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# Analytical solution of the mean field Ising model for finite systems

**Dalía S Bertoldi**[1,2], **Eduardo M Bringa**[1,2] **and E N Miranda**[1,3]

[1] Instituto de Ciencias Básicas, UNCuyo 5500-Mendoza, Argentina
[2] CONICET-Mendoza, Argentina
[3] IANIGLA-CONICET, CCT Mendoza, 5500-Mendoza, Argentina

E-mail: emiranda@mendoza-conicet.gov.ar

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**Abstract**
The Ising model for finite systems, e.g. for clusters of different sizes and crystal lattices, was solved analytically by the mean field approach. The magnetization was calculated from the number of accessible microstates, using the gamma function and its derivatives, unlike the usual solution in the microcanonical which uses the Stirling approximation. We determined a scaling exponent of $\sim 1/3$, which shows how the Curie temperature decreases with decreasing nanoparticle size. Moreover, the model predicts the behaviour of surface and core regions and it explains in simple terms several effects previously observed in experiments and Monte Carlo simulations of small magnetic systems.

## 1. Introduction

During the last few decades much effort has been devoted to the understanding of the physical phenomena appearing in magnetic nanoparticle systems [1, 2]. The development of novel techniques and the refinement of synthesis methods have allowed the experimental study of particles at the nanoscale [3, 4]. The behaviour of nanoparticles is influenced by finite size effects. The crystal lattice, the magnetization processes, the Curie temperature, the supermagnetism, etc, are all affected by the size of the system. Additionally, the behaviour of the surface is different from the behaviour of the core of a nanoparticle due to, among other things, differences in the coordination number and ordering of spins. The competition between core and surface effects determines the state of the particle, which can go from paramagnetic to ferromagnetic.

The Curie temperature is one of the most important magnetic parameters because it describes the stability of the paramagnetic and ferromagnetic phases. There are theoretical models that predict the dependency of the Curie temperature on system size [5–8], but most of them use the entropy of the system, which is not a magnitude that can be changed experimentally in a direct way.

Recently, there has been an increasing focus on the study of ferromagnetic nanostructures due their application in diverse areas of high-density data storage, ferrofluids, colour imaging, devices of UHF and drug carriers for specific drug administration [9, 10]. However, if the system size is lower than a given critical size the useful ferromagnetic properties are lost. Therefore, to determine the technological limitations of current devices, there is a need to know how size controls ferromagnetism.

Experimental studies for different materials show that the Curie temperature in nanoparticles is strongly size dependent and decreases as the particle radius is reduced [11–14]. At the same time, different numerical calculations have shown that at finite temperature the profile of the magnetization across a small particle is non-uniform, with the magnetization decreasing towards the surface [15–17].

As usual in the study of magnetic systems, the Ising model plays a central role in trying to understand the behaviour of such systems. This model has been solved analytically for systems in one and two dimensions [18, 19]. For higher dimensionality different approaches are used, with the mean field approach standing out. At the same time, there are many simulations of finite systems using Monte Carlo simulations [20–22]. For example, in the work by Velásquez *et al* [23], 3D clusters are studied in the framework of the Ising model, using variational and Monte Carlo methods.

Here we propose a new approach: to solve analytically the Ising model for small systems in the microcanonical formalism, using the mean field formalism and considering the presence of surfaces. The main idea is simple. On the

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one hand, the count of the accessible microstates of the system is carried out exactly in closed form, avoiding the Stirling approximation which is inadequate for systems of few particles. On the other hand, a number of non-uniform coordination corrections is introduced in the application of mean field theory. The theory of phase transitions and finite size effects have been discussed extensively within the framework of statistical mechanics (see for instance the book by Brankov *et al* [24] and references therein), but here we aim only to explain some phenomenological properties of nanoparticles.

In the following sections we first obtain a new magnetization expression for small systems, then we apply it to clusters of different sizes, and finally we present a discussion and some conclusions.

## 2. Ising model in the microcanonical formalism

### 2.1. Preliminary considerations

The Ising model consist of $N$ particles with spin $\pm 1/2$ that can be in one of two states, with energy $\varepsilon_+ = -\mu H$ or $\varepsilon_- = \mu H$, respectively. The particles with spin $-1/2$ (namely $N_-$) are located anti-parallel to the field $H$ and those with spin $1/2$ parallel (namely $N_+$).

The total energy of the system is $U = \mu H(N_- - N_+)$ and the magnetization is the difference between the number of particles in the two states $m = \mu(N_+ - N_-)$. Then, the numbers of particles in each state are:

$$N_+ = \frac{N}{2}\left(1 + \frac{m}{N\mu}\right) \tag{2.1}$$

$$N_- = \frac{N}{2}\left(1 - \frac{m}{N\mu}\right) \tag{2.2}$$

and the energy is rewritten as $U = -mH$.

For convenience we introduce a change of variable. Let the net magnetization $M$ be defined as $M = (N_+ - N_-)/N = m/N\mu$, such that the number of accessible microstates can be written as:

$$\Omega = \frac{N!}{N_-!N_+!} = \frac{N!}{[\frac{N}{2}(1+M)]![\frac{N}{2}(1-M)]!}. \tag{2.3}$$

### 2.2. Approximate solution

The first step is to introduce equation (2.3) in the well-known definition of the Boltzmann entropy

$$S = k_B \ln \Omega \tag{2.4}$$

where $S$ and $k_B$ are the entropy and the Boltzmann constant, respectively. Then, applying the Stirling approximation $\ln(N!) \simeq N \ln N$ we obtain:

$$S = k_B\left\{N\ln N - \frac{N}{2}(1+M)\ln\left[\frac{N}{2}(1+M)\right] - \frac{N}{2}(1-M)\ln\left[\frac{N}{2}(1-M)\right]\right\}. \tag{2.5}$$

On the other hand, the temperature is given by the thermodynamic relation $1/T = \partial S/\partial U$ or, applying the chain rule,

$$\frac{1}{T} = \frac{\partial S}{\partial M}\frac{\partial M}{\partial U}. \tag{2.6}$$

Using the above equation, it is easy to show that:

$$\frac{1}{T} = \frac{k_B}{2\mu H}\left\{\ln\left[\frac{N}{2}(1+M)\right] - \ln\left[\frac{N}{2}(1-M)\right]\right\}. \tag{2.7}$$

Solving equation (2.7),

$$M = \tanh\left(\frac{\mu H}{Tk_B}\right). \tag{2.8}$$

There are two contributions to the field $H$: on the one hand, the local field of each spin which is generated by the magnetization of its neighbours, and on the other hand the external field $B$ that affects all the lattice.

The mean field theory supposes that each spin feels an effective uniform field generated by the rest of the spins. A direct summation can be carried out using this hypothesis:

$$H = JMz + B \tag{2.9}$$

where $z$ is the coordination number and $J$ the spin–spin interaction. Finally, the transcendent equation for the magnetization of the system is:

$$M = \tanh\left(\frac{\mu(JMz + B)}{Tk_B}\right). \tag{2.10}$$

Strictly speaking, to assume that the effective field is uniform the lattice must have translational invariance. However, in small systems there is not translational invariance and surface effects are important. To consider these factors, we used a non-uniform coordination number that depends on the position of each atom in the system. On the other hand, in the exact calculation of the number of microstates that is developed in the following section, we avoided the Stirling approximation.

### 2.3. Exact solution

The use of the Stirling approximation is justified in the limit $N \to \infty$, i.e. when the number of particles in the system is very large. However, it generates significant errors for small systems, even a system of $N = 10^8$ particles presents a relative error of 10% with respect to the exact solution for the $\ln(N!)$ [25] which is:

$$\ln(N!) = \ln[\Gamma(N+1)] \tag{2.11}$$

where $\Gamma$ is the gamma function. We repeat the calculation of the previous section using the equation (2.11) and the gamma function derivatives, denoted $\psi^n(z)$ and defined as [26]:

$$\psi^n(z) = \frac{\mathrm{d}^{n+1}}{\mathrm{d}z^{n+1}}\ln[\Gamma(z)]. \tag{2.12}$$

Then, instead of equation (2.5) we obtain:

$$S = k_B \ln\left\{\frac{\Gamma(N+1)}{\Gamma[\frac{1}{2}(N - MN + 2)]\Gamma[\frac{1}{2}(N + MN + 2)]}\right\}. \tag{2.13}$$

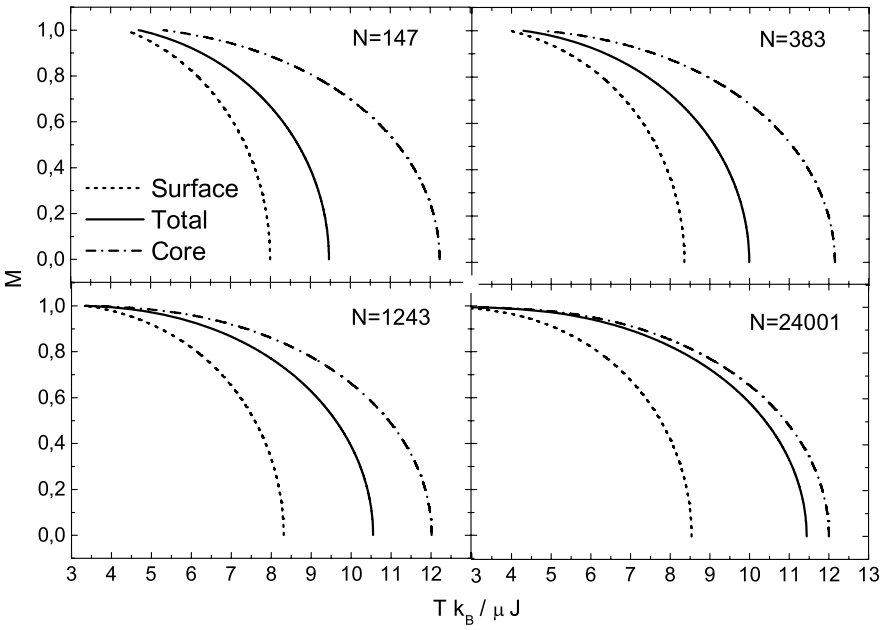


**Figure 1.** Thermal variation of the surface and core magnetization (lower and upper lines, respectively) and net magnetization (solid line) for four different sizes of hcp clusters at $B = 0$. The net magnetization was calculated as the weighted average of the core and surface contributions. Similar behaviour is seen for fcc and bcc lattices.

Using the relationship (2.6) we find that:

$$\frac{1}{T} = \frac{k_B}{2\mu H}\left\{\psi^0\left[\frac{N}{2}(1+M)+1\right] - \psi^0\left[\frac{N}{2}(1-M)+1\right]\right\}. \tag{2.14}$$

Then, replacing $H$ by (2.9) we get a new transcendent equation for the magnetization which has an explicit dependency with the number of particles $N$,

$$\frac{JzM+B}{Tk_B/\mu} = \frac{1}{2}\left\{\psi^0\left[\frac{N}{2}(1+M)+1\right] - \psi^0\left[\frac{N}{2}(1-M)+1\right]\right\} \tag{2.15}$$

where the value of $z$ might not be the same for all the atoms in the system. The equation (2.15) is the central result of this paper; in section 3 it is solved for different cases.

## 3. Magnetic clusters

Finite size effects dominate the magnetic behaviour of clusters, increasing their relevance as the particle size decreases.

Equation (2.15) was used to study the magnetization of different sized clusters. In addition it was possible to establish a relationship between the value of $M$ and the number of particles. The core and surface contributions to the total magnetization have been identified.

Three types of clusters were studied, each one with a different crystal lattice: (1) hexagonal (hcp), with a lattice parameter $a_0 = 0.251$ nm characteristic of cobalt, (2) body-centred cubic (bcc), with $a_0 = 0.287$ nm characteristic of iron and (3) face-centred cubic (fcc), with $a_0 = 0.352$ nm characteristic of nickel. Clusters and the coordination number for each atom were obtained using the LAMMPS code [27]. Clusters were created as spheres of different radius at the origin of the appropriate crystal lattice. The coordination number was obtained by counting atoms within spheres with a cut-off radius between the first and second neighbours' shells, according to crystal lattices.

The coordination numbers at the surface are smaller than in the bulk. Therefore, we differentiated the number of atoms with each coordination and for each of them we calculated the equation (2.15). The net magnetization was obtained by a weighted average of all the contributions.

In figure 1 the magnetization of the surface and core are shown for four different cluster sizes without external field, i.e. $B = 0$. At higher temperatures, i.e. temperatures near the critical temperature, the contribution of the surface becomes smaller than the core contribution. The core contribution was calculated by solving (2.15) with $z = z_{\text{bulk}}$, while the surface contribution is the average of equation (2.15) for each coordination number that satisfies $z \neq z_{\text{bulk}}$. Therefore, the net magnetization is lower than in the bulk case, as a consequence of surface and finite size effects.

The surface magnetization is relevant in very small clusters, where the ratio between the number of atoms at the surface $N_S$ and in the core $N_C$ is greater than unity ($N_S/N_C > 1$), and the net magnetization approaches the surface magnetization. The opposite occurs for large clusters, where core effects are more significant, i.e. $N_S/N_C \ll 1$, and the net magnetization is almost not influenced by the surface. The values of core and surface magnetization change for different crystal lattices, with those lattices that have a greater coordination number showing higher magnetization values.

The Curie temperature of the core ($T_c^C$) is about 30% larger than that corresponding to surface atoms ($T_c^S$) for the cases shown in figure 1, and as a result the

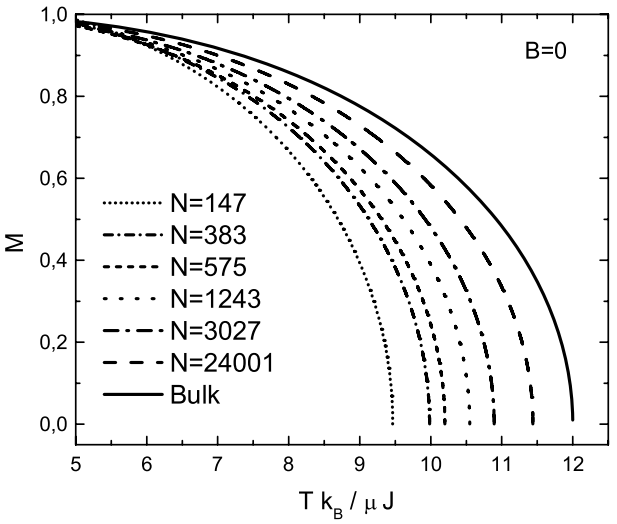


**Figure 2.** Thermal variation of the net magnetization for different sizes of hcp clusters without external field. The solid line corresponds to the magnetization of the bulk system, calculated by the equation (2.10). Clusters must be greater than $N = 3000$ for the equation (2.10) to have errors of less than 10% with respect to the equation (2.15). Similar behaviour is seen for fcc and bcc clusters.

ferromagnetic–paramagnetic transition is not homogeneous. For example, if the temperature is $Tk_B/J = 10$, the core is in the ferromagnetic phase, since $T_c^C > 10$, whereas the surface still is in the paramagnetic phase, i.e. $T_c^S < 10$.

In figure 2 the behaviour of the magnetization is shown for clusters of different sizes, with hcp structure, for zero external field. The solid line is the magnetization in the bulk limit, calculated by equation (2.10). As expected, the magnetization estimated by equation (2.15) tends to the magnetization calculated by equation (2.10) as the number of particles increases. However, even for clusters of 3000 particles there are differences of about 10% at temperatures near to the critical temperature.

For a given temperature the value of $N$ determines the cluster magnetic phase. For example, at $Tk_B/J = 10.5$ the clusters shown in figure 2 with more than 1243 particles are in the ferromagnetic phase whereas those that have fewer particles are in the paramagnetic phase.

This tendency of the magnetization with size is consistent with previous work using Monte Carlo simulations [16, 17] and using the large-scale spin–lattice dynamics (SLD) algorithm for iron thin films [28].

Figure 3 shows the shift of the Curie temperature with size. A decrement in the value of $N$ results in a transition to the paramagnetic phase at lower temperatures than in bulk. The exponents $\phi$ were obtained by drawing $(T_c^{bulk} - T_c^N)$ and $1/N$ on a log–log scale. For illustrative purpose we also show the tendencies for small clusters, i.e. 150 atoms, but these values were excluded from the scaling, which only included clusters with ∼300 or more atoms. We find that $\phi \simeq 0.32$ regardless of the crystal lattices characteristic.

Although the values of $\phi$ are concentrated around 0.32, the confidence intervals suggest that the value of the scaling exponent could be $1/3$. This would imply that the relationship decreases inversely proportional to the radius of the cluster. Let $R$ be the radius of the cluster and $r$ the atomic radius, $4/3\pi R^3 \propto 4/3\pi r^3 N$, then $(1/N)^{1/3} \propto 1/R$, so $(T_c^{bulk} - T_c^N) \sim 1/R$. The scaling laws for the surface and core regions are the same as that for the net magnetization.

Our model correctly predicts the tendencies observed in systems of relatively few particles. The dependence of the Curie temperature on size also has been observed experimentally and most of these studies confirm the declining of $T^C$ as the particle sizes decrease. This behaviour has been found in thin films [13, 28] and in nanoparticles, such as magnetite [11], FePt in fct phase [12] and Ni nanocrystals [14]. For the case of $MnFe_2O_4$ there are works which observed that $T_c$ increased with decreasing particle size [29]. However, $MnFe_2O_4$ is not a good system for studying finite size scaling effects. Chen *et al* [30] explained this increase as a result of different degrees of inversion of the spinel lattice due to $Mn^{2+}$ to $Mn^{3+}$ oxidation as a function of size.

Velásquez *et al* [23] also present a theoretical model for finite size effects in nanoparticles' properties. Their study was performed in the framework of an Ising model in the canonical formalism, using the variational approach based on the Bogoliubov inequality.

The particle diameters, $D$, that were studied in their work overlap with the ones used in our work. They obtained that the critical exponent associated to the correlation length is $\nu = 1.0001 \pm 0.0001$ for both bcc and fcc lattices. This value is consistent with our result, an exponent of $\phi \simeq 1/3$ with respect to the number of particles implies that the critical temperature decreases proportionally to the inverse of the cluster radius and therefore $\nu \simeq 1$. They used an effective coordination, $z_{eff}$, computed as the average coordination number in the cluster $z_{eff} = 1/N\sum_{i=1}^{N} n_i z_i$, where $N$ is the number of atoms. The idea is the same as we developed applying the equation (2.15) to each set of atoms with a specific coordination. In fact, if we calculate $z_{eff}$ for our clusters, we find that $z_{eff}$ is linear with the inverse of the radius, as in their work.

Unlike our model, the study from [23] can describe structures with atomic disorder and competing interactions. Competition is given, essentially, by the difference in sign and magnitude of the $J$ parameter. If we consider that $J(D) = J(\infty)$, equation (19) in [23] gives,

$$\frac{T_c(D)}{T_c(\infty)} \simeq \frac{z_{eff}}{z} \tag{3.1}$$

where $T_c(\infty)$ is the critical temperature in the bulk material. The value of $T_c(D)$ obtained from equation (3.1) is consistent with the values obtained from our model. For the studied radii, the difference is less than 2%.

The scaling exponent for the 3D Ising model is $\nu = 0.6289$ and for mean field is $\nu = 0.5$ [31], using the canonical ensemble. These values are lower than those obtained with our model and in [23], which are very similar to the exponent of a two-dimensional Ising model. The difference between our exponent and the expected value for mean field can be attributed to the fact that we solve the problem

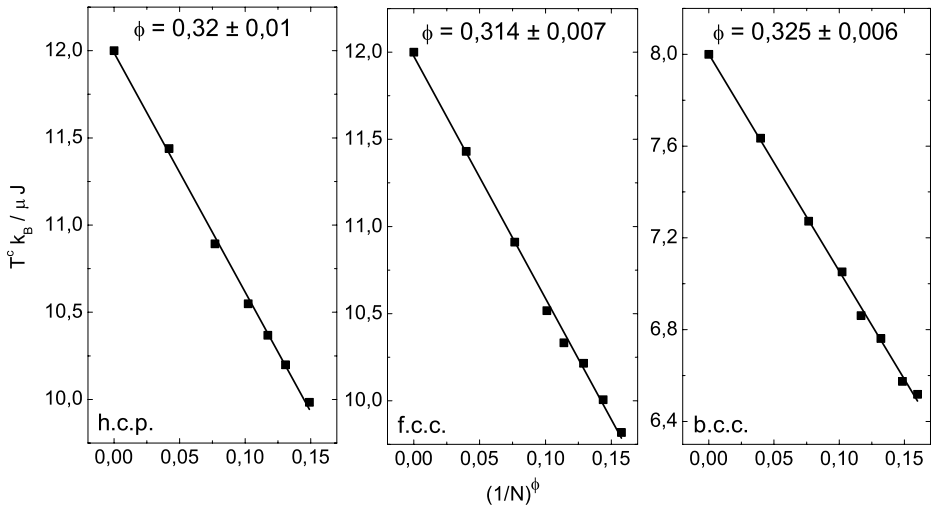


**Figure 3.** Size effect of the Curie temperature for different crystal lattices. The exponent $\phi$ shows how $T^{\mathrm{C}}$ varies with the number of particles $N$. The relation decreases in inverse proportion to the radius of the cluster.

in the microcanonical ensemble. The non-universality of the exponents for different ensembles has also been found in other cases, for example in the study of the critical behaviour of rigid rods. Almarza *et al* [32, 33] performed a Monte Carlo simulation in the grand canonical ensemble and found that the critical exponents of the polydisperse rods belong to the two-dimensional Ising universality class, while Lopez *et al* [34, 35] performed their calculations in the canonical ensemble and found that the exponents belong to the percolation universality class.

An example of a Monte Carlo study of finite size effects in nanoparticles is the work by Iglesias and Labarta [17]. They found that the critical exponent associated with the particle diameter for the maghemite structure is $\nu = 0.49 \pm 0.03$. However, the difference with our scaling exponent value is not surprising. The unit cell of maghemite is formed by two different sublattices with ions $Fe^{3+}$ and $O^{2-}$. Our model only applies to pure crystalline structures in situations where the spin–spin interaction $J$ does not vary for different couplings. In those cases, our model can only predict trends.

We performed simulations of larger clusters (up to ∼3 million atoms) and we compared the critical temperature obtained by equation (2.15) with that resulting from Monte Carlo simulations for bcc and fcc lattices, using periodic boundaries [36]. Both temperatures were normalized with the corresponding bulk temperatures and in the thermodynamic limit the two approaches tend towards the same value, as expected.

From the values of critical temperature and spontaneous magnetization it is possible to determine how the scaling exponent of the magnetization varies with the system size. The theory of critical phenomena predicts that the magnetization varies like a power law $M(T) \propto (T_{\mathrm{c}} - T)^{\beta}$ [31] in a second-order transition, where $\beta$ is the critical exponent. This exponent was determined by the slope on a log–log scale and for all the values of $N$, we obtained $\beta \simeq 0.5$, which is the standard value for the scaling exponent within the mean field approach.

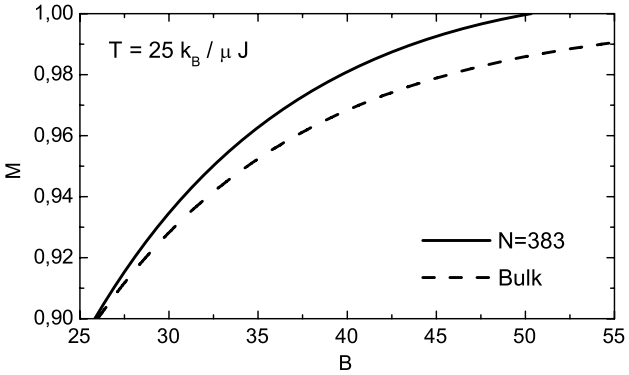


**Figure 4.** Net magnetization versus applied field. The dashed line corresponds to the bulk system while the solid curve to a hcp cluster with $N = 383$.

Finally, we solved the equation (2.15) for non-zero external fields. As shown in figure 4, the saturation of the field is more easily reached for small systems than for the bulk case. At $B = 0$ the spins on the surface are more disordered than the internal spins. However, at $B \neq 0$ the surface spins are more sensitive to the field and they are aligned more easily. This happens because surface bonds have a lower coordination number than bulk bonds, then the surface spins behave more independently while the core spins align towards the field direction in a more coherent way. Thus, the small clusters are easier to magnetize and to demagnetize than systems of many particles.

## 4. Summary and conclusions

We solved analytically the Ising model for small systems in the microcanonical formalism, using the mean field theory and considering the presence of surfaces. The magnetization was calculated from the number of accessible microstates, using the gamma function and its derivatives, unlike the usual

solution with the Stirling approximation which is inadequate for systems with relatively few particles.

Our model explains in simple terms several of the effects observed in experiments and Monte Carlo simulations of magnetic clusters. There are several experimental studies that confirm the declining of $T_c$ as the particle sizes decrease. We explain the observed shift of the critical temperature with nothing more than the change in the coordination number of the surface atoms. We found that $T_c$ follows a finite scaling with an exponent between 0.32 and 0.33, which indicates that $T_c$ decreases proportionally to the inverse of the radius of the cluster. This result is an agreement with the work by Velásquez *et al* [23].

There are Monte Carlo simulations that identified different contributions to the total magnetization, arising from the core and surface of nanograins. Our model predicts the behaviours of surface and core regions, and their relationship with the number of particles in each regions.

We obtain a new magnetization expression for small systems as a function of their size. At $B = 0$ we determined: (1) the reduction of $T_c$ with decreasing size; (2) the increase of the core magnetization with respect to the surface magnetization with increasing size; (3) the dependency of the net magnetization with size. On the other hand, at $B \neq 0$, we demonstrated that the net magnetization increases as the particle size decreases, indicating that small particles are easier to magnetize and demagnetize than bulk systems. Overall, our model provides simple qualitative predictions which facilitate the understanding of small-scale magnetization phenomena.

Preliminary works suggest that our result can be generalized to Heisenberg spins, but our model does not describe magnetic anisotropy or other situations where the spin–spin interactions vary.

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