Spontaneous magnetization in presence of nanoscale inhomogeneities in diluted magnetic systems

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Abstract

The presence of nanoscale inhomogeneities has been experimentally evidenced in several diluted magnetic systems, which in turn often leads to interesting physical phenomena. However, a proper theoretical understanding of the underlying physics is lacking in most of the cases. Here we present a detailed and comprehensive theoretical study of the effects of nanoscale inhomogeneities on the temperature dependent spontaneous magnetization in diluted magnetic systems, which is found to exhibit an unusual and unconventional behavior. The effects of impurity clustering on the magnetization response have hardly been studied until now. We show that nano-sized clusters of magnetic impurities can lead to drastic effects on the magnetization compared to that of homogeneously diluted compounds. The anomalous nature of the magnetization curves strongly depends on the relative concentration of the inhomogeneities as well as the effective range of the exchange interactions. In addition we also provide a systematic discussion of the nature of the distributions of the local magnetizations.

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Interest in diluted magnetic systems, such as diluted magnetic semiconductors (DMSs)\textsuperscript{1-3} and diluted magnetic oxides (DMOs)\textsuperscript{4,5}, has continued to surge over the past couple of decades owing to their huge potential for spintronics applications. The prime requisite of room-temperature ferromagnetism for feasible spintronics devices has led to Curie temperature ($T_C$) being the primary focus of interest in these diluted magnetic materials. Contrary to a longstanding belief of defect and inhomogeneity free samples leading to high Curie temperatures, recent studies suggest otherwise. Experimental studies have revealed the formation and existence of magnetic clusters on the nanoscale order, in materials like (Ge,Mn)\textsuperscript{6} and (Zn,Co)O\textsuperscript{7}, which in turn gave rise to very high $T_C$’s ($\geq 300$ K). In fact, very recently we have theoretically shown that incorporating magnetic nanoclusters, can lead to drastically high critical temperatures (often above room-temperature) in diluted magnetic systems with effective short-ranged exchange interactions\textsuperscript{8}. Now, another important aspect which can help to further reveal the physical intricacies of these strongly disordered and complex systems is the temperature dependence of the spontaneous magnetization $M(T)$. Among the many interesting features that magnetization possesses, some worth mentioning are the convex or concave nature of the $M(T)$ curves and their critical behavior close to the transition point.

In the particular case of DMSs, one of the very first observations of ferromagnetism in (In,Mn)As\textsuperscript{9}, revealed a surprising non-mean-field like behavior of the spontaneous magnetization with temperature. The experimentally determined magnetization curve had an unusual outward concave-like shape which is in stark contrast to the typical convex behavior obtained within the standard Weiss mean-field theory\textsuperscript{10}, as well as that observed in conventional ferromagnetic materials. The (In,Mn)As samples studied in Ref.\textsuperscript{9} were reported to be insulating. Similar concave $M(T)$ behavior was also observed in insulating samples of Ge$_{1-x}$Mn$_x$ determined by superconducting quantum interference device (SQUID) magnetometry and magnetotransport measurements\textsuperscript{11}. Theoretical predictions, based on a percolation transition of bound magnetic polarons in the strongly localized regime\textsuperscript{12}, were made to explain this non-mean-field like magnetization behavior. Similar magnetization behavior in DMS systems, in the insulating regime, was also predicted by other theoretical studies based on numerical calculations\textsuperscript{13-15}. The deviation in the $M(T)$ behavior from the standard Brillouin-function shape was believed to be partly due to the small carrier density compared to the localized spin density, as well as due to the wide distribution of the exchange interactions and hopping integrals\textsuperscript{15}. On the other hand, in metallic DMSs, for
example \( \text{Ga}_{1-x}\text{Mn}_x\text{As} \) for \( x=0.05-0.10 \), the magnetization behavior is found to decrease almost linearly with temperature\(^{16,17} \). This behavior is somewhat intermediate between the concave-like \( M(T) \) curves in the insulating regime and the classic convex magnetization. However, experimental studies suggest that annealing treatments can have a strong effect on the nature of the magnetization behavior\(^{18,19} \). During annealing the magnetization curves are found to become more convex and Brillouin-function-like compared to their linear behavior before annealing. This anomalous behavior of the magnetization highlights the importance of disorder in these systems.

However, the effects of correlations in disorder or impurity clustering have hardly been considered barring a few cases. In Ref\(^{20} \) the authors reported that correlated defects lead to “mean-field-like” magnetization curves in \( \text{(Ga,Mn)As} \). In another theoretical study\(^{21} \) on Co doped ZnO, inhomogeneous phases were shown to be responsible for high Curie temperatures in comparison to the homogeneous samples. Now a majority of the existing theoretical studies are based on mean-field-like approaches, which is known to be inadequate to treat thermal and/or transverse fluctuations and disorder reliably in these systems. In Ref\(^{22} \) the authors have used some complementary theoretical approaches in addition to the mean-field theory, which include the dynamical mean-field theory (DMFT), to study the temperature dependent magnetization in doped magnetic semiconductors. Nonetheless, the effects of clustered defects were not taken into account. The presence of nanoscale inhomogeneities can give rise to very interesting and new physics in these diluted systems, as was seen in the case of the Curie temperatures\(^{8} \).

The primary objective of the current manuscript is to investigate the effects of these inhomogeneities on the spontaneous magnetization behavior. Here we first study the nature of the magnetization in homogeneously diluted systems with no correlation in impurity positions, and then extend this to systems containing clusters of magnetic impurities. We observe very interesting as well as strong deviations from the homogeneous magnetization curves. A non-trivial nature of the spontaneous magnetization in these inhomogeneous diluted systems is found to strongly depend on the relative concentration of the inhomogeneities as well as the effective range of the magnetic exchange interactions.

For the sake of simplicity we have assumed here a simple cubic lattice with periodic boundary conditions. We fix the total concentration of impurities in the system to \( x=0.07 \) and the total number of impurities in the system is denoted by \( N_{\text{imp}} \). The inhomogeneities
are assumed to be of spherical shape of radii $r_0$. The concentration of impurities inside each nanosphere is denoted by $x_{in}$. To avoid additional parameters, we restrict ourselves to nanospheres of fixed radii $r_0=2a$ ($a$ is the lattice spacing) and $x_{in}=0.8$. The concentration of nanospheres in the system is defined by $x_{ns}=N_S/N$, where $N_S$ is the total number of sites included in all the nanospheres and $N=L^3$ is the total number of sites. We define a variable $P_N=(x_{in}/x)x_{ns}$, which denotes the total fraction of impurities contained within the nanospheres. For a particular configuration, the nanospheres are distributed in a random fashion on the lattice, the only restriction being to avoid any overlap with each other.

In order to calculate the spontaneous magnetization, we start with the effective diluted Heisenberg Hamiltonian describing $N_{imp}$ interacting spins ($S_i$) randomly distributed on a lattice of $N$ sites, given by

$$H = -\sum_{i,j} J_{ij} p_i p_j S_i \cdot S_j$$

where the sum $i,j$ runs over all sites and the random variable $p_i=1$ if the site is occupied by an impurity or otherwise 0. We assume magnetic couplings of the form $J_{ij}=J_0\exp(-|\mathbf{r}|/\lambda)$, where $\mathbf{r} = \mathbf{r}_i - \mathbf{r}_j$. Both, the choice for such couplings and the values of the damping parameter $\lambda$ are discussed below.

This Hamiltonian (Eq.1) is treated within the self-consistent local random phase approximation (SC-LRPA) theory, which is essentially a semi-analytical approach based on finite temperature Green’s functions. More details on the SC-LRPA theory can be found in

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FIG. 1: (Color online) Illustration of the self-consistent local random phase approximation method at a given temperature $T$. 
Ref.3,23 A schematic illustration of the method is given in Fig. 1. We briefly summarize the method in the following. We start the self-consistent loop with a fully polarized, collinear ferromagnetic ground state at \( T=0 \) K. The impurity spins are assumed to be classical in this case, although the theory is valid for quantum spins as well. Within the SC-LRPA formalism, for a given disorder configuration, the local magnetizations at each site \( \langle S^z_i \rangle \) \((i=1,2,\ldots,N_{\text{imp}})\) are calculated self-consistently at each temperature. The local magnetization is evaluated using a Callen-like expression\(^{24}\), which relates the local Green’s function at site \( i \) to the local magnetization at this site,

\[
\langle S^z_i \rangle = \frac{(S - \Phi_i)(1 + \Phi_i)^{2S+1} + (1 + S + \Phi_i)\Phi_i^{2S+1}}{(1 + \Phi_i)^{2S+1} - \Phi_i^{2S+1}}
\]  

(2)

where the local effective magnon occupation number is given by

\[
\Phi_i = -\frac{1}{2\pi} \left\langle S^z_i \right\rangle \int_{-\infty}^{+\infty} \frac{\Im G_{ii}(\omega)}{\exp(\omega/k_B T) - 1} d\omega
\]  

(3)

We define the retarded Green’s function as \( G_{ij}(\omega) = \int_{-\infty}^{\infty} G_{ij}(t)e^{i\omega t} dt \), where \( G_{ij}(t) = i\theta(t)\langle [S^+_i(t),S^-_j(0)] \rangle \), which describes the transverse spin fluctuations, and \( \langle \ldots \rangle \) denotes the expectation value. By the self-consistent treatment we obtain, for a given temperature and disorder configuration, the average magnetization which is defined by

\[
\langle S^z_{\text{avg}} \rangle = \frac{1}{N_{\text{imp}}} \sum_{i=1}^{N_{\text{imp}}} \langle S^z_i \rangle
\]

The accuracy and reliability of the SC-LRPA to treat thermal/transverse fluctuations and disorder/dilution have been shown on several occasions.\(^{3,25}\) Moreover it has also been applied to calculate the Curie temperatures in inhomogeneous diluted systems in a recent study.\(^{8}\)

The assumption of the magnetic interactions of the form \( J_{ij} = J_0 \exp(-|\mathbf{r}_i - \mathbf{r}_j|/\lambda) \) is based on the fact that both \textit{ab initio} studies\(^{26,27}\) as well as model calculations\(^{28}\) have shown that the exchange couplings in III-V DMSs are relatively short ranged and non-oscillating in nature. This non-oscillating behavior results from the existence of a preformed impurity band (IB)\(^{29}\). The existence/absence of an IB has been a longstanding debate for several years. But in our view, the still often quoted RKKY interactions in these compounds, only consistent with the valence band (VB) or perturbative picture, can be ruled out. As shown in Ref.\(^{25}\) only the IB scenario could explain the proximity of (Ga,Mn)As to the metal-insulator transition as well as the observed red shift in the optical conductivity in this compound. Furthermore, recent experimental findings\(^{30}\) definitely corroborate the fact that the Fermi level is located within an IB in (Ga,Mn)As. Now, in (Ga,Mn)As, for about 5% Mn, a fit
of the *ab initio* exchange couplings provides a value of $\lambda$ of the order of $a/2$. It should be noted that in the case of (Ga,Mn)N the exchange interactions are even shorter ranged. In the following we will consider a range of $\lambda$’s, corresponding to relatively long-ranged couplings down to shorter ranged ones, and try to analyze their effects on the magnetization behavior. In fact the magnetic couplings could also depend on the nanoscale inhomogeneities. However, computing these would require extensive calculations involving finite size analyses, systematic average over disorder configurations and especially diagonalizing considerably large matrices. This is beyond the scope of the present manuscript. Nevertheless, we believe that the overall nature of the exchange couplings, especially in wide gap compounds like (Ga,Mn)N, will remain essentially unchanged. The interesting feature which we want to focus on in the current work is the role the inhomogeneities play in determining the nature of the magnetization curves. In the following, the average magnetization is always plotted as a function of the reduced temperature $T/T^*$, where $T^*$ is the temperature corresponding to the case when $\langle S^z_{\text{avg}} \rangle = 0.001S$. We have chosen $T^*$ instead of $T_C$ as it is difficult to determine accurately the critical temperatures from the magnetization curves. However, we have checked for some cases, that $T^*$ is relatively close to the $T_C$ directly calculated from the semi-analytical expression (Eq.(1) in Ref.[8]).

To begin with we consider the homogeneously diluted case where the magnetic impurities are randomly distributed on the lattice. Fig. 2 shows the average magnetization as a function of the reduced temperature $T/T^*$, for different values of $\lambda$. The magnetization shown corresponds to a single configuration of disorder and the system size is $L=24$. We observe that for relatively long-ranged couplings ($\lambda=2a$), the magnetization curve has a pronounced convex shape which is the usual behavior predicted by the mean-field theories of Weiss and Stoner[10], and observed commonly in conventional ferromagnetic materials. On decreasing $\lambda$, we notice that the convexity decreases and for $\lambda=a/3$ the magnetization is more linear-like over a broad temperature range. In fact similar behavior (linearity) of the magnetization was observed in metallic samples of (Ga,Mn)As by magnetic circular dichroism (MCD) studies[16]. This shows that the relatively short ranged interactions are more relevant for the case of DMS materials and also vindicates the choice of our exchange couplings. In order to have a qualitative idea of the relative change in the behavior of the magnetization with $\lambda$, we have plotted in the inset of Fig. 2 the curvature ($\kappa$) of the magnetization curves at the spe-
FIG. 2: (Color online) Average magnetization as a function of $T/T^*$ for different values of $\lambda$, corresponding to the homogeneous case for a single configuration. Inset: Curvature of the $\langle S_z^{\text{avg}} \rangle$ curves as a function of $\lambda$, calculated at $T/T^*=0.5$. The blue dashed line indicates the saturation value of the curvature. (Here $N=24^3$).

cific value of $T/T^*=0.5$. The curvature is defined by $\kappa = \left| \frac{\partial^2 \langle S_z^{\text{avg}} \rangle / S}{\partial u^2} \right| \left( 1 + \left[ \frac{\partial \langle S_z^{\text{avg}} \rangle / S}{\partial u} \right]^2 \right)^{-3/2}$, where $u = \frac{T}{T^*}$. As can be clearly seen, with increase in $\lambda$ the curvature changes significantly, increasing by almost a factor of five from $\lambda=a/3$ to $\lambda=2a$. For $\lambda \geq 2a$, one sees that $\kappa$ has already saturated and the magnetization has a standard Brillouin shape. This implies that $\lambda \geq 2a$ corresponds to the long range coupling regime.

In Fig. 2, we have shown the magnetization for a single configuration of disorder. However, we know that in diluted materials the magnetic properties are often very sensitive to the random impurity configurations. Fig. 3 shows the average magnetization calculated for 25 configurations of disorder corresponding to three different values of $\lambda$. We have also calculated $\Delta S / \langle S_z^{\text{avg}} \rangle$, which gives a measure of the fluctuations of the extremal magnetization curves from the configuration averaged one ($\langle S_z^{\text{avg}} \rangle$), at the particular value of $T/T^*=0.6$. For $\lambda=a$ (Fig. 3(a)), we observe that the spontaneous magnetization is weakly sensitive to
the disorder configurations, the overall shape of the curves is unchanged. $\Delta S_{z}^{\langle S_{z} \rangle}$ is found to vary within 10% of the configuration averaged magnetization. For $\lambda=2a$ (not shown here), these fluctuations were found to be even smaller, varying within less than 5%. For the short ranged couplings, $\lambda=a/2$, the magnetization curves are still convex but the fluctuations are stronger now. $\Delta S_{z}^{\langle S_{z} \rangle}$ is more than doubled as compared to the intermediate range of $\lambda=a$.

As can be seen from Fig. 3(b), some of the curves have a regular convex behavior while some are more linear in nature. Now on further reducing $\lambda$ (Fig. 3(c)), the deviations become even stronger, and a significant number of configurations exhibit a clear linear temperature dependence. The fluctuation at $T/T^*=0.6$ increases by almost a factor four, compared to the case of $\lambda=a$. Even, in some cases, the magnetization profiles are slightly concave toward the high temperature. It should be noted that the more linear or concave magnetization curves correspond to relatively high $T_C$’s. This figure clearly shows that the disorder effects are significantly enhanced in the case of short-ranged interactions. The primary reason is that the probability to find regions of weakly interacting impurities increases significantly for the case of short-ranged interactions. This is the case in most of the DMSs, where a non-trivial behavior of the magnetization is observed.

So far we have only considered homogeneously diluted systems assuming a fully random distribution of the impurities. Now we move to the case of nanoclusters of magnetic impurities. Fig. 4 shows for $\lambda=a$, the average magnetization of the whole system, $\langle S_{z}^{\text{tot}} \rangle$, as a function of $T/T^*$, for four different concentrations of nanospheres, $x_{ns}=1\%, 3\%, 5\%$,
FIG. 4: (Color online) $\langle S^c_z \rangle$ stands for the total average magnetization ($\langle S^\text{tot}_z \rangle$), magnetization inside the nanospheres ($\langle S^\text{in}_z \rangle$) and outside the nanospheres ($\langle S^\text{out}_z \rangle$). Four different concentrations of nanospheres ($x_{\text{ns}}$): (a) 0.01, (b) 0.03, (c) 0.05, and (d) 0.07, are considered. $P_N$ is the percentage of total impurities contained in the nanospheres. Here $\lambda=a$, $r_0=2a$, $x_{\text{in}}=0.8$ and $N=24^3$. The x-axis is in units of $T/T^*$. and 7%. In addition, we have also shown the average magnetization inside and outside the clusters denoted by $\langle S^\text{in}_z \rangle$ and $\langle S^\text{out}_z \rangle$, respectively. The curves shown here correspond to a single configuration of disorder, the variation with disorder configurations will be discussed in what follows. We immediately observe that in the presence of inhomogeneities the spontaneous magnetization has a non-trivial behavior and exhibits a drastically different nature when compared to the homogeneous case (Fig. 2). This can be clearly seen even for the lowest concentration of nanospheres. For $x_{\text{ns}}=0.01$, for which 11% of the total impurities are inside the nanospheres, $\langle S^\text{tot}_z \rangle$ decreases rapidly till about $T/T^* \sim 0.5$, then it becomes concave and decays slowly toward the higher temperatures. By gradually increasing the concentration of the nanospheres, an interesting change in the average magnetization behavior
is observed. For $x_{ns} = 3\% \ (P_N = 0.34)$, $\langle S^\text{tot}_z \rangle$ falls off less sharply at low temperature, for 5\% it is almost linear over the entire temperature range, and for 7\% it becomes more convex. Thus a crossover in the curvature of $\langle S^\text{tot}_z \rangle$ appears at $x_{ns} \approx 0.05$. On the other hand, $\langle S^\text{in}_z \rangle$ exhibits a clear convex nature which does not change with $x_{ns}$. This indicates that the average magnetization inside the clusters remains almost uniform and is mainly controlled by the intra-cluster couplings. We can clearly see that the inhomogeneities have a very strong effect on the impurities outside the clusters. $\langle S^\text{out}_z \rangle$ has a very pronounced concave nature which can even be seen for relatively small $x_{ns}$. The slope at low temperatures becomes steeper with increasing concentration of nanospheres. For example, at $T / T^* \sim 0.3$, $\langle S^\text{out}_z \rangle$ has a value of 0.85 for $x_{ns} = 0.01$, 0.62 for $x_{ns} = 0.03$, and about 0.4 for $x_{ns} = 0.05$. Similar concave behavior of the temperature dependent magnetization is observed in the case of some insulating DMS materials. However, in most of the cases studied until now clustering effects have hardly been considered. Note that we have also performed the calculations for $\lambda = 2a$ (not shown here). In this extended coupling regime, it was found that the effect of inhomogeneities are very weak. $\langle S^\text{tot}_z \rangle$, $\langle S^\text{in}_z \rangle$, and $\langle S^\text{out}_z \rangle$ exhibit a convex nature and were found to be relatively close to each other.

In an earlier study, we have shown that relatively short-ranged couplings appear to have spectacular effects on the Curie temperatures in the presence of nanoscale inhomogeneities. Short-ranged interactions are more relevant from the practical point of view. As mentioned above, the magnetic couplings in most III-V DMS materials ((Ga,Mn)As, (Ga,Mn)N, (Ga,Mn)P,...) are effectively short-ranged in nature. In Fig. 5 we show $\langle S^\text{tot}_z \rangle$, $\langle S^\text{in}_z \rangle$, and $\langle S^\text{out}_z \rangle$ as a function of $T / T^*$, for four different concentrations of nanospheres, in the case of relatively short-ranged couplings, namely $\lambda = a/2$. To start with, we first discuss the results for a single configuration of disorder. For the lowest $x_{ns}$ (Fig. 5(a)), the behavior of $\langle S^\text{tot}_z \rangle$ is almost similar to that observed in the case of $\lambda = a$ (Fig. 4(a)). But on increasing $x_{ns}$ further (Fig. 5(b)), we immediately observe that $\langle S^\text{tot}_z \rangle$ decreases much more rapidly at lower temperatures followed by a slow decay toward the high temperatures. Also, in Fig. 5(c) and (d), an inflection appears in $\langle S^\text{tot}_z \rangle$ around $T / T^* \sim 0.6$. In this case $\langle S^\text{in}_z \rangle$ too exhibits a non-trivial behavior for all values of $x_{ns}$, which is unlike the case of $\lambda = a$. For example, for $x_{ns} = 0.05$ (Fig. 5(c)), there is a shoulder-like feature in $\langle S^\text{in}_z \rangle$ around $T / T^* \sim 0.05$, which is absent for $\lambda = a$ (Fig. 4(c)). Thus unlike the case of $\lambda = a$, where the intra-cluster couplings dominate, there are other relevant couplings, like the inter-cluster ones and those between...
FIG. 5: (Color online) $\langle S_z^{\text{tot}} \rangle$ stands for the total average magnetization ($\langle S_z^{\text{tot}} \rangle$), magnetization inside the nanospheres ($\langle S_z^{\text{in}} \rangle$) and outside the nanospheres ($\langle S_z^{\text{out}} \rangle$). Four different concentrations of nanospheres ($x_{ns}$): (a) 0.01, (b) 0.03, (c) 0.05, and (d) 0.07, are considered. $P_N$ is the percentage of total impurities contained in the nanospheres. Here $\lambda=a/2$, $r_0=2a$, $x_{in}=0.8$ and $N=24^3$. The x-axis is in units of $T/T^*$.

the cluster impurities and bulk impurities, which come into play. On the other hand, the $\langle S_z^{\text{out}} \rangle$ curves are typically concave for all considered $x_{ns}$, and exhibit a long tail toward the higher temperatures. They exhibit a sharp fall-off at low temperatures with increasing $x_{ns}$. At $T/T^* \sim 0.2$, for $x_{ns}=0.01$, the value of $\langle S_z^{\text{out}} \rangle$ is about 0.8 which falls rapidly to almost 0.3 for $x_{ns}=0.05$. With increasing $x_{ns}$, the concentration of impurities outside the clusters gradually decreases, leading to an increase of the typical distance between them. Consequently the impurities outside interact more weakly with each other and this explains the sharp fall-off in $\langle S_z^{\text{out}} \rangle$ at lower temperatures.

In the previous two figures we have discussed the cases for a single configuration of disorder only. Let us now analyze how the results depend on the random cluster configurations.
Fig. 6 shows the average magnetization calculated for 50 configurations of disorder, for $x_{ns} = 0.05$. We compare the case of the intermediate couplings to the short-ranged ones. One sees for the case of $\lambda = a$ (Fig. 6(a)), that the magnetization curves have an almost linear similar shape over the entire temperature range. In fact the configuration averaged magnetization can be well approximated by $\langle S_{z}^{\text{tot}} \rangle \approx (1 - T/T^*)$. On the other hand, the configuration averaged magnetization has a pronounced concave nature for $\lambda = a/2$ (Fig. 6(b)). The decay slope at low temperatures ($T/T^* \leq 0.2$) is twice that of $\lambda = a$. Concerning the disorder fluctuations, one clearly sees that they are much stronger in the case of the short-ranged couplings. For example, the fluctuation of $\langle S_{z}^{\text{tot}} \rangle$ at $T/T^* = 0.6$ is found to be more than doubled compared to that of $\lambda = a$. For $\lambda = a/2$, most of the magnetization curves are concave in nature, while some exhibit a linear-like behavior. The typical separations between the clusters play a decisive role, in the presence of short-ranged couplings. This has been discussed in the context of Curie temperatures in inhomogeneous systems\(^8\). Here it should be noted that the concave-like curves correspond to higher critical temperatures, while the linear ones coincide with relatively low $T_C$'s. Finally, we have found that for relatively extended couplings ($\lambda \approx 2a$) (not shown here), $\langle S_{z}^{\text{tot}} \rangle$ exhibits a more convex-like behavior. The fluctuations resulting from the disorder configurations were found to be much smaller than
FIG. 7: (Color online) Configuration averaged magnetization $\langle S_{z}^{\text{tot}} \rangle$ for different system sizes $N=L^3$, corresponding to (a) $\lambda=a$, and (c) $\lambda=a/2$. The total number of configurations is of the order of few hundred. The respective fluctuations $\frac{\Delta S}{\langle S_{z}^{\text{tot}} \rangle}$ at $T/T^*=0.6$, as a function of $1/N$ is shown in (b) for $\lambda=a$, and (d) for $\lambda=a/2$. (Here $x_{ns}=0.05$, $r_0=2a$, and $x_{in}=0.8$).

We now proceed further with the study of the finite size effects. Indeed, in inhomogeneous systems one naturally expects finite size effects to be much stronger than in homogeneously diluted ones. These effects can be even more drastic for larger size of inhomogeneities. The configuration averaged total magnetizations as a function of $T/T^*$, corresponding to $\lambda=a$ and $\lambda=a/2$, are shown respectively in Fig. 7(a) and (c). The calculations are performed over different system sizes varying from $N=12^3$ up to $N=28^3$. $\langle S_{z}^{\text{tot}} \rangle$ is the averaged magnetization obtained over a sample of few hundred disorder configurations. In both cases we observe that the magnetization curves are very similar and the finite size effects are very weak. Let us now discuss the size dependence of the fluctuations, $\frac{\Delta S}{\langle S_{z}^{\text{tot}} \rangle}$, (with respect to disorder configurations) of the average magnetization $\langle S_{z}^{\text{tot}} \rangle$. Fig. 7(b) and (d) show $\frac{\Delta S}{\langle S_{z}^{\text{tot}} \rangle}$.
FIG. 8: (Color online) Distribution of local magnetizations $\langle S^z_i \rangle$ for $\lambda=a/2$, at three different temperatures $T_1$, $T_2$, and $T_3$, which correspond to $\langle S^{tot}_z \rangle=0.75S$, $0.4S$ and $0.1S$ respectively. Left column: Inhomogeneous case for $x_{ns}=0.05$ (with $r_0=2a$ and $x_{in}=0.8$). Right column: Homogeneous case. (The red dashed lines indicate the values of $\langle S^{tot}_z \rangle/S$ corresponding to the three different temperatures).

at $T/T^*=0.6$ as a function of $1/N$, corresponding to $\lambda=a$ and $\lambda=a/2$, respectively. For $\lambda=a$, it is almost constant by varying the system sizes, with a value of around 0.42. Interestingly for the short-ranged couplings, $\frac{\Delta S}{\langle S^{tot}_z \rangle}$ decays monotonously, and indicates that it saturates to a value of about 0.7 in the thermodynamic limit. It is important to note that for homogeneously diluted systems $\frac{\Delta S}{\langle S^{tot}_z \rangle}$ vanishes in the thermodynamic limit. This means that the total average magnetization is not a self averaging quantity in the presence of inhomogeneities.

In order to get a deeper insight into the temperature dependence of the magnetization in the presence of inhomogeneities, we analyze the nature of the distributions of the local
magnetizations. In Fig. 8, we compare the distributions of $\langle S^i_z \rangle$'s in the inhomogeneous systems with those of the homogeneously diluted ones, for $\lambda=a/2$. The distributions are shown for three different temperatures $T_1$, $T_2$, and $T_3$, which correspond to $\langle S^\text{tot}_z \rangle=0.75S$, $0.4S$, and $0.1S$, respectively. The distributions at the lowest temperature $T_1$ are almost similar for both cases, except for an extended tail toward the small magnetization values in the inhomogeneous system. At the intermediate temperature $T_2$, the distributions reveal a more pronounced difference. It has a Gaussian-like shape for the homogeneous systems, whereas it has a clear bimodal nature in the inhomogeneous ones. It is also wider than that of the homogeneous case. There is a significantly higher weight below $\langle S^i_z \rangle=0.2$ for the inhomogeneous systems. This relatively high weight corresponds to the impurities outside and far from the clusters. The broad peak around $\langle S^i_z \rangle\approx 0.7$ is associated with the impurities inside the clusters. Finally, for the highest temperature $T_3$, the distribution in the homogeneous systems is unimodal with a small tail at large $\langle S^i_z \rangle$'s and the half-width is of the order of $\langle S^\text{tot}_z \rangle=0.1$. On the other hand, in the inhomogeneous compounds, we observe a very strong weight for $\langle S^i_z \rangle\leq 0.05$ and an extended tail which goes up to $4\langle S^\text{tot}_z \rangle$. For $T=T_3$, an estimate of the percentage of impurities within the range of $0.05 \leq \langle S^i_z \rangle \leq 0.15$ gives 65% for the homogeneous systems and only 15% in the presence of inhomogeneities.

To conclude this discussion, we provide in Fig. 9 a real space illustration of the variation of the local magnetizations. The 2D-snapshots, are shown for both kinds of systems, at the temperatures $T_1$, $T_2$, and $T_3$, corresponding to $\lambda=a/2$.

Thus, to summarize, we have presented a detailed and comprehensive study of the effects of nanoscale inhomogeneities on the spontaneous magnetization in diluted magnetic systems, which had hitherto been unexplored. We have compared the average magnetization behavior in inhomogeneous systems to that of the homogeneously diluted case, for different ranges of the magnetic couplings. Unlike the convex nature of the spontaneous magnetization in homogeneous systems a linear temperature dependence (over the entire temperature range) is obtained in the inhomogeneous compounds for intermediate couplings. Whereas it exhibits a pronounced concave shape in systems with short-ranged interactions. Additionally, the local magnetizations show bimodal and wider distributions in contrast to that of the homogeneously diluted systems, where it remains unimodal. The finite size analyses have revealed that the fluctuations of the average magnetization (with respect to disorder configurations) remain finite in the thermodynamic limit in inhomogeneous systems, unlike the
FIG. 9: Snapshots of the local magnetizations in a 2D-plane for $\lambda=a/2$. The top panel corresponds to the homogeneous case, and the bottom panel to the inhomogeneous case for $x_{ns}=0.05$. $T_1$, $T_2$, and $T_3$ denote the temperatures when $\langle S_{z}^{tot}\rangle=0.75S$, 0.4$S$ and 0.1$S$ respectively. (Here $N=24^3$).

homogeneously diluted case for which it vanishes. We believe it would be of great interest to corroborate our findings by future experimental studies.

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