Metamagnetism in one dimensional systems with edge sharing CuO polyhedra

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We study a Heisenberg chain with nearest-neighbor (NN) $J_1$ and next-NN $J_2$ exchange interactions with anisotropies $\Delta_1$ and $\Delta_2$ respectively. We investigate by analytical and numerical methods the region of parameters for which there is a jump in the magnetization $M$ as a function of magnetic field $B$. Some materials with edge sharing CuO polyhedra are candidates to show an abrupt change in $M(B)$. The magnetization as a function of applied magnetic field in several materials shows a discontinuity or very rapid increase at a certain field $B_c$. Gerhardt et al. have shown that for certain parameters, a magnetization jump is also present in the spin-1/2 XXZ chain with NN and next-NN exchange coupling (keeping $\Delta_1 = \Delta_2 = \Delta$):

$$H = \sum_i [J_1(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y + \Delta_1 S_i^z S_{i+1}^z)$$
$$+ \sum_i J_2(S_i^x S_{i+2}^x + S_i^y S_{i+2}^y + \Delta_2 S_i^z S_{i+2}^z)] - B \sum_i S_i^z. \quad (1)$$

For a metamagnetic transition to occur at very low temperatures, the zero-field ground-state energy per site $E$ as a function of the magnetization $M = \sum_i S_i^z/L$ ($L$ is the number of sites), should satisfy two conditions: I) $\partial^2 E/\partial M^2 < 0$ in a finite interval of values of $M$. Then one can draw a straight line $E'(M)$ which is tangent to $E(M)$ in at least two points (the Maxwell construction, see Fig. 1(a)) in such a way that $E'(M) > E(M)$ for $M_1 \leq M \leq M_2$. II) $E(M_2) > E(M_1)$. If these two conditions are satisfied, jumps from $M_1$ to $M_2$ at the critical field $B_c = [E(M_2) - E(M_1)]/(M_2 - M_1)$.

From the general behavior of $E(M)$, Gerhardt et al. have found that when metamagnetism exists, $M_2 = 1/2$ and the condition II ceases to be satisfied when $M_1 = 0$. More precisely, from their finite-size results for $E(M, \alpha, \Delta)$, with $\alpha = J_2/J_1$, they obtained a critical value of $\Delta$ ($\Delta_f(\alpha)$) from the equation $E(0, \alpha, \Delta_f) = E(1/2, \alpha, \Delta_f)$. For $\Delta < \Delta_f$ the system is ferromagnetic at $B = 0$. Another critical value $\Delta_f(\alpha)$ was obtained from the condition $\partial^2 E/\partial M^2|_{M=1/2} = 0$. For $\Delta > \Delta_f$ the curvature $\partial^2 E/\partial M^2$ is positive for all $M$. The discretized $\partial^2 E/\partial M^2|_{M=1/2} = 0$ has some finite-size effects.

From the numerical solution of the problem of two spin excitations on the ferromagnetic state for $L \rightarrow \infty$, more accurate values of $\Delta_f(\alpha)$ were obtained recently for $\alpha \leq 1/2$. In the region of the $(\alpha, \Delta)$ plane where $\Delta_f(\alpha) < \Delta < \Delta_f(\alpha)$ a metamagnetic transition occurs in the model.

We have studied the two-magnon problem for generic values of $\Delta_1$ and $\Delta_2$, and found analytical results for the condition $\partial^2 E/\partial M^2|_{M=1/2} = 0$ if $\alpha \leq 0.75$. When $\Delta_1 = \Delta_2 = \Delta$, in the region $\alpha \leq 1/4$, the function $\Delta_f(\alpha)$ can be accurately approximated by:

$$\Delta_f = -1 + 2 \sum_{i=1}^4 \alpha^i + 6 \alpha^5 + O(\alpha^6), \text{ if } \alpha \leq 0.2$$

Finally in the region $1/2 \leq \alpha \leq 0.75$, $\Delta_f(\alpha)$ is very flat. Near $\alpha = 1/2$ it can be approximated as $\Delta_f = -1 + 0.309(x - 1/2)^2$. These results show that metamagnetism is not possible if $\Delta > (-5 + \sqrt{17})/4 = -0.219$. Unfortunately, such a large anisotropy of $J_2$ seems unrealistic. Instead, $\Delta_1 = -1$ corresponds to isotropic ferromagnetic $J_1$, since a rotation of every second spin in $\pi$ around the $z$ axis changes the sign of the $x$ and $y$ components of $J_1$.

The main purpose of this work is to extend the previous results to negative $\Delta_1$ and positive $\Delta_2$. Since it is expected that the parameters for several copper oxides containing edge sharing Cu-O chains lie near the isotropic limit $\Delta_1 = -1, \Delta_2 = 1$, we consider this limit in what follows. From numerical diagonalization of 20 sites, we obtain that spontaneous ferromagnetism does not take place for $\alpha > 1/4$. If in addition $\alpha \leq 0.7$, there is a bound state in the two-magnon problem at wave vector $q_2 = 2q_1$, where $q_1 = \pm \arccos[-1/(4\alpha)]$ are the wave vectors of the one-magnon states of lowest energy. For $\alpha > 0.7$, there might be a two-magnon bound state with $q_2 \neq 2q_1$, but we have not studied this alternative because it seems not possible to solve the problem analytically for large $\alpha$. Thus, we expect a jump in $M(B)$ for $1/4 < \alpha \leq \alpha_c$ with $\alpha_c \geq 0.7$.

In Fig. 1(a) we show $E(M)$ for a chain of $L = 20$ sites with periodic boundary conditions for $\alpha = 0.425$, chosen in such a way that $q_1 = \pm 7\pi/10$ are allowed wave vectors of the finite chain. For other values of $\alpha$, one
might obtain a numerical negative $\partial^2 E/\partial M^2 |_{M=1/2}$ because of frustration effects which increase $E(M - 1/L)$. In spite of this precaution, the results show a significant even-odd effect: the energies for odd (even) total spin $S = |M|L$ seem to be shifted to higher (lower) energies. If this effect persists in the thermodynamic limit (keeping $L$ even) states with odd $S$ become irrelevant (because they do not minimize $E - MB$ for any $B$) and a bound state in the two-magnon problem does not necessarily imply $\partial^2 E/\partial M^2 |_{M=1/2} < 0$. From $E(S/L)$ for the three highest even $S$ with $L = 28$, minimized with respect to the optimum twisted boundary conditions to allow for incommensurate wave vectors \[7\], we obtain a very small curvature which is negative for $\alpha < \alpha_c = 0.359$ but positive for $\alpha > \alpha_c$. If $\alpha_c$ remains finite in the thermodynamic limit, $M(B)$ would increase abruptly for $\alpha > \alpha_c$, but without showing a true jump. While this difference is hard to distinguish experimentally, it would be of interest to calculate $\partial^2 E/\partial M^2 |_{M=1/2}$ using larger clusters.

To obtain a continuous curve $E(M)$ from which $M(B)$ can be derived, we have fitted the eleven numerical values represented in Fig. 1(a) by a polynomial of even powers of $M$ up to $M^{10}$. This function satisfies the physical condition $E(M) = E(-M)$ and has six fitting parameters (nearly half of the number of points to be fitted, to average the even-odd effect). The resulting $B = \partial E/\partial M$ is represented in Fig. 1(b). At a critical field $B_c = 0.192 J_1$, the magnetization jumps from $M_1 = 0.347$ to $M_2 = 1/2$. While the numerical values of $B_c$ and particularly $M_1$ depend on the particular fitting procedure used and the size of the system, the general shape of $M(B)$ is robust: around $B/J_1 = 0.19 \pm 0.01$ there is a sudden increase of $M$ from $\sim 0.25$ to $1/2$. While the shape of $M(B)$ does not depend very much on $\alpha > 1/2$, $B_c$ increases strongly with $\alpha$.

To conclude, while the existence of a real jump in $M(B)$ requires a study of larger clusters, we have shown that the magnetization of the model for parameters appropriate to edge-sharing Cu-O chains has a sudden increase at a magnetic field $B_c$. Using parameters calculated for La$_6$Ca$_8$Cu$_{24}$O$_{41}$ \[6\] and assuming a gyromagnetic factor $g = 2$ we obtain $B_c \approx 14$ Tesla.

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FIG. 1. Energy per site as a function of total spin per site for a chain of 20 sites (solid circles). The full line is a fit (see text). Dashed line and diamonds correspond to the Maxwell construction. (b) Magnetic field as a function of the magnetization. Parameters are $J_1 = 1$, $J_2 = 0.425$, $\Delta_1 = -1$ and $\Delta_2 = 1$.

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