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Magnetic properties of Heisenberg clusters

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Abstract. The effects of finite size on the thermodynamic properties of ferromagnetic clusters have been calculated on the basis of a self-consistently solved spin-wave spectrum for clusters of various sizes. The finite size leads to an effective power law for the temperature dependence of the magnetization ($M \sim T^{\alpha}$; $3/2 < \alpha < 3$), and to a neutron-scattering cross section with a wavevector broadened discrete spin-wave spectrum very different from that of the bulk. Predictions of the implications of finite size are extended to the size range of nanoparticles. The effects should be readily observed in experiments.

1. Introduction

The ability to produce metallic clusters consisting of only a few of atoms has opened a new field of research in the border region between molecular and traditional solid state physics [1]. Most theoretical efforts in the physics of clusters have been devoted to the investigation of electronic, magnetic, and structural ground-state properties [1–6]. Recently, the magnetic properties of metallic clusters have received much attention [7–13]. Experimental findings of temperature, field and size dependencies of the magnetic moment of iron and cobalt in clusters [8, 9], have been explained by a theoretical model of superparamagnetic relaxation [11]. However, a number of problems still remain unsolved. The thermodynamic properties of clusters, and their change with size, have previously been addressed in a number of Monte Carlo model studies of Heisenberg [14, 15] and Ising [10,16] spin systems of finite size. These studies predict a rounding of the critical behaviour and non-uniform magnetization profiles for the clusters. However, the Monte Carlo studies did not describe the detailed dynamical behaviour, and no detailed analysis of the low-temperature behaviour of the magnetization has been given. The motivation for the present study is to clarify these questions.

Through a self-consistent determination of the eigenstates of the Heisenberg cluster (the spin-wave spectrum) the thermodynamic properties of clusters with up to 749 spins are calculated. In order to compare with available experimental data of Fe clusters/particles the calculations are performed for clusters with a BCC structure. We find the temperature dependence of the magnetization and its variation with cluster size. The dynamical behaviour, and the corresponding neutron-scattering cross section, are predicted. These properties are yet to be measured.

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2. Model

The clusters are built by spherical cut-outs from a BCC lattice around a central spin. A cluster with 27 spins is illustrated in figure 1. Atoms with different coordination numbers are indicated by the different shades. Figure 1 is further discussed in section 4.1. The exchange interaction between nearest-neighbour spins is modelled using the Heisenberg Hamiltonian

$$H = -\frac{1}{2}\sum_{ij} J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{1}$$

where J_{ij} is the exchange energy constant and $\boldsymbol{S}_i$ and $\boldsymbol{S}_j$ are the spins on sites i and j. The quantum mechanical equation of motion, in a site-dependent random-phase approximation, can be written [17]

$$\omega S_j^+ = \sum_i J_{ij} M_i S_j^+ - \sum J_{ij}\frac{M_i + M_j}{2} S_i^+ \tag{2}$$

where $S^+ = S_x + iS_y$ and $M_i = \langle S_{zi}\rangle$ is the thermally averaged mean value of the spin at site i. In a translational invariant system, where M_i is the same at all sites, the equation of motion can be diagonalized by a Fourier transformation to the reciprocal $\boldsymbol{q}$-space; the eigenstates are plane waves. As translational invariance is lacking for clusters, (2) must be solved numerically in real space; the wavevector $\boldsymbol{q}$ is no longer a good quantum number. One should, therefore, expect modifications of the bulk dispersion curve, $\omega(\boldsymbol{q}) = S(J_0 - J_{\boldsymbol{q}})$, where $J_{\boldsymbol{q}} = \sum_r J_r \exp(-i\boldsymbol{q}\cdot\boldsymbol{r})$ and $J_0 = J_{\boldsymbol{q}=0}$ is the exchange energy constant times the number of nearest neighbours (the coordination number).

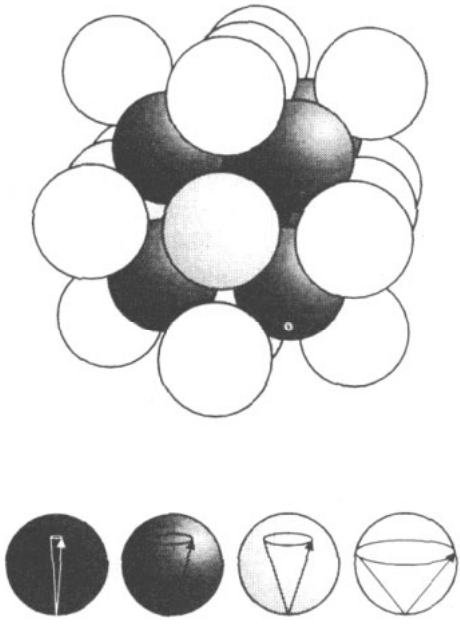

Figure 1. Illustration of a 27-spin cluster set up by a spherical cut-out in a BCC lattice. The spins in the cluster belong to four shells with different coordination numbers as illustrated by the different shades of the spheres. The cones in the lower part illustrate the thermal mean value of the cones on which the spins precess in a classical interpretation of the spin-wave states calculated at $T = 1.0J/k_B$.

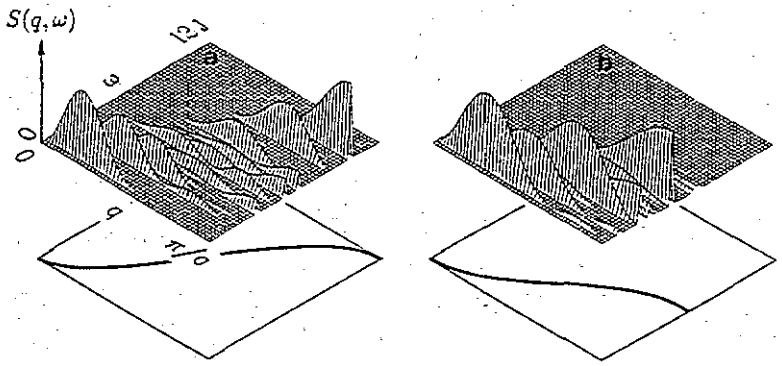

Figure 2. The neutron scattering cross section $S(q, \omega)$ calculated for $\hat{q}$ along the [100] (*a*) and [110] direction (*b*) for a BCC cluster containing 59 spins. The full curves in the lower plane are the bulk dispersion curves in the same directions.

3. Ground-state properties

3.1. Neutron-scattering cross section

A direct diagonalization of (2) for a cluster of N spins gives N discrete eigenvalues (E_p) and the corresponding eigenvectors (ψ_i^p) for S^+. The neutron-scattering cross section at $T = 0$ is [18]

$$S(q, \omega) \propto \sum_p |S^+_{p,q}|^2 \delta(\hbar\omega - E_p) \tag{3}$$

where

$$S^+_{p,q} = \sum_i \psi_i^p \exp(\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{r}_i). \tag{4}$$

In figure 2 $S(q, \omega)$ is shown at $T = 0$ for a cluster with 59 spins for $\hat{q} = q/|q|$ along the [110] and [100] directions. $S(\mathbf{q}, \omega)$ is strongly modified compared to the bulk behaviour, depicted as full curves in the lower plane of the figure. A continuous distribution of q values is needed in order to describe the spin-deviation pattern of the discrete eigenstates. An energy gap (ΔE) between the ground state and the first excited state is seen, and this is of particular importance for the low-temperature behaviour of the temperature dependence of the magnetization. In figure 2(*a*) the highest-energy eigenvalue for the cluster clearly lies below the bulk value. This is an effect of the smaller coordination number at the surface. On the other hand, when spin-waves perpendicular to the most close-packed planes ([110]) are considered (figure 2(*b*)), the highest-energy eigenstates of the cluster are at higher energies than for the bulk. The eigenstates of the cluster cannot be characterized by just one q-vector, as in the case of the bulk. Each cluster eigenstate is characterized by a large number of q vectors, and hence spin-waves in energetically less favourable directions than [110] may be mixed into the cluster state. This explains why the cluster states, in spite of the reduced coordination number in the cluster, can be more energetic than the bulk [110] states.

3.2. The energy gap

Since the thermodynamic behaviour of clusters at low temperatures is dominated by the presence of the energy gap (ΔE) in the spin-wave spectrum, it is important to understand the cluster-size dependence of ΔE. In figure 3 ΔE is plotted as a function of reciprocal size ($1/r_c$); where r_c is defined as the radius of a sphere, with bulk density, that contains N spins. The variation of ΔE with size can be understood by considering the largest possible wavelength in the clusters. For the smallest clusters the first eigenstate resembles a standing wave with $\hat{q}$ perpendicular to the most close-packed planes, i.e. along the [110] direction. It has maximum amplitude, antinodes, at the surface and a node in the centre. The half wavelength, $\lambda/2$, thus equals the cluster dimension. As the size of the clusters increases the antinodes move inward. For increasing cluster size the eigenstates approach the eigenstates of a magnetic field in a continuum sphere (due to the similarity of the equation of motions). The first eigenstate of a magnetic field in a sphere has a node in the centre and antinodes positioned within the sphere [19]. Now, let us assume that ΔE is proportional to the square of the wave number, $\Delta E = JSa^2q^2$, as for the bulk (a is the lattice constant). We further assume that the maximum half-wavelength, $\lambda(r_c)/2 = \pi/q_c$, varies from being equal to the diameter (d) for the smallest cluster ($d = 2r_c = 2r_0$), to being a constant fraction, ξ, of the diameter for large clusters ($d = 2r_c \gg 2r_0$), as is the case for the electromagnetic eigenstates for a continuum sphere. One then arrives at an expression for ΔE as a function of cluster size:

$$\frac{\Delta E(r_c)}{Ja^2S} = q_c^2 = \left(\frac{2\pi}{\lambda(r_c)}\right)^2 = \tfrac{1}{4}\pi^2[\xi r_c + (1-\xi)r_0]^{-2}. \tag{5}$$

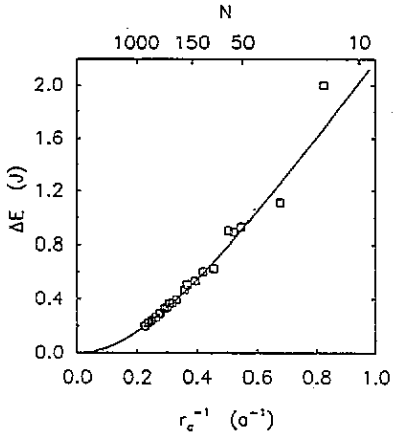

Figure 3. The energy gap, $\Delta E(r_c)$, between the ground state and the first exited state in the spin-wave spectrum as a function of reciprocal size, $1/r_c$. The full curve is the theoretical curve based on (5).

Figure 3 shows $\Delta E(r_c)$ for clusters of various size compared with (5) (for $\xi = 0.71$). This simple picture accounts very well for the variation of ΔE with size. Therefore, (5) allows a prediction of ΔE for cluster sizes beyond the largest ($N = 749$) studied in this work. Assuming the exchange energy constant (J) to be equal to 11.6 meV [20] as for bulk iron, the energy gap of a 725 cluster corresponds to a temperature $\Delta E/k_B$ of about 30 K (see figure 3).

4. Thermodynamic properties

4.1. Magnetization profile

To evaluate the temperature dependence of the magnetization the eigenvalues of (2) must be evaluated, and weighted using the proper statistical weighting (Bose statistics). As the coefficients in (2) depend on M_i the problem must be solved self-consistently. Due to the lack of neighbours at the surface, the spin deviations are expected to be larger in the outer shells than at the centre. Therefore, the thermally averaged spin projections, M_i, must be allowed to vary with position, as well as with temperature. The problem is solved self-consistently in the following way. The eigenvalues (E_p) and eigenvectors (ψ_i^p) are found by diagonalizing (2), starting from an initial assumption of the M_i profile. The thermal mean value of the spin projection on the z axis, M_i, is then found by adding the statistically weighted spin deviations of all eigenstates (p) [21]:

$$M_i = S - S\sum_p |\psi_i^p|^2[\exp(E_p/k_BT) - 1]^{-1}. \tag{6}$$

The diagonalization of (2) is then repeated with the new values of M_i, leading to new eigenvalues, which, after the statistical weighting of (6), lead to a new determination of the profile of M_i. The procedure is repeated until the mean value $M_{mean} = \sum_i M_i/N$ converges. The convergence test is double-sided, such that the final M_{mean} is independent of whether the starting guess is smaller or larger than the accepted solution.

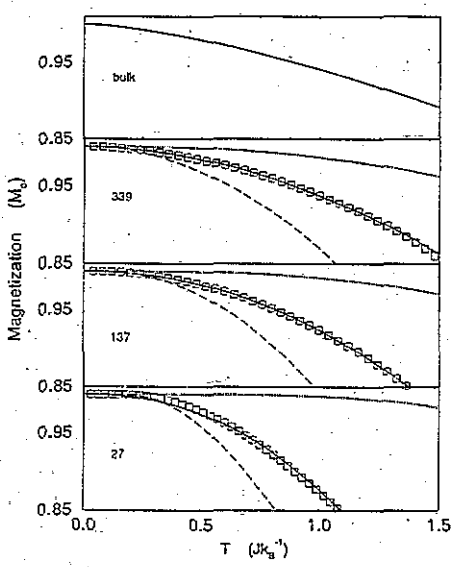

Figure 4. The figure shows the mean magnetization (squares), the magnetization at the centre (dotted) and the magnetization of the surface layer (broken) as a function of temperature for clusters containing 27, 137 and 339 spins. The full curves are best-fit curves using an effective power law (7). The power law describes the behaviour of the large clusters very well. Only for the smallest cluster ($N = 27$) is the expected exponential behaviour evident. The bulk behaviour is shown at the top of the figure.

The temperature dependence of the magnetization determined in the above-described manner is found to deviate substantially from the behaviour of bulk materials. At high temperature the magnetization of the clusters decreases faster with temperature than for the bulk, due to excitation of large-amplitude surface modes. At low temperature the magnetization decreases slower for the clusters than for the bulk [22], as a consequence of the presence of ΔE in the spin-wave spectrum for clusters. These effects are readily seen

in figure 4, where calculated magnetization profiles for different sized clusters are shown. The magnetization of the surface layer and of the central spin, as well as M_{mean}, are shown as a function of temperature. The surface layer is seen to be much softer than the interior shells. The magnetization of the central spin, on the other hand, decreases much more slowly than the bulk magnetization, indicating that the spin fluctuations are transferred to the outer spins in the clusters. As the size of the cluster increases the bulk behaviour (top curve) is approached; the curves for the surface and for the centre move closer together, and the mean magnetization decays more slowly. These effects are also seen in figure 1. In a classical picture the magnitude of S^+ signifies the opening angle of the cone on which the spin precess [17]. The cones in the lower part of figure 1 illustrate the thermal averages of the opening angles in the classical spin precession movement for the four types of spins in the 27-spin cluster at a given temperature. Clearly, as the number of nearest neighbours decreases towards the surface of the cluster the average opening angle of the precession cones increases corresponding to a larger reduction of the magnetization.

4.2. Temperature dependence of the mean magnetization

The energy gap leads to an exponential temperature dependence of the magnetization ($M_{mean} \propto 1-\exp(-\Delta E/k_B T)$) at temperatures ($T \ll \Delta E/k_B$) low enough for only the first excited state to be appreciably populated. However, we find that the temperature dependence of the mean magnetization of the clusters over a larger temperature range (from 0–25% of the mean field Curie temperature, Θ) is well described by the effective power law

$$M_{mean}(T)/M_0 = 1 - BT^{\alpha}. \tag{7}$$

The energy gap (ΔE) and the lacking coordination are the key features of the finite size. Their effects on the temperature dependence of the magnetization can be effectively described by (7) through enhanced (compared to bulk) values of α and B. Furthermore, the expression is simple, has the right form in the bulk limit, and it is in accordance with experimental praxis in the field. Fitting with the above expression gives very good fits except for the smallest clusters for $T < \Theta/25$, where a systematic deviation is observed due to the exponential behaviour at low temperatures. This is illustrated in figure 4 where the fits (using (7)) to the $M_{mean}(T)$ data are drawn as full curves.

The results of the fitting procedure are summarized in figure 5, where α and B are plotted against $1/r_c$. Using the known value ($\alpha = 3/2$) for the infinite system ($1/r_c = 0$) a straight line can be drawn, which allows a prediction of the effective power-law exponent, α, for cluster sizes beyond those for which the full calculations have been performed. The B coefficient varies in a less systematic way with size than α, but it is, generally, somewhat larger than the bulk value. The B parameter depends on the number of nearest neighbours in the system, which varies from spin to spin in the cluster. The mean coordination number increases with increasing cluster size. The scatter of the points in figure 5 reflects the structural variations in the mean coordination number. For example, the clusters with $N = 27$ ($r_c^{-1} = 0.68$) and $N = 89$ ($r_c^{-1} = 0.46$) have, for their size, extraordinarily low effective coordination numbers (see figure 1). Therefore, the temperature dependence of the mean magnetization for these clusters are characterized by relatively large B values. As the size of the cluster increases the B parameter approaches the bulk value (broken curve in figure 5).

4.3. Experimental situation

In recent years several studies on the structure and magnetic properties of clusters and ultrafine particles [8, 9, 20–32] have been reported. Free unsupported clusters of iron were

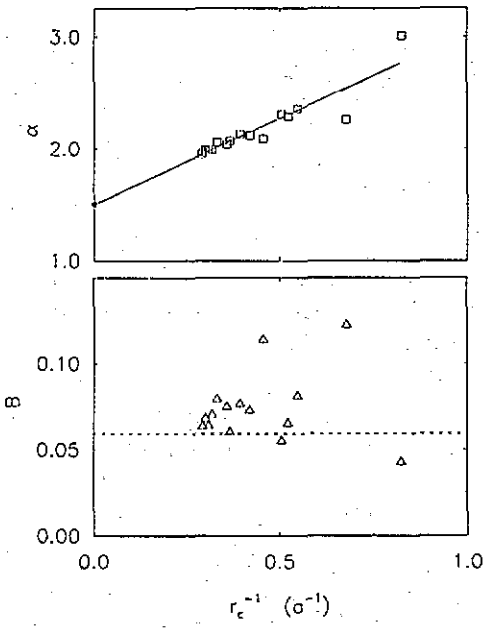

Figure 5. Upper half: the size dependence of the effective power-law exponent, α, obtained from a best fit of (7) to the calculated mean magnetization in the temperature interval from 0–25% of the mean-field Curie temperature (Θ). The full line is a straight line extrapolated to the bulk value. Lower half: the size dependence of the parameter B of the power-law fits. The broken line is the bulk value.

first made by Cox and co-workers [23], who studied the magnetic moment for clusters consisting of 2–17 atoms. The clusters investigated by Cox and co-workers [23] were made by pulsed laser evaporation of an iron rod. The magnetic moments of the clusters were determined by measuring the deflection of the clusters in a Stern–Gerlach experiment. The results indicated that the magnetic moment per atom was at least that of bulk iron. This kind of experiment has now also been carried out by other groups. During the past two years clusters of iron [8], cobalt [9] and gadolinium [35] have been investigated by studying the Stern–Gerlach profiles of the deflected cluster beams. The magnetic moment of the cluster will perform superparamagnetic relaxation [11, 36]. When analysing the results of the deflection experiments [8, 9] on this basis the magnetic moments of the iron and cobalt atoms are deduced to be 10–50% larger (at $T = 0$) than the atomic moments found in the bulk material [11, 36]. The cluster beam studies, in conjunction with the model of superparamagnetic relaxation, have thus confirmed the theoretical prediction [2, 4, 37] that the magnetic moments of cluster atoms should be at least that of bulk atoms. The results indicate that even very small clusters preserve ferromagnetic behaviour.

Structural studies of unsupported clusters/particles are yet to be performed. However, structural investigations of supported clusters have been made. The general conclusion of these studies has been, at least for cluster sizes down to about 1.0 nm, that the structure of iron clusters is the same as for bulk iron, i.e. the BCC structure. Clusters of this size contain about 50 atoms.

It is clear from the available experimental work that the preparation of ultra-small particles/clusters is not an easy task. Each preparation technique has its complicating factors which have to be taken into account when interpreting the results. The ultra-fine particles are typically supported, hence the outermost atoms of the clusters are interacting with 'foreign' atoms, thereby introducing an extra component to the magnetization profile and to the Mössbauer spectrum. These interfacial properties are often difficult to separate from the intrinsic properties of the clusters and, moreover, the properties of the interface will also influence the properties of the interior of the clusters.

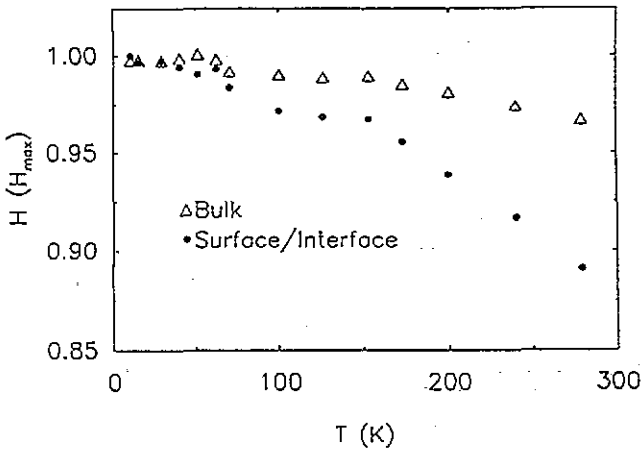

Figure 6. Experimental data of Herr and co-workers [26] for the temperature dependence of the hyperfine fields of atoms in the bulk (triangles) and atoms at the surface (circles) of 6 nm α-iron particles. The hyperfine fields have been normalized by the maximum value in each series.

However, it seems by now to be well established that the magnetization of the surface layers decreases faster, with increasing temperature, than does the magnetization in the interior of the particles. Studies of 3–6 nm iron particles have shown that the Mössbauer spectra should be analysed by using at least two sextets [26, 30, 32]: one sextet with a magnetic hyperfine field typical of that for bulk iron, and one sextet with a magnetic hyperfine field which is about 5% larger than that of bulk iron (at $T = 5$ K) [26]. The latter sextet is ascribed to surface/interface atoms [26]. The 5% enhanced magnetic hyperfine field at a surface is in good accordance with theoretical findings [38]. It is a result of band narrowing, brought about by the lack of orbital overlap at the surface. At elevated temperatures the magnetic hyperfine field (which can be considered to be proportional to the magnetization) decreases faster with increasing temperature at the surface than in the interior of the particles [26, 30, 32]. This has been clearly demonstrated in Mössbauer studies by Herr and co-workers [26] on approximately 6 nm large iron particles produced by an inert-gas condensation technique. Their results are reproduced in figure 6. The observation of the presence of a non-uniform magnetization profile, with the magnetization decreasing towards the surface, is in accordance with the presented model calculations, as well as with previous Monte Carlo simulations [14, 15]. Experiments on thin iron films have, similarly, revealed a more rapid decrease of the magnetization at the surface than in the bulk, as the temperature is increased [39, 40].

5. Discussion

From the previous section it is clear that, as yet, no firm conclusion on the effects of finite size on the temperature dependence of the magnetization of clusters/ultrafine particles has been reached experimentally. To address the most important effects of finite size in the statistical properties we have made a number of simplifying assumptions in the present theoretical treatment. The clusters were assumed to have a simple BCC structure, the magnetic moments (at $T = 0$) to be the same at all sites, and the strength of the exchange interaction the same among all neighbouring spins. Whereas the assumption on the structure seems justified on the basis of experimental results, the two latter assumptions may not be fulfilled in real clusters, as indicated in the above discussion of the Mössbauer studies of small iron particles. Structural relaxations may occur close to the surface, which

will then influence the atomic moments ($g\mu_B S$) and the exchange energy constants (J_{ij}). Such local variations (electronic finite-size effects) will affect the magnetic properties of the clusters. Such electronic finite-size effects should show up in experimental studies in addition to the statistical effects deduced from our model calculations. The calculations have been extrapolated to the size regime of nanoscale particles, where effects due to structural and electronic changes near the surface of the particles will be of less importance for the mean magnetic properties of the particles.

Although experimental studies of the magnetic and structural properties of clusters and ultra-small particles of iron, fabricated by several different preparation techniques, have grown considerably in number in recent years, the rather simple and straightforward experiments that would yield the best tests of the model calculations presented in this paper are yet to be performed, e.g. precise measurements of the mean magnetization as a function of temperature. In order to reduce the problems due to interactions of surface spins with a support, one suggestion for an experimental study that would be valuable for a comparison with our spin-wave calculations, is to collect iron clusters, e.g. size-selected clusters from a cluster beam, in a matrix of a condensed inert gas—quite similar to the procedure employed by Montano and co-workers [29] in their XAFS studies of iron monomers and dimers. The finite-size modifications of the neutron-scattering cross section and the substantial energy gap between the ground state and the first excited state should also be accessible to experimental studies.

6. Summary

We have elucidated the effects of finite size on the dynamics and thermodynamic magnetic properties of clusters. We have chosen a simple model for the exchange interaction to address, especially the statistical properties.

We have found large effects of the finite size on the magnetic properties of clusters. The neutron-scattering cross section is strongly modified compared to the bulk. The eigenstates are wavevector broadened and there is an overall softening of the states due to the importance of the surface. The energy gap between the ground state and the first excited state can be quite substantial, and a simple model accounting for its variation with size is suggested. The magnetization in the clusters is found to be non-uniform decreasing towards the surface of the clusters, and the overall behaviour (the temperature dependence of the mean magnetization) is found to be well described by a power law with a size-dependent exponent larger than the bulk value (1.5).

The results allow theoretical predictions of as yet unmeasured experimental quantities, such as the neutron-scattering cross section, the energy gap in the spin-wave spectrum, and the effective power law for the temperature dependence of the magnetization. The predictions are extrapolated to the size-regime of nanoscale particles.

Acknowledgments

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