[13(a)]Recent specific-heat measurements (Ref. 6) of La_3Tl and La_3In in 90 kOe yielded $\gamma \sim 17$ and 20 mJ/°K² mole La, respectively. Therefore, correct values for $\Delta C/\gamma T_s$ in Fig. 2 are ~2.5 and 2.7, respectively.

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PHYSICAL REVIEW B VOLUME 6, NUMBER 7 1 OCTOBER 1972

Induced-Moment Ferromagnetism in Pr_3Tl

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It is shown that the intermetallic compound Pr_3Tl is a case of an induced-moment ferromagnet in which the nonmagnetic singlet ground state of Pr^{3+} is spontaneously polarized below 11 °K due to slightly overcritical exchange interactions. By substituting only 7 at.% of La for Pr, both the ordered moment m_0 (at 0 °K) and the Curie temperature T_C are reduced to zero, in contrast to the usual behavior of local-moment systems. The observed ratios of m_0/T_C for different La concentrations are compared with theoretical expectations. Specific-heat and resistivity anomalies at the transition are shown to be much reduced from what one would expect in an ordinary ferromagnet.

I. INTRODUCTION

The magnetic behavior of rare-earth intermetallic compounds and alloys depends mainly on two interactions: (a) the exchange interaction between the conduction electrons and the rare-earth ions and (b) the interaction of the rare-earth ion with its surrounding crystal field. If the resulting magnetic interaction between the rare-earth ions (via conduction electrons) is larger than the crystal field interaction, the latter is usually neglected at first and then reintroduced to explain the magnetic anisotropies in the ordered state. Examples of this case are the heavy-rare-earth metals from Gd to Tm. This paper deals with an example of the opposite situation, where the magnetic exchange interaction between the ions is smaller than the crystal field interaction. In this case one must first find the crystal field splitting of the lowest multiplet of the rare-earth ion and then consider the effect of magnetic exchange interactions on the crystal field levels. Of particular interest is the case of a crystal-field-singlet ground state, which can be realized in non-Kramers rare-earth ions (even J). Since such a ground state has no magnetic moment, it takes a finite critical magnitude of magnetic exchange interaction between neighboring ions in order to introduce a magnetically ordered ground state. If the exchange interaction is undercritical, the singlet ground state remains the stable state down to zero degrees, one then has Van Vleck paramagnetism and no magnetic order. For overcritical exchange, the singlet state mixes spontaneously with higher-lying crystal-field states to produce an energetically lower-lying polarized ground state. The ground-state moment m_0 must be determined self-consistently; it disappears at some finite ordering temperature T_C. In such an exchange-induced ferromagnet both m_0 and T_C are expected to go to zero continuously as the exchange interaction approaches the critical value (see Sec. III). The main purpose of this paper is to experimentally investigate this behavior in the

compound Pr_3Tl, in which the singlet ground state of Pr^{3+} is polarized ferromagnetically due to slightly overcritical exchange interactions. The critical value for the exchange interaction is approached experimentally by diluting the compound with La. In Sec. II we describe the experimental procedure. In Sec. III we summarize the theoretical results obtained for singlet-ground-state systems with exchange interactions, and in Sec. IV we compare the experimental results with the theoretical expectations.

II. EXPERIMENTAL PROCEDURE

A. Sample Preparation

Details of the sample preparation technique are given in the preceding paper by Bucher *et al.*[1] X-ray diffraction patterns confirmed the cubic Cu_3Au structure with a lattice constant of $a = 4.935$ and 5.06 Å for Pr_3Tl and La_3Tl, respectively. These values are noticeably smaller than the ones reported in the literature.[2,3]

B. Magnetic Measurements

Magnetic susceptibility measurements were carried out in a new type of magnetometer which has a temperature range from 0.5 to 100 °K and a field range from 0 to 50 kOe. It consists of a superconducting magnet of $1\frac{1}{8}$-in. bore into which a second small helium Dewar is inserted. Two niobium-titanium pickup coils wound onto this Dewar form a superconducting transformer with a third coil outside the magnet. Both pickup coils are centered in the magnet; they are 1 in. apart. The third coil is located in a magnetic shield outside the superconducting magnet in liquid helium. Sample flux changes generate field changes in this third coil. These field changes are monitored with a Hewlett-Packard flux gate magnetometer whose probe is located inside the third coil. The sensitivity is such that an indication of 1 G on the gaussmeter corresponds to a sample magnetic moment of 0.674 emu ($G\,cm^3$). Samples are mounted on a copper holder inside the insert helium Dewar. By pumping on this Dewar and heating up the copper holder, temperatures of about 100 °K can be reached. With this apparatus, susceptibility data can be taken directly by keeping the field fixed and sweeping the temperature. Magnetization data at constant temperature must be taken point by point by first setting the field and then lifting the sample in and out of the pickup coil. The superconducting magnet has to be operated in persistent mode to ensure stability of the magnetic field.

C. Specific-Heat Measurements

Specific-heat measurements have been taken with a heat-pulse calorimeter described elsewhere.[4]

D. Resistivity Measurements

Resistivity measurements were made with the conventional four-lead technique in the magnetometer cryostat described above, using the same copper holder. Samples were cut from the arc-melted buttons with a jeweler's saw and the four leads (platinum) were attached by means of spot welding.

III. THEORY

The problem of a system of singlet-ground-state ions coupled by exchange interactions has been treated by various authors.[5–8] Trammell[5] and Bleaney[6] treated it in the molecular-field approximation. Grover[7] has applied the technique of second quantization to the problem and has given solutions for the ground state and the excited states for both under- and overcritical values of the exchange interaction. He has treated both cases of hexagonal and cubic crystal field symmetries. More recently, Wang and Cooper[8] have treated the problem by starting from the molecular-field ground state and calculating the spectrum of the excited collective modes by the equation-of-motion technique in both a random phase as well as in a two-site correlation approximation. Since the crystal field symmetry around the Pr^{3+} ion in Pr_3Tl is close to cubic (see Sec. IV and preceding paper), so that the lowest-lying crystal-field levels (in the absence of exchange interactions) would be a Γ_1 singlet and a higher-lying (weakly split) Γ_4 triplet, we like to compare our results with Grover's calculation for the singlet-triplet case. In what follows, we first recall the molecular-field results for the singlet-triplet case, then briefly discuss Grover's calculation and use his results to derive the ordered moment at zero degrees and the ferromagnetic transition temperature as the exchange interaction approaches the critical value.

A. Molecular-Field Approximation

We neglect higher-lying crystal-field levels and consider only the ground-state singlet Γ_1 and the next-higher-lying state, a Γ_4 triplet. With the notation in Fig. 1, the Hamiltonian is diagonal in the crystal field eigenfunctions in the absence of exchange interactions:

$$H_0 = \sum_i \Delta(|\tilde{1}\rangle\langle\tilde{1}| + |\tilde{0}\rangle\langle\tilde{0}| + |-\tilde{1}\rangle\langle-\tilde{1}|)\,. \quad (1)$$

We now add an exchange interaction between ions of the form

$$H_{ex} = -\sum_i K_{ij}\,\vec{J}_i \cdot \vec{J}_j\,. \quad (2)$$

In the molecular-field approximation, H_{ex} simplifies to

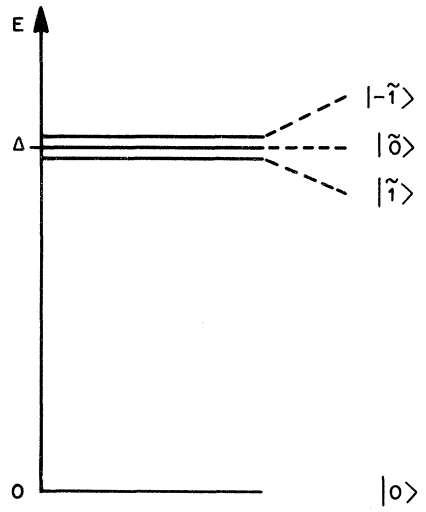

FIG. 1. Definition of wave functions for the singlet-triplet case.

$$H_{ex} = -2Kz\langle J\rangle \sum_i \tilde{J}_i \ . \tag{3}$$

Here z is the number of next-nearest neighbors and

$$H_M = -2Kz\langle J\rangle / g_J \mu_B \tag{4}$$

is the molecular field created by the z nearest-neighbor ions. In the Hamiltonian $H_0 + H_{ex}$ the angular-momentum operator $\tilde{J}_z$ (we choose the z axis as the self-polarization axis) connects the ground state $|0\rangle$ with the excited state $|\tilde{0}\rangle$. The new eigenstates will therefore be $|\tilde{1}\rangle$, $|-\tilde{1}\rangle$ as well as combinations $|\epsilon\rangle$ and $|g\rangle$ of $|0\rangle$ and $|\tilde{0}\rangle$. Calculation shows that the latter combinations are

$$\begin{aligned} |g\rangle &= \cos\theta\,|0\rangle + \sin\theta\,|\tilde{0}\rangle \ , \quad \text{ground state} \\ |\epsilon\rangle &= \sin\theta\,|0\rangle - \cos\theta\,|\tilde{0}\rangle \ , \quad \text{excited state} \end{aligned} \tag{5}$$

with $\tan(2\theta) = 4Kz\alpha^2\langle J_z\rangle/\Delta$ and $\alpha = \langle 0|J_z|\tilde{0}\rangle$. The angular momentum $\langle J_z\rangle$ of the ground state at zero degrees is

$$\langle J_z\rangle = \langle g|J_z|g\rangle = 2\cos\theta\sin\theta \ . \tag{6}$$

This self-consistent equation for $\langle J_z\rangle$ only has a solution if $4Kz\alpha^2/\Delta \geq 1$, which defines the magnitude of the critical exchange interaction $(K\cdot z)_{crit} = \Delta/4\alpha^2$ for spontaneous polarization of the ground state. For overcritical exchange interactions one finds

$$\langle J_z\rangle_{T=0} = \alpha\left(\frac{\eta^2-1}{\eta^2}\right)^{1/2}, \quad \eta = \frac{4Kz\alpha^2}{\Delta} \ . \tag{7}$$

The ground-state energy $E_g = \langle g|H|g\rangle$ becomes

$$E_g = \tfrac{1}{2}\Delta(1-\eta) \ . \tag{8}$$

In Fig. 2 the energies of the eigenstates are plotted schematically versus the exchange parameter η. In order to find the transition temperature one has to calculate $\langle J_z\rangle$ at finite temperatures:

$$\begin{aligned} \langle J_z\rangle &= p_g\langle g|J_z|g\rangle + p_e\langle e|J_z|e\rangle \\ &= 2\cos\theta\sin\theta(p_g - p_e) \ , \end{aligned} \tag{9}$$

where p_g and p_e are the thermal population probabilities of the ground and excited states, respectively. The solution of (9) is

$$\langle J_z\rangle^2 = \alpha^2[(p_g - p_e)^2 - 1/\eta^2] \ . \tag{10}$$

The transition to the paramagnetic state is defined by $\langle J_z\rangle = 0$ or by

$$p_g(T) - p_e(T) = 1/\eta \ . \tag{11}$$

One finds for the transition temperature the equation

$$(1 - e^{-\Delta/kT_C})/(1 + 3e^{-\Delta/KT_C}) = 1/\eta \ . \tag{12}$$

Of special interest is the relation between the ordered moment of the ground state and the transition temperature. This relation is obtained by eliminating η from (7) and (12); it is plotted in Fig. 3. At first sight this result is surprising and seems to be unphysical in that the ordered moment vanishes much faster than the transition temperature; it is in fact in strong disagreement with the experimental results. The reason for this behavior is that close to the critical exchange the magnetic condensation energy $\frac{1}{2}\Delta(1-\eta)$ is very much smaller than the gap Δ to the next-higher excited state: Even for a small ordered moment, one has to heat up to a temperature much larger than k_B^{-1} times the magnetic condensation energy in order to populate the oppositely polarized higher excited state $|e\rangle$ and compensate the ground-state moment.

B. Collective Excitation Model

In reality one can expect collective crystal-field excitations to exist which have energies comparable to the magnetic condensation energy of the ground state. In order to obtain the dispersion relations for such collective excited states, one can either use an equation-of-motion technique or the method of second quantization. The latter method has been applied by Grover[6] to the case of ferromagnetic Pr^{3+} in a cubic crystal field, and we can use his solution to compute the ferromagnetic transition temperature as a function of the ordered moment. His calculation applies for a simple-cubic lattice of Pr^{3+} ions when only isotropic next-nearest-neighbor exchange interactions are considered. In Pr_3Tl, the Pr^{3+} ions form three interpenetrating simple cubic lattices which, from the point of view of

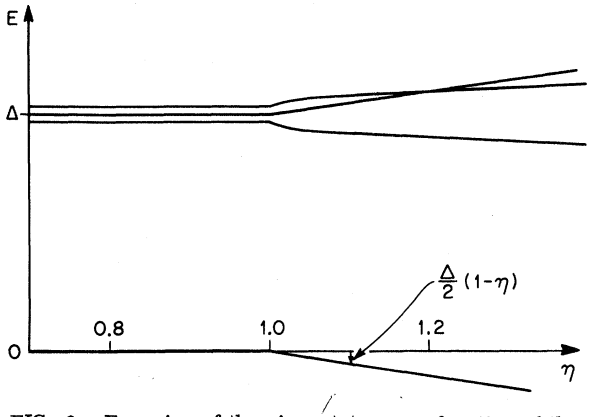

FIG. 2. Energies of the eigenstates as a function of the exchange parameter η (molecular-field approximation).

next-nearest-neighbor ions, are not exactly equivalent to each other. This leads to anisotropies in the dispersion relations for the collective modes in k space. However, in the long-wavelength limit which we shall use below, these anisotropies disappear and the dispersion relations of the three cubic sublattices are degenerate with each other. Grover starts by writing the Hamiltonian H_0+H_{ex} [Eqs. (1) and (2)] in terms of creation and destruction operators, by means of which an ion can be excited from the ground state to one of the triplet states. By definition, then,

$$a_i^\dagger|0\rangle_i=|\tilde{0}\rangle_i,\quad b_i^\dagger|0\rangle_i=|1\rangle_i,\quad c_i^\dagger|0\rangle_i=|-1\rangle_i\ , \tag{13}$$

and $a_i^\dagger a_i$, $b_i^\dagger b_i$, $c_i^\dagger c_i$ are the occupation numbers of the three triplet states of ion i. These operators actually satisfy the commutation relations for bosons as long as the occupation probability of the triplet states is small compared to the occupation probability of the ground state (low temperatures). It turns out that this condition is well satisfied in Pr_3Tl below its Curie point of 11 °K. Grover finds that the Hamiltonian can be diagonalized by means of a Bogoliubov transformation:

$$\alpha_k^\dagger=u_k a_k^\dagger-v_k a_{-k},\quad \beta_k^\dagger=w_k b_k^\dagger+x_k c_k\ ,\quad \gamma_k^\dagger=w_k c_k^\dagger+x_k b_k\ . \tag{14}$$

Here a_k, etc., are the coefficients in the Fourier expansion $a_i=(1/N^{1/2})\sum_k a_k e^{i\vec{k}\cdot\vec{r}}$. The dispersion relations of the new modes α_k, β_k, and γ_k which he finds are sketched in Fig. 4. In the limit of long wavelengths, they are given by

$$\hbar\omega_{k,\alpha}=\Omega(1+A^2a^2k^2)^{1/2}\ ,$$

with

$$\Omega=(W_0-\Delta)^{1/2},\quad W_0=4Kz\alpha^2\ ,$$

$$A^2=1/z(\eta^2-1),\quad \eta=W_0/\Delta\ , \tag{15}$$

$$\hbar\omega_{k,\beta}=\hbar\omega_{k,\gamma}=Dak\ ,$$

with

$$D=\left(\frac{\Delta+W_0}{2z}\right)^{1/2}\left(\frac{77W_0+83\Delta}{24\alpha^2}\right)^{1/2}\ .$$

This solution is valid in the range $\eta\geq 1$, where a magnetically ordered ground state exists. The longitudinal modes α_k are modes with magnetization along the z axis. They show a gap at $k=0$ in the ordered state because of the presence of the internal field. The transverse modes β_k and γ_k have magnetization normal to the z axis. They do not interact with the internal field and have no gap at $k=0$. To compute the transition temperature to the paramagnetic state we again use Eq. (10) where we set $\langle J_z\rangle=0$:

$$\alpha^2[(p_g-\textstyle\sum p_e)^2-1/\eta^2]=0\ , \tag{16}$$

where now

$$\textstyle\sum p_e=\langle p_{k,\alpha}\rangle$$

is the thermal average of the excitation probability of all excitations α_k. Since

$$p_g+\langle p_{k,\alpha}\rangle+\langle p_{k,\beta}\rangle+\langle p_{k,\gamma}\rangle=1\ , \tag{17}$$

the transition temperature is given from (16) and (17) by the equation

$$2\langle p_{k,\alpha}\rangle+\langle p_{k,\beta}\rangle+\langle p_{k,\gamma}\rangle=1-1/\eta\ . \tag{18}$$

Now $\langle p_{k,\alpha,\beta,\gamma}\rangle$ are the thermal averages of the products $\alpha^\dagger\alpha$, $\beta^\dagger\beta$, and $\gamma^\dagger\gamma$, respectively. One finds, in the long-wavelength limit $[(\eta-1)\ll 1]$:

$$\langle p_{k,\alpha}\rangle=\frac{1}{N}\sum_k(u^2+v^2)\,\frac{1}{e^{\hbar\omega_{k,\alpha}/kT-1}}\ ,$$
$$\langle p_{k,\beta}\rangle=\langle p_{k,\gamma}\rangle=\frac{1}{N}\sum_k(w^2+x^2)\,\frac{1}{e^{\hbar\omega_{k,\beta}/kT-1}}\ , \tag{19}$$

where

$$u=\cosh\theta,\quad v=\sinh\theta,\quad \tanh 2\theta=1/(2\eta^2-1)\ ,$$
$$w=\cosh\phi,\quad x=\sinh\phi,\quad \tanh 2\phi\cong\eta\ .$$

Transforming the sums into integrals over k space we finally find

$$\langle p_{k,\alpha}\rangle=\frac{2z^{3/2}(u^2+v^2)}{2\pi^2}\int_{(\eta^2-1)}^{\infty}\frac{x[x^2-(\eta^2-1)]^{1/2}}{e^{\Delta x/kT-1}}\,dx\ , \tag{20}$$

$$\langle p_{k,\beta}\rangle=\langle p_{k,\gamma}\rangle=\frac{(w^2+x^2)\Delta^3}{2\pi^2D^3}\int_0^{\infty}\frac{x^2dx}{e^{\Delta x/kT-1}}\ . \tag{21}$$

The integrals in (20) have been evaluated numerically as a function of kT/Δ, and Eq. (18) has been solved graphically for the transition temperature as a function of η. The angular mo-

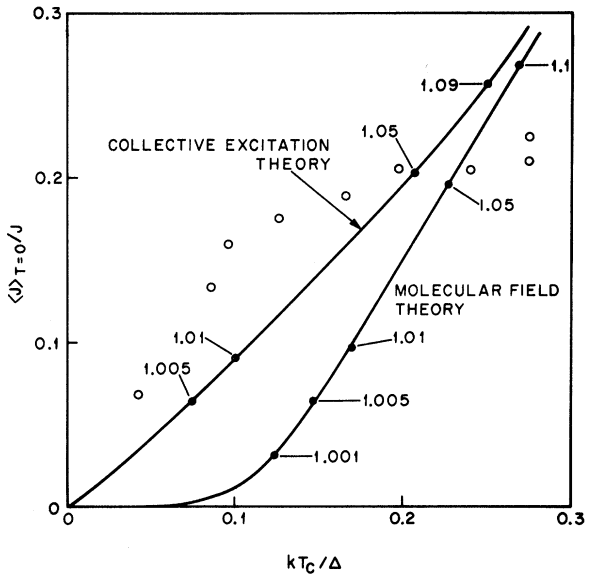

FIG. 3. Reduced ordered moment at zero degrees vs reduced ordering temperature. The numbers denote values for the exchange parameter η.

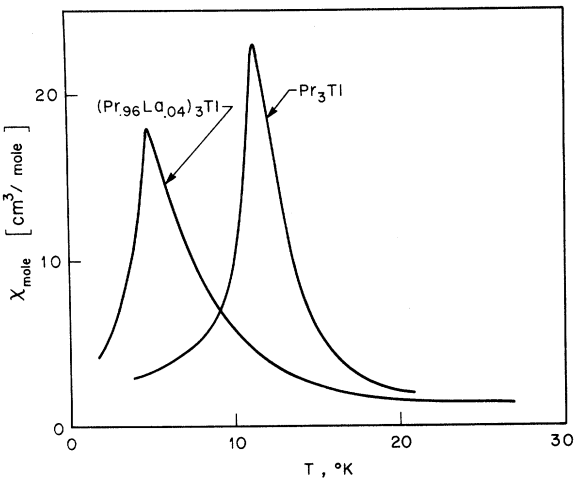

FIG. 5. Initial susceptibility vs temperature of Pr_3Tl and $(Pr_{0.96}La_{0.04})_3Tl$.

mentum in the ordered state is given (as in the molecular-field approximation) by Eq. (7). The resulting relationship between $\langle J_z \rangle_{T=0}$ and T_C for the singlet-triplet case with overcritical exchange interactions, calculated according to Eqs. (7), (18)–(20), is again plotted in Fig. 3. As expected, this result differs drastically from the molecular-field result. The transition temperatures are much reduced for small ordered moments. This is due especially to the existence of the transverse modes, which are still appreciably populated at low temperatures. Although these modes do not contribute directly to the magnetization in the z direction, they help in depopulating the ground state at low temperatures, thereby reducing its moment quite effectively.

IV. DISCUSSION OF EXPERIMENTAL RESULTS

A. Susceptibility and Magnetization

Figure 5 shows the initial susceptibility versus temperature of pure Pr_3Tl and of $(Pr_{0.96}La_{0.04})_3Tl$, measured with an ac technique ($H = 10$ Oe, $\nu = 30$ Hz). The Curie temperatures T_C have been taken to be the temperatures of maximum susceptibility. The variation of T_C with La concentration is tabulated in Table I and shown in Fig. 6:

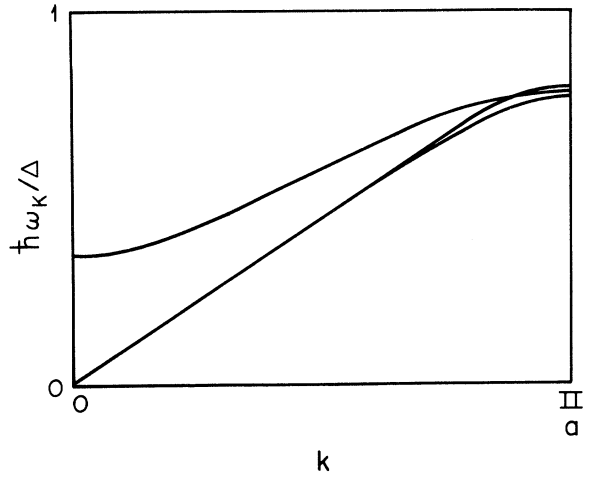

FIG. 4. Plot of the dispersion relations of the modes α_k, β_k, γ_k for $\eta = 1.06$ (case of Pr_3Tl).

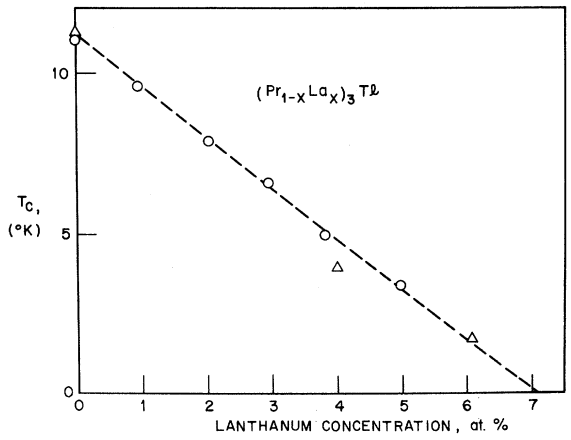

FIG. 6. Ferromagnetic transition temperatures of $(Pr_{1-x}La_x)Tl$ alloys.

TABLE I. Reduced ordered moment, ferromagnetic transition temperature, and critical parameter for $(Pr_{1-x}La_x)_3Tl$ alloys.

La conc. (at. %)	$\langle J\rangle/J$	T_C (°K)	$\eta=\left(\frac{1}{1-(J/\alpha)^2}\right)^{1/2}$
0	0.212(0.224)	11(11.3)	1.058
0.95	0.206	9.6	1.055
2.0	0.208	7.9	1.056
2.95	0.190	6.6	1.046
3.8	0.176	4.9	1.039
4.0	0.159	3.95	1.031
4.97	0.135	3.4	1.022
6.06	0.069	1.65	1.0057

An almost linear decrease is found. A La concentration of only 7% is necessary to depress T_C to zero. This behavior is typical for an induced-moment ferromagnet; it should be contrasted with the case of exchange-coupled local moments, where the corresponding decrease in the Curie temperature would be much smaller. In Fig. 7 we show magnetization measurements taken at 1.1 °K on Pr_3Tl and on $(Pr_{0.95}La_{0.05})_3Tl$. The ordered moment has to be determined by extrapolation, since the ordered ground state is still Van Vleck paramagnetic in high fields. "Saturation" is achieved in comparatively low fields, indicating a small magnetic anisotropy. Small magnetic anisotropy can be expected, at least for small ordered moments, since the Van Vleck susceptibility in the paramagnetic state and in small fields is isotropic. In Grover's theory, this is reflected by the fact that the energy of the $k=0$ mode of the transverse excitations is zero (which leads to infinitely large transverse magnetic susceptibility), while the energy of the $k=0$ longitudinal mode is finite (leading to a finite longitudinal susceptibility). For larger ordered moments one can expect cubic anisotropy to develop in the ordered state. In that case it is no longer sufficient to consider only the ground state and the Γ_4 excited triplet, one also has to take into account the Γ_3 doublet (lying above the Γ_4 triplet) which couples to the Γ_4 triplet. One then can expect the $k=0$ transverse modes to develop energy gaps. A further source of magnetic anisotropy in the ordered ground state is the magnetoelastic energy, which we have neglected so far. Judging from the observed "soft" magnetic behavior of Pr_3Tl, however, it seems that both the singlet-triplet approximation as well as the assumption of a rigid lattice are still good enough to explain the static magnetic measurements. A plot of the observed moments measured in units of the full Pr^{3+} moment $g\mu_B J$ $(3.2\mu_B)$ is shown in Fig. 8. It can be seen that the ordered moment in Pr_3Tl is only 22% of the full Pr^{3+} moment, a fact which is typical of induced-moment ferromagnetism. According to Eq. (7) we compute a value of 1.06 for the parameter η for Pr_3Tl. We feel that the singlet-triplet case treated in Sec. III is a good approximation to the case of Pr_3Tl for the following reason: Each unit cell in the Cu_3Au structure of Pr_3Tl (see Fig. 3 of preceding paper by Bucher *et al.*[1]) contains three different Pr sites which have four equidistant Tl and eight Pr nearest neighbors each. This makes the local symmetry tetragonal. However, if one assumes that both the Tl and Pr ions are in a triply ionized state, the deviation from cubic symmetry (with 13 equal nearest neighbors) should be small and a singlet ground state with a next-higher excited triplet state should be a good approximation to the actual crystal field level scheme. To compare our results with theory, we plot the reduced moment $\langle J\rangle_{T=0}/J$ vs kT_c/Δ in Fig. 3. A value of $k\times 40$ °K has been chosen in the plot. This value is considerably lower than the value of 78 °K which has been deduced from measurements of the specific heat in dilute $(La_{1-x}Pr_x)_3Tl$ alloys where exchange effects can be neglected (see preceding article[1]). It can be seen from Fig. 3 that the experiments do not follow the predicted curve for the collective excitation model. However, the

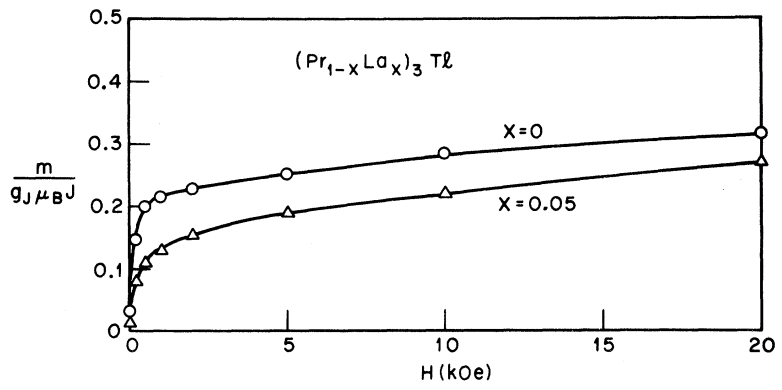

FIG. 7. Magnetization measurements of $(Pr_{1-x}La_x)_3Tl$ alloys.

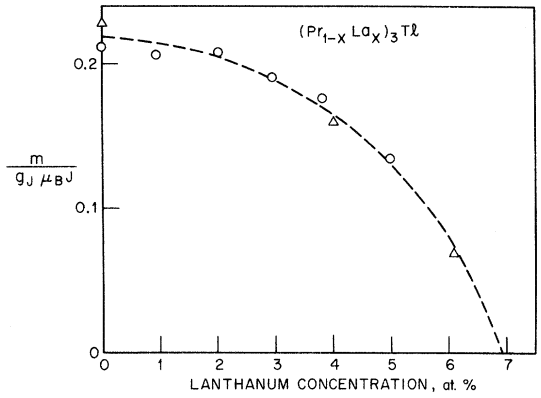

FIG. 8. Reduced ordered moment at 1.1 °K of $(Pr_{1-x}La_x)_3Tl$ alloys.

latter model does come much closer to the experimental results than the molecular-field model, especially in that it reproduces the observed nearly linear relationship between m_0 and T_C in the near-critical case. As pointed out in Sec. III, this improvement is due mostly to the presence of the gapless transverse excitation modes.

B. Specific Heat

Specific-heat measurements on pure Pr_3Tl are shown in Fig. 9. The anomaly observed at the transition temperature of 11 °K is much smaller than what one would expect in the case of magnetic ground-state ions which undergo magnetic order. This is in agreement with the prediction for an induced-moment system: If the ordering temperature is much smaller than the gap Δ, little magnetic entropy is left at the transition temperature since the higher-excited collective crystal-field excitations are already thermally depopulated. In the molecular-field approximation, the transition is of second order, and the discontinuity of the specific heat at T_C is given approximately by

$$\left(\frac{\Delta C}{R}\right)_{T=T_C} = 2x^3 e^{-2x}[1 + 2(x-1)e^{-x} + O(e^{-2x})], \tag{22}$$

where x equals Δ/kTc. For pure Pr_3Tl the transition temperature is 11 °K. Taking a value of Δ/k of 40 °K, one finds for $\Delta C/R$ at the transition a value of 0.076. The observed "hump" in the specific heat of Pr_3Tl around 10 °K is indeed of this order of magnitude. With a value of Δ/k of 78 °K, Eq. (21) would yield a value of 4.9×10^{-4} for $\Delta C/R$ at the transition. We cannot use Eq. (21), however, to determine a value of Δ/k from the observed specific-heat anomaly, since this equation neglects entirely the existence of the collective excited modes. Unfortunately, no explicit calculations for $(\Delta C/R)_{T=T_C}$ are available at present which take into account the collective excited modes.

C. Resistivity

Electrical resistivity measurements on Pr_3Tl are shown in Fig. 10. No anomaly at all can be detected at the transition temperature. This again is in qualitative agreement with the fact that the magnetic entropy change at the transition is small. Of special interest is the temperature dependence of the resistivity at low temperatures. In Fig. 11, $\log\Delta\rho(T)$ is plotted vs $\log T$ below 7 °K $[\Delta\rho(T) = \rho(T) - \rho_0]$. We find that in this range $\Delta\rho(T)$ varies with $T^{2.8}$. The resistivity of the nonmagnetic analogue of Pr_3Tl, namely, La_3Tl, is also plotted in Fig. 11. These latter measurements had to be taken in a field of 50 kOe in order to suppress superconductivity. Zero-field data below the superconducting transition temperature of 8.9 °K were obtained by subtracting the magnetoresistive part, using Kohler's rule. We find that $\Delta\rho(T)$ of La_3Tl is about an order of magnitude smaller than $\Delta\rho(T)$ of Pr_3Tl below 7 °K and varies as $T^{3.7}$. In Pr_3Tl, a higher value of $\Delta\rho(T)$ is to be expected because of the additional scattering processes between conduction electrons and the collective magnetic excitations. Below the ferromagnetic transition tem-

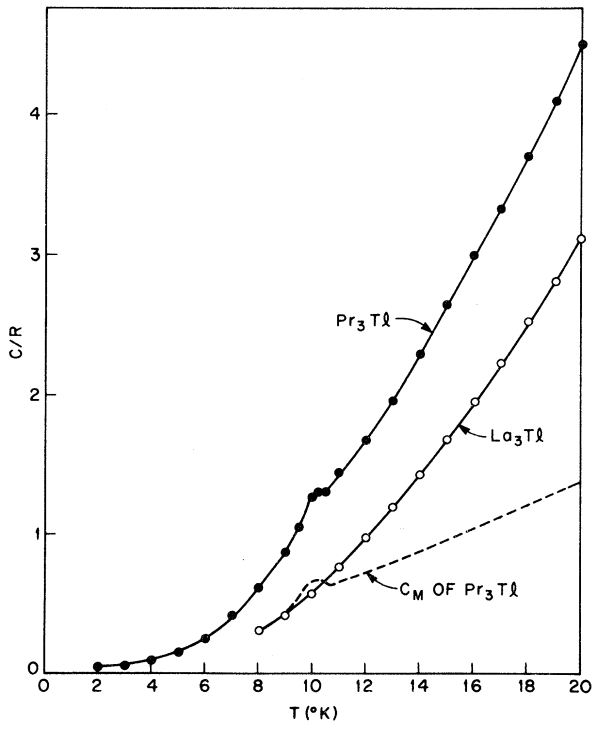

FIG. 9. Specific heat of Pr_3Tl and La_3Tl.

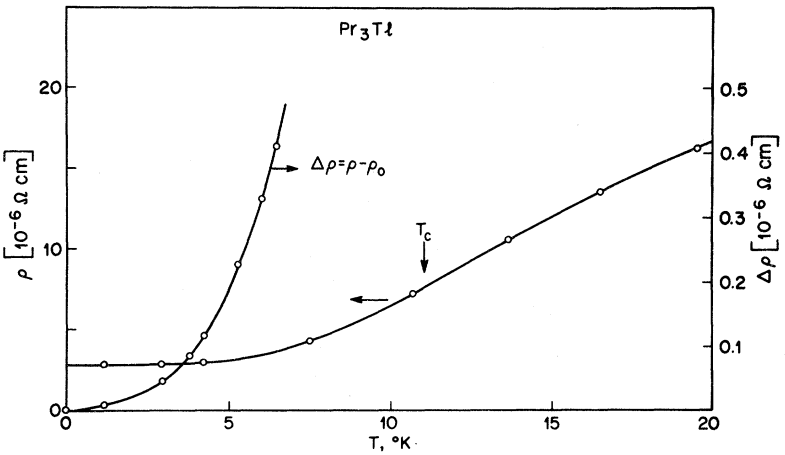

FIG. 10. Resistivity of Pr_3Tl below 20 °K.

perature, the dominant "magnetic" contribution to $\Delta\rho(T)$ comes from scattering with the transverse modes β_k and γ_k. If those modes have phononlike dispersion relations, one would expect the "magnetic" contribution $\Delta\rho_{mag}(T)$ of Pr_3Tl to have the same temperature dependence as $\Delta\rho(T)$ of La_3Tl. Experimentally, $\Delta\rho_{mag}(T)$ varies as $T^{2.8}$, whereas $\Delta\rho(T)$ of La_3Tl varies as $T^{3.7}$. This would mean that the low-lying transverse modes die out somewhat slower than predicted by theory as one approaches zero degrees.

V. CONCLUSIONS

The data presented here on Pr_3Tl show conclusively that this compound is a case of an induced-moment ferromagnet. One of the interesting features of such a magnet is that both the ordered moment per ion and the Curie temperature approach zero as the exchange interaction becomes critical. We have, to our knowledge, for the first time demonstrated this behavior experimentally in the alloy system $(Pr_{1-x}La_x)_3Tl$. We find that the molecular-field theory is inadequate to explain the results since it completely neglects the presence of low-lying collective excited modes. A comparison with Grover's theory which predicts the presence of such modes yields better agreement with experiment. It should be pointed out that recent neutron scattering experiments have been carried out on polycrystals of Pr_3Tl in an attempt to observe the dispersion of the collective excited modes directly.[8,9] Modes similar to the longitudinal one depicted in Fig. 4 have been found, but no temperature dependence was seen and there was so far no evidence of low-lying transverse modes.

As pointed out before, we have analyzed our results in the singlet-triplet (Γ_1 and Γ_4) approximation and neglected the higher-lying Γ_3 state which couples to the Γ_4 triplet. The inclusion of the Γ_3 state introduces cubic anisotropy and creates energy gaps in the transverse modes. However, from our observed relationship between ordered moment and transition temperature, from the small magnetic anisotropy, and from the magnetic contribution to the resistivity at low temperatures, we conclude that these gaps cannot be too large and that collective excitations with energies considerably smaller than kT_C (T_C = transition temperature) must exist.

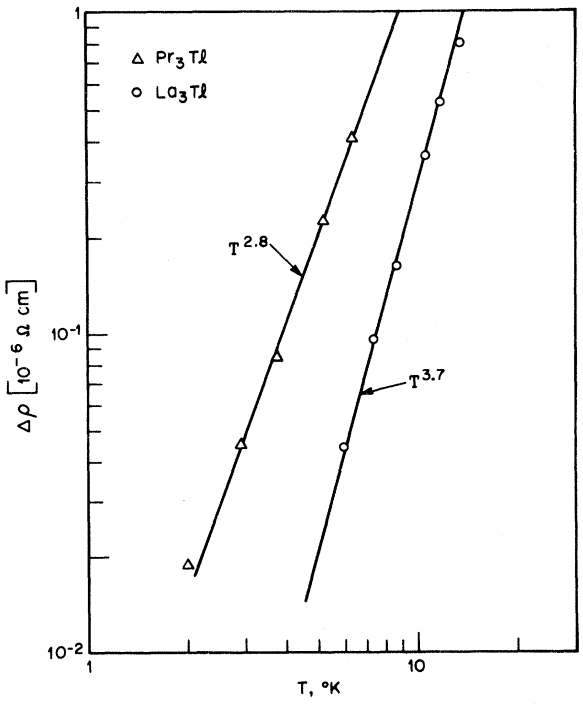

FIG. 11. Logarithmic plot of the temperature-dependent part of the resistivity in Pr_3Tl and La_3Tl vs temperature.

The true excitations may possibly have a very short lifetime and no well-defined wave number. This would make it more difficult to observe them by means of neutron scattering techniques.

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Neutron Scattering from fcc Pr and Pr_3Tl

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Elastic-neutron-scattering measurements on the singlet-ground-state ferromagnets fcc Pr and Pr_3Tl are reported. Both exhibit magnetic phase transitions, possibly to a simple ferromagnetic state at 20 and 11.6 °K, respectively. The transitions appear to be of second order although that in fcc Pr is clearly anomalous. Additional information on the inelastic scattering studies of the Γ_1-Γ_4 excitons in these systems is presented. dhcp Pr is also briefly discussed.

In a recent paper,[1] we have reported inelastic-neutron-scattering studies of the elementary magnetic excitations, crystal field excitons, in the singlet-ground-state ferromagnets fcc Pr and Pr_3Tl. It was found that well-defined excitons existed in both the ferromagnetic and paramagnetic regimes but with energies which were nearly independent of temperature. This latter result was most surprising in light of our expectations based on the existing theory which predicted both soft-mode behavior at the phase transition and a marked overall temperature dependence of the exciton energies.[2] In this paper we report complementary elastic-neutron-scattering results which elucidate the magnetic structure; we also present additional experimental information on the inelastic studies in order to facilitate any hoped for attempts at a theoretical interpretation of these results. In two accompanying papers by Bucher, Maita, and Cooper[3] and by Andres, Bucher, Darack, and Maita[4] the bulk magnetic properties of the system $(Pr_{1-x}La_x)_3Tl$ as a function of x are presented. In a third paper Cooper[5] discusses an effective boson theory for the excitations of singlet ground state Pr in a cubic environment. This set of papers, therefore, gives a complete exposition of the available experimental and theoretical information on induced-moment ferromagnetism in Pr_3Tl. It is hoped that they will lay an effective groundwork for future studies.

For completeness we briefly review the theory. The Pr^{3+} ion has the configuration $4f^2$ with the free-ion $^3H_4$ multiplet lying lowest. For a cubic system the crystal-field Hamiltonian may be written

$$\mathcal{H}_{CF} = A_4\langle r^4\rangle \chi_4 [O_4^0(J) + 5\,O_4^4(J)] + A_6\langle r^6\rangle \chi_6 [O_6^0(J) - 21\,O_6^4(J)], \quad (1)$$

where the O_n^m are Stevens operator equivalents and the χ_n are reduced matrix elements.[6] The consequent single-ion energy levels for fcc Pr with the