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Magnetic order near surfaces and corners of a planar Heisenberg antiferromagnet

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Abstract. We study the isotropic spin one-half antiferromagnet on square lattices with (10) surfaces and 90° corners. The magnetization and correlations are calculated using the quantum Monte Carlo (QMC) method of Suzuki and Trotter.

The behaviour of the magnetic order near a surface has been the subject of many studies [1]. For the case of localized magnetic moments, however, most investigations have dealt with systems of classical spins. Then the general feature is a monotonous decrease of the order from the bulk towards the surface, unless the interactions are enhanced there [2, 3]. This is the behaviour one expects intuitively. For Ising models with ferromagnetic bonds it follows rigorously from Griffiths' inequalities [4]. For quantum systems these equalities can be violated [5] and a more complicated variation of the order may occur.

There are a few studies of this problem for Heisenberg models using the spin-wave approximation or Green-function methods. The results for ferromagnetic systems [6, 7] resemble those of the Ising case. As there, only finite temperatures are interesting since the ground state is completely ordered. The situation is different for antiferromagnets where quantum fluctuations exist in the ground state. These fluctuations are modified by a boundary and a spatial variation of the order results even at zero temperature. For a chain with free ends and uniaxial anisotropy a spin-wave calculation gave the following result [8]: the order decreases towards the ends, but does it in a oscillatory way. Moreover, it increases again at the last spin and is larger there than in the bulk. Calculations on BCC and SC films [7, 9, 10] showed that such effects also exist in higher dimensions and depend on the details of the surface geometry. On the other hand, exact calculations for open chains with a fixed spin at one end show no enhancement of the magnetization at the opposite end for $S = \frac{1}{2}$, although the effect is found for $S = 1$ [11].

We have investigated the problem for a system that is more complicated than a chain but still allows us to proceed beyond simple approximations. This is the case for the planar spin one-half Heisenberg antiferromagnet with isotropic exchange between nearest neighbours. Thus we studied the Hamiltonian

$$\mathcal{H} = J \sum_{\langle jk \rangle} \boldsymbol{S}_j \cdot \boldsymbol{S}_k \tag{1}$$

on a square lattice. The extensive work of the past years has shown that the ground state is ordered with a bulk magnetic moment of 0.30 in units of $g\mu_B$ [12–14]. This is a 40% reduction from the maximum (Néel) value. Thus the quantum fluctuations are strong and

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one can also expect sizeable boundary effects. To study them, we have used the quantum Monte Carlo (QMC) method [15], through which the bulk properties were also obtained.

In the following we will consider the magnetic order near a (10) surface and near a 90° corner formed by two such surfaces. While previous simulations dealing with periodic boundary conditions always considered the square of the total staggered magnetization, we are interested here in local quantities. The two geometries are shown in figure 1. For the case of a (10) surface, the lattice was closed to form a cylinder. The spins along one boundary were fixed in staggered positions. This breaking of the symmetry makes it possible to calculate directly the magnetization $m_j = |\langle S_j^z \rangle|$ in the layers [2]. Upon increasing the size of the $L \times L$ system the profile will approach its infinite-lattice limit. This procedure is analogous to the way one calculates classical order parameters via the corner transfer matrix method [16]. For the case of the corner, a different method was used. There the spins along all boundaries were left free and the magnetization was determined from the asymptotic correlation functions between spins on equivalent positions j, j' via $m_j = \sqrt{3|\langle S_j^z S_{j'}^z \rangle|}$. For spins at opposite corners, the distances are relatively large and thus convenient for the extrapolation to $L = \infty$. It was also checked that the two procedures give consistent results.

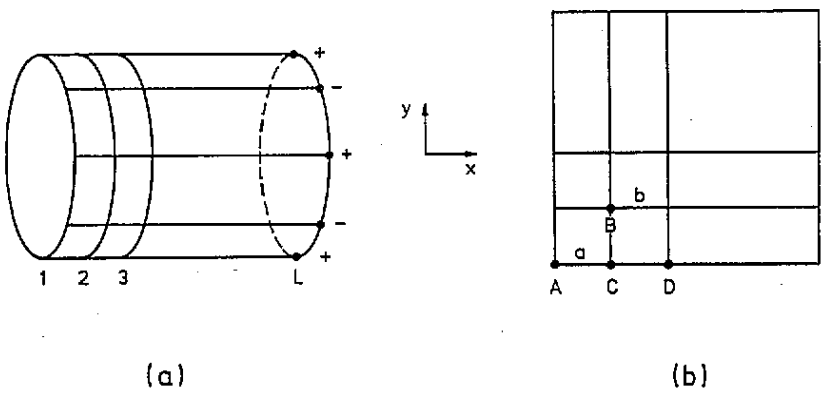

Figure 1. Square lattices with a free (10) surface and fixed spins at layer L (a) and 90° corners formed by free boundaries (b).

In the QMC method one calculates the thermal expectation values of observables $\langle A \rangle = Z^{-1} \operatorname{Tr}(A e^{-\beta \mathcal{H}})$ from which the ground state values can be extracted in the limit $\beta \to \infty$. The trace is rewritten by decomposing $\mathcal{H}$ into two non-commuting parts, $\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2$. For this one uses a chequer-board decomposition of the lattice [12, 17]. Then the Trotter formula [18]

$$e^{-\beta(\mathcal{H}_1+\mathcal{H}_2)} = \lim_{m\to\infty} \left(e^{-\bar{\beta}\mathcal{H}_1} e^{-\bar{\beta}\mathcal{H}_2}\right)^m \tag{2}$$

is applied, where $\bar{\beta} = \beta/m$. Inserting complete sets of eigenstates of S_{tot}^z between the exponentials, one obtains the partition function of a classical (Ising) system with an additional (Trotter) dimension. For fixed m one then deals with a lattice of $L \times L \times 2m$ sites. Since S_{tot}^z commutes with $\mathcal{H}_1$ and $\mathcal{H}_2$, the number of (+) spins remains constant in the $2m$ Trotter planes and one may connect them by so-called world lines, which are closed

in the Trotter direction. The Monte Carlo procedure can then be viewed as a process where these lines are moved around in the classical system. This is done via a number of local spin flips which simulate the quantum fluctuations. They are carried out in cubes of interacting spins forming the three-dimensional system, as described in [17].

At free or fixed boundaries, these processes have to be modified. The flip probabilities, which depend on the local surroundings, are changed and certain types of flips which would move the world lines out of the system, do not appear. Non-zero winding numbers [12, 17], which occur for periodic boundary conditions (in a particular spatial direction), are excluded automatically. Thus for the corner problem only local flips that turn around four spins were used. In the surface case, a global flip corresponding to cutting and reconnecting the word lines was also applied. Thereby the winding number in the y-direction was changed. However, the acceptance rate decreases exponentially in larger systems and the process may be omitted for $L > 12$. All simulations were performed in the subspace $S^z_{\mathrm{tot}} = 0$ with $L^2/2$ world lines which contains the exact ground state [19].

The Monte Carlo averaging involved up to 3.5×10^4 spins and $3\text{–}7.5 \times 10^5$ sweeps with the first $1\text{–}3 \times 10^5$ sweeps used for thermalization. The Trotter limit $m \to \infty$ was taken by a linear fit of the data versus $\bar{\beta}^2$, using five or six values of m so that $\bar{\beta}^2 \ll 1$. To obtain the ground-state values, the temperature was then lowered until no further change of the data occurred. In the surface case, values $T/J = 0.067, 0.067, 0.050$ were found sufficient for $L = 8, 10, 12$, respectively. In the case of the corner, the necessary temperatures could also be estimated by calculating the internal energy in two ways, from nearest neighbour correlations and as a derivative of the partition function. The two results have to agree only for $T \to 0$ due to the restriction to $S^z_{\mathrm{tot}} = 0$. A comparison of the data leads to values $T/J = 0.067, 0.050, 0.030, 0.025$ for $L = 4, 6, 8, 10$, respectively. These temperatures lie well below the gap $E_1 - E_0$, which vanishes as $1/L^2$ [13]. Due to these low temperatures, the Trotter numbers have to be quite large, e.g. up to $m = 179$ for $L = 10$. This was the factor that prohibited the simulation of larger systems. The thermodynamic limit, finally, was taken assuming a $1/L$ dependence of the magnetization as in linear spin-wave theory. It is believed that the exact results also follow this law [12].

We first present some results for the short-range order as measured by the nearest-neighbour correlations $\langle S^z_j S^z_k \rangle$. In figure 2 these correlations are shown for a 12×8 lattice at $T/J = 0.067$. The absolute values at this temperature are actually a few percent larger than in the ground state. One notices oscillations within two or three lattice constants from the surface and the corner. Right at the corner (bond a) the correlation is strongest with a value of -0.151, which is 32% higher than the value -0.114 in the centre of the system. This can be attributed to the constrained motion of the world lines in the corner. The weakest correlation is found for the bond b with a value of -0.106. Altogether the effects at the surface, where the spins have fewer neighbours, dominate and the energy per bond is lower than in the system with periodic boundary conditions. The same has been found for systems with static holes [20], which can be viewed as having inner surfaces. It is also in accord with the results for other antiferromagnets where the number of neighbours can be varied [21, 22].

Oscillations in the short-range order also occur in Heisenberg chains with open ends [23, 24] where the boundary effects decay only slowly (as $1/j$) into the interior. For a chain the basic mechanism can be seen quite easily in the resonating valence bond (RVB) picture. The exact ground-state wave function is a superposition of singlet products [25], or Rumer functions [26]. The simplest case is a ring of four sites. Then there are only two such functions and one has

$$|\Psi_0\rangle = c_1 \{12\}\{34\} + c_2 \{23\}\{41\} \tag{3}$$

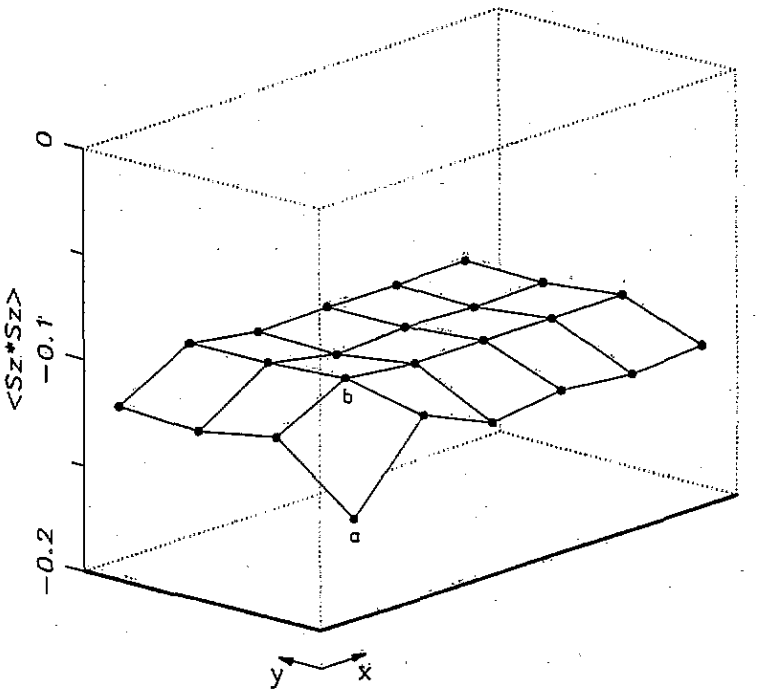

Figure 2. Nearest-neighbour correlations $\langle S_j^z S_k^z \rangle$ in the x-direction. The numerical values are denoted by points and plotted above their lattice positions (see also figure 1(b)).

where $\{ij\}$ denotes a spin singlet. Symmetry then gives $c_1 = c_2$ and the correlations are $\langle S_i^z S_{i+1}^z \rangle = -0.166$. If one cuts the bond between spins 1 and 4, one finds $c_2/c_1 = 0.366$. Thus the singlet $\{41\}$ enters with a lower, and $\{12\}$ and $\{34\}$ with a higher weight than in the ring. As a result the correlation is strongest at the open end: $\langle S_1^z S_2^z \rangle = -0.228$, $\langle S_2^z S_3^z \rangle = -0.083$. The situation is analogous in longer chains.

Within a spin-wave calculation one may argue that there are two competing effects at the end of a chain and at arbitrary surfaces in general [27]. In the Hamiltonian the uniaxial term $J S_1^z S_N^z$ which favours the Néel order, is missing. But obviously this is more than compensated for by the absence of the flip term $J(S_1^+ S_N^- + S_1^- S_N^+)$. This term causes the quantum fluctuations and has a stronger effect on the correlation.

Table 1. Numerical values of the surface and corner magnetization at positions marked in figure 1.

	(10) surface		90° corner	
L	m_1	m_2	m_A	m_B
4	—	—	0.320(3)	—
6	—	—	0.261(3)	0.293(3)
8	0.242(3)	0.255(2)	0.230(4)	0.249(4)
10	0.238(2)	0.250(2)	0.214(5)	0.239(5)
12	0.235(3)	0.244(3)	—	—
∞	0.223(9)	0.224(8)	0.143(6)	0.130(10)

We now turn to the long-range order, i.e. the staggered magnetization. Table 1 gives the numerical results for four selected positions (marked in figure 1) together with the extrapolations to the infinite lattice size. Compared with the nearest-neighbour correlations one sees a clear difference: the magnetization is reduced near the boundary. The effect is quantitatively quite large, about 25% for the (10) surface and over 50% for the corner, compared to the bulk. For correlations between spins in the surface the crossover from enhancement to reduction occurs for distances longer than three lattice constants. Such effects have also been noticed in [20].

It is remarkable that the surface value is practically the same as in the bulk of a honeycomb lattice [28], which has the same coordination number (3) as the boundary spins here. For the surface, one also notices that the first two layers show virtually the same moment, thus forming a small plateau. Beginning with the third layer the magnetization then increases monotonously towards its bulk value. By contrast, a spin-wave calculation does not give this plateau, although it leads to a very similar result (0.217) for the moment in the first layer. Rather it predicts a continuous increase of the order and beyond the first few layers it approaches its bulk limit approximately as $1/j$. More interesting are the corner results. There one finds indeed a small 'oscillation'. The corner spin has a slightly larger magnetization than the next one diagonally inward at position B. From this point of view, the plateau found at the surface can be interpreted as an incipient increase of the boundary magnetization, which then becomes visible at the corner. In figure 3 the value of the order parameter is indicated for some additional sites in the lattice. One sees that also as one moves away from the corner along the surface (A $\rightarrow$C$\rightarrow$D in figure 1) the magnetization first decreases a little and then increases again monotonously.

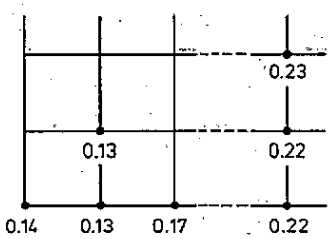

Figure 3. Spontaneous magnetization at various lattice positions in the thermodynamic limit.

In summary, we have found sizeable surface effects resulting from the influence of the geometry on the quantum fluctuations. The results are different, however, for the short and the long-range order. The correlations are enhanced at the surface and show clear oscillations whereas the order parameter displays a more conventional profile. The appearance of oscillations is, in principle, not so surprising. They can be viewed as a result of the antiferromagnetic nature of the order, which favours unit cells with two atoms. Correspondingly, oscillations have also been found around static holes [20, 27]. They also occur in magnets with itinerant electrons. In this case, however, non-trivial order-parameter profiles appear for ferro- and for antiferromagnets [29–31]. Thus the mechanism is different in detail.

Band magnets also show a surface enhancement of the order, which can be related to an increased density of states. In our case the situation is not so simple and the form of the profiles not obvious. Comparing the various results, it seems that also here an 'open' surface with no bonds inside a layer favours a large magnetization [9]. The boundary spins

then have only a minimal number of neighbours to which they must adapt. Whether it is actually possible that the surface order exceeds the bulk value is, however, still not clear. It is therefore planned to study the (11) surface of the square lattice with the QMC method in order to clarify the situation.

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