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Quantum solitons in $CuGeO_3$: a density matrix renormalization group study

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Abstract

The one-dimensional spin–Peierls model with quantum phonons is studied with an optimal phonon method combined with the density matrix renormalization group. By calculating the ground state in the sector $S_z = 1$ I obtain a soliton–antisoliton pair. The soliton is compared to nuclear magnetic resonance experiments which image the soliton lattice present in $CuGeO_3$ under a high magnetic field. Reasonable agreement with experiment can be achieved by choosing a large value of the phonon frequency and the spin phonon coupling constant. This indicates that $CuGeO_3$ is in a non-adiabatic strongly coupled parameter regime. A robust feature of the calculations is an attraction between solitons even without interchain coupling.

1. Introduction

The study of spin–Peierls systems has been invigorated by the discovery [1] of the remarkable spin–Peierls material $CuGeO_3$. A variety of experimental techniques can be applied to this material due to the fact that very good large single crystals can be grown. One striking experiment is the imaging of the soliton lattice using nuclear magnetic resonance (NMR) [2,3]. At low temperature and strong magnetic fields, the magnetization of spin–Peierls materials develops a periodic modulation [4]. In the above-mentioned NMR experiment, the observable is the average value of the spin S_z as a function of spin position along the one-dimensional chains of $CuGeO_3$.

It is natural to try to describe these experiments in terms of a one-dimensional spin–Peierls model, i.e. an isotropic spin-$\frac{1}{2}$ Heisenberg chain coupled to Einstein oscillators (see below for a more complete description). Due to the difficulty of dealing with both quantum spins and quantum mechanical oscillators, as a first approximation it seems reasonable to treat the oscillators classically. Such a model, however, is in gross disagreement with experiment. For example, the calculated spin height W, $W \equiv \max(i, j)|S_i^z - S_j^z|$, where S_i^z is the spin on the ith site, is roughly four times too large in comparison to experiment [5,6]. It has been suggested

that this discrepancy is due to the classical treatment of the oscillators [7,8]. Other experimental observations, e.g. the triplet excitation gap dependence on dimerization [9], seem to require that the ratio of the oscillator frequency Ω to the spin coupling J is of order one, therefore one is in a non-adiabatic regime where the oscillators must be treated with quantum mechanics.

In a previous paper, I have investigated the spin–Peierls model with quantum mechanical Einstein oscillators [10] using the density matrix renormalization group (DMRG) [11]. A special optimal basis technique [12] was used to reduce the dimension of the state space of a single oscillator from infinity to a finite (small) value after which standard DMRG techniques could be applied. As a result of this study, it was verified that quantum fluctuations greatly reduced the value of W and for very large chains there was reasonable agreement with experiment.

Although the above treatment of $CuGeO_3$ seems encouraging, there are several aspects that require further investigation. Firstly, in my previous study, the optimal phonons were determined from a short chain with periodic boundary conditions while the DMRG calculation was done with the infinite-system algorithm with free boundary conditions. Secondly, there was a large finite-size effect with free boundary conditions and a rather uncertain extrapolation to long chains was required to obtain agreement with experiment. Most importantly, there is additional information contained in the NMR measurements. For example, consider the ratio R defined as $R = W/T$ where T is defined as the maximum over i of $|S_i^z + S_{i+1}^z|$. According to my previous calculation (see figure 4 of [10]) for a 100-site chain with reasonable parameter values, the ratio is approximately 5 and extrapolation to larger chains gives an even larger value. This is in poor agreement with the measured R of about 1.5 [7].

The purpose of this paper is improve my previous DMRG study and see whether the improved calculations can produce a better comparison to experiment. The technical improvements will consist of using periodic boundary conditions and the finite-system algorithm. The paper is organized as follows. In the next section I will recall the model and the computational method that I have used. The third part of the paper gives the results of the calculation and the final section consists of conclusions.

2. The model and calculational method

The model I will take for $CuGeO_3$ is the one-dimensional spin–Peierls Hamiltonian. Isotropic spin-$\frac{1}{2}$ operators will represent the Cu atoms while the lattice degrees of freedom will be treated as quantum Einstein operators. To set up the notation, the Hamiltonian is

$$H = H_{ph} + J\sum_i [(1 + g(b_i^+ + b_i))(S_i S_{i+1}) + \alpha S_i S_{i+2}] \qquad H_{ph} = \Omega \sum_i b_i^+ b_i .$$

S_i is the spin operator on the ith site and b_i^+ creates a bond phonon on the ith bond. In this choice of units, $b_i^+ + b_i = x_i$, x_i being the coordinate of the ith oscillator. The four parameters, which must be set by comparison to experiment, are J, the nearest-neighbour spin–spin coupling, g, the spin phonon interaction parameter, α, the second-nearest-neighbour spin–spin coupling, and Ω, the bare frequency of bond phonons. All energies are measured in terms of J (i.e. $J = 1$) so there remain three parameters.

For many of my calculations I will use the parameter value $\alpha = 0.36$. This is consistent with spin susceptibility experiments where a reasonably large next-nearest-neighbour spin–spin coupling is indicated [13, 14]. However, to determine α one has to choose a Hamiltonian and the Hamiltonians used to determine α have assumed classical oscillators. One can imagine that if quantum oscillators are used, a different value of α is necessary. The parameters Ω and g are not very well known and I will report calculations with a range of values. It is important to note that I am working with a highly renormalized Hamiltonian; hence there is no direct relationship between Ω and the phonon frequencies measured in neutron or Raman scattering.

Let us recall what a soliton 'looks like' in this context. First consider the phonon subspace. A soliton then corresponds to a region of transition from one sense of dimerization (i.e. the two values of $\langle x_i \rangle$) to the other sense. Hand in hand with the modulation of expectation value of the oscillator coordinates is a modulation of the expectation value of S_i^z with the maximum value occurring where the minimum coordinate expectation value arises [10]. The soliton is distinguished from the antisoliton in that for the soliton, $\langle S_i^z \rangle$ for sites i even is positive and for i odd is negative, while for the antisoliton $\langle S_i^z \rangle$ is negative for i even and positive for i odd.

The computational approach that I have used is to first find the optimal phonons by the method of Zhang *et al* [12] by studying a ten-site chain with periodic boundary conditions. From this calculation I extract two optimal phonons per site. These phonons are used for calculations on long chains. For every choice of parameter values the optimal phonons need to be recalculated. My previous work [10], where full details of the method are provided (see also [12]), indicates that the transfer of the optimal phonons from short to long chains and working with a small number of optimal phonons per site are both reasonable approximations.

Once the optimal phonons are obtained, I do a standard finite-system algorithm DMRG calculation. The only slightly non-standard aspect of the calculation is the use of periodic boundary conditions. To illustrate how the boundary conditions are handled, consider an infinite-system algorithm calculation. Indeed, to provide input for the finite-system algorithm I first do an infinite-system calculation up to the size of chain in which I am interested. Explicitly, my DMRG for periodic boundary conditions is as follows: start with an eight-site chain (or any other chain where one can find the ground state). There are eight spins and eight bond oscillators with the leftmost oscillator denoted as n_0 and the leftmost spin denoted s_1. Calculate the ground state. From the ground state, form the density matrix for the system consisting of the five leftmost oscillators and the four leftmost spins and find the optimal basis for the system block. Now proceed to a chain with twelve sites, where there are twelve spins and twelve bond oscillators, and consider a truncated state space consisting of product states of the form $|n_0\rangle|s_1\rangle|\lambda\rangle|s_6\rangle|n_6\rangle|s_7\rangle|\lambda'\rangle|s_{12}\rangle$. Here $|\lambda\rangle$ and $|\lambda'\rangle$ are optimal states of the five-oscillator, four-spin block calculated from the eight-site chain. After calculating the ground state in this state space, again compute the density matrix, the system here consisting of the leftmost six spins and seven oscillators. This procedure is then iterated to reach the desired system size, after which one does finite-algorithm sweeps. There is a choice involved for the state $|\lambda'\rangle$. One can choose the states for the right block (i.e. $|\lambda'\rangle$) to be either reflections (as when one uses free boundary conditions) or translations of the states of the left block. I have found that reflected states provide lower energies and better numerical stability; hence reflected states have been used. With my present implementation I am limited to chains of about 200 sites and 80 states in the blocks due to computer time limitations.

3. Results of the DMRG calculations

In this section I will give the results of my DMRG calculations. The calculations are done in the subspace $S_z = 1$; this subspace is of the greatest interest since one expects for $S_z = 1$ the ground state to contain a soliton–antisoliton pair. Since the Hamiltonian is rotationally invariant, we can take a magnetic field along the z-direction. The energies of eigenstates in the magnetic field are equal to the energies when no field is present minus a term proportional to the magnetic field and the z-component of spin S_z. If the magnetic field is large enough, the ground state in the sector $S_z = 1$ is the ground state. From the soliton–antisoliton pair I can extract the quantities W and R discussed in the introduction. W and R can be directly compared to the NMR experiments that image the soliton lattice.

Let us first examine rather adiabatic parameter values ($J/\Omega = 10$), namely $\Omega = 0.1$, $g = 0.2$ and $\alpha = 0.36$. More adiabatic parameter values are easier to compute in the sense that the ground state consists of a soliton–antisoliton pair even for rather small chains. For these parameter values, I will display results for a 100-site chain; however, similar results are obtained for a 52-site chain. For a 100-site chain, with 80 states in the blocks, the ground state energy (for $S_z = 1$) is -77.483. This calculation involved finding the ground state of matrices of rank 100 000 using the Davidson algorithm [15]. Computing and finding the ground state of these matrices for each step in a sweep imposes a size limit of order 80 states for the dimension of the blocks. By way of comparison, the infinite-chain algorithm with 120 states in the block gives an energy of -77.475.

In figures 1(a) and (b) I plot the expectation value of the z-component of the spin at the ith site $\langle S_i^z \rangle$ versus the site. Figure 1(a) is the result of the infinite-system algorithm, while figure 1(b) gives the results after one sweep, additional sweeps having a small effect on $\langle S_i^z \rangle$. It took about three days on an Intel 500 MHz Pentium 2 Xenon processor to compute this figure. In an exact calculation, $\langle S_i^z \rangle$ would be a constant, since the model is translationally invariant for periodic boundary conditions. However, a DMRG calculation breaks translational invariance, allowing a soliton–antisoliton pair to form. Qualitatively, figure 1(a) and (b) are similar; however, there are significant quantitative differences. From figure 1(a), one can extract $W = 0.32$, $R = 2.7$ and from figure 1(b), $W = 0.22$, $R = 2.9$. Since the finite-system algorithm gives a lower energy (-77.483 versus -77.468) I take it to be more accurate and the greater accuracy of the finite-system algorithm is in agreement with standard DMRG 'wisdom' [11]. In comparison to experiment, W at 0.22 is a factor of approximately four times too large and $R = 2.9$ is too big by a factor of approximately 2. It is perhaps not too surprising that I do better with R than with W since the ratio g/Ω has been chosen to be fairly large. From my experience with free boundary conditions, larger ratios g/Ω give smaller R-values. For example, in figure 4 of [10] $g/\Omega = 0.5$, a small ratio, and the R-value is roughly 5. In addition, I have done calculations for $\Omega = 0.1$, $g = 0.1$, $\alpha = 0.36$ using the finite-system algorithm and periodic boundary conditions for a chain with 200 sites. With this smaller value of g/Ω, I find $R = 4.2$. Hence, to reduce the value of R, it seems that the ratio g/Ω has to be large.

A striking feature of figure 1(b), as compared to the less accurate figure 1(a), is that the two solitons appear to attract each other. In experiments, a soliton–antisoliton bound state has been observed [16]. A previous treatment of the spin–Peierls model with quantum phonons, which treated smaller chains, indicated no binding between solitons and the attraction observed in experiment was attributed to interchain interaction [17].

To cure the problem of too large W and R, I have studied the possibility of decreasing the second-neighbour coupling α and the possibility of increasing the phonon frequency Ω. Let us first consider the experimentally less plausible alternative, decreasing α. In figure 2, I have plotted $\langle S_i^z \rangle$ for the parameters $g = 0.2$, $\Omega = 0.1$, $\alpha = 0$ for 100 sites. One notices that the solitons are wider than for $\alpha = 0.36$, with W decreasing to 0.19 and R increasing to 5.3. Hence W improves slightly but R gets much worse. This behaviour can be rationalized by noting that both g and α tend to increase the energy gap between the ground state and excited states. Hence decreasing α tends to decrease the gap which in turn leads to a larger correlation length, that is, wider solitons. Since increasing g decreases R, it can also be expected that a decrease of α will tend to increase R.

To obtain better agreement experiment, one is led to increase the phonon frequency Ω. In figures 3(a) and (b) $\langle S_i^z \rangle$ is shown for $g = 0.4$, $\Omega = 0.2$, with $\alpha = 0$ in figure 3(a) and $\alpha = 0.36$ in 3(b). For the parameter values in figures 3(a), (b), $W = 0.17$ (0.12) and $R = 3.7$ (2.4). It seems that we need, whatever α does, to further increase the phonon frequency. The physical motivation is that larger Ω leads to larger quantum fluctuations and therefore W is reduced.

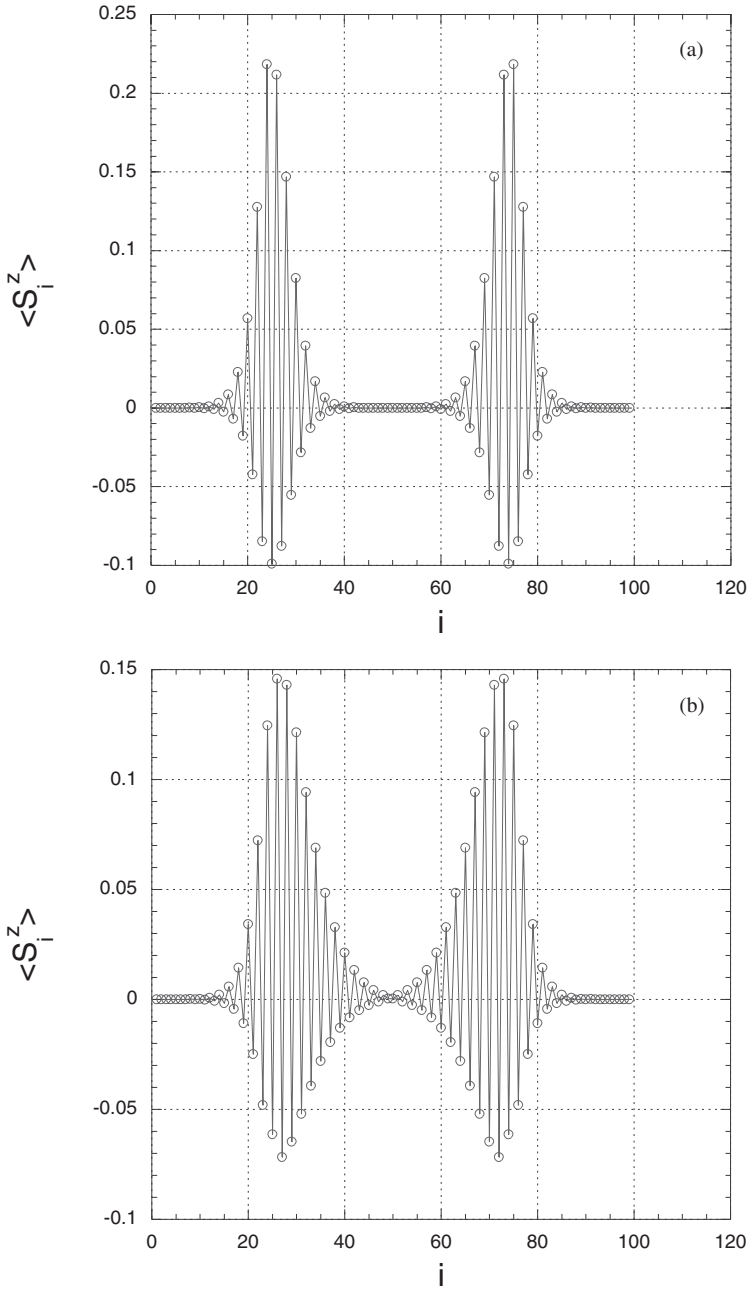

Figure 1. Soliton–antisoliton profiles for a 100-site chain. The parameters are $\Omega = 0.1$, $g = 0.2$ and $\alpha = 0.36$. $\langle S_i^z \rangle$ is plotted as a function of i. (a) was calculated with the infinite-system algorithm while in (b) the finite-system algorithm was used.

I have thus studied the parameter values $g = 1$, $\Omega = 0.5$ and $a = 0.36$ for 100 sites with 80 states in the blocks. However, in this case, the calculation does not converge to a soliton–antisoliton pair. To remedy this problem I have considered a 200-site chain, the rationale being

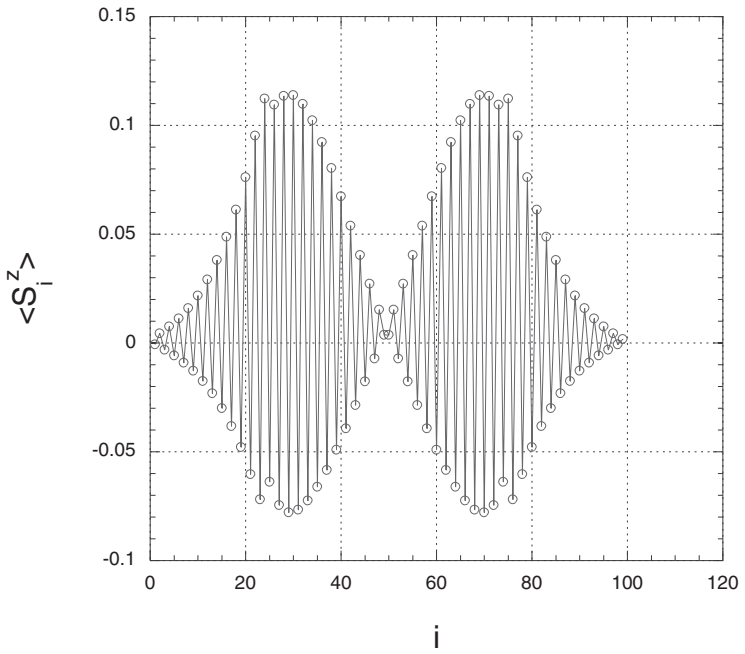

Figure 2. $\langle S_i^z \rangle$ for the parameter values $\Omega = 0.1$, $g = 0.2$, $\alpha = 0$.

that as Ω increases the soliton width increases, and it becomes difficult for the soliton to fit into the smaller system. For 200 sites one does obtain a soliton–antisoliton pair with $W = 0.06$ and $R = 2$. Figure 4 is a graph of $\langle S_i^z \rangle$ for these parameter values. It appears that with a large Ω and g one can get reasonable agreement with experiment.

4. Conclusions

I have made calculations for a soliton–antisoliton pair for a variety of values of the phonon frequency Ω and the spin phonon coupling g. By making proper choices of the parameters, basically large Ω and large g, one can obtain reasonable agreement with NMR experiments for the height W and the ratio R of the soliton. I do not claim that the parameter values that I have found that fit experiment are unique. However, it does seem that fairly large values of Ω are necessary to fit the NMR data. It is interesting that a recent neutron scattering experiment gives different values of W and R, $W \sim 0.2$ and $R \sim 5$ [18]. Therefore, according to the neutron scattering results, one is in a rather adiabatic regime. It has been suggested that this discrepancy between the two experiments arises from the differing timescales probed by NMR and neutron scattering. The timescales probed by neutron scattering are approximately one hundred times shorter than those probed by NMR. Hence, in the neutron scattering experiment there is insufficient time for quantum fluctuations to be important [18].

A robust feature of my calculations is the attraction between solitons even without interchain coupling. For example, there is clear evidence of attraction in figures 1(b), 3(b) and 4. In figure 4 the solitons, at first glance, appear to have a repulsive interaction. However, I am using periodic boundary conditions and hence the solitons are 'touching' at the 0th (200th) site. This attraction between solitons is in agreement with experiment [16] and in disagreement with previous theoretical calculations [17] where shorter chains were considered.

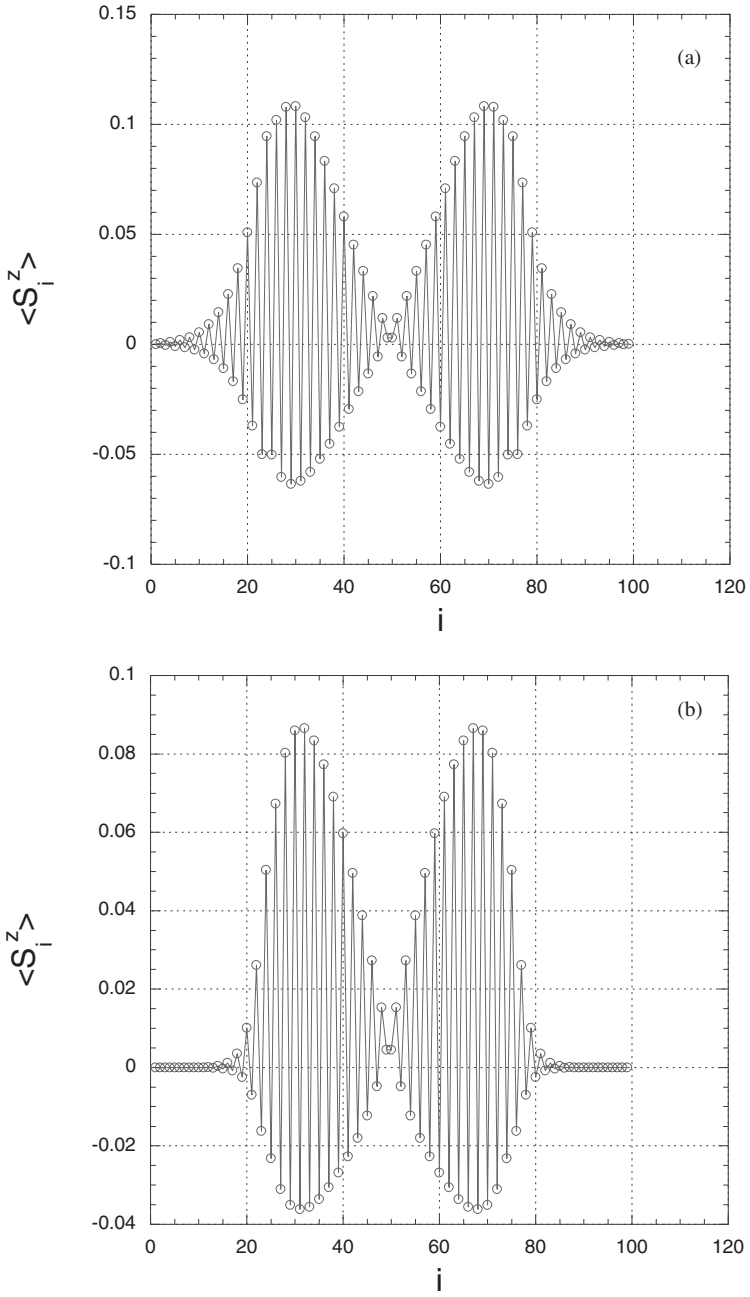

Figure 3. $\langle S_i^z \rangle$ for the parameter values $\Omega = 0.2$, $g = 0.4$. In (a) $\alpha = 0$, while in (b) $\alpha = 0.36$.

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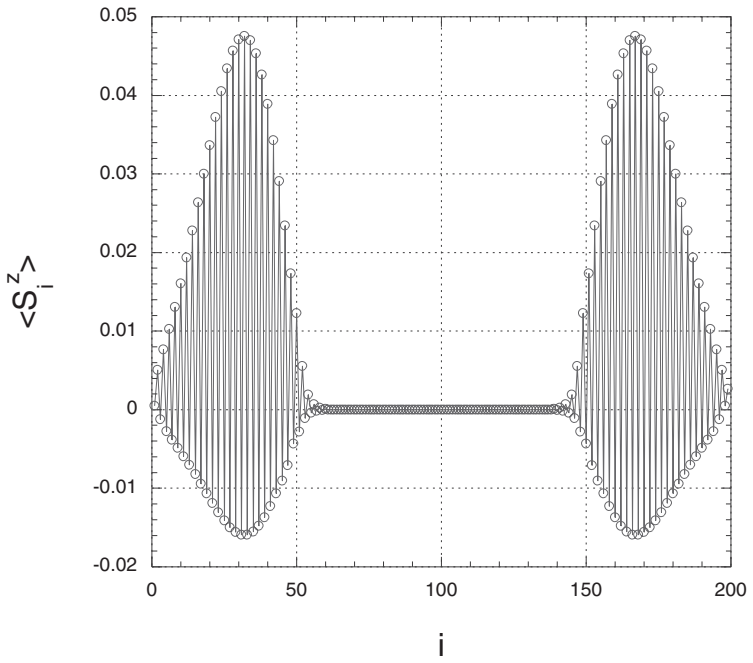

Figure 4. $\langle S_i^z \rangle$ for the parameter values $\Omega = 0.5$, $g = 1$, $\alpha = 0.36$ for a 200-site chain.

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