Nonclassical effects and off-diagonal couplings in a model for $FeBr_2$

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Using Monte Carlo techniques, we show that the recently experimentally observed transitionlike phenomena in the transverse spin ordering close to the anomalies in the antiferromagnetic phase of $FeBr_2$ [O. Petracic *et al.*, Phys. Rev. B **57**, R11 051 (1998)] may result from the quantum nature of the $S=1$ spins and an off-diagonal exchange between axial and planar spin components. [S0163-1829(99)12913-8]

Recently, Petracic *et al.*[1] presented experimental evidence for a weakly first-order phase transition in the antiferromagnetic phase of $FeBr_2$, when applying a magnetic field under nonvanishing tilting angle with respect to the c axis of the hexagonal crystal, extending previous work with an axial field.[2,3] They argue that the measured jumps in the magnetization parallel and perpendicular to the field are related to the surprisingly sharp peaks near the anomalies in the specific heat which had been measured before in axial magnetic fields.[4]

They suggest that quantum effects in a $S=1$ anisotropic Heisenberg magnet and off-diagonal exchange between axial and planar spin components allowed by the crystal symmetry[5] may be crucial in explaining the transitionlike phenomena.

Indeed, our analyses on an appropriate model support their suggestion. In particular, we considered the following $S=1$ Hamiltonian, being believed to describe quite realistically $FeBr_2$ (Refs. 5–10)

$$\mathcal{H}=-\sum J_{ij}(\alpha^{-1}S_i^zS_j^z+S_i^xS_j^x+S_i^yS_j^y)-\sum D(S_i^z)^2$$

$$-\sum (H_xS_i^x+H_zS_i^z)+\mathcal{H}_{\rm od}(J_{xz}). \qquad (1)$$

The first term describes anisotropic, $\alpha<1$, exchange interactions, J_{ij}, between nearest (J_1), next-nearest (J_2), and third (J_3) neighbor spins in the triangular layers perpendicular to the c axis and between spins in adjacent layers connected by equivalent exchange paths (J').[6–10] The second term describes the single-ion anisotropy of strength D. The third term refers to the tilted field, having components both perpendicular (H_z) and parallel (H_x) to the triangular planes. The last term $\mathcal{H}_{\rm od}(J_{xz})$ describes the off-diagonal bilinear couplings between axial (S_z) and planar (S_x and S_y) components of neighboring spins in the layers, with the strength J_{xz}.[5] For instance, for neighboring spins with the same value of y, one has

$$\mathcal{H}'_{\rm od}=-J_{xz}(S_i^zS_j^x+S_i^xS_j^z). \qquad (2)$$

The corresponding terms for other pairs of nearest-neighbor spins follow from appropriate translations and rotations.[5]

We performed Monte Carlo simulations on that Hamiltonian in the quasiclassical approximation, i.e., fixing the spin length to be $\sqrt{2}$, with $S_z=\pm 1,0$. The exchange constants J_{ij} and the anisotropy factor α were chosen as before[10] (based on spin-wave analyses[6,8]), with the tenfold-coordinated antiferromagnetic couplings J' between adjacent triangular layers, with nearest-neighbor, $J_1=-16.75J'$, and competing third-neighbor, $J_3=-0.29J_1$, interactions in the planes perpendicular to the c axis, and with an Ising-type anisotropy factor $\alpha=0.78$. We then varied the degree of the Ising-like anisotropy due to the single-ion term D, the axial H_z and planar H_x components of the external magnetic field, as well as the strength J_{xz} of the off-diagonal exchange between the axial and planar spin components.

In an axial field, $H_x=0$ and H_z being close to the critical field, $H_{z0}=-20J'$, at zero temperature, the peculiar shape of the temperature-dependent specific heat $C(T)$ found experimentally[4,1] can, indeed, be reproduced by tuning the remaining parameters of the model, as shown in Fig. 1. In particular, the maximum at the lower temperature shows a pronounced shoulder, associated with the anomaly in the z components of the spins[2–4] originating from the relatively weak, albeit dominating, intralayer couplings as compared to the interlayer couplings as well as from the high interlayer coordination number.[9–11] The superimposed peak shifts towards the critical temperature T_c and gets sharper as J_{xz} increases. That peak, at T_{xy}, reflects the disordering of the

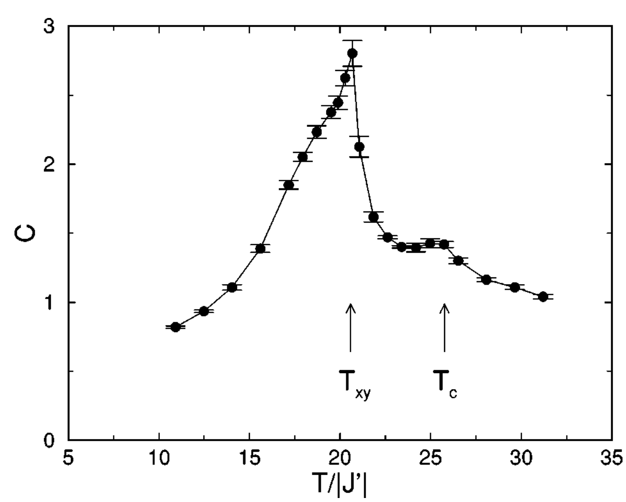

FIG. 1. Temperature-dependent specific heat C as obtained from Monte Carlo simulations of Hamiltonian (1) with $J_{xz}=16.2J'$, $D=-8.1J'$, and $H_z=-18J'(=0.9H_{z0})$, for systems of 20^3 spins.

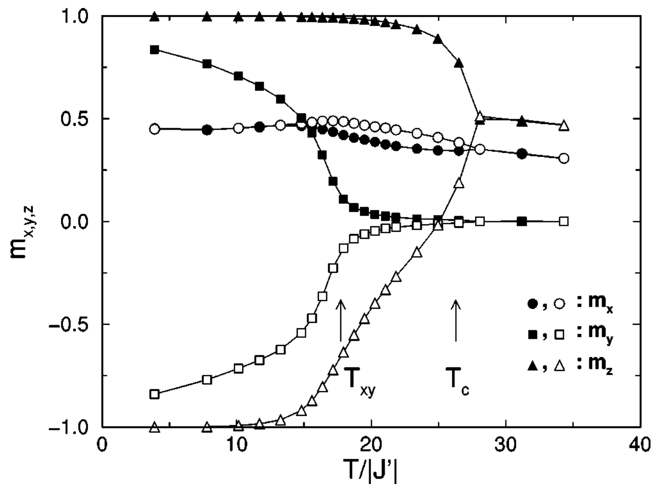

FIG. 2. Temperature dependence of the x,y, and z components of the magnetization per layer in odd (full symbols) and even (open symbols) planes, using the same parameters as in Fig. 1, with an additional planar component of the field, $H_x = 0.75H_z$, simulating systems of 30^3 spins.

antiferromagnetic low-temperature state of the xy or planar components of the spins, being aligned ferromagnetically in each in the triangular planes, with an antiferromagnetic arrangement between subsequent layers. The corresponding disordering of the z or axial components occurs at T_c. The two processes are largely decoupled due to the quantization of the spins. Note that the anomaly and the transitionlike peak are not intimately related, and their positions can be moved relatively to each other by changing, for instance, the off-diagonal exchange J_{xz} and the degree of Ising-like anisotropy D.

Applying a nonaxial field, $H_x > 0$ and keeping H_z fixed, the xy components of the spins order in the antiferromagnetic phase, at low temperatures, in a spin-flop state, in which the x components of the magnetization per layer are almost identical in all layers, but the y components of the magnetization in adjacent planes have opposite signs. Increasing the temperature, the y component of the magnetization per layer changes rapidly at the temperature T_{xy}, well below the transition temperature T_c of the antiferromagnetic phase, see the Monte Carlo data depicted in Fig. 2. The change in the y component may be associated with a first-order transition, characterized by a jump in the magnetization per layer, when the off-diagonal exchange J_{xz} exceeds a critical value (at about $J_{xz} = 18J'$). Otherwise, one finds a drastic, but presumably analytic change. Experimentally,[1] one is at the border of these two scenarios, observing a weakly first-order transition at T_{xy}. Note that the experimental findings[1] are compatible with the spin-flop state.

In summary, our simulational data show that inclusion of the off-diagonal couplings in the spin components allowed by the crystal symmetry and of the quantum nature of the $S = 1$ spins in the Hamiltonian for $FeBr_2$ is, indeed, sufficient to describe the transitionlike features in the specific heat and the magnetization recently observed experimentally. The features are related to a rapid change or transition in the planar spin components, which, in turn, seems to occur rather fortuitously closely to the anomaly of the axial spin components in the antiferromagnetic phase of $FeBr_2$ discussed extensively before.

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[1] O. Petracic, Ch. Binek, W. Kleemann, U. Neuhausen, and H. Lueken, Phys. Rev. B **57**, R11 051 (1998).
[2] M.M.P. de Azevedo, Ch. Binek, J. Kushauer, W. Kleemann, and D. Bertrand, J. Magn. Magn. Mater. **140-144**, 1557 (1995).
[3] K. Katsumata, H. Aruga Katori, S.M. Shapiro, and G. Shirane, Phys. Rev. B **55**, 11 466 (1997).
[4] H. Aruga Katori, K. Katsumata, and M. Katori, Phys. Rev. B **54**, R9620 (1996).
[5] D. Mukamel, Phys. Rev. Lett. **46**, 845 (1981).
[6] W.B. Yelon and C. Vettier, J. Phys. C **8**, 2760 (1975).
[7] L. Hernández, H.T. Diep, and D. Bertrand, Europhys. Lett. **21**, 711 (1993).
[8] S. Pouget, Ph.D. thesis, I.N.S.A. Toulouse, France, 1993.
[9] W. Selke and S. Dasgupta, J. Magn. Magn. Mater. **147**, L245 (1995); W. Selke, Z. Phys. B **101**, 145 (1996).
[10] M. Pleimling and W. Selke, Phys. Rev. B **56**, 8855 (1997).
[11] K. Held, M. Ulmke, N. Blümer, and D. Vollhardt, Phys. Rev. B **56**, 14 469 (1997).