High efficiency parallel quantum transfer matrix simulations of chromium-based molecular rings beyond the giant spin approximation*

P. Kozłowski[1], G. Musial[1], M. Antkowiak[1], G. Kamieniarz[1], and D. Gatteschi[2]

[1] Faculty of Physics, A. Mickiewicz University, ul. Umultowska 85, PL-61-614 Poznań, Poland
[2] Department of Chemistry, INSTM and Universitá di Firenze, via della Lastruccia 3, I-50019 Sesto Fiorentino, Italy

Abstract. The symmetric parallel processing is employed to facilitate the numerically exact quantum transfer matrix (QTM) calculations. The message passing is applied and the MPI library is exploited, ensuring full portability and functionality of an application. Different processes of the parallelized job calculate the parts of the partition function and the mean values of total spin components. We demonstrated its functionality by calculating torque and specific heat for the s=3/2 quantum spin system modeling Cr_8 molecular ring. The Hamiltonian considered contains single ion anisotropy and alternating nearest neighbor exchange integrals. We present results obtained in low temperatures where the convergence of the QTM method is slower and sequential calculations become very time consuming. The parallel processing allows obtaining results in real time. The dependence of speedup and of efficiency versus the number of parallel processes is studied and discussed.

Key words: parallelization of processing; MPI; magnetic rings; Heisenberg model; numerical simulations

1 Introduction

Molecular rings of transition-metals immersed in a non-magnetic environment are very good examples of mesoscopic systems in which quantum phenomena can be observed on a macroscopic scale. Therefore they became of interest as potential building blocks of envisaged quantum computers or as a basis of an efficient storage device [1]. The intra-cluster ferro- or antiferromagnetic interactions are usually modelled with a Heisenberg Hamiltonian. Yet, there are subtle, but important for future applications, phenomena like e.g. S-mixing, which cannot be described within the giant spin approximation [2]. Therefore the modeling of molecular rings requires inclusion of anisotropic terms to the Heisenberg Hamiltonian. Then, the total spin is not any more a good quantum number and the

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symmetry of the model lowers, which makes exact algebraic approaches unfeasible. Instead, the perturbative methods are used, but it is difficult to estimate their accuracy.

We present a non-perturbative quantum transfer matrix numerically exact approach. We have introduced the symmetric parallel processing which is necessary to obtain results in real time, especially in low temperatures where the convergence of the QTM method is slower and sequential calculations become too time consuming.

We have used the MPI library [4] to parallelize the computational process in the subject simulations. The main reason for this choice is that message passing is the most effective and universal among the parallel computational models, as it effectively works in every computer system with the distributed, shared or mixed type of memory and fits well separate processors connected by fast or slow communication network.

To show high efficiency of the method, we present results for the magnetic torque and specific heat calculations in low temperatures, obtained for chromium ring Cr_8. The good scalability of so scheduled parallel simulations is demonstrated by studying the dependence of the speedup versus the number of parallel processes.

2 Physical setup

To model the Cr_8 ring we use the following quantum spin ($s = 3/2$) Hamiltonian:

$$\mathcal{H} = \sum_{j=1}^{4} \left(J^{\mathrm{o}} \boldsymbol{s}_{2j-1} \cdot \boldsymbol{s}_{2j} + J^{\mathrm{e}} \boldsymbol{s}_{2j} \cdot \boldsymbol{s}_{2j+1} \right) + \sum_{j=1}^{8} \left[D(s_j^z)^2 + g\mu_{\mathrm{B}} B \left(s_j^x \sin\theta + s_j^z \cos\theta \right) \right] , \tag{1}$$

where $J^{\mathrm{o,e}}$ are nearest-neighbor exchange integrals for 'odd' and 'even' pairs, respectively, D is the (site-independent) single-ion anisotropy, B is the external magnetic field applied in the $x - z$ plane and forming an angle θ with the z axis. g is the corresponding Landé factor and μ_{B} stands for Bohr magneton. We assume periodic boundary conditions ($j + 8 \equiv j$). We are interested in the magnetic torque τ and the specific heat C:

$$\tau = -g\mu_{\mathrm{B}} \left(\langle S^x \rangle \cos\theta - \langle S^z \rangle \sin\theta \right) , \quad C = -T \left(\frac{\partial^2 F}{\partial T^2} \right)_B . \tag{2}$$

The free energy F and the thermal averages of the total spin components $\langle S^x \rangle$ and $\langle S^z \rangle$ are defined with the help of the partition functions Z, Z_x and Z_z:

$$F = -k_{\mathrm{B}} T \ln Z , \quad \langle S^{x,z} \rangle = \frac{Z_{x,z}}{Z} , \quad Z = \mathrm{Tr} \mathrm{e}^{-\beta\mathcal{H}} , \quad Z_{x,z} = \mathrm{Tr} S^{x,z} \mathrm{e}^{-\beta\mathcal{H}} , \tag{3}$$

with traces over 4^8 dimensional spin space. Computation of above traces is difficult due to the large size of a spin space and the fact that $\mathcal{H}$ consists of non-commuting operators. To overcome these problems we employ the transfer matrix technique to factorise $\exp(-\beta\mathcal{H})$ and run our program in many parallel processes to speedup the calculations.

3 Quantum transfer matrix technique

The Hamiltonian (1) can be written as a sum of two non-commuting Hamiltonians $\mathcal{H}^{\mathrm{o}}$ and $\mathcal{H}^{\mathrm{e}}$. Then, using the Trotter formula, one can express the partition function Z as the limit:

$$Z = \lim_{m\to\infty} Z_m = \lim_{m\to\infty} \mathrm{Tr}\left(\mathrm{e}^{-\frac{\beta}{m}\mathcal{H}^{\mathrm{o}}}\mathrm{e}^{-\frac{\beta}{m}\mathcal{H}^{\mathrm{e}}}\right)^m . \tag{4}$$

To optimise numerical calculations the m-th approximant Z_m can be represented as a product of the matrix V and shift operators $\mathcal{P}$, $\mathcal{P}^\dagger$[3]

$$Z_m = \mathrm{Tr}\left[\left(V\mathcal{P}^\dagger\mathcal{P}^\dagger\right)^4\mathcal{P}^\dagger\left(V\mathcal{P}^\dagger\mathcal{P}^\dagger\right)^4\mathcal{P}\right]^m \tag{5}$$

with $V = \mathrm{e}^{H_{12}} \otimes \hat{1} \otimes \hat{1} \otimes \hat{1}$[3], a sparse matrix with no more than 16 non-zero elements in each column and each row. H_{12} is a two spin Hamiltonian. The trace in (5) is calculated by acting with the operators V, $\mathcal{P}$ and $\mathcal{P}^\dagger$ on all the base vectors. In practice, however, we exploit the symmetry of the system and take into account only non-equivalent vectors. In this way we reduce the number of operations approximately by a factor of 1/10. Due to the sparsity of V, $\mathcal{P}$ and $\mathcal{P}^\dagger$ the number of operations in (5) scales like $mn = m4^8$, whereas with a regular matrix-vector multiplication mn^2 operations are required. Yet in low temperatures, where the slow convergence of Z_m compels calculations for large m, the sequential execution time becomes unrealistic. Therefore we prepared a parallel version of our Fortran code using the MPI library. Different processes of the parallelized job calculate independently their contributions to the partition function.

4 Results and conclusions

Our simulations have been performed on the multicomputer (built out of units with two four-core Intel Xeon 3GHz CPUs) with Open MPI software [4, 5] and InfiniBand hardware and we have been submitting our jobs through PBS. A good parameter to determine the scaling of our computations is the speedup u, which is defined as $u = t_{seq}/t_{par}$, where t_{seq} and t_{par} denote the sequential and parallel execution time of our program, respectively. The dependence of the speedup u on the number of parallel processes p for our program (solid circles) and the ideal speedup (broken line) is presented in Fig. 1. As shown in Fig. 1, the scaling

[3] $\otimes$ stands for the tensor product and $\hat{1}$ is a 16×16 identity operator.

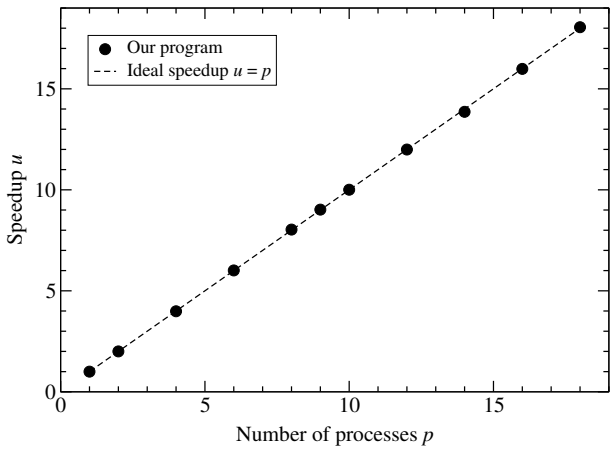

Fig. 1. The dependence of the speedup u on the number of parallel processes p for our program (solid circles) and the ideal speedup (broken line).

is almost perfect, also when processors from different nodes are engaged. This demonstrates high efficiency of our parallel quantum transfer matrix simulations.

Using the application with parallel processing described above, we were able to calculate the torque τ for very low temperatures (around 50mK). At the lowest temperatures we had to reach values of the parameter m in Eq. (4) of the order 2500. Calculations of the specific heat C are feasible in slightly higher temperatures (around 1K) since the scaling versus m is more complex.

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References

1. Meier, F., Levy, J., Loss, D.: Quantum computing with antiferromagnetic spin clusters. Phys. Rev. B 68, 134417 (2003)
2. Carretta, S., Santini, P., Affronte, M., Ghirri, A., Sheikin, I., Piligkos, S., Timco, G., Winpenny, R. E. P.: Topology and spin dynamics in magnetic molecules. Phys. Rev. B 72, 060403(R) (2005)
3. Delica, T., Leshke H.: Formulation and numerical results of the transfer-matrix method for quantum spin chains. Physica A 168, 736-767 (1990)
4. http://www.mpi-forum.org/ – MPI Forum Home Page
5. http://www.open-mpi.org/ – Open MPI Home Page