Fig. 1a, PRB, Acharyya, Nowak, Usadel
Fig. 1b, PRB, Acharyya, Nowak, Usadel
Fig. 2a, PRB, Acharyya, Nowak, Usadel
Fig. 2b, PRB, Acharyya, Nowak, Usadel
Fig. 3a, PRB, Acharyya, Nowak, Usadel
Fig. 3b, PRB, Acharyya, Nowak, Usadel
Fig. 3c, PRB, Acharyya, Nowak, Usadel
Fig. 4, PRB, Acharyya, Nowak, Usadel
Fig. 5, PRB, Acharyya, Nowak, Usadel
Transverse ordering of an antiferromagnet in a field with oblique angle to the easy axis

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Abstract

Motivated by the recent experimental observations [Phys. Rev. B 57 R11051 (1998)] of transverse spin ordering in FeBr₂ induced by a magnetic field with oblique angle to the easy axis of the system, we performed extensive Monte Carlo simulations of a classical anisotropic Heisenberg model. We have calculated the specific heat and the parallel and perpendicular components of the magnetisation as well as the antiferromagnetic order parameter and studied these quantities as a function of temperature. A tilted spin-flop phase is obtained for certain parameter values. Many of the effects occurring in connection with this phase agree qualitatively well with the experimental facts.

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I. INTRODUCTION

Among many other magnetic materials, so called metamagnets show interesting phase transitions induced by an external magnetic field. Especially, the multicritical behavior in the field-temperature-plane of the phase diagram is subject of immense interest. In recent experiments the metamagnet FeBr₂ was studied. Cooling the sample in zero field
the well-known transition from the paramagnetic to the antiferromagnetic state leading to a divergence of the specific heat at the respective Néel temperature was observed. For finite applied magnetic fields (along the crystallographic c-axis) the magnetic part of the specific heat of this system showed a peculiar shape. As the field increases, the specific heat develops an anomalous peak (the structure containing a broad noncritical anomaly at $H_-(T)$, and a sharp peak at $H_1(T)$, where $H_- < H_1 < H_c$) at a temperature lower than the corresponding critical temperature. This anomalous peak may indicate an additional second phase transition besides the usual transition from the paramagnetic (saturated) to the antiferromagnetic phase. To identify the nature of a possible third phase is the main goal of both, theoretical and experimental work.

For a simple and qualitative understanding, Monte Carlo simulations have been performed for an Ising model. Considering the hexagonal lattice structure of FeBr$_2$, ferromagnetic intra-planar interaction and antiferromagnetic inter-planar interactions, it has been shown that the anomalous peak of the specific heat (at $H_-(T)$) can be reproduced with interaction parameters obtained from spin wave analysis and neutron scattering experiments. The "phase boundary" obtained from Monte Carlo simulations agreed qualitatively well with the experimental one. It has been conjectured that the anomaly line is the "border" between antiferromagnetic at low temperature and a "mixed phase", where it was speculated that due to the positive axial field small clusters of positive spins in the negative sea may form a stable phase. The detailed characterization of this "intermediate phase" is missing in the literature.

However, the recent experimental observations of a transverse spin ordering associated with a weak first order transition (at $H_1(T)$) and a sharp peak of the specific heat cannot be explained by a simple Ising model. A model with transverse spin components is necessary. A disorder-order transition of the $m_s = 0$ spin components probably due to off-diagonal exchange was conjectured. Motivated by this conjecture, the so-called semiclassical Heisenberg model including off-diagonal exchange interactions has been studied recently by Monte Carlo simulation. In this model, the axial component of the spin vector...
is quantized (it can take values -1, 0 and +1) while the planar component is a classical vector which can rotate continuously in the transverse plane. One can consider this model to be a (de)coupled combination of a $S = 1$ Ising model with a kind of classical XY- model and consequently, with Ising-like anisotropy, one observes always two sharp peaks in the specific heat even at zero axial field (surprisingly also with ferromagnetic interaction and no off-diagonal exchange interaction). The appearance of these two peaks at zero field is in contradiction to the experimental evidence of a critical end point on the anomaly line at nonzero axial field (see phase diagram of ref. 7). Also, the sequence of the different orderings (planar and axial) with temperature seems to be reversed as compared with the experimental facts. The microscopic description of the spin configuration in different phases has not been worked out so far.

These shortcomings of the semi-classical model led us to search for a different approach. We found that a much simpler model, namely a classical Heisenberg model can explain some of the recent experimental facts. In our paper, we report on our results from Monte Carlo simulations of an anisotropic classical Heisenberg model in the presence of a magnetic field where the field may have an oblique angle to the easy axis of the system. We study the temperature variations of the specific heat, the transverse and axial magnetisations and antiferromagnetic order parameters and compare directly with experimental observations. We are especially interested in the nature (microscopic configuration) of the phase in between the critical line and the so-called anomaly line of the phase diagram of FeBr$_2$. Our results show quite close resemblance to the recent experimental facts. The paper is organized as follows: in the next section we present the model, in section (III) the Monte Carlo simulation scheme is discussed, section (IV) contains the simulational results and the comparison with experimental facts. At the end of this section, the microscopic spin configuration in different phases is shown. The paper ends with a summary and concluding remarks in section V.
II. THE CLASSICAL ANISOTROPIC HEISENBERG MODEL

The classical, anisotropic Heisenberg model with competing interactions in presence of a magnetic field can be represented by the Hamiltonian

\[ H = -J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - J' \sum_{\langle ij \rangle'} \vec{S}_i \cdot \vec{S}_j - D \sum_i (S^z_i)^2 - \vec{H} \cdot \sum_i \vec{S}_i, \]  

(2.1)

where \( \vec{S}_i \) represents a classical spin vector of magnitude unity at site \( i \) of the lattice. This spin vector may point into any direction in spin space continuously. For simplicity, we have chosen a tetragonal lattice of linear size \( L \). The ranges of interactions are limited to the nearest neighbors only where the first sum is over the intra-planar exchange interactions which are ferromagnetic \((J > 0)\) and the second sum is over the inter-planar exchange interactions which are antiferromagnetic \((J' < 0)\). \( D \) is the uniaxial anisotropy constant favoring the spin to be aligned either parallel or antiparallel to the \( z \)-axis and \( \vec{H} \) is the external, uniform magnetic field. We use periodic boundary conditions in all directions.

We have performed Monte Carlo simulations for the system described above where we used a system size of \( L = 20 \). Measuring all energetical quantities in units of the ferromagnetic intra-planar interaction \( J \) we set the antiferromagnetic inter-planar interaction to \( J' = -0.5J \) and the anisotropy to \( D = 0.3J \). It should be noted here that very large values of \( D \) will yield Ising-like behavior. In order to be able to observe transverse ordering one has to choose lower values for the anisotropy. We have suitably chosen the parameter values in such a way that the anisotropy is high enough to yield a longitudinal antiferromagnetic phase at low magnetic field and low enough to allow for reasonably large transverse spin components so that the qualitative behavior of the transverse components of magnetisation and order parameter can be observed within the Monte Carlo method. The specific choice of the parameter values was optimized by trial-and-error. We are aware of the fact that our choice of parameters is not realistic compared to FeBr\(_2\). Especially, the value of \( D \) is much too low in our simulations. On the other hand, it is known that the exchange interaction
takes place between a large number of spins, it is not restricted to the nearest neighbours only. The transverse ordering in experimental systems is much smaller compared with the longitudinal one (less than $1\%$). These effects are too small to be observed in a realistic, quantitative simulation. Hence, we restrict ourselves to a pure qualitative description of certain effects that might be comparable to those found experimentally.

### III. MONTE CARLO SIMULATION SCHEME

We performed extensive Monte Carlo simulations of the system above using the following algorithm. At fixed temperature $T$ and field $\vec{H}$, we choose a lattice site $i$ randomly and updated the spin value $\vec{S}_i$ to $\vec{S}_i'$ (randomly chosen on an unit sphere) by using the Metropolis rate:

$$W(\vec{S}_i \rightarrow \vec{S}_i') = \text{Min}[1, \exp(-\Delta \mathcal{H}/k_B T)]$$

where $\Delta \mathcal{H}$ is the change of energy due to the change of the direction of the spin vector from $\vec{S}_i$ to $\vec{S}_i'$. We set the Boltzmann constant to $k_B = 1$. $L^3$ such random updates of spins is defined as one Monte Carlo Step per site (MCSS).

Starting from an initially random configuration (corresponding to a high temperature phase) we equilibriate the system up to $4 \times 10^4$ MCSS and calculated thermal averages and fluctuations from further $4 \times 10^4$ MCSS. Hence, the total length of the simulation for one fixed temperature $T$ is $8 \times 10^4$ MCSS. Then we decrease the temperature and use the last spin configuration of the simulation at this temperature as the initial configuration for the calculation for the next temperature. In this way we simulate a cooling procedure which is closer to equilibrium compared to starting at each temperature with a random spin configuration. The CPU time needed for $8 \times 10^4$ MCSS is approximately 1 hour on an IBM RS/6000-590 workstation.

We have calculated the following quantities:

1. Sublattice magnetisation components for odd and even labelled planes:
\[ m^q_{o,e} = \frac{2}{L^3} \sum_{i \in \{e\}, \{o\}} \langle S^q_i \rangle \]

where \( q \in \{x, y, z\} \) and the sum is over all sites in either even or odd labeled planes. \(< \ldots >\) denotes an average over time (MCSS) (assuming ergodicity and, hence, that an ensemble average and the time average yields the same results).

2. Longitudinal antiferromagnetic order parameter:
\[ O^z_{AF} = \frac{1}{2} |(m^z_o - m^z_e)| \]

3. Longitudinal ferromagnetic order parameter:
\[ M^z_F = \frac{1}{2} (m^z_o + m^z_e) \]

4. Transverse antiferromagnetic order parameter:
\[ O^{xy}_{AF} = \frac{1}{2} \sqrt{(m^x_o - m^x_e)^2 + (m^y_o - m^y_e)^2} \]

5. Transverse ferromagnetic order parameter:
\[ M^{xy}_F = \frac{1}{2} \sqrt{(m^x_o + m^x_e)^2 + (m^y_o + m^y_e)^2} \]

6. Total energy per lattice site
\[ E = \frac{1}{L^3} \langle \mathcal{H} \rangle \]

7. Specific heat per site:
\[ C = L^3 \delta E^2 / (k_B T^2) \]

where, \( \delta E^2 = \langle \frac{1}{L^3} \mathcal{H}^2 \rangle - \langle \frac{1}{L^3} \mathcal{H} \rangle^2 \) are the fluctuations of the energy.

Note that the specific heat \( C \) can also be obtained from the temperature derivative of the energy, \( dE/dT \). Interestingly, it turns out to be a criterion for equilibrium that the two definitions of \( C \) are identical during our simulations.
IV. NUMERICAL RESULTS

As a primary check, we show in Fig. 1 the temperature variation of the longitudinal antiferromagnetic order parameter, \( O_{AF}^z \), the transverse antiferromagnetic order parameter, \( O_{AF}^{xy} \), and the magnetic specific heat, \( C \), at zero field, \( \vec{H} = 0 \).

Our results indicate that at zero field only one transition is observed from a paramagnetic to an antiferromagnetic state where the spins of odd and even planes are aligned alternate parallel and antiparallel to the z-axis (Fig. 1a). The transverse antiferromagnetic and ferromagnetic order parameters remain zero for all temperatures. Consequently the temperature variation of the specific heat (Fig. 1b) shows one single peak at the Néel temperature \( T_N \approx 1.28 \). This is also observed in experiments as a well-known fact. It should be emphasized here that the semi-classical Heisenberg model with Ising-type anisotropy shows two peaks following from two transitions for zero field (see Fig. 1 of Ref. 7), which is not consistent with the experimental facts.

In an applied field parallel to the easy axis the (longitudinal) antiferromagnetic ordering is stable for fields up to \( H_z \leq 0.64 \). The peak position of the specific heat shifts towards lower temperature as one increases the axial field \( H_z \). This result is also consistent with the experimental observations.

To compare with recent experimental observations, we now apply a small transverse field (\( H_x = 0.1 \)) in addition to an axial field of \( H_z = 0.7 \). It should be noted that in real experiments the effect of a transverse field has been incorporated just by tilting the sample by a certain angle \( \theta \) with respect to the direction of the field. Fig. 2a shows the temperature variation of the magnetic specific heat measured from both, the fluctuations of the energy and the temperature derivative of the energy. Both results agree reasonably well and show two peaks in agreement with experimental facts. The high temperature peak is usually called the critical one while the low temperature sharp peak close to the broad anomalous maximum of the specific heat (not reproduced in our simulations) is not yet explained. For a direct comparison we refer to see Fig. 2 of Ref. 8, keeping in mind that in our simulation...
we fixed the field and varied the temperature whereas the reverse was done in experiments

The low temperature sharp peak can be identified as signature of a first order phase transition while the high temperature peak seems to be associated with a second order phase transition. This follows immediately from the temperature variation of the total energy $E$ which is presented in Fig. 2b. At low temperature there is a jump of the energy - a latent heat - which appears as a sharp peak in the specific heat. To characterize the nature of this phase in the intermediate temperature range (in between the two peaks of the specific heat) we have studied also the temperature variation of the longitudinal and transverse order parameters, respectively.

Fig. 3a shows the temperature variation of the longitudinal antiferromagnetic order parameter $O_{AF}$. The behavior of $O_{AF}$ clearly indicates two phase transitions, one at higher temperature ($T \sim 1.0$) which is continuous and a second one which is of first order (or discontinuous) at lower temperature ($T \sim 0.78$). The temperature variations of the longitudinal ferromagnetic order parameter ($M_z$) and the transverse antiferromagnetic order parameter ($O_{xy}^{AF}$) are shown in Fig. 3b. The transverse antiferromagnetic spin ordering is evident in the intermediate range of temperature. This result is very similar to recent experimental observations made by neutron diffraction.

We conclude that during cooling from high temperatures, the system first orders continuously to a transverse antiferromagnetic phase. The corresponding ordering temperature is marked as $T_c$. This transverse antiferromagnetic order increases as the temperature decreases and at lower temperature a second transition occurs where the transverse antiferromagnetic order jumps to a lower value leading to a mainly longitudinal antiferromagnetic order. In other words, this second transition corresponds to a discontinuous rotation of the staggered magnetisation vector from a mainly transverse direction to a mainly longitudinal one. It should be mentioned here that the opposite scenario was observed in the semi-classical model with off-diagonal interaction studied recently (see Fig. 9 of Ref. 7).

For a direct comparison with the earlier experiments, we have calculated the magnetisation components parallel ($M_\parallel$) and perpendicular ($M_\perp$) to the total applied field $\vec{H} =$
\(H_x \hat{x} + H_z \hat{z}\) from the longitudinal and transverse magnetisation components. In experiments, the latter are termed as \(M_{\text{ax}}\) and \(M_{\text{pl}}\), respectively. We have, \(\theta = \tan^{-1}(H_x/H_Z) \approx 8.2^\circ\). In the experiment this tilting angle was even larger (approximately 30\(^\circ\)) but our choice for this angle \(\theta\) is restricted by the parameter values used in the simulation. \(M_\parallel\) and \(M_\perp\) can be readily calculated just by applying a rotation of angle \(\theta\) which yields \(M_\parallel = M_x^F \cos \theta + M_{xy}^F \sin \theta\) and \(M_\perp = -M_x^F \sin \theta + M_{xy}^F \cos \theta\). The temperature variations of \(M_\parallel\) and \(M_\perp\) obtained in this way are shown in Fig.3c. The weak first order jump is evident and the data agree qualitatively with the experimental diagram (see Fig. 3 of Ref.\(^4\)). The transition at higher temperature is indicated by a marker \(T_c\), where the slope of \(M_\parallel\) (i.e., \(dM_\parallel/dT\)) becomes maximal.

What will be the microscopic spin structure in all different phases? The high temperature phase is disordered, of course with a paramagnetic response to the external field. Hence, as the temperature decreases the longitudinal component of total magnetisation increases. At \(T_c\), the transverse antiferromagnetic order starts to develop and consequently, the longitudinal component of the total magnetisation decreases. The spin structure of this phase is sketched in Fig. 4 (marked as TSF). It is a spin-flop (SF) phase, slightly tilted along the positive \(x\)-direction due to presence of the transverse field. We call it a tilted spin-flop phase (TSF).

To understand this phase let us first recall the structure of a spin-flop phase. In a pure spin-flop phase (drawn and marked as SF in Fig. 4), one finds longitudinal ferromagnetic order and transverse antiferromagnetic order as follows from the \(x\)- and \(z\)-components of the spin vector which are also shown. Lowering the temperature from a paramagnetic phase, first the longitudinal magnetisation will increase and then will remain constant if the angle between two spins remains constant or increases if the angle between two spins decreases. At the transition temperature \(T_c\) the slope \(dM_z^{AF}/dT\) will change rapidly. The longitudinal antiferromagnetic order parameter remains zero since one has equal values of \(m_z^\alpha\) and \(m_z^\varepsilon\). One can characterize the spin-flop (SF) phase by, \(M_x^F \neq 0\), \(O_{xy}^{AF} \neq 0\), \(O_x^{AF} = 0\) and \(M_{xy}^F = 0\). It is mainly a coexistence of axial ferromagnetic order and transverse antiferromagnetic...
order.

However, in the tilted spin-flop phase, i.e., in presence of a transverse field, the spins in one layer will be more aligned along the positive $x$-direction compared to the spins in the neighbouring layer (see TSF in Fig. 4). This will increase the angle between the two spins and as a result, the longitudinal magnetisation will start to decrease as one decreases the temperature. Almost the same effect can be observed in the temperature variation of $M_\parallel$ (see our Fig. 3c and for comparison also the experimental situation, Fig. 3 of Ref. 4). Due to unequal values of $m^z_o$ and $m^z_e$ one obviously will find nonzero values of the longitudinal antiferromagnetic order parameter in the TSF phase (see Fig. 3a). But nevertheless the system is effectively ferromagnetically ordered since the signs of the values of $m^z_e$ and $m^z_o$ are the same even when the absolute values are different so that the longitudinal antiferromagnetic order parameter is nonzero in this phase. Since the absolute values of the transverse magnetisations of the two different sublattices are different (although they are oppositely directed), the transverse magnetisation is nonzero. This observation has also been made in experiments. Hence in the TSF phase it is $M^z_F \neq 0$, $O^x_{AF} \neq 0$, $O^z_{AF} \neq 0$ and $M^{xy}_F \neq 0$.

After a further decrease of temperature one will encounter a phase with longitudinal antiferromagnetic (AF) order. The transition from TSF- to AF-phase is of first order. This is consistent with the experimental observations. The weak jumps of $M_\parallel$ and $M_\perp$ (see our Fig. 3c and for comparison with experiments Fig. 3 of Ref.) is a signature of a discontinuous transition from a tilted spin-flop (TSF) phase to a longitudinal antiferromagnetic (AF) phase. In a pure longitudinal antiferromagnetic (AF) phase, $M^z_F = 0$, $O^{xy}_{AF} = 0$, $O^z_{AF} \neq 0$ and $M^{xy}_F = 0$. Strictly speaking, due to the application of a small $H_z$ one will have very small but nonzero value of $M^{xy}_F$.

In addition, we have also studied the temperature variation of the transverse antiferromagnetic susceptibility ($\chi^{xy}_{AF} = \frac{L^3(\delta O^{xy}_{AF})^2}{k_BT}$) shown in Fig. 5. The two transitions, i.e., at high temperature from saturated paramagnetic to tilted spin-flop and at low temperature from a tilted spin-flop to longitudinal antiferromagnetic phase, are evident from the figure.
V. SUMMARY

Motivated by recent experimental observations in the metamagnet FeBr$_2$, we have studied a classical anisotropic Heisenberg model with ferromagnetic intra-planar interaction and antiferromagnetic inter-planar interaction by Monte Carlo simulations. We focused on the temperature variations of the magnetic specific heat, longitudinal and transverse order parameters (both ferromagnetic and antiferromagnetic) and $M_\parallel$ and $M_\perp$, where the system is in a magnetic field tilted to the easy axis of the system.

Transverse spin ordering and a weak first order transition (additional to the well known antiferromagnetic transition) associated with a very sharp peak of the magnetic specific heat at low temperature are observed in agreement with experiments. The high temperature phase transition is identified as a continuous transition from a paramagnetic phase to a tilted spin-flop phase while the low temperature transition is discontinuous and from tilted spin-flop phase to a longitudinal antiferromagnetic phase.

None of the models studied so far theoretically can provide a reasonably well explanation for all experimental facts observed in the FeBr$_2$ metamagnet at the same time. Monte Carlo calculations in an Ising model on a hexagonal lattice with realistic interaction parameters can reproduce the broad anomalous maximum of the specific heat at $H_\perp(T)$. This anomaly is not reproduced within our simulations. It was shown that this anomaly is due to a strong Ising character of FeBr$_2$ and it is due to the fact that one needs a large number of interlayer interaction neighbors.

On the other hand, recent experimental observations of transverse ordering cannot be explained by an Ising model. The semi-classical Heisenberg model with off-diagonal interaction contains the anomaly of the specific heat as well as the sharp peak additional to the usual transition. But the sequence of different ordering seems to be in contradiction with recent neutron diffraction results. Most importantly, it gives two transitions (associated with two peaks of the specific heat) even in zero field. However, very probably, the phase line $H_1(T)$ ends up at a critical end point at nonzero field. On the other hand, our
much simpler approach, with a classical anisotropic Heisenberg model, can explain some of the recent experimental facts and it can also provide a microscopic description of the different ordering. To find a model which can explain the entire phase diagram of the FeBr$_2$, in our opinion is still an open problem.

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FIGURES

FIG. 1. Temperature variations of (a) longitudinal ($O^z_{AF}$) and transverse ($O^{xy}_{AF}$) antiferromagnetic order parameter (solid line is just connecting the data points) and (b) specific heat $C$ (solid line represents $\frac{dE}{dT}$). $\vec{H} = 0$.

FIG. 2. Temperature variations of (a) specific heat $C$ (the continuous line represents $dE/dT$) and (b) total energy $E$, for $H_z = 0.7$ and $H_x = 0.1$.

FIG. 3. Temperature variations of (a) longitudinal antiferromagnetic order parameter ($O^z_{AF}$), (b) longitudinal ferromagnetic ($M_z$) and transverse antiferromagnetic ($O^{xy}_{AF}$) order parameter, and (c) $M_\parallel$ and $M_\perp$ as explained in the text. Solid lines in (a) and (b) are just connecting the data points. $H_z = 0.7$ and $H_x = 0.1$.

FIG. 4. Schematic representation of an antiferromagnetic (AF) phase, a spin-flop (SF) phase, and a tilted spin-flop (TSF) phase. Each vector may represents the magnetisation of one plane of the system.

FIG. 5. Temperature variation of the transverse antiferromagnetic susceptibility ($\chi^{xy}_{AF}$) for $H_x = 0.1$ and $H_z = 0.7$. The solid line is just connecting the data points.