS

All materials are magnetic in nature. The magnetic properties arise mainly due to the electrons present in the atom, which have small magnetic moment by virtue of their motion. Nucleus also has a small magnetic moment, but it is insignificant to that of the electrons and it does not affect the gross magnetic properties.

1 Magnetic Moments of Electrons

There are two kinds of electron motions namely the orbital and the spin motion and each has a magnetic moment associated with it. The orbital motion of an electron is similar to the current in a loop of wire without resistance, and both are equivalent to the circulation of charge. Therefore, the magnetic moment of an electron due to its orbital motion is given by,

$$\mu = (\text{area of loop})/(\text{current in emu})$$

If e is the charge on the electron in esu and c is velocity of light, then e/c is the charge in emu. The current or charge passing through a given point per unit time is thus

$$((e/c)(v/2\pi r))$$

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Therefore,

$$\mu_{orbit} = \pi r^2 \left(\frac{ev}{2\pi rc}\right) = \frac{evr}{2c}$$

The angular momentum of an electron is given by,

$$mvr = nh/2\pi$$

Combining these equations, we have the orbital magnetic moment for an electron in the first Bohr orbit,

$$\mu_{orbit} = eh/4\pi mc$$

Spin is a universal property of electrons in all states of matter at all temperatures. The electrons behave as if they were spinning about its own axis, as well as moving in an orbit about the nucleus and associated with this spin are definite amounts of magnetic moments and angular momentum.

2. Diamagnetism

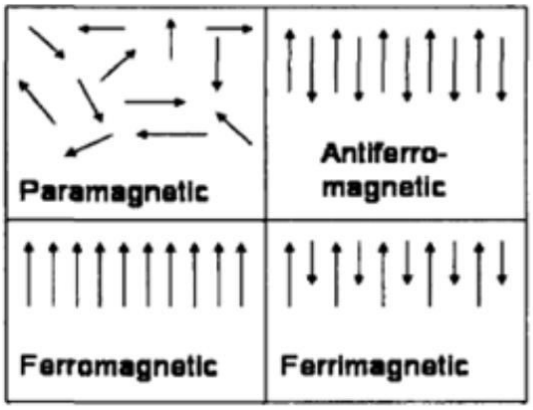
There is an inherent connection between dimagnetism and all the materials. A material's diamagnetic properties are due to the interaction of two electrons in the interior of the material's shells. The field induces a very faint magnetization in them, and it points in the opposite direction of the applied field. An applied magnetic field induces a shift in electronic orbital motion,

which results in this type of magnetism. As stated by Lenz's law, the induced currents produce an induced magnetic flux that is in the opposite direction of the applied magnetic field.. Even though diamagnetism is inherent in all magnetic materials, magnetic atoms obscure it.

Temperature has little effect on the diamagnetic material's negative susceptibilities, which can range from 10- 6 to 10- 6.

3. Paramagnetism

In paramagnetic compounds, the magnetism comes from the atomic constituents' permanent magnetic moments. If there are no interactions between these moments, then they can go in any direction they choose. This phenomenon is known as free atom paramagnetism. A net magnetic moment is connected with the atoms that make up a paramagnetic material. Magnetic fields induce an induced magnetization along the field's axis when applied to an object. Magnetization decreases as temperature rises. The variation in magnetization as a function of field becomes increasingly linear with increasing temperature. It was French physicist Pierre Curie who first discovered that paramagnetic susceptibilities change as a function of temperature in 1895. He argued that, on the whole, the populace was to blame.



(a)

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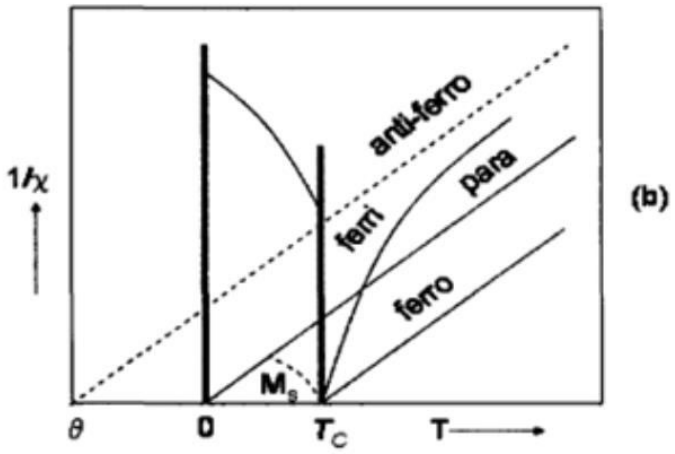

Figure 3: (a) Alignment of magnetic moments in different magnetic materials and (b) variation of the inverse susceptibility with temperature of para-, antiferro-,ferro- and ferrimagnetic materials.

V. ANTIFERROMAGNETISM

In antiferromagnetic materials, the magnetic moments of adjacent atoms align in opposing directions, resulting in a net zero magnetic moment for the bulk material. However, the degree to which this susceptibility varies with temperature is odd. They have a slight positive susceptibility at all temperatures. Neel is credited with developing the theory of antiferromagnetism. The two sublattices undergo spontaneous magnetic ordering as in ferromagnetic materials below a certain temperature, dubbed the Neel temperature, T/vi, but the net magnetization is zero due to the opposite directions of the sublattices magnetizations. A little amount of net magnetization can be detected in the presence of an external magnetic field. Antiferromagnetic materials become paramagnets when heated above the Neel temperature limit. Figure 3 depicts the magnetic moment alignment and temperature dependence of antiferromagnetic material susceptibility.

VI. CONCLUSION

Magnetization data on a superparamagnetic nickel oxide and two lines of ferrihydrite nanoparticles is analysed by taking into account the distribution of the nanoparticles' magnetic moments. In determining the concentration of different components in a magnetic nanocomposite, the analysis described here is extremely useful. Anisotropic effects can potentially alter the magnetization process in magnetic nanoparticles. An anisotropic impact is most significant when it is magnetocrystalline. In the superparamagnetic state, when the anisotropy energy is bigger than the thermal energy, this dependence is frequently neglected. Anisotropy energy's impact on magnetization is also ignored in this study. A future study of magnetic nanoparticle magnetization data may include taking this effect into account. For systems with more than two constituents, the analysis of magnetization data on magnetic nanocomposite. Nanoparticles of antiferromagnetic materials show some surprising and unusual behavior. These behaviors are not observable in ferro and ferrimagnetic nanoparticles. Magnetization of antiferromagnetic nanoparticles should be described by the modified Langevin function. But fitting the magnetization as a function of applied magnetic field data in superparamagnetic region results unphysical values of different fit parameters. For example, this leads particle magnetic moment of 5 nm nickel oxide particles to be about 2000 μB. Such values are much larger than the expected values due to uncompensated spins on particle surface. There is also an alternate method to estimate particle magnetic moment of magnetic nanoparticles. In this method, low temperature saturation magnetization is divided by particles number. But this method also leads large value of the particle magnetic moment. To explain this observation, a multisublattice model has been proposed.

But still there is no experimental support for this model. Magnetization of antiferromagnetic nanoparticles is expected to be only function of temperature T and applied magnetic field B and should scale as (B T). But experimental observations do not show this scaling. Particle magnetic moment of antiferromagnetic nanoparticles is found to increase with increasing temperature. A thermo-induced contribution is reported to be responsible for this behavior.

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