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Comparison of the $S=\frac{1}{2}$ xy and antisymmetric exchange chains in an external field

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Green's-function techniques are used to calculate the static correlation functions for the $S=\frac{1}{2}$ xy and antisymmetric exchange chains in a transverse magnetic field. The spin correlation functions are the same for the zero-field case, and the correlation functions are different when there is a symmetry-breaking external field. Possible compounds are proposed where this effect could be observed experimentally.

The compounds $AM\mathrm{Cl}_3\cdot 2\mathrm{H}_2\mathrm{O}$, where $A=\mathrm{Cs,Rb}$ and $M=\mathrm{Co,Fe}$, are examples of chainlike magnetic compounds with a weak interchain exchange interaction and a stronger anisotropic (Ising-like) intrachain exchange interaction. The compound with $A=\mathrm{Rb}$ and $M=\mathrm{Co}$ (RCC) has recently generated a bit of controversy. McElearney and Merchant[1] concluded, on the basis of susceptibility and magnetic specific-heat measurements, that RCC is an example of a linear-chain Dzialoshinsky-Moriya (DM) magnet with no measurable symmetric exchange. This was determined by comparison of experimental results with exact theoretical calculations for the spin-$\frac{1}{2}$ DM chain.

The usual mechanism leading to antisymmetric exchange is spin-orbit coupling which is small compared to the symmetric exchange interaction and can be treated as a perturbation.[2] Indeed, it has been shown that the magnitude of the DM interaction relative to symmetric exchange is $D/J\simeq\Delta g/g$, where g is the g value at the metal-ion site. Therefore, the conclusion in Ref. 1 is very surprising. Recently, Nijhof, van der Ulist, Puertolas, and Gerritsma[3] have published dynamic susceptibility and electron-spin-resonance data for RCC, and their data were analyzed in terms of a spin-$\frac{1}{2}$ xy Hamiltonian. The experimental data obtained to date appear to indicate that RCC might be either a pure DM chain or an xy chain. The existence of a pure DM chain would be very interesting to experimentalists, although it is very unlikely that one would exist. This theoretical work investigates the possibility of experimentally determining if a chain is represented by a DM or xy Hamiltonian.

The spin-$\frac{1}{2}$ xy model[4] and the DM antisymmetric exchange model[5] in one dimension represented by the following interactions have exact solutions.

$$H_{xy}=J\sum_i\left(S_i^xS_{i+1}^x+S_i^yS_{i+1}^y\right)\ ,$$

$$H_{\mathrm{DM}}=\sum_i \mathbf{D}(i,i+1)\cdot\mathbf{S}_i\times\mathbf{S}_{i+1}\ .$$

Since the DM Hamiltonian is related to the symmetric exchange Hamiltonian by a coordinate rotation about the vector $\mathbf{D}$, it is not surprising that these two solutions are similar except for a relative shift of the peak of the k-dependent spin-correlation function.[6] If, however, there is an external field not parallel to the $\mathbf{D}$ vector, the symmetry is lowered, the coordinate rotation cannot be done, and in this case one expects field-dependent differences in the thermodynamic quantities for the two interactions. The corresponding classical problem for both the xy and DM interactions has attracted attention because of the existence of sine-Gordon solitons. Transfer-integral techniques have been used to obtain the static thermodynamic properties for the classical easy-plane symmetric[7] and antisymmetric exchange[8] models, and not surprisingly there is a difference between these two interactions in the presence of an external field. In particular, the DM interaction will modify the soliton contribution to the free energy.

In this paper we compare the static properties of both the spin-$\frac{1}{2}$ xy and DM chains. The spin-$\frac{1}{2}$ case is chosen because exact zero-field correlation functions are used in this calculation. The method used to obtain the exact results transforms the spin operators to Fermi operators and because of the symmetry of the Hamiltonian, complicated phase terms will cancel and the problem is equivalent to the free-fermion problem. However, the lower symmetry introduced by the external field results in phase operators which do not cancel and the transformation to Fermi operators becomes unwieldy. For this reason spin Green's functions are used to obtain the static thermodynamics. We obtain the external field dependence of the static spin-correlation function for both the xy and DM chains. As expected, the correlation function is the same for the two systems in the absence of an external field and differences occur for nonzero field.

First consider the xy interaction

$$H_{xy}=J\sum_i\left(S_i^xS_{i+1}^x+S_i^yS_{i+1}^y\right)-h\sum_i S_i^x\ ,$$

with the field defining the x direction. Since we will consider the zz spin-correlation function, begin with the Green's function[9] $G_{ij}(t)=\langle\langle S_i^z\,|\,S_j^z\rangle\rangle$, where operators to the right of the bar are evaluated at time $t=0$ and operators to the left of the bar are evaluated at time t. The equation of motion for $G_{ij}(t)$ results in a system of equations involving higher-order Green's functions and this set of equations is truncated by two approximations. First, a modified symmetric decoupling is defined which will lower the order of the higher-order functions. Second, only terms up to second order in h/J are retained. The decoupling procedure is illustrated for one of the higher-order Green's functions

$$\langle\langle S^y_{i-1} S^z_i S^y_{i+1} \mid S^z_j \rangle\rangle = f(T) G_{ij} ,$$

where $f(T)$ is a function of temperature that will be determined from the exact zero-field results. A list of the Green's functions and their space- and time-Fourier-transformed equations of motion is

$$\begin{aligned}
EG_k &= iJ(A_k - R_k) - ihF_k , \\
EF_k &= iJ\lambda_k + ihG_k , \\
E\lambda_k &= iJ\{\alpha + 2h(T)[1+\cos(2ka)] - 4g(T)\cos(ka)\} F_k - ihA_k , \\
EA_k &= 2i\psi_1^{xx}[1-\cos(ka)] - iJ\alpha[1-\cos(ka)]G_k + ih\lambda_k , \\
ER_k &= -2i\psi_1^{xx}[1-\cos(ka)] + iJ\alpha[1-\cos(ka)]G_k + ihS_k , \\
ES_k &= -iJ\{2g(T) - [\alpha + 4h(T)]\cos(ka) + 2g(T)\cos(2ka)\} F_k - ihR_k ,
\end{aligned} \tag{1}$$

where

$$G_{ij} = \langle\langle S^z_i \mid S^z_j \rangle\rangle,\ F_{ij} = \langle\langle S^y_i \mid S^z_j \rangle\rangle,\ \lambda_{ij} = \langle\langle S^x_{i-1} S^z_i + S^x_{i+1} S^z_i \mid S^z_j \rangle\rangle ,$$

$$A_{ij} = \langle\langle S^x_{i-1} S^y_i + S^y_i S^x_{i+1} \mid S^z_j \rangle\rangle,\ R_{ij} = \langle\langle S^y_{i-1} S^x_i + S^y_{i+1} S^x_i \mid S^z_j \rangle\rangle,\ S_{ij} = \langle\langle S^z_{i-1} S^x_i + S^x_i S^z_{i+1} \mid S^z_j \rangle\rangle ,$$

the functions $h(T)$ and $g(T)$ are defined by

$$\langle\langle S^x_i S^x_{i+1} S^y_i \mid S^z_j \rangle\rangle = g(T) F_{ij},\ \langle\langle S^z_i S^z_{i+1} S^y_i \mid S^z_j \rangle\rangle = h(T) F_{ij},\ \alpha = 2\langle S^{z2}_i \rangle + 2f(T) ,$$

and $\psi_n^{\alpha\beta} = \langle S^\alpha_i S^\beta_{i+n} \rangle$ is a static correlation function. From the Green's functions and the spectral theorem, the k-dependent spin-correlation functions are obtained. For the zero-field case, the zz correlation function peaks at $ka = \pi$ and the value of the correlation function at the peak can be related to quantities such as the correlation length and the static susceptibility. Therefore, Eqs. (1) are solved for the special case $ka = \pi$ and after a bit of algebra we arrive at the Green's function for the xy chain

$$G_J(E) = \frac{-4J\psi_1^{xx}(2E^2 - 2\Delta J^2 + h^2)}{E^2(E^2 - h^2 - \Delta J^2) - 4\alpha J^2(E^2 - \Delta J^2) - 2h^2 J^2(\Delta + \alpha) - h^2(E^2 - h^2)} , \tag{2}$$

where $\Delta = \frac{1}{2} + 2f + 4g + 4h$.

Next the functions α and Δ are determined for the zero-temperature and zero-field case as follows: when $h = 0$, Eq. (2) has poles at $\pm 2J\sqrt{\alpha}$ and from the spectral theorem we get the spin-correlation function $\psi^{zz}_{k=\pi/a}$. For $T = 0$ this is

$$\psi^{zz}_{k=\pi/a} = -4\pi\psi_1^{xx}\frac{1}{\sqrt{\alpha}} \tag{3}$$

from which α can be obtained since ψ_1^{xx} and $\psi^{zz}_{k=\pi/a}$ are known from Ref. 1. The Green's-function results are in this way forced to be exact for zero field and approximate for nonzero field. The xx correlation function is used to determine Δ in terms of α as follows: the Green's function $M_{ij} = \langle\langle S^x_i \mid S^x_j \rangle\rangle$ can be determined from the second-order equation of motion

$$M_{k=\pi/a}(E) = -\frac{2J(\psi_1^{xx} - \psi_1^{zz})}{E^2 - 2\Delta J^2} \tag{4}$$

for the space- and time-Fourier-transformed M_{ij} evaluated at $k = \pi/a$, and from Eq. (4) the corresponding static correlation function is

$$\psi^{xx}_{k=\pi/a} = -2\pi\frac{\psi_1^{xx} - \psi_1^{zz}}{\sqrt{2\Delta}} . \tag{5}$$

The exact correlation functions in Eqs. (3) and (5) from Ref. 1 yield a number for the ratio $\Delta/\alpha = 0.17$. This ratio is then used to approximate the correlation function at any small field in terms of the parameter α. The temperature dependence of α is determined by forcing the zero-field Green's function results to agree with the exact results at various temperatures, as we have done for zero temperature. We will not go this far; instead, the correlation functions are determined in terms of α, and the field dependence rather than the temperature dependence of thermodynamic quantities will be of interest. Using the Green's function in Eq. (2), the correlation function at $k = \pi/a$ is

$$\psi^{zz}(J) \cong -\frac{4\pi\psi_1^{xx}}{\sqrt{\alpha}}\left[1 - \frac{2.18}{\alpha}\frac{h^2}{J^2}\right] , \tag{6}$$

where only terms second order in h/J are kept.

Next consider the DM interaction

$$H_{\mathrm{DM}}=D\sum_i (S_i^x S_{i+1}^y - S_i^y S_{i+1}^x) - h\sum_i S_i^x \tag{7}$$

with the external field perpendicular to the D vector. The same zz correlation function will be determined at $k=\pi/a$, but the Green's functions and their equations of motion will be different. The list is

$$\begin{aligned}
&EG_{k=\pi/a}=iD(B+C)-ihF \ , \\
&EF=iD\Gamma+ihG \ , \\
&EB=-4i\psi_1^{xy}+2iaDG+ih(\Gamma+\Lambda) \ , \\
&EC=-4i\psi_1^{xy}+2iaDG \ , \\
&E\Gamma=-iDaF-ih(B-L) \ , \\
&E\Lambda=iDaF-ih(B-L) \ , \\
&EL=-ih(\Gamma+\Lambda) \ ,
\end{aligned} \tag{8}$$

where

$$\begin{aligned}
&\Gamma=\langle\langle S_{i-1}^y S_i^z - S_i^z S_{i+1}^y \,|\, S_j^z\rangle\rangle \ , \\
&C=\langle\langle S_{i-1}^x S_i^x - S_i^x S_{i+1}^x \,|\, S_j^z\rangle\rangle \ , \\
&\Lambda=\langle\langle S_{i-1}^z S_i^y - S_i^y S_{i+1}^z \,|\, S_j^z\rangle\rangle \ , \\
&L=\langle\langle S_{i-1}^z S_i^z - S_i^z S_{i+1}^z \,|\, S_j^z\rangle\rangle \ , \\
&B=\langle\langle S_{i-1}^y S_i^y - S_i^y S_{i+1}^y \,|\, S_j^z\rangle\rangle \ ,
\end{aligned}$$

and F and G are defined in Eqs. (1). $G(E)$ is determined by solving Eqs. (8):

$$G_D(E)=-\frac{4D\psi_1^{xy}(2E^2-2aD^2-5h^2)}{(E^2-4aD^2-4h^2)(E^2-aD^2-h^2)} \ . \tag{9}$$

Notice that if $h=0$ and $J=D$ Eq. (9) will be the same as Eq. (2) since ψ_1^{xy} for the pure DM chain is the same as ψ_i^{xx} for the xy chain. This is expected because both systems are related by a coordinate rotation. However, for nonzero h the Green's functions for the two chains are different. Using Eq. (9), the zz correlation function for the DM chain is

$$\psi^{zz}(D)\cong-\frac{4\pi\psi_1^{xy}}{\sqrt{a}}\left[1-\frac{1}{2a}\frac{h^2}{D^2}\right] \tag{10}$$

to second order in h/D. Comparison of Eqs. (6) and (10) shows that they are the same if $h=0$. However, for nonzero h the xy chain correlation function has a stronger field dependence than the DM chain.

The correlation functions obtained above are exact if $h=0$ and they should be a good approximation for small nonzero h. Possibly one could distinguish between the two models by susceptibility versus field measurements. It would perhaps be useful to compare RCC data with data obtained from the same measurements of other known spin-$\frac{1}{2}$ xy magnetic chains. Two possible compounds that we are aware of are $\mathrm{Co(N_2H_5)_2(So_4)_2}$, which was determined to be an xy chain from zero-field specific-heat measurements[10] and $\mathrm{Cs_2CoCl_4}$, which was again studied by specific-heat measurements.[11,12]

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