Universal scaling behavior of interacting nanomagnets

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In this Letter we report how the influence of the interparticle dipolar interactions, computed by the volume sample concentration c, can be described in a general way if scaled by the dimensionless parameter $c_0 = 2K/M_S^2$. This scaling $c_0$-parameter is derived from the energy equation governing the system by using simple arguments, and its meaning is analogous to the normalization of the influence of the external magnetic field $H$ by the anisotropy field of the particles $H_A = 2K/M_S$. We use a Monte Carlo technique to test the usefulness of this $c_0$ parameter for interpreting the controversial evolution of the blocking temperature ($T_B$) vs. c reported in the literature, finding that the $T_B(c)$-curves of different particle types follow the same trend if scaling $c/c_0$.

Nanosized magnetic materials constitute the working body of several nanotechnological applications as the widely-used magnetic data storage, and the promising nanomagnetic logic, which rely on a very precise control of the magnetic entities and of their relative interaction. The problem is that for these closely-arranged systems the dominant interaction is the magnetic dipole-dipole coupling, which lacks an analytical solution and exhibits a complex interplay with the other more relevant energies in the system (anisotropy, Zeeman). Finding a precise and easy description of its features arises, therefore, as a crucial task to address in the current nanomaterialization race towards ever-denser concentrated designs.

Such a purpose has often been pursued in the basis of the Néel model. Dipolar interactions were investigated in relation with the anisotropy energy barrier of single-domain superparamagnetic (SPM) systems and its effect on the characteristic blocking temperature, $T_B$. The interparticle coupling strength was varied by changing the sample concentration of the system, c. The reasons to choose this method are its ease to carry out the experimental measurements and to correlate them with the theoretical background.

Despite the intense research devoted to this field in the recent years, no concluding results have been yet achieved, finding in the literature very different $T_B(c)$ trends for a priori very similar samples. These trends range from, i) a regular increase; ii) a rapid increase followed by a saturation; iii) a non-monotonic behaviour; iv) a decrease; etc. Although analytical models have been developed that describe reasonably well specific cases, a general model able to account for all the different shapes has not been reported so far (see Ref. [12]).

In an effort to shed light on this complex scenario, we present in this work an alternative approach: by simply rewriting the energies governing the system in an adequate way, the influence of the dipolar interactions in the magnetic behavior of the system (introduced as proportional to c) is described in a general way by the unitless parameter $c_0 = 2K/M_S^2$. In order to demonstrate the universality of this scaling parameter, we build up our arguments in the context of the usual single-domain particles SPM frame.

The SPM model represents the particles by their large magnetic supermoment and also by their magnetic anisotropy $\vec{K} = K\hat{n}$, with K the anisotropy constant and $\hat{n}$ the easy axis direction. The magnetic supermoment results from the coherent rotation of the inner atomic moments and is in first approximation proportional to the particle volume $V_i$, as $\vec{\mu}_i = M_S V_i \hat{\vec{\mu}}_i$, with $M_S$ the saturation magnetization and $\hat{\vec{\mu}}_i$ the unitary vector that describes its orientation.

The energy per particle $E^{(i)}$ for a non-interacting system is given by the anisotropy ($E_A$) and Zeeman ($E_Z$) energies as

$$E^{(i)} = E_A^{(i)} + E_Z^{(i)} = -K V_i \left( \frac{\vec{\mu}_i \cdot \hat{n}_i}{|\vec{\mu}_i|} \right)^2 - \vec{\mu}_i \cdot \vec{H}$$

Both energies determine the magnetic evolution of the system, being more important their relative weight than their absolute values. This is easily emphasized if rewriting Eq. (1) as

$$e^{(i)} = \frac{E^{(i)}}{2K V_i} = -\frac{1}{2} \cos^2 \varphi_i - \frac{H}{H_A} \cos \theta_i$$

where $\varphi_i$ and $\theta_i$ are the angles that $\vec{\mu}_i$ forms with respect to $\vec{H}$ and $\vec{K}$, respectively. Likewise, the anisotropy field is defined as $H_A = 2K/M_S$, which accounts for the relative importance between Zeeman and anisotropy energies. This is highlighted in the field dependence of $T_B \propto (1 - H/H_A)^{3/2}$ [14].
This expression illustrates that the influence of $H$ on the particle system does not depend on the absolute $H$-value but rather on the ratio $H/H_A$. Thus, particles with different values of $K$ and $M_S$ may still follow a similar $T_B(H)$ shape depending on their characteristic $H_A$ value, i.e. $T_B \equiv T_B(H/H_A)$.

We follow the same line of reasoning to investigate the influence of the dipolar interaction. In this case, for an $N$-particle system the energy per particle is

$$E^{(i)} = -KV_i \left( \frac{\hat{\mu}_i \cdot \hat{n}_i}{|\hat{\mu}_i|} \right)^2 - \hat{\mu}_i \cdot \vec{H} +$$

$$+ \sum_{j \neq i}^N \left( \frac{\hat{\mu}_i \cdot \hat{\mu}_j}{r_{ij}^3} - 3 \left( \frac{\hat{\mu}_i \cdot \hat{r}_{ij}}{r_{ij}^3} \right) \left( \frac{\hat{\mu}_j \cdot \hat{r}_{ij}}{r_{ij}^3} \right) \right)$$  \hspace{1cm} (3)

where $\hat{r}_{ij}$ is the vector connecting particles $i$ and $j$, and the summation extends to the neighboring particles. Our procedure is to rewrite Eq. (3) also in terms of $H/H_A$, trying to find also an easy dependence for the dipolar energy term. For such a purpose, we assume i) a monodisperse system; ii) the particles are located into a cubic box of side $L$, so that we may use normalized units $\bar{a}_{ij} = \hat{r}_{ij} N^{\frac{3}{2}}/L$; iii) the dimensionless sample concentration $c$ results in $c = \sum_{i=1}^N V_i / L^3 \equiv NV/L^3$. Thus, the reduced energy $e^{(i)}$ reads now

$$e^{(i)} = \frac{E^{(i)}}{2KV} = -\frac{1}{2} \cos^2 \varphi_i - \frac{H}{H_A} \cos \theta_i +$$

$$+ \frac{c}{c_0} \sum_{j \neq i}^N \left( \frac{\bar{e}_{\mu_i} \cdot \bar{e}_{\mu_j}}{a_{ij}^3} - 3 \left( \frac{\bar{e}_{\mu_i} \cdot \bar{a}_{ij}}{a_{ij}^3} \right) \left( \frac{\bar{e}_{\mu_j} \cdot \bar{a}_{ij}}{a_{ij}^3} \right) \right)$$  \hspace{1cm} (4)

where we have introduced $c_0 = 2K/M_S^2$. This $c_0$-parameter weights the importance of the dipolar interaction energy as $c/c_0$, analogously as $H/H_A$ weights the importance of the Zeeman energy. Therefore, we can conclude that same that $H/H_A$ provides a general behaviour for the magnetic field dependence of the system, it is possible to define also a general behaviour for the influence of the dipolar interaction, given by $c/c_0$. This result demonstrates the existence of an inner scale of energies in the thermomagnetic properties of those nanostructured systems as a function of the $H_A$ and $c_0$ parameters.

The physical meaning of $c_0$ can be interpreted as an estimation of the relative importance between the anisotropy and dipolar energies, i.e. $c_0 = 2K/M_S^2 \propto E_A/E_D$. It is worth to note that the $2K/M_S^2$ ratio had been noticed in the literature as a relevant magnitude concerning interacting SPM systems. However, neither its meaning as a general scaling factor nor its key-role to solve controversial results in the literature (as the $T_B$ vs. $c$ evolution) had been reported up to now.

The next step is to demonstrate that the above assertion on $c_0$ as a scaling factor for the influence of magnetic dipolar interactions can effectively solve controversial results in the literature. With this purpose in mind, we decided to tackle the $T_B$ vs. $c$ problem based on its central role amongst theoretical-nanoparticle problems as to dipolar interaction concerns.

Our procedure consists on comparing the $T_B(c)$ data for different types of particles, in order to study the different trends reported in the literature. We represent the particle types by their corresponding $c_0$-values, and examine their magnetic behaviour both as a function of $c$ and $c/c_0$. Since experimental data is usually subjected to uncontrolled secondary effects (polidispersity [16], aggregation [17]) that might mask the influence of the dipolar interactions, we have used a Monte Carlo technique that allowed us to perfectly control the characteristics of the sample and hence to be sure about the nature of the different $T_B$ data. The computational procedure is the same as described in Ref. [12], and the results are presented in the usual reduced temperature units $t = k_B T/2K$. $T_B$ is roughly evaluated as the maximum of the zero field cooling (ZFC) curves under a low applied field $H = 100$ Oe, as usual, and hence to obtain the $T_B$ vs. $c$ data we simulated ZFC processes for a systematic variation of $\Delta c = 2.5\%$.

To represent different types of particles we characterized them by their $K$ and $M_S$ values, and associated each type to the corresponding $c_0$. To select the $K$ and $M_S$ values (and hence the $c_0$ cases) we decided to keep a common value of $K$ and vary $M_S$. By performing this way we can expect that in the non-interacting limit case $T_B$ will be the same for the different particle types, since $T_B \approx KV/2k_B$ (i.e. independent of $M_S$), so that we can study the evolution of $T_B$ with $c$ from this common non-interacting point. It is worth to note that although this is an ideal assumption, it is in fact possible to design different materials so that they have a common (or very similar) $K$ value but different $M_S$. Also, for computational purposes it is easier to keep the same value of $K$ (included in the reduced temperature units) and vary only $M_S$. The $K$ value is relatively low so that dipolar interactions influence easily the system, using $K = 1.5 \times 10^5 \text{erg/cm}^3$. The $M_S$ values considered and the corresponding $H_A$ and $c_0$ parameters are shown in Table I together with $H/H_A$ ratio corresponding to $H = 100$ Oe.

The $M_S$ values were chosen so that they represent realistic physical values, and the equivalent $c_0$ cases cover a wide range of $E_A$ vs. $E_D$, so that $c_0$ varies between the $E_A$-dominating case ($c_0 = 4$),
ZFC curves for specific field and concentration conditions and well intermediate cases. In Fig. 1, we show some ZFC curves for the non-interacting case under the same absolute field $H$ and the maximum of the curves; however, if using the same reduced field $H/H_A = 0.18$, the ZFC curves separate if using the same absolute $c$ and overlap if using the same normalized $c/c_0$. This result demonstrates the existence of an inner scale of energies as a function of $H_A$ and $c_0$.

We focus now on the $T_B$ ($t_B$ in reduced units) vs. $c$ problem. Following the same procedure as in Fig. 1 we simulate ZFC processes for the different cases of Table I and systematically vary $c$, evaluating $t_B$ as the maxima of the curves. In Fig. 3(a), we observe that the $t_B(c)$ data follows different trends under the same absolute values $H = 100$ Oe and $c = 0.10$, but that share however a common origin in the non-interacting $c = 0.0$ case if using the same reduced field $H/H_A = 0.18$ (Fig. 3(b)). It is worth to emphasize the different trends observed in the $t_B$ vs. $c$ data, which reproduce a rich variety of tendencies as mentioned in the introduction. A complete characterization of all different types would need however a more detailed analysis of the specific shapes of the curves.

In Fig. 3(c) it is shown that the apparently different trends of Fig. 3(a) share however a more similar tendency if plotted vs. $c/c_0$, even for the
same absolute field \( H = 100 \text{ Oe} \). Furthermore, all of them pretty much overlap if using both the same \( H/H_A = 0.18 \) value and plotted \( c/c_0 \), as shown in Fig. 3(d). These results suggest (within the precision of the results) the existence of a general \( t_B(H/H_A; c/c_0) \) curve, with the different trends of Figs. 3(a),(b) being only different fractions of it.

It might be argued, however, that the precision of the \( t_B(c) \) data is not enough to demonstrate the scaling of the SPM phenomena, since there is some deviation between the \( t_B(c/c_0) \) curves. To erase such possible doubts we have followed a different approach: we analyzed the maxima of the curves at \( t_B \), \( m(t_B) \), which is far more precise to evaluate than \( t_B \). The results displayed in Fig. 4 demonstrate with high precision the role of the \( c_0 \) parameter as a scaling factor for the magnetic dipolar interaction: the \( m(t_B) \) data, which follows different trends if using absolute \( H = 100 \text{ Oe} \) values, as shown in Figs. 4(a),(b); or absolute \( c = 0.1 \) values, as in Figs. 4(a),(c); however perfectly overlaps in a common \( m(t_B) \) curve if using the same relative \( H/H_A = 0.18 \) value and plotted \( c/c_0 \), as shown in Fig. 4(d).

In summary, we report the existence of a general scaling factor for the influence of the dipolar interaction energy in single-domain entities. It is defined by the dimensionless parameter \( c_0 = 2K/M_S^2 \), and together with the anisotropy field \( H_A \) serves to define a general behavior for such systems as governed by the dipolar and Zeeman energies. Specifically, we applied this parameter to study the controversial results on the evolution of \( T_B \) vs. \( c \) in assemblies of SPM nanoparticles, finding that the apparently dis-similar tendencies reported in the literature, belong in fact to the same general \( T_B(c/c_0) \) curve. It remains as the subject of a future work the detailed analysis of the shape of such \( T_B(c/c_0) \) curve.

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[15] See e.g., Phys. Rev. Lett. 84, 167 (2000); J. Magn. Magn. Mater. 226, 1936 (2001); Adv. Chem. Phys. 128, 191 (2004).
[16] A. Wang, J. Li, and R. Gao, Appl. Phys. Lett. 94, 212501 (2009).
[17] C. E. Hoppe et al., J. Phys. Chem. C 112, 13099 (2008).