Model for spin coupling disorder effects on the susceptibility of antiferromagnetic nanochains

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The temperature dependence of the static magnetic susceptibility of exchange-disordered antiferromagnetic Heisenberg spin-1/2 finite chains with an odd number of spins is investigated as a function of size and type of disorder in the exchange coupling. Two models for the exchange disorder distribution are considered. At sufficiently low temperatures each chain behaves like an isolated spin-1/2 particle. As the size of the chains increases, this analogy is lost and the chains evolve into the thermodynamic limit behavior. The present study provides a simple criterion, based on susceptibility measurements, to establish when odd-sized chains effectively simulate a single spin-1/2 particle. © 2009 American Institute of Physics. [DOI: 10.1063/1.3072361]

Precise placement of individual atoms in a host material would allow improvements in the performance of currently available devices,1 fabrication of new devices,2 as well as experimental verification of basic properties predicted for low-dimensional systems.3 Control over atomic positioning on surfaces was first achieved over a decade ago. More recently, significant progress has been reported in the contexts of magnetic nanochains4,5 and of donor placement in Si.6–8

Accurate dopant positioning (e.g., P) in Si is motivated not just by the trend dictated by Moore’s law, requiring fabrication of smaller and increasingly precise devices in Si, but also by proposals for Si-based quantum computers,2 where the qubits involve the electronic and nuclear spins of shallow donors. Spin 1/2 particles are ideal candidates for qubits in Si because of their limited interactions with their environment, leading to long coherence times.9 However, the proposed one- and two-qubit gates, driven by electric and magnetic fields, would have to be controlled within the length scale of a single spin.2,10 An alternative definition of a qubit, for which conditions on field control would be less severe, has been proposed by Meier et al.11,12 They have investigated the magnetic behavior of antiferromagnetic (AF) clusters of spin-1/2 particles, which exhibit a $S^z = \pm 1/2$ doublet ground state13 and could be used to define a logical qubit. Quantum gate operations would not affect the coupling between spins within the chains and would require the control of electric and magnetic fields on the length scale of the spin array.

Linear arrays with odd number of P atoms in Si, in which the coupling between the electronic spins on adjacent P donors is AF,2 meet in principle the above conditions for defining qubits. However, the intensity of such coupling is highly sensitive to the relative position of the donors.14,15 Changes in the donor positioning of just one lattice parameter may alter the strength of the coupling by orders of magnitude. Therefore, at the level of precision so far achieved in the techniques of sample preparation, which is of about 1 nm,6,8 these chains are bound to exhibit some degree of disorder in their structure, leading to fluctuations in the exchange interaction $J$ between magnetic moments in the chain. When the P atoms are positioned along a single [100] crystal axis, with the interdonor separations distributed around some target value, $J$ remains restricted to an interval around the average value $J_0$.9,15 Assuming that fluctuations in the impurity positioning along the [100] axis are of the order of 1 nm, the probability distribution of the exchange interaction can be well described by a trimal one,

$$P_{\text{tri}}(J) = (1/3)\{\delta(J - J_0) + \delta(J - (1 + W)J_0) + \delta(J - (1 - W)J_0)\},$$

(1)

where $0 < W < 1$ is a parameter giving the degree of dispersion of $J$. On the other hand, slight deviations on the P positioning, on the order of the interatomic distance with respect to the perfectly aligned chain along a [100] axis, lead to important differences. The distribution of values of $J$ turns out to be peaked near $J=0$,15 and can be modeled by an exponential one,

$$P_{\text{exp}}(J) = \frac{1}{J_0} e^{-J/J_0} \Theta(J),$$

(2)

where $\Theta(J)$ is the unitary step function. In any case, the occurrence of disorder in the exchange interaction within the cluster is a relevant ingredient in determining its magnetic behavior. It is therefore a key issue regarding the practical use of such clusters, in particular to define qubits.

In the case of ordered linear chains of atomic spins assembled on an insulating surface, scanning tunneling microscopy measurements have been recently used to investigate the magnetic excitation spectra of such systems, on the basis of which the strength of the coupling between the spins could be assessed.5 However, for chains buried in the host bulk material, as in the case of donor-based spin qubits,2 such an approach would not be applicable.

We show here that susceptibility measurements on samples with chains of odd number of magnetic spin 1/2 particles constitute a valuable tool for investigating their magnetic behavior, in particular to determine the conditions under which these chains behave as an effective spin 1/2 particle. Susceptibility measurements on a different nano-
scale system, namely, molecular nanomagnets, have proven useful to characterize their magnetic behavior.\textsuperscript{16}

We have carried out a detailed study of the temperature behavior of the averaged susceptibility per spin $\langle \chi_N \rangle$ of AF spin-1/2 chains with odd number $N$ of spins. We have focused our attention on two fabrication-related factors affecting the magnetic properties of such chains, namely, their length and type of disorder in the exchange interaction. Both trimodal and exponential exchange distributions, as given by Eqs. (1) and (2), respectively, have been considered. For the former, $W$ was set equal to 0.5, which corresponds to a relatively wide disorder distribution. For $N=3$ average susceptibilities have been calculated analytically, whereas for larger sizes, quantum Monte Carlo simulations have been performed.\textsuperscript{17} In what follows, susceptibilities are given here in units of $\chi_0=g^2\mu_B^2/J_0$, and temperature in units of $J_0/k_B$, where $\mu_B$ is the Bohr magneton and $g(=2)$ is the Landé factor. In these units, the static susceptibility of a single Heisenberg magnetic moment is given by $\chi_1=S(S+1)/3T$, so that $T\chi_1=1/4$ when $S=1/2$.

At low temperatures ($T \ll 1$), the behavior of $\langle \chi_N \rangle$ should be analogous to that of a single $S=1/2$ spin.\textsuperscript{11,12} We have investigated the extent to which such analogy holds under the combined effects of disorder, temperature, and chain length $N$. Figure 1(a) shows the susceptibility ratio $\chi_1/(N\langle \chi_N \rangle)$ at $T=1/32$ for odd values of $N$ ranging from 1 to 17, and for the two exchange distributions. Such ratio provides us with a simple quantitative criterion for assessing if the clusters behave collectively as a single spin. In this case the ratio should be 1. We notice that for the trimodal distribution, the susceptibility ratio remains close to 1 over a relatively wide range of values of $N$. However, in the case of exponential disorder, significant deviations from 1 rapidly occur as $N$ increases above 3. It is also interesting to look at the behavior of the susceptibility ratio as a function of $T$, for fixed $N$. Results for $N=9$ are presented in Fig. 1(b) for the two exchange distributions. We clearly see that for the trimodal distribution the ratio remains close to 1 over a temperature range wider than the one corresponding to the exponential one. As $T$ increases, results for the two disorder distributions merge and the susceptibility ratio tends to $1/N$, indicating that in the high-$T$ limit the chain breaks into $N$ independent spins. In all cases, the deviation of the susceptibility ratio from 1, for a given temperature and type of disorder, increases with $N$. We conclude that using longer chains as qubits,\textsuperscript{11,12} with the advantage of facilitating qubit control by external fields, also brings more severe requirements in terms of achieving sufficiently low temperatures.

For both low and high temperature regimes, $\langle \chi_N \rangle$ exhibits a Curie-like behavior, though with different prefactors. In the former, the coefficient is equal to 1/4$N$, whereas in the latter, where thermal fluctuations overcame the exchange interaction, it is equal to 1/4. This can be clearly seen by plotting $(N\langle \chi_N \rangle)^{-1/2}$ as a function of log($T$) over a rather wide range of temperatures, as in Fig. 2. In this sort of plotting, Curie-like ($\sim 1/T$) behavior is represented by a horizontal line. The figure shows results for different values of $N$ and for exponential (a) and trimodal (b) distributions. In both cases, we notice that in the intermediate temperature region, $(N\langle \chi_N \rangle)^{-1/2}$ shows a linear behavior with log($T$), which becomes more pronounced as $N$ increases. We remark that such behavior represents a precursor of the Fisher\textsuperscript{18,19} scaling law, according to which in the thermodynamic limit ($N \rightarrow \infty$) the plot of $(N\langle \chi_N \rangle)^{-1/2}$ versus log($T$) results in a straight line whose slope depends on just the exchange coupling disorder distribution.\textsuperscript{17,20} Departure from the Fisher scaling behavior below some characteristic temperature $T^*$ signals the onset of the single spin behavior. Such temperature should provide an estimate for the magnitude of the gap between the ground state doublet and the first excited state. A plot of $\langle \chi_N \rangle$ as a function of $NT$ in a double-log scale for several odd values of $N$ is presented in Fig. 3. We note that for $NT \leq 1$ all curves collapse onto a single one, indicating the onset of the single spin behavior. It follows that $T^*=1/N$, supporting our interpretation, in terms relating $T^*$ to the first excitation gap, which is known to scale with the inverse of the number of sites in the chain.\textsuperscript{12}
In conclusion, we have studied the temperature behavior of the magnetic susceptibility of odd-numbered linear spin chains. Our calculations encompass a wide range of values of $N$ and of $T$, and clearly establish the existence of three temperature regions where the spin susceptibility of the chains exhibit distinct behaviors. Disorder in the intrachain exchange couplings plays an important role in our results. For a disorder distribution that does not include extremely small values of $J$, the spin cluster analogy with single spin 1/2 particles remains robust at low $T$, even if the distribution is considerably wide, as in the case of the trimodal distribution considered here. However, if arbitrarily small values of $J$ occur, which is probably unavoidable under current samples fabrication capabilities, restricting the system dynamics to the doublet ground state manifold would require unrealistically low operation temperatures. We recall that in the case of donor-based spin qubits, sample preparation should include an overgrowth stage after atomic positioning at a surface. We have also determined how the temperature below which the cluster behaves as a single spin $T^*$ scales with $N$, establishing its relation with the first excitation gap. The present work sheds light on relevant points regarding the magnetic behavior of AF nanochains and its perspective for application as qubits in spin-based solid-state devices.

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FIG. 3. (Color online) Average susceptibility for odd number of spins chains vs $NT$ in the case of trimodal disorder. The data suggest that $T^*(N) \sim 0.8/N$ for this particular type of disorder.

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