Comparative study of thermal and magnetic properties of CeT_yX_{4-y} ferromagnets

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Low-temperature thermal (specific heat, C_P) and magnetic [ac susceptibility, χ_{ac}, and isothermal magnetization $M(B)$] measurements on CeT_yX_{4-y} ferromagnets (with $T=$ Ni, Cu, Pd, Ag, Au, and $X=$ Ga, Al, with $0.3 < y < 0.7$) are reported. With the exception of $T=$ Ni, all compounds with $X=$ Ga show a typical second-order ferromagnetic transition. In the case of $T=$ Ni, magnetic correlations set in at $T \approx 3T_C$ (T_C being the Curie temperature) and the $C_P(T)$ and $\chi_{ac}(T)$ maxima (at 3.3 and 4.2 K, respectively) do not coincide, while the $M(B)$ curves reveal an antiferromagnetic character at lower temperatures. In these systems, the magnetic entropy of the ordered phase is larger than 85% of $R\ln2$, in coincidence with the low values of the Sommerfeld coefficient $\gamma \lesssim 10$ mJ/mol K^2. For $T=$ Ag and $X=$ Al, a strong diamagnetic signal was observed at $T_s = 0.63$ K in $\chi_{ac}(T)$, with the characteristics of a superconducting component. The thermodynamical properties of these compounds confirm the lack of hybridization when the Ce atom is in a ferromagnetic ground state. [S0163-1829(97)08230-1]

I. INTRODUCTION

In spite of the large number of Ce systems exhibiting coexistence of magnetic order and Kondo effect, the great amount of antiferromagnets (AF) recognized up to now with respect to ferromagnets (F) remains a puzzling problem. In coincidence with the fact that the Kondo effect is related to a negative-type electron coupling parameter J, none of the known F systems show conclusive indications of hybridization effects in their respective ground states (GS) and therefore F order is generally taken as the hallmark of trivalent Ce GS.[1,2] In most cases, the magnetic behavior of Ce-based intermetallics is successfully described in terms of the competition between two types of interactions via the Doniach-Lavagna[3,4] picture, i.e., an *intersite* one [e.g., like the Ruderman-Kittel-Kasuya-Yosida (RKKY)] which tends to order magnetically the $4f$ moments and an *intrasite* one that screens the local moments (related to the Kondo effect). Regardless of the ''sign'' of J, the oscillatory nature of the RKKY long-range interaction includes the possibility of F as well as AF states formation. As a consequence, a F or AF magnetic structure should occur periodically as the interatomic Ce-Ce spacing D changes and the sign of the intersite interaction oscillates (independently of the degree of hybridization of the $4f$ level with the band states). In spite of that, it is observed that in Ce binary compounds the F cases only appear within a well defined Ce-Ce spacing range: $3.7 < D < 4.1$ Å.[2] For such a spacing the Ce atoms are their mutual first neighbors, allowing a more direct exchange than the electron-mediated RKKY interaction (which is strongly related to the hybridization mechanism). In any case, for distances larger than $D > 4.1$ Å, an electron-mediated mechanism is required and coincidentally *no* F order is observed.[2]

Further information concerning the interplay between F or AF states with the Kondo effect can be obtained by comparing their respective magnetic to nonmagnetic GS transformations induced by alloying the Ce ligands. The T_C evolution as a function of concentration in a F system of the type Ce $Y_{1-x}Z_x$ differs from the Doniach-Lavagna description mainly in two features: a broad ''plateau'' around the maximum of $T_C(x)$ is followed by a drastic drop of the ordering temperature, as observed in $CePd_{1-x}Ni_x$, $CePt_{1-x}Ni_x$, $Ce(Ge_{1-x}Si_x)_2$, and $CeSi_{2-x}$.[5] This behavior suggests that the F exchange and the Kondo effect do not coexist and basically they ''exclude'' each other, whereas an AF-GS seems to favor that effect. For comparison, one can quote that the evolution of the concentration dependence of the Néel temperature $T_N(x)$ of $CeIn(Ag_{1-x}Cu_x)_2$ and $CeRh_2(Ge_{1-x}Si_x)_2$, behaves according to the Doniach-Lavagna description. Another distinction between the evolution of F and AF systems under alloying is the intrinsic difference between their respective ''final'' (nonmagnetic) GS. In the former, one systematically finds an *intermediate-valent* behavior, whereas for the latter a *heavy-fermion* GS.[6] Such a difference indicates that a strong hybridization strength ($T_K > 100$ K, that includes charge fluctuations) is required to overcome a F-GS, while in an AF system spin fluctuations (with $T_K \simeq 10$ K) are enough to screen the Ce magnetic moment.[6,7]

All these empirical features point to the exclusion of F and Kondo effect, however whether such exclusion is intrinsically related to the nature of the hybridization mechanism remains under discussion. Theoretical studies indicate that AF correlations tend to stabilize the Kondo effect, whereas F ones tend to destroy it,[7] though there are also arguments to the contrary.[8]

In this context, some CeT_yGa_{4-y} systems, with $T=$ Ni and Cu, were claimed to be F Kondo lattices,[9] whereas for $T=$ Pd and Ag the influence of the Kondo effect was found to be negligible.[10] From the analysis of the environmental

TABLE I. Thermal and magnetic data of CeT_yX_{4-y} compounds: Curie temperature, T_C (K); temperature of the maximum of C_{el} or χ'', T_{max} (K); Sommerfeld coefficient, γ (mJ/mol K^2); coefficient of the C_{el} $\propto T^{-2}$, A (JK/mol); coefficients of the C_{ph} contribution as $\beta = 0.1B_3$ (mJ/mol K^4), $\beta' = 10^{-7}B_5$ (mJ/mol K^6), $\beta'' = 10^{-12}B_7$ (mJ/mol K^8); Curie-Weiss temperature extracted from low temperatures, Θ_p^{LT} (K); entropy gain at $2T_C$, ΔS_m ($R\ln 2$); magnetization at 2 K for $B = 0.2$ T and $B = 5T$, M (μ_B/mol).

Compound	T_C	γ	A	β,β',β''	Θ_p^{LT}	ΔS_m	M
$CeNi_{0.5}Ga_{3.5}$	4.16	9.2	46	6, 7, -3	3.1	0.96	0.25 & 1.03
$T_{max}(C_{el})$	3.25						
$CeCu_{0.5}Ga_{3.5}$	6.05	8	76	8, 15, 18	6.2	0.95	0.78 & 0.96
$CePd_{0.3}Ga_{3.7}$	4.09	8.4	56	13, -7, 12	4.7	0.94	0.78 & 0.98
$CeAu_{0.6}Ga_{3.4}$	3.46	8.3	32	9, 46, -40	3.6	0.93	0.79 & 1.02
$CeAg_{0.75}Ga_{3.25}$	3.78	6	42	6, 48, -35	3.8	0.92	0.78 & 1.06
$CeAg_{0.6}Al_{3.4}$	3.19	8	47	10, -15, 30	3.5	0.86	0.67 & 1.06
$T_{max}(\chi'')$	0.63						

conditions of an ion in its crystallographic site one can extract information about the expected state of valence. Within this family of compounds, the YbT_yGa_{4-y} compounds form with Yb in its lowest valence configuration (i.e., Yb^{2+}) due to the low electronegativity at the lanthanide site.[11] Then from the correlation between Ce and Yb valencies,[12] the presence of a divalent Yb ion in that structural site allows us to deduce the trivalency of Ce in the systems at hand, excluding the possibility of hybridization effects.

In this work we have performed complementary low-temperature magnetic and thermal measurements on CeT_yGa_{4-y} compounds (with $T =$ Ni, Cu, Pd, Ag, Au), which show the aforementioned properties. Although these compounds were recognized as being F from their magnetic properties, the specific heat was measured only in a few of them by different authors and different sample preparation procedures.[13–16] Therefore, in order to compare their respective values of ΔS_m and γ, a systematic study in specimens having the same sample preparation and measuring performances is required. The choice of different transition metals intends to produce volume and chemical variations on the Ce atom environment (keeping the same structural symmetry) and to check whether hybridization mechanism occurs or not in these F systems. The $CeAg_yAl_{4-y}$ ferromagnet[13] was also included in this study as a variation of the richest metalloid component.

II. EXPERIMENTAL DETAILS AND RESULTS

All investigated samples have been prepared using an argon-triarc furnace in a Ti-gettered atmosphere by melting appropriate amounts of the constituents. In order to obtain homogeneous samples, the buttons were remelted several times and annealed (except for $CeAg_{0.6}Al_{3.4}$) for two weeks at 600 °C under partial ^{4}He pressure. By means of x-ray diffraction, using the Cu Kα radiation, all samples showed the proper $BaAl_4$ structure without observable foreign phases. The concentration value is the minimum value of the

T element for which single phases are formed in the respective compounds.[13–16] Microprobe analysis was performed in the two $CeAg_{0.6}Al_{3.4}$ samples in order to check the respective sample homogeneity and local Al concentration. Energy-dispersive spectroscopy (EDS) analysis shows an excess of Al in some regions of the sample, up to a 30% with respect to the nominal concentration. The specific-heat measurements were performed in semiadiabatic calorimeters using the heat-pulse method within the 1.5–15 K range of temperature for most of the samples. In order to better define the temperature dependence of the electronic specific heat in the magnetically ordered phase, to detect an eventual superconducting transition and to analyze the phonon contribution, one of the samples (cf. $CeAg_{0.6}Al_{3.4}$) was measured in a calorimeter working between 0.4 and 30 K. The ac susceptibility was measured in a ^{3}He cryostat operating between 0.4 and 30 K, with an inductance bridge working with an excitation field of 10 μT at 128 Hz. The magnetization curves were measured in a superconducting quantum interference device magnetometer with fields up to $B = 5$ T, in isotherms ranging between 2 and 10 K.

The temperature dependence of the measured specific heat (C_P) was analyzed in the paramagnetic ($T > T_C$) phase as due to electronic (C_{el}) phonon (C_{ph}) contributions, $C_P(T) = C_{el}(T) + C_{ph}(T)$, with $C_{el} = A/T^2 + \gamma T$ and $C_{ph} = B_3 T^3 + B_5 T^5 + B_7 T^7$. The A/T^2 term accounts for the short-range magnetic interactions right above T_C, and γT is the Sommerfeld coefficient. The respective fits were performed for $T > 1.3T_C$ and the resulting values are listed in Table I. The *Debye* temperatures (extracted from the B_3 coefficient) range between 200 and 270 K. The ac susceptibility (χ_{ac}) and C_{el} data around the ferromagnetic transition of the compounds under study are compared in Fig. 1 for $T =$ Ni and Cu, Fig. 2 for $T =$ Pd and Au, and in Fig. 3 for $T =$ Ag and $X =$ Ga, Al. Their respective F character is confirmed by the rapid rise of the inductive component (χ') of $\chi_{ac}(T)$, which shows a maximum at $T \simeq T_C$, in coincidence with the maximum slope of the dissipative signal χ'' (as
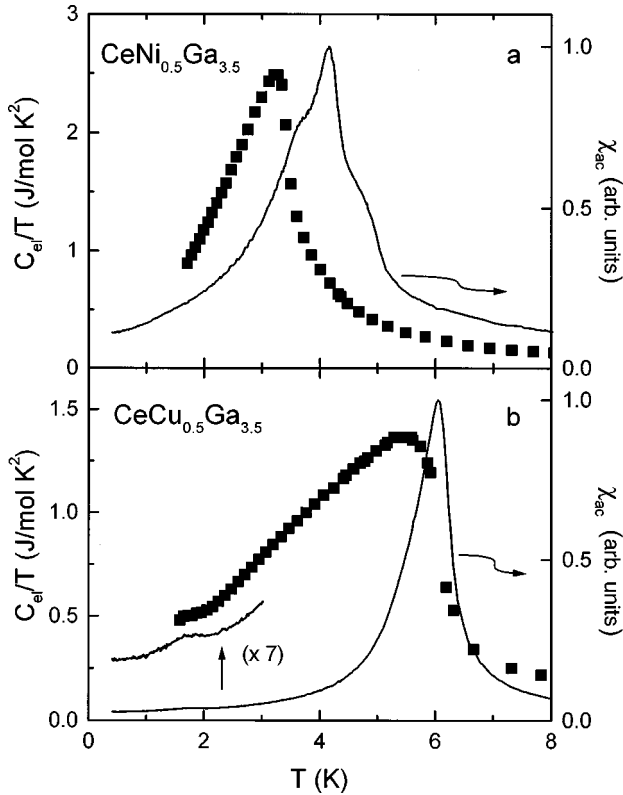
FIG. 1. Low-temperature specific-heat divided temperature and inductive component of the ac susceptibility of CeT_yGa_{4-y}, for $T =$ Ni (a) and Cu (b).

FIG. 2. Low-temperature specific-heat divided temperature and inductive component of the ac susceptibility of CeT_yGa_{4-y}, for $T =$ Pd (a) and Au (b).

shown in Fig. 3). Except for $CeNi_{0.5}Ga_{3.5}$, all the $C_{el}(T)$ transitions have the characteristics of a second-order type, with the ordering temperature at the inflexion point of the $C_{el}(T)$ broadened jump. The respective transition temperatures, listed in Table I, are in agreement with previously reported T_C values.[14–16] In the case of $CeNi_{0.5}Ga_{3.5}$, the $C_{el}(T)$ transition is not *mean-field*-like and the maximum of $\chi_{ac}(T)$ is found at 30% higher temperature than the maximum of $C_{el}(T)$, see Table I. Also $CeAg_{0.6}Al_{3.4}$ exhibits an unexpected behavior showing a diamagnetic $\chi_{ac}(T)$ signal below 0.63 K, but without a correspondent jump in $C_{el}(T)$ [see Fig. 3(b)]. The respective magnetization curves (M vs H) at temperatures ranging between 2 and 10 K, under fields up to 5 T are displayed in Fig. 4 for $T =$ Ni and Cu, in Fig. 5 for $T =$ Pd and Au, and in Fig. 6 for those with $T =$ Ag and $X =$ Ga, Al.

III. DISCUSSION AND CONCLUSIONS

Some thermodynamical conditions that have to be fulfilled to recognize the Ce-GS as a nonhybridized $4f$ state are[2] (i) to have the total expected entropy for the Ce-doublet GS ($\Delta S_m = R\ln 2$) because all the electronic degrees of freedom should be condensed in the magnetically ordered phase, (ii) accordingly, at $T \to 0$ one should observe a Sommerfeld coefficient (γ) comparable to that of a nonmagnetic (usually La-based) reference compound, and (iii) one should have a "nonscreened" magnetic moment of the order of $1\mu_B$ per atom as the saturation value, depending on the crystal-field (CF) GS. These three characteristics have been observed in
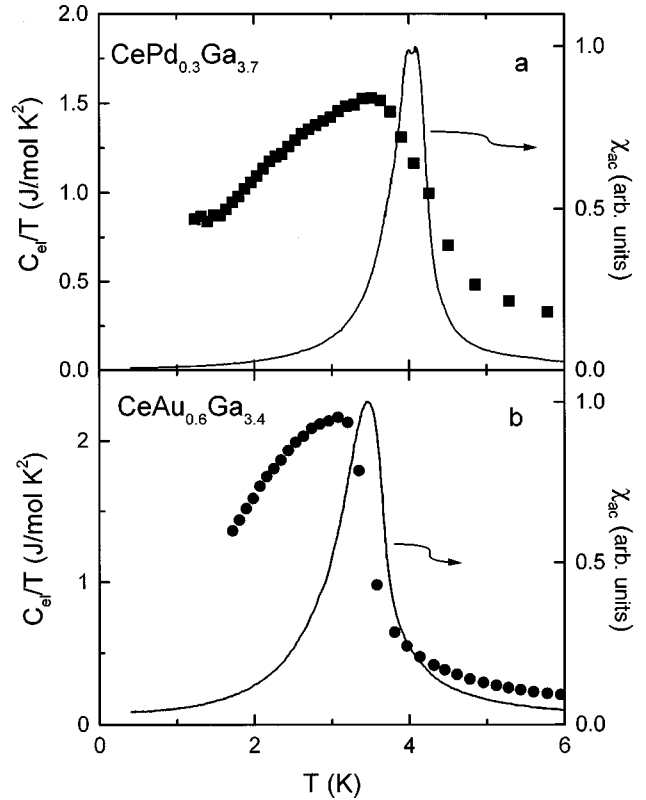
all F Ce binaries[2] and ternaries such as $CePdSb$,[17] $CeCuSi$,[18] and $CeRu_2Ge_2$.[19,20] (iv) Another distinctive feature observed in F Ce-based compounds is that the electrical resistivity (ρ) does not show the characteristic Kondo-like negative slope above the Curie temperature T_C (as verified in $CePt$,[21] $CePd$,[22] or $CePdSb$.[23]) Nevertheless a maximum of $\rho(T)$ at high temperatures may be present due to the eventual hybridization of the excited CF levels. Finally, (v) the Curie-Weiss temperature [extracted from a low-temperature extrapolation of the inverse magnetic susceptibility $\chi^{-1}(T)$] should be close to T_C, whereas the extrapolation from temperatures above the CF splitting may have a larger but negative value also due to hybridization of exited CF levels.[21–23] The only exception to these general properties is found in $CePd_2Ga_3$,[24] which shows a large γ_{LT} value ($= 0.3$ J/mol K^2), despite the fact that it fulfils all the other conditions.

Concerning the thermodynamical analysis of the magnetic GS of these compounds, the entropy gain of the magnetic phase was evaluated as $\Delta S_m = \int C_{el}(T)/T dT$. Within the experimental indetermination, all the studied compounds show $\Delta S_m \geq 0.86R\ln 2$ (see Table I) and then can be considered as fully magnetic, i.e., with practically no degrees of freedom involved in hybridization effects. This is confirmed by the $\gamma \leq 10$ mJ/mol K^2 values (evaluated as C_{el}/T for $T \to 0$, see also Table I), which are comparable to those observed in stable-valent rare-earth compounds.[2]

With respect to the magnetic properties, all the studied compounds reach a magnetization value of about $1\mu_B$ at 2 K under a field of 5 T (see Table I). The positive Curie-Weiss temperatures (Θ_p^{LT}, see Table I), extrapolated from tempera-
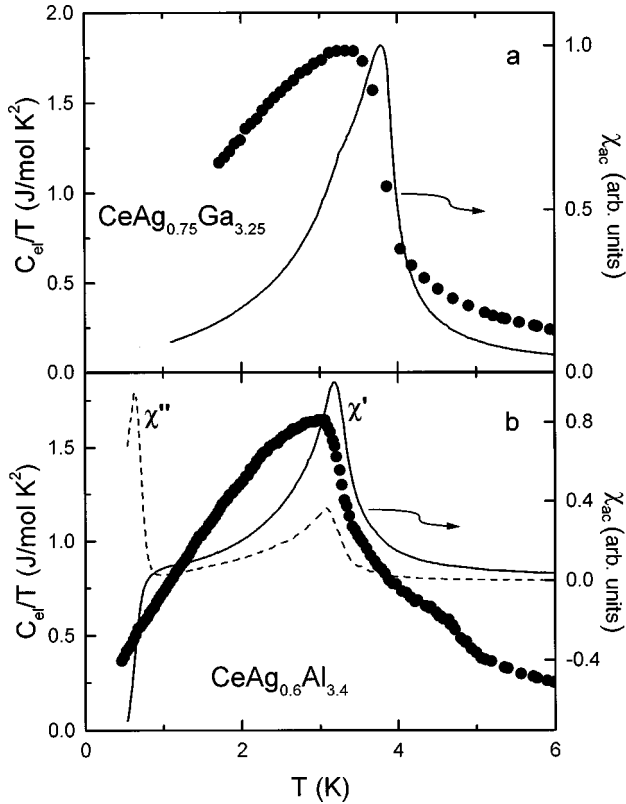
FIG. 3. Low-temperature specific-heat divided temperature and inductive component of the ac susceptibility of $CeAg_yX_{4-y}$, for $X=Ga$ (a) and Al (b).

FIG. 4. Field dependence of the magnetization of CeT_yGa_{4-y}, for $T=Ni$ (a) and Cu (b).

tures well below the CF splitting[9,10,14–16] confirm the development of ferromagnetic interactions right above the ordering temperature. However, one must remark that for the "$3d$" T elements, the Θ_p values obtained from a high-temperature extrapolation (Θ_p^{HT}) are negative,[20,10] suggesting that the excited CF level could be hybridized. This possibility is supported by the $\rho(T)$ dependence at temperatures on the order of the CF levels splitting that correspond to some broadened levels.[9,10] Such a situation is also observed in other Ce compounds, e.g., in the $Ce(Pd_{1-x}Ni_x)$ F system, whose CF excited levels hybridize as x increases without losing the F character of its GS, according to the zero slope of $\rho(T)$ right above T_C.[25]

Although the general behavior of this family of compounds is dominated by ferromagnetic interactions, not all of these compounds can be recognized as clearly having a long-range ferromagnetic order parameter. A basic structural factor of these systems is that they do not have a stoichiometric composition and therefore there is an intrinsic disorder in there, mostly depending on the relative size of the T atoms with respect to the X ones. This atomic disorder occurs at the $4e$ sites, which in these compounds is the Ce next-neighbor atom.[16] As expected, the strongest distortion is presented by $T=Ni$ (the smallest element of this group). The difference in temperature between magnetic and thermal maxima of $CeNi_{0.5}Ga_{3.5}$ indicates a strong magnetic instability in this compound. An AF character for the $T=2$ K isotherm of the magnetization is confirmed by an Arrott's plot[26] (M^2 vs H/M, see Fig. 7), which contrasts with the presence of an incipient spontaneous magnetization observed in the $T=3$
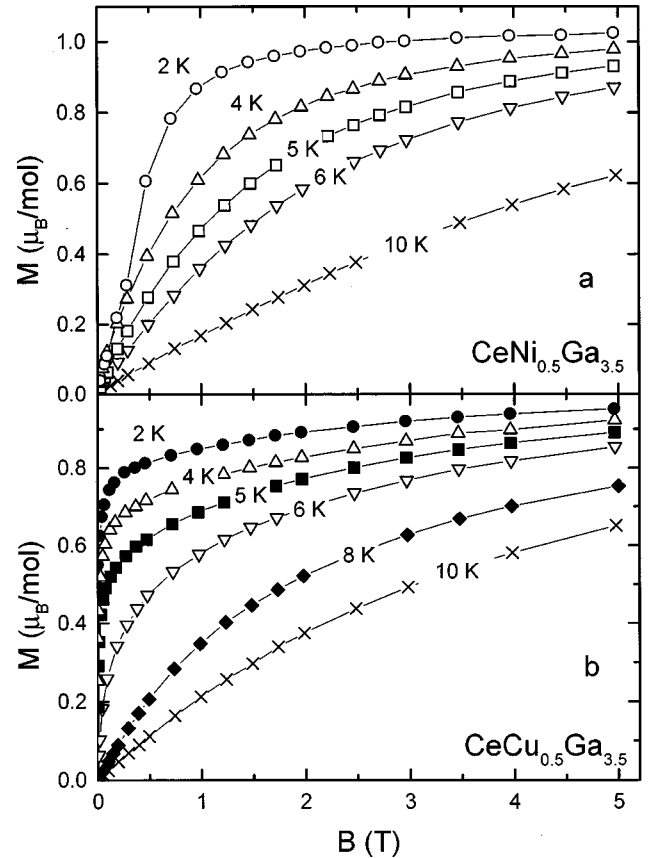
and 4 K isotherms. Further evidence of an AF GS in this compound is given by the $C_{el}(T)$ dependence below the ordering temperature [T_m, defined as the temperature of the maximum of $C_{el}/T(T)$ in Fig. 1] which follows a T^n law with $n=2.7$ (as shown in the inset of Fig. 8), close to the AF dispersion relation. Above T_m, the $C_{el}(T)$ dependence is dominated by magnetic fluctuations. The tail of the C_{el} jump extends well above T_m following an AT^{-2} dependence (with $A=46$ J K/mol, see Table I) up to at least four times T_m, see Fig. 8. In this compound, no anomaly in $C_{el}(T)$ is observed where $\chi(T)$ shows its maximum. A similar situation was observed in CePdSb where this shift in temperature of the respective maxima was attributed to the strong anisotropy of the magnetic interactions.[17] Then a tentative description of the magnetic evolution of this compound can be given as due to ferromagnetic fluctuations below about 10 K, which reach their maximum at 4.2 K. At $T_m \approx 3.2$ K the thermodynamic transition results in an AF structure.

It is remarkable that, with the exception of $T=Ni$, the exponent of the $C_{el}(T) \sim T^n$ dependence of the ordered phase ranges between $1.8 < n < 2$, higher than expected for an ideal F dispersion relation. These values are extracted from a log vs log representation, as shown in detail in Fig. 10 for $CeAg_{0.6}Al_{3.4}$. In coincidence with the thermal properties, the respective magnetic moment measured at 2 K and $B=5$ T (see Table I) are the expected for a full trivalent Ce-ion GS (Γ_{t7}^1) in a tetragonal crystal symmetry.[27] In the case of CeNi$_y$Ga$_{4-y}$ and $CeAg_yAl_{4-y}$ a weaker magnetization was observed at low fields (0.2 T, see also Table I).
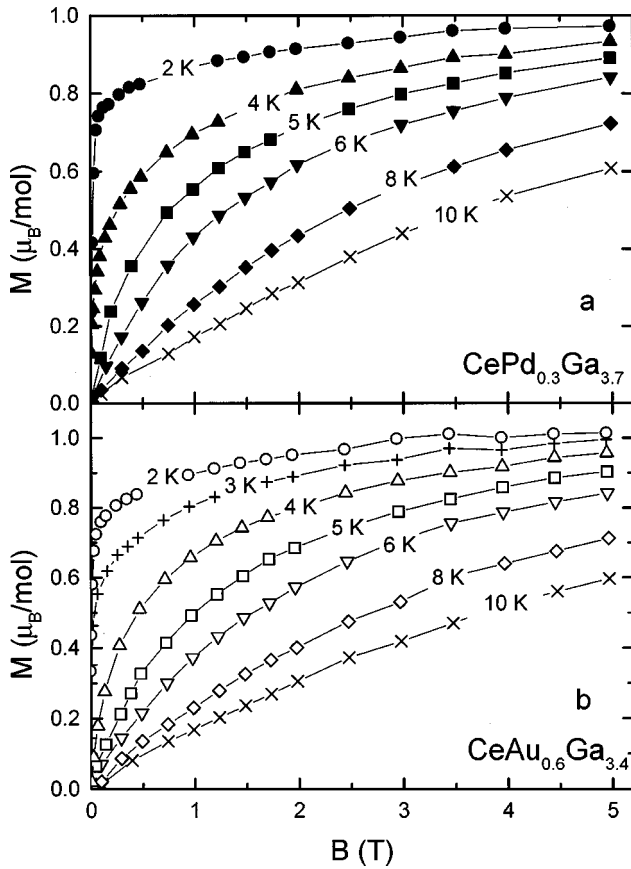
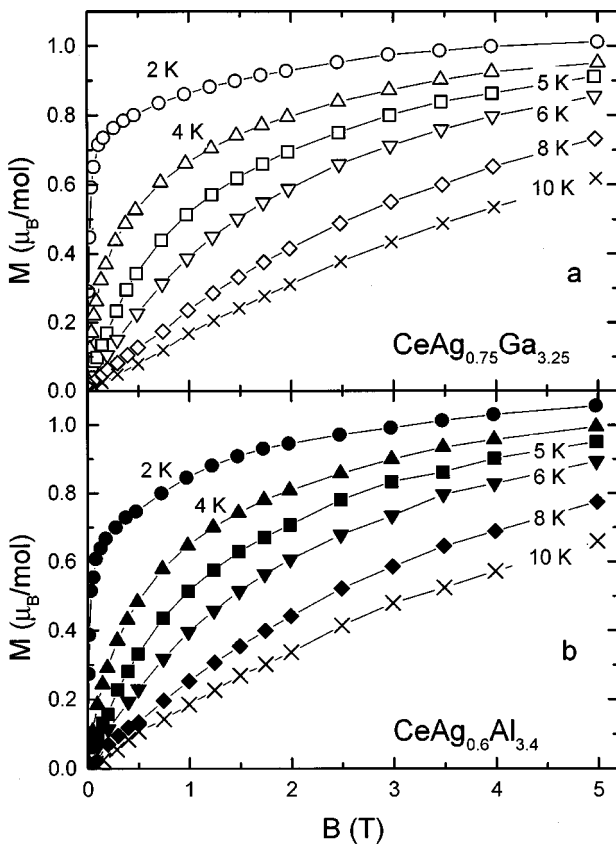FIG. 5. Field dependence of the magnetization of CeT_yGa_{4-y}, for $T = Pd$ (a) and Au (b).

FIG. 6. Field dependence of the magnetization of $CeAg_yX_{4-y}$, for $X = Ga$ (a) and Al (b).
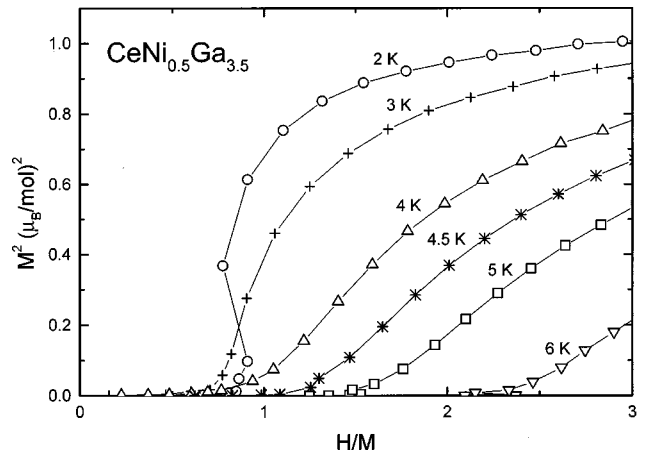

FIG. 7. Low-temperature magnetic-field dependence of the $CeNi_{0.5}Ga_{3.5}$ magnetization in an Arrott's plot.

In this family of compounds "frustration" effects should not be excluded as the possible origin of the F state. From the crystallographic information one sees that the "c/a" ratio of the lattice parameters ranges between 2.42 and 2.49 (Refs. 14–16,28). It is known that for a critical value of $c/a = 2.449$, the second and third Ce neighbors lie at the same distance, then a triangular network is formed in the [110] plane, which gives the conditions for frustration of an eventual AF interaction. Despite the fact that the first Ce neighbors do not belong to the same [110] plane they should be affected by the same frustration mechanism. Again $CeNi_{0.5}Ga_{3.5}$ escapes from this situation because its "c/a" ratio has a lower value (i.e., $c/a = 2.40$) and because the structural disorder produced by the difference in size of Ni and Ga is important. This possibility is supported by the fact that, as the y concentration increases, the F interaction weakens (as in $CeAu_yGa_{4-y}$, Ref. 28) in coincidence with the c/a increase. In any case the stability of the F phase is weakened by the increase of the T concentration. As it was observed through the $\rho(T)$ dependence right above T_C in $CeNi_yGa_{4-y}$, $CeCu_yGa_{4-y}$ (Ref. 9) and $CeAu_yGa_{4-y}$ (Ref. 28), the characteristic Kondo negative slope develops
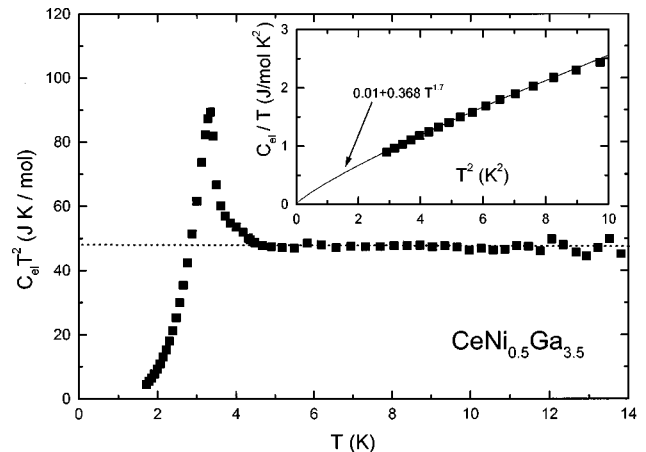

FIG. 8. High-temperature specific heat of $CeNi_{0.5}Ga_{3.5}$ in a $C_{el}T^2$ versus T representation to show the $C_{el} = f(T^{-2})$ dependence for $T > T_m$. Inset: low-temperature ($T < T_m$) $C_{el}(T)$ dependence analysis.
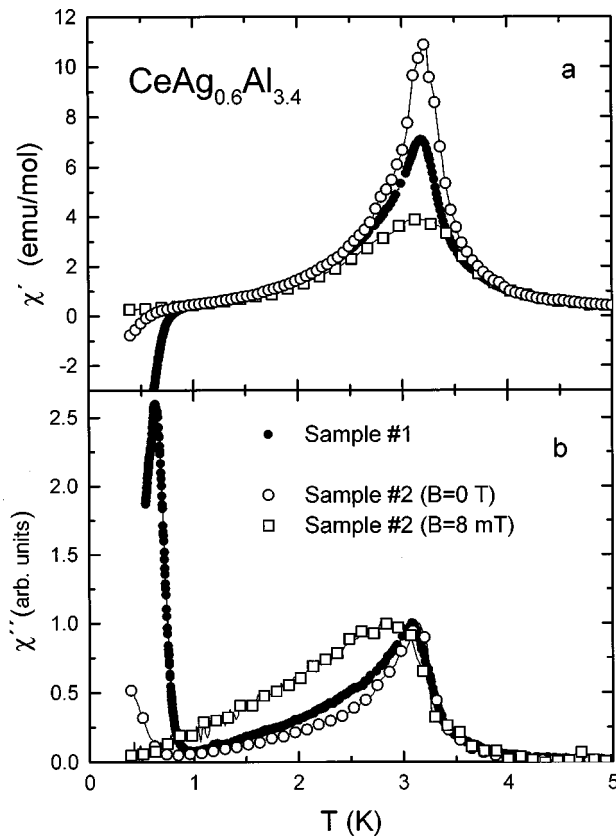
FIG. 9. Low-temperature inductive (a) and dissipative (b) components of the ac susceptibility of $CeAg_yAl_{4-y}$, to show the ferromagnetic and superconducting transitions.
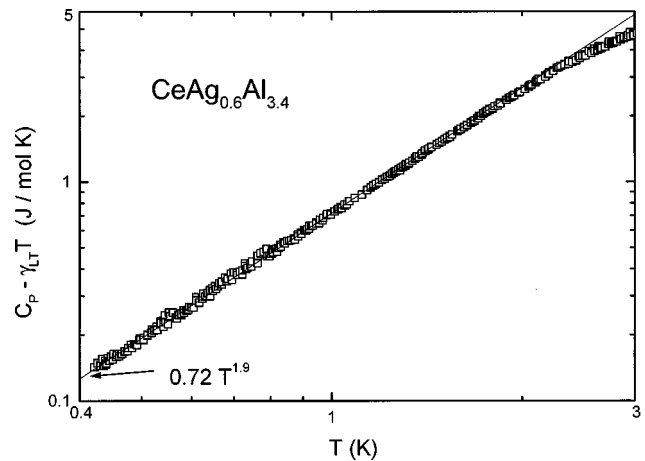

FIG. 10. Low-temperature $C_p(T)$ dependence of $CeAg_yAl_{4-y}$, with $\gamma = 8$ mJ/mol K, in a log-log representation to show the low-temperature dependence analysis.

as soon as the F character disappears.

As mentioned before, $CeAg_{0.6}Al_{3.4}$ also shows an abnormal behavior. In this compound the $\chi_{ac}(T)$ signal becomes diamagnetic at $T_s = 0.63$ K [see Fig. 3(b)]. To this end we have analyzed inductive and dissipative components of χ_{ac} in zero and applied dc fields of 0.8 and 8 mT (see Fig. 9) and concluded that they indicate a superconductive transition. Notice that, regardless of the arbitrary units of χ_{ac}, the diamagnetic signal is comparable to the F one, though it is practically not detected in the $C_{el}(T)$ measurements. Such a diamagnetic signal can be attributed to two possible origins. One is related to the presence of Al layers formed by these atoms in the $4d$ crystallographic position of the $CaZn_2Al_2$ variant of the $BaAl_4$-type structure,[13] that involves nearly

70% of the atoms per formula unit. The other is related to some Al segregation observed in the EDS analysis. Even in the first case, no jump is expected to be seen in $C_{el}(T)$, because 3.4 Al atoms/mol would produce a $\Delta C_{el}(T_s) \lesssim 10$ mJ/mol K^2, a few percent of the ferromagnetic contribution at that temperature; see Fig. 10. Concerning the second possibility, pure Al becomes a superconductor at $T_s = 1.175$ K, but magnetic impurities could reduce and broaden such a transition like in sample 2, whereas in sample 1 the transition looks quite sharp; see Fig. 9. In any case, the suppression of superconductivity under an external field of 8 mT agrees with the critical field $B_{co} = 10$ mT of pure Al.

We conclude that this family of ferromagnetic compounds confirm the fact that the F exchange weakens the hybridization strength, as predicted by some theoretical models.[7] The absence of hybridization effects on the Ce-GS is confirmed by the AF compound $CeNi_yGa_{4-y}$, through its ΔS_m, γ and $d\rho/dT = 0$ values. Nonetheless, in each case a detailed analysis of the origin of ferromagnetism has to be done and the eventual hybridization of the excited CF levels need to be taken into account for a realistic description of the system.

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