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LETTER TO THE EDITOR

Theory of the surface spin-flop transition in antiferromagnetic spin chains

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Abstract. A new formulation based on the transfer-matrix method is presented for antiferromagnetic spin chains. The magnetic susceptibility and the spin correlation functions are calculated exactly, and the surface spin-flop phase in the even-spin case is identified as the energetically degenerate or nearly degenerate single-domain wall states, the degeneracy arising from the translational degree of freedom of the domain wall centre. The surface spin-flop to bulk spin-flop transition is found to be a first-order-like transition.

The surface spin-flop (SSF) transition in a semi-infinite antiferromagnetically coupled ferromagnetic layer was first discussed theoretically quite some time ago [1–3]. Unlike the bulk system, in which it is known that the Néel state makes a first-order transition to the bulk spin-flop (BSF) or canted spin state at a critical magnetic field H_{BSF} applied parallel to the ferromagnetic layers, the system of an even number of layers is predicted to have yet another phase, the SSF phase, between these two phases due to the presence of free surfaces. According to the perturbative analysis of the case $H_a \ll H \ll H_e$ in the model 1D spin Hamiltonian (equation (1) taken from [2]), this intermediate phase appears at $H_{SSF} \simeq H_{BSF}/\sqrt{2}$ and develops with an increase in the magnetic field *abruptly*, namely via a first-order transition, to the BSF state at H_{BSF}. Wang *et al* [4, 5] recently studied antiferromagnetically coupled Fe/Cr superlattices experimentally and theoretically. The experimental result for the magnetic susceptibility of the even-layer superlattices confirmed the existence of the transition from Néel to SSF states. Wang *et al* also conducted a numerical analysis of the model spin Hamiltonian (equation (1)) for the SSF transition using a self-consistent mean-field method to determine the ground-state spin configuration. The result reproduced the experimental result for the magnetic susceptibility but argued that, unlike the Keffer–Chow picture, the SSF to BSF transition is continuous. A similar self-consistent mean-field calculation was also done for finite temperatures by Carriço, Camley and Stamp [6] in the study of the phase diagram of thin antiferromagnetic films such as FeF_2 and MnF_2 [7–9]. More recently, Trallori *et al* [10] analysed the ground states of the same spin Hamiltonian in terms of a 2D area-preserving map. To our surprise, however, Trallori *et al* concluded that the SSF state does not exist for the semi-infinite system.

It is to be noted that, for the Fe/Cr superlattice [4, 5], the intralayer exchange coupling is thousands of times stronger than the interlayer one. Moreover, an important fact is that *the experimentally observed magnetization has little temperature dependence* [13]. Thus the system is quasi-1D in the perpendicular direction, giving support to the 1D model spin Hamiltonian description. However, for detailed comparison with experiments, one would still need to include certain effects of intralayer spin fluctuations.

In this letter we present a new analysis of the model spin Hamiltonian (1) below in terms of the transfer-matrix method [11, 12]. Our findings are as follows. (i) The SSF phase does exist for even-spin systems, although we identify the SSF states as *energetically degenerate or nearly degenerate, due to finite-size, one-domain wall* states (due to the translational degree of freedom of the domain wall centre); therefore the SSF state is not necessarily localized to the surface. In contrast, the previous theories [2, 4, 5, 10] considered only the ground state. Departing from the unrealistic $T = 0$ and taking into account excited states nearly degenerate with respect to the ground state, the conclusion drawn by Trallori *et al* should be altered to support the existence of the SSF phase. (ii) In the Ising limit, for which calculation can be done analytically, the nature of the transition from the SSF to the BSF state is found to be of *first order* in that the magnetization is discontinuous over the transition. For the planar Heisenberg case with even spins, the numerically calculated spin correlation functions and the size dependence of the magnetization per site demonstrate that the SSF to BSF transition is *also of first order*. The latter finding (ii) is consistent with our intuition that (i) in the large-size, bulk limit, there should be no difference between cases with even and odd numbers of spins: both should exhibit first-order transition to the BSF phase; and (ii) suppose that a domain wall is localized near the surface for some reason, energetical or dynamical, then again the response thereafter of an even or an odd number of spins to the elevated magnetic field should be the same as that of the bulk, that is, both should undergo a first-order transition to the BSF phase. Throughout the present paper, the term *first order* should be understood as *first-order-like* for the finite-size system.

Let us consider the spin Hamiltonian

$$\frac{\mathcal{H}}{g\mu_\beta S} = \frac{H_e}{2}\sum_{l=1}^{N-1}\cos(\phi_l - \phi_{l+1}) - \frac{H_a}{2}\sum_{l=1}^{N}\cos^2\phi_l - H\sum_{l=1}^{N}\cos\phi_l \qquad (1)$$

where S is the spin size, ϕ_l describes the orientation of the lth spin, H_e the antiferromagnetic exchange coupling, H_a the in-plane anisotropy and H the magnetic field applied in the direction of the easy axis (z axis) of ferromagnetic layers. We evaluate the partition function by the transfer-matrix method:

$$\begin{aligned} Z &= \mathrm{Tr}\,\mathrm{e}^{-\beta\mathcal{H}} \\ &= \int_0^{2\pi}\mathrm{d}\phi_N\cdots\int_0^{2\pi}\mathrm{d}\phi_1\mathrm{e}^{-\beta\mathcal{H}}\int_0^{2\pi}\mathrm{d}\Delta\sum_m\delta(\phi_N-\phi_1+2\pi m-\Delta) \\ &= \int_0^{2\pi}\mathrm{d}\Delta\int_0^{2\pi}\mathrm{d}\phi_N\cdots\int_0^{2\pi}\mathrm{d}\phi_1\mathrm{e}^{-\beta\mathcal{H}}\sum_n\psi_n^*(\phi_N-\Delta)\psi_n(\phi_1) \qquad (2)\end{aligned}$$

where $\{\psi_n\}$ is a 2π periodic orthonormal complete set yet to be determined. Now re-write the Hamiltonian as

$$\frac{\mathcal{H}}{g\mu_B SH_e} = \sum_{l=1}^{N-1}\mathcal{H}_{l,l+1} - \frac{\xi}{4}\left[\cos^2\phi_N + \cos^2(\phi_N-\Delta)\right] - \frac{\zeta}{2}\left[\cos\phi_N + \cos(\phi_N-\Delta)\right] \qquad (3)$$

where $\xi \equiv H_a/H_e$, $\zeta \equiv H/H_e$ and

$$\mathcal{H}_{l,l+1} = \frac{1}{2}\cos(\phi_l-\phi_{l+1}) - \frac{\xi}{4}\left(\cos^2\phi_l+\cos^2\phi_{l+1}\right) - \frac{\zeta}{2}\left(\cos\phi_l+\cos\phi_{l+1}\right). \qquad (4)$$

The set $\{\psi_n\}$ is then chosen to satisfy the transfer-matrix (TM) equation

$$\int_0^{2\pi}\mathrm{d}\phi_l\exp(-q\mathcal{H}_l)\psi_n(\phi_l) = \lambda_n\psi_n(\phi_{l+1}) \qquad (5)$$

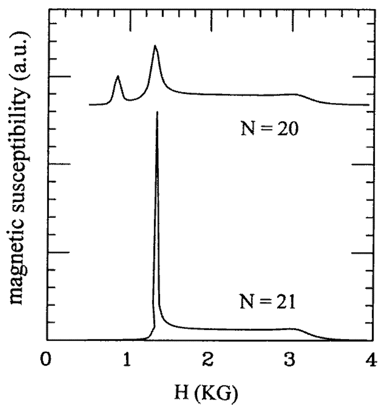

Figure 1. The longitudinal magnetic susceptibility versus the applied magnetic field for $N = 20$ (top) and 21 (bottom) spins.

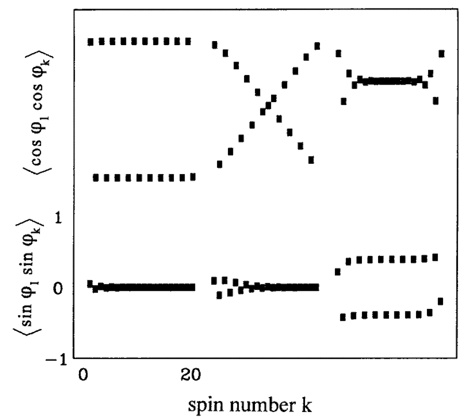

Figure 2. The longitudinal (top) and transverse (bottom) spin correlations for the $N = 20$ planar Heisenberg case. The applied magnetic fields are 0.5 kG (left-hand side), 1.0 kG (middle) and 1.6 kG (right-hand side). All graphs share the same scale: from -1 to 1 along the vertical axis and from 0 to 20 along the transverse axis.

where $q \equiv g\mu_B S H_e \beta$. It is to be noted that $\mathcal{H}_{l,l+1}$ is real and symmetrical with respect to the interchange of ϕ_l and ϕ_{l+1}; therefore the eigenvalues λ_n are real and ψ_n can be taken as real 2π periodic functions.

By using the TM equation (5) repeatedly, the partition function can be reduced to

$$Z = \sum_n \lambda_n^{N-1} G_n^2 \tag{6}$$

where

$$G_n \equiv \int_0^{2\pi} \mathrm{d}\phi \psi_n(\phi) \exp\left[q\left(\frac{\xi}{4}\cos^2\phi + \frac{\zeta}{2}\cos\phi\right)\right]. \tag{7}$$

The spin correlation functions are calculated likewise. Let $f(\phi)$ be any 2π periodic function of ϕ. Then, by repeatedly using the TM equation (5) and the expansion

$$f(\phi)\psi_n(\phi) = \sum_p \langle \psi_p | f(\phi) | \psi_n \rangle \psi_p(\phi) \tag{8}$$

we have

$$\langle f(\phi_k) f(\phi_l) \rangle = \sum_{n,p,m} G_m \lambda_m^{N-k} \langle \psi_m | f(\phi) | \psi_p \rangle \lambda_p^{k-l} \langle \psi_p | f(\phi) | \psi_n \rangle \lambda_n^{l-1} G_n / Z. \tag{9}$$

By choosing $f = S\sin\phi$ and $S\cos\phi$, one can calculate the spin correlation functions $\langle S_{x,k} S_{x,l} \rangle$ and $\langle S_{z,k} S_{z,l} \rangle$. When f is put equal to unity, equation (9) gives expectation values of the x and z components of spins which, when summed over the entire lattice, give the magnetizations in the transverse (x) and longitudinal (z) directions.

To solve the TM equation (5) numerically, we divide the interval $[0, 2\pi]$ into M segments, the corresponding ψ_n being an M-dimensional vector, and equation (5) is transformed into a matrix eigenvalue problem. The case studied by Wang *et al* [4, 5] had $S = 1$, $H_e = 2.0$ kG, $H_a = 0.5$ kG and temperature $T = 0$. Note that the magnetic energy scale is $g\mu_B H_e \simeq 0.3$ K so we take $T = 5 \times 10^{-3}$ K as a low enough temperature to

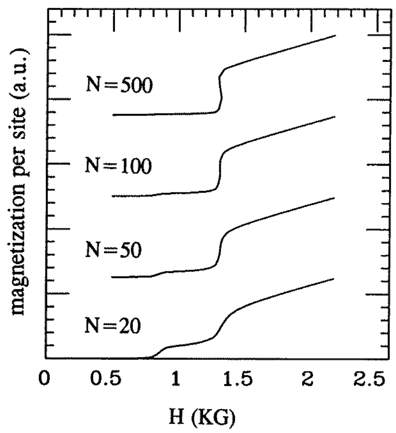

Figure 3. The longitudinal magnetization per site versus the applied magnetic field for $N = 20$, 50, 100 and 500.

be regarded as $T = 0$, although a sharp phase transition occurs only at $T = 0$. It turns out that there are numerous excited states nearly degenerate with respect to the ground state, the excitation energies being less than $T = 5 \times 10^{-3}$ K; thus the latter temperature is more relevant to the experiment than $T = 0$ would be. The numerical accuracy was checked by studying the $M = 400$, 500 and 600 cases, which gave exactly the same results.

Figure 1 shows the longitudinal magnetic susceptibility versus the applied magnetic field for $N = 20$ and 21, reproducing the experimental findings of Wang *et al* [4, 14]. The sharp peak in the $N = 21$ case is attributed to the Néel-to-BSF transition, whereas those in the $N = 20$ case are attributed to the Néel-to-SSF and SSF-to-BSF transitions. Figure 2 shows the spin correlation functions $\langle \cos\phi_l \cos\phi_k \rangle$ and $\langle \sin\phi_l \sin\phi_k \rangle$ versus $k = 1, 2, \ldots, N = 20$ for the three magnetic fields $H = 0.5$, 1.0 and 1.6 kG. The spin correlations on the left- and right-hand sides of figure 2 are clearly characteristic of the Néel state and of the BSF or canted spin state. On the other hand, the peculiar correlation pattern of the middle graph in figure 2, which should be attributed to the SSF state, provides useful information about the SSF state. The local antiferromagnetic order is clear, but the correlation decreases with the spin–spin separation. In comparison with the Ising case, as we will see below, this longitudinal correlation pattern is due to a spatially fluctuating domain wall; that is, the SSF state is composed of energetically degenerate or nearly degenerate single-domain wall states. This can also be seen from the longitudinal magnetization, which shows that about one spin is flopped from down to up during the Néel-to-SSF transition. It is to be noted that the inside spins become affected by the spatial location of the domain wall, leading to a decreasing correlation with distance, but the correlation should again become strong towards the other end because then we know that there is definitely just one domain wall inside and the outermost spin should definitely be up. On the other hand, the small but non-zero transverse correlation indicates that spins are not quite oriented along the z direction but rather are slightly canted. Now a crucial question is that of whether the SSF-to-BSF transition is continuous or discontinuous, namely of first order. Fortunately, we can answer this question without going to the $T = 0$ limit. The size dependence of the longitudinal magnetization suffices for us to answer the question. If the ratio of the magnetizations between the SSF and BSF states did not change with the number of spins N, that would mean a continuous transition, whereas, if the contrary applies, the transition must be discontinuous. We thus calculated the N dependence of the longitudinal magnetization per site, figure 3, from which we conclude that the transition is of first order.

Finally, one can see these points analytically in the limit of strong anisotropy, namely, the Ising case in the limit $T \to 0$ ($q \to \infty$). In this case the spin orientation is limited to $\phi = 0$ and π and the TM equation (5) reduces to a 2×2 matrix eigenvalue problem:

$$\begin{pmatrix} g_{00} & g_{\pi 0} \\ g_{0\pi} & g_{\pi\pi} \end{pmatrix} \begin{pmatrix} \psi_{n0} \\ \psi_{n\pi} \end{pmatrix} = \lambda_n \begin{pmatrix} \psi_{n0} \\ \psi_{n\pi} \end{pmatrix} \tag{10}$$

where

$$g_{00} = \exp\left[-q\left(\frac{1}{2} - \zeta\right)\right] \qquad g_{\pi 0} = g_{0\pi} = \exp\left(\frac{1}{2}q\right) \qquad g_{\pi\pi} = \exp\left[-q\left(\frac{1}{2} + \zeta\right)\right].$$

For N even, the partition function can then be calculated to leading order in the limit $q \to \infty$ as

$$Z = \begin{cases} 2\exp\left(\dfrac{N-1}{2}q\right) & \text{for } \zeta < \frac{1}{2} \\ \dfrac{N}{2}\exp\left(\dfrac{N-1}{2}q + (2\zeta - 1)q\right) & \text{for } \frac{1}{2} < \zeta < 1 \\ \exp\left(N\zeta q - \dfrac{N-1}{2}q\right) & \text{for } \zeta > 1. \end{cases} \tag{11}$$

We find from (11) and energy considerations that, for $\zeta < \frac{1}{2}$, the ground state is doubly degenerate:

$$\downarrow\uparrow\downarrow\uparrow \ldots \downarrow\uparrow \qquad \text{or} \qquad \uparrow\downarrow\uparrow\downarrow \ldots \uparrow\downarrow . \tag{12}$$

For $\frac{1}{2} < \zeta < 1$, one has $N/2$ *degenerate single-domain wall states*. For $N = 6$, for example, these states are

$$\uparrow\uparrow\downarrow\uparrow\downarrow\uparrow \qquad \uparrow\downarrow\uparrow\uparrow\downarrow\uparrow \qquad \uparrow\downarrow\uparrow\downarrow\uparrow\uparrow . \tag{13}$$

Finally, for $\zeta > 1$, the system is ferromagnetic, all the spins pointing in the z direction.

Upon identifying these ground states as the Néel, SSF and BSF states, we again reach the conclusion that *the SSF-to-BSF transition is of first order* in that the magnetization is discontinuous there. Now we turn to the spin correlation functions. Starting with equation (9), after a lengthy but straightforward calculation, the spin correlation function in the Ising case for N even, $T = 0$ and $\frac{1}{2} < \zeta < 1$ is given by

$$\langle \cos\phi_l \cos\phi_k \rangle = \left[1 + (-1)^{k-1}(N - 2k + 1)\right]/N \tag{14}$$

which is plotted in figure 4. We thus find that it is identical to the corresponding spin correlation function, shown in the middle of figure 2, for the planar Heisenberg case. This result demonstrates a robust character of the SSF state, in that it is composed of energetically degenerate or nearly degenerate single-domain wall states.

To conclude, the SSF phase does exist in the even-spin system, in contradiction to the conclusion of Trallori *et al* [10]. We would like to point out that the mapping method cannot handle the energetically degenerate single-domain wall states such as (13). The observation by Trallori *et al* of a chaotic behaviour within certain parameter ranges for ξ and ζ may be related to this degeneracy. On the other hand, the SSF-to-BSF transition is found to be of first order, confirming the Keffer–Chow prediction from perturbation theory. We also observed a finite-size effect in the Néel-to-SSF transition; that is, the critical magnetic field decreases with increasing system size N. Since there is no such effect in the Ising case,

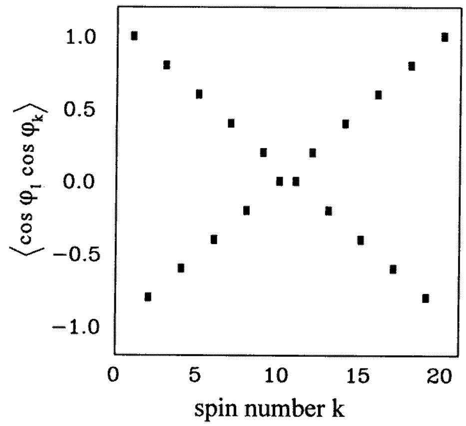

Figure 4. The spin correlation for the $N = 20$ Ising case with the applied magnetic field $H_e/2 < H < H_e$.

H_{SSF} being always $H_{BSF}/2$ irrespective of N, the size dependence is clearly related to the spatial extent of a domain wall.

I would like to express my thanks to Eric Fullerton, who ignited my interest in layered magnetic systems and gave me a timely comment and some of the most recent references. I have also benefited from discussions with Jun Akimitsu and Hiroshi Matsuoka.

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[14] We note that the transverse magnetization is strictly zero for all the cases considered. This is in contradiction to the results of [6], in which a non-zero transverse magnetization was reported. It appears that the non-zero transverse magnetization is due to the authors having overlooked the ground-state degeneracy, which was not properly handled by the self-consistent mean-field method.