Ising model in transverse magnetic field and the properties of singlet ferromagnets of the praseodymium type

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A perturbation theory for the calculation of the correlation and thermodynamic functions in the Ising model with a transverse magnetic field is developed for a large exchange interaction radius by using the diagram technique for spin operators.[8] The temperature dependence of the excitation spectrum, of the excitation damping, and of the heat capacity of the magnetic system are calculated in a broad temperature range both above and below the Curie temperature. It is shown that if only interaction between nearest neighbors is important then the temperature dependences of the various quantities should be correct to within a factor on the order of unity. A phase transition with respect to the coupling constant should occur in the Ising model with a transverse magnetic field at $T=0$. The singularities of the terms of the perturbation-theory series in $|\eta-1|$ are found to be logarithmic (η is a dimensionless coupling constant, the critical value of which is unity). The results of a number of experimental investigations[2,3] can be explained by the developed theory.

1. INTRODUCTION

Many presently-known systems can be described by an Ising model with a transverse magnetic field, i.e., by a Hamiltonian in the form

$$\mathscr{H}=-\sum_{i,j} V(\mathbf{r}_i-\mathbf{r}_j)S_i^xS_j^x-\Delta\sum_i S_i^z. \tag{1}$$

Included among these systems are, in particular, certain rare-earth metals and intermetallic compounds[1]. The most typical and well investigated substances of this class are Pr and Pr_3Tl, which we shall henceforth bear primarily in mind. In these magnets, the crystals of which have cubic symmetry, a strong crystal field splits the magnetic f level of the praseodynium atom, the total angular momentum of which is J = 4. The sublevel scheme cited, e.g., in the paper of Birgenau et al.[2] is shown in Fig. 1. The number at each sublevel is the multiplicity. The most essential fact in this scheme is that the lower sublevel is a singlet and therefore $\langle 1|J^\alpha|1\rangle = 0$. Here α = x, y, z, and $\langle 1|$ is the wave function of the singlet state. In one of the states of the first-excited triplet, the average of the projections of the angular momentum is also equal to zero. The wave function of this state will be designated $\langle 2|$. The only nonzero matrix element between the states $\langle 1|$ and $\langle 2|$ is $\langle 1|J^z|2\rangle = \langle 2|J^z|1\rangle = \alpha$. The Hamiltonian of the system, with allowance for the exchange interaction, is

$$\mathscr{H}=-\frac{1}{2}\sum_{i,j}\tilde{V}(\mathbf{r}_i-\mathbf{r}_j)\mathbf{J}_i\mathbf{J}_j-\sum_i V_{ci},$$

where V_{ei} is the crystal-field operator. As will be shown below, the energy of the exchange interaction and the splitting of the f-level of praseodymium are approximately equal.

We simplify the problem by retaining in the level scheme only the states $\langle 1|$ and $\langle 2|$. Following[1], we introduce Pauli matrices S that act on the wave functions of these states. Then the Hamiltonian of the system reduces to the form (1), where $V(\mathbf{r}_i - \mathbf{r}_j) = 1/2\alpha^2\tilde{V}(\mathbf{r}_i - \mathbf{r}_j)$ and Δ is the difference between the energies of our levels. For Pr we have Δ = 70°K. Our simplification of the level scheme should not alter the results significantly. This question was discussed in detail in Sec. 4.

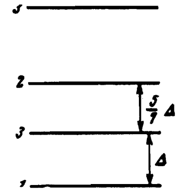

FIG. 1

Pr and Pr_3Tl go over into the ferromagnetic state at 20 and 11.6°K, respectively[2]. Their magnetic moment at saturation is small, one-quarter of the moment of the free Pr^{3+} ion per Pr atom[2,3]. Inelastic scattering of neutrons has shown[2] that the spectrum branch corresponding to spins waves at $T < T_C$ does not vanish even at $T > T_C$ (T_C is the Curie temperature). At large momenta the excitation energy turns out to be the same in the paramagnetic and ferromagnetic phases. We note also that there are substances that differ quite negligibly from Pr with respect to all the essential parameters, including the magnitude of the exchange interaction, but these substances do not go over into ferromagnetic states.

To understand these unusual properties of singlet magnets such as Pr, we assume that the exchange-interaction radius r_0 is large. Then we can obtain from (1) the macroscopic energy density

$$E=-V(0)M_x^2-\Delta M^z. \tag{2}$$

Here V(0) is the zeroth Fourier component of the potential V(r), and M is the density of the spin S. Minimizing the energy density from (2), we can easily find that at T = 0 in the ground state we have $M_x = 0$ if $\eta = \Delta/V(0) > 1$, and $M_x = 1/2n_0(1-\eta^2)^{1/2}$ if $\eta < 1$ (n_0 is the concentration of the magnetic atoms. Since the magnetic moment is $\langle J\rangle = \alpha\langle S^x\rangle$, it follows that a sufficiently strong exchange interaction causes a restructuring of the ground state and leads to the appearance of a magnetic moment, which can be called induced. A phase transition with respect to the coupling constant η takes place in the system at T = 0. An estimate of η for Pr and Pr_3Tl from the data on the saturation moment leads to a value $\eta^{-1} = 1.06$[2], which is close to the critical value. Therefore a slight change of the parameters causes the ferromagnetism to vanish even at T = 0.

The molecular-field method has been used for the Hamiltonian[1] for quite some time; a detailed description of this method can be found, e.g., in the book by Vaks[4]. Excitations in such a system can be regarded as low-energy Frenkel excitons; they exist, naturally, both above and below T_c. The excitation spectrum was obtained in a large number of papers[5, 6] with the aid of the Holstein-Primakoff transformation or methods equivalent to it. However, as noted in a number of the cited papers, these methods cannot be used in this case. The point is that the operators α_k and α_k^+, which diagonalize (1) are linear combinations of the Holstein-Primakoff operators

$$\alpha_k{}^+ = u_k a_k + v_k a_k{}^+.$$

Therefore

$$\gamma = \sum_k \langle a_k{}^+ a_k \rangle \neq 0$$

even at T = 0. If Δ and V(0) are of the same order of magnitude, then the quantity γ, and with it all the corrections to the spectrum obtained by transforming the operators, has no parametrically small quantity in the nearest-neighbor-interaction approximation. A similar situation obtains in antiferromagnets[7]. In both cases, the corrections at T = 0 are connected with the fact that the ground state of the system is not known. The "molecular field," i.e., the second term of (1), was chosen by a number of workers[5, 6] as the zeroth Hamiltonian, but it does not yield the correct ground state of the system even at T = 0, since neither S^z nor $M^z = \Sigma S_i^z$ commutes with $\mathscr{H}$. The lack of a consistent perturbation theory has not made it possible to determine the temperature dependence of the spectrum and of the thermodynamic quantities. For example, different decoupling methods lead to different temperature dependences[6]. Moreover, the resultant phase transition can be either of first or second order, depending on the method of decoupling in[6].

In this paper, using the diagram technique of Vaks, Larkin, and Pikin[8], a successive-approximation method is constructed for the calculation of the correlation and thermodynamic functions, assuming a large exchange-interaction radius. The development of a successive-approximation method is made possible by the fact that at a large interaction radius the ground state can be obtained, accurate to r_0^{-3}, from the macroscopic expression for the energy density. The corrections to the ground state obtained from (2) are proportional to different powers of r_0^{-3} and are small if r_0 is large enough.

The temperature dependence of the spectrum, the damping of the excitations, and the specific heat of the magnetic system are calculated both above and below T_c. The question of the phase transition with respect to η is examined. It turns out that the perturbation-theory terms have a logarithmic singularity in $|\eta - 1|$, unlike the power-law singularity in $\tau = (T - T_c)/T_c$, which is obtained for phase transitions at finite temperatures. The parameter of the successive-approximation method (we shall call this the molecular-field method) at T = 0 is $(a/r_0)^3 \ln|\eta - 1|$, where a is the lattice constant.

In Sec. 4 is discussed the applicability of the model to real singlet ferromagnets of the Pr type. It is shown that in spite of the fact that $(a/r_0)^3 \approx 1$ for these substances, the formulas derived in the paper are good estimates of the temperature dependence of the spectrum, of the damping, and of the specific heat at $|\tau| \gtrsim 1$. The results of the neutron-diffraction measurements[2, 3] are also explained.

In a recent paper, Stinchcombe[9] has also constructed a perturbation theory, assuming a large interaction radius. However, the temperature dependence of the spectrum was not calculated in[9]; only an integral equation for the excitation energy $\epsilon(q)$ was obtained, but without citing a solution. The exciton damping is calculated in[9] in first order in r_0^{-3}. At the same time, it is shown in the present paper that the contribution to the damping, due to the next order in r_0^{-3}, is larger than that calculated in[9] and has a different temperature dependence. In addition, the parameter of the perturbation theory in[9] is taken to be the reciprocal number of nearest neighbors, which is incorrect even far from the critical region of the phase transition. The phase transition with respect to η is not considered in[9].

2. EXCITATION SPECTRUM AND CORRELATION FUNCTIONS IN THE PARAMAGNETIC STATE

The Hamiltonian (1) can be transformed, by introducing the operators $S^\pm = 2^{-1/2}(S^x \pm iS^y)$, to the form

$$\mathscr{H} = -\frac{1}{2}\sum_{i,j} V(\mathbf{r}_{ij})\,(2S_i{}^+S_j{}^- + S_i{}^+S_j{}^+ + S_i{}^-S_j{}^-) - \Delta\sum_i S_i{}^z.$$

We introduce in the usual manner the temperature correlation function of the spins:

$$K^{\alpha\gamma}(k, i\omega_n) = \frac{1}{2}\int_{-\beta}^{\beta} e^{i\omega_n\tau}\,d\tau \sum_{r_2} e^{i\mathbf{k}(\mathbf{r}_1-\mathbf{r}_2)}\langle T(S_{r_2}{}^\alpha(\tau) - \langle S^\alpha\rangle),\ (S_{r_1}{}^\gamma(\tau) - \langle S^\gamma\rangle)\rangle. \tag{3}$$

Differentiating the expression for $K^{\alpha\gamma}(\mathbf{r}, \tau)$, which is obtained from (3) by a Fourier transformation, and replacing $\partial S/\partial\tau$ by $[\mathscr{H}, S]$, we obtain the following exact relation between the correlation functions:

$$(\Delta - i\omega)K^{+-}(k, i\omega) = (\Delta + i\omega)K^{--}(k, i\omega) + \langle S^z\rangle. \tag{4}$$

It is also seen from (3) that

$$K^{-+}(k, i\omega) = K^{+-}(k, -i\omega), \quad K^{--}(k, i\omega) = K^{++}(k, -i\omega).$$

To construct the perturbation theory, we choose $\mathscr{H}_0 = -\Delta\Sigma S_i^z$ as the zero-order Hamiltonian. The diagram technique of Vaks, Larkin, and Pikin[8] can then be used to calculate the correlation and thermodynamic functions. The rules for constructing the diagrams for the spin S = 1/2 consist in the following:

1) All the diagrams consist of single-cell blocks joined by interaction lines.

2) Each vertex S^+ of a single-cell block has one outgoing line, while the vertex S^- has either one incoming line or else two incoming lines and one outgoing line. The vertex S^z has either one incoming and one outgoing line, or else no lines at all.

3) Each line corresponds to a Green's function $G = (\Delta - i\omega_n)^{-1}$. If the diagram breaks up into N singly-connected diagrams, and the total number of vertices S^z and triple vertices S^- on the solid lines is equal to l, then the diagram has as a common factor $(-1)^l b^{N-1}$, where b^{N-1} is the (N−1)-st derivative of b = (1/2) tanh(Δ/2T) with respect to Δ/T.

The system of Dyson equations for the considered problem is of the form

$$K^{--} = \Sigma^{--} + V\Sigma^{--}(K^{--} + K^{+-}) + V\Sigma^{-+}(K^{--} + K^{+-}),$$
$$K^{+-} = \Sigma^{+-} + V\Sigma^{+-}(K^{--} + K^{+-}) + V\Sigma^{++}(K^{--} + K^{+-});$$

it follows therefore that

$$K^{--}=[\Sigma^{--}+V(\Sigma^{+-}\Sigma^{-+}-\Sigma^{--}\Sigma^{++})](1-2V\Sigma^{xx})^{-1},$$
$$K^{+-}=[\Sigma^{+-}+V(\Sigma^{--}\Sigma^{++}-\Sigma^{+-}\Sigma^{-+})](1-2V\Sigma^{xx})^{-1},$$
$$\Sigma^{xx}={}^1/_2(\Sigma^{++}+\Sigma^{--}+\Sigma^{+-}+\Sigma^{-+}).$$

Here $\Sigma^{\alpha\beta}$ is the aggregate of the irreducible diagrams.

Since the single-cell blocks with odd number of vertices S^+ and S^- and with one vertex S^z are equal to zero, we have in the paramagnetic phase

$$K^{z+}=K^{z-}=0,\quad K^{zz}=\Sigma^{zz}.$$

In the zeroth approximation of the molecular-field method we have

$$\Sigma_{(0)}^{++}=\Sigma_{(0)}^{--}=0,\quad \Sigma_{(0)}^{+-}(k,i\omega_n)=bG(i\omega_n),$$

from which it follows that

$$K_{(0)}^{+-}(k,i\omega_n)=b\frac{\Delta-bV(k)+i\omega_n}{\omega_n{}^2+\varepsilon_0{}^2(k)},$$
$$\varepsilon_0{}^2(k)=\Delta^2-2b\Delta V(k). \tag{5}$$

Just as in[8], in the calculation of $\Sigma^{\alpha\gamma}$ it is consistent to join the vertex with a "screened" interaction

$$\mathrm{V}(k,i\omega_n)=V(k)+V^2(k)(K_{(0)}^{++}+K_{(0)}^{--}+K_{(0)}^{+-}+K_{(0)}^{-+})=V(k)\frac{\Delta^2+\omega_n{}^2}{\omega_n{}^2+\varepsilon_0{}^2(k)}. \tag{6}$$

The interaction $\mathrm{V}(k, i\omega_n)$, unlike $V(k)$, can join vertices pertaining to the same node. It corresponds to a wavy line in the diagrams.

If $\eta > 1$, then, as seen from (5), we have $\epsilon_0^2(0) > 0$ at all temperatures, and consequently the system is paramagnetic at all temperatures, i.e., $\langle S^x\rangle = 0$. On the other hand, if $\eta < 1$, then at a certain temperature T_c we have $\epsilon_0^2(0) = 0$, and at $T < T_c$ we have $\epsilon_0^2(0) < 0$, thus indicating a phase transition in the system. The correlators $K^{\alpha\gamma}$ ($\alpha, \gamma = +, -$) near the transition have an Ornstein-Zernike form, and the corresponding susceptibilities have pole singularities in τ at $\eta < 1$ and in $(\eta - 1)$ at fixed T. Therefore the phase transitions, both with respect to temperature and with respect to the coupling constant (the latter will be considered at T = 0) are second-order transitions. This conclusion is confirmed also by the structure of the perturbation-theory series, which will be considered below.

The diagrams of first order in r_0^{-3} are obtained for $\Sigma^{\alpha\gamma}$ if the interaction $\mathrm{V}(k, i\omega_n)$ is used to join, by various methods, the vertices of the block Γ^{++--}, the diagrams for which are shown in Figs. 2a and 2b. For example, Σ^{--} (1) is determined by diagram 2c, and is equal to

$$\Sigma_{(1)}^{--}=-\frac{1}{2}bT\sum_{n,\mathbf{k}}G(i\omega_n)G(-i\omega_n)\mathrm{V}(k,i\omega_n)[G(i\omega)+G(-i\omega)]$$
$$=-\frac{b\Delta\Omega_0}{2(\omega^2+\Delta^2)}\int\frac{V(k)}{\varepsilon_0(k)}[1+2N(\varepsilon_0(k))]\frac{d^3\mathbf{k}}{(2\pi)^3}. \tag{7}$$

Here Ω_0 is the unit cell volume and $N(\epsilon) = (\epsilon^{\beta\epsilon} - 1)^{-1}$.

The temperature-dependent term in (7) is proportional to $T^{3/2}\exp[-\beta\epsilon_0(0)]$ if $\epsilon_0(0) > T$, and to T^2 if $\epsilon_0(0) < T$. At the same time, the contribution of the multiply-connected diagrams of the type 2b is proportional to $e^{-\beta\Delta}$. As already indicated in the introduction, we assume that $|1 - \eta| \ll 1$. Therefore $T_c \ll \Delta$ and in the temperature range $T < \Delta$, which is considered in this paper, we have

$$\varepsilon_0{}^2(0)=\Delta^2(\eta-1)+2\Delta^2\,\mathrm{ch}\left(\beta\frac{\Delta}{2}\right)\ll\Delta^2.$$

Thus, the contribution of the multiply-connected diagrams is exponentially small in comparison with the contribution of the singly-connected diagrams, and will be disregarded.

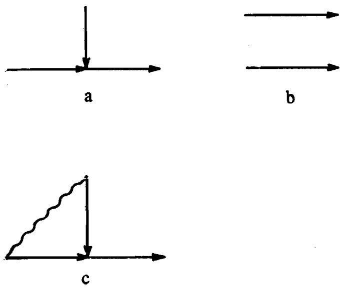

FIG. 2

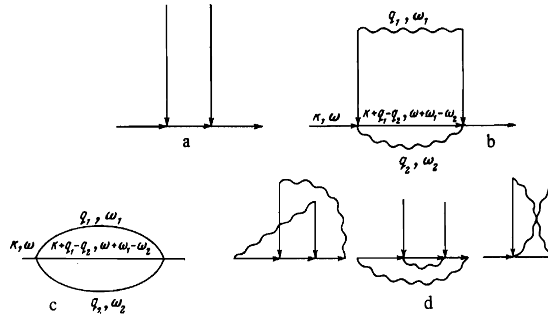

FIG. 3

Calculating in similar fashion the remaining quantities $\Sigma^{\alpha\gamma}$ and substituting them in (2) and (4), we obtain for the spectrum and for $\langle S^z\rangle$

$$\varepsilon^2(k)=\varepsilon_0{}^2(k)+\frac{V(k)\Delta\Omega_0}{2}\int\frac{V(q)}{\varepsilon_0(q)}\frac{3\Delta+\varepsilon_0(q)}{\Delta+\varepsilon_0(q)}[1+2N(\varepsilon_0(q)]\frac{d^3\mathbf{q}}{(2\pi)^3}, \tag{8}$$

$$\langle S^z\rangle=b-b\,\Omega_0\int\frac{V(q)}{\varepsilon_0(q)}\frac{\Delta-\varepsilon_0(q)}{\Delta+\varepsilon_0(q)}\frac{d^3\mathbf{q}}{(2\pi)^3}-\Omega_0\int\frac{V(q)}{\varepsilon_0(q)}\frac{\Delta^2+\varepsilon_0{}^2(q)}{\Delta^2-\varepsilon_0{}^2(q)}\times N(\varepsilon_0(q))\frac{d^3\mathbf{q}}{(2\pi)^3}. \tag{9}$$

It is seen from (8) and (9) that even at T = 0 there are corrections proportional to r_0^{-3} to the zeroth approximation of the spectrum and to $\langle S^z\rangle$. The presence of these corrections is due to the fact that the Hamiltonian $\mathscr{H}_0$ does not yield the correct ground state. This question was discussed in detail in Sec. 1.

The diagrams of second order, r_0^{-6}, are obtained for $\Sigma^{\alpha\gamma}$ by joining the vertices of the block Γ^{+++---} (Fig. 3a) with an interaction. Let us consider, e.g., the correction Σ^{+-} (Fig. 3b). To calculate it it is necessary to replace the internal solid line by a thick line, i.e., to set it in correspondence with

$$\tilde{G}(k,i\omega)=G(i\omega)(1+\mathrm{V}(k,i\omega)G(i\omega))=K_{(0)}^{+-}(k,i\omega).$$

After summing over the frequencies we obtain

$$\Sigma_{(3b)}^{+-}=\frac{bG^2(i\omega)\Delta^3\Omega_0{}^2}{4}\int\frac{V(q_1)V(q_2)}{\varepsilon_1\varepsilon_2\varepsilon_3}\Big\{\Phi(\varepsilon_1,\varepsilon_2,\varepsilon_3)+\Phi(\varepsilon_2,\varepsilon_3,\varepsilon_1)+\Phi(\varepsilon_3,\varepsilon_2,\varepsilon_1)+\frac{N(\varepsilon_1)N(\varepsilon_2)N(\varepsilon_3)-(1+N(\varepsilon_1))(1+N(\varepsilon_2))(1+N(\varepsilon_3))}{\omega^2+(\varepsilon_1+\varepsilon_2+\varepsilon_3)^2}\times(\varepsilon_1+\varepsilon_2+\varepsilon_3)\Big\}\frac{d^3q_1\,d^3q_2}{(2\pi)^6}. \tag{10}$$

$$\Phi(\varepsilon_1,\varepsilon_2,\varepsilon_3)=\frac{(1+N(\varepsilon_1))N(\varepsilon_2)N(\varepsilon_3)-N(\varepsilon_1)(1+N(\varepsilon_2))(1+N(\varepsilon_3))}{\omega^2+(\varepsilon_1-\varepsilon_2-\varepsilon_3)^2}\times(\varepsilon_1-\varepsilon_2-\varepsilon_3),$$

$\epsilon_1 = \epsilon_0(\mathbf{q}_1)$, $\epsilon_2 = \epsilon_0(\mathbf{q}_2)$, $\epsilon_3 = \epsilon_0(\mathbf{k} + \mathbf{q}_1 - \mathbf{q}_2)$. It is seen from (10) that $\Sigma_{(3b)}^{+-}$, in the language of diagram technique for ordinary particles, has the meaning of the self-energy correction necessitated by the two-particle interaction. It can be set in correspondence with diagram 3c, where the solid line corresponds to the propagator $(\omega_n^2+\epsilon_0^2(q))^{-1}$. The diagrams of Fig. 2 yield the Hartree correction to $\Sigma^{\alpha\gamma}$.

We consider first $\Sigma(0, 0)$ at $T = 0$. Then only the last term in the curly brackets of (10) differs from zero. As $\eta \to 1$ and at small q, we have $\epsilon_0(q) \sim q$, so that the integral with respect to q_2 in (10) diverges logarithmically if $\eta \to 1$ and $q_1 \to 0$. It is possible to estimate expression (10) by putting $\eta = 1$ in all the $\epsilon_0(q)$ and by replacing the lower limit of integration with respect to q^2 by $q_0 = r_0^{-1}|\eta-1|^{1/2}$. We obtain

$$\Sigma^{+-}_{(3b)}=\frac{G^2(i\omega)\Omega_0^2\Delta^3}{8(2\pi)^4u^6}[(\eta-1)\ln|\eta-1|+C],\qquad u=\left(-\Delta\frac{\partial V(k)}{\partial k^2}\right)^{1/2}\Bigg|_{k=0},$$

C is a constant on the order of unity.

It is easily seen that in general, in n-th order or perturbation theory, $\Sigma^{+-}(0, 0)$ has terms that contain singularities in $\eta-1$, and the largest of them is proportional to $(\eta-1)[(a/r_0)^3\ln|\eta-1|]^{n-1}$. Indeed, when the order of perturbation theory is increased by unity, there is added to the diagrams one effective interaction, and one of the solid lines can become thick, i.e., two additional factors of the type $\omega_n^2+\epsilon_0^2(q)$ appear in the denominator. After summing over m there are in the denominator three frequencies that depend on the new integration momentum q, i.e., roughly speaking, q^3, and this yields an additional $\ln|\eta-1|$ as $\eta\to 1$. Thus, the structure of the expression for the spectrum at small q and as $\eta \to 1$ is the following:

$$\begin{aligned}\varepsilon^2(q)&=\varepsilon_0^2(q)+\Delta^2\sum_{m=1}^{\infty}A_m\left(\frac{a}{r_0}\right)^m\\&+\Delta^2(\eta-1)\sum_{m=1}^{\infty}B_m\left[\left(\frac{a}{r_0}\right)^3\ln|\eta-1|\right]^m\left(\frac{a}{r_0}\right)^3.\end{aligned}\tag{11}$$

Here A_m and B_m are numerical coefficients of the order of unity. The terms of the last series in (1) diverge as $\eta \to 1$. Unlike the phase transitions at finite temperatures, when the terms of the perturbation theory diverge in proportion to powers of $\tau = (T-T_c)/T_c$, in the phase transition with respect to η at $T = 0$, which is considered here, the singularities are logarithmic. We have a successive-approximation method if

$$(a/r_0)^3\ln|\eta-1|\ll 1.\tag{12}$$

At finite temperatures, putting $N(x) = T/x$ in (10), we obtain power-law divergences in terms of $\epsilon_0^{-1}(0)$. The criterion for the applicability of the molecular-field method is in this case

$$(a/r_0)^3T/\varepsilon_0(0)\ll 1.\tag{13}$$

The terms of the first series in (11) appear upon integration over large momenta $q \approx r_0^{-1} \gg q_0$, and therefore are independent of $\eta-1$ or of τ.

The integral-containing term of (8) yields the Hartree correction to the spectrum. The temperature-independent term under the integral sign in (8) leads to a significant, on the order of $(a/r_0)^3$, renormalization of V(k), which is of no interest to us. The integral with $N(\omega)$ in (8), as will be shown presently, can determine the dependence of $\epsilon(k)$ on T, and, in addition, as $\eta \to 1$ it can become larger than $\epsilon_0^2(0)$. In this case $\Sigma^{XX}_{(1)} > \Sigma^{XX}_{(0)}$, and it is therefore necessary to renormalize the bare frequency $\epsilon_0(k)$ by including also $\Sigma^{\alpha\gamma}_{(1)}$ in the zeroth approximation. The Hartree terms in the higher orders of perturbation theory are smaller than $\Sigma^{\alpha\gamma}_{(1)}$ in terms of the parameter $(a/r_0)^3$ (see the series (11)) and, just as $\Sigma^{\alpha\gamma}_{(1)}$, depend on the temperature and can therefore be disregarded. It might seem that in order to carry out the renormalization it is necessary to replace $\epsilon_0(k)$ by $\epsilon(k)$ in $V(k, i\omega)$, meaning also in the integral of formula (8), and this would lead to an integral equation for $\epsilon(k)$. However, as will be shown later on, the temperature-dependent part of (k) is always smaller than T^2, and the contribution to the integral is made by values $\epsilon \approx T$. This means that replacement of $\epsilon_0(k)$ by $\epsilon(k)$ in the integral of (8) is not necessary, and formula (8) yields the renormalized frequency $\epsilon(k)$. We note that the mentioned integral equation was derived in[9], but its solution was not given.

Let us consider several limiting cases.

1) $\eta > 1$. Since $\epsilon_0(0) < T$, we can put $\epsilon_0(q) = qu$ in (8), after which we obtain

$$\varepsilon^2(k)=\varepsilon_0^2(k)+V(k)\Delta^2\Omega_0T^2/4u^3.\tag{14}$$

The second term in (14) is of the order of $T^2(a/r_0)^3$ at small k.

Replacing $\tanh(\beta\Delta/2)$ by unity and equating $\epsilon(0)$ to zero, we obtain an expression for T_c:

$$T_c=2[(1-\eta)u^3/\Delta\Omega_0]^{1/2}\sim\Delta[(1-\eta)(r_0/a)^3]^{1/2}.\tag{15}$$

It is seen from (15) that replacement of $\tanh(\beta\Delta/2)$ by unity is permissible at $[(1-\eta)(r_0/a)^3]^{1/2} \ll 1$. In this case the temperature dependence of the spectrum and the connection of T_c with η differ substantially from the result obtained from the zeroth approximation. The spectrum at $T > T_c$ is practically activationless, since

$$\varepsilon(0)=\frac{1}{2}(\Delta^3\Omega_0/u^3)^{1/2}(T^2-T_c^2)^{1/2}\ll T.\tag{15'}$$

If $\tau = (T-T_c)/T_c \ll 1$, then at $q \ll r_0^{-1}$ we have

$$\varepsilon^2(q)=(qu)^2+2(1-\eta)\tau\Delta^2\tag{16}$$

and the criterion (13) reduces to

$$(a/r_0)^3\tau^{-1}\ll 1.\tag{17}$$

We note that the condition for the applicability of the self-consistent field method takes in[8] the form $(a/r_0)^3\tau^{-1/2} \ll 1$, which does not coincide with (17). The reason for this difference is that in [8] T_c does not depend on r_0, whereas in our case $T_c \sim r_0^{3/2}$.

Writing down the denominator in the formula for K^{+-} in the form $q^2+q_0^2$, we obtain for the reciprocal radius q_0 the expression

$$q_0=\Delta u^{-1}[2(1-\eta)\tau]^{1/2}\sim r_0^{-1}[(1-\eta)\tau]^{1/2}.$$

The criterion (12) takes the form

$$(a/r_0)^3\ln q_0r_0\approx(a/r_0)^3\ln[(1-\eta)\tau]\ll 1.\tag{18}$$

When the conditions (17) and (18) are satisfied, the molecular-field method is applicable in the paramagnetic region. It is seen from these criteria that at sufficiently small $1-\eta$ and not too small τ, the logarithmic singularities of the perturbation-theory series terms are more substantial than the power-law singularities.

2) $\eta > 1$, $\epsilon_0(0)|_{T=0} > T$. In this case we have

$$\begin{aligned}\varepsilon^2(k)&=\varepsilon_0^2(k)+\frac{3\Omega_0V(k)\Delta}{\pi^{3/2}\varepsilon_0(0)}(mT)^{3/2}\exp(-\beta\varepsilon_0(0)),\\m^{-1}&=-\frac{\Delta}{\varepsilon_0(0)}\frac{\partial V(k)}{\partial k^2}\Bigg|_{k=0}.\end{aligned}\tag{19}$$

The dependence of ϵ on T remains exponential, but the exponent in the last term of (19) is smaller by a factor $(1-\eta)^{1/2}$ than the exponent of $e^{-\beta\Delta}$ contained in $\epsilon_0(k)$. Therefore the Hartree correction contains in this case likewise a dependence of $\epsilon(k)$ on T in a wide range of temperatures.

3) $\eta > 1$, $\epsilon_0(0)|_{T=0} < T$. In this case formula (4) is valid.

An analysis of the experimental data, carried out in Sec. 4, shows that for Pr_3Tl we have $(a/r_0)^3 \approx 1$. However, the structure of the perturbation-theory series is such that in this case the temperature dependences outside the critical region (i.e., at $\tau \gtrsim 1$) coincide with (14), (15), and (19). Indeed, at $\epsilon(0) \gtrsim T$ the temperature-dependent parts of the corrections to $\epsilon(k)$ in n-th order of perturbation theory are proportional to $e^{-\beta\epsilon(0)}T^{3/2}(a/r_0)^{3n}$. All the corrections are equally dependent on the temperature and are of the same order of magnitude at $(a/r_0)^3 \approx 1$. Therefore the temperature-dependent term in $\epsilon(k)$ is correctly calculated apart from a coefficient that is a function of a/r_0 only and is probably of the order of unity. Similar arguments can be advanced also in the case of $\epsilon(0) \approx T$. The relation $\epsilon(0) \gtrsim T$ is violated only in the critical region $|\tau| < 1$.

The damping of the excitations is determined by the imaginary part of the analytic continuation of the quantity $\Sigma^{xx}(k, i\omega)$ to real frequencies. The significant diagrams for $\Sigma^{\alpha\gamma}(k, i\omega)$ in second order differ from $\Sigma^{+-}_{(3b)}(k, i\omega)$ only by a numerical factor. Examples of such diagrams are shown in Fig. 3d.

If the conservation laws permit the decay of an excitation into three excitations, then the last term of (10) yields the damping connected with this process and equal to

$$\gamma_{\text{dec}}(k, \varepsilon(k)) = \frac{-\pi\Omega_0{}^2\Delta^4}{2\varepsilon(k)} \int \frac{\delta(\varepsilon(k)-\varepsilon_1-\varepsilon_2-\varepsilon_3)V(q_1)V(q_2)}{\varepsilon_1\varepsilon_2\varepsilon_3}$$
$$\times[N(\varepsilon_1)N(\varepsilon_2)N(\varepsilon_3)-(1+N(\varepsilon_1))(1+N(\varepsilon_2))(1+N(\varepsilon_3))]\frac{d^3\mathbf{q}_1\,d^3\mathbf{q}_2}{(2\pi)^6}.$$

If we confine ourselves in the calculation of $\epsilon(k)$ to the first term in the expansion of V(k) in k^2, then the argument of the δ-function in the expression written out above differs from zero at arbitrary k, q_1, q_2, and $\gamma_{\text{dec}} = 0$. On the other hand, if the term proportional to k^4 in the expansion of V(k) in k^2 is positive and its contribution $\epsilon^4(k)/\Delta^2$ to $\epsilon^2(k)$ is larger than $\epsilon^2(0)$, then we have a decay spectrum at these values of k. Thus, decay is possible only at $\epsilon(k) > (\Delta\epsilon(0))^{1/2}$.

The first three terms in (10) yield the damping of the excitations connected with their interaction. It is equal to

$$\gamma(k, \varepsilon(k)) = \frac{3\Delta^4\Omega_0{}^2}{2\varepsilon(k)} \int \frac{[\delta(\varepsilon(k)+\varepsilon_1-\varepsilon_2-\varepsilon_3)+\delta(\varepsilon(k)+\varepsilon_1+\varepsilon_2-\varepsilon_3)]}{\varepsilon_1\varepsilon_2\varepsilon_3}$$
$$\times V(q_1)V(q_2)[N(\varepsilon_1)(1+N(\varepsilon_2))(1+N(\varepsilon_3)-N(\varepsilon_2)N(\varepsilon_3)(1+N(\varepsilon_1))]\frac{d^3\mathbf{q}_1\,d^3\mathbf{q}_2}{(2\pi)^6}$$

At $\epsilon(q) \gg \epsilon(0)$, the exciton spectrum is of the phonon type and, analogously, the phonon damping $\gamma(k, \epsilon(k))$ depends essentially on the ratio of the different parameters (e.g., such as ϵ, $(\Delta\epsilon(0))^{1/2}$, etc.

A detailed analysis of $\gamma(k, \epsilon(k))$ at $\epsilon \gg \epsilon(0)$ is meaningful apparently in connection with some particular experimental situation. We confine ourselves to the frequency region $\epsilon - \epsilon(0) \lesssim \epsilon(0)$. Two cases are possible.

1) If $\epsilon(0) > T$, then

$$\gamma(k, \varepsilon(k)) = \frac{3\Omega_0{}^2}{2(2\pi)^4}\frac{\Delta^6 m(2mT)^2}{\varepsilon^4(0)\varepsilon(k)}e^{-\beta\varepsilon(0)}$$
$$\sim \varepsilon(k)\left(\frac{a}{r_0}\right)^6\left(\frac{T}{\varepsilon(0)}\right)^2 e^{-\beta\varepsilon(0)}.$$

2) On the other hand if $\epsilon(0) < T$, then the terms proportional to $N(\omega)$ and proportional to the product of two Bose functions yield contributions of the same order of magnitude to the damping. The essential quantities are $q_1 \sim q_2 \sim T/u$ in the integral of the first terms and $q_1 \sim q_2 \sim q_0$ and q_T in the integral of the second. An estimate of the damping yields

$$\gamma(k, \varepsilon(k)) \approx \frac{\Omega_0{}^2\Delta V(k)}{\varepsilon(0)u^6}T^2.$$

In the calculation of the damping, Stinchcombe[9] considered only diagrams of first order in r_0^{-3}, one of which is shown in Fig. 2b. Therefore the damping obtained by him[9] is proportional to $e^{-\beta\Delta}$ and is small in comparison with damping calculated in the present paper in both the paramagnetic and the ferromagnetic region.

To calculate the free energy F, we replace V(r) by gV(r) and differentiate F with respect to g[10]:

$$\frac{\partial F}{\partial g} = -\frac{T^2\Omega_0}{2}\sum_n\int\frac{d^3\mathbf{q}}{(2\pi)^3}K^{xx}(q, i\omega_n)V(q).$$

Using the fact that

$$1-2gV(q)\Sigma^{xx}(q, i\omega_n) = \frac{\omega_n{}^2+\varepsilon^2(q)}{\omega_n{}^2+\Delta^2},$$

we obtain

$$\frac{\partial F}{\partial g} = \frac{\Omega_0 T}{2}\int\frac{d^3k}{(2\pi)^3}\frac{\partial}{\partial g}\ln\operatorname{sh}\frac{\beta\varepsilon(k)}{2}$$
$$+\frac{\Omega_0}{2}\int\frac{d^3k}{(2\pi)^3}\frac{1+2N(\varepsilon(q))}{\varepsilon(k)}\left[\frac{\varepsilon^2(k)-\varepsilon_0{}^2(k)}{g}-\frac{\partial(\varepsilon^2-\varepsilon_0{}^2)}{\partial g}\right]. \tag{20}$$

The appearance of the second term in (20) is due to the fact that it is impossible to carry out a graphic summation in the diagram technique for the thermodynamic potential. Therefore the characteristic frequency renormalization does not reduce, generally speaking to a replacement of $\epsilon_0(k)$ by $\epsilon(k)$ in the expression for the free energy of the Bose gas of the noninteracting particles.

Let us calculate the specific heat, taking into account only the first term of (20); it will be shown below that the contribution of the second term is small. We have

$$C = -T\frac{\Omega_0}{2}\int\frac{d^3\mathbf{q}}{(2\pi)^3}\left\{\frac{\partial^2\varepsilon(q)}{\partial T^2}(1+2N(\varepsilon(q)))\right.$$
$$\left.-\frac{e^{\beta\varepsilon(q)}}{(e^{\beta\varepsilon(q)}-1)^2}\left(\frac{\partial\varepsilon(q)}{\partial T}-\frac{\varepsilon(q)}{T}\right)^2\right\}. \tag{21}$$

If $\eta < 1$ and $\tau \ll 1$, then we obtain from (14) and (21)

$$C = \frac{2\pi^2\Omega_0}{15u^3}T^3 - 4\frac{(1-\eta)^{3/2}}{\tau^{1/2}}. \tag{22}$$

On the other hand if $\epsilon(0) > T$, then

$$C = \frac{\Omega_0(2mT)^{3/2}}{4\pi^{3/2}}\left(\frac{\varepsilon(0)}{T}\right)^2 e^{-\beta\varepsilon(0)}. \tag{22'}$$

In the derivation of (22) and (22'), an important role was played in the integrals of (21) by small momenta $q \sim T/u$ and $q \sim q_0$. Let us take the derivatives of $\partial F/\partial g$ with respect to T. The integrand in the first term of (20) then has an extra factor $V(q)/\epsilon(q)$ in comparison with the integrand in the second term. Since small momenta play an important role in the integral, the second term can indeed be neglected.

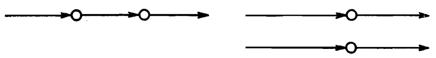

FIG. 4

In concluding this section, let us consider the correlator K^{zz}. The diagrams for K^{zz} are obtained from the blocks shown in Fig. 4. Summing over the frequency and carrying out an analytic continuation, we obtain

$$K^{zz}(k,\omega)=\frac{3}{2}\Delta\Omega_0\int\frac{V(q)}{\varepsilon_1\varepsilon_2}\left\{\frac{\varepsilon_1+\varepsilon_2}{\omega^2-(\varepsilon_1+\varepsilon_2)^2}\right.$$

$$\times[N(\varepsilon_1)N(\varepsilon_2)-(1+N(\varepsilon_1))(1+N(\varepsilon_2))]+\frac{\varepsilon_1+\varepsilon_2}{\omega^2-(\varepsilon_1-\varepsilon_2)^2} \qquad (22'')$$

$$\left.\times[N(\varepsilon_1)(1+N(\varepsilon_2))-N(\varepsilon_2)(1+N(\varepsilon_1))]\right\}\frac{d^3\mathbf{q}}{(2\pi)^3},$$

$$\varepsilon_1=\varepsilon(q),\quad \varepsilon_2=\varepsilon(\mathbf{k}-\mathbf{q}).$$

At T = 0 we obtain from (22) the following expression for $\mathrm{Re}K^{zz}(0, 0)$:

$$\mathrm{Re}\,K^{zz}(0,0)=-\frac{3\Omega_0\Delta^2}{4(2\pi)^2u^3}(\ln q_0r_0+C),$$

where $C \approx 1$. Thus, the series for K^{zz} begins with a term that has a singularity in $\eta - 1$ and, unlike in (10), there is no factor $\eta - 1$. Therefore each term of the series diverges as $\eta \to 1$.

3. FERROMAGNETIC PHASE

At $T < T_c$, the system goes over into the ferromagnetic state. The zeroth Hamiltonian in the ferromagnetic phase is chosen to be as follows:

$$\mathscr{H}_0=-\Delta\sum_i S_i^z-y\sum_i S_i^x,$$

where $y = 2\langle S^x\rangle V(0)$. We rotate the axes x and z in such a way that the molecular field $h = (\Delta^2 + y^2)^{1/2}$ is directed along the z axis. The Hamiltonian (1) in terms of the new coordinates takes the form

$$\mathscr{H}=-h\sum_i S_i^z-y\sum_i\langle S_i^{x'}\rangle-\sum_{i,j}V^{(1)}(\mathbf{r}_i-\mathbf{r}_j)(2S_i^+S_j^-+S_i^-S_j^-+S_i^+S_j^+)$$

$$-\frac{1}{2}\sum_{i,j}V^{(2)}(\mathbf{r}_i-\mathbf{r}_j)(S_i^z-\langle S^z\rangle)(S_j^z-\langle S^z\rangle)-$$

$$-\sum_{ij}V^{(3)}(\mathbf{r}_i-\mathbf{r}_j)[S_i^+(S_j^z-\langle S^z\rangle)+S_i^-(S_j^z-\langle S^z\rangle)], \qquad (23)$$

$$V^{(1)}(r)=V(r)\frac{\Delta^2}{y^2+\Delta^2},\qquad V^{(2)}(r)=\frac{2y^2}{y^2+\Delta^2}V(r),$$

$$V^{(3)}(r)=\frac{\sqrt{2}\,y\Delta}{y^2+\Delta^2}V(r),\qquad \langle S_i^{x'}\rangle\equiv y/2V(0).$$

Writing down the system of Dyson's equations in the usual manner, we can express the correlators in terms of the self-energy parts. We present only the expression for K^{+-}:

$$K^{+-}=[\Sigma^{+-}+V^{(1)}(k)(\Sigma^{++}\Sigma^{--}-\Sigma^{+-}\Sigma^{-+})$$
$$-V^{(2)}(k)\Sigma^{+-}(\Sigma^{z+}+\Sigma^{z-})+V^{(3)}(k)(\Sigma^{++}\Sigma^{z-}-\Sigma^{+-}\Sigma^{z+})]$$
$$\times[1-2V^{(1)}(k)\Sigma^{xx}-V^{(2)}(k)\Sigma^{zz}-V^{(3)}(k)(\Sigma^{z+}+\Sigma^{z-})]^{-1}.$$

In the zeroth approximation we have

$$K^{+-}(k,i\omega_n)=b(\beta h)\frac{z-V^{(1)}(k)+i\omega_n}{\omega_n^2+\varepsilon_0^2(k)},$$

$$\varepsilon_0^2(k)=h^2[1-2bV^{(1)}(k)\Delta^2h^{-3/2}]. \qquad (24)$$

The free energy in this approximation is[8]

$$F^{(0)}=-T\ln\mathrm{ch}\,\beta\frac{h}{2}+\frac{y^2}{4V(0)}.$$

Determining y from the condition $\partial F(0)/\partial y = 0$, we obtain an equation for h:

$$h=V(0)\,\mathrm{th}\,(\beta h/2).$$

At T = 0 we have

$$\langle S^{x'}\rangle={}^1/_2(1-\eta^2)^{1/2},\qquad y/\Delta=(1-\eta^2)^{1/2}\ll 1.$$

The structure of the perturbation-theory series is the same as in the paramagnetic region. The molecular-field method is valid if the criteria (12) and (13) are satisfied.

We confine ourselves to low temperatures $T \ll T_c$, when the temperature-dependent increment to $\epsilon_0(q)$ is small. The Hartree correction to the spectrum leads to expression (8) for $\epsilon(q)$, in which $\epsilon_0(q)$ is now determined by formula (24), and V(q) is replaced by $V^{(1)}(q)$. To determine the temperature dependence of the spectrum it is necessary to substitute for h in (24) its first-approximation value.

The free energy in the first approximation is

$$F^{(1)}=F^{(0)}+\frac{\Omega_0}{2}\int\ln\mathrm{sh}\frac{\beta\varepsilon_k}{2}\frac{d^3\mathbf{k}}{(2\pi)^3}+O(e^{-\beta\Delta}).$$

Neglecting terms of order $(a/r_0)^3$ and $e^{-\beta\Delta}$, we obtain

$$\frac{\langle S^{x'}\rangle_{T=0}-\langle S^{x'}\rangle}{\langle S^{x'}\rangle_{T=0}}=\frac{\langle J\rangle_{T=0}-\langle J\rangle}{\langle J\rangle_{T=0}}=(1-\eta^2)^{-1}\Gamma(T),$$

$$\Gamma(T)=\begin{cases}{}^3/_8\Omega_0(2mT/\pi)^{3/2}(1-\eta^2)^{-1/2}e^{-\beta\varepsilon_0(0)}, & \varepsilon_0(0)>T\\ (T/T_c)^2, \quad \varepsilon_0(0)<T\ll T_c\end{cases} \qquad (25)$$

Substituting (24) and (25) in (8) we get

$$\varepsilon^2(k)=\varepsilon_0^2(k)-2\Delta^2\Gamma(T). \qquad (26)$$

As expected, $\epsilon(k)$ increases with decreasing temperature. With the aid of (26) we can easily verify that the criterion (13) is satisfied in the entire temperature region $T \ll T_c$.

Since $y \ll \Delta$, it follows that $V^{(1)} \approx V \gg V^{(2)} \gg V^{(3)}$. Therefore the specific heat is described by formula (22') at $\epsilon_0(0) > T$, and is equal to the first term of (22) if $\epsilon_0(0) < T$. The damping connected with $\mathrm{Im}\,\Sigma^{xx}$ is determined by the corresponding formulas of the preceding section with $\epsilon(q)$ from (24).

The quantities $\Sigma^{z\alpha}$ (α = z, +, −) are determined, apart from a numerical factor, by expression (22"). Thus, corrections to the spectrum and damping exist in the ferromagnetic region and are all connected with the "triple anharmonicity" of the excitations. As seen from (23), these quantities have a small factor $y^2/\Delta^2 = 1 - \eta^2$ in comparison with $V^{(1)}_{(k)}\,\Sigma^{xx}$ and cannot change Re ω significantly. For the same reasons as for the two-particle interaction, a decay of the excitation into two is possible only if $\epsilon_0 > (\epsilon_0(0)\Delta)^{1/2}$. The imaginary part of the second term under the integral sign in (22') is proportional to $[N(\epsilon_1) - N(\epsilon_2)]\delta(\epsilon + \epsilon_1 - \epsilon_2)$. The conservation laws can be satisfied in this case, too, only if $\epsilon_0(k)$ or ϵ_1 is larger than $(\Delta\epsilon_0(0))^{1/2}$. Therefore at $\epsilon_0(k) \ll (\Delta\epsilon_0(0))^{1/2}$ the contribution made to the damping by $\mathrm{Im}\Sigma^{z\alpha}$ is of the order of $\exp[-\beta(\Delta\epsilon_0(0))^{1/2}]$ and can be neglected.

4. SINGLET FERROMAGNETISM OF PRASEODYMIUM IN Pr_3Tl

We discuss first the applicability of the two-level model for the description of real magnets such as Pr. An analysis carried out by Cooper[5] and valid in first approximation in $(a/r_0)^3$ has shown that the Hamiltonian of the system, with allowance for all nine sublevels, can be broken up into two parts in such a way that in the first part, $\mathscr{H}_1$, there are only operators of the transition between states with wave functions $\langle 1|$ and $\langle 2|$, while the second part $\mathscr{H}_2$ does not contain these operators. Therefore the spectrum branch determined by the Hamiltonian $\mathscr{H}_1$ can be found without considering the remaining sublevels.

The spectrum of the excitations in Pr_3Tl at $q > 0.4\pi/a$ was obtained in[2,3] with the aid of inelastic scattering. The appreciable dispersion of the excitation energy at such momenta shows that $(a/r_0)^3 \approx 1$. It was shown in Sec. 2 that in spite of this fact, the formulas of the present paper yield the correct dependence of $\epsilon(q)$ on T. This conclusion is valid also for the specific heat and for the damping.

In the zeroth approximation of the molecular-field method, all the quantities depend on the temperature via the exponential $e^{-\beta\Delta}$. From the values Δ and T_C given in Sec. 1, it follows that there is indeed a wide range of temperatures in the paramagnetic and ferromagnetic phase, where the relations obtained by us, namely the power-law relation and the exponential function $e^{-\beta\epsilon^{(0)}}$, are more appreciable than the results of the zeroth approximation.

The developed theory explains why no temperature dependence of the spectrum was observed in[2,3] in the rather wide temperature range 4.5–40°K. The point is that the spectrum was measured at large momenta, when, as seen from (14), (19), and (26), the spectrum depends weakly on the temperature. $\epsilon(q)$ depends on T via y, $b = \tanh(\beta h/\Delta)$, and the second term of (8). But the presence of y introduces in V(q) a relative correction $y^2/\Delta^2 = 1 - \eta^2 = 0.06$, which is much less than the measurement accuracy. The second term in (8) can change the effective value of V(q) by not more than $\delta V = V(q)(T/\Delta)^2\zeta$, where ζ is a numerical factor that does not contain the parameters (see, e.t., (8)). The measurements on Pr_3Tl were performed at $T < 20°K$, and therefore $1 - \tanh(\beta h/2) = 0.06$ and $\delta V/V \approx 0.05\zeta$, so that the temperature dependence of $\epsilon(q)$ can escape observation if the coefficient ζ is not large.

For Pr, the measurements were performed up to 40°K, so that $1 - \tanh(\beta h/2) \approx 0.2$ and $\delta V/V \approx 0.2\zeta$, but the spectra was measured at such large momenta that $\epsilon(q)/\Delta \geq 0.75$. This means that the temperature dependence of $\epsilon(q)$ is weaker by approximately a factor of 3 than the dependence of the effctive V(q) on T, so that the former was not observed.

To compare the theory with experiment, it would be quite desirable to measure the temperature dependence of the spectrum at $q < q_0 \approx \pi/4a$.

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