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Success of a simulation approach for magnetic nanosystems: Power of physical laws

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HIGHLIGHTS

- Our new quantum simulation method was successfully applied to a ferromagnetic nanowire.
- To verify the correctness of the new approach, all simulations were started from a random magnetic configuration.
- The total free energies of the nanosystem were found to decrease spontaneously in simulations.
- So our hypothesis that the new approach is built on the principle of lowest free energy is proved.
- Using the new approach, the computing speed may be considerably accelerated.

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ABSTRACT

The new quantum simulation model and the self-consistent algorithm (SCA) for magnetic nanosystems, that we proposed 2 years ago, were extended to study the magnetic properties of a nanowire consisting of 3d ions which are coupled ferromagnetically. To test the applicability of the algorithm, our simulations in the present work were started from a magnetic structure in which all spins in the whole nanosample were *randomly* oriented (defined as the random magnetic configuration for later use) as other authors have been doing with Monte Carlo or micromagnetism method, and such calculated results were all reasonable. Especially, the free energies evaluated at the chosen temperatures were found to attenuate *spontaneously* and quickly, as the program ran, towards the minima according to the principle of lowest free energy as expected. This suggests that the computational algorithm is able to lead the code to converge rapidly to the equilibrium state automatically without the need to minimize the total (free) energy of the system elaborately that must be done if the Monte Carlo or micromagnetism method is used, demonstrating the great power of natural laws.

1. Introduction

The exotic and peculiar physical properties of magnetic nanosystems are originated from their very tiny sizes, lowered symmetry and broken bonds at the surfaces [1–6]. The spins within the surface shell tend to order randomly in space which gives rise to strong surface effects, and the large surface portion with respect to the tiny sample volume greatly modifies the magnetic structures of the nanosystem since the spins in the core are strongly influenced by the surface anisotropy via the exchange interaction among the neighboring magnetic ions [1].

So far, the micromagnetism theory and the Monte-Carlo method (MC) have been employed worldwide as the two main tools to study the magnetic properties of nanosystems quantitatively [7–10,12,13]. However, these two methods are actually built upon the classical physics [14], as they are implemented in a computing program, the magnetic moments in the nanomagnets are always treated as classical vectors of fixed length, though their orientations can be rotated spatially in a simulating process. For the same sake, when micromagnetism is employed, for instance, the total free energy and its derivatives with respect to the spin orientations of the nanosystem have to be calculated in every computing loop in order to lower the total free energy and thus determine the new orientations of the magnetic moments, until the computation converges down to the equilibrium state where the total free energy of the nanosample is minimized. Obviously, in such a simulating process, many computing loops must be performed, the convergence speed is usually very slow. Moreover, as indicated by Usov, due to the classical nature the above approaches are naturally not flexible to handle the complex

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nanosystems, such as the nanomagnets of tiny size or composed of different magnetic ions in a unit cell [14].

To cope with the problems, Usov and Gudoshnikov [14] proposed a quantum simulation model, where a Hamiltonian

$$\mathcal{H} = -\sum_{i,j\neq i} \mathcal{J}_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - g\mu_B\sum_i \mathbf{h_0}\cdot\mathbf{S}_i - \sum_i D_i(S_i^z)^2, \quad (1)$$

based on Hartree–Fock approximation, was used to simulate the magnetic structures of nanoparticles consisting of 3d ions, which are coupled by Heisenberg exchanges with the strength of $\mathcal{J}_{ij}$ and possibly under the interaction of an applied magnetic field denoted by $\mathbf{h_0}$. In addition, an ion is affected by the single-ion anisotropy of strength D_i. Using this model, he simulated the magnetic configurations of nanoballs containing 3d ions in the absence of external magnetic field, and calculated the hysteresis loops for the nanograins with different particle diameters. In his article, however, only the results for $S=1/2$ were presented. Thus, in other cases the effects of the single-ion anisotropy, that are absent when $S=1/2$, remain to be investigated.

To solve the problems, we have built a new *quantum* simulation model, developed a self-consistent algorithm (SCA), and applied them firstly to the nanoparticles consisting of rare-earth ions ($PrAl_2$, $DyFe_2Si_2$ and $DyNi_2B_2C$ nanoballs) [15–17], and then to the nanosystems containing 3d ions [18–20], respectively. Though no experimental data were available to make direct comparison, our computed magnetic configurations, specific heats, hysteresis curves, etc., for the studied nanosystems, all look reasonable. Especially the calculated results for $DyNi_2B_2C$ of tetragonal crystal structure in the core showed good agreement with those observed in the bulk sample: below the magnetic transition temperature T_M, the ions on an *ab*-plane in the core region align ferromagnetically in the [1 1 0] direction, but the two adjacent magnetic *ab* layers order antiferromagnetically, and those moments on the surface order perpendicular to the surface outwards due to the single-ion anisotropy as a positive value for k_S was used in simulations.

We have assumed that the SCA approach was based on the principle of lowest free energy [15–20]. As a program implemented with the algorithm is running, all magnetic moments in the sample are rotated and their magnitudes adjusted by the local effective magnetic field to minimize the total free energy of the whole nanosystem spontaneously according to the law of lowest free energy. Therefore using this new technique we need neither to calculate the total free energy nor to minimize it elaborately which is however imperative if the MC or micromagnetism method is used, thus the computational speed can be accelerated considerably.

Our new simulation approach works usually well, but in some cases under the condition that the magnetic structure must be initialized properly when a simulation is started from a temperature well below the phase transition. For a simple nanosystem proper initialization can be easily done by considering its magnetic and geometrical symmetries. However, in general we do not know how to do it since the nanosystem to be simulated may be very complicated both magnetically and geometrically. In such a case, a simulation started from an arbitrarily initialized magnetic structure might be trapped in a local minimum instead of the globally least minimum of the total free energy. To overcome the difficulty, our simulation approach was improved with aid of the quantum Monte Carlo (QMC) technique, and used to study the nanoparticles consisting of 3d ions which are ferromagnetically or antiferromagnetically coupled [19]. Thus in a simulation, being started from a random magnetic configuration, Metropolises algorithm helps the code pass through the energy barrier from its one side to another side to get rid of being trapped in a local minimum of the total free energy, the SCA module invoked later leads the program to converge quickly down to another local minimum. After sufficient number of loops, all local minima have been visited and compared, the state with globally least free total energy is finally reached and thus chosen as the equilibrium state of the nanosystem. The calculated magnetic structures, magnetization and specific heat curves for the ferromagnetic and antiferromagnetic nanoballs all look reasonable as presented in the article [19].

However, until now we have not checked if the calculated total free energy really decreases as a simulation is going on when only the SCA algorithm is employed. To test the hypothesis all computations, for a nanowire in the present work, were performed by using the algorithm, and started from a *random* magnetic configuration (RMC). To generate reasonable results, all simulations were started above the magnetic transition temperature as other authors have been doing with the Monte Carlo method for instance. It turns out that such evaluated total free energies at all chosen temperatures indeed decrease quickly towards minima while the code runs according to the principle of lowest free energy, and all our calculated results are reasonable, verifying the correctness of the computing algorithm and especially demonstrating the great power of the natural laws.

2. The quantum model and self-consistent computational algorithm

Now we consider a nanowire consisting of 3d ions with spin S=3/2 which are coupled ferromagnetically through the Heisenberg exchange interaction. For simplicity the nanowire is chosen as a cylinder cut out of a bulk crystal with simple cubic structure, the symmetric axial line of the cylinder is along the *c*-axis of the crystalline, its center is taken as the coordinate origin and the principal axes as the coordinate axes. The radius of its cross section $R=5a$ and the length $L=8R=40a$ where a is the crystal lattice parameter. The Hamiltonian of the whole nanosystem is given by Refs. [18–20]

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

where the first two terms denote the single-ion anisotropy on the surface and inside the nanowire with strengths k_S and k_C, respectively, the third term represents the Heisenberg exchange interaction among the 3d magnetic ions in which the exchange interaction constant λ_{ij} was assumed to be the same through out the wire in all calculations, and the last one stands for the Zeeman energy when placed in an external magnetic field. The above parameters were set to be $k_C=20$ K, $k_S=50$ K, $\lambda_{ij}=\lambda=k_C/2$ to perform all simulations. On the surface, the crystal symmetry is destroyed, so the uniaxial anisotropy, assumed to be along the *z*-axis in the above Hamiltonian, is absent, but all ions on the surface are still subjected to the surface anisotropy which tends to rotate the magnetic moments normal to the surface since k_S is taken to be positive in our simulations. The above Hamiltonian has the same form as that widely employed by most researchers in the classical Monte-Carlo simulations [12,13]. However here **S** is a spin operator rather than a classical vector, so the thermal average of a physical quantity A at temperature T must be calculated with

$$\langle A\rangle = Z^{-1}\sum_n\langle n|\exp(-\mathcal{H}/k_BT)\hat{A}|n\rangle, \quad (3)$$

where Z is the partition function.

3. Numerical results

Following the procedure described above, simulations were performed for the nanowire from $T=100$ K, above the magnetic transition temperature $T_M\approx 90.2$ K which was determined later,