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Canted-paramagnetic boundary of anisotropic antiferromagnets in a field of arbitrary direction

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Abstract. A general asymptotic expression for the low-temperature, canted-paramagnetic boundary of a single-ion uniaxial antiferromagnet is obtained as a function of the angle between the external magnetic field and the easy axis. If this angle is other than zero it is shown that in general the critical field behaves asymptotically as a T^2 law. Depending on the values of this angle and of the anisotropy parameter a $T^{3/2}$ dependence can be observed for experimentally accessible low temperatures. These results are considered for the uniaxial antiferromagnetic $NiCl_2 \cdot 6H_2O$.

1. Introduction

The phase boundary between the spin-flop and paramagnetic phases of anisotropic antiferromagnets has been the subject of several experimental and theoretical investigations in recent years. For uniaxial antiferromagnets, the critical magnetic field along the easy direction behaves asymptotically according to a $T^{3/2}$ law (Anderson and Callen 1964, Feder and Pytte 1968, Oliveira Jr *et al* 1978). On the other hand, for transverse anisotropies this asymptotic dependence changes to T^2 (Cieplak 1977, Figueiredo and Salinas 1984). If the field is applied normal to the easy axis the dependence of the critical magnetic field on temperature follows a T^2 law even for uniaxial antiferromagnets (Becerra *et al* 1988).

In this paper a calculation is reported for the canted-paramagnetic boundary of a uniaxial antiferromagnet in a field at an arbitrary angle to the easy magnetic axis of the system. It is shown that whatever the angle θ between the magnetic field and the easy axis, the paramagnetic critical field behaves asymptotically as T^2. However, depending on the relative values of the anisotropy and of the angle θ, a $T^{3/2}$ dependence can be observed for experimentally accessible low temperatures. This is possible because the energy spectrum of magnons can become quadratic in the wavevector in this region. These results are applied to the predominantly uniaxial antiferromagnetic crystal $NiCl_2 \cdot 6H_2O$.

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2. Calculation

We consider the following spin-Hamiltonian on the paramagnetic phase of an antiferromagnet:

$$\mathcal{H} = J\sum_{i,j} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + D\sum_i (S_i^z)^2 + gu_{\mathrm{B}}\sum_i \boldsymbol{H}\cdot\boldsymbol{S}_i \tag{1}$$

where J is the exchange interaction between nearest-neighbour pairs of spins on a simple cubic lattice, and D is a single-ion uniaxial term ($D<0$). The last sum is the usual Zeeman term and the field $\boldsymbol{H}$ is at an angle θ with respect to the easy direction z. In the paramagnetic phase we must consider the elementary excitations of the system around the equilibrium positions of the spins. First, the spin axes are rotated an angle φ about the y direction. The magnetic field is in the xz plane, and the new axis of quantisation is z', which is at an angle φ to the easy direction. The following transformation is obtained for the spin operators:

$$\begin{aligned} S_i^x &= S_i^{x'}\cos\varphi + S_i^{z'}\sin\varphi \\ S_i^y &= S_i^{y'} \\ S_i^z &= S_i^{z'}\cos\varphi - S_i^{x'}\sin\varphi. \end{aligned} \tag{2}$$

Now, defining the raising and lowering spin operators

$$S_j^{\pm} = S_j^{x'} \pm \mathrm{i}\, S_j^{y'} \tag{3}$$

introducing the Holstein–Primakoff representation

$$\begin{aligned} S_j^{z'} &= S - a_j^+ a_j \\ S_j^+ &= (2S)^{1/2} f_j(S) a_j \\ S_j^- &= (2S)^{1/2} a_j^+ f_j(S) \end{aligned} \tag{4}$$

and taking the Fourier transform of the creation and destruction operators of spin deviations a_j^+ and a_j, the following expression for the Hamiltonian up to terms of order S^{-1} is obtained:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_2 + \mathcal{H}_4 \tag{5}$$

where

$$\mathcal{H}_0 = \tfrac{1}{2}NzS^2J + \tfrac{1}{2}NDS^2\cos^2\varphi - NSgu_{\mathrm{B}}\cos(\theta-\varphi) \tag{6}$$

$$\begin{aligned} \mathcal{H}_2 = \sum_k \{&[zSJ(v_k - 1) + (2S-1)D(\tfrac{3}{2}\sin^2\varphi - 1) + gu_{\mathrm{B}}H\cos(\theta-\varphi)]a_k^+ a_k \\ &+ \tfrac{1}{4}(2S-\tfrac{1}{2})D\sin^2\varphi(a_k a_{-k} + a_k^+ a_{-k}^+)\} \end{aligned} \tag{7}$$

$$\begin{aligned} \mathcal{H}_4 = -\frac{1}{4SN}\sum_{k,r,s} \{&[zSJ(v_s + v_{s+r-k} - 2v_{k-r}) + 4SD(\tfrac{3}{2}\sin^2\varphi - 1)]a_{s+r-k}^+ a_k^+ a_r a_s \\ &+ SD\sin^2\varphi(a_{k+r+s}^+ a_k a_r a_s + a_{s-r-k}^+ a_k^+ a_r^+ a_s)\}. \end{aligned} \tag{8}$$

In these equations, N is the number of lattice points and

$$v_k = \frac{1}{z}\sum_d \exp(-\mathrm{i}\,\boldsymbol{k}\cdot\boldsymbol{d}) \tag{9}$$

is the structure factor for the z nearest neighbours on a simple cubic lattice.

We can determine the equilibrium positions of the spins by minimising $\mathscr{H}_0$ with respect to the angle φ. We then obtain

$$(SD/gu_{\mathrm{B}}H)\sin(2\varphi) + \sin(\theta - \varphi) = 0. \tag{10}$$

Only if $SD/gu_{\mathrm{B}}H \ll 1$, we will have $\varphi = \theta$. In this approximation $\mathscr{H}_0$ represents the classical ground-state energy of an antiferromagnet in its paramagnetic phase. The spin-wave excitations will then be generated from the deviations around the equilibrium positions of the spins, given by the angle φ in (10). Within this approximation it is easy to show that the terms with one and three spin-wave operators, which appear due to the rotation of the spin axes, are eliminated from the spin-wave Hamiltonian.

The magnon energy spectrum can be determined by solving the equations of motion for the Green functions (Zubarev 1960) $\langle\langle a_k; a_r^+\rangle\rangle$ and $\langle\langle a_{-k}^+; a_r^+\rangle\rangle$, where $\boldsymbol{k}$ and $\boldsymbol{r}$ are vectors of the first Brillouin zone. These equations have been decoupled by the application of Wick's theorem (Tyablikov 1967) to the four point averages. Other approximations could be used to break the chain of Green functions like the 'Callen decoupling' (Anderson and Callen 1964) but the problem would become very difficult to handle and the results in the very low temperature region are essentially the same as in the random phase approximation which is used here. We then have

$$\langle\langle a_k; a_r^+\rangle\rangle = (\delta_{k,r}/2\pi)[(E + A_k)/(E^2 - E_k^2)] \tag{11a}$$

$$\langle\langle a_{-k}^+; a_r^+\rangle\rangle = -(\delta_{k,r}/2\pi)B_k/(E^2 - E_k^2) \tag{11b}$$

where the magnon energy spectrum is given by

$$E_k = (A_k^2 - B_k^2)^{1/2} \tag{12}$$

and

$$\begin{aligned} A_k = {} & zSJ(v_k - 1) + (2S-1)D(\tfrac{3}{2}\sin^2\varphi - 1) + gu_{\mathrm{B}}H\cos(\theta - \varphi) \\ & - \frac{1}{4SN}\sum_r \{6SD\sin^2\varphi\langle a_r a_{-r}\rangle + [4zSJ(v_k + v_r - v_{r-k} - 1) \\ & + 16SD(\tfrac{3}{2}\sin^2\varphi - 1)]\langle a_r^+ a_r\rangle\} \end{aligned} \tag{13}$$

$$\begin{aligned} B_k = {} & (S - \tfrac{1}{4})D\sin^2\varphi - \frac{1}{4SN}\sum_r \{6SD\sin^2\varphi\langle a_r^+ a_r\rangle \\ & + [2zSJ(v_k + v_r - 2v_{k+r}) + 8SD(\tfrac{3}{2}\sin^2\varphi - 1)]\langle a_r a_{-r}\rangle\}. \end{aligned} \tag{14}$$

We also have

$$\langle a_k^+ a_k\rangle = \tfrac{1}{2}(A_k/E_k - 1) + A_k m_k/E_k \tag{15}$$

and

$$\langle a_k a_{-k}\rangle = -(B_k/2E_k)(1 + 2m_k) \tag{16}$$

where

$$m_k = [\exp(E_k/k_{\mathrm{B}}T) - 1]^{-1} \tag{17}$$

is the magnon occupation number.

The magnitude of the canted-paramagnetic critical field $H_c(T, \theta)$ is detemined by the limit of stability of the paramagnetic phase, namely by the equation

$E_{k_0}(T, H, \theta) = 0$, where the vector $\boldsymbol{k}_0$ labels the corners of the first Brillouin zone. Neglecting small zero-point corrections (Cieplak 1977), the asymptotic form of the critical field at low temperatures is given by

$$H_c(T, \theta) = H_c(0, \theta) - \Delta H_c(T, \theta) \tag{18}$$

where

$$g u_B H_c(0, \theta) = [12SJ - (2S-1)D(\tfrac{3}{2}\sin^2\varphi - 1) - S(1 - 1/4S)D\sin^2\varphi]/\cos(\theta - \varphi) \tag{19}$$

and

$$\begin{aligned} g u_B \Delta H_c(T, \theta) = {} & (3\xi(2)/2^{3/2}\pi^2) \\ & \times \{[6J - D(2\sin^2\varphi - 1)]/[-(1 - 1/4S)(D/J)\sin^2\varphi]^{1/2}\} \\ & \times [1/\cos(\theta - \varphi)](k_B T/SJ)^2 \end{aligned} \tag{20}$$

where we have put $z = 6$ for a simple cubic lattice and $\xi(2) = \Sigma_{n=1}^{\infty} n^{-2}$. As we can see, the paramagnetic critical field varies asymptotically as T^2, if the angle between the external magnetic field and the easy axis is other than zero. In the case where θ is equal to zero, the magnon energy spectrum becomes quadratic around the corners of the Brillouin zone, and we obtain the well known result for the critical field (Feder and Pytte 1968)

$$g u_B H_c(T) = 12SJ - (1 - 2S)D - [\Gamma(\tfrac{3}{2})\,\xi(\tfrac{3}{2})/\pi^2](6J + D)(k_B T/SJ)^{3/2}. \tag{21}$$

3. Discussion and conclusions

The magnon energy spectrum for non-interacting spin-waves near the corners of the first Brillouin zone is given by

$$E_q = (\tfrac{1}{6}zSJ\,q^2 + 2S|D|(1 - 1/4S)\sin^2\varphi)^{1/2}(\tfrac{1}{6}zSJ)^{1/2}q \tag{22}$$

where the vector $\boldsymbol{q}$ is measured from the corners of the Brillouin zone. We can consider the energy spectrum to be quadratic if

$$\tfrac{1}{6}zSJ\,q^2 \gg 2S|D|(1 - 1/4S)\sin^2\varphi \tag{23}$$

that is

$$\sin^2(\varphi(\theta)) \ll E_q/2S(1 - 1/4S)|D| \tag{24}$$

where φ as a function of θ is given by (10).

It is worth mentioning that a quadratic spectrum gives a $T^{3/2}$ dependence for the critical field, while a linear spectrum gives a T^2 dependence (Figueiredo 1984). In this way, a $T^{3/2}$ dependence can be observed for angles between the magnetic field and the easy axis which satisfy the inequality (24). In order to compare this result with experimental data we must relate the energy spectrum with temperature. Considering that to excite a magnon of energy E_q we must have a thermal energy of order $k_B T$ (that is, $E_q \simeq k_B T$), we can write the inequality (24) in the following form:

$$\sin(\varphi(\theta)) \ll [k_B T/2S(1 - 1/4S)|D|]^{1/2}. \tag{25}$$

If $D \rightarrow 0$, we can note that the inequality (25) is satisfied by all the angles between 0° and

90°. In this case the paramagnetic critical field always exhibits a $T^{3/2}$ dependence. This result was to be expected on physical grounds because an isotropic antiferromagnet does not present any privileged direction.

We now apply the former inequality to $NiCl_2 \cdot 6H_2O$, which is a single-ion uniaxial antiferromagnet. For this crystal in its paramagnetic phase at low temperature, we always have $S|D|/gu_BH < 0.07$, and (10) gives $\varphi \leqslant \theta$. Therefore, the inequality (25) applied to $NiCl_2 \cdot 6H_2O$, becomes

$$\theta \leqslant \sin^{-1}(T/2.42)^{1/2}. \tag{26}$$

For this antiferromagnet the observed dependence of the critical field on temperature is of the form $T^{3/2}$ in the accessible low-temperature range, that is, T larger than 0.3 K (Oliveira Jr *et al* 1978). For this temperature, the inequality (26) gives $\theta \leqslant 20°$. Even if the alignment between the external field and the easy axis is not perfect, we expect a behaviour of the type $T^{3/2}$ for temperatures larger than 0.3 K. On the other hand, if new measurements are performed on this antiferromagnet at very low temperatures, for instance, $T < 0.1$ K ($T = 0.1$ K, $\theta \leqslant 11°$) the $T^{3/2}$ law can be observed only for an almost perfect alignment of the field with the easy axis. If this perfect alignment is not obtained, we expect a T^2-dependence for $T < 0.1$ K.

We conclude by saying that if θ is other than zero, the canted-paramagnetic critical field behaves asymptotically according to a T^2 law for uniaxial antiferromagnets. We know that the alignment of the field with the easy axis is of fundamental importance in order to observe the spin-flop transition. Here, we also note that the determination of the canted-paramagnetic boundary as a function of the temperature depends on this same alignment. The inequality (25) serves as our guide in this case because it relates the magnitude of the anisotropy, the absolute temperature and the angle θ between the external magnetic field and the easy magnetic axis. As far as this author knows the measurements on the paramagnetic phase boundaries of antiferromagnets are usually made with the field parallel or perpendicular to the easy crystalline axis. See, for instance, the measurements performed on $NiCl_2 \cdot 4H_2O$ (Becerra *et al* 1988), on $MnCl_2 \cdot 4H_2O$ (Rives and Benedict 1975) and on $CoCl_2 \cdot 6H_2O$ (Rives and Bathia 1975). It would be interesting to do new measurements on the canted-paramagnetic phase boundary of anisotropic antiferromagnets, now as a function of the temperature and of the angle θ, in order to test the validity of these arguments.

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