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Mutual verification of two new quantum simulation approaches for nanomagnets

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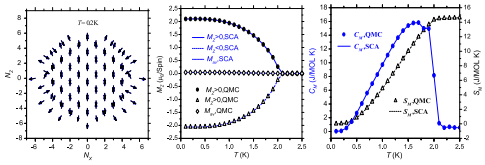
HIGHLIGHTS

- Our new quantum Monte Carlo approach was improved significantly in this work.
- The two new quantum simulation approaches were applied separately to the same nanoparticle.
- All simulations were started from a random magnetic configuration above T_M.
- The two simulation approaches have generated exactly identical results.
- Therefore, the two simulation approaches are now verified by each other.

GRAPHICAL ABSTRACT

We applied our two new quantum simulation approaches to the same antiferromagnetic nanoparticle and obtained exactly identical calculated results.



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ABSTRACT

Two new quantum simulation methods, which we developed recently based on the Metropolis and self-consistent algorithms defined as QMC and SCA approaches respectively, were employed to investigate the magnetic properties of an antiferromagnetic nanoparticle with strong surface anisotropy. All simulations were started from a random magnetic configuration and carried out from a temperature well above the phase transition stepwise down to very low temperatures as other researchers have been doing in classical Monte Carlo (CMC) simulations. It turns out that the magnetic structures, magnetizations, total (free) energy, magnetic entropy and specific heat calculated by means of the two approaches are well consistent with each other, thereby verifying their correctness mutually.

1. Introduction

Over the last half century, classical Monte Carlo and micromagnetic simulations [1–3] have served as the two main tools to study the physical properties of magnetic materials numerically. However, these two methods were built upon classical physics. When they are implemented in computing programs, all magnetic moments in the sample are treated as classical vectors of fixed lengths, though their orientations can be rotated by the local effective magnetic fields. As indicated by Usov and Gudoshnikov [4], due to their classical nature the two methods are naturally neither able to accurately describe various interactions involved nor flexible enough to handle complex nanosystems, such as those of very tiny size or composed of different magnetic ions in a unit crystal cell.

Although a quantum Monte-Carlo model for spin systems was proposed 20 years ago [5,6], few persons have utilized it to study the magnetism of nanosystems at least to our best knowledge,

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perhaps because it is too abstruse theoretically and very time-consuming computationally. Moreover, when it is employed, only the ground state of the system is considered. However, in fact at finite temperatures many excited states of real systems are populated, they must be taken into account in order to evaluate the physical quantities more precisely.

To cope with the problems, we have built a new quantum simulation approach based on the self-consistent algorithm and have successfully applied it to study the magnetic nanoparticles consisting of the 4*f* [7–9] and 3*d* [10–13] ions, respectively. The SCA method was assumed to be built upon the principle of the lowest free energy [7–13]. That is, as the computational program implemented with the SCA algorithm runs, all magnetic moments in the nanosample are rotated and their magnitudes adjusted by the local effective magnetic fields to minimize the total free energy of the nanosystems spontaneously according to the law of the lowest free energy. This hypothesis has been proved by our recent simulations performed for a pair of ferromagnetic and antiferromagnetic nanowires [12,13]. Though no experimental data of the nanomagnets are available to make direct comparison, our calculated magnetic structures, temperature dependence of specific heat, and magnetization hysteresis curves, etc., all look reasonable. Especially, our simulated magnetic structures for a $DyNi_2B_2C$ nanoball, assumed to be cut out of the body-centered tetragonal crystalline, showed good agreement with that observed in the bulk sample: below the magnetic transition temperature T_M, the ions on an *ab*-plane inside the core align ferromagnetically in the [110] direction, and two adjacent *ab* layers orders antiferromagnetically.

For a simple nanosystem, its magnetic structure can be easily initialized properly before starting simulation, the SCA algorithm is able to lead the computing code to converge very quickly to the equilibrium state. However, in reality a nanosystem may be very complicated both magnetically and geometrically, then we hardly know how to initialize it. A simulation started from an improperly initialized magnetic structure might be trapped in a local minimum of the total free energy so that the code converges to a wrong state. To avoid such case, we have developed a new quantum Monte Carlo (QMC) method using the Metropolis algorithm [11]. The new quantum approach enables us to start simulations from a random magnetic structure and leads the computing program to reach the correct equilibrium state finally.

However, when the new QMC approach was first employed [11], the computations were mostly done with the *combined* simulation method. That is, at every temperature, the QMC module is firstly called to run for a certain number of loops to help the code tunnel through the energy barriers one by one, then the SCA module is invoked which leads the code to converge quickly down to a local minimum of the total free energy. Such procedure must be repeated many times so that all local minima are visited and compared, the state of the globally least minimum is finally chosen as the equilibrium state. Moreover, at that time all simulations, including those using the SCA approach, were carried out from a very low temperature stepwise up to high temperatures.

But, for a complicated nanosystem if we start simulations from a very low temperature with a random magnetic structure, it is sometimes very hard to generate any reasonable results [4]. For the sake, in the current work all simulations have been started from a temperature well above the magnetic transition temperature T_M, then stepwise down to very low temperatures as other researchers have been doing by means of the classical Monte Carlo and micromagnetic methods. Moreover, the SCA and the new QMC approaches have been applied *separately* to the magnetic nanograin, and the calculated magnetic structures, magnetization, free energy, specific heat, etc., in the absence of external magnetic field are well consistent with each other, thereby verifying the correctness of the two simulation methods.

2. The new quantum Monte Carlo model

In our new quantum Monte Carlo model, the Metropolis algorithm is employed and incorporated with quantum theory. In every QMC step, a spin is selected randomly from the considered magnetic system and then rotated randomly in a narrow cone around its original direction. If this operation is able to lower the energy of the ion, the new orientation is accepted. Otherwise, a random number r in the interval (0,1) is generated to compare with $p_i = \exp(-\Delta\varepsilon_i/k_BT)$, where $\Delta\varepsilon_i$ denotes the energy increment of the considered i-th ion resulted from the rotation. If $r \le p_i$, the new direction is also accepted, otherwise discarded. Such steps are repeated many times until a certain accuracy is reached. In our previous work [11], we used the acceptance ratio as the criterion of convergency. However we found that the accepting ratio could not be reduced efficiently after many loops as expected especially at high temperatures. For the reason we now define new criteria. Assuming the i-th spin $\mathbf{s}_i$ originally orients along $\hat{\mathbf{s}}_i = \vec{i}\cos\alpha + \vec{j}\cos\beta + \vec{k}\cos\gamma$, where α, β and γ are the angles between $\mathbf{s}_i$ and the Cartesian coordinate axes, and after one QMC loop it has been rotated to a new direction $\hat{\mathbf{s}}_i' = \vec{i}\cos\alpha' + \vec{j}\cos\beta' + \vec{k}\cos\gamma'$. To monitor the rotations of all spins a quantity

$$\tau = [(\cos\alpha - \cos\alpha')^2 + (\cos\beta - \cos\beta')^2 + (\cos\gamma - \cos\gamma')^2]^{1/2}, \quad (1)$$

is defined and calculated for every spin. If the τ's of all spins are less than a given value τ_0, convergency is assumed to be reached. On the other hand, if the total energy only changes slightly in a few hundreds of loops, convergency is also considered to be reached. Afterwards, the magnetization, total (free) energy and magnetic entropy of the system are evaluated at the temperature by averaging over the last a few hundred or thousand loops as done in CMC simulations. Since this new QMC approach is able to calculate all eigenstates of any nanosystems containing arbitrary large spins, it thus enables us to evaluate the physical properties of real magnetic systems at high temperatures more accurately than the QMC method proposed by previous authors.

3. Calculated results

Now we consider a nanoball consisting of 3*d* ions with spin $S=5/2$ which are coupled antiferromagnetically through Heisenberg exchange interaction. For simplicity the nanoball, whose radius $R=6a$, is assumed to be cut out of a bulk cubic crystalline. A coordinate system is built with its origin at the center of the ball and its *z*- and *x*- axes along the *c*- and *a*- axes of the crystalline, respectively. The interaction in the nanosystem can be described by a Hamiltonian [10–13]

$$\mathcal{H} = -K_S\sum_i(\mathbf{n}_i\cdot\mathbf{S}_i)^2 - K_C\sum_i(\hat{\mathbf{z}}\cdot\mathbf{S}_i)^2 - \frac{1}{2}\sum_{i,j\neq i}\mathcal{J}_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - \mu_B g\sum_i\mathbf{S}_i\cdot\mathbf{B}, \quad (2)$$

where the first two terms denote the anisotropy on the surface and inside the nanoparticle with strength K_S and K_C, respectively, the third term represents the Heisenberg exchange interaction among the 3*d* magnetic ions, and the last one stands for the Zeeman energy under the interaction of an external magnetic field. For convenience, only the nearest Heisenberg exchange interaction among the spins are considered and the exchange strength was assumed to be same through out the whole nanoparticle, that is, $\mathcal{J}_{ij} = \mathcal{J}$, and the above parameters were assigned to $\mathcal{J}/k_B = -0.1$ K, $K_C/k_B = 0.2$ K, and $K_S/k_B = 0.3$ K, to perform all