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Phase diagram of the antiferromagnetic *XY* model with a triangular lattice in an external magnetic field

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Abstract. The ordered states of a planar antiferromagnet with a triangular lattice are investigated in the presence of a magnetic field. The spin wave free energy is taken into account and proves to be important for determining the properties of the system. The phase diagram is constructed. It contains four different phases with rigorous long-range order. Three of them are characterised by different configurations of mean magnetic moments for the three sublattices. The existence of one more non-trivial phase with an algebraic decay of the correlation functions is very probable.

1. Introduction

In the exchange approximation a planar antiferromagnet can be described with the Hamiltonian:

$$H = J \sum_{(jj')} \boldsymbol{m}_j \cdot \boldsymbol{m}_{j'} = J \sum_{(jj')} \cos(\varphi_j - \varphi_{j'}) \tag{1}$$

where $\boldsymbol{m}_j = (\cos \varphi_j, \sin \varphi_j)$ are unit planar vectors defined on lattice sites and the summation is performed over pairs of nearest neighbours. In the case of a flat triangular lattice (and this is the case we are interested in) the ground state consists of three sublattices. The magnetic moments (spins) $\boldsymbol{m}_j$ belonging to different sublattices form the angles 120° with respect to each other:

$$\phi_2 = \phi_1 \pm 120° \qquad \phi_3 = \phi_1 \mp 120°. \tag{2}$$

Here ϕ_l ($l = 1, 2, 3$) denote the common values of φ_j in each of the three sublattices. In addition to continuous degeneracy caused by the invariance of the Hamiltonian with respect to homogeneous rotation of all spins the ground state also possesses two-fold discrete degeneracy (upper and lower signs in (2)). The order parameter degeneracy space R is, accordingly, a pair of circumferences:

$$R = Z_2 \times S^1.$$

It proves convenient to describe this additional (discrete) degeneracy by introducing the helicity (vorticity) v_n of each triangular plaquette Γ_n:

$$v_n = (1/2\pi) \sum_{(jj') \in \Gamma_n} \{\varphi_j - \varphi_{j'} - \pi\}. \tag{3}$$

Here the curly brackets imply that the difference $\varphi_j - \varphi_{j'} - \pi$ must be reduced to an interval $(-\pi, \pi)$. If one neglects the possibility of the strictly ferromagnetic configuration of spins, the helicities v_n prove to acquire the values $\pm\frac{1}{2}$ only. Our definition of helicity (equation (3)) seems to be more adequate than the one used by other authors (see, e.g., Lee *et al* 1984b) because it excludes the possibility of zero helicity and stresses the Ising character of this variable.

In the ground state the positive and negative helicities alternate regularly (figure 1).

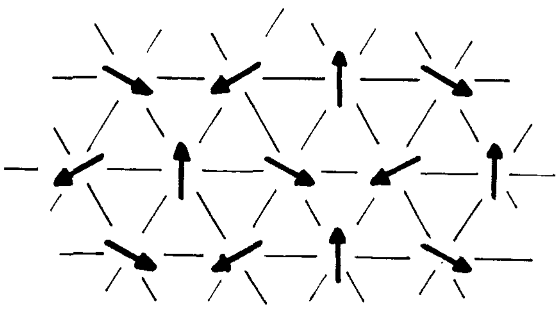

Figure 1. The ground state of the AF XY (t)-model in zero magnetic field.

The two-fold discrete degeneracy corresponds to a simultaneous changing of signs of all v_n.

Miyashita and Shiba (1984) studied phase transitions in the antiferromagnetic XY model with a triangular lattice (AF XY(t) model) by means of a numerical simulation. They have shown that the Berezinskii–Kosterlitz–Thouless transition (BKT transition) associated with the restoring of the continuous symmetry takes place at slightly lower temperature than the Ising-type transition associated with the breaking of antiferromagnetic ordering in v_n (Lee *et al* (1984b) failed to discover the difference between the temperatures of these transitions).

In the presence of a magnetic field the Hamiltonian of the AF XY(t) model should be rewritten as:

$$H = J \sum_{(jj')} \boldsymbol{m}_j \cdot \boldsymbol{m}_{j'} - \boldsymbol{h} \cdot \sum_j \boldsymbol{m}_j = J \sum_{(jj')} \cos(\varphi_j - \varphi_{j'}) - h \sum_j \sin \varphi_j \qquad (4)$$

where angles φ_j are measured from the direction perpendicular to $\boldsymbol{h}$. A rather striking feature of the AF XY(t) model is that the continuous degeneracy of the ground state is retained in the presence of the magnetic field. The minimum of (4) is achieved when the conditions:

$$\begin{aligned} &\cos \phi_1 + \cos \phi_2 + \cos \phi_3 = 0 \\ &\sin \phi_1 + \sin \phi_2 + \sin \phi_3 = h/(3J) \end{aligned} \qquad (5)$$

are fulfilled. These conditions keep one free parameter (see, e.g., Lee *et al* 1984b). With increasing h up to $h_{c1} \equiv 3J$ the two circumferences of the order parameter degeneracy space merge in three points. With further increasing temperature these junctions split again crosswise, the order parameter of the degeneracy space becoming only one circumference. At $h = h_{c2} \equiv 9J$ this circumference merges into a point corresponding to the configuration with all spins aligned parallel (paramagnetic state).

It seems worth mentioning that the continuous degeneracy of the ground state in the presence of the magnetic field is accidental (i.e. not caused by any symmetry). It is removed with an arbitrary change of the form of the interaction. However, in the case of the pure exchange interaction it is retained even when the interaction of more distant neighbours is taken into account.

The phase diagram of the model (4) was investigated by means of a numerical simulation (Lee *et al* 1984b) and analytically: in a mean-field approximation (Lee *et al* 1984a), by means of constructing the phenomenological Hamiltonians (Lee *et al* 1984b) and by studying relevant topological excitations (Dotsenko and Uimin 1984, 1985). However, it has been overlooked by the authors of these analytical studies that at finite temperatures the continuous degeneracy is removed due to the free energy of the spin waves.

In § 2 of this paper the spin wave free energy is calculated in the simplest (harmonic) approximation, which is valid at low temperatures. This free energy proves to be dependent on the particular kind of vacuum state. The spin configurations required to minimise it are found. In § 3 disordering due to thermal fluctuations is taken into account and the phase diagram of the model (4) is constructed on the basis of the analysis of § 2. Section 4 is devoted to the discussion of the results obtained and of the experimental situation.

Some of the results of this paper have been published previously (Korshunov 1985), and similar results have been obtained by Kawamura (1984).

2. Minimisation of the spin wave free energy

We would like to calculate the spin wave free energy in the harmonic approximation. In order to do this it is convenient to supplement the Hamiltonian (4) with a fictitious kinetic energy in the form, say:

$$K = (M/2) \sum_j \dot{\varphi}_j^2 .$$

A variation of the Lagrangian

$$L = K - H$$

with respect to φ_j yields the equations of motion, which in the linearised form can be written as

$$\begin{aligned}
-M\delta\ddot{\varphi}_1 &= h \sin\phi_1 \delta\varphi_1 - 3J[\cos(\phi_1 - \phi_2)(\delta\varphi_1 - a\delta\varphi_2) \\
&\qquad + \cos(\phi_1 - \phi_3)(\delta\varphi_1 - a^*\delta\varphi_3)] \\
-M\delta\ddot{\varphi}_2 &= h \sin\phi_2 \delta\varphi_2 - 3J[\cos(\phi_2 - \phi_3)(\delta\varphi_2 - a\delta\varphi_3) \\
&\qquad + \cos(\phi_2 - \phi_1)(\delta\varphi_2 - a^*\delta\varphi_1)] \\
-M\delta\ddot{\varphi}_3 &= h \sin\phi_3 \delta\varphi_3 - 3J[\cos(\phi_3 - \phi_1)(\delta\varphi_3 - a\delta\varphi_1) \\
&\qquad + \cos(\phi_3 - \phi_2)(\delta\varphi_3 - a^*\delta\varphi_2)]
\end{aligned} \tag{6}$$

where

$$a = a(k) = \tfrac{1}{3} \sum_{l=1}^{3} \exp(\mathrm{i}\boldsymbol{e}_l \cdot \boldsymbol{k})$$

and we have introduced the Fourier transforms for the deviations $\delta\varphi_j$ of φ_j from their equilibrium values separately for each of the three sublattices (denoted by the indices 1, 2, 3). The numbering of the sublattices and basis vectors $\boldsymbol{e}_l$ ($\boldsymbol{e}_1 + \boldsymbol{e}_2 + \boldsymbol{e}_3 = 0$) are shown in figure 2. The particular kind of ground state is characterised by the ϕ_l—the equilibrium values of φ_j in each of the three sublattices. The ϕ_l are assumed to satisfy (5). The equation for the eigen-frequencies of small oscillations can be easily obtained as the

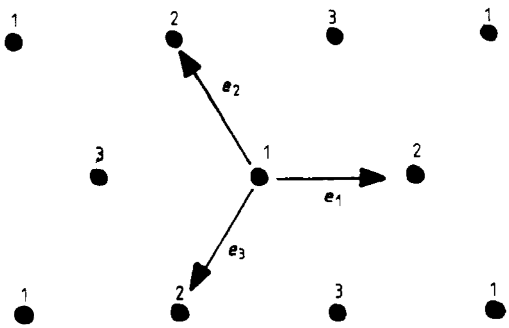

Figure 2. The numbering of sublattices and the basis vectors used in equations (6).

condition of compatibility of (6):

$$\det\begin{Vmatrix} 1-\frac{1}{3}M\omega^2/J & a\cos(\phi_1-\phi_2) & a^*\cos(\phi_1-\phi_3) \\ a^*\cos(\phi_2-\phi_1) & 1-\frac{1}{3}M\omega^2/J & a\cos(\phi_2-\phi_3) \\ a\cos(\phi_3-\phi_1) & a^*\cos(\phi_3-\phi_2) & 1-\frac{1}{3}M\omega^2/J \end{Vmatrix} = 0. \qquad (7)$$

It has three solutions, one of them corresponding to the gapless mode and the other two, to the modes with gaps. The existence of the gapless mode is caused by the continuous degeneracy of the ground state.

The free energy of the harmonic oscillator with the frequency ω in the classical limit is given by:

$$F_0 = -k_BT\ln(k_BT/\hbar\omega) = \tfrac{1}{2}k_BT\ln[\omega^2/(k_BT/\hbar)^2]. \qquad (8)$$

The total free energy of all the three modes for a given value of the wave-vector $\boldsymbol{k}$ is dependent only on the product of the squared eigen-frequencies which can be obtained from (7) without solving it:

$$\omega_1^2\omega_2^2\omega_3^2 = (3J/M)^2[1-(a^3+a^{*3})/2 - AS] \qquad (9)$$

where

$$A = A(\boldsymbol{k}) = aa^* - (a^3+a^{*3})/2 \qquad (10)$$

and

$$S = S(\phi_1,\phi_2,\phi_3) = \cos^2(\phi_1-\phi_2)+\cos^2(\phi_2-\phi_3)+\cos^2(\phi_3-\phi_1). \qquad (11)$$

The function A being non-negative, the minimal free energy is achieved for the maximum value of S. As one could expect, the term connected with M can be separated in the free energy (8) and proves to be independent of the ϕ_l and therefore should not be taken into consideration.

The attentive reader can get an impression that at finite temperatures performing a quadric expansion on the background of the fixed ground state produces a result that is inconsistent due to the absence of a rigorous long-range order in φ. We want to stress that when the anisotropy found is self-consistently taken into account a gap appears in the gapless mode. The long-range order becomes rigorous. It seems worth remembering that in two-dimensional systems with a continuous Abelian symmetry an arbitrary small anisotropy proves to be relevant if the temperature is small in comparison with the constant in the gradient energy term (Pokrovsky and Uimin 1973a, b, José *et al* 1977).

Let us now consider what kind of states have the minimal free energy of the spin waves. The configurations of spins in different sublattices maximising $S(\phi_1, \phi_2, \phi_3)$ with constraints (5) being taken into account are shown in figure 3. For $h < h_{c1}$ the three

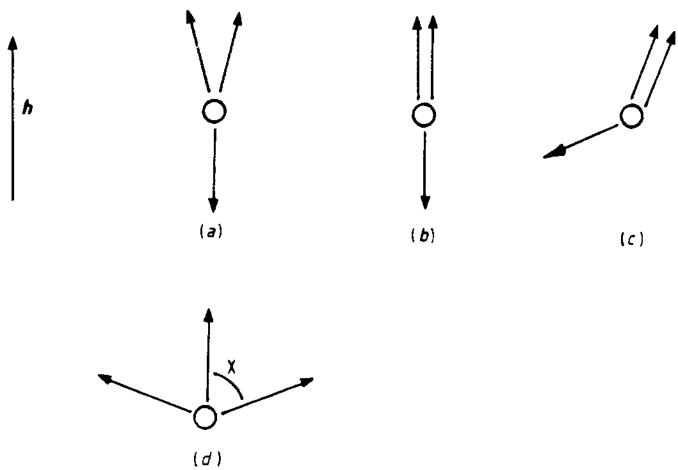

Figure 3. Configuration of spins of the three sublattices with the minimal spin wave free energy: (*a*), $0 < h < h_{c1}$; (*b*), $h = h_{c1}$; (*c*), $h_{c1} < h < h_{c2}$; (*d*), in the case of the opposite sign of the anisotropic part of the free energy.

sublattices are non-equivalent. On one of them the spins are antiparallel to the field, and on the two others they have the perpendicular-to-field components of opposite signs (figure 3(*a*)). This state is six-fold degenerate in accordance with a number of possible permutations of non-equivalent sublattices.

With increasing h the angle between the spins, which are not antiparallel to the field, diminishes, and at $h = h_{c1}$ vanishes (figure 3(*b*)). Two of the sublattices become equivalent, the spins on them being parallel to the field. The degeneracy multiplicity of this state is equal to three.

For $h_{c1} < h < h_{c2}$ the equivalence of two of the sublattices is retained, but the symmetry in the direction perpendicular to the field becomes broken (figure 3(*c*)). That leads to the degeneracy multiplicity being increased up to six, as for $0 < h < h_{c1}$. The anharmonicities being taken into account, the magnetic moment in this state is not parallel to the field (at finite temperatures).

Thus we have considered which states are preferred by the spin wave free energy at the lowest temperatures.

3. Phase diagram

For magnetic fields only slightly exceeding h_{c1} the first phase transition to occur with increasing temperature should be the Ising-type transition restoring the broken symmetry in the perpendicular-to-field direction. This can be explained by the small value of the domain wall free energy for the states that differ only by the signs of the perpendicular-to-field components of the magnetic moment. The transition considered restores the equivalence of two such states, being thus of the Ising type. At higher temperatures the degeneracy multiplicity is equal to three. The temperature of this transition should decrease with decreasing h (like the corresponding domain wall free energy) and at $h = h_{c1}$ should become zero.

Analogously, for the fields slightly lower than h_{c1} the domain wall with minimal free energy also separates the states differing by the sign of the perpendicular-to-field component of the magnetic moments of different sublattices. In this case however both states are symmetric in the perpendicular-to-field direction. They can be transformed into each other by the permutation of the two sublattices. With an increase in temperature the equivalence of these states should be restored. This transition is also of the Ising type, and the temperature of the transition should also be zero at the point $h = h_{c1}$.

In the phase diagram of the model (4) obtained by Lee *et al* (1984b) by means of numerical simulation both of these lines are present. The analysis carried out in the previous section allows one easily to identify the three ordered phases discovered by these authors. In each of them the mean values of the magnetic moments for each of the three sublattices should be directed as figure 3(*a*), 3(*b*) or 3(*c*) shows.

The phase diagram resulting from our analyses with the results of numerical simulations of Miyashita and Shiba (1984) and of Lee *et al* (1984b) being taken into account is shown in figure 4 in coordinates T/J, h/J. On the curves DB and DC the Ising-type transitions described above take place.

The phase b can be treated as a commensurate crystal $\sqrt{3} \times \sqrt{3}$ of spins antiparallel to the field on the background of the parallel ones (we are speaking of the mean values of the spins). The exact solution of the hard hexagon model (Baxter 1980) shows that the melting of such structure (line BC) should proceed with the same critical exponents as the phase transition of the three-state Potts model. An assumption however was put forward (Huse and Fisher 1984) that the breaking of the chiral symmetry (i.e. the dependence of the domain wall free energy both on the particular states that are separated and on the orientation of the wall) may lead to a change in the universality class.

The line AB corresponds to the phase transition in the Z_6 model with chiral asymmetry, so the type of transition cannot be predicted unambiguously either. For h close to h_{c2} the values of the ϕ_l satisfying (5) in the leading order can be expressed parametrically as:

$$\phi_1 = b \sin\theta \qquad \phi_2 = b \sin(\theta + 120°) \qquad \phi_3 = b \sin(\theta - 120°)$$

where $b = 2[(h_{c2} - h)/h_{c2}]^{1/2}$ (Dotsenko and Uimin 1985), so the original AF XY(t) model can be approximated by the ordinary XY model for variable θ. When the spin wave free energy is taken into account the effective sixth-order anisotropy field emerges for variable θ. Thus the splitting of the phase transition into two seems very probable, the intermediate phase being characterised by the algebraic decay of the correlation function for θ (just as it should be in the case of the ordinary six-state clock model, see José *et al*

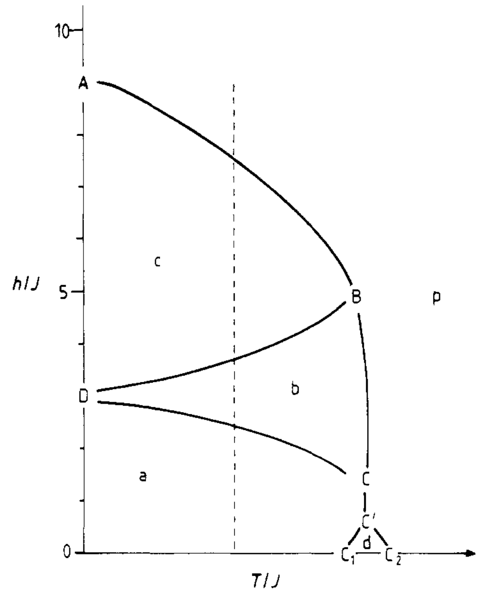

Figure 4. Phase diagram of the AF XY(t) model in an external magnetic field. In phases a, b and c the mean magnetic moments of different sublattices form configurations similar to those shown in figures 3(a), 3(b) and 3(c) respectively. In phase d only the long-range order with respect to helicities is retained. Phase p is paramagnetic.

1977). So the curve BA (or at least part of it) should split into two curves joining in the point A.

Let us now consider the form of the phase diagram for the low fields. The numerical simulation of Miyashita and Shiba (1984) shows that in zero field the BKT transition precedes that of the Ising type. This Ising-type transition restores the equivalence between the states differing in signs of the helicities in each elementary cell and therefore is essentially the same transition as the transition on the line DC which, so it seems, should terminate at the point C_2 of the Ising-type transition in zero field.

For small fields the influence of the spin wave free energy is equivalent to that of the three-fold anisotropy field for a continuous variable. So the point C_1 of the BKT transition in zero field should not be an isolated singular point but it should serve as an end-point for a line of phase transitions between the six-fold-degenerate and two-fold-degenerate states (cf José *et al* 1977). This makes us conclude that one more ordered phase should exist (phase d in figure 4). In this phase the mean values of magnetic moments are equal for all the sublattices, but the 'antiferromagnetic' order with respect to the helicities is retained. The point of coexistence of four phases (phases a, b, d and paramagnetic phase p) is likely to split into two triple points (points C and C′ in figure 4).

The line $C'C_2$ is the line of the Ising-type transitions, and the line $C'C_1$ that of the three-state Potts model transitions. The phase transitions on the line CC′ are likely to be of first order, just as in the six-state Potts model (Baxter 1973) or in a six-state cubic model (Nienhuis *et al* 1983).

When the interaction of more distant neighbours is taken into account the sequence and types of the transitions in zero field can change (Korshunov and Uimin 1986). The existence of the phase d is possible only if in zero field the BKT transitions take place at a lower temperature than that of the Ising type.

4. Discussion

In the two previous sections we have studied the phase diagram of the AF XY(t) model in an external magnetic field and have found four different ordered phases with rigorous long-range order (in one of them—only with respect to helicities). The existence of one more non-trivial phase with an algebraic order for the variable seems very probable.

Our conclusions about the nature of the ordered phases coincide with those of Lee *et al* (1984b) with respect to the phase b and contradict them for phases a and c. It is claimed by these authors that the configurations of mean magnetic moments in each of the three sublattices to be achieved in phases a and c are those that correspond to the other sign of the anisotropy energy (i.e. to free energy being minimal for minimal values of S). We would like to stress that if such was the case, only one ordered phase would exist, with a configuration of spins such as in figure 3(d). The spins in one of the three non-equivalent sublattices would be parallel to the field and the angle χ would vary continuously from 120° to zero with increasing h. The phase diagram would have no non-trivial structure at low temperatures.

The AF XY(t) model is not of purely academic interest. The triangular lattice is typical of the absorbed monolayer forming two-dimensional crystal. It may be expected that the dense monolayer of O_2 on a graphite substrate (Stephens *et al* 1980) or a thin layer of the solid O_2 (α-phase) will exhibit the peculiar behaviour described in this paper.

One more physical system that can be described by the AF XY(t) model is the layer of Eu intercalated in graphite. The dependence of the magnetic moment on h experimentally observed in EuC_6 at low temperatures (see Suematsu *et al* (1981) and references therein) agrees with the existence of three different ordered phases. The corresponding path is shown in the phase diagram (figure 4) by the broken line.

Acknowledgment

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