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Quantum magnets with large single-ion easy-plane anisotropy in magnetic field

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We propose a theory describing low-temperature properties of magnets with integer spin and large single-ion easy-plane anisotropy D in magnetic field H directed parallel to the hard axis. Considering the exchange interaction between spins as a perturbation and using the bosonic spin representation proposed in our recent paper [1] we find thermal corrections to the elementary excitation spectrum, magnetization and specific heat in the vicinity of the quantum critical point (QCP) $H = H_{c1}(0) \sim D$ in the first nonvanishing orders of the perturbation theory. An expression is found for the boundary of the paramagnetic phase $H_{c1}(T)$ in the H–T-plane. The effective interaction between bosons is derived near the QCP. The proposed theory describes well experimental data obtained in $NiCl_2$–$4SC(NH_2)_2$ (DTN).

Introduction. The topic of quantum criticality has received much attention in recent two decades. Of particular interest are quantum critical points (QCPs) which can be reached in experiments by varying easily controllable parameters such as external magnetic field, pressure, level of doping, etc. The equivalence between a spin system and a diluted gas of bosonic particles proved to be very useful in describing field-induced QCPs in magnets [2]. This equivalence is revealed and exploited using appropriate representation of spin operators via bosonic ones.

We discuss in the present paper properties of a system on a 3D lattice with an integer spin and large single-ion anisotropy which is described by the Hamiltonian

$$\mathcal{H} = D\sum_i (S_i^z)^2 + \frac{1}{2}\sum_{i,j} J_{i,j}\mathbf{S}_i\mathbf{S}_j + g\mu_{\rm B}H\sum_i S_i^z, \quad (1)$$

where g is the Lande factor and $D > 0$ is assumed to be much larger than exchange constants ($D \gg J$) so that the ground state at $H = 0$ is paramagnetic (all spins are mainly in the quantum state with $S^z = 0$). This system has at least two field-induced QCPs corresponding to transitions from the paramagnetic (at $H = H_{c1}(T = 0)$) and from the fully polarized (at $H = H_{c2}(T = 0)$) phases to other phases which nature depends on the details of the exchange coupling and the lattice geometry. We propose in our recent paper [1] a bosonic integer spin representation that is convenient for the paramagnetic phase discussion. Using this representation and considering the exchange interaction as a perturbation we find in Ref. [1] the spectrum of the Hamiltonian (1) in the paramagnetic phase at $H = 0$ in the third order in the perturbation theory (hereafter referred to as expansion in terms of J/D for shot).

We continue our study of the model (1) in the present paper and address its low-temperature properties in the vicinity of the QCP $H = H_{c1}(T = 0)$ using the proposed bosonic spin representation. Expressions are derived below in the first nonvanishing orders in J/D for thermal corrections to the elementary excitation spectrum, magnetization and specific heat. An expression is found for $H_{c1}(T)$ that is the boundary of the paramagnetic phase in the H–T-plane. The effective interaction is derived between bosons near the QCP which can be extracted from experiment. We demonstrate that the proposed theory describes well the corresponding experimental data obtained in $NiCl_2$–$4SC(NH_2)_2$ (DTN) [3–18] which is the most extensively studied compound of the type under discussion.

The magnetic subsystem of DTN consists of Ni ions with $S = 1$ and $g = 2.26$. Magnetic ions form a body-centered tetragonal lattice which can be viewed as two interpenetrating tetragonal sublattices. The exchange interaction between spins inside one sublattice is antiferromagnetic and strongly anisotropic: the exchange constant along the tetragonal hard axis (z-axis) is much larger than those along x- and y-axes. Then, DTN is a quasi-1D material having two QCPs at H equal to[2)]

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[2)] It should be noted that there is a certain discrepancy in values of $H_{c2}(0)$ in the experimental literature on DTN. Specific heat and magnetocaloric effect measurements give $H_{c2}(0) \approx 12.6$ T [9, 5]. On the other hand magnetization measurements [11] give the value of $H_{c2}(0)$ very close to Eq. (3) which was obtained in ac susceptibility measurements [7]. Besides, anomalies in the sound velocity

$$H_{c1}^{\mathrm{DTN}}(T=0)=2.05\,\mathrm{T}, \tag{2}$$
$$H_{c2}^{\mathrm{DTN}}(T=0)=12.175\,\mathrm{T} \tag{3}$$

with a canted antiferromagnetic phase between them. It was found [5, 7] that the QCP $H=H_{c1}(0)$ belongs to the 3D BEC universality class: $H_{c1}(T)-H_{c1}(0)\propto T^{\alpha}$ with $\alpha\approx 1.5$. The strength of the effective interaction between long-wavelength bosons was extracted in Ref. [8] from measurements of magnetization and $H_{c1}(T)$ (see below). The specific heat at small T was measured in Refs. [9] at $H\sim H_{c1}(0)$ and $H\sim H_{c2}(0)$.

Theoretical description of these experimental data was given using a number of self-consistent calculations (valid for $S=1$ only) with the following parameters of the Hamiltonian (1): $D=8.9\,\mathrm{K}$, $J_z=2.2\,\mathrm{K}$ and $J_{xy}=0.18\,\mathrm{K}$, where J_z is the exchange coupling constant along the chains, J_{xy} is that between chains inside one tetragonal sublattice and the interaction between the tetragonal sublattices is neglected. However results of ESR [18] and inelastic neutron scattering experiments [5] cannot be described by the Hamiltonian (1) with the conventional parameters (see discussions in Refs. [18, 19, 1]). Then, we propose in Ref. [1] the following set of parameters that differs from the conventional one using which we fit experimentally obtained spectrum and explain qualitatively the ESR results:

$$\begin{aligned} D&=7.72\ \mathrm{K},\\ J_z&=1.86\ \mathrm{K},\\ J_{xy}&=0.2\ \mathrm{K},\\ V&=0.1\ \mathrm{K}, \end{aligned} \tag{4}$$

where V is the exchange coupling constant between neighboring spins from different tetragonal sublattices which is introduced first in Ref. [19]. Then, it is tempting to reexamine the above mentioned experimental results for DTN at $T\neq 0$ using parameters (4) that is one of the aims of the present paper.

Method and technique. It is convenient to use the following spin representation of integer S which is proposed in our previous study [1]:

[16], the sound attenuation [16] and the thermal conductivity [9, 17] at a given $T<0.5$ K were observed at fields smaller than those in the specific heat [9]. The origin of this discrepancy is not discussed in the literature and we have no explanation for this situation either. We choose in our recent [1] and the present consideration of DTN the value (3) because it provides better agreement between our expressions for the spectrum found in the third order in J/D and the experimentally observed spectrum at $H=0$ [5] (see Ref. [1] for detail).

$$S_i^z=b_i^\dagger b_i-a_i^\dagger a_i, \tag{5}$$

$$\begin{aligned} S_i^+&=S_i^x+iS_i^y=b_i^\dagger\sqrt{\frac{(S-b_i^\dagger b_i)(S+1+b_i^\dagger b_i)}{1+b_i^\dagger b_i}}+\\ &+\sqrt{\frac{(S-a_i^\dagger a_i)(S+1+a_i^\dagger a_i)}{1+a_i^\dagger a_i}}a_i\approx\\ &\approx b_i^\dagger\left(c_1-c_2\,b_i^\dagger b_i\right)+\left(c_1-c_2\,a_i^\dagger a_i\right)a_i, \end{aligned} \tag{6}$$

where a_i and b_i are bosonic operators, $c_1=\sqrt{S(S+1)}$ and $c_2=\sqrt{S(S+1)}-\sqrt{(S-1)(S+2)/2}>0$. Representation (5), (6) reproduces the spin commutation relations on the physical subspace which is constrained by the following additional term in the Hamiltonian (see discussion in Ref. [1]):

$$\mathcal{H}_U=\frac{U}{N}\sum_i a_i^\dagger b_i^\dagger a_i b_i,\quad U\to+\infty, \tag{7}$$

where N is the number of unit cells. Substituting Eqs. (5), (6) into Eq. (1) and taking into account Eq. (7) one obtains for the Hamiltonian

$$\mathcal{H}=\sum_{\mathbf{p}}\left[\epsilon_{1a,h}(\mathbf{p})a_{\mathbf{p}}^\dagger a_{\mathbf{p}}+\epsilon_{1b,h}(\mathbf{p})b_{\mathbf{p}}^\dagger b_{\mathbf{p}}\right]+$$
$$+\sum_{\mathbf{p}}\frac{c_1^2}{2}J_{\mathbf{p}}\left(a_{\mathbf{p}}^\dagger b_{-\mathbf{p}}^\dagger+a_{\mathbf{p}}b_{-\mathbf{p}}\right)+ \tag{8a}$$
$$+\frac{1}{N}\sum_{\mathbf{p}_1+\mathbf{p}_2+\mathbf{p}_3+\mathbf{p}_4}\left\{\left[D+\frac{1}{2}J_{3-1}-\frac{c_1c_2}{2}(J_1+J_3)\right]\times\right.$$
$$\left.\times\,(a_1^\dagger a_2^\dagger a_3a_4+b_1^\dagger b_2^\dagger b_3b_4)+(U-J_{3-1})a_1^\dagger b_2^\dagger a_3b_4\right\}- \tag{8b}$$
$$-\frac{1}{N}\sum_{\mathbf{p}_1+\mathbf{p}_2+\mathbf{p}_3=\mathbf{p}_4}\frac{c_1c_2}{2}J_1\times$$
$$\times\left(b_1^\dagger a_2^\dagger a_3^\dagger a_4+a_1^\dagger b_2^\dagger b_3^\dagger b_4+a_4^\dagger a_3a_2b_1+b_4^\dagger b_3b_2a_1\right), \tag{8c}$$

where $J_{\mathbf{p}}=\sum\limits_j J_{ij}e^{i\mathbf{p}\mathbf{R}_{ij}}$,

$$\epsilon_{1a,h}(\mathbf{p})=D+\frac{c_1^2}{2}J_{\mathbf{p}}-h, \tag{9}$$

$$\epsilon_{1b,h}(\mathbf{p})=D+\frac{c_1^2}{2}J_{\mathbf{p}}+h, \tag{10}$$
$$h=g\mu_{\mathrm{B}}H$$

are spectra of a and b particles in the first order in J/D (here and below the number in the lower index of the spectrum indicates its order in J/D). It is convenient for the following to introduce three Green's functions

$$G_{a,h}(p)=-i\langle a_pa_p^\dagger\rangle, \tag{11a}$$
$$G_{b,h}(p)=-i\langle b_pb_p^\dagger\rangle, \tag{11b}$$
$$F_h(p)=-i\langle b_{-p}^\dagger a_p^\dagger\rangle, \tag{11c}$$

where $p = (\omega, \mathbf{p})$ and a_p is the Fourier transform of $a_{\mathbf{p}}(\tau)$. Dyson equations for one couple of these Green's functions have the form

$$G_{a,h}(p) = G_{0a,h}(p)\left[1 + \Sigma_{a,h}(p)G_{a,h}(p) + \Pi_h(p)F_h(p)\right],$$
$$F_h(p) = G_{0b,h}(-p)\left[\overline{\Pi}_h(p)G_{a,h}(p) + \Sigma_{b,h}(-p)F_h(p)\right], \quad (12)$$

where $G_{0a,h}(p) = [\omega - \epsilon_{1a,h}(\mathbf{p}] + i\delta)^{-1}$, $G_{0b,h}(p) = [\omega - \epsilon_{1b,h}(\mathbf{p}) + i\delta]^{-1}$, Σ and Π are normal and anomalous self-energy parts, respectively. Solving Eqs. (12) and the couple of equations for $G_{b,h}(p)$ and $F_h(p)$ one obtains

$$G_{a,h}(p) = \frac{\omega + \Sigma_{b,h}(-p)}{\mathcal{D}_h(p)}, \quad (13)$$

$$G_{b,h}(p) = \frac{\omega + \Sigma_{a,h}(-p)}{\mathcal{D}_h(-p)}, \quad (14)$$

$$F_h(p) = -\frac{\overline{\Pi}_h(p)}{\mathcal{D}_h(p)}, \quad (15)$$

$$\mathcal{D}_h(p) = \left[\omega - \epsilon_{1a,h}(\mathbf{p}) - \Sigma_{a,h}(p)\right] \times \quad (16)$$
$$\times \left[\omega + \epsilon_{1b,h}(\mathbf{p} + \Sigma_{b,h}(-p)\right] + |\Pi_h(p)|^2. \quad (17)$$

Spectra of a and b particles should be found from equations

$$\mathcal{D}_h[\epsilon_{a,h}(\mathbf{p}), \mathbf{p}] = 0, \qquad \mathcal{D}_h(-\epsilon_{b,h}(\mathbf{p}), \mathbf{p}) = 0. \quad (18)$$

***$T = 0$* and *$H = 0$*.** Because a and b particles are equivalent at $H = 0$ one has

$$G_{a,h=0}(p) = G_{b,h=0}(p) = G(p), \quad (19a)$$
$$\Sigma_{a,h=0}(p) = \Sigma_{b,h=0}(p) = \Sigma(p), \quad (19b)$$
$$\Pi_{h=0}(p) = \Pi(p), \quad (19c)$$
$$\epsilon_{a,h=0}(\mathbf{p}) = \epsilon_{b,h=0}(\mathbf{p}) = \epsilon(\mathbf{p}). \quad (19d)$$

We calculate in our previous paper [1] $\Sigma(p)$, $\Pi(\mathbf{p})$ and the spectrum $\epsilon(\mathbf{p})$ up to the third order in J/D.

***$T = 0$* and *$H \neq 0$*.** Taking into account that $[\mathcal{H}, \sum_i S_i^z] = 0$ and using Eq. (5) one concludes that h and $-h$ play the role of chemical potentials for a and b particles, respectively, so that

$$G_{a,h}(\omega, \mathbf{p}) = G(\omega + h, \mathbf{p}), \quad (20a)$$
$$G_{b,h}(\omega, \mathbf{p}) = G(\omega - h, \mathbf{p}), \quad (20b)$$
$$F_h(\omega, \mathbf{p}) = F(\omega + h, \mathbf{p}), \quad (20c)$$

and, correspondingly, $\Sigma_{a,h}(\omega, \mathbf{p}) = \Sigma(\omega + h, \mathbf{p})$, $\Sigma_{b,h}(\omega, \mathbf{p}) = \Sigma(\omega - h, \mathbf{p})$, $\Pi_h(\omega, \mathbf{p}) = \Pi(\omega + h, \mathbf{p})$,

$$\epsilon_{a,h}(\mathbf{p}) = \epsilon(\mathbf{p}) - h, \quad (21a)$$
$$\epsilon_{b,h}(\mathbf{p}) = \epsilon(\mathbf{p}) + h, \quad (21b)$$

where $\epsilon(\mathbf{p})$, $\Sigma(p)$, $\Pi(p)$, $G(p)$ and $F(p)$ are defined in Eqs. (19).

It is seen from Eq. (21a) that the spectrum of a particles has a gap which vanishes at $H = H_{c1}(0)$ and the spectrum becomes unstable at larger fields, where

$$g\mu_{\mathrm{B}} H_{c1}(T = 0) = \epsilon(\mathbf{p}_0) \quad (22)$$

and $\mathbf{p}_0$ is the momentum at which $\epsilon(\mathbf{p})$ has a minimum. This instability signifies a transition to another phase. One has $\mathbf{p}_0 = (\pi, \pi, \pi)$ in DTN because exchange couplings are antiferromagnetic.

One concludes from Eqs. (20), (21) that magnetic field lifts the equivalence between a and b particles. However magnetization $M(H, T = 0) = g\mu_{\mathrm{B}}\langle S_i^z\rangle$ remains zero in the paramagnetic phase as it can be readily seen from Eqs. (5), (20a) and (20b). Thermal fluctuations make finite the magnetization.

***$T \neq 0$* and *$H \sim H_{c1}(0)$*.** Let us consider the dispersion equation (18) for a particles. We have found its solution in our previous paper [1] at $T = 0$ up to the third order in J/D. The aim of the present discussion is to find temperature corrections to the spectrum $\delta_T\epsilon_{a,h}(\mathbf{p})$ in the first nonvanishing orders in J/D and T considering the temperature to be small enough $T \ll J$. Then, it is convenient to represent the spectrum and self-energy parts using Eqs. (20) in the following form:

$$\epsilon_{a,h}(\mathbf{p}) = \epsilon(\mathbf{p}) - h + \delta_T\epsilon_{a,h}(\mathbf{p}), \quad (23)$$

$$\Sigma_{a,h}[\epsilon_{a,H}(\mathbf{p}), \mathbf{p}] = \Sigma[\epsilon(\mathbf{p}), \mathbf{p}] + $$
$$+ \delta_T\epsilon_{a,h}(\mathbf{p}) \left.\frac{\partial\Sigma(\omega, \mathbf{p})}{\partial\omega}\right|_{\omega=\epsilon(\mathbf{p})} + \delta_T\Sigma_{a,h}[\epsilon(\mathbf{p}) - h, \mathbf{p}], \quad (24)$$

where $\delta_T\Sigma_{a,h}(\omega, \mathbf{p})$ is the temperature correction to $\Sigma_{a,h}(\omega, \mathbf{p})$. Expressions similar to Eq. (24) can be written for $\Sigma_{b,h}[-\epsilon_a(\mathbf{p}), \mathbf{p}]$ and $\Pi_h[\epsilon_a(\mathbf{p}), \mathbf{p}]$. Substituting these equations into Eq. (18) and using results of our previous calculation [1] of self-energy parts we have in the first order in J/D $\delta_T\epsilon_{a,\mathbf{p},h} = \delta_T\Sigma_{a,h}[\epsilon(\mathbf{p}) - h, \mathbf{p}]$ and $H_{c1}(T) = \epsilon(\mathbf{p}_0) + \delta_T\Sigma_a(0, \mathbf{k}_0)$. The first order correction in J/D to $\delta_T\Sigma_a\left[\epsilon(\mathbf{p}) - h, \mathbf{p}\right]$ is given by the Hartree–Fock diagram shown in Fig. 1a. As a result one obtains

$$\delta_T\epsilon_{a,h}(\mathbf{p}) = 4\Gamma_a\left[\epsilon(\mathbf{p}) - h, 0, \mathbf{p}\right]\frac{M(H,T)}{g\mu_{\mathrm{B}}}, \quad (25)$$

$$g\mu_{\mathrm{B}} H_{c1}(T) = \epsilon(\mathbf{p}_0) + 4\Gamma_a(0, 0, \mathbf{k}_0)\frac{M(H,T)}{g\mu_{\mathrm{B}}}, \quad (26)$$

where $\Gamma_a(\Omega, \mathbf{p}, \mathbf{q})$ is the vertex and

$$\frac{M(H,T)}{g\mu_B} = \frac{1}{N}\sum_{\mathbf{k}} N[\epsilon(\mathbf{k}) - h] \quad (27)$$

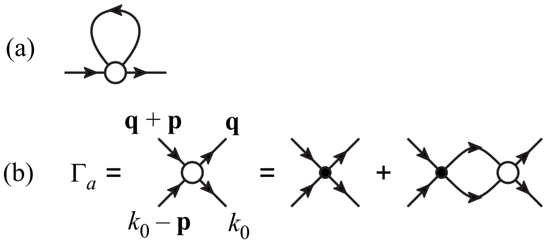

Fig. 1. (a) – The Hartree–Fock diagram giving the first order correction in J/D to the self-energy part. (b) – Diagram equation for the vertex $\Gamma_a(\omega, \mathbf{p}, \mathbf{q})$ which is involved in the Hartree–Fock diagram. Lines are Green's function (11a) of a particles. Black dots are bare vertexes given by term (8b) in the Hamiltonian

is equal to the magnetization in the second order in J/D at $T \ll \epsilon(0) + h$. It is explained in our previous paper [1] that ladder diagrams give the main contribution to the vertex leading to the Bethe–Salpeter equation for $\Gamma_a(\Omega, \mathbf{p}, \mathbf{q})$ that is shown in Fig. 1b. To calculate the vertex in the leading order in J/D one can use Green's function in the form $G_a(\omega, \mathbf{p}) = 1/[\omega - \epsilon_1(\mathbf{p}) + h + i\delta]$. When $\Omega \sim J$, the solution can be tried in the form $\Gamma_a(\Omega, \mathbf{p}, \mathbf{q}) = A(\Omega) + (J_\mathbf{p} - J_{\mathbf{p}+\mathbf{q}})/4 + B^z(\Omega) J^z_{\mathbf{p}+(\mathbf{q}-\mathbf{k}_0)/2} + B^{xy}(\Omega) J^{xy}_{\mathbf{p}+(\mathbf{q}-\mathbf{k}_0)/2}$. The solution is quite cumbersome and we do not present it here. We point out only that $\Gamma_a \sim J$ when $\Omega \sim J$. The value

$$v_0 = 2\Gamma_a(0, 0, \mathbf{k}_0) \tag{28}$$

is an effective two-particle interaction which can be found experimentally at small T as a slope of the plot of $H_{c1}(T)$ vs $M_c(T) = M[H = H_{c1}(T), T]$ (see Eqs. (26) and (27)).

The specific heat can be obtained using Eq. (1) and the formula $C(H, T) = d\langle\mathcal{H}\rangle/dT$ with the following result in the first order in J/D:

$$C(H,T) = \frac{d}{dT}\left\{\frac{1}{N}\sum_{\mathbf{k}}\left[\epsilon_1(\mathbf{k}) - h\right] N\left[\epsilon_3(\mathbf{k}) - h\right]\right\}, \tag{29}$$

where $\epsilon_1(\mathbf{k})$ and $\epsilon_3(\mathbf{k})$ are spectra at $T = 0$ in the first and in the third orders in J/D, respectively.

Application to DTN. Equations for $M(H, T)$, $H_{c1}(T)$ and $C(H, T)$ obtained above are applicable at $H \approx H_{c1}(0)$ only at small enough T. In the case of a quasi-1D system it usually means that the temperature cannot exceed the value of the exchange constant between spin chains ($\sim$ 0.2 K in DTN). At such T the quasi-1D system behaves like a 3D one and we obtain the proportionality to $T^{3/2}$ of $H_{c1}(T) - H_{c1}(0)$, $M_c(T) = M[H = H_{c1}(T), T]$ and $C[H = H_{c1}(T), T]$ expected for QCP of 3D BEC universality class.

This proportionality was really observed experimentally in DTN. Fig. 2 shows the experimental data for

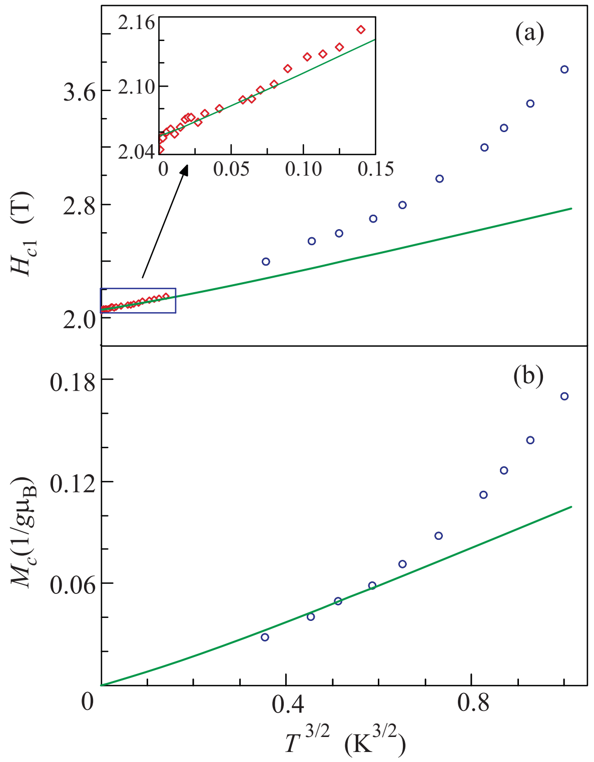

Fig. 2. Plots of (a) H_{c1} and (b) M_c (magnetization at $H = H_{c1}(T)$) vs $T^{3/2}$ in DTN. Circles and diamonds are experimental data from Ref. [8] and Ref. [7], respectively. Lines are drawn using Eqs. (26), (27) and parameters (4)

$M_c(T)$ and $H_{c1}(T)$ obtained in Ref. [8] and Ref. [7] for $0.5\,\mathrm{K} < T < 1\,\mathrm{K}$ and $1\,\mathrm{mK} < T < 300\,\mathrm{mK}$, respectively, together with results of our calculations with Eqs. (26), (27) and parameters (4). The agreement between the theory and experiment is very good at $T < 0.3$ K. Large temperature fluctuations come into play at greater T which are not taken into account in Eqs. (26) and (27). As a result the deviation from experimental data is noticeable at $T > 0.5$ K for $h_{c1}(T)$ and at $T > 0.7$ K for $M_c(T)$.

Although (and quite expectedly) neither $H_{c1}(T)$ nor $M_c(T)$ do not depend linearly on $T^{3/2}$ at $T > 0.7$ K in DTN (see Fig. 2), it is observed experimentally [8] that $H_{c1}(T)$ is a linear function of $M_c(T)$ at $0.5\,\mathrm{K} < T < 1\,\mathrm{K}$ as is demonstrated in Fig. 3. The effective two-particle interaction is extracted from this plot in Ref. [8] as it is explained above with the result $v_0 \approx 0.61$ meV. However, most likely that v_0 is renormalized by thermal fluctuations in DTN at such large T. We plot in Fig. 3 also $H_{c1}(T)$ vs $M_c(T)$ using the low-temperature experimental data of Ref. [7] for $H_{c1}(T)$ and results of our compu-

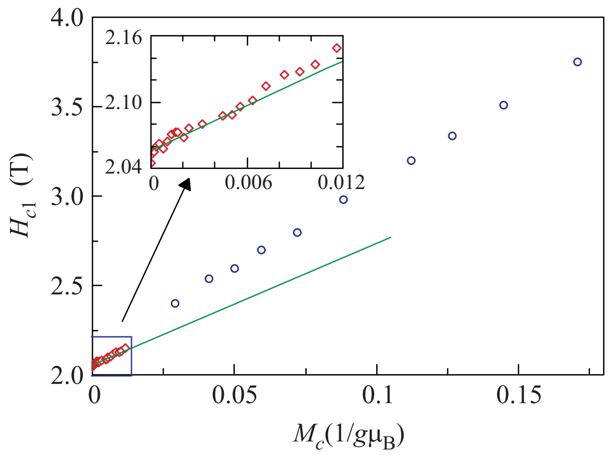

Fig. 3. $H_{c1}(T)$ versus $M_c(T)$ in DTN. Circles are experimental data from Ref. [8]. Diamonds correspond to experimental data of Ref. [7] for $H_{c1}(T)$ and results of our computation of $M_c(T)$ by Eq. (27) with parameters (4). Lines are drawn using Eqs. (26), (27) and parameters (4)

tation of $M_c(T)$ by Eq. (27) with parameters (4). The effective interaction obtained in this way is equal approximately to 0.44 meV that is 28% smaller than the value experimentally found in Ref. [8] and that is in excellent agreement with the result of our calculation of v_0 by Eq. (28) and parameters (4).

Experimental data of Ref. [9] for $C(H,T)$ at $H \approx H_{c1}(0)$ are shown in Fig. 4 together with results of our

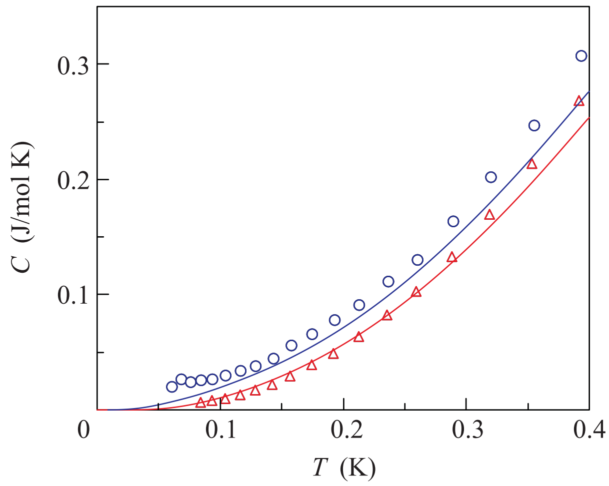

Fig. 4. Specific heat in DTN. Experimental data are taken from Ref. [9]. Lines are drawn using Eq. (29) and parameters (4)

calculations with Eq. (29), parameters (4) and expression for $\epsilon_3(\mathbf{p})$ found in Ref. [1]. A reasonable agreement between the theory and experiment is seen at $T < 0.3$ K. The specific heat of the model (1) with parameters (4) is proportional to $T^{3/2}$ at $H = H_{c2}(T)$ as well because the spectrum in the fully polarized phase given exactly (at $T = 0$) by $\varepsilon(\mathbf{p}) = h - (2S-1)D - SJ_\mathbf{0} + SJ_\mathbf{p}$ is also quadratic near its minimum. It is demonstrated in Ref. [9] that due to the strong renormalization of the spectrum in the paramagnetic phase $C[H = H_{c2}(T), T]$ is about 6 times larger than $C[H = H_{c1}(T), T]$. Simple calculation of the specific heat with the spectrum $\varepsilon(\mathbf{p})$ and parameters (4) shows that in agreement with the experiment $C[H = H_{c2}(T), T]$ is approximately 5.7 times larger than $C(H = H_{c1}(T), T)$ given by Eq. (29).

To conclude, we develop a theory describing low-temperature properties of the model (1) with integer spin and large D in magnetic field. We find thermal corrections to the elementary excitation spectrum, magnetization and specific heat in the vicinity of the QCP $H = H_{c1}(0) \sim D/(g\mu_B)$ in the first nonvanishing orders in J/D. The proposed theory with parameters (4) suggested in Ref. [1] describes well low-temperature experimental data obtained in $NiCl_2$–$4SC(NH_2)_2$.

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