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Proceedings of TiMP 2021

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Preface

The Springer International Conference on Trends in Modern Physics (TiMP) 2021, the third annual conference of the Physics Department of Assam Don Bosco University (ADBU), was organised from 26 to 27 February, 2021, by the department, in collaboration with Indian Association of Physics Teachers, after successfully organising TiMP 2019 and TiMP 2020. A large number of participants, from various universities, colleges and institutes of India and abroad, presented around hundred research papers in the event. Due to the restrictions imposed by the COVID-19 pandemic, the conference was held in hybrid mode, with half of the participants making their presentations online and the remaining half presenting offline, in person. The organisers of the conference made all possible efforts to ensure that every delegate is able to seamlessly access all the presentations, irrespective of whether the presenter or the presentation is online or offline. To this end, all the offline presentations were also streamed live via web-conferencing and all the posters were made available online. Selected papers of TiMP 2021 have found their place in the proceedings after going through the due processes of peer reviews.

It was a felt need by the department to hold yearly national conferences on TiMP, as in this region there were no such yearly conferences of physics, where young researchers can share their ideas and get suggestions and help from renowned academicians of the country and other parts of the world. It may be noted, in this context, that the Physics Department, ADBU, ever since its inception in 2018, has been working at different levels to popularise elementary as well as advanced physics though various other approaches, like symposiums, workshops, refresher course, etc. The TiMP conference series has not been confined to any specific branch of physics, but, practically, to all the major disciplines of physics with the following underlying philosophy. While it is true that each discipline of physics has become so highly specialised that it is not easily legible to a person of another discipline, we must remember that over the history of the development of modern science, physicists' contributions were not only across different branches of physics but also to various other fields of science. For example, Marie Curie, a physicist, won a Nobel Prize in chemistry, apart from a Nobel Prize in physics. James Watson who got the Nobel Prize for proposing the double helix structure of the DNA molecule was actually inspired by physics Nobel Laureate Erwin Schrödinger's book, "What Is Life?". World Wide Web (WWW) was invented in a physics research institute, CERN. The first computer simulation was developed in nuclear physics. Physicists' contribution to mathematics can be exemplified by the development of calculus by Isaac Newton. Venki Ramakrishnan, a Ph.D. in physics, got the Nobel Prize for his studies of the structure and function of the ribosome which is important in the production of antibiotics. The list of physicists contributing to other fields of science is actually too long. Thus, it is evident that if people from physics can make so many contributions to additional domains of science outside the realm of physics, it is both productive and likely for them, even if they are from specific branches of physics, to contribute to and collaborate with other disciplines within physics. With that philosophy in mind, this multidisciplinary physics conference series was conceptualised and has been being implemented successfully.

We thank the convener of the international conference, Mr. Parag Bhattacharya, together with the co-conveners, Dr. Debajyoti Dutta and Dr. Ngangom Aomoa. We also express our gratitude to all the reviewers, Dr. Lalthakimi Zadeng, Dr. Yubaraj Sharma, Dr. Simanta Chutia, Prof. Sunandan Baruah, Dr. Sumita Kumari Sharma, Dr. Kaustubh Bhattacharyya, Dr. Shantu Saikia, Dr. Debajyoti Dutta, Dr. Ngangom Aomoa, Prof. Atri Deshamukhya, Dr. Subhankar Roy, Dr. Debasish Borah, Dr. Wandahun Longtrai Reenbohn, Dr. Rashi Borgohain, Prof. Pritam Deb, Dr. Pralay Kumar Karmakar, Dr. Ashok Kumar Jha, Dr. Umananda Dev Goswami, Dr. Hemen Kumar Kalita and Dr. Subhaditya Bhattacharya. Finally, we thank all the authors for their contributions in the proceedings.

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Chapter 32
Review on Magnetism in Nanomaterials and Superparamagnetism

Bandana Gogoi and Upamanyu Das

Abstract Nanotechnology plays a prominent role in the fabrication of novel materials by controlling the structure of matter at the nanometric scale changing properties at a molecular level. The particles with nanodimensions change their material properties in a dramatic way showing uniqueness in behaviour with modified properties. In many ferromagnetic materials when the size is reduced to the nanoscale level, the magnetic properties enhance in a unique way, thus leading to a superparamagnetic state. The magnetic moment of the material randomly flips the direction of their magnetization, and the random orientations of magnetic spins inside the particles result in zero remanent magnetization and zero coercivity. An unusual change in the hysteresis loop shows the magnetization curve passing through the origin, showing the state of zero magnetization.

32.1 Introduction

At the nanoscopic dimension, the magnetic behaviours of magnetic nanomaterial show significant differences from those observed at bulk scale with the same chemical composition [1–3]. As the size is reduced material property gradually moves from the regime of bulk material behaviour to molecular-level material behaviour. Drastic changes in properties took place at the molecular level.

With the reduction of size, the basic magnetic properties or magnetism connected to different bulk ferro and anti-ferromagnetic material changes to develop in different modified ways. These changes may be shown to occur from the dimension of the material which becomes comparable to some of the basic fundamental characteristic lengths of one or more of various physical properties that are more relevant to the magnetic properties (e.g. the size of magnetic domains, exchange length etc.). With reduced size to nanoscale level, translation symmetry of the magnetic material breaks giving rise to specific sites with reduced coordination numbers, broken exchange and a higher proportion of atoms on the surface increasing surface effect. A high surface

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to volume ratio brings more close contact with the exterior system. The spin-wave spectrum of the nanometric material also tends to change and this change can be observed as spin-wave energy becomes comparable to the thermal energy which plays a prominent role in developing new magnetic properties [3–12].

Nanoscale magnetism has been basically studied to be developing from unpaired d-orbital electrons as well as the coupling effect of these electrons with nuclear spins. At nanosized dimension, the surface area gets more exposure to the exterior neighbouring system, as a result of which the material achieves higher modified functionalities like increased reactivity, higher catalytic action and decreased melting point due to surface effect. The surface energy increases with increased surface area and hence surface effect dominates over the other observed effects at nanosized dimension. This increased surface energy effect also helps in developing extraordinary magnetic behaviours in magnetic nanomaterials. Particle size plays a significant role in determining the basic material property of any material like magnetic, electrical, optical or electronic property. Therefore increasing interest has been developed to study magnetic nanomaterials in recent times due to their size-dependent properties [13, 14].

Figure 32.1 shows the typical hysteresis loop of any bulk ferromagnetic material. In order to observe the changes of magnetic properties of ferro or anti-ferromagnetic materials with particle size, the magnetic hysteresis curve of normal bulk ferromagnetic material needs to be understood. In normal ferromagnetic behaviour, the curve is observed to have a remanence and a coercive field, i.e. the line of magnetization curve does not pass through zero or origin.

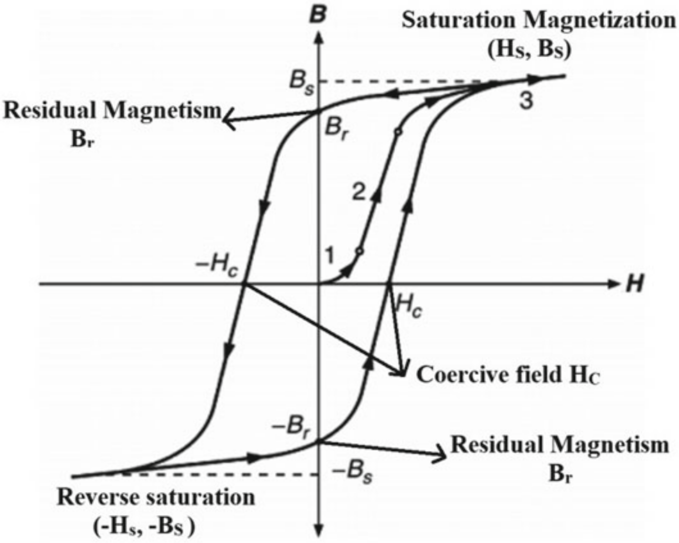


Fig. 32.1 Magnetization curve of ferromagnetic material in a bulk state

The development of uniqueness in the magnetic property of magnetic materials can be studied to be originated from the division of magnetic material into distinct and well-separated magnetic domains with magnetic moment alignment. The change in magnetic moment directions in magnetic domains may correspond to the total cancellation of the magnetic moment or may tend to minimize the total average magnetization to become nearly zero.

There are various interaction terms in a magnetic system that contribute to the total internal energy of a magnetic material and can be expressed as

$$E_{tot} = E_{ex} + E_A + E_{ms} + E_{ext} \tag{32.1}$$

Exchange interaction (E_{ex}) is responsible for the establishment of magnetic order in magnetic materials. This interaction arises from a quantum effect due to the indistinguishability of the electron.

$H = -2JS_iS_j$ where J is the exchange constant. S_i and S_j are spins.

Magnetostatic energy (E_{ms}) or dipolar energy is the measure of the magnetic energy of a magnetic sample because of its own magnetic field. This field is the demagnetizing field that arises from the divergence of magnetization.

Magnetic anisotropy (E_A) is crystallogenic in origin. The shape of the sample, the stress in the material and atomic segregation determine the value of magnetic anisotropy energy. The energy of a magnetically ordered sample depends on the relative direction of the magnetization and the structural axes; for example, a solid has an axis along which the energy is at a minimum. The anisotropy energy E_A is written as a function of the direction cosines $\alpha_{1,}\alpha_2$ and α_3 defined in relation to the axes of the crystal.

Uniaxial anisotropy is the approximation that in some samples anisotropy depends only on the angle θ between the magnetization and a given axis. The anisotropy energy per unit volume takes the form

$$E_A/V = K_1 sin^2\theta + K_2 sin^4\theta \tag{32.2}$$

where θ is the angle of magnetization with the single axis, and depending on the magnitude of anisotropy constants K_1 and K_2, the sample can have an easy axis or easy plan.

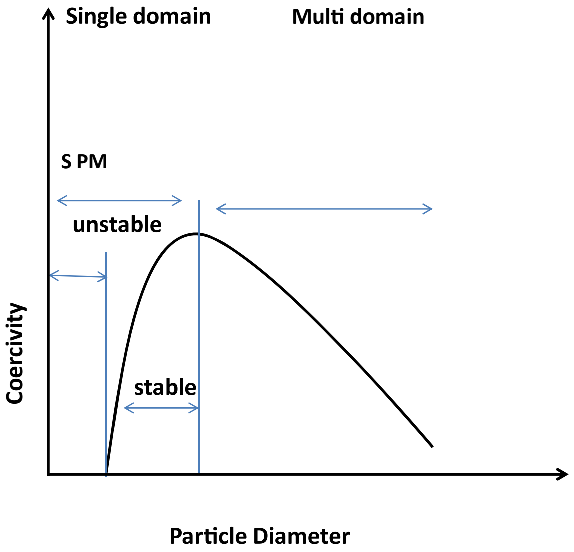
Magnetoelastic energy and magnetostriction is the elastic energy of a magnetic material that arises from the interaction between the magnetization and the strains. Magnetoelastic energy is the increase in the anisotropy energy of magnetic material when submitted to stress. Magnetostriction, the intrinsic property of magnetic material, is the coupling between magnetic and elastic energy, i.e. when subjected to magnetic field magnetic material changes shape [3, 14–16].

Exchange interaction is responsible for the ordering of atomic magnetic moments which causes the atomic magnetic moments to become parallel and showing magnetic ordering in ferromagnetic material. But the presence of other interactions such as anisotropy, dipolar, magnetoelastic etc. leads to the formation of magnetic domains, where the magnetic moments are perfectly ordered [3]. As the size of the particle is reduced, the energy necessary to divide itself into magnetic domains is higher than the energy needed to remain as a single magnetic domain or monodomain [10, 17].

The size of the magnetic material has a great influence in determining its magnetic behaviour, e.g. a ferromagnetic material below a critical particle size (15 nm) can possess a single magnetic domain and can show paramagnetic behaviour above a characteristic temperature called blocking temperature (T_B). With the increase of the size of the magnet, the number of magnetic domains increases, and as a result, the number of domain walls also increases. During the whole process, there is a decrease in magnetostatic energy while there is an increase in the exchange and the anisotropy energies because of the more number of domain walls [8, 18]. This dependency of magnetic property on the size of the magnetic material can be illustrated by considering the coercivity of the magnet and the dependence of coercivity on the size of the magnet as shown in Fig. 32.2 [3, 16, 19].

For very small particles, with a diameter smaller than the critical diameter of superparamagnetism (D_{spm}), the particle shows unstable magnetization with flipping spin and it results in zero coercivity (H_c). For the diameter in the range between D_{spm} and the critical diameter of a single domain (D_{sd}), the magnetic moment shows stable nature and hence coercivity (H_c) does not become zero. Coercivity increases with the increase of single-domain diameter D_{sd} and after reaching the multidomain region with the increasing diameter, coercivity again decreases. Hence the magnet shows the maximum coercivity when the diameter is equal to the single-domain diameter, D_{sd} [8, 10, 20–22].

Fig. 32.2 Dependency of coercivity on the size of the particle diameter of magnetic nanomaterial



32.2 Basic Concept of Superparamagnetism

Superparamagnetism (SPM) is a type of magnetism that develops in small nanoparticles of ferromagnetic or anti-ferromagnetic materials which possess single-domain non-interacting magnetic moment grains. Nanosized material with a single magnetic domain can show superparamagnetic behaviour below T_B (blocking temperature) also when the size is sufficiently reduced below blocking volume (V_B), which is the maximum volume below which superparamagnetism starts at a particular temperature and that possibly arises due to spin-based momentum of the unpaired electrons present in the material [21–23].

The energetic stability of a single magnetic domain was theoretically predicted and established by Kittel in 1946 [24]. Magnetic nanoparticles generally show a preference along the direction where magnetic alignment takes place and are said to be anisotropic along these directions. Nanoparticles generally show uniaxial anisotropy, which means that there are two easy directions of magnetization pointing in opposite directions (antiparallel) and are separated by an energy barrier. For single-domain magnetic material, all the magnetic moments are aligned along the preferred anisotropy axis, therefore the free energy contribution from exchange and anisotropy becomes zero. Hence the magnetostatic energy becomes the only relevant energy term.

The critical diameter of the single domain (D_{sd}) of magnetic material has a close relationship with the anisotropy constant K. For identical saturation magnetization (M_s) single-domain diameter D_{sd} increases with domain wall energy, i.e. D_{sd} is proportional to the domain wall energy. When the size reduction of the particle is sufficiently large then thermal energy overcomes the anisotropy energy. At this stage of magnetization magnetic moment shows fluctuating nature rather than stable nature [8, 20, 21].

At a given temperature, as the size is reduced to a large extent, spin-wave energy modifies and becomes comparable to thermal energy in single-domain non-interacting magnetic grain or particle, and thermal energy becomes insufficient to overcome the spin–spin interaction and can lead to random orientations of magnetic spins inside the particles. The critical diameter D_{spm} is the maximum size below which the superparamagnetic behaviour starts at a particular temperature and the corresponding volume at which a particle goes from blocked to unblocked state is called blocking volume (V_B) [22, 23].

At blocking temperature (T_B)*,* thermal energy overcomes the anisotropy barrier of nanoparticles. Above blocking temperature (T_B)*,* thermal fluctuations dominate and magnetic moments are randomly orient. Nanoparticles with a uniaxial anisotropy randomly flip the direction of their magnetization and show a spontaneous reversal of magnetization when thermal fluctuation is sufficient enough to overcome the barrier potential that is supposed to arise from magneto crystalline in origin and due to magnetoelastic and shape anisotropy. It was Neel [25] who shows that above T_B a stable magnetization cannot be established due to thermal fluctuations acting on small particles, and as a result, the system shows *superparamagnetic* behaviour. The typical time of average laps between two flips is called *Neel-relaxation time* τ_N. If τ_m is considered to be the measuring time of the magnetic effect of a particular nanomagnetic material for its observed magnetic behaviour, then the following observations can be made in a nanomagnetic material [25, 26].

If $\tau_m < \tau_{N,}$ the material is in a blocked state and the magnetization flip does not take place.

If $\tau_m > \tau_{N},$ magnetic flip occurs and magnetic behaviour alters. The material shows superparamagnetism.

This shows that the observed magnetic behaviour in nanomagnetic material depends on measuring time τ_m. In most practical applications the measuring time τ_m is tried to keep constant. The transition between superparamagnetic and blocked state is used to study as a function of applied temperature.

The first and basic theory that describes the basic understanding of nanoparticle magnetism is the *Stoner-Wolfforth model* [14, 27]. In this model, each nanoparticle is considered as an ellipsoidal homogenous single-domain non-interacting grain. According to this theory, depending on the spin configuration nanoparticles may have a single domain, vortex or multidomain state. Nanoparticles in the smallest range of diameter do not behave as stable magnet but exhibit the phenomenon of superparamagnetism [3]. However, this model is suitable at $T = 0$ K and is applicable to nanoparticles with uniaxial anisotropy only. The large surface to volume ratio in nanoparticles enhances the magnetic moment and anisotropy [27].

32.2.1 BasicTheory

The energy expression for single-domain magnetic grain with uniaxial anisotropy in the presence of external magnetic field H can be expressed as the sum of magnetic anisotropy energy and Zeeman energies:

$$E = KV\sin^2(\Phi - \theta) - \mu_0 M_s VH\cos\Phi \tag{32.3}$$

where V is the grain volume of the nanoparticle, K being the uniaxial anisotropy constant parameter and M_s is the saturation magnetization. All the three quantities, external magnetic field *H,* grain magnetization and magnetization easy axis lie in the same plane. Φ represents the angle between magnetization and magnetizing field and θ represents the angle between magnetization easy axis and magnetizing field [10, 17, 25, 28].

In absence of an external magnetic field, two equally energetically favourable directions exist. Both directions are parallel to the energetically favourable spontaneous magnetization direction also called magnetization easy axis for anisotropic magnetic material and there possess the energy barrier ΔE between them in KV. At temperatures higher enough, the thermal energy kT is capable of overcoming the barrier potential and alteration of magnetization direction takes place. While in presence of an external magnetic field, the symmetry of the two magnetization easy axis directions breaks down. When magnetization direction is along the external magnetic field, domain energy of the nanoparticle grain decreases and therefore energy barrier for spin fluctuation becomes high. The reverse is the case when magnetization directs in opposite direction to an externally applied magnetic field, the energy barrier for spin fluctuation decreases and reversal of magnetization takes place spontaneously. So nanoparticles with uniaxial anisotropy flip the direction of their magnetization randomly and spontaneously. The thermally initiated fluctuations of the magnetization direction between the two easy axis directions are called *superparamagnetic (Neel) relaxation* and the typical expression for *Neel-relaxation time* τ_N can be expressed by the Neel-Brown expression as

$$\tau_N = \tau_0 exp\,(\Delta E\,/kT) \tag{32.4}$$

where τ_0 is the length of time and is the function of the characteristic of the material and usually lies between 10^{-12} s and 10^{-9} s. τ_0 depends weakly on temperature and various material parameters such as magnetic anisotropy constant, particle volume and saturation magnetization [10, 16, 17, 25, 28].

In this regard, a definition of blocking temperature T_B can be given as the temperature at which the relaxation time τ_N equals the experimental time τ_m or T_B can be defined as the temperature between the blocked and the superparamagnetic state [25, 28].

Equation (32.4) represents the connection between the time τ_N and the temperature *T.*

$$\text{At}\tau_{N\,=}\;\tau_m \quad T_B = \Delta E/kln\big(\tau_{m/}\tau_0\big), \tag{32.5}$$

A clear distinction between the two states can be expressed as

- The state is blocked when $\tau_m < < \tau_N or\; T < T_B$.
- The state becomes superparamagnetic when $\tau_m > > \tau_N or\; T > T_B$.

The first reveal of single-domain particle magnetization presented by *Stoner* and *Wohlfarth* [14] suggested the existence of high coercivity fields below $T_{B.}$ The anisotropy energy arising from magnetocrystallogenic origin becomes comparable to thermal energy and the direction of the magnetic moment starts fluctuating spontaneously and goes through a rapid superparamagnetic relaxation. The supposed system of uniform non-interacting nanoparticles at $T > T_B$ overcomes barrier energy and the magnetic moments started flipping between the easy magnetization directions. At $T < T_B$, the magnetocrystalline-originated anisotropy energy barrier cannot be overcome by the thermal energy and the magnetic moment of each particle rotates from the field direction back to the nearest easy magnetization axis because of which non-zero coercivity results. The total magnetization decreases with increasing temperature as the nanoparticles and the corresponding easy magnetization directions are randomly

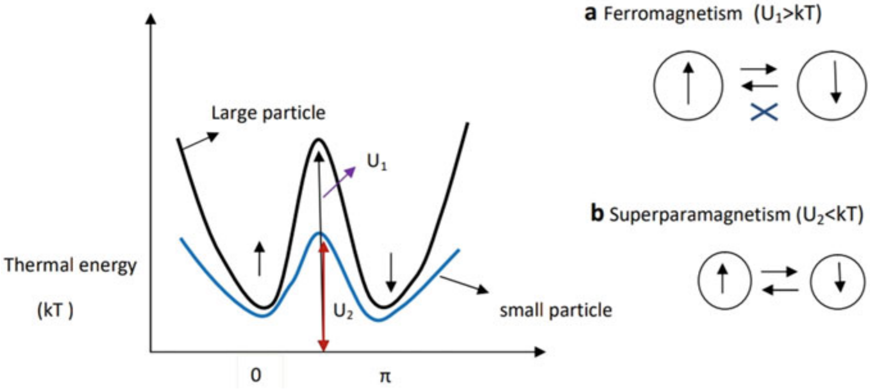


Fig. 32.3 Ferromagnetism in large and small magnetic particle,**a** large particle magnetism (U$_{1.}$ > kT)**,** no spin flipping takes place, **b** small particle superparamagnetism (U$_{2.}$ < kT), with spin flipping

oriented and the randomness increases with temperature [20, 21, 28]. Figure 32.3 shows the ferromagnetism in large particles and superparamagnetism in small nanoparticles.

32.3 Brief Discussions

The typical behaviour of large particle ferromagnetism and small particle ferromagnetism can be observed from the thermal energy transition curve. For two vectors spin directions (vector upward ↑ and vector downward ↓), if spin vector cannot move from one direction to other, there exist some net magnetization and the material shows ferromagnetism, i.e. when exchange energy $U_1 > kT$ vector spin cannot flip or re-orient. In the energy plot (thermal energy as a function of the orientation of the spin) if one spin magnetic moment vector lies in the first stability zone (minimum energy) and if it has to come to the next stability zone (with minimum energy) it has to overcome the energy barrier. The energy barrier is proportional to the grain particle volume (V). The energy barrier potential for a large particle is large. If the spin has to change its direction it has to overcome the large energy barrier. For a small-sized particle of nanodimension, this energy difference is much smaller; therefore, it is easy for the magnetic vector to change its direction crossing the potential barrier. At room temperature also the thermal energy is much greater than the exchange energy between the magnetic vectors [10, 28].

Hence for large particles, at room temperature, the thermal energy is much less than the energy required to cross the barrier, but this energy is sufficiently more than the energy required crossing the barrier in small particles changing the magnetic vector. Hence there is an automatic reversible change in the direction of magnetic vector or spin. At normal temperature also this spin flipping can take place in a

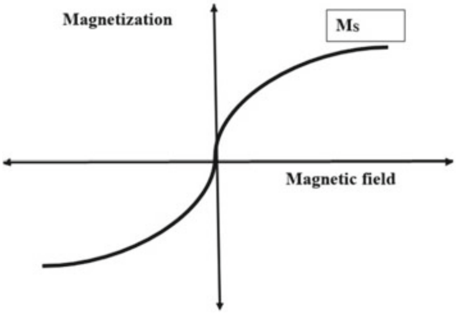


Fig. 32.4 Curve showing superparamagnetism with remanence $M_R = 0$ and $H_C = 0$

nanodimensional particle; hence both the possibilities of stable state are possible in the system. This spin flipping leads to the property of superparamagnetism in the nanosystem. Whenever the energy required or exchange energy U_2 is less than kT (thermal energy) then it can have spin fluctuation and it results in superparamagnetism. Figure 32.4 presents the typical behaviour of the superparamagnetism nature of magnetic nanoparticles, where the magnetization curve passes through the origin.

32.4 Conclusions

For large particle, the hysteresis loop possesses a particular area. When the particle size is reduced sufficiently (around 10–12 nm), these particles do not show the hysteresis loop but a plot that goes through the origin, which is like both remanence and coercive field are zero. This represents the typical paramagnetic behaviour. Although these particles are small they have several moments comprising ions or molecules. These moments are flipping among themselves and the resultant is a paramagnetic behaviour. This is one of the important aspects of the magnetic properties of nanostructures [29]. When the particle size is large it shows the hysteresis loop, but for the same material particle when the size is reduced to nanodimension it does not show hysteresis but passes through the origin with no remanence and coercive field, i.e. superparamagnetism is a function of the size of the particle.

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