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Combined Electron-Nuclear Magnetic-Ordering Phenomena in Singlet-Ground-State Systems

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The magnetic behavior of a system of singlet-ground-state ions coupled by exchange interactions is considered in the molecular-field approximation. It is shown that when hyperfine interactions are included, the critical exchange necessary for spontaneous induced-moment order is shifted to slightly lower values. For still smaller values of exchange, cooperative nuclear magnetic order occurs in the system of the singlet-ground-state ions. The recently reported results on nuclear magnetic order in $PrCu_2$ can be understood by assuming that in $PrCu_2$ the exchange interactions are very close to the threshold value which divides between cooperative electronic and nuclear magnetic order.

I. INTRODUCTION

In a recent letter[1] we reported the observation of spontaneous nuclear magnetic order of the Pr nuclei at 54 mK in the compound $PrCu_2$. This surprisingly high ordering temperature could not be understood on the basis of a second-order perturbation calculation, according to which the hyperfine-induced $4f$ moments[2] in the singlet ground state (which have the nuclear degeneracy $2I+1$) are coupled by the $4f$-$4f$ exchange interactions. Such a calculation yielded an ordering temperature of 2.3 mK. The enhancement to 54 mK is due to the proximity of the exchange interactions in $PrCu_2$ to the critical value for electronic-induced-moment order. In this note we show that as one approaches this critical value, one continuously goes from a state of cooperative nuclear magnetic order to a state of induced-moment order, the transition temperature rising roughly as $(\lambda_{crit}-\lambda)^{-3}$ in the ferromagnetic case (λ is the molecular-field constant). It is shown that to a good approximation the transition temperature can be simply related to the molecular-field constant, the nuclear susceptibility, and the crystal-field-only Van Vleck susceptibility. In what follows we first recall briefly the molecular-field (MF) results for the two singlet level system with exchange and then consider the same system including hyperfine interactions. We calculate the hyperfine-enhanced nuclear susceptibility above the ordering temperature and finally discuss the nature of the phase transition in the pure electronic, mixed electronic-nuclear, and pure nuclear ordering regimes.

II. INDUCED-MOMENT SYSTEM WITHOUT HYPERFINE INTERACTIONS

We consider the Hamiltonian

$$H=\sum_i V_{c,i}-\sum_{i,j} K_{ij}J_iJ_j \tag{1}$$

of a system of non-Kramers rare-earth ions subjected to the local crystal field V_c and coupled by exchange interactions K_{ij}. In the MF approximation this Hamiltonian is written, per ion, as

$$H=V_c-2K_0\langle J\rangle J, \quad K_0=\sum_i K_{ij}, \tag{2}$$

$\langle J\rangle$ being the average angular momentum of all ions. We now suppose that the eigenstates of V_c are two singlets designated by $|1\rangle$ and $|-1\rangle$ and separated by an energy Δ:

$$\langle 1|V_c|1\rangle=0, \quad \langle -1|V_c|-1\rangle=\Delta.$$

Such systems have been treated first by Trammell[3] and Bleaney.[4] Diagonalization of H leads to the new eigenfunctions

$$\begin{aligned}|1'\rangle&=\cos\alpha|1\rangle+\sin\alpha|-1\rangle,\\ |-1'\rangle&=-\sin\alpha|1\rangle+\cos\alpha|-1\rangle,\end{aligned} \tag{3}$$

with

$$\tan 2\alpha=\frac{\eta\langle J\rangle}{c}, \quad \eta=\frac{4K_0c^2}{\Delta}, \quad c=\langle 1|J|-1\rangle.$$

The new ground-state moment is

$$\langle 1'|J|1'\rangle=c\sin 2\alpha=\frac{c(\eta^2-1)^{1/2}}{\eta}. \tag{4}$$

Thus the singlet ground state remains stable for $\eta < 1$ and the critical exchange interaction for the appearance of a self-induced moment is given by $\eta = 1$. (For simplicity, we assume here only positive values of η, i.e., ferromagnetic exchange interactions.) The new ground-state energy is

$$\langle 1' \mid H \mid 1' \rangle = -\tfrac{1}{2}\Delta(\eta - 1), \quad 0 < \eta - 1 \ll 1 . \tag{5}$$

The transition temperature to the paramagnetic state can be computed from the requirement that the thermal average of the moment of both singlet states vanishes:

$$\langle J \rangle_{T_c} = p_1 \langle J_1 \rangle + p_2 \langle J_2 \rangle = 0, \tag{6}$$

where $p_{1,2}$ are the population probabilities of two states. In the limit $|\eta| - 1 \ll 1$, Eq. (6) yields the relation

$$\tanh \frac{\Delta}{2kT_c} = \frac{1}{\eta} . \tag{7}$$

The Van Vleck susceptibility of the two singlet states is given by

$$\chi_v(T) = g_J \mu_B \langle J \rangle_T / H.$$

Calculation yields

$$\chi_v(T) = \frac{\chi_c(T)}{1 - \lambda \chi_c(T)} , \tag{8}$$

with

$$\lambda = \frac{2K_0}{g_J^2 \mu_B^2} \quad \text{and} \quad \chi_c(T) = \frac{2c^2}{\Delta} g_J^2 \mu_B^2 \tanh \frac{\Delta}{2KT} .$$

$\chi_c(T)$ is the crystal-field-only susceptibility (without exchange interactions). It can be seen that Eq. (7) is identical to the equation

$$\chi_c(T)\lambda = 1. \tag{9}$$

Equation (9) says that the transition temperature to the induced-moment state can directly be read off a plot of $\chi_c(T)^{-1}$ vs T, as sketched in Fig. 1. It is that temperature at which $\chi_c(T_c)^{-1}$ equals λ.

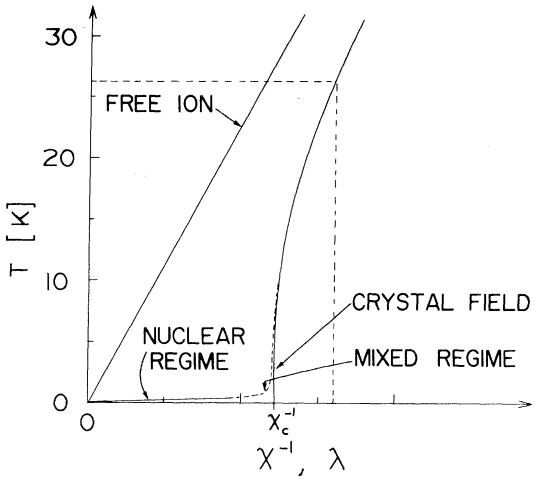

FIG. 1. Inverse crystal-field-only Van Vleck susceptibility plotted against temperature. For overcritical exchange interactions $[\lambda > \chi_c^{-1}(0)]$ the transition temperature to the induced-moment state is given by $\chi_c^{-1}(T_c) = \lambda$. Also schematically indicated in the figure are the nuclear-ordering temperatures as a function of λ for undercritical values $[\lambda < \chi_c^{-1}(0)]$ in both the nuclear and (dashed) the mixed regime (see text).

III. INDUCED-MOMENT SYSTEMS WITH HYPERFINE INTERACTIONS

Instead of the Hamiltonian (2) we now consider the Hamiltonian

$$H = V_c - 2K_0 \langle J \rangle J + AJI . \tag{10}$$

A Hamiltonian similar to Eq. (10) has been treated for the first time by Murao.[5,6] He assumed that the eigenstates of V_c were a low-lying singlet and excited doublet at energy Δ, and that these three states could be described as the result of the orbital splitting of an $S = 1$ triplet.

We again start from the eigenstates of V_c which we assume to be singlets separated by an energy Δ. We further assume a nuclear spin of $I = \frac{1}{2}$, so that each singlet state has two nuclear substates, which we designate by $|-1^+\rangle$, $|-1^-\rangle$. Since the nuclear spin will remain a good quantum number, all matrix elements involving products of nuclear wave functions of unlike spin will be zero. The new eigenstates therefore can be written in the form

$$\begin{aligned}
|1'^+\rangle &= \cos\alpha^+ \mid 1^+\rangle + \sin\alpha^+ \mid -1^+\rangle , \\
|-1'^+\rangle &= -\sin\alpha^+ \mid 1^+\rangle + \cos\alpha^+ \mid -1^+\rangle , \\
|1'^-\rangle &= \cos\alpha^- \mid 1^-\rangle + \sin\alpha^- \mid -1^-\rangle , \\
|-1'^-\rangle &= -\sin\alpha^- \mid 1^-\rangle + \cos A^- \mid -1^-\rangle ,
\end{aligned} \tag{11}$$

with

$$\tan 2\alpha^+ = 2c(2K_0 \langle J \rangle - AI^+)/\Delta ,$$

$$\tan 2\alpha^- = 2c(2K_0 \langle J \rangle - AI^-)/\Delta .$$

The ground-state moment is given by

$$\langle J_1^+ \rangle = c \sin 2\alpha^+ . \tag{12}$$

Equation (12) is a self-consistent equation for $\langle J_1^+ \rangle$, which at $T = 0$ can also be written in the form

$$y = (\eta y - a)/[1 + (\eta y - a)^2]^{1/2} , \tag{13}$$

with

$$y = \langle J_1^+ \rangle / c, \quad a = 2AcI^+/\Delta, \quad \eta = 4K_0 c^2/\Delta ,$$

or in the form

$$y = (\eta^{*2} - 1)^{1/2}/\eta^*, \quad \eta^* = \eta - a/y . \tag{14}$$

Equation (14) is similar to Eq. (4) and indicates that the critical value of η for a spontaneous in-

duced moment is now shifted to lower values by an amount a/y. For small values of η the approximate solution of Eq. (13) is

$$y=\frac{\langle J_1^+\rangle}{c}=\frac{-a}{1-\eta}=\frac{-2AcI^+}{\Delta(1-\eta)} \quad . \tag{15}$$

Equation (15) tells us that $-ac$ is the $4f$ angular momentum that the hyperfine interaction alone admixes to the two nuclear substates of the singlet ground state. Exchange interactions enhance this moment by $1/(1-\eta)$. The solution of the self-consistent equation (13) for the ground-state moment is shown in Fig. 2, together with the solutions (4) and (15). The transition temperature to the paramagnetic state can again be obtained by computing $\langle J\rangle_T$ of all states and asking for a zero at T_c. This is easy to do in the nuclear regime where one only has to consider the two lowest nuclear substates with $4f$ angular moments $\mp a/(1-\eta)$ and with the energy separation $4K_0[a^2/(1-\eta)^2]$ (at 0 K). The calculation of the transition temperature yields

$$kT_c=\frac{2K_0a^2c^2}{1-\eta} \quad . \tag{16}$$

This expression is similar to the one found by Murao.[5]

If one had assumed the hyperfine-induced and exchange-enhanced moments $\mp ca/(1-\eta)$ to be rigid, one would have obtained the ordinary MF formula

$$kT_c=2K_0\left(\frac{ac}{1-\eta}\right)^2 \tag{17}$$

for the transition temperature (for the special case of $I=\frac{1}{2}$). The difference between Eq. (16) and Eq. (17) comes from the fact that in obtaining (16) one reduces the exchange enhancement $1/(1-\eta)$ to zero as one approaches T_c from below. This is somewhat unphysical since the nuclear susceptibility is still exchange enhanced above T_c. To calculate the nuclear susceptibility one must include the terms $-g_J\mu_BHJ-g_N\mu_NHI$ in Eq. (10) and again calculate the moments of the two lowest nuclear substates. We obtain

$$\langle J_1^{+-}\rangle=\pm\left(|I|-\frac{|ac|}{1-\eta}\right)+b \ , \quad b=2cg_J\mu_BH\Delta, \tag{18}$$

with the parameter a defined in Eq. (13). Here b is the field-induced Van Vleck moment which is superimposed upon the hyperfine-enhanced nuclear moment $[I-ac/(1-\eta)]$. The (hyperfine- and exchange-enhanced) nuclear susceptibility is obtained from the Zeeman splitting of the latter moments. At temperatures much higher than the nuclear-ordering temperature (but still in the regime where the Van Vleck susceptibility is nearly temperature independent), calculation yields

$$\chi_{N,\infty}=\frac{g_N^2\mu_N^2I^2(1+K)^2}{kT} \ , \quad K=\frac{g_J\mu_B}{g_N\mu_N}\,\frac{2Ac^2}{\Delta}\,\frac{1}{(1-\eta)} \tag{19}$$

for our special case of $I=\frac{1}{2}$. Equation (19) says that in the Van Vleck paramagnetic regime the local field at the nucleus is enhanced by the factor $(1-K)$, where K can be interpreted as a Knight shift due to the $4f$ electrons which is proportional to the Van Vleck susceptibility

$$K=h_f\chi_v \ , \quad h_f=\frac{-A}{g_J\mu_Bg_N\mu_N} \quad . \tag{20}$$

If one guesses that closer to the nuclear-ordering temperature, the nuclear susceptibility assumes the MF form

$$\chi_N=\frac{\chi_{N,\infty}}{1-\lambda'\chi_{N,\infty}} \ , \tag{21}$$

then the nuclear-ordering temperature should be given by the condition

$$\lambda'\chi_{N,\infty}(T_c)=1. \tag{22}$$

If $\lambda'=\lambda[K/(1+K)]^2$, then Eq. (22) indeed yields Eq. (17). The reason that λ' instead of λ enters in Eq. (21) is that the exchange forces act only on the hyperfine-induced $4f$ moments and not on the bare nuclear moments.

Experimentally[7,8] it has been found that the nuclear susceptibility does indeed follow the form (21) over a wide temperature range down to temperatures close to the ordering temperature. The parameter K determined experimentally from Eq. (21) always agrees with the one found from relation (20). Examples of such measurements are

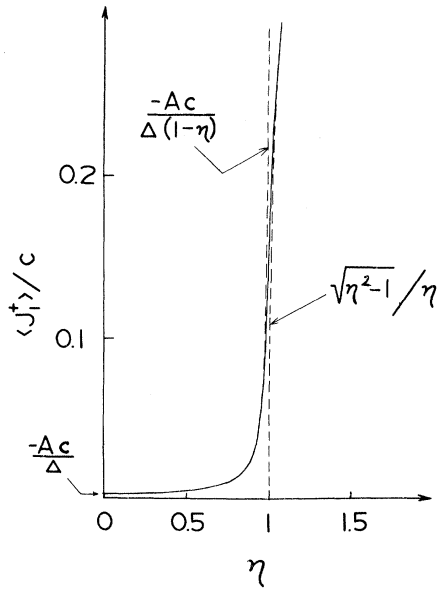

FIG. 2. Exchange-induced ground-state moment as a function of the critical parameter $\eta=\lambda\chi_c=4K_0c^2/\Delta$.

shown in Figs. 3 and 4, where the inverse of the nuclear susceptibility is plotted versus temperature at very low temperatures. The nuclear susceptibility is the difference between the observed susceptibility and the Van Vleck susceptibility (the latter being the observed susceptibility above 1 K). We therefore believe that Eq. (22) is a better approximation to the actual nuclear-ordering temperature than Eq. (16).

For exchange interactions closer to the critical value, the exact calculation of the transition temperature is more complicated and has to be done numerically by iterative procedures. However, we can anticipate that ordering temperatures in the critical regime can approximately be obtained by simply looking for singularities in the *total* susceptibility $\chi_v + \chi_N$:

$$\chi_v + \chi_N = \frac{\chi_c(T) + \chi_{N,\infty}(T)}{1 - \lambda[\chi_c(T) + \chi_{N,\infty}(T)]} \quad . \tag{23}$$

Here we have neglected the difference between λ' and λ, which is usually small (for typical Pr intermetallic compounds, K is usually between 10 and 20). Going back to Fig. 1 this now means that in order to find the ordering temperature we have to intersect the ordinate at η with the inverse of the *total* susceptibility, and it is clear that one always will find a solution even if $\eta < 1$ because of the nuclear tail of χ^{-1}. The denominator in Eq. (23) is zero for

$$T_c = \frac{\lambda c_{N,\infty}}{1 - |\lambda|\,\chi_c(T)} \simeq \frac{\lambda c_{N,\infty}}{1 - |\eta|} \quad , \tag{24}$$

$$c_{N,\infty} = g_N^2 \mu_N^2 I^2 (1+K)^2/k$$

$$= g_N^2 \mu_N^2 I^2 \left(1 - \frac{g_J \mu_B}{g_N \mu_N} \frac{2Ac^2}{\Delta(1-\eta)}\right)\Big/ k. \tag{25}$$

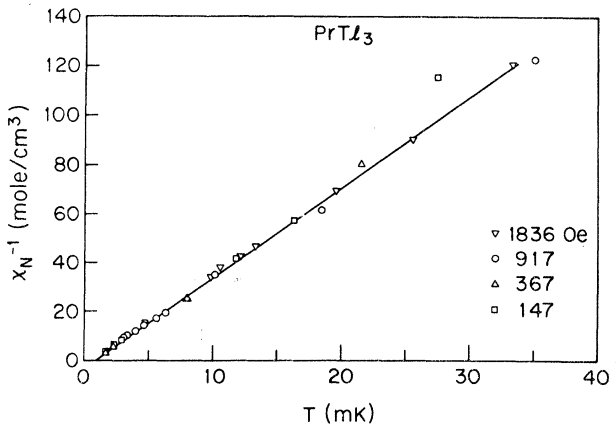

FIG. 3. Inverse nuclear magnetic susceptiblilty of $PrTl_3$ at very low temperatures, plotted against temperature.

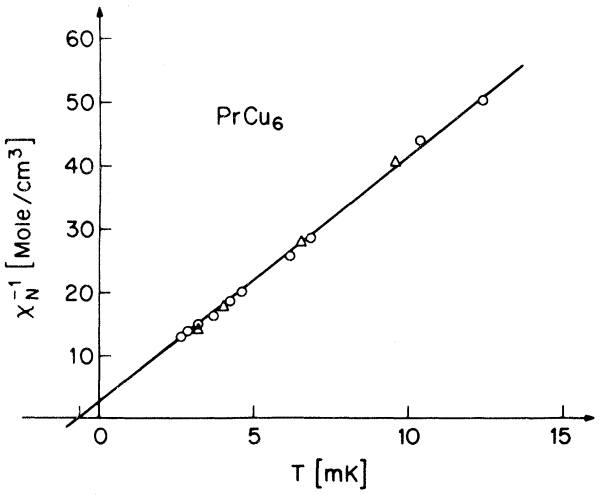

FIG. 4. Inverse nuclear magnetic susceptibility of $PrCu_6$ at very low temperatures, plotted against temperature. Shown are the results of two runs in $H = 550$ Oe.

Equation (24) holds at temperatures low enough so that $\chi_c(T)$ can be replaced by $\chi_c(0)$. It shows that for η close to 1, the nuclear-ordering temperature (22) is enhanced by $1/(1-\eta)$ in the ferromagnetic case. In the antiferromagnetic case, the absolute value of η will enter (24), whereas a negative value will enter (25) (the nuclear susceptibility is deenhanced by antiferromagnetic exchange interactions). This means that for the same absolute value of η the transition temperature is higher for ferromagnetic interactions. Equation (24) should be a good approximation to about $\eta = 0.9$. Between $\eta = 0.9$ and $\eta = 1.0$, T_c can easily be determined by interpolation. In the case of a nuclear spin different from $\frac{1}{2}$, I^2 in (25) has to be replaced by $I(I+1)/3$. Figure 5 is a log-log plot of T_c vs η determined in this way for the case of Pr compounds with $\Delta = k \times 52.5$ K ($I = \frac{5}{2}$, $A/\Delta = 10^{-3}$). This result is qualitatively similar to that obtained by Murao.[6]

IV. DISCUSSION OF PHASE TRANSITION AND COMPARISON WITH EXPERIMENTS

In the regime of electronic order ($\eta > 1$ and slightly below 1) the relation

$$kT_c > 2A \langle J_1^+ \rangle I$$

holds, which means that the thermal energy at the transition is larger than the energy splitting of the lowest two nuclear substates. In this case both the electronic and nuclear entropy reduction at the transition will be small and the bulk of the nuclear entropy comes out only below T_c when the nuclear moments gradually align in the hyperfine field of the exchange-induced $4f$ moment of the ground state. There will be a gradual increase in

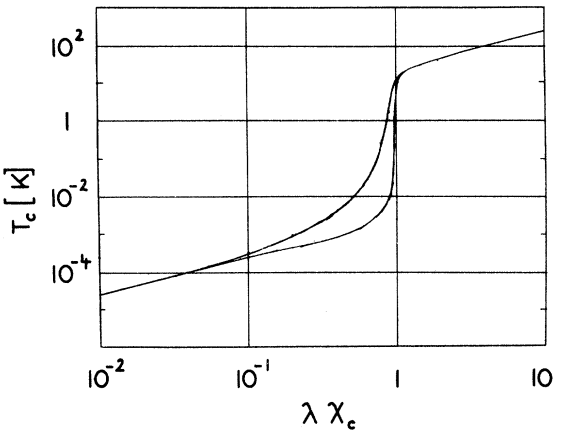

FIG. 5. Approximate variation of the transition temperature with the critical parameter $\lambda\chi_c$ $(=\eta)$ [solutions of Eqs. (7) and (24)] for both ferromagnetic (upper curve) and antiferromagnetic exchange interactions (lower curve). The threshold value of η which divides between the nuclear- and electronic-order regimes is 0.65 and 0.89, respectively (see text). The values $\Delta/k = 52.5$ K, $A/k = 52.5 \times 10^{-3}$ K and $I = \frac{5}{2}$ (for ^{141}Pr) have been used.

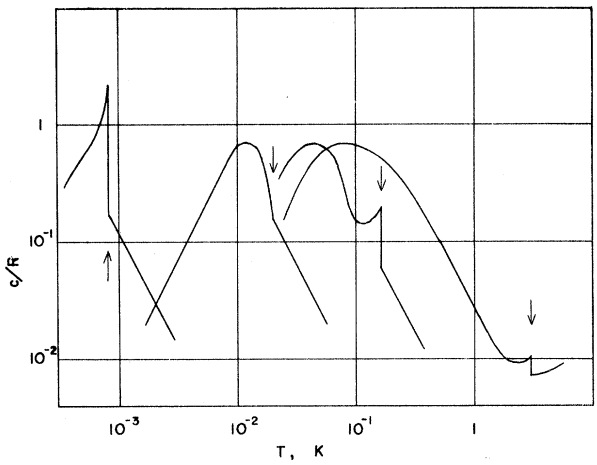

FIG. 6. Specific-heat anomalies expected in MF approximation in the electronic regime (right-hand curve), and the mixed regime (center curves). The arrows indicate the magnetic-ordering temperatures.

magnetization below T_c, which can be interpreted as the gradual alignment of the hyperfine-enhanced moments (15) in the molecular field of the ordered state. This behavior has also been found by Murao in the exact MF calculation of his model. In the regime of nuclear order on the other hand, the relation

$$kT_c < 2A\langle J_1^+\rangle I$$

holds. In this case the two nuclear sublevels spontaneously split at T_c and one has a large nuclear entropy reduction at the transition. The threshold temperature between the two regimes is approximately given by

$$kT_c = 2A\langle J_1^+\rangle I. \tag{26}$$

Using the approximate forms (15) and (24), respectively, for $\langle J_1^+\rangle$ and T_c as a function of η, one can compute from (26) a threshold value for η. For ferromagnetic exchange one obtains (for $I = \frac{5}{2}$) in this way $\eta \simeq 0.62$, and for antiferromagnetic exchange $\eta \simeq 0.89$, irrespective of the value of Ac^2/Δ. For $A/\Delta = 10^{-3}$ and $c^2 = 20/3$, which is typical for many Pr compounds, these η values correspond to ordering temperatures of 24 and 4.6 mK, respectively, for ferromagnetic and antiferromagnetic exchange interactions. In Fig. 6 we plot the expected shape of the specific-heat anomaly in a log-log plot in both the electronic and the nuclear regime as well as at the threshold between the two. In the electronic regime, the nuclear specific heat will look like an ordinary Schottky-type anomaly, while in the nuclear regime it will look like a second-order magnetic transition. At the threshold it will have a shape in between those two. It is about this shape (the one with an ordering temperature of 2×10^{-2} K in Fig. 6) that was recently observed in $PrCu_2$, which orders antiferromagnetically at 54 mK. With the parameters $A/\Delta = 4\times10^{-3}$ and $c^2 \simeq 9$ appropriate for

TABLE I. Comparison of observed and calculated nuclear and electronic magnetic-ordering temperatures in singlet-ground-state systems. The values of $|\eta|$ have been estimated from the exchange enhancement of the Van Vleck susceptibility (i.e., by comparing the calculated crystal-field-only susceptibility with the observed one).

Material	$\lvert\eta\rvert$	T_c Calculated	T_c Observed	Characterization of transition
$PrTl_3$	0.1	0.41 mK[a]	1.0–1.5 mK (ferro)[b,c]	Nuclear
PrBi	0.15	0.49 mK	~9 mK (ferro)[d]	Nuclear
$PrPt_5$	0.51	2.66 mK	<3 mK (antiferro)[c]	Nuclear
$PrCu_6$	0.56	1.2 mK	<2 mK (antiferro)[e]	Nuclear
$PrCu_2$	Between 0.8 and 1	~Between 10 mK and 1 K	54 mK (antiferro)[f]	Mixed electronic-nuclear
Pr_3Tl	1.058	$kT_c \simeq 0.21\,\Delta$	11 K[g]	Electronic

[a]Reference 14. [b]Reference 8. [c]Reference 12. [d]Reference 13. [e]Reference 7. [f]Reference 1. [g]Reference 11.

$PrCu_2$, the threshold value of T_c would be 37 mK. This would mean that in $PrCu_2$ one is very close to the threshold between electronic and nuclear order, somewhat closer to the electronic side, which explains the shape of the nuclear-specific-heat anomaly observed in $PrCu_2$.

To date, nuclear and electronic magnetic-ordering phenomena have been observed in a number of singlet-ground-state systems.[9-12] A summary of the results that we have obtained so far in praseodymium intermetallic compounds is given in Table I.

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[14]A nuclear ordering temperature for $PrTl_3$ of 1.2 mK has been calculated by Landesman [J. Phys. (Paris) **32**, 671 (1971)] in a different way, namely, by calculating the collective excitations in the singlet-triplet system with exchange interactions and then considering the hyperfine effects to second order. Unfortunately, a critical value of $\eta=0.56$ has been assumed, which we believe does not apply to $PrTl_3$.

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Dielectric Response of the Electron Liquid in Generalized Random-Phase Approximation: A Critical Analysis

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We critically examine various approximate theories which have been put forward within the spirit of generalized random-phase approximation (GRPA) for dielectric response of the degenerate electron liquid at metallic densities. The exchange-correlation contribution to the effective field acting on an electron is expressed in terms of a frequency-independent function $G(\vec{q})$ in GRPA. There are requirements of certain sum rules, e.g., the compressibility sum rule, the fluctuation-dissipation theorem, and the third-frequency-moment sum rule, which impose restrictions on $G(\vec{q})$. The theory of Vashishta and Singwi for $G(\vec{q})$ satisfies the compressibility sum rule and the fluctuation-dissipation theorem, while another recent theory by Pathak and Singwi satisfies the third-moment sum rule and the fluctuation-dissipation theorem. The second work neglects the correlation contribution to the kinetic energy, while the first one takes it into account through the introduction of an *ad hoc* parameter. In this paper, we show that if the correlation kinetic energy part is correctly taken into account then $G(\vec{q})$ cannot be made to satisfy the compressibility sum rule and the third-moment sum rule simultaneously, in the sense that doing this would violate the ground-state-energy theorem of Ferrell.

The dielectric response of the electron liquid is conveniently discussed through a frequency- and wave-vector-dependent dielectric function.[1] The random-phase approximation (RPA) of Nozières and Pines[2] was the first useful theory for this dielectric function, which is given by

$$\frac{1}{\epsilon_{\mathrm{RPA}}(\vec{q},\omega)} = 1 + \phi(\vec{q})\chi_{\mathrm{RPA}}(\vec{q},\omega), \tag{1a}$$

where

$$\chi_{\mathrm{RPA}}(\vec{q},\omega) = \frac{\chi^0(\vec{q},\omega)}{1-\phi(\vec{q})\chi^0(\vec{q},\omega)}, \tag{1b}$$

$\phi(\vec{q})$ is the Fourier transform of the Coulomb potential, and $\chi^0(\vec{q},\omega)$ is the polarizability of free electrons. All the other attempts[3] that have been made to improve upon the RPA result have started from the following form of the density-density response function:

$$\chi(\vec{q},\omega) = \frac{\chi^0(\vec{q},\omega)}{1-\psi(\vec{q})\chi^0(\vec{q},\omega)}, \tag{2a}$$

$$\psi(\vec{q}) = \phi(\vec{q})\,(1-G(\vec{q})), \tag{2b}$$

where an effective potential $\psi(\vec{q})$ enters. The additional term $-\phi(\vec{q})G(\vec{q})$ is the correction due to exchange-correlation effects. It is also called the local-field correction.

There are a number of exact results which $\chi(\vec{q},$