ISSN 1063-7761, Journal of Experimental and Theoretical Physics, 2007, Vol. 104, No. 5, pp. 751–757. © Pleiades Publishing, Inc., 2007.
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Dynamics of Paramagnets at Zero Temperature

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Received October 9, 2006

Abstract—A macroscopic theory of low-frequency excitations in nearly unstable exchange spin systems with the singlet ground state is developed. Examples of dynamics near the pressure- and magnetic-field-instability points are considered. The development of the theory for the case of the instability of the system with respect to the appearance of magnetization is complicated.

PACS numbers: 75.10.-b, 75.30.Ds, 76.20.+q

DOI: 10.1134/S1063776107050093

1. INTRODUCTION

Owing to developed quantum fluctuations, the paramagnetic state of a spin system can be observed at temperatures much lower than the characteristic parameters of the spin–spin interaction down to absolute zero. When the exchange effects are much larger than the relativistic effects, excitations in such (singlet) ground state have a certain spin S. In the exchange approximation, excitations with the certain spin are degenerate in the spin projection and, as a rule, have a finite energy for any quasimomentum. The external magnetic field leads to the Zeeman splitting of the excitation spectrum (for $S \neq 0$). When the magnetic field reaches the critical value, the energy of a single excitation vanishes at a certain quasimomentum. In higher fields, the singlet state becomes unstable and a certain spin-ordered state appears in dependence of the type of this softening mode.

The theory of triplet excitations in the singlet ground state of one-dimensional systems was developed by Affleck [1] on the basis of analysis of the semiclassical limit of the microscopic model (see also [2, 3]). For the cases, where the singlet ground state is close to instability at zero temperature, the macroscopic description of the low-frequency spin dynamics of paramagnets is possible without any model representations. In this work, such theory is developed for singlet, triplet, and quintuple excitations in paramagnets, where the relativistic effects are much smaller than the exchange effects. We use the Landau theory of the second-order phase transitions [4] and exchange symmetry representations [5]. The results correspond to the general properties of the singlet ground state of the spin system [6]. It is worth noting that the singlet state exists not only in paramagnets, but also in scalar magnetics [7]. The theory is directly applicable to such spin structures.

2. EXCHANGE APPROXIMATION

Excitations with $S = 0, 1, 2$, etc. correspond to the oscillations of the spin scalars $\eta^{(i)}$, spin vectors $\boldsymbol{\eta}^{(i)}$, symmetric second-rank spin tensors $\eta^{(i)}_{\alpha\beta}$ with zero trace, etc., respectively, which are transformed according to the irreducible representations of the crystal group G (the superscript i specifies the fields belonging to a given n dimensional representation). It is worth noting that there is a difficulty in the development of the macroscopic theory for the pseudovector representation of the group G for $S = 1$ (when the vector $\boldsymbol{\eta}$ is transformed as the magnetization $\mathbf{M}$, see the last section). This case is not considered in this work.

The spin-dynamics equations are derived using the variational principle. Let us consider the derivation of the spin-dynamics equations for $S = 0, 1$, and 2 and only one-dimensional representations of the group G.

The excitations for $S = 0$ correspond to oscillations of the spin scalar η. It can be both nonmagnetic (its sign does not change under time reversal) and magnetic (changes sign) quantities. The former case corresponds to the spin-isotropic, G-noninvariant part of the spin–spin correlation function $\eta \propto \delta_{\alpha\beta}\langle S_\alpha(t, \mathbf{r}_1)S_\beta(t, \mathbf{r}_2)\rangle$, where $S_\alpha(t, \mathbf{r})$ is the spin density operator. Averaging over high frequencies of spin motion is implied in the correlation function, so that a slow function of time remains. The latter case corresponds to the spin-isotropic part of the three-point correlation function $\eta \propto e_{\alpha\beta\gamma}\langle S_\alpha(t, \mathbf{r}_1)S_\beta(t, \mathbf{r}_2)S_\gamma(t, \mathbf{r}_3)\rangle$.

The Lagrangian of the scalar field has the form

$$L = \frac{I}{2}\dot{\eta}^2 - \frac{A}{2}\eta^2 - \frac{G_{ij}}{2}\partial_i\eta\partial_j\eta. \qquad (1)$$

The exchange constant A is positive ($\eta = 0$ corresponds to the ground state) and is assumed small. This smallness ensures the possibility of developing the macroscopic theory of low-frequency spin dynamics. The

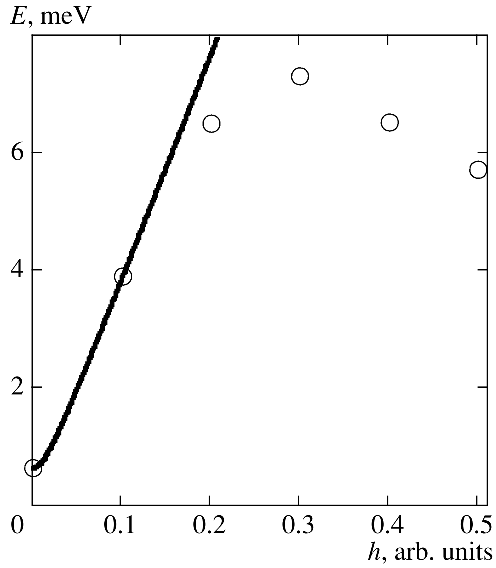

Fig. 1. Spin-excitation energy vs. the wave vector $q = (h, 0, 1)$ in the $TlCuCl_3$ paramagnetic crystal. The circles are the inelastic neutron scattering data [8] and the line is spectrum (3) with the parameters $\omega_0 = 0.16$ THz and $s = 0.5$ km/s.

exchange tensor G_{ij} determining the inhomogeneity energy has a natural order of magnitude and satisfies the stability condition for the homogeneous state (specifies a positively definite form). The tensor G_{ij} has the symmetry of the paramagnetic crystal class. To shorten expressions, the positive exchange constant I determining the kinetic energy is set to one in all below formulas.

Lagrangian (1) corresponds to the spin dynamics equations

$$\ddot{\eta} + A\eta - G_{ij}\partial_i\partial_j\eta = 0, \tag{2}$$

from which the magnon spectrum is obtained in the form

$$\omega(\mathbf{q}) = \sqrt{\omega_0^2 + s^2q^2}, \tag{3}$$

where $\omega_0 = \sqrt{A}$ is a small gap and s is the magnon velocity depending on the excitation propagation direction ($s^2 = G_{ij}q_iq_j/q^2$).

The gap in the magnon spectrum must change under pressure and can decrease to zero as the pressure increases to $P = P_c$. Under the assumption that $A = a(P_c - P)$, where $a > 0$ is a constant, near the critical point, the gap in the magnon spectrum is expressed as

$$\omega_0 = \sqrt{a(P_c - P)}, \quad P < P_c. \tag{4}$$

For $P > P_c$, the paramagnetic state becomes unstable with respect to the order parameter $\eta = \eta_0 \neq 0$.

When the field η is associated with the pair correlation function, a peculiar structure transition occurs and crystal-group elements responsible for change in the sign of the field η are lost (remember that only one-dimensional representations are considered). As a rule, the transition must be accompanied by change in the coordinates of nuclei corresponding to lowering of symmetry. However, the symmetry breaking can be manifested only in the electronic subsystem if no quantity that is transformed according to a representation of the group G corresponding to the field η can be composed from the displacements of the nuclei. When the field η is associated with the triple correlation function, the transition leads to the scalar-magnetic state [7].

Taking into account the fourth-order exchange invariant $B\eta^4/4$ in the potential energy (under the assumption that $B > 0$), near the critical point for $P > P_c$, we obtain

$$\eta_0^2 = -\frac{A}{B} = \frac{a(P - P_c)}{B}. \tag{5}$$

After the transition, the magnon spectrum holds form (3), but the gap is given by the expression

$$\omega_0 = \sqrt{2a(P - P_c)}, \quad P > P_c. \tag{6}$$

It is worth noting that the above description of low-frequency dynamics is applicable not only to spin excitations but also to any Bose degrees of freedom close to instability in condensed matter. The specificity of the spin dynamics is manifested for nonzero excitation spins, particularly in an external magnetic field.

The excitations for $S = 1$ are described by the vector $\boldsymbol{\eta}$ in the spin space. It can be either a magnetic quantity whose sign changes under time reversal (in this case, the vector $\boldsymbol{\eta}$ is related to the spin density as $\eta_\alpha \propto \langle S_\alpha(t, \mathbf{r})\rangle$) or a spin vector dual to the spin-antisymmetric part of the spin–spin correlation function $\eta_\alpha \propto e_{\alpha\beta\gamma}\langle S_\beta(t, \mathbf{r}_1)S_\gamma(t, \mathbf{r}_2)\rangle$ invariant under time reversal.

The Lagrangian of the vector field has the form

$$L = \frac{1}{2}\dot{\boldsymbol{\eta}}^2 - \frac{A}{2}\boldsymbol{\eta}^2 - \frac{G_{ij}}{2}\partial_i\boldsymbol{\eta}\partial_j\boldsymbol{\eta}. \tag{7}$$

The corresponding spin dynamics equations

$$\ddot{\boldsymbol{\eta}} + A\boldsymbol{\eta} - G_{ij}\partial_i\partial_j\boldsymbol{\eta} = 0 \tag{8}$$

provide the triply degenerate magnon spectrum of form (3). Such spectrum corresponds to the data reported in [8] on inelastic neutron scattering in $TlCuCl_3$ (see Fig. 1). The small ratio of the excitation-energy minimum (for $q = 0$) to the maximum (for $q \sim 0.3$ arb. units) is a main small parameter of the macroscopic theory. In $(CH_3)_2CHNH_3CuCl_3$ crystals, a quasi-one-dimensional exchange spin system with the singlet ground state is implemented. In this system, the triplet branch of magnetic excitations with a deep minimum in the wave vector at the edge of the Brillouin zone is found (see Fig. 2) [9]. It is worth noting that the macroscopic description of low-frequency spin dynamics is the same for both these cases.

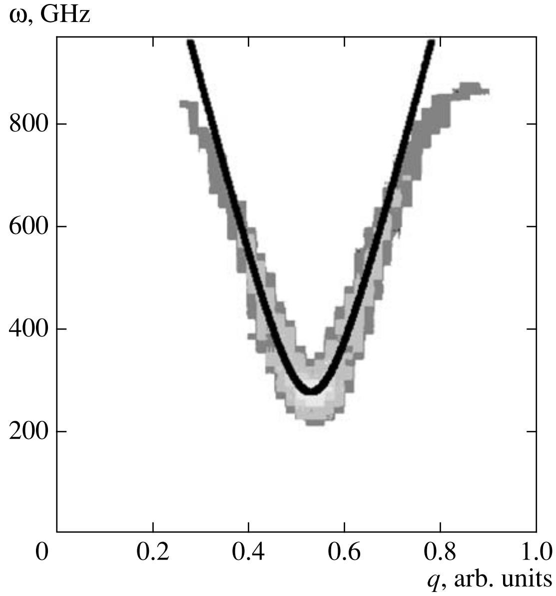

Fig. 2. Spin-excitation energy $E = \hbar\omega$ vs. the wave vector along spin chains in the $(CH_3)_2CHNH_3CuCl_3$ paramagnetic crystal. The points are the inelastic neutron scattering data [9] and the line is spectrum (3) where the wave vector is measured from the edge of the Brillouin zone with the parameters $\omega_0 = 0.28$ THz and $s = 0.41$ km/s.

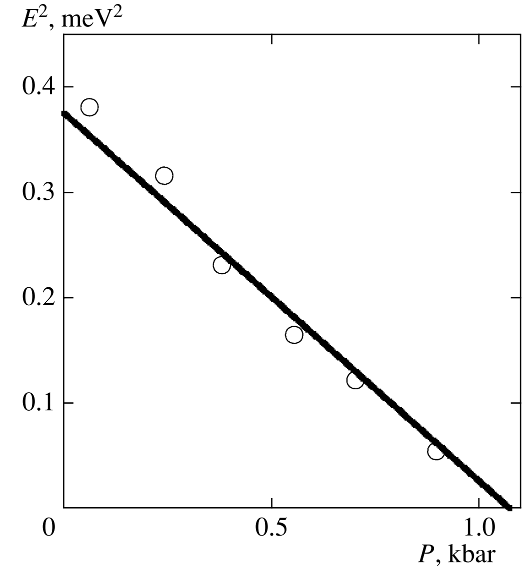

Fig. 3. Pressure dependence of the magnon-spectrum gap squared, $E^2 = a(P_c - P)$, in the $TlCuCl_3$ paramagnetic crystal according to data from [10] for the parameters P_c = 1.1 kbar and a = 0.35 meV^2/kbar.

The gap near the critical pressure is also given by Eq. (4). Such a simple law is in agreement with the experimental results for $TlCuCl_3$ (see Fig. 3) [10]. For $P > P_c$, the paramagnetic state becomes unstable with respect to the appearance of the antiferromagnetic (or nematic [12]) order parameter $\boldsymbol{\eta} = \boldsymbol{\eta}_0 \neq 0$. The absolute value $|\boldsymbol{\eta}_0|$ is also determined by Eq. (5), where B is the coefficient of the fourth-order exchange invariant $(\boldsymbol{\eta}^2)^2/4$ in the potential energy. After the transition, the triple degeneration is removed. A usual gapless doubly degenerate spectrum $\omega = sq$ of orientation oscillations of a collinear antiferromagnet (or a spin nematic with axial symmetry) appears along with the longitudinal oscillations of the antiferromagnetic vector given by Eq. (3) with the gap specified by Eq. (6).

The excitations of the system for $S = 2$ are described by the traceless symmetric spin tensor $\eta_{\alpha\beta}$. The Lagrangian has the form

$$L = \frac{1}{2}\dot{\eta}_{\alpha\beta}^2 - \frac{A}{2}\eta_{\alpha\beta}^2 - \frac{G_{ij}}{2}\partial_i\eta_{\alpha\beta}\partial_j\eta_{\alpha\beta}. \quad (9)$$

The corresponding spin dynamics equations

$$\ddot{\eta}_{\alpha\beta} + A\eta_{\alpha\beta} - G_{ij}\partial_i\partial_j\eta_{\alpha\beta} = 0 \quad (10)$$

provide the magnon spectrum of form (3) with quintuple $(2S + 1 = 5)$ degeneration.

When approaching the critical pressure, the gap goes to zero according to Eq. (4). If the tensor $\eta_{\alpha\beta}$ is transformed according to the unit representation of the group G, the cubic invariant $\eta_{\alpha\beta}\eta_{\beta\delta}\eta_{\delta\alpha}$ exists. As a result, the critical point is unattainable under the equilibrium conditions, because the first-order phase transition must occur earlier. For the one-dimensional nonunity representation, the behavior of the system for $P > P_c$ is somewhat more complicated than that for scalar and vector cases, because the expansion of the potential energy must include not only the fourth-order term but also the sixth-order exchange invariant [11]

$$\frac{B}{4}(\eta_{\alpha\beta}^2)^2 + \frac{C}{6}(\eta_{\alpha\beta}\eta_{\beta\delta}\eta_{\delta\alpha})^2. \quad (11)$$

If $C < 0$, the order parameter of the spin nematic appears [12] with the axial symmetry

$$\eta_{\alpha\beta} = \frac{\eta_0}{\sqrt{6}}(3c_\alpha c_\beta - \delta_{\alpha\beta}),$$

where $\mathbf{c}$ is the unit vector in the spin space. The absolute value of the order parameter, η_0, is determined by Eq. (5). In this case, the magnon spectrum contains the gapless doubly degenerate mode $\omega = sq$ of orientation oscillations of the vector c and three optical modes given by Eq. (3) with the gaps

$$\omega_0^{(1)} = \sqrt{2|A|} \propto \sqrt{P - P_c},$$

$$\omega_0^{(2)} = \omega_0^{(3)} = \frac{2|A|}{B}\sqrt{\frac{|C|}{3}} \propto P - P_c.$$

If $C > 0$, the tensor

$$\eta_{\alpha\beta} = \frac{\eta_0}{\sqrt{2}}(a_\alpha a_\beta - b_\alpha b_\beta) \tag{12}$$

appears, where **a** and **b** are the mutually orthogonal unit spin vectors. The absolute value of the order parameter η_0 is determined by Eq. (5). The spin symmetry [13] of the order parameter given by Eq. (12) is the group D_2^S. In this case, the magnon spectrum contains the gapless doubly degenerate mode $\omega = sq$ of orientation oscillations of the spin structure and two optical modes given by Eq. (3) with the gaps

$$\begin{aligned}\omega_0^{(1)} &= \sqrt{2|A|} \propto \sqrt{P - P_c},\\ \omega_0^{(2)} &= \frac{|A|}{B}\sqrt{\frac{C}{2}} \propto P - P_c.\end{aligned} \tag{13}$$

3. EXCHANGE DYNAMICS IN THE MAGNETIC FIELD

In the presence of the magnetic field lower than the characteristic exchange field, the Lagrangian of the scalar degree of freedom for $S = 0$ can be supplemented by the single exchange invariant $H^2\eta^2$. However, the appearance of such invariant contradicts the general requirement of the exchange approximation: the magnetization of the spin system $\mathbf{M} = \partial L/\partial \mathbf{H}$, where $\mathbf{H}$ is the magnetic field, and the mechanical spin moment $\mathbf{S} = \partial L/\partial \dot{\boldsymbol{\theta}}$, where $\dot{\boldsymbol{\theta}}$ is the spin-space rotation velocity, are related as $\mathbf{M} = \gamma\mathbf{S}$ (Larmor theorem), where γ is the gyromagnetic ratio for the free electron. Thus, the magnetic field does not affect the dynamics of the scalar spin field in the exchange approximation.

The invariants $(\dot{\boldsymbol{\eta}}[\boldsymbol{\eta} \times \mathbf{H}])$, $\boldsymbol{\eta}^2 H^2$, and $(\boldsymbol{\eta} \cdot \mathbf{H})^2$ can appear in the Lagrangian of the vector field $\boldsymbol{\eta}$ for $S = 1$ in the presence of the magnetic field. However, according to the Larmor theorem, appearing terms, along the kinetic energy, must be representable as $(\dot{\boldsymbol{\eta}} + \gamma[\boldsymbol{\eta} \times \mathbf{H}])^2/2$. Correspondingly, the spin dynamics equations for the vector field have the form

$$\begin{aligned}&\ddot{\boldsymbol{\eta}} + 2\gamma[\dot{\boldsymbol{\eta}} \times \mathbf{H}] + \gamma^2[\mathbf{H} \times [\boldsymbol{\eta} \times \mathbf{H}]]\\ &\quad + A\boldsymbol{\eta} - G_{ij}\partial_i\partial_j\boldsymbol{\eta} = 0.\end{aligned} \tag{14}$$

From this equation, the spectrum of the spin waves $\omega(\mathbf{q}, \mathbf{H})$ is obtained in the form

$$\omega = \sqrt{\omega_0^2 + s^2q^2} + \gamma S_{\mathbf{H}} H, \tag{15}$$

where the excitation spin projections $S_{\mathbf{H}} = 0$ and $S_{\mathbf{H}} = \pm 1$ onto the magnetic field direction correspond to the field oscillations $\boldsymbol{\eta}$ polarized along $\mathbf{H}$ and circularly polarized in the plane perpendicular to $\mathbf{H}$, respectively.

When the critical magnetic field $H_{c0} = \omega_0/\gamma$ is reached, the minimum frequency of frequencies (15) corresponding to $\gamma S_{\mathbf{H}} < 0$ vanishes and the $\boldsymbol{\eta} = 0$ state becomes unstable. For this reason, the next term of the expansion, $B(\boldsymbol{\eta}^2)^2/4$ (it is assumed that $B > 0$) should be taken into account in the Lagrangian. For fields $H > H_{c0}$, the homogeneous-state energy minimum

$$U_0 = -\frac{1}{2}\gamma^2[\boldsymbol{\eta} \times \mathbf{H}]^2 + \frac{A}{2}\boldsymbol{\eta}^2 + \frac{B}{4}(\boldsymbol{\eta}^2)^2 \tag{16}$$

is reached when the antiferromagnetic order parameter $\boldsymbol{\eta} = \boldsymbol{\eta}_0 \perp \mathbf{H}$, where $\boldsymbol{\eta}_0^2 = \gamma^2(H^2 - H_{c0}^2)/B$, appears.

The new term in the Lagrangian leads to the appearance of the nonlinear addition $B\boldsymbol{\eta}^2\boldsymbol{\eta}$ in spin dynamics equation (14). Linearizing the equation in the amplitude of small oscillations near the equilibrium position $\delta\boldsymbol{\eta} = \boldsymbol{\eta} - \boldsymbol{\eta}_0$, we determine the spectrum of spin waves in the antiferromagnetic state as $\omega_0 = \sqrt{\gamma^2H^2 + s^2q^2}$ for oscillations $\delta\boldsymbol{\eta} \parallel \mathbf{H}$ (the magnetic-resonance frequency $\omega = \gamma H$) and as [1]

$$\omega^{\pm} = \sqrt{\frac{\omega_\perp^2}{2} + s^2q^2 \pm \sqrt{\frac{\omega_\perp^4}{4} + 4\gamma^2H^2s^2q^2}} \tag{17}$$

for oscillations $\delta\boldsymbol{\eta} \perp \mathbf{H}$ with elliptic polarizations. Here, $\omega_\perp = \gamma\sqrt{2(3H^2 - H_{c0}^2)}$ is the frequency of the second magnetic resonance. In the approximation under consideration, the frequency ω_- vanishes at $q = 0$ for $H > H_{c0}$ (Goldstone mode, i.e., rotations of the spin structure about the magnetic field [14]). The observed splitting of the triplet gap in $TlCuCl_3$ [15, 16] and in $(CH_3)_2CHNH_3CuCl_3$ [9] is shown in Fig. 4 together with the obtained theoretical curves.

According to the Larmor theorem, the following terms appear in the Lagrangian of the tensor field $\eta_{\alpha\beta}$ for $S = 2$ in the presence of the magnetic field:

$$2\gamma e_{\alpha\beta\nu}\eta_{\alpha\mu}\dot{\eta}_{\beta\mu}H_\nu + 2(\gamma H_\alpha\eta_{\beta\mu})^2 - 3(\gamma H_\alpha\eta_{\alpha\beta})^2. \tag{18}$$

For fields $H < H_c = \sqrt{A}/2\gamma = \omega_0/2|\gamma|$, the magnon spectrum is given by general expression (15), where the projection $S_H = -2, -1, 0, 1$, or 2.

For fields higher than H_c, axial symmetry about the field direction is spontaneously broken and the order parameter given by Eq. (12) with the amplitude $\eta_0 = 2\gamma\sqrt{(H^2 - H_c^2)/B}$ appears. The vectors **a** and **b** lie in the plane perpendicular to the magnetic field. The magnon spectrum in this case has the Goldstone mode for rotations about the magnetic field and four optical modes with the gaps $\omega_0^{(1)} = \gamma H$, $\omega_0^{(2)} = 2\gamma H$, $\omega_0^{(3)} = 3\gamma H$, and $\omega_0^{(4)} = 2\gamma\sqrt{5H^2 - H_c^2}$.

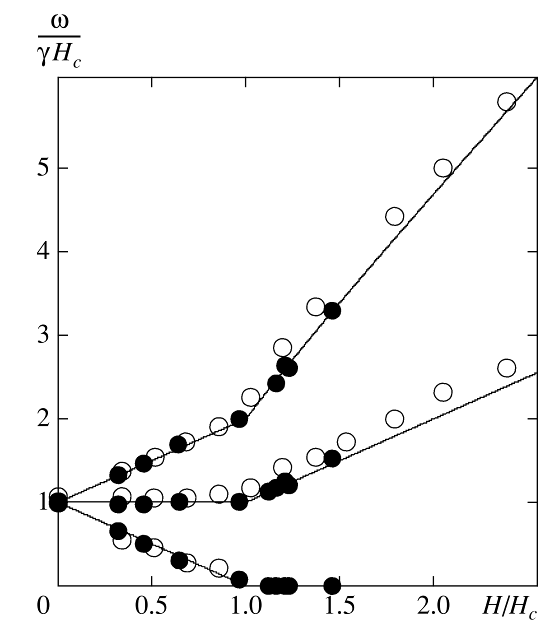

Fig. 4. Splitting of the triplet gap in the magnetic field for (open circles) $TlCuCl_3$ [15, 16] ($H_c = 5.9$ T) and (closed circles) $(CH_3)_2CHNH_3CuCl_3$ [9] ($H_c = 9.9$ T).

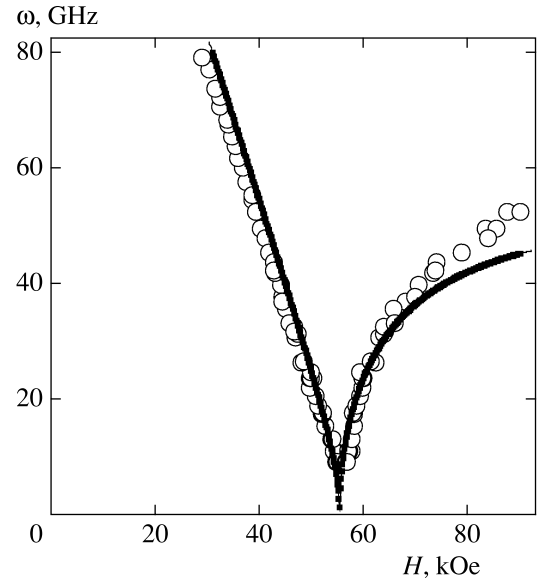

Fig. 5. Behavior of the low-frequency branch of the magnetic resonance in $TlCuCl_3$ near the critical magnetic field $\mathbf{H} \parallel C_2$.

4. RELATIVISTIC CORRECTIONS

The procedure for including of relativistic corrections will be considered on the example of $TlCuCl_3$ for which the detailed experimental investigations for the gap of the softening branch were performed [17] by the magnetic resonance method near the transition in the magnetic field.

The exchange symmetry of the field $\boldsymbol{\eta}$ is determined by the transformation properties under the permutations of atoms corresponding to the lattice symmetry elements [5]. The magnetic cell of the antiferromagnetic state of $TlCuCl_3$ coincides with the paramagnetic cell. For this reason, it is sufficient to specify only the action of the rotation elements of the C_{2h} crystal class. It follows from the data of the magnetic elastic scattering of neutrons [18] that $C_2\boldsymbol{\eta} = \boldsymbol{\eta}$ and $\sigma_h\boldsymbol{\eta} = -\boldsymbol{\eta}$. It is clear that the antiferromagnetic state has no Dzyaloshinskii invariants leading to weak ferromagnetism. In this case, the main quadratic relativistic contribution to the Lagrangian is reduced to anisotropy $(\beta_{\alpha\beta}/2)\eta_\alpha\eta_\beta$, where the tensor $\beta_{\alpha\beta}$ has the symmetry C_{2h}. Since the state is close to instability even in the absence of the field, the tensor components $\beta_{\alpha\beta}$ can be comparable with a small exchange constant A. To shorten the expressions, let us join the designations of the relativistic and exchange terms by setting $\beta_{\alpha\alpha} = A$. Since the length of the vector $\boldsymbol{\eta}$ is small, the relativistic effects can be disregarded in the fourth-order terms.

The tensor $\beta_{\alpha\beta}$ has the crystal symmetry C_{2h}. Let us direct the **z** axis along the second-order axis and the **x** and **y** axes along the principal axes of the tensor $\beta_{\alpha\beta}$ so that the inequality $\beta_{xx} < \beta_{yy}$ is satisfied. According to the experimental data [18], the easy magnetization axis lies in the (xy) plane at an angle of 13° to the [201] crystallographic direction. In our notation, it is the **x** axis.

For an arbitrary direction of the magnetic field, the very lengthy dispersion equations are obtained. We only present the analytical expressions for the frequencies of homogeneous oscillations for the case, where the field is directed along one of the proper axes of the anisotropy tensor, for example, the **z** axis. For fields lower than the critical value (for a given direction), the frequency of oscillations polarized along the field (η_z) is equal to $\omega_0 = \sqrt{\beta_{zz}}$. The frequencies of the elliptically polarized oscillations of the components η_x and η_y are determined by the equations

$$\omega_\pm^2 = \gamma^2 H^2 + \frac{\beta_{xx} + \beta_{yy}}{2} \pm \sqrt{\left(\frac{\beta_{xx} - \beta_{yy}}{2}\right)^2 + 2\gamma^2 H^2(\beta_{xx} + \beta_{yy})}. \tag{19}$$

The frequency ω_- vanishes for $H_c = \sqrt{\beta_{xx}}/\gamma$. The equilibrium antiferromagnetic vector components after the transition are $\eta_x = \gamma\sqrt{(H^2 - H_c^2)/B}$ and $\eta_y = \eta_z = 0$. In this case, the frequencies of small oscillations near the equilibrium state are equal to $\omega_0 = \sqrt{\beta_{zz} - \beta_{xx} + \gamma^2 H^2}$ for the component $\delta\eta_z$

$$\omega_\pm^2 = 3\gamma^2 H^2 - \frac{3\beta_{xx} - \beta_{yy}}{2}$$

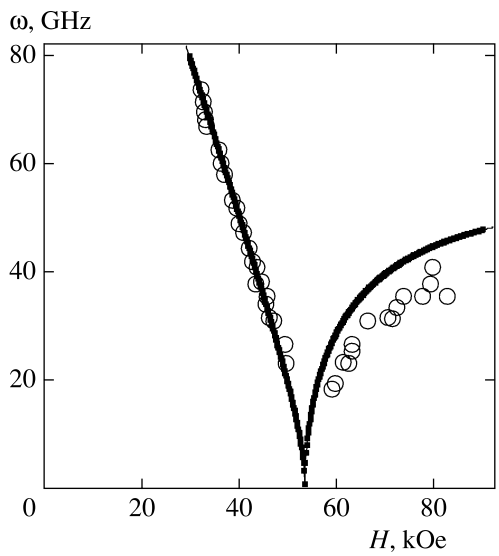

Fig. 6. Behavior of the low-frequency branch of the magnetic resonance in $TlCuCl_3$ near the critical magnetic field $\mathbf{H} \parallel [201]$.

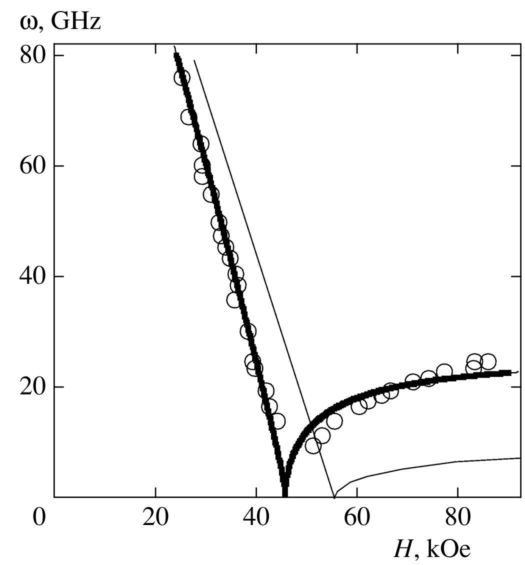

Fig. 7. Behavior of the low-frequency branch of the magnetic resonance in $TlCuCl_3$ near the critical magnetic field $\mathbf{H} \perp (10\bar{2})$. The thin curve is plotted by using only the parameters β_{xx}, β_{yy}, and β_{zz} obtained from a fit for two other directions of the magnetic field.

$$\pm\left\{\left(3\gamma^2H^2 - \frac{3\beta_{xx} - \beta_{yy}}{2}\right)^2 - 2(\beta_{yy} - \beta_{xx})(\gamma^2H^2 - \beta_{xx})\right\}^{1/2} \qquad (20)$$

for the elliptically polarized components $\delta\eta_x$ and $\delta\eta_y$. The frequencies ω_- given by Eqs. (19) and (20) with the parameters $\beta_{xx} = 3.3 \times 10^4$ GHz^2 and $\beta_{yy} = 2.4 \times 10^4$ GHz^2 correspond to the experimental data (Fig. 5).

Glazkov et al. [17] also studied the low-frequency magnetic resonance for two directions of the magnetic field: first, along the [201] crystallographic axis in the plane perpendicular to the C_2 axis and, second, perpendicular to the plane $(10\bar{2})$. The behavior of the frequency for $\mathbf{H} \parallel [201]$ is shown in Fig. 6. The agreement with the theoretical curves is ensured by the third parameter $\beta_{zz} = 2.3 \times 10^4$ GHz^2.

The description of the behavior of the frequency for the field direction $\mathbf{H} \perp (10\bar{2})$ appears to be impossible when the main anisotropy effects are taken into account. This means that the contribution from next corrections is anomalously large in this case. However, since the symmetry of the crystal is low, there are many such relativistic and exchange-relativistic terms $\dot{\eta}_\alpha\dot{\eta}_\beta$, $[\boldsymbol{\eta} \times \dot{\boldsymbol{\eta}}]_\alpha H_\beta$, $(\mathbf{H} \cdot \boldsymbol{\eta})H_\alpha\eta_\beta$, $H^2\eta_\alpha\eta_\beta$, and $\boldsymbol{\eta}^2 H_\alpha H_\beta$. For fitting, it is sufficient to take into account only one relativistic exchange term $(\tilde{\beta}_{xx}/2)H_x^2\boldsymbol{\eta}^2$, which does not affect the spectrum for the field direction $\mathbf{H} \parallel C_2$ and almost does not affect the spectrum for the field direction $\mathbf{H} \parallel [201]$. The theoretical curves in Fig. 7 are plotted for the fourth parameter $\tilde{\beta}_{xx} = -0.42$ (GHz/kOe)^2.

5. CONCLUSIONS

The theory developed in this work completely corresponds to the general properties of the spin structures with the singlet ground state at zero temperature [6]: Zeeman removal of the energy degeneration of spin multiplets (spin excitations with an arbitrary wavenumber) in the external field and the absence of the magnetic susceptibility up to the critical magnetic field (pressure). The low-frequency spectra of appearing (above the critical magnetic field) spin structures correspond to the spectra of antiferromagnets, spin nematics, or tensor magnetics. The macroscopic theory cannot answer the question of which excitations exist in a given singlet state and which of them become unstable.

As indicated above, the situation of the instability of the singlet state with respect to the appearance of the magnetization is a peculiar case. We could not find the Lagrangian formulation of the spin dynamics of the paramagnets that would lead to the Landau–Lifshitz equation for the low-frequency mode after the transition point. If a macroscopic spin degree of freedom with the magnetization symmetry exists in the system, the susceptibility in the singlet state at zero temperature must generally be nonzero. There is a precedent of such singlet state in microscopic theory. It is the one-dimensional Heisenberg model for the case of the antiferromagnetic sign of the exchange constant for the chain of spins $S = 1/2$. In this model, the magnon spectrum has zeros at the point $q = 0$ and at the edge of the Brillouin zone, as well as the linear dispersion same at both points. The nature of such spectrum is not yet understood in the framework of the macroscopic theory, because the spontaneous breaking of symmetry and, correspondingly, the Goldstone mode are absent in the system.

ACKNOWLEDGMENTS

We are grateful to A.F. Andreev, V.N. Glazkov, A.I. Zheludev, M.E. Zhitomirskiĭ, and A.I. Smirnov for stimulating discussions and valuable remarks and comments. This work was supported by the Russian Foundation for Basic Research (project nos. 04-02-17294, 06-02-16509, and 06-02-17281), the Landau Foundation (Forschungszentrum Jülich, Germany), and the Council of the President of the Russian Federation for Support of Young Scientists and Leading Scientific Schools.

REFERENCES

1. I. Affleck, Phys. Rev. B **41**, 6697 (1990); **43**, 3215 (1991).
2. M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist, Phys. Rev. B **69**, 054423 (2004).
3. A. K. Kolezhuk, V. N. Glazkov, H. Tanaka, and A. Oosawa, Phys. Rev. B **70**, 020403(R) (2004).
4. L. D. Landau and E. M. Lifshitz, *Course of Theoretical Physics*, Vol. 5: *Statistical Physics*, 4th ed. (Nauka, Moscow, 1995; Butterworth, London, 1999), Part 1.
5. A. F. Andreev and V. I. Marchenko, Usp. Fiz. Nauk **130**, 39 (1980) [Sov. Phys. Usp. **23**, 21 (1980)].
6. V. I. Marchenko, Pis'ma Zh. Éksp. Teor. Fiz. **59**, 590 (1994) [JETP Lett. **59**, 618 (1994)].
7. V. I. Marchenko, Pis'ma Zh. Éksp. Teor. Fiz. **48**, 387 (1988) [JETP Lett. **48**, 427 (1988)].
8. A. Oosawa, T. Kato, H. Tanaka, et al., Phys. Rev. B **65**, 094426 (2002).
9. V. O. Garlea, A. Zheludev, T. Masuda, et al., cond-mat/0608566.
10. Ch. Rüegg, A. Furrer, D. Sheptykov, et al., Phys. Rev. Lett. **93**, 257201 (2004).
11. V. Barzykin, L. P. Gorkov, and A. V. Sokol, Europhys. Lett. **15**, 869 (1991).
12. A. F. Andreev and I. A. Grishchuk, Zh. Éksp. Teor. Fiz. **84**, 87 (1984) [Sov. Phys. JETP **60**, 267 (1984)].
13. A. M. Farutin and V. I. Marchenko, Zh. Éksp. Teor. Fiz. **127**, 1106 (2005) [JETP **100**, 977 (2005)].
14. A. M. Farutin and V. I. Marchenko, Pis'ma Zh. Éksp. Teor. Fiz. **83**, 282 (2006) [JETP Lett. **83**, 238 (2006)].
15. Ch. Rüegg, N. Cavadini, A. Furrer, et al., Appl. Phys. A **74**, S840 (2002).
16. Ch. Rüegg, N. Cavadini, A. Furrer, et al., Nature **423**, 62 (2003).
17. V. N. Glazkov, A. I. Smirnov, H. Tanaka, and A. Oosawa, Phys. Rev. B **69**, 184410 (2004).
18. H. Tanaka, A. Oosawa, T. Kato, et al., J. Phys. Soc. Jpn. **70**, 939 (2001).

Translated by R. Tyapaev