Vol. 97 (2000) *ACTA PHYSICA POLONICA A* No. 3

Proceedings of the European Conference "Physics of Magnetism '99", Poznań 1999

LIEB–MATTIS MODEL FOR TWO INTERPENETRATING ANTIFERROMAGNETS

D. Schmalfuss, S.E. Krüger and J. Richter

Institut für Theoretische Physik, Universität Magdeburg
PF 4120, 39016 Magdeburg, Germany

We investigate a frustrated Lieb–Mattis-like spin-1/2 model that is a reference model for the corresponding square-lattice Heisenberg model describing the unusual magnetic properties of $Ba_2Cu_3O_4Cl_2$. Due to frustration we obtain a rich magnetic phase diagram. We find two critical temperatures in accordance with recent experiments on $Ba_2Cu_3O_4Cl_2$.

PACS numbers: 75.10.Jm

1. Introduction

In contrast to La_2CuO_4 the layered $Ba_2Cu_3O_4Cl_2$ cuprate is built by copper oxide planes which contain two kinds of copper sites, Cu(A) and Cu(B). The Cu(A) atoms form a square lattice corresponding to the CuO_4 plane in La_2CuO_4. The additional Cu(B) atoms are centered in every second Cu(A) plaquette. Together they form two interpenetrating subsystems of spins 1/2. As in La_2CuO_4 the 180° superexchange J_{AA} between neighbouring Cu(A) spins is strongly antiferromagnetic. On the other hand, the Cu(B) spins interact much weaker with each other, i.e. $J_{BB} \ll J_{AA}$ [1]. The tight binding analysis of the band structure of $Ba_2Cu_3O_4Cl_2$ yields a frustrating ferromagnetic coupling J_{AB} between the subsystems comparable with J_{BB} [1]. Though we have a finite coupling J_{AB} between the subsystems in recent experiments, two critical transition temperatures have been observed which differ in one order of magnitude, $T_A \approx 330$ K and $T_B \approx 40$ K [2] and can be related to the subsystems A and B, respectively. Additionally, there is a weak ferromagnetic moment found for temperatures $T_B < T < T_A$ which might be attributed to an additional pseudodipolar coupling between the two subsystems [3]. In our paper we study a simplified version of the square-lattice Heisenberg model for $Ba_2Cu_3O_4Cl_2$ studied in [4, 5] which allows us to calculate the phase diagram exactly following [6].

2. Model

We consider a system of $3N$ spins 1/2 consisting of two antiferromagnetic subsystems A and B. The number of spins in subsystem A is $N_A = 2N$ and the number of spins in B is $N_B = N$. Both subsystems A and B consist of two sublattices A_1, A_2, and B_1 and B_2 respectively. The number of spins in $A_{1(2)}$ is N and in $B_{1(2)}$ is $N/2$. The corresponding three-parameter model is

$$H = \frac{4J_{AA}}{3N} \sum_{i\in A_1, j\in A_2} \boldsymbol{S}_i \boldsymbol{S}_j + \frac{8J_{BB}}{3N} \sum_{i\in B_1, j\in B_2} \boldsymbol{S}_i \boldsymbol{S}_j + \frac{2J_{AB}}{3N} \sum_{i\in A, j\in B} \boldsymbol{S}_i \boldsymbol{S}_j . \quad (1)$$

This kind of long-range coupling was introduced by Lieb and Mattis [7] and makes the model in the thermodynamic limit molecular field like, however, with the full rotational invariance. In correspondence to the situation in $Ba_2Cu_3O_4Cl_2$ we choose antiferromagnetic $J_{AA} > J_{BB} > 0$ and ferromagnetic $J_{AB} < 0$. Introducing the total spin operators $\boldsymbol{S}^2_{A_{1(2)}}, \boldsymbol{S}^2_{B_{1(2)}}, \boldsymbol{S}^2_{A(B)} = (\boldsymbol{S}_{A(B)_1} + \boldsymbol{S}_{A(B)_2})^2$ and $\boldsymbol{S}^2 = (\boldsymbol{S}_A + \boldsymbol{S}_B)^2$ with $\boldsymbol{S}_{A_{1(2)}} = \sum_{i\in A_{1(2)}} \boldsymbol{S}_i$, $\boldsymbol{S}_{B_{1(2)}} = \sum_{i\in B_{1(2)}} \boldsymbol{S}_i$ we find after some algebra the eigenvalues of H

$$\begin{aligned} E = {} & 2J_{AA}\left[S_A\left(S_A+1\right) - S_{A_1}\left(S_{A_1}+1\right) - S_{A_2}\left(S_{A_2}+1\right)\right]/3N \\ & +4J_{BB}\left[S_B\left(S_B+1\right) - S_{B_1}\left(S_{B_1}+1\right) - S_{B_2}\left(S_{B_2}+1\right)\right]/3N \\ & -\left|J_{AB}\right|\left[S\left(S+1\right) - S_A\left(S_A+1\right) - S_B\left(S_B+1\right)\right]/3N, \end{aligned} \quad (2)$$

where the competition between the terms proportional to J_{AA}, J_{BB} and the term proportional to $|J_{AB}|$ becomes obvious. The first two terms tend to minimize $S_{A(B)}$, the last one tends to maximize these quantum numbers. To consider the thermodynamic limit $N \to \infty$ we introduce normalized quantum numbers $\in [0,1]$ which may serve as order parameters

$$a_{1(2)} = \frac{2S_{A_{1(2)}}}{N}, \quad b_{1(2)} = \frac{4S_{B_{1(2)}}}{N}, \quad a = \frac{S_A}{N}, \quad b = \frac{2S_B}{N}. \quad (3)$$

3. Ground state

Minimizing (2) we set $S_{A_{1(2)}} = N/2$, $S_{B_{1(2)}} = N/4$ (i.e. $a_{1(2)} = b_{1(2)} = 1$) and $S = S_A + S_B$. For dominating inter-subsystem coupling $|J_{AB}|$ we obtain a state with fully polarized subsystems, i.e. $a = b = 1$. More interesting is the limit of small $|J_{AB}|$ which corresponds to the situation in $Ba_2Cu_3O_4Cl_2$. Then we have antiferromagnetic singlet states in the subsystems, i.e. $a = b = 0$. For intermediate $|J_{AB}|$ we have a state where the subsystem A is partially polarized (canted spin structure) and the subsystem B is fully polarized, $a = |J_{AB}|/4J_{AA}$, $b = 1$. The antiferromagnetic state and the state with the canted spin structure are separated from each other by a first-order transition line given by $8J_{AA}J_{BB} - |J_{AB}|^2 = 0$. On the other hand, the canted state is separated from the fully polarized state by a second-order transition.

4. Thermodynamics

In the thermodynamic limit the saddle point approximation becomes exact. Then the partition function is determined by its largest term. Due to symmetry we have $a_1 = a_2$ and $b_1 = b_2 \in [0,\ 1]$. Furthermore, the main contribution to the partition function is given for $S = S_A + S_B$. The remaining quantum numbers have to be chosen according a $(b) \in [0, a_1\ (b_1)]$. Taking this into account the remaining task is to find the minimum of the free energy given by

$$
\begin{aligned}
F = & -Nk_BT\ln\frac{N^3}{2} + Nk_BT(1-a_1)\ln\left[\frac{N}{2}(1-a_1)\right] \\
& + Nk_BT(1+a_1)\ln\left[\frac{N}{2}(1+a_1)\right] + \frac{Nk_BT}{2}(1-b_1)\ln\left[\frac{N}{4}(1-b_1)\right] \\
& + \frac{Nk_BT}{2}(1+b_1)\ln\left[\frac{N}{4}(1+b_1)\right] + \frac{J_{AA}N}{3}\left(2a^2-a_1^2\right) \\
& + \frac{J_{BB}N}{6}\left(2b^2-b_1^2\right) - \frac{|J_{AB}|\,N}{3}ab. \qquad (4)
\end{aligned}
$$

As a result, we get the minimizing quantum numbers as functions of the couplings and the temperature. In Fig. 1 the phase diagram is presented. We focus our interest on the parameter region where the intra-subsystem couplings dominate the inter-subsystem coupling, namely $8J_{AA}J_{BB} - |J_{AB}|^2 > 0$. In this region we have $a_1 = \tanh(J_{AA}a_1/3k_BT)$ and $b_1 = \tanh(J_{BB}b_1/3k_BT)$ with $a = b = 0$. According to the experimental findings we put $J_{AA} = 10J_{BB}$. Then three phases occur. For temperatures k_BT smaller than $J_{BB}/3$ both subsystems are antiferromagnetically ordered. At $k_BT_B = J_{BB}/3$ a second-order transition takes place and the weaker coupled subsystem B becomes paramagnetic while subsystem A remains antiferromagnetic.

There is another second-order transition at $k_BT_A = J_{AA}/3$ where the magnetic ordering in A disappears and both subsystems are paramagnetic. Obviously, the spin system exhibits two critical temperatures corresponding to the behaviour of $Ba_2Cu_3O_4Cl_2$. Due to $J_{AA} = 10J_{BB}$ the corresponding critical

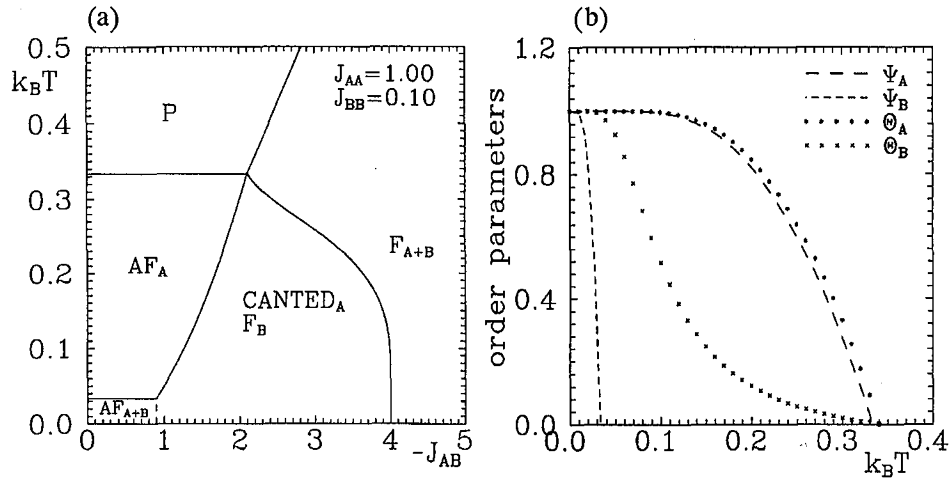

Fig. 1. (a) Phase diagram of two coupled antiferromagnets as a function of temperature k_BT and frustrating coupling. (b) Antiferromagnetic order parameters $\Psi_{A(B)} = \langle(S_{A(B)_1} - S_{A(B)_2})^2\rangle/N^2_{A(B)}$ $(J_{AA} = 10J_{BB} = 1)$ compared with the ferromagnetic order parameters $\Theta_{A(B)} = \langle(S_{A(B)_1} + S_{A(B)_2})^2\rangle/N^2_{A(B)}$ of the corresponding ferromagnetic model $(J_{AA} = 10J_{BB} = -1)$ versus temperature k_BT and $J_{AB} = -0.2$.

temperatures differ in one order of magnitude. According to recent calculations based on the LDA+U scheme [8] even a parameter relation $J_{AA} \gg |J_{AB}| \gg J_{BB}$ may be realized in $Ba_2Cu_3O_4Cl_2$. Within the considered model (1) for very small $J_{BB} \to 0$ and low temperatures subsystem A is in the canted phase and B is fully polarized. We emphasize that in this case still we have two critical temperatures, however the first second-order transition is between $(CANTED_A, F_B)$ and AF_A (see Fig. 1). The starting point of this second-order transition line between $(CANTED_A, F_B)$ and AF_A is shifted close to the origin. This line is given by $k_B T = |J_{AB}|^2/12J_{AA} - J_{BB}/3$.

Next we consider the order parameters. In addition to the model with antiferromagnetic J_{AA} and J_{BB} we consider a corresponding model with ferromagnetic couplings, i.e. we change the signs of J_{AA} and J_{BB}. This ferromagnetic model exhibits no frustration. Comparing the order parameters of the antiferromagnetic model $\Psi_{A(B)}$ and the ferromagnetic model $\Theta_{A(B)}$ it becomes evident that the existence of two critical temperatures is connected with frustration inherent in the antiferromagnetic model but not in the ferromagnetic model.

5. Conclusions

Motivated by the real structure and the unusual magnetic behaviour of $Ba_2Cu_3O_4Cl_2$ we have studied a Heisenberg spin system consisting of two interpenetrating antiferromagnets. Assuming Lieb–Mattis-like long-ranged interactions the Hamiltonian is exactly solvable. The competition between the three couplings and thermal fluctuations leads to an interesting phase diagram. In agreement with experimental data we have found two critical temperatures indicating magnetic phase transitions of second order. A necessary condition for that is the frustrating nature of the couplings. Finally, we notice that the ferromagnetic moment is not reproduced. As argued in [3] one needs pseudodipolar coupling for that.

Acknowledgment

The authors are indebted to R. Hayn for valuable discussions. This work was supported by the DFG (project Ri 615/7-1).

References

[1] H. Rosner, R. Hayn, J. Schulenburg, *Phys. Rev. B* **57**, 13660 (1998).

[2] K. Yamada, N. Suzuki, J. Akimitsu, *Physica B* **213-214**, 191 (1995).

[3] F.C. Chou, A. Aharony, R.J. Birgeneau, O. Entin-Wohlman, M. Greven, A.B. Harris, M.A. Kastner, Y.J. Kim, D.S. Kleinberg, Y.S. Lee, Q. Zhu, *Phys. Rev. Lett.* **78**, 535 (1997).

[4] J. Richter, A. Voigt, J. Schulenburg, N.B. Ivanov, R. Hayn, *J. Magn. Magn. Mater.* **177-181**, 737 (1998).

[5] J. Richter, D. Schmalfuss, S.E. Krüger, *Physica B* **259-261**, 911 (1999).

[6] J. Richter, S.E. Krüger, A. Voigt, C. Gros, *Europhys. Lett.* **28**, 363 (1994).

[7] E. Lieb, D. Mattis, *J. Math. Phys.* **3**, 749 (1962).

[8] A. Jaresko, A. Perlov, S. Khalilov, R. Hayn, private communication.