PHYSICAL REVIEW B VOLUME 29, NUMBER 9 1 MAY 1984

Quenching of spin fluctuations in the highly enhanced paramagnets $R\mathrm{Co}_2$ (R =Sc, Y, or Lu)

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(Received 5 December 1983)

The low-temperature (1.3—20.0 K) high-magnetic-field (0—10 T) heat capacity and the magnetization and magnetic susceptibility (1.7—300 K) of the strongly Pauli paramagnetic $R\mathrm{Co}_2$ (R =Sc, Y, or Lu) compounds with the MgCu_2-type structure were measured. The heat-capacity results for ScCo_2, YCo_2, and LuCo_2 show that the electronic specific-heat constant decreases with increasing magnetic fields (by 7%, 4%, and 10%, respectively, at 10 T). For YCo_2 the coefficient of the T^3 term (β) in the heat capacity is found to increase by 18% at 10 T, but for ScCo_2 and LuCo_2 β remains constant within experimental error. Analyses based on several theoretical models of the quenching of spin fluctuations by high magnetic fields suggest that the characteristic spin-fluctuation temperature is ~20 K for ScCo_2, ~35 K for YCo_2, and ~16 K for LuCo_2. The magnetization and the field dependence of the magnetic susceptibility of the same samples as used in the heat-capacity measurements indicate the presence of ferromagnetic impurities in the samples, but the estimated concentrations are sufficiently low that they probably have no effect on the observed heat capacities. Maxwell's thermodynamics relationship between the field dependence of the heat capacity and the temperature dependence of the magnetic susceptibility has been examined.

I. INTRODUCTION

The anomalous enhancement of the electronic contribution to the heat capacity due to spin fluctuations in the strongly Pauli paramagnetic metals has been of considerable interest for about a decade.[1] It was pointed out by Brinkman and Engelsberg[2] and by Béal-Monod *et al.*[3] that the application of high magnetic fields offers one way of testing the spin-fluctuation theory. Brinkman and Engelsberg[2] explained that a magnetic field of the order of characteristic spin-fluctuation temperature T_s is required to quench spin-fluctuation enhancements. The effective field H_{eff} is given by

$$H_{\mathrm{eff}}=k_B T_s/\mu_B S^{1/2}\ , \tag{1}$$

where k_B is the Boltzmann constant, μ_B is the Bohr magneton, and S is the Stoner-exchange—enhancement factor.[4] The characteristic spin-fluctuation temperature is defined as

$$T_s=T_F/S\ , \tag{2}$$

where T_F is the Fermi temperature. Béal-Monod *et al.*[3] showed that the decrease in the heat capacity at 0 K of a nearly ferromagnetic Fermi liquid is proportional to H^2.

If the magnetic field is sufficiently large so that the Zeeman splitting energy of opposite spin states is comparable to or larger than the characteristic spin-fluctuation energy, then the paramagnons no longer have sufficient energy to flip spins, and therefore, the inelastic spin-flip scattering is quenched. Thus the specific-heat enhancement is expected to decrease with increasing magnetic field. Recently Hertel *et al.*[5] made a more detailed mathematical analysis and found that the electronic contribution to the heat capacity would be depressed by a few percent at 10 T, if the Stoner enhancement and the mass enhancement due to spin fluctuations are large (~4 and ~1.5, respectively), and also if the spin-fluctuation temperature T_s is small (~15 K). The discovery in 1980 of the quenching of spin fluctuations by high magnetic fields in the electronic heat capacity of LuCo_2 (Ref. 6) has substantiated these theoretical predictions.[2-5] The discovery of this lowering of the electronic heat capacity by magnetic fields has subsequently been confirmed in other highly enhanced paramagnetic materials, i.e., Sc,[7] CeSn_3,[8] and Pd-Ni.[9] The lowering of the electronic heat capacity reported in "pure Pd" by Hsiang *et al.*[10] has not been observe by Stewart and Brandt[11] on another Pd sample. Furthermore, these results (for Sc, CeSn_3, and Pd-Ni) show that this effect occurs at lower fields than has been expected. In particular, the electronic specific-heat constant decreases by 10.3% at 10 T for LuCo_2, 11.5% for Sc, 26.9% for CeSn_3, 13.4% for 0.47-at.% Ni-Pd, and 11.9% for 0.97-at.% Ni-Pd. On the other hand, Barnea[12] has estimated, from the temperature dependence of the magnetic susceptibility, that $T_s \approx 630$ K (i.e., $H_{\mathrm{eff}} \approx 230T$) for LuCo_2. This inconsistency requires further detailed theoretical and experimental studies on the quenching of the spin-fluctuation enhancement caused by high magnetic fields.

The strongly Pauli paramagnetic $R\mathrm{Co}_2$ (R denotes Sc, Y, or Lu) compounds with the cubic MgCu_2-type structure were chosen to investigate the quenching of spin fluctuations by high magnetic field because of the large magnetic susceptibility and unusual temperature dependence of the $R\mathrm{Co}_2$ phases. The magnetic susceptibility shows a flat maximum of the order of $\sim 3\times10^{-3}$ emu/mol at a fairly high temperature, $T_{\max}$ (~250, ~370, and ~600 K for YCo_2,[13,14] LuCo_2,[15] and ScCo_2,[16] respectively). For

YCo_2 the low-temperature zero-field heat-capacity and electrical-resistivity measurements have revealed a large electronic specific-heat constant (12 mJ/g-at. K^2) (Refs. 17 and 18) and a T^2-dependent resistivity below ~20 K.[19] Because of these unusual properties, several authors[12,20–22] have considered and treated the RCo_2 (R denotes Sc, Y, or Lu) compounds as a typical Fermi liquid with a large Stoner-enhancement factor $S \approx 10$.

Block *et al.*[23] have predicted, on the basis of high-field (up to 15 T) magnetization measurements on YCo_2, that the itinerant-electron metamagnetism, which was proposed first by Wohlfarth and Rhodes,[24] will occur in an applied field of ~140 T. The magnetic measurements on YCo_2 and $LuCo_2$ in fields up to 38 T by Schinkel[25] have shown that the field dependence of magnetization for both compounds can be approximated by two linear regions at 4.2 K, and even at 77 K for YCo_2. These results suggest that these compounds are typical nearly ferromagnetic materials.

In this paper the experimental results of the low-temperature (1.3–20 K) heat-capacity measurements in magnetic fields up to 10 T, and the magnetic susceptibility (1.7–300 K) measurements, are reported in detail for the RCo_2 compounds (R denotes Sc, Y, or Lu). The quenching of spin fluctuations by magnetic fields is discussed in connection with the theoretical predictions.[2–5]

II. EXPERIMENTAL

The preparation of stoichiometric YCo_2 is difficult because it forms peritectically and it is almost impossible to avoid the presence of the ferromagnetic YCo_3 phase.[17,22,26] In order to avoid the contamination of the samples by the ferromagnetic RCo_3 (pure Co for Sc-Co alloys) phase, rare-earth-rich nonstoichiometric samples of $ScCo_2$ (35.0 at. % Sc), YCo_2 (35.0 and 36.0 at. % Y), and $LuCo_2$ (35.0 at. % Lu) were prepared by arc-melting. The starting scandium, yttrium, and lutetium metals used in this investigation were prepared in Ames Laboratory by the calcium reduction of fluoride, followd by vacuum-casting and sublimation.[27] The cobalt metal was purchased from Johnson Matthey, Ltd. The major impurities in the starting materials are given in Table I.

The arc-melted ingots were sealed in a tantalum capsule, annealed for 1 week at 1000 °C, and then furnace-cooled. X-ray powder photographs showed only the presence of the RCo_2 Laves phase. However, metallographic examination and electron-microprobe analysis revealed the presence of second phases in all four samples. The concentrations of the rare earth and cobalt in the second phase relative to the matrix were determined by measuring the relative intensities of the Sc $K\alpha$, Y $K\alpha$, Lu $L\alpha$, and Co $K\alpha$ lines while scanning across appropriate sections of the samples during the microprobe analysis. These data indicate that the second phases in $ScCo_2$ and $LuCo_2$ are Sc- and Lu-rich phases, respectively (probably ScCo and Lu_4Co_3), while in the 35-at. % Y-YCo_2 sample the second phase is Co rich (probably YCo_3). The 36-at. % Y-YCo_2 sample was not examined by microprobe analysis, but magnetization measurements (to be discussed later) indicate that the ferromagnetic phase present in the 35-at. % Y-YCo_2 sample was not present in the 36-at. % Y-YCo_2 sample.

TABLE I. Impurity levels in the starting materials (in at. ppm). The impurity levels for those elements not listed, or for those listed for which no value is reported (unless otherwise noted), are 1 at. ppm or less.

Impurity	Sc	Y	Lu	Co
H	490	616	1385	a
C	83	104	277	a
N	6	13	75	a
O	188	250	1853	a
F	28	131	28	a
Al	3		20	
Si	2	<2	8	
Cl	2	2	3	
Ti				10
V				3
Cr	5.6	2.1		
Fe	53	27	34	3
Ni	5	4.6	10	15
Cu	50	4	17	
Y	2	matrix	3	
Zr	<5	<3		
Nb			<6	
Pd		<3		
Sn				3
Ba				3
La	5	5.3	2.8	
Ce	4.2	2.8	3.7	
Pr		8		
Nd	1.8		4	
Gd	<10	8.1	9.2	
Tb		4		
Er	1.1			
Yb			<4	
Ta	7.3			
W	2.3	20	2.5	
Os			<2	
Tl		2		
Pb				10

[a] No analyses available.

Resistance ratios, $\rho_{300\,K}/\rho_{4.2\,K} = \Gamma_{4.2\,K}$, were determined for all four samples. They varied in value from a high of 26 for $ScCo_2$ to a low of 7.1 for YCo_2 (36 at. % Y). The $\Gamma_{4.2\,K}$ value for $LuCo_2$ was 23 and that for YCo_2 (35 at. % Y) was 9.4. These values are not unreasonable considering that all of the samples have a nonstoichiometric composition and contained some second phases.

An isolation heat-pulse–type low-temperature calorimeter with a mechanical heat switch was used in this study. The temperature was measured by using a germanium-resistance thermometer which had been calibrated at magnetic fields of 0, 2.50, 5.39, 7.62, and 9.98 T.[28] The heat capacity of the 1965 Calorimetry Conference standard copper sample[29] was measured to serve as a check of the apparatus and experimental technique. These low-temperature heat-capacity results were in agreement within ±1% at 0 T and ±2% at the four nonzero magnetic fields with the previous zero-field data.[29,30]

Both magnetization and magnetic-susceptibility measurements were made on small samples cut out of the heat-capacity samples. The magnetization data were obtained at three temperatures (4.2, 77, and ~285 K) by using the Foner method. An electromagnet was used to generate magnetic fields ranging from 0.5 to 0.3 T. The magnetic-susceptibility measurements were made at a number of temperatures from 1.7 to 300 K by using the Faraday technique. An electromagnet with specially tapered pole pieces was used to produce a uniform gradient and magnetic fields ranging from 0.9 to 1.52 T. All of the results shown in the figures in this paper were obtained at 1.52 T. More details concerning the apparatus and the experimental procedure can be found in an earlier publication.[31]

III. RESULTS

A. Heat capacity

The heat-capacity measurements on $R\mathrm{Co}_2$ (R denotes Sc, Y, or Lu) were made between 1.3 and 20 K at magnetic fields of 0, 2.50, 5.39, 7.62, and 9.98 T. Figures 1,2, and 3 show the experimental results for ScCo_2, YCo_2, and LuCo_2, respectively. As can be seen, all the C/T-vs-T^2 curves are linear and there is no evidence for a $T^3\ln T$ term, except possibly for the slight up turn evident in the 5.39-, 7.62- and 9.98-T magnetic field data for ScCo_2 and LuCo_2. This up turn, however, is probably due to the superparamagnetic behavior of magnetic impurities such as iron. The magnitude of the upturn in ScCo_2 and LuCo_2 and the lack of it in YCo_2 is consistent with the iron concentrations in starting materials, i.e., ScCo_2, which has about 60% more iron then LuCo_2 (Table I), exhibits a larger up turn than LuCo_2; YCo_2, which has the least amount of iron present, exhibits no up turn.

For ScCo_2 and LuCo_2 one can distinctly see that the curves are parallel to one another and that they fall with increasing magnetic fields, especially for $H > 2.5$ T. However, for YCo_2 the straight lines for the various fields tend to cross one another with the intercept decreasing and the slopes increasing with increasing field.

In order to examine the influence of the magnetic fields on the heat capacity in the $R\mathrm{Co}_2$ phases, the electronic specific-heat constant γ, and the Debye temperature at 0 K, Θ_D, were calculated from a least-squares fitting of the data for each field between 1.3 and 6.0 K to the equation

$$C/T=\gamma+\beta T^2 , \tag{3}$$

where

$$\beta=1944/\Theta_D^3 \ (\mathrm{J/g\text{-}at.\ K^4}) . \tag{4}$$

The γ, β, and Θ_D values are presented in Tables II, III, and IV for ScCo_2, YCo_2 (both samples), and LuCo_2, respectively. For the results obtained at fields of 5.39, 7.62, and 9.98 T, the up turn at temperatures below ~2 K was neglected when fitting the ScCo_2 and LuCo_2 data to Eq. (3). Solid lines in Figs. 1 and 3 are the least-squares-fit results.

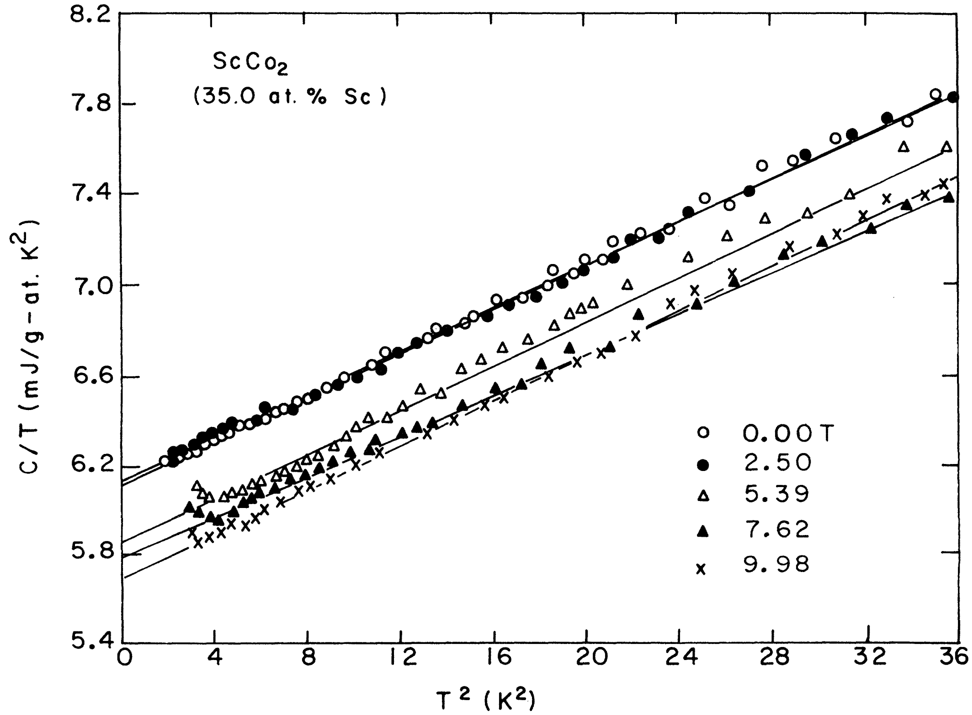

FIG. 1. Heat capacity of ScCo_2 at five magnetic fields. The solid lines are the results of a least-squares fitting of the data for each of the five magnetic fields to Eq. (3).

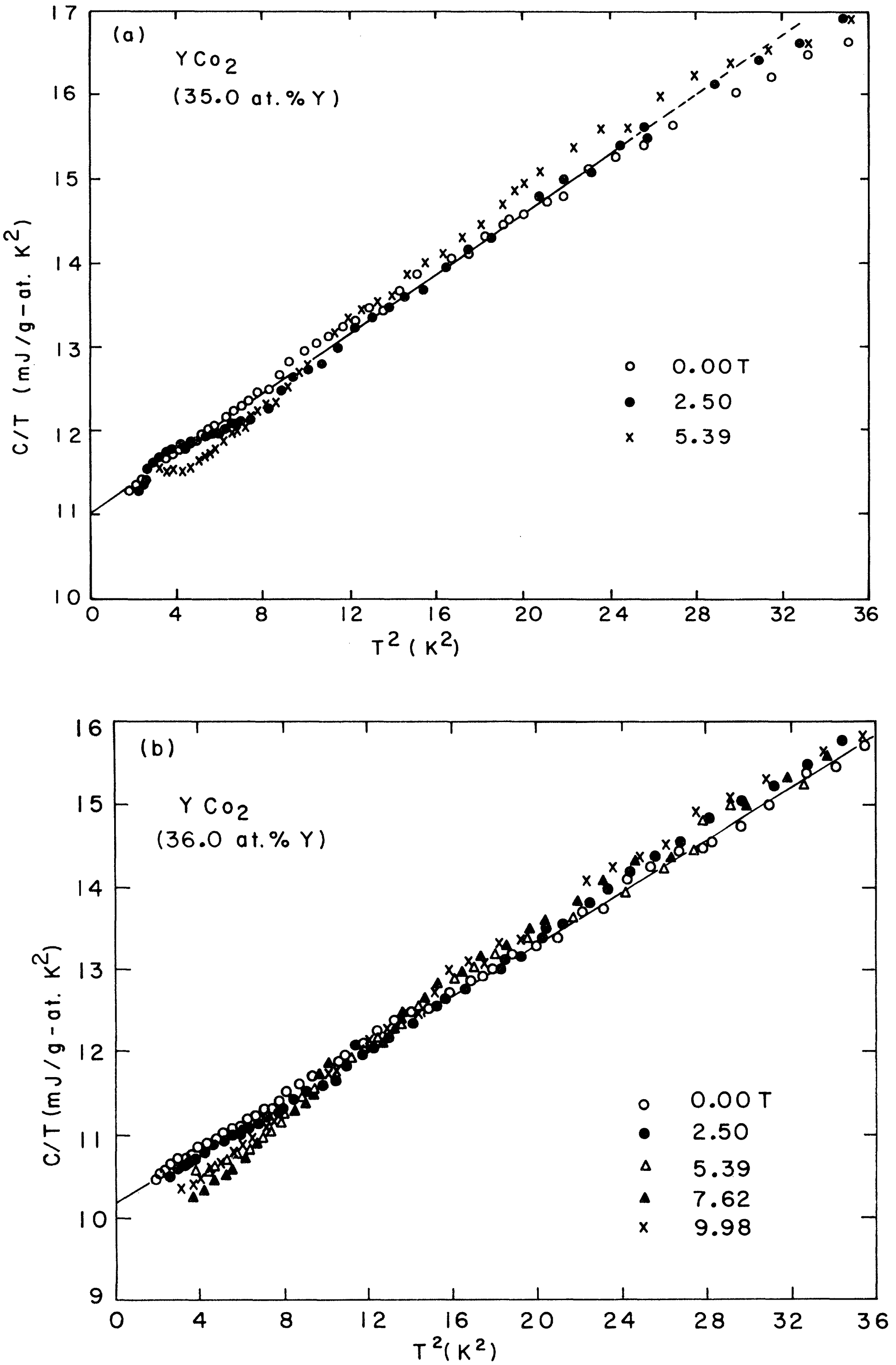

FIG. 2. Heat capacity of YCo_2 containing (a) 35 at. % Y and (b) 36 at. % Y at several magnetic fields. The solid line is the result of a least-squares fitting of the zero-field data to Eq. (3).

The resultant γ and β results for the two YCo_2 samples are presented along with the previous results in Table III. The zero-field γ and Θ_D values of our YCo_2 samples are somewhat smaller than the results reported by Block *et al.*[17] and Muraoka *et al.*[18] It is difficult to ascertain the reason for these discrepancies, but differences in the purities of the starting materials (they were not given by the other authors) and the distribution of and the chemical composition of the second phases present could account for the variation in γ and Θ_D.

The most interesting observation which can be made is that the slopes for the C/T-vs-T^2 plots for both YCo_2

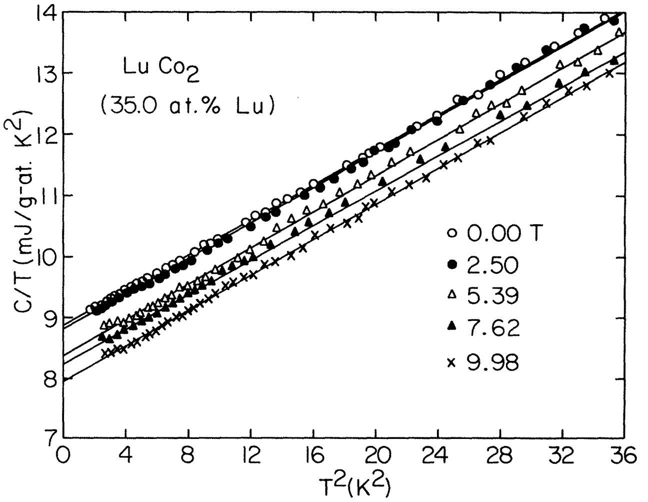

FIG. 3. Heat capacity of LuCo$_2$ at five magnetic fields. The solid lines are the results of a least-squares fitting of the data for each of the five magnetic fields to Eq. (3).

TABLE II. Results of a least-squares fitting of the heat-capacity data of ScCo$_2$ at five magnetic fields to Eq. (3).

H (T)	γ (mJ/g-at. K^2)	β (mJ/g-at. K^4)	Θ_D (K)
0.00	6.13±0.01	0.0479±0.0005	344±1
2.50	6.11±0.02	0.0489±0.0007	341±2
5.39	5.85±0.01	0.0488±0.0004	341±1
7.62	5.78±0.01	0.0453±0.0003	350±1
9.98	5.69±0.01	0.0496±0.0008	340±2

samples change with magnetic field, while those for ScCo$_2$ and LuCo$_2$ remain constant. This is more evident when the upper portions of Figs. 4 and 5 are compared [remembering that β is inversely proportional to Θ_D^3—Eq. (4)] where it is seen that $\Theta_D(\beta)$ remains constant as a function of field for ScCo$_2$ (within 1.7%) and LuCo$_2$ (within 0.9%) (Fig. 4), while there is no doubt, in spite of the scatter in the data, that β increases as the applied field increases for YCo$_2$ (Fig. 5). Furthermore, the relative change in β, as well as γ, with increasing magnetic field, appears to be larger in the sample with the higher cobalt concentration, which is to be expected since it is thought that the d electrons of cobalt are undergoing the spin fluctuations. Initially we thought that the variation of β for the YCo$_2$ samples with the applied magnetic field was due to the presence of YCo$_3$ in our samples. Although this cannot be ruled out, we now believe that the more reasonable explanation for the variation in β is due to an induced moment on the cobalt atoms by the applied magnetic field. The β variation shown here is essentially identical to that found in Sc,[7] CeSn$_3$,[8] and Pd-Ni alloys,[9] and in the cases of Sc and CeSn$_3$, it has been suggested that a magnetic moment is induced on the Sc or Ce atoms by the applied magnetic field.

The analytical results for the field dependence of γ are shown in the lower portions of Figs. 4 and 5 for the three RCo$_2$ compounds. The application of an applied field of 10 T lowers the γ value from the zero-field value by 7.2% for ScCo$_2$, 3.8% for YCo$_2$ (36 at.% Y), and 10.3% for LuCo$_2$ (both the Sc and Lu compounds contain 35 at.% R).

In addition, our measurements up to 20.0 K show that the difference of the C/T-vs-T^2 curves at 0 and 9.98 T is a maximum at ~10 K for ScCo$_2$, at ~11 K for YCo$_2$, and at ~12 K for LuCo$_2$, e.g., see Fig. 6 for LuCo$_2$. We have also found that at higher temperatures there is no

TABLE III. Results of a least-squares fitting of the heat-capacity data of YCo$_2$ at five magnetic fields to Eq. (3).

H (T)	γ (mJ/g-at. K^2)	β (mJ/g-at. K^4)	Apparent Θ_D (K)	Ref.
		35 at.% Y		
0	11.02±0.01	0.179±0.001	222±1	a
2.50	11.07±0.03	0.169±0.002	226±1	a
5.39	10.59±0.02	0.220±0.002	207±1	a
		36 at.% Y		
0	10.19±0.02	0.157±0.001	232±1	a
2.50	10.11±0.04	0.164±0.002	228±1	a
5.39	10.19±0.06	0.135±0.003	244±2	a
7.62	9.97±0.08	0.158±0.004	231±2	a
9.98	9.80±0.03	0.185±0.002	219±1	a
		Literature values		
0	12.1[b]		256[b,c]	17
0	12.0		240	18

[a] This study.

[b] For the composition YCo$_{1.8}$ (36 at.% Y).

[c] Θ_D value was erroneously reported by the authors (Ref. 17) as 276 K; the correct value is listed here.

TABLE IV. Results of a least-squares fitting of the heat-capacity data of $LuCo_2$ at five magnetic fields to Eq. (3).

H (T)	γ (mJ/g-at. K^2)	β (mJ/g-at. K^4)	Θ_D (K)
0.00	8.85±0.01	0.1434±0.0004	238.4±0.2
2.50	8.80±0.03	0.1449±0.0011	237.6±0.6
5.39	8.36±0.02	0.1473±0.0009	236.3±0.5
7.62	8.22±0.04	0.1419±0.0018	239.3±1.0
9.98	7.94±0.01	0.1450±0.0008	237.5±0.4

difference in the heat capacities at 0 and 9.98 T. The temperature at which they merge is ~13 K for $ScCo_2$, ~14 K for YCo_2, and ~20 K for $LuCo_2$ (Fig. 6). This means that the enhanced electronic specific-heat constant is predominant at low temperatures, and also that the enhancement is suppressed because of the renormalization of spin fluctuations as the temperature increases.[32]

B. Magnetization and magnetic susceptibilities

Typical magnetization, σ, curves for the RCo_2 compounds are shown in Fig. 7 (for $LuCo_2$). The $ScCo_2$ and $LuCo_2$ samples, which contained 35 at.% R, and the YCo_2 sample which contained 36 at.% Y, exhibited essentially identical σ-vs-H curves, in that they were linear with a zero intercept for temperatures equal to or greater than 77 K, and that the 4.2-K curve showed a nonlinear increase at low fields, which became linear at high fields. The curvature for the 4.2-K magnetization results suggest the presence of a ferromagnetic impurity, which is not detected in the heat-capacity results, and this indicates the impurity concentration must be quite small, namely less than 1%. The impurity could be a RCo_x phase with $x > 2$, or it could be free Co on the surface as suggested by Collings *et al.*[16] in their study of $ScCo_2$. Since our alloy composition and microprobe analyses indicate that the second phase is richer in R than RCo_2, and since RCo_x compounds with $x < 3$ do not order magnetically,[13-16] we believe that free Co on the surface of the RCo_2 compounds might account for the 4.2-K magnetization results. This was confirmed by an Auger depth-profile analysis of the $LuCo_2$ sample, which clearly showed the presence of free Co on the surface, which sat on a layer of Lu_2O_3, which, in turn, rested on the bulk $LuCo_2$ material. The amount of free Co on the surface can be estimated by extrapolating the high-field data to $H = 0$ T (dashed line in Fig. 7) to give the saturation mag-

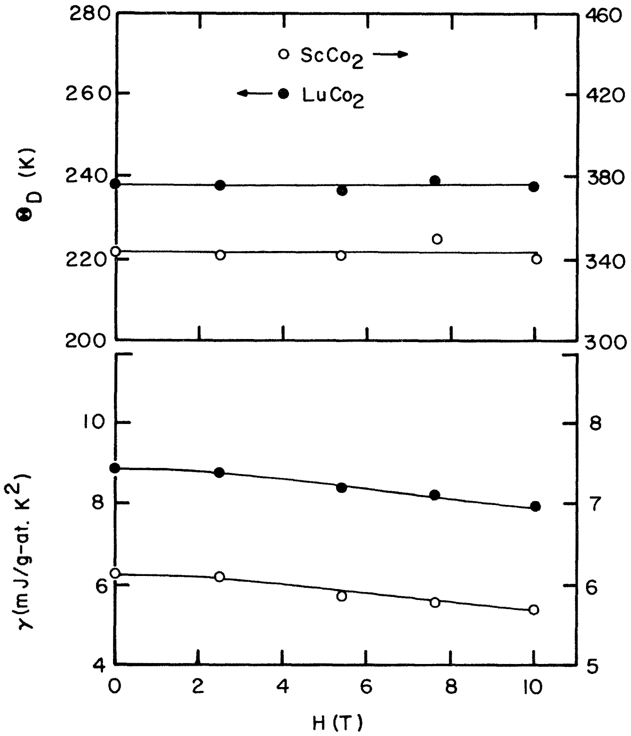

FIG. 4. Electronic specific-heat constant γ and the Debye temperature Θ_D of $ScCo_2$ and $LuCo_2$ as a function of the magnetic field.

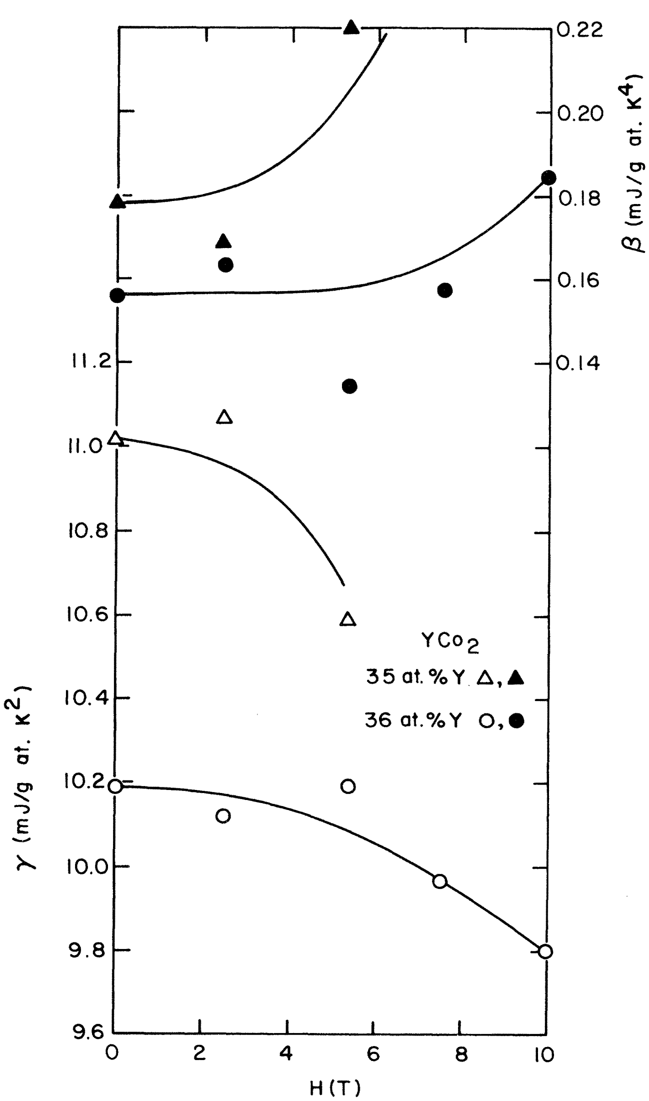

FIG. 5. Electronic specific-heat constant γ and the coefficient of the T^3 term β of two YCo_2 samples as a function of the magnetic field.

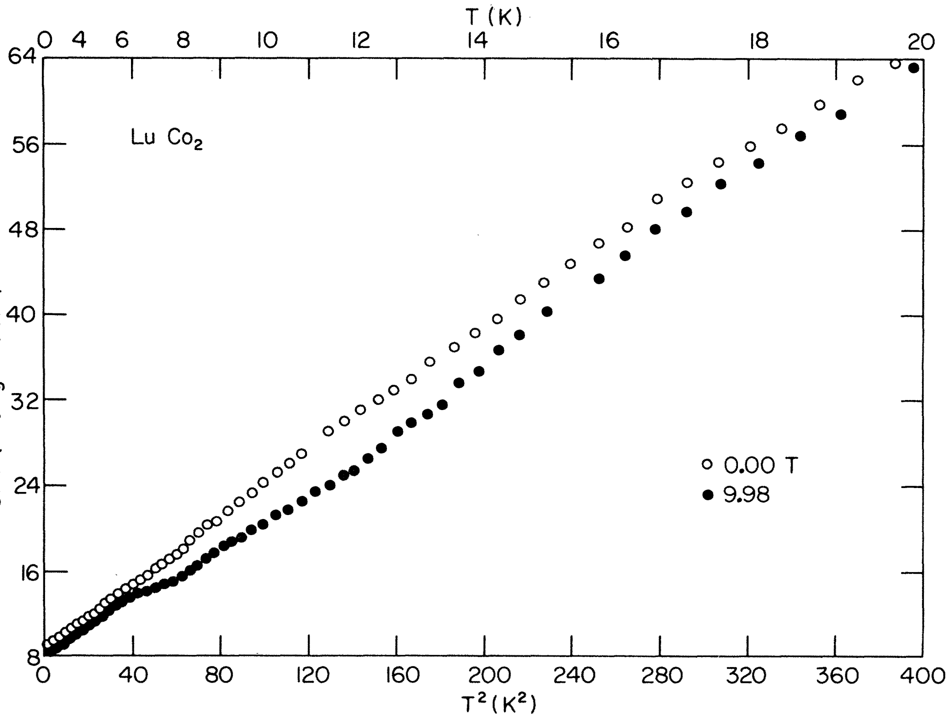

FIG. 6. Heat capacity of $LuCo_2$ from 1.3 to 20 K at 0 and 9.98 T.

netization value for the impurity, σ_i. Assuming an effective magnetic moment for the Co of $1.6\mu_B$ we find that $\sim 10^{-5}$ g per gram of sample of free Co lies on the surface for all three samples (see Table V). From the size of the $LuCo_2$ magnetization sample, and assuming that the Co atoms completely cover the surface in a close-packed arrangement of atoms, it is estimated that there are about 90 layers of free Co on the surface of the $LuCo_2$ specimen.

The magnetization results for the YCo_2 sample containing 35 at. % Y differ from the other three samples in that the σ-vs-H curves at 4.2, 77, and 280 K indicate the presence of a ferromagnetic second impurity phase. In this case we believe, at least for the 77- and 280-K results, that σ_i is due to YCo_3, which orders magnetically at 301 K.[13] The presence of YCo_3 was suggested by the microprobe analysis, as discussed earlier. At 4.2 K we believe that σ_i is due to YCo_3 and free Co on the surface. As shown in Table VI the amount of YCo_3 present in the alloy is 5.6×10^{-4} g per gram of alloy and the amount of free cobalt on the surface is 3.7×10^{-5} g per gram of alloy, which is nearly the same value found for the free Co in the 36-at. % Y-YCo_2 and $LuCo_2$ samples (Table V).

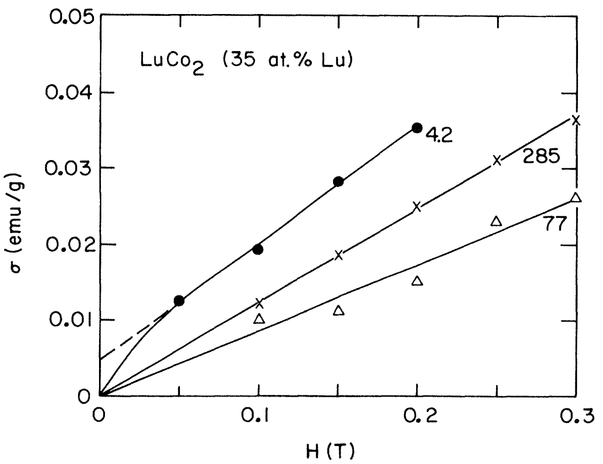

FIG. 7. Magnetization vs applied magnetic field for $LuCo_2$ at three temperatures (in K).

The Faraday magnetic susceptibilities from 1.7 to 300 K of the same four RCo_2 samples, which were used in the heat-capacity measurements (but are different from the magnetization samples, which were also cut from the heat-capacity samples), are shown in Figs. 8—10, along with the results reported in literature.[13—16,23,25] The magnetization results are in good agreement (within a few percent) with the susceptibility values shown in Figs. 8—10. There is, however, a considerable difference between our results and those published in the literature. However, this is to be expected since our alloys are deficient in Co, while those reported in the literature were for the RCo_2 stoichiometry. This is readily obvious when one compares

TABLE V. Impurity saturation magnetization value σ_i and mass of free cobalt on RCo_2 sample surface.

Compound	σ_i (emu/g)	Mass of Co (g/g)
$ScCo_2$ (35 at. % Sc)	0.012	7.9×10^{-5}
YCo_2 (36 at. % Y)	0.0051	$3.3_6\times10^{-5}$
$LuCo_2$ (35 at. % Lu)	0.0048	$3.1_6\times10^{-5}$

TABLE VI. Impurity saturation magnetization value σ_i and the masses of YCo_3 present in the bulk sample and free cobalt on the surface of the 35-at. % Y-YCo_2 alloy.

Temp. (K)	σ_i (emu/g)	p_{eff}[a] YCo_3 (μ_B/YCo_3)	Mass of YCo_3 (g/g)
280	0.0095	0.8	5.6×10^{-4}
77	0.0164	1.4	5.6×10^{-4}
4.2	0.0220	1.4	b

[a] Reference 13.

[b] Assumed mass of YCo_3 to be 5.6×10^{-4} g per gram of alloy from the 77- and 280-K results, which acounts for 0.0164 emu/g of the 0.0220 emu/g measured for σ_i. The difference, 0.0056 emu/g, is assumed to be due to the free cobalt on the sample surface (i.e., 3.7×10^{-5} g per gram of alloy).

our results for the two YCo_2 samples which differ by 1 at. % (Fig. 9).

The field dependence of the magnetic susceptibility was examined at 4.2 K. In all cases the susceptibility was found to decrease with increasing field from 0.91 to 1.52 T (see Fig. 11). These field dependencies suggest the presence of hard ferromagnetic impurities other than the free Co found on the surfaces or the YCo_3 impurity phase found in the 35-at. % Y-YCo_2 sample as discussed above, since the saturation for these impurities occurs at or below 0.15 T (see Fig. 7). It is doubtful that this effect at high fields is due to the Fe impurities in the starting metals because the effect observed in the compounds is inversely proportional to the amount of Fe in the materials (see Table I), thus ruling out the presence of free Fe. We believe the effect shown in Fig. 11 is due to pairs of Co atoms occupying R-metal-atom sites, which has been observed by others in the closely related RCo_x phases (where x varies from 2 to 5), which when carried out in a systematic manner will lead to the intermediate phases RCo_2, R_2Co_7, and R_2Co_{17}.[33] An upper limit for the number of such pairs can be estimated in the following way by using the method of Honda and Owen.[34] From the χ_{meas}-vs-$1/H$ plot (Fig. 11), one can calculate the true susceptibility of RCo_2 (χ_{RCo_2}) by extrapolation to $1/H=0$, where the intercept$=\chi_{RCo_2}$. At any given field we have

$$\chi_{\text{meas}}=\chi_{RCo_2}+\Delta\chi_{\text{ferro}} , \tag{5}$$

where $\Delta\chi_{\text{ferro}}$ is the contribution to the susceptibility by the ferromagnetic impurity and is given by

$$\Delta\chi_{\text{ferro}}=c\sigma_{\text{sat}}/H , \tag{6}$$

where σ_{sat} is the saturation magnetization of the impurity phase at the same temperature of the susceptibility measurement, and c is the impurity concentration. We have assumed that the pair of Co atoms has the same effective moment as pure Co metal. This is quite likely to be too low since the Co-Co separation in the Co pair is much smaller than in Co metal, and thus one would expect a higher effective magnetic moment. With the use of this value we have, at least, an upper limit on c and thus the number of Co atom pairs. From the results shown in Fig. 11, we calculate the upper limit of Co-atom pairs occupy-

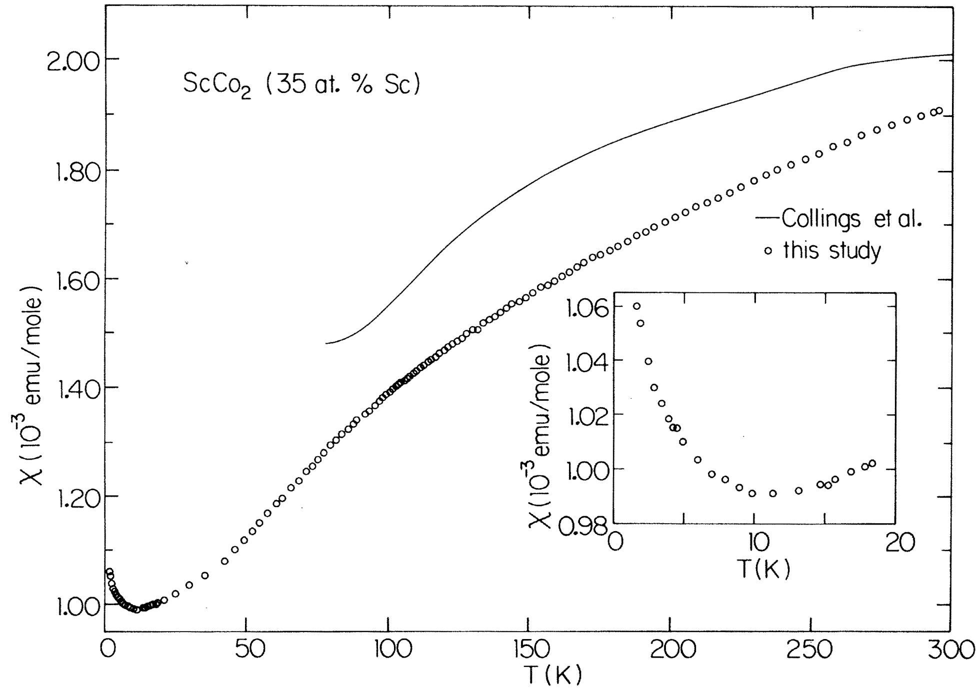

FIG. 8. Magnetic susceptibility of $ScCo_2$ from 1.7 to 300 K.

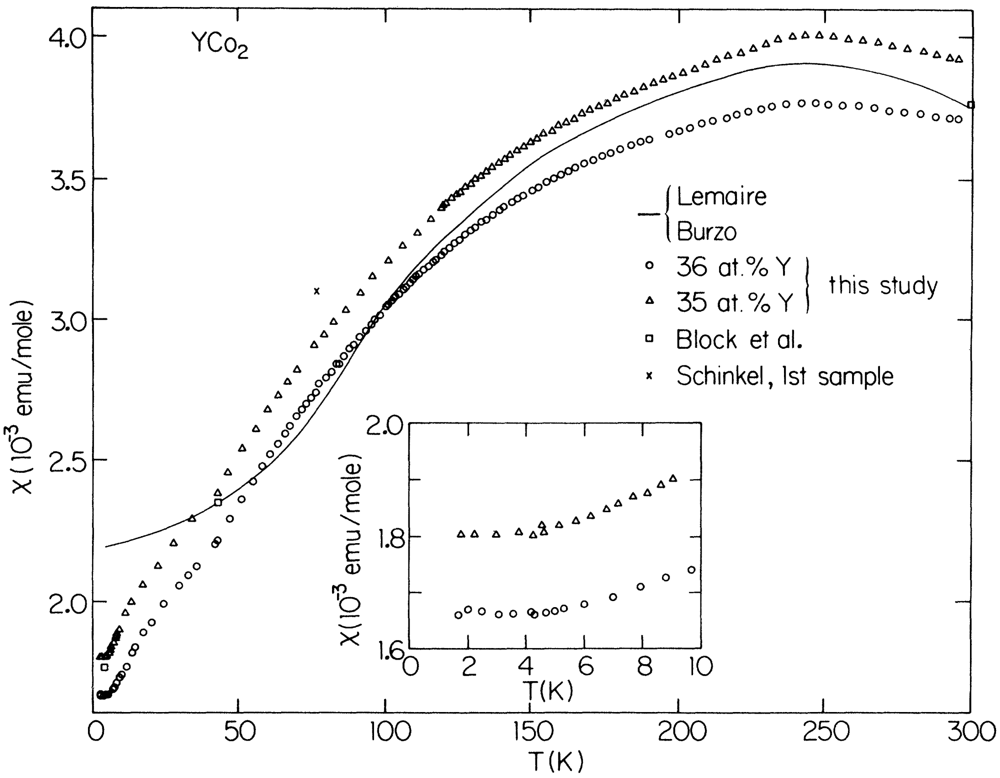

FIG. 9. Magnetic susceptibility of two YCo$_2$ samples from 1.7 to 300 K.

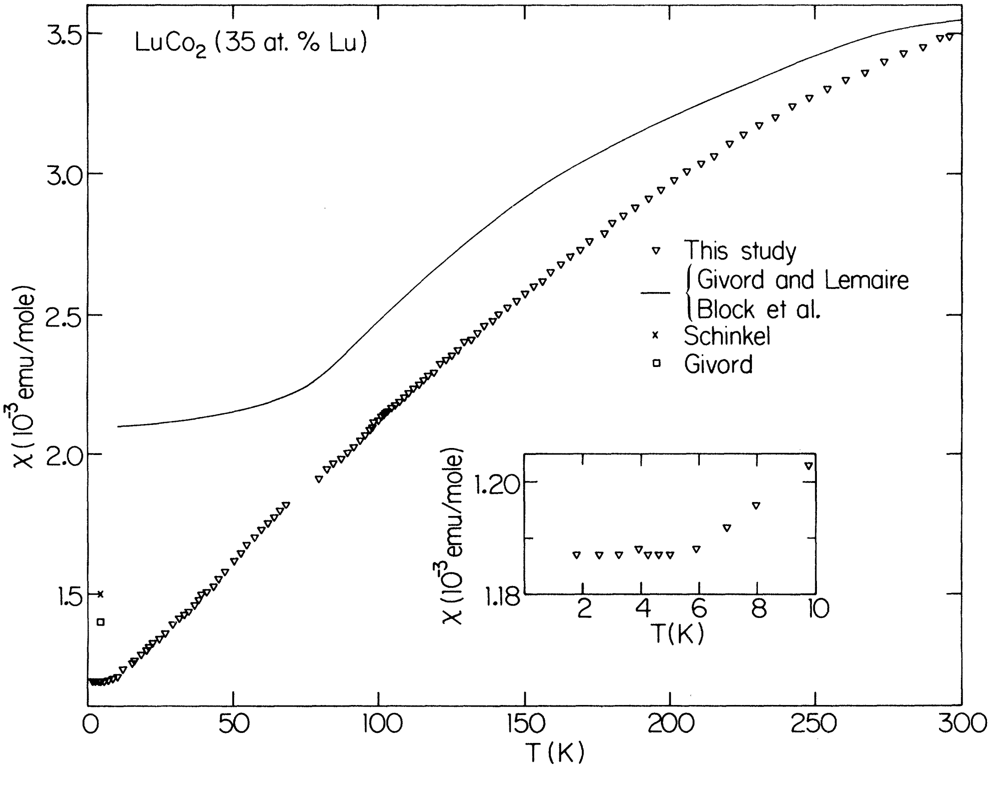

FIG. 10. Magnetic susceptibility of LuCo$_2$ from 1.7 to 300 K.

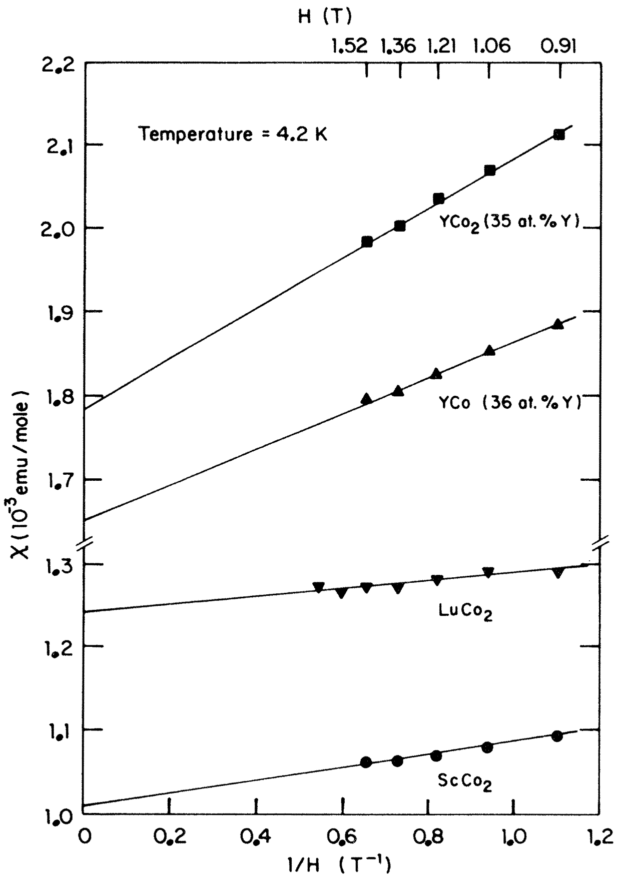

FIG. 11. Reciprocal-field plots for $ScCo_2$, two YCo_2 samples, and $LuCo_2$.

ing a Sc site to be 0.45×10^{-4} per g-at. in $ScCo_2$, a Y site in 36-at. % Y- and 35-at. % Y-YCo_2 to be 1.25×10^{-4} and 1.75×10^{-4} per g-at., respectively, and a Lu site in $LuCo_2$ to be 0.27×10^{-4} per g-at.

Collings *et al.*[16] noted similar effects in their high-field susceptibility measurements on $ScCo_2$. They suggested that the ferromagnetic impurity was Co atoms which precipitated out of solution during heat treatment. This is quite possible in their material since it was stoichiometric, but this is not likely to be the case for our sample, which is Co deficient. Thus, Co-atom pairs occupying R sites is the most likely explanation for the field dependence we have observed at high magnetic fields.

IV. DISCUSSION

In this section, the Debye temperature and some electronic parameters (such as the mass-enhancement and Stoner-enhancement factors) of the RCo_2 compounds are examined first. The discussion then centers on the Maxwell relation between the variation of the heat capacity as a function of magnetic field and the temperature dependence of the magnetic susceptibility, as well as the quenching of spin fluctuations by magnetic fields in connection with three theoretical models.

A. Debye temperature

The Debye temperature at 0 K, Θ_D, is calculated from the heat-capacity data by using Eq. (4). As shown in Fig. 4, the Θ_D values for $ScCo_2$ and $LuCo_2$ exhibit no magnetic field dependence within the accuracy of measurements. In the case of the two YCo_2 samples, one finds a variation in the β values [related to Θ_D via Eq. (4)] with the applied field (see Fig. 5 and Table III). Based upon our experience with $CeSn_3$ (Ref. 8) and Sc (Ref. 7) the zero-field β value yields the true Θ_D value, while the high-field data do not because the induced magnetic moment increases β and thus yields a lower "apparent Θ_D."

Lindemann[35] has shown that the Debye temperature is related to the melting point T_m, the mean atomic weight M, and the mean atomic volume V of a compound by the following expression:

$$\Theta_D = B(T_m/MV^{2/3})^{1/2} . \tag{7}$$

The constant B varies with the crystal type, but it is considered to be invariant for an isostructural closely related series of compounds.[36] In Fig. 12 the Debye temperatures of $ScCo_2$, YCo_2, and $LuCo_2$ are plotted as a function of $(T_m/MV^{2/3})^{1/2}$, where one finds a linear relationship. This shows that the Lindemann relation [Eq. (7)] is obeyed by these compounds. The slope of the straight line in Fig. 12 gives $B=119$, which is slightly larger than the value for ionic crystals ($B\approx115$).[36]

B. Paramagnon (spin-fluctuation) enhancement factor

For a nearly ferromagnetic metal, the low-temperature heat capacity well below T_s may be compared with the theory[2,32,37,38] which predicts

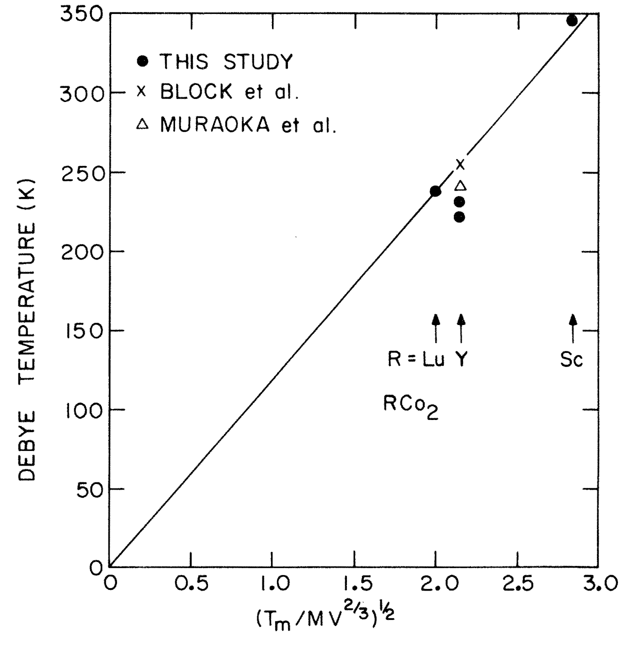

FIG. 12. Debye temperature of the RCo_2 phases as a function of $(T_m/MV^{2/3})^{1/2}$.

$$C=\gamma_0 T[m^*/m+\alpha(T/T_s)^2\ln(T/T_s)]+\beta_0 T^3\ . \tag{8}$$

Here, $m^*/m=1+\lambda_{\text{spin}}+\lambda_{\text{e-p}h}$ is the 0-K many-body mass enhancement, which includes the spin fluctuation and electron-phonon contributions (λ_{spin} and $\lambda_{\text{e-ph}}$, respectively), γ_0 is the electronic specific-heat constant determined from the band-structure density of states, α is proportional to $S(1-S^{-1})^2$, and $\beta_0 T^3$ is the usual lattice contribution. As shown by Lederer and Mills,[39] and by Engelsberg *et al.*,[40] Eq. (8) is valid only for the case of an uniform enhancement throughout the lattice, but if one takes into account the local enhancement on one atom (a magnetic solute atom), then the $T^3\ln T$ term is negligible except for $T<T_s/100$; thus the next leading term is small and has a T^3 dependence. Therefore, for the local-enhancement case at low temperatures, not only γ but also β is effected by the spin fluctuations, and the resultant value extracted from β is not the true Debye temperature. The theoretical predictions also suggest that the effective λ_{spin} value and the $T^3\ln T$ term both decrease with increasing magnetic fields.[2,3,5]

The band-structure calculation by Cyrot and Lavagna[41] for the cubic Laves-phase ($MgCu_2$-type) compounds RM_2, where R is a rare-earth element and M is Fe, Co, or Ni, show (1) that the density-of-states curves for these compounds are nearly the same as their respective pure metal M and are independent of the rare-earth element R, and (2) that there is a good agreement between the calculated and experimental γ values for YFe_2 and YNi_2, but not YCo_2. Assuming that their density of states at the Fermi level for YCo_2 is also valid for $ScCo_2$ and $LuCo_2$, we have estimated the mass-enhancement factor due to spin fluctuations.

The total-enhancement factor λ_{total} $(=\lambda_{\text{spin}}+\lambda_{e\text{-ph}})$ is given by

$$1+\lambda_{\text{total}}=m^*/m=N(E_F)/N_0(E_F)\ , \tag{9}$$

where $N(E_F)$ and $N_0(E_F)$ is the experimental and calculated density of states, respectively, at the Fermi level. By using the $N_0(E_F)$ value of Cyrot and Lavagna[41] (0.367 states/eV atom spin) and the $N(E_F)$ values calculated from the zero-field γ values, we determined the λ_{total} values shown in Table VII for the three RCo_2 compounds. The λ_{total} values are quite large. The $\lambda_{e\text{-ph}}$ value for pure Co is 0.2,[42] and Bennemann and Garland[43] have reported $\lambda_{e\text{-ph}}=0.14$, 0.35, and 0.42 for pure Sc, Y, and Lu, respectively. The $\lambda_{e\text{-ph}}$ values for the RCo_2 compounds were calculated as the average value from the component elements and are shown in Table VII. These estimated values of $\lambda_{e\text{-ph}}$ are not unreasonable, especially since these compounds are not superconducting, and they are probably correct within a factor of 2. The resultant λ_{spin} values for these compounds are quite large, consistent with our observations of the decrease in γ with increasing magnetic fields.

C. Stoner-enhancement factor

The Stoner-exchange—enhancement factor $S=\chi/\chi_0$ can be estimated from the observed magnetic susceptibility extrapolated to 0 K (χ) (see Table VII) and the Pauli paramagnetic susceptibility at 0 K as determined from the calculated density of states at the Fermi level (χ_0). The χ_0 value of 0.237×10^{-4} emu/g-at. is calculated from the $N_0(E_F)$ value, which was reported for YCo_2 by Cyrot and Lavagna.[41] The calculated values of the Stoner-enhancement factor for RCo_2 (R denotes Sc, Y, or Lu) are given in Table VII and are seen to range from 15.5 to 25.3.

D. Maxwell's relation

Recently Béal-Monod[44] suggested that the change in the heat capacity as a function of magnetic field and the change in the temperature dependence of the magnetic susceptibility as $T\rightarrow 0$ K for $LuCo_2$ apparently does not follow Maxwell's thermodynamic relationship:

$$H\left[\frac{\partial^2\chi}{\partial T^2}\right]_{H=0\ \text{T}}=\left[\frac{\partial C/T}{\partial H}\right]_{T=0\ \text{K}}\ . \tag{10}$$

However, she thought this might be due to the fact that different samples were used for the two measurements. This apparent violation of Maxwell's relation was an additional incentive to make both measurements on the same samples, which we have done.

As shown by Béal-Monod[44] and Acker *et al.*[45] the field dependence of the heat capacity is related to the temperature dependence of the susceptibility via Maxwell's thermodynamic relation

$$C/T(H)-C/T(0)=(H^2/T^2)[\chi(T)-\chi(0)]\ , \tag{11}$$

TABLE VII. The density of states at the Fermi level [$N_0(E_F)$ and $N(E_F)$], the mass-enhancement factor (λ_{total}, $\lambda_{e\text{-ph}}$, and λ_{spin}), the magnetic susceptibility at 0 K (χ_0 and χ), and the Stoner-exchange—enhancement factor (S) of RCo_2 (R denotes Sc, Y, or Lu).

Compounds	$N_0(E_F)$ (states/eV atom spin)	$N(E_F)$ (states/eV atom spin)	λ_{total}	$\lambda_{e\text{-ph}}$[a]	λ_{spin}	χ_0 (10^{-4} emu/g-at.)	χ[b] (10^{-4} emu/g-at.)	S
$ScCo_2$		1.30	2.54	0.18	2.36		3.67	15.5
YCo_2 (35 at. % Y)	0.367[c]	2.34	5.38	0.25	5.13	0.237[d]	6.00	25.3
YCo_2 (36 at. % Y)	0.367[c]	2.16	4.89	0.25	4.64	0.237[d]	5.33	22.5
$LuCo_2$		1.88	4.12	0.27	3.85		3.96	16.7

[a] See text.

[b] Corrected for ferromagnetic impurity.

[c] Reference 41.

[d] Calculated from density of states, Ref. 41.

assuming that the low-temperature susceptibility varies as T^2. Experimentally, below ~4 K, χ varies as T^2 for all four samples studied here, and thus in the limit as $T \to 0$ K we can use Eq. (11) to estimate the change in the heat capacity as a function of the applied field. The results are summarized in Table VIII, where it appears that Maxwell's relation holds for all four samples in sign, and in magnitude for three samples—the calculated depression of the heat capacity for $ScCo_2$ is too large. It is noted, however, that the maximum field of the susceptibility measurement (1.52 T) is significantly below the lowest field of the heat-capacity measurements (2.50 T). Furthermore, considering (1) the error limits on the heat-capacity measurements (all measured changes in the heat capacity as the field is increased from 0 to 2.50 T are essentially zero), and (2) the error in determining $\chi(T)-\chi(0)$ (the difference at best is only a few percent), it is difficult to say whether or not there is agreement. Because of these three reasons nothing definitive can be said about the validity of Maxwell's thermodynamic relation at this time. But since more precise heat-capacity measurements at lower fields are extremely difficult to make, and since significant changes in the heat capacities have been observed above 2.5 T (see Figs. 1—5 and Tables II—IV), susceptibility measurements on these four samples as a function of temperature at high magnetic fields are clearly needed to check Maxwell's relation [Eq. (11)].

E. Quenching of spin fluctuations by magnetic fields

Béal-Monod *et al.*[3] have shown that the shift of the electronic specific-heat constant at 0 K by an applied field H,

$$[C(0)-C(H)]/C(0) \approx [\gamma(0)-\gamma(H)]/\gamma(0) = \delta\gamma/\gamma(0) ,$$

should be

$$\delta\gamma/\gamma(0) = 0.1(S/\ln S)h^2 , \tag{12}$$

where h is the reduced field and is equal to $\mu_B H/k_B T_s$. In 1980 Hertel *et al.*[5] made a more detailed mathematical analysis of the quenching effect of the spin-fluctuation enhancement of the electronic specific heat by magnetic fields. They have given the effective mass enhancement $\lambda_{\text{spin}}(H)/\lambda_{\text{spin}}(0)$ as a function of the reduced field h.

Quite recently, Béal-Monod and Daniel[4] extended the older work of Béal-Monod *et al.*[3] and gave the following expression for the change in the electronic specific-heat constant with field:

$$\frac{\partial\gamma}{\gamma(0)} = -2.47\frac{\chi_P S}{\gamma(0)}\left[\frac{H}{T_s}\right]^2 + 1.05\frac{\chi_P S^2}{\gamma(0)}\left[\frac{H}{T_s}\right]^4 , \tag{13}$$

where χ_P is the Pauli susceptibility at $T=0$ K. The constants were taken for the case $T \ll H$.

TABLE VIII. Change in heat capacity predicted by Maxwell's relation.

Compound	$\Delta(C/T)^{\text{calc}}_{1.52\ \text{T}}$ (mJ/g-at. K^2)	$\Delta(C/T)^{\text{expt}}_{2.50\ \text{T}}$ (mJ/g-at. K^2)
$ScCo_2$	−0.20	−0.02±0.02
YCo_2 (35 at. % Y)	+0.01	+0.04±0.04[a]
YCo_2 (36 at. % Y)	−0.04	−0.07±0.06[a]
$LuCo_2$	−0.001	−0.05±0.04

[a]For $T=1$ K; the YCo_2 experimental values vary with temperature because β changes with applied field. At higher temperatures the two values listed tend toward zero.

As shown in Figs. 4 and 5, the electronic specific-heat constant decreases with increasing fields and at 9.98 T, $\delta\gamma/\gamma(0)$ is 10.3% for $LuCo_2$, 7.2% for $ScCo_2$, and 3.8% for YCo_2 (36 at. % Y). This strongly suggests that the reported T_s value for $LuCo_2$ (630 K) is unreasonably large, by about 2 orders of magnitude. Namely, we calculate a value of 0.05% for $\delta\gamma/\gamma(0)$ for $LuCo_2$ by using Eq. (12), and $S=16.7$ $H=10$ T, and $T_s=630$ K, which compares to the 10% value noted above for $LuCo_2$.

The characteristic spin-fluctuation temperature T_s for $ScCo_2$, YCo_2, and $LuCo_2$ can be estimated by comparing our experimental results with the theoretical models. Figure 13 shows the shift of the electronic specific-heat constant at 0 K, $\delta\gamma/\gamma(0)$, of $ScCo_2$, YCo_2, and $LuCo_2$, caused by an applied field H. The solid curves are taken from Hertel *et al.*[5] ($S=10$ and $\lambda_{\text{spin}}=0.37$). The dashed lines are the results of fitting the model of Béal-Monod *et al.*[3] [Eq. (12)] by assuming $S=15.5$ and $T_s=17$ K for $ScCo_2$, $S=22.5$ and $T_s=29$ K for YCo_2 (36 at. % Y), and $S=16.7$ and $T_s=15$ K for $LuCo_2$. T_s values which differ by more than ±1 K would lead to poor fits of the experimental data for the model of Béal-Monod *et al.*[3]

An attempt was made to fit the data to the model of Béal-Monod and Daniel[4] [Eq. (13)], but substituting the experimental values for χ_P, $\gamma(0)$, and S leads to large coefficients (of the order of 10^4) for the $(H/T_s)^n$ terms, and the resultant values for $\delta\gamma/\gamma(0)$ are extremely sensitive to the choice of T_s [large changes occur in $\delta\gamma/\gamma(0)$ for changes of 0.1 K in T_s]. Thus, it was impossible to fit the field data to a single T_s value. However, values of T_s similar to those noted above (after the model of Béal-Monod *et al*[3]) or below (after the model of Hertel *et al.*[5]) will give $\delta\gamma/\gamma(0)$ values which are small and positive.

The experimental results are in good agreement (considering the experimental error) with the curves of Hertel *et al.*[5] for $T_s=(23\pm2)$ K for $ScCo_2$, $T_s=(40\pm5)$ K for YCo_2, and $T_s=(18\pm2)$ K for $LuCo_2$. The agreement of the observed $\delta\gamma/\gamma(0)$ values with the H^2 dependence [Eq. (12)] is also good, but the curvature is less satisfactory than that given by Hertel *et al.* It is noted that the T_s values derived from the model of Hertel *et al.*[5] are approximately slightly larger than those obtained from the first model of Béal-Monod *et al.*,[3] for the respective RCo_2 compounds.

The spin-fluctuation temperatures derived from the two models are more than an order of magnitude smaller than the values calculated by Eq. (2). For example, for $LuCo_2$ a value between 15 and 18 K is derived depending on the model, which compares to a value of ~630 K estimated from the Fermi temperature. These spin-fluctuation temperatures correspond to effective fields [Eq. (1)] of 5.5—6.6 and 230 T, respectively. This inconsistency may require a new definition of the characteristic spin fluctua-

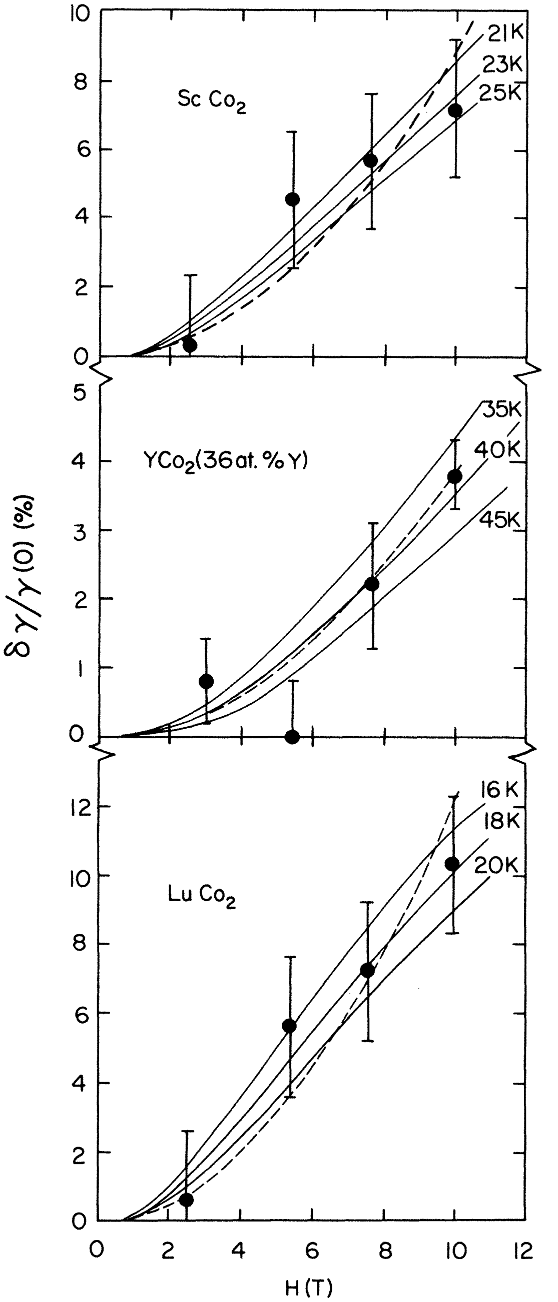

FIG. 13. Shift of the electronic specific-heat constant at 0 K, $\delta\gamma/\gamma(0)=[\gamma(0)-\gamma(H)]/\gamma(0)$, of $ScCo_2$, YCo_2, and $LuCo_2$, caused by an applied field H. The theoretical curves are taken from Hertel *et al.* (Ref. 5) [solid lines for $S=10$, $\lambda_{spin}=0.37$, and various T_s values (listed)] and from Béal-Monod *et al.* (Ref. 3) (dashed curve). See text for the S and T_s values used to construct the dashed curves for the three RCo_2 compounds.

tion which contains some details of the band structure in addition to T_F and S. It is possible, however, as has been recently pointed out by Gschneidner and Ikeda,[46] that both temperatures may be characteristic of the solid, with the higher T_s value being associated with 99% of the electron states of the material, and the lower T_s value being due to a small area of the Fermi surface (involving ~0.01 of an electron per atom) having an extremely high density of states (~1000 mJ/g-at. K^2) and a correspondingly high λ_{spin}. It is this λ_{spin} which is quenched by the applied field, leading to a smaller λ value at high fields.

F. Uniform versus local enhancement

In view of the magnetization and magnetic-susceptibility studies one may wonder what the influence of magnetic impurities might have on the observed heat-capacity changes with increasing field. The total ferromagnetic impurity levels vary from ~40 to ~350 at. ppm, but from our measurements on other spin-fluctuation materials [Sc (Ref. 7), $CeSn_3$ (Ref. 8), and Pd—0.5 and Pd—1.0 at. % Ni (Ref. 9)], we have shown that 0.08 to 0.001 electrons per atom account for the observed heat-capacity changes. Assuming that a similar number of electrons are involved in the RCo_2 compounds, one might expect the number of impurity atoms (assuming they contribute one electron per atom) necessary to account for the observed effects in the RCo_2 to number somewhere between 10^3 to 10^5 at. ppm. This is about a factor of 100 larger than the number of observed ferromagnetic impurity atoms. Thus the uniform-enhancement model would appear to account for the observed effects. However, if these ferromagnetic impurity atoms polarize a large volume (~100 neighbors), then the local-enhancement model would be the appropriate model. Although we cannot rule out the local-enhancement model we feel that the uniform-enhancement model best describes the quenching of spin fluctuations in the RCo_2 phases. Additional studies on RCo_2 phases of much higher quality probably could answer the question of whether the observed behaviors are due to impurities (the local-enhancement model) or if they are an intrinsic effect of the RCo_2 phases (the uniform-enhancement model).

ACKNOWLEDGMENTS

The authors wish to thank Professor B. N. Harmon, Professor D. K. Finnemore, Dr. S. K. Dhar (Ames Laboratory), Dr. A. I. Schindler (U.S. Naval Research Laboratory), Professor T. Moriya (Institute for Solid State Physics, Tokyo University), and Professor M. T. Béal-Monod (Université de Paris-Sud) for their helpful comments and discussions. Thanks are also due Dr. T. Ito for his cooperation in the magnetization measurements, Dr. H. Horie for his cooperation in the metallurgical analyses by the electron-probe microanalyzer, Mr. J. J. Holl for the sample preparation, and Dr. A. J. Bevolo for carrying out the Auger analysis of the $LuCo_2$ sample. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-ENG. This research was supported by the Director of Energy Research, Office of Basic Energy Sciences, WPAS-KC-02-01.

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[1] T. Moriya, J. Magn. Magn. Mater. 14, 1 (1979).

[2] W. F. Brinkman and S. Engelsberg, Phys. Rev. 169, 417 (1968).

[3] M. T. Béal-Monod, S.-k. Ma, and D. R. Fredkin, Phys. Rev. Lett. 20, 929 (1968).

[4] M. T. Béal-Monod and E. Daniel, Phys. Rev. B 27, 4467 (1983).

[5] P. Hertel, J. Appel, and D. Fay, Phys. Rev. B 22, 534 (1980).

[6] K. Ikeda and K. A. Gschneidner, Jr., Phys. Rev. Lett. 45, 1341 (1980).

[7] K. Ikeda, K. A. Gschneidner, Jr., T.-W. E. Tsang, and F. A. Schmidt, Solid State Commun. 41, 889 (1982).

[8] K. Ikeda and K. A. Gschneidner, Jr., Phys. Rev. B 25, 4623 (1982).

[9] K. Ikeda, K. A. Gschneidner, Jr., and A. I. Schindler, Phys. Rev. B 28, 1457 (1983).

[10] T. Y. Hsiang, J. W. Reister, H. Weinstock, G. W. Crabtree, and J. J. Vuillemin, Phys. Rev. Lett. 47, 523 (1981).

[11] G. R. Stewart and B. L. Brandt, Phys. Rev. B 28, 2266 (1983).

[12] G. Barnea, J. Phys. F 7, 315 (1977).

[13] R. Lemaire, Cobalt [33], 201 (1966).

[14] E. Burzo, Int. J. Magn. 3, 161 (1972).

[15] F. Givord and R. Lemaire, Solid State Commun. 9, 341 (1971); D. Bloch, F. Chaisse, F. Givord, J. Voiron, and E. Burzo, J. Phys. (Paris) Colloq. 32, C1-659 (1971).

[16] E. W. Collings, R. D. Smith, and R. G. Lecander, J. Less-Common Metals 18, 251 (1969).

[17] D. Block, D. L. Camphausen, J. Voiron, J. B. Ayasse, A. Berton, and J. Chaussy, C. R. Acad. Sci. Ser. B 275, 601 (1972).

[18] Y. Muraoka, M. Shiga, and Y. Nakamura, J. Phys. Soc. Jpn. 42, 2067 (1977).

[19] K. Ikeda, J. Phys. Soc. Jpn. 42, 1541 (1977).

[20] E. Burzo and D. P. Lazar, Solid State Commun. 18, 381 (1976).

[21] S. Misawa and K. Kanematsu, J. Phys. F 6, 2119 (1976); S. Misawa, *ibid.* 8, L263 (1978).

[22] H. R. Kirchmayr and C. A. Poldy, in *Handbook on the Physics and Chemistry of Rare Earths,* edited by K. A. Gschneidner, Jr. and L. Eyring (North-Holland, Amsterdam, 1979), Vol. 2, p. 55.

[23] D. Block, D. M. Edwards, M. Shimizu, and J. Voiron, J. Phys. F 5, 1217 (1975).

[24] E. P. Wohlfarth and P. Rhodes, Philos. Mag. 7, 1817 (1962).

[25] C. J. Schinkel, J. Phys. F 8, L87 (1978).

[26] K. Ikeda, J. Less-Common Metals 52, 101 (1977).

[27] B. J. Beaudry and K. A. Gschneidner, Jr., in *Handbook on the Physics and Chemistry of Rare Earths,* edited by K. A. Gschneidner, Jr. and L. Eyring (North-Holland, Amsterdam, 1978), Vol. 1, p. 173.

[28] K. Ikeda, K. A. Gschneidner, Jr., B. J. Beaudry, and U. Atzmony, Phys. Rev. B 25, 4604 (1982).

[29] D. W. Osborne, H. E. Flotow, and F. Shreiner, Rev. Sci. Instrum. 38, 159 (1967).

[30] D. L. Martin, Can. J. Phys. 47, 1253 (1969).

[31] R. J. Stierman, K. A. Gschneidner, Jr., T-W. E. Tsang, F. A. Schmidt, P. Klavins, R. N. Shelton, J. Queen, and S. Legvold, J. Magn. Magn. Mater. 36, 249 (1983).

[32] K. Makoshi and T. Moriya, J. Phys. Soc. Jpn. 38, 10 (1975).

[33] B. Barbara, D. Gignoux, D. Givord, F. Givord, and R. Lemaire, Int. J. Magn. 4, 77 (1973).

[34] L. F. Bates, *Modern Magnetism* (Cambridge University Press, Cambridge, England, 1951), p. 133.

[35] F. A. Lindemann, Z. Phys. 11, 609 (1910).

[36] M. Blackman, in *Encyclopedia of Physics,* edited by S. Flügge (Springer, Berlin, 1955), Vol. VII, Part 1, p. 325.

[37] N. F. Berk and J. R. Schrieffer, Phys. Rev. Lett. 17, 433 (1966).

[38] S. Doniach and S. Engelsberg, Phys. Rev. Lett. 17, 750 (1966).

[39] P. Lederer and D. L. Mills, Phys. Rev. Lett. 20, 1036 (1968).

[40] S. Engelsberg, W. F. Brinkman, and S. Doniach, Phys. Rev. Lett. 20, 1040 (1968).

[41] M. Cyrot and M. Lavagna, J. Phys. (Paris) 40, 763 (1979).

[42] D. A. Papaconstantopoulos, L. L. Boyer, B. M. Klein, A. R. Williams, V. L. Morruzzi, and J. F. Janak, Phys. Rev. B 15, 4221 (1977).

[43] K. H. Bennemann and J. W. Garland, in *Superconductivity in d- and f-Band Metals (Rochester),* Proceedings of the Conference on Superconductivity in *d*- and *f*-Band Metals, edited by D. H. Douglas (AIP, New York, 1972), p. 103.

[44] M. T. Béal-Monod, Physica 109&110B, 1837 (1982).

[45] F. Acker, R. Huguenin, J. L. Smith, and G. R. Stewart, Phys. Rev. B 27, 5883 (1983).

[46] K. A. Gschneidner, Jr. and K. Ikeda, J. Magn. Magn. Mater. 31-34, 265 (1983).