QUANTITATIVE DESCRIPTION OF THERMODYNAMICS OF LAYERED MAGNETS IN A WIDE TEMPERATURE REGION

V.Yu. Irkhin[†] and A.A. Katanin
Institute of Metal Physics, 620219 Ekaterinburg, Russia

The thermodynamics of layered antiferro- and ferromagnets with a weak interlayer coupling and/or easy-axis anisotropy is investigated. A crossover from an isotropic 2D-like to 3D Heisenberg (or 2D Ising) regime is discussed within the renormalization group (RG) analysis. Analytical results for the (sublattice) magnetization and the ordering temperature are derived are obtained in different regimes. Numerical calculations on the base of the equations obtained yield a good agreement with experimental data on La$_2$CuO$_4$ and layered perovskites. Corresponding results for the Kosterlitz-Thouless and Curie (Néel) temperatures in the case of the easy-plane anisotropy are derived.

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The problem of layered magnetic systems is of interest both from theoretical and practical point of view [1]. Here belong, e.g., quasi-two-dimensional (quasi-2D) perovskites, ferromagnetic monolayers and ultrathin films. We consider the Heisenberg model with small parameters of the interlayer coupling $\alpha = J'/J$ and easy-axis anisotropy $\eta = 1 - J^z/J^{x,y}$ ($J$ is the in-plane exchange parameter). This case permits a regular consideration since the magnetic transition temperature is small, $T_M \ll |J|S^2$, which is reminiscent of weak itinerant magnets.

At not too low temperatures $T$ the standard spin-wave theory (SWT) is insufficient to describe correctly thermodynamics of such systems. Somewhat better results can be obtained within the self-consistent theory (SSWT) [2, 3], which takes into account the temperature renormalization of $\alpha$ and $\eta$. However, the values of the ordering temperature in SSWT are still too high in comparison with experimental ones, and the critical behavior is quite incorrect.

To improve radically SSWT, the summation of leading contributions in all orders of perturbation theory should be performed. To this end we use the renormalization group (RG) approach similar to that of Ref. [4]. This approach is valid outside the critical region which is very narrow for layered systems. The same results can be obtained by direct summation of RPA-type corrections to spin-wave interaction vertex.

The result for the relative (sublattice) magnetization $\sigma_r \equiv \bar{S}/S(T = 0)$ reads [3, 5]

$$\sigma_r = 1 - \frac{T}{4\pi \rho_s} \left[ \ln \left( \frac{\Gamma(T)}{\Delta(f_r, \alpha_r)} \right) + 2 \ln(1/\sigma_r) + 2(1 - \sigma_r) + \Phi \left( \frac{T}{4\pi \rho_s \sigma_r} \right) \right]$$  \hspace{1cm} (1)

with $\Delta(f, \alpha) = f + \alpha + \sqrt{f^2 + 2\alpha f}$, $f = 4\eta$, other quantities are given in the Table 1:

| AFM, quantum regime ($T \ll |J|S$) | $T^2/c^2$ | $\gamma |J|SS_0$ | $f_{S_0}/S^2$ | $\alpha S_0/S$ |
|-----------------------------------|-----------|-----------------|-----------------|-----------------|
| FM, quantum regime ($T \ll JS$)   | $T/JS$    | $JS^2$          | $f$             | $\alpha$       |
| FM, AFM, classical regime ($T \gg |J|S$) | $32$      | $JS^2Z_{L_1}$  | $fZ_{L_2}$     | $\alpha Z_{L_3}$ |

†Corresponding author. Fax: +7 (3432) 74 52 44; e-mail: Valentin.Irkhin@imp.uran.ru
where \( c = \sqrt{8|J|\gamma S} \) is the spin-wave velocity, \( \gamma \simeq 1 + 0.078/S \) is the renormalization parameter for intralayer coupling, \( Z_{L1} = Z_{L2} = Z_{L3} = 1 - T/8\pi|J|S^2 \). The inequality \( \Gamma \gg \Delta \) should to be satisfied for the validity of (1). Note that the classical regime is realized only for very large \( S \). The quantities \( f_T \) and \( \alpha_T \) in (1) are the temperature-renormalized values of interlayer coupling and anisotropy parameters, and for \( T \ll 4\pi\rho_s\sigma_r \) (i.e. beyond the critical region, see below)

\[
f_T/f_r = (\alpha_T/\alpha_r)^2 = \sigma_r^2 \tag{2}
\]

Thus the anisotropy and interlayer coupling are strongly renormalized with temperature which should be taken into account when treating the experimental data. In the quantum regime, the parameters \( \alpha_r \) and \( f_r \) are ground-state (quantum-renormalized) anisotropy and interlayer coupling (for ferromagnetic case the ground-state renormalizations are absent). In the classical regime, \( f_r \) and \( \alpha_r \) are ‘lattice-renormalized’ parameters of anisotropy and interlayer coupling, i.e. the corresponding parameters of the continuum model with the same thermodynamic properties as the original lattice model.

Three temperature regimes can be distinguished.

(i) low temperatures, \( T \ll T_M \sim 2\pi|J|S^2/\ln(\Gamma/\Delta) \). The analysis of non-uniform susceptibility shows that the excitations in the whole Brillouin zone have spin-wave nature. Only first term in the square brackets in (1) is to be taken into account and the magnetization also demonstrates the spin-wave behavior.

(ii) intermediate temperatures, \( (\sqrt{S}/S)/\ln(\Gamma/\Delta) \ll T/2\pi|J|S^2 \ll \sqrt{S}/S (T \text{ is of the same order as } T_M) \). Close to the center of the Brillouin zone the excitations still have the spin-wave nature, while for large enough momenta they have non-spin-wave character. Only in-plane (two-dimensional) fluctuations are important in this regime. All the terms in (1), except for the last, are important, which leads to significant modification of the dependence \( \sigma_r(T) \) in comparison with SWT.

(iii) critical region, \( T/(2\pi|J|S^2) \gg \sqrt{S}/S \ (1 - T/T_M \ll 1) \). In this regime the spin-wave excitations are present only for momenta \( q^2 \ll \Delta \) (hydrodynamic region) whereas at all other \( q \) excitations have non-spin-wave character. The thermodynamics in this case is determined by 3D (or Ising-like) fluctuations. The contribution of \( \Phi_a \) is of the same order as other terms in the square brackets of (1) and the RG approach is unable to describe the thermodynamics in this regime. The values of critical exponents can be corrected in comparison with SSWT with the use of the \( 1/N \) expansion. For the isotropic quasi-2D antiferromagnet we obtain

\[
\sigma_r^2 = \left[ \frac{T_N}{4\pi|J|SS_0\gamma} \right]^{1-\beta_3} \left[ \frac{1}{1 - A_0 \left( 1 - \frac{T}{T_N} \right)} \right]^{2\beta_3} \tag{3}
\]

with \( A_0 \simeq 0.9635 \) and \( \beta_3 = (1 - 8/\pi^2N)/2 \simeq 0.36 \).

Up to some (unknown) constant \( C(f/\alpha) \) we have for the transition temperature the result

\[
1 = \frac{T_M}{4\pi\rho_s} \left[ \ln \frac{2\Gamma(T_M)}{\Delta(f_1, \alpha_1)} + 2\ln \frac{4\pi\rho_s}{T_M} + C(f/\alpha) \right]. \tag{4}
\]

Note that all the logarithmic terms are included in (1) and \( C \) gives only a small contribution to this result. For the quantum antiferromagnets the value of \( C(\infty) \) can be calculated by the \( 1/N \) expansion \( \Sigma \), \( C(\infty) \approx -0.0660 \). The value of \( C(0) \) for the same case can be deduced from experimental data on layered magnetic compounds \( \Sigma \), \( C(0) \approx -0.7 \).
For practical purposes, simple interpolation expressions for the functions \( \Phi(x) \), which permit to describe the crossover temperature region, are useful. We obtain at \( x < 1 \):

\[
\Phi(x)|_{\alpha=0} = \frac{x}{\sqrt{x^2 + 1}} [C(\infty) - 2 + 8 \ln 2],
\]

\[
\Phi(x)|_{f=0} = \frac{x}{\sqrt{x^2 + 1}} [C(0) - 1 + 3 \ln 3].
\]

(5)

Numerical calculations with the use of the equations obtained yield a good agreement with experimental data on the layered perovskites (see Figs. 1-2 and the Table 2), and the Monte-Carlo results for the anisotropic classical systems.

| Compound | \( T_{M}^{\text{SSWT}}, \text{K} \) | \( T_{M}^{\text{RG}}, \text{K} \) | \( T_{M}^{\exp}, \text{K} \) |
|----------|-------------------|-------------------|-------------------|
| \( \text{La}_2\text{CuO}_4 \) | 527 | 343 | 325 |
| \( \text{K}_2\text{NiF}_4 \) | 125 | 97.0 | 97.1 |
| \( \text{Rb}_2\text{NiF}_4 \) | 118 | 95.0 | 94.5 |
| \( \text{K}_2\text{MnF}_4 \) | 52.1 | 42.7 | 42.1 |
| \( \text{CrBr}_3 \) | 51.2 | 39.0 | 40.0 |

In the easy-plane case (\( \eta < 0 \)) finite-temperature magnetic transition is absent at \( \alpha = 0 \). At the same time, the Kosterlitz-Thouless transition where unbinding of topological excitations (vortex pairs) occurs. For small anisotropy value the temperature of this transition is small, and to leading logarithmic accuracy [7]

\[
T_{KT}^{(0)} = \frac{4\pi |J| S^2}{\ln |\pi^2/\eta|}
\]

which is similar to the result for \( T_M \) in the easy-axis case. As well as for magnets with small easy-axis anisotropy, topological excitations are important only close to \( T_{KT} \). Using the renormalization group approach, the result similar to (4) can be obtained [8]

\[
1 = \frac{T_{KT}}{4\pi \rho_s} \left[ \ln \left( \frac{\Gamma(T_{KT})}{|f_r|} \right) + 4 \ln \frac{4\pi \rho_s}{T_{KT}} + C' \right]
\]

(7)

For the magnetic ordering temperature in the presence of interlayer coupling we obtain

\[
1 = \frac{T_M}{4\pi \rho_s} \left[ \ln \left( \frac{\Gamma(T_M)}{|f_r|} \right) + 4 \ln \frac{4\pi \rho_s}{T_M} + C' - \frac{2A^2}{\ln^2(f/\alpha)} \right]
\]

(8)

The comparison of our results with the experimental data [1] is presented in the Table 3 (the parameters \( C' \approx -1.0 \) and \( A = 3.5 \) are fitted for the first compound).

| Compound | \( \text{K}_2\text{CuF}_4 \) | stage-2 NiCl_2 | \( \text{BaNi}_2(\text{PO}_4)_2 \) |
|----------|-------------------|--------------|-----------------|
| \( T_{M}^{(0)}, \text{K} \) | 11.4 | 35.3 | 45.0 |
| \( T_{M}^{\text{RG}}, T_{C}^{\text{RG}}, \text{K} \) | 5.5; 6.25 | 17.4; 18.7 | 23.0; 24.3 |
| \( T_{M}^{\exp}, T_{C}^{\exp}, \text{K} \) | 5.5; 6.25 | 18±20 | 23.0; 24.5 |

One can see that the two-loop corrections improve radically the result [3].

Figure captions

Fig.1. The theoretical temperature dependences of the relative sublattice magnetization \( \sigma_r \) from the spin-wave theory (SWT), SSWT, Tyablikov theory (TT), RG approach [1], and the experimental points for \( \text{La}_2\text{CuO}_4 \) [9]. The RG' curve corresponds to inclusion
of the function $\Phi_{AF}^a(t/\sigma)$ given by (3). The $1/N$ curve is the critical behavior predicted by $1/N$-expansion (3). The result of $1/N$-expansion at intermediate temperatures practically coincides with RG'.

Fig.2. Temperature dependence of the relative sublattice magnetization $\sigma(T)$ of $K_2NiF_4$ in the SWT, SSWT, RG approaches and $1/N$-expansion for $O(N)$ model as compared with the experimental data (circles). The RG' curve corresponds to inclusion of the function $\Phi_{AF}^a(t/\sigma)$ given by (3). Short-dashed line is the extrapolation of the $1/N$-expansion result to the critical region.

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