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THERMODYNAMIC PROPERTIES OF SPIN-ONE UNIAXIAL ANTIFERROMAGNETS

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ABSTRACT

The thermodynamic properties of spin-one Heisenberg antiferromagnet with uniaxial anisotropy are investigated in the mean field approximation. The anisotropy is not approximated by a local molecular field but is considered exactly. The phase diagrams are presented for different anisotropy constants D with the applied magnetic field H parallel to the easy axis of magnetization. It is shown, that the phase diagram in the H vs. D plane exhibits a new critical point. At this critical point the nature of the phase transition from the paramagnetic to the spin-flop phase changes from the second to the first order.

1. INTRODUCTION

The uniaxial Heisenberg antiferromagnet has been investigated intensively [1) -4)], however with the anisotropy term approximated by a molecular field usually. This approximation has been subjected to criticism; therefore, Devlin [5)], Kenan [6)], and Vettier [7)] considered the uniaxial anisotropy term exactly but in the absence of the external magnetic field. They were able to obtain an expression for the Néel temperature T_N, which did not diverge with increasing anisotropy, as was the case in the previous studies.

In this paper we investigate the uniaxial Heisenberg antiferromagnet in the presence of an external magnetic field applied along the easy axis of magnetization. We consider the anisotropy term exactly. The Hamiltonian of the antiferromagnet is given in the form

$$\mathcal{H} = \frac{1}{2} \sum_{ab} J \vec{S}_a \vec{S}_b - D\left(\sum_a (S_a^z)^2 + \sum_b (S_b^z)^2\right)$$
$$- g\mu_B H^z \left(\sum_a S_a^z + \sum_b S_b^z\right) . \qquad (1)$$

Here, the two alternating sublattices are denoted by a and b. The first term describes the exchange interaction between the nearest neighbouring a and b lattice sites and J is the exchange coupling constant. The second term in $\mathcal{H}$ is due to the crystal-field uniaxial anisotropy with the easy axis in the z direction, D is the anisotropy constant. The last term is the Zeeman energy, where H^z is the external magnetic field corrected for the demagnetization effects. Within the mean field approximation (MFA) the Hamiltonian splits into two terms, each corresponding to a given sublattice,

$$\mathcal{H} = \sum_a \mathcal{H}_a + \sum_b \mathcal{H}_b - NJ \langle \vec{S}_a \rangle \langle \vec{S}_b \rangle . \qquad (2)$$

The sublattice Hamiltonian $\mathcal{H}_\alpha$ ($\alpha \in$ a or b) is a function of the local molecular field $\vec{\Lambda}_\alpha$

$$\mathcal{H}_\alpha = - \vec{\Lambda}_\alpha \vec{S}_\alpha - D(S_\alpha^z)^2 , \qquad (3)$$

and N is the number of sites within one sublattice.
In the following section we shall briefly consider the method of obtaining the critical fields.

2. MAGNETIC PHASE TRANSITIONS

The magnetic phase diagram of a uniaxial antiferromagnet with magnetic field applied along the easy axis can present three distinct phases: antiferromagnetic (AF), spin flop (SF), and paramagnetic (PM). In the phase diagram these phases are separated by second- or by first-order phase boundaries. Each second order phase boundary is given by one critical field H^c. On the other hand, the first order phase boundary consists of two phase stability limits (H^c) and one

thermodynamic critical field H^{th} at which the free energies of two phases in contact (denoted by 1 and 2) become equal

$$F(1) = F(2). \tag{4}$$

The critical fields are determined by vanishing of the inverse susceptibility tensor,

$$\mathrm{Det}\,(\chi^{-1}) = 0\,, \tag{5}$$

which is related to the free energy F by

$$\chi^{-1}_{\alpha\alpha'} = \frac{1}{N}\frac{\partial^2 F}{\partial\langle\vec{S}_\alpha\rangle\partial\langle\vec{S}_{\alpha'}\rangle}\,. \tag{6}$$

The free energy is

$$F = \langle\mathcal{H}\rangle + kT\cdot\langle\ln\rho\rangle\,. \tag{7}$$

Here ρ is the density matrix, which in the MFA factorizes into individual-site density matrices

$$\rho = \prod_a \rho_a \prod_b \rho_b\,, \tag{8}$$

where

$$\rho_\alpha = \frac{1}{Z}\exp\left\{\beta\left[\vec{\Lambda}_\alpha\vec{S}_\alpha + D(S^z_\alpha)^2\right]\right\}\,. \tag{9}$$

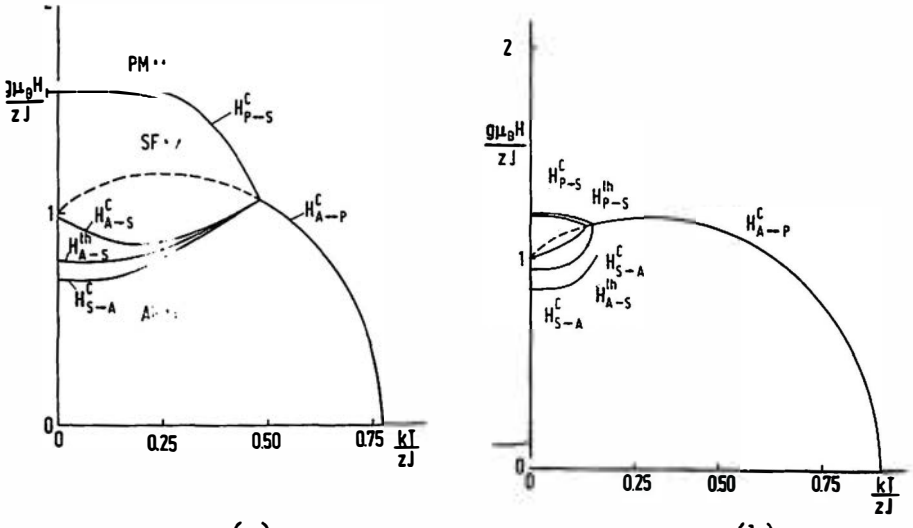
It should be noted that the free energy is a function of the variational parameter $\vec{\Lambda}_\alpha$. The equlibrium spin configuration is obtained from the condition, that F has a local minimum in the $\vec{\Lambda}_\alpha$ space,

$$\frac{\partial F}{\partial\vec{\Lambda}_\alpha} = 0\,. \tag{10}$$

Conditions (5) and (10) determine completely the critical fields H^c. The thermodynamic critical field is, on the other hand, determined by the conditions (4) and (10).

3. RESULTS

The magnetic phase boundaries of S = 1 Heisenberg antiferromagnet for the anisotropy constants D/zJ = 0.4 and D/zJ = 0.8 (z is the number of nearest neighbours in the crystal lattice) are presented in Fig. 1. We see that, as usually,



(a) (b)

Fig. 1. Phase diagrams in the H-T plane of uniaxial Heisenberg antiferromagnet with the anisotropy constants: (a) D = 0.4zJ and (b) D = 0.8zJ

the phase transition between the AF and the SF phase is of the first order. On the other hand, the phase transition between the SF and the PM phase is of the second order only for the anisotropy $D < D_c = \frac{2}{3} zJ$ and becomes of the first order for $D > D_c$. This change in the critical behaviour has not been known up to now because the crystal field anisotropy has not been treated properly in earlier calculations. The anisotropy term causes the precession of spins when the local molecular field is not parallel to the easy axis of magnetization. This is the case in the SF phase. At the SF to PM phase transition the precession and also equilibrium spin orientation change to the configuration of the PM phase continuously for small D and discontinuously for $D > D_c$.

The critical fields in the limit $T \to 0$ are presented in Fig. 2. It is seen, that the SF to PM phase transition changes its nature at $D_c = \frac{2}{3} zJ$. The phase stability limit of the PM phase is given by

$$g\mu_B H^c_{P \to S} = 2zJ - D \, . \tag{11}$$

The second-order critical field between the AF and PM phase

$$g\mu_B H^c_{A \leftrightarrow P} = zJ \tag{12}$$

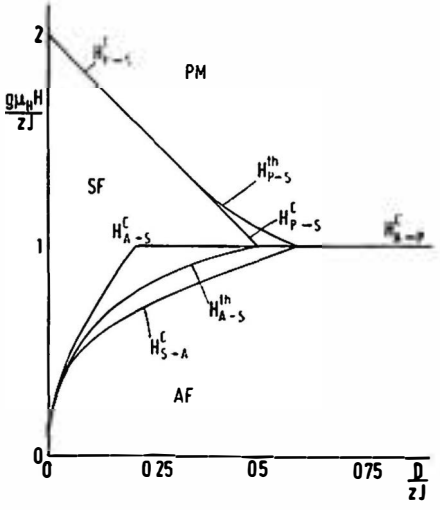


Fig. 2. Phase diagram in the H-D plane at zero temperature

is independent on D, since in the MFA both phases are completely ordered along the easy axis of magnetization and therefore the spins do not feel the anisotropy field at T = 0 K. The anisotropy field does not favour the SF configuration, therefore the critical field separating the SF from AF phase increases with anisotropy. Analytic expression is obtained for the stability limit of the AF phase only

$$g\mu_B H^c_{A\to S} = \left[D\,(D + 2zJ)\right]^{\frac{1}{2}} . \qquad (13)$$

No simple expression can be obtained for the thermodynamic critical fields and for the phase stability limits of the SF phase. The reason is in the spin precession, which is present in the SF phase even at T = 0 K.

In conclusion let us compare the present results with the previous analyses where the anisotropy field has been approximated by a mean field [1]. Contrary to previous results we have found that the PM to SF phase transition becomes of the first order when the anisotropy exceeds $D_c = 2zJ/3$. We have also shown that the phase stability limit of the AF phase against the SF phase $H^c_{A\to S}$ never exceeds the critical field at which the AF phase changes to PM phase ($H^c_{P\leftrightarrow A}$). The reason is in the fact, that beyond the critical field $H^c_{A\leftrightarrow P}$ the PM phase instead of the AF phase exists and that there the PM phase is unstable against the SF phase.

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