

The Effect of Heavy Fermion or F-Semiconductor Systems on Gd-Intermetallic Systems

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Abstract. Based on the instability of magnetic behavior through the concept of local exchange and hybridization, a new class of heavy fermion is constructed with a stable local magnetic ion, "Gd". The lattice constant, DC/AC- magnetic susceptibility, $\chi(T)$, and electrical resistivity, $\rho(T)$, measurement in the magnetic unstable intermetallic compounds of $Gd_2A_xB_{1-x}$, show that (I) The hidden magnetic internal energy is manifested by shape and field dependence is strongly at $x = 0.4$, (II) The lattice parameter of a crystal and magnetic structure, as well as high transition temperature, " T_c ", strongly depends on the conductive electron concentration. Both the functional change of the lattice parameter (non-Vegard behavior) and magnetic character with electron concentration suggest that the free electrons are not completely free and are somehow participating in the band functions and magnetic character, and that (III) There is a coexistence of Kondo behavior and magnetic ordering "re-entrant antiferromagnet" for $x = 0.4$ in the range of temperature $30 < T_k < 90$ K with $T_N = T_{max} = 30$ K, so that (IV) Finally, the metal insulator-like behavior with a complete quench of magnetic ordering occurs antiferromagnetically, named superparamagnet, at a certain conductive electron, " $x = 0.3$ ", where the sample is field dependent (on which, it is suggested, $\sum \varepsilon_{ij} J_{ij} = 0$).

Keyword: Possibility of Kondo effect on "Gd".

INTRODUCTION

The instability of three dimensional intermetallic compounds of an "F-atomic" state, in which some of the F-electrons become itinerant at low temperatures, has dominated heavy fermion "HF" research since 1980, based on the unstable ground state magnetic ion, "Ce, U" [1-3]. In contrast, no new families of "HF" on a true stable ground state with an integral F-electron and ordered local moments on the F-state, such as "Gd", have been discovered. This experimental report is part of a report to observe the Kondo lattice behavior, which results in HF-formation for Gd-Intermetallic on the duality of conductive electron concentration "c.e.c". A key area of investigation is the study of exchange fluctuation and magnetic moment instability, named "energetic moment", caused by the width of the V.B.S (Virtual Bond State), in terms of inter band mixing, due to strong spin polarization as F-d exchange and

d-d interaction [4], which is manifested by the hidden energy of shape dependence [5]. The characteristic competition (or even completeness) between the magnetic intersite interaction "RKKY-exchange" and demagnetizing on site, named the "Kondo interaction", could be the cause of instability. The competition and, finally, completeness is related to the strength and sign of coupling between the conductive electrons, "c.e.", and local 4F-state (L.F), which gives rise to the magnetic polarization of (c.e.) [6] ($J_{f-c} J_{ex}^{DI} + J_{ex}^{DI}$) direct and indirect exchange interaction, "DI, IDI". The relative strength of the "exchange interaction" and the mixing of potential " V_{f-c} " manifest themselves on the boundary of duality of the Localized Conduction (L-C) electron, which belongs to either the "HF" or mixed (s-d) valence region [7,8]. This is observed on the resistivity anomalies near the magnetic ordering temperature, as well as the instability of the magnetic structure, where it is described in Anderson [9,10] and Kondo [11] models, in terms of hybridization or electron transfer within a defined ground state and moment compensation, respectively. However, in the Kondo/Anderson theory of magnetic impurities, this occurs due to virtual excitation, whereby an F-electron is promoted to a band state at the Fermi level and

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the F-hole is filled by a conduction electron with spin of the opposite sign. Such virtual fluctuations in the valence, thus, resulting in spin fluctuations, quench the magnetic moment at low temperatures [12]. However, the relative localization and delocalization of the F-electron will affect both the F-F interaction via the interatomic space, " r_c ", comprised of the mean free path, " λ_m ", of conduction electrons and the interband mixing, by which it strongly affects the character of "c.c.", as in the mixing of " $5d - 6s$ " on the Gd-intermetallic system [13]. The dominant interaction between the $4f$ and $5d$ character of the conduction electron is also pointed out [14].

In order to understand the cause and rule of the above theoretical assumption on a stable ground state, the similarity in the reported data on the selected ternary intermetallic compound, $\text{Gd}_2\text{Au}_x\text{Al}_{1-x}$ ($0 < x < 1$), offers an opportunity to:

- Investigate a system similar to both transition metals, where itinerant electrons play an important role and to those with well-localized spin, in terms of the intermediate of a $6s - 5d$ electron (duality of c.e.c);
- Prove that the sign and strength of the exchange parameter, $J(0)$, determining the nature of the conduction electron, has affected the topological position of the magnetic ions;
- Show the peculiarity of the onset of unstable ferromagnetic ordering, "F.M", at a fairly high temperature and the being stable re-entrant "AF.M" at a low temperature, where $\mu_{\text{exp}} < \mu_{\text{free-ion}}$;
- Consider the non-Vegard rule behavior, which is caused by a distortion of the topological position of magnetic ions in the " $a - b$ " plane;
- Clear out the hidden energy, due to the energetic moment, which is caused by the internal and the fluctuation field.

SAMPLE PREPARATION

The samples were prepared by the melting stoichiometric method, using a $4N$ -element for Gd and a $5N$ for Al and Au in a standard arc-melting furnace in an Ar-atmosphere. The X-ray diffraction of these compounds, which were taken before and after the annealing process, revealed an orthorhombic crystal structure with a Pnma space group [15]. The lattice parameters were found to be strongly sensitive to the annealing processes, especially for the compositional range of $0.2 < x < 0.7$ (Figures 1a and 1b), where the shape dependence of the magnetic structure manifested, as described in detail, in [5]. The annealing temperature, T_a , was 650°C for 96 hours.

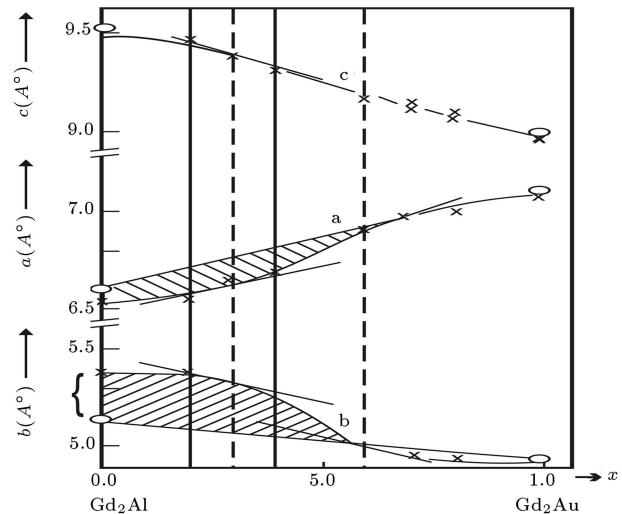


Figure 1a. Lattice constant of the system $\text{Gd}_2\text{Au}_x\text{Al}_{1-x}$.

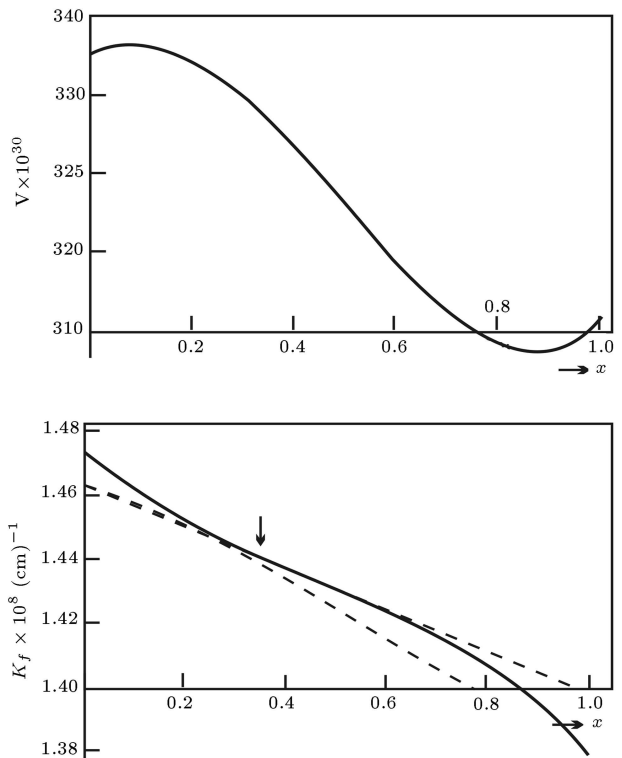


Figure 1b. Variation of " K_f " of Fermi sphere with x .

Magnetic measurements were performed by (i) Vibrating the sample magnetometer in the range of temperature 4.2 to 300 K and in a field up to 20 kOe; and (ii) A.C. magnetic susceptibility by the standard induction method in a closed refrigerator down to 10 K and in an A.C. field up to 30 Oe.

Electrical resistivity measurements were carried out using (i) The closed refrigerator method; and (ii) Two selected samples by a super conducting magnet cryostat (cryogenic liquid He). The current and voltage contacts were made with silver paint.

RESULTS AND DISCUSSION

The measured magnetic susceptibility, $\chi(T)$, and resistivity, $\rho(T)$, of the selected samples are given in Figures 1 to 4.

The lattice constants, magnetic behavior and the strength of their correlation are evaluated where:

- The lattice parameters of these systems show strong deviation from Vegard's rule, which is one of the most frequently used methods in studying the effect of intermediate valence (the unstable magnetic behavior of Ce, Yb and Sm intermetallic systems). Figure 1a shows the lattice constants dependence on the conduction electron concentration. The volume of the unit cell increases as "c.e.c" increases (Al^{3+} was substituted for Au^{1+}), while the ionic radius $R(\text{Au}^{1+}) > R(\text{Al}^{3+})$ (Figure 1b).
- A typical Curie-Weis (C-W) behavior with a corresponding high paramagnetic Curie temperature ($\theta = T$) is observed with low value of effective moment, $\mu_{\text{eff}} = 4.2 - 5.2\mu$, in the range of available temperature, which is a function of c.e.c, as in Figure 2. Discontinuous linear distribution of $\chi^{-1}(T)$ reveals chemical clusters, which may introduce short range interaction.
- The effects of increasing c.e.c on $\chi(T)$ are strongly noticeable at relatively high magnetic transition temperature, T_c , in respect of $\chi(T)$, to a lower value (increasing T_c) with increasing c.e by $\Delta T_c = 20^\circ\text{C}$ with $\Delta x = 0.2$ (Figure 2). It becomes more destabilized in the range of $x = 0.2 - 0.4$. This variation is more prominent for $x = 0.4$, where the main magnetic instability occurs and T_c is difficult to be estimated (Figure 3a).
- At temperatures below 300°C and in low magnetic

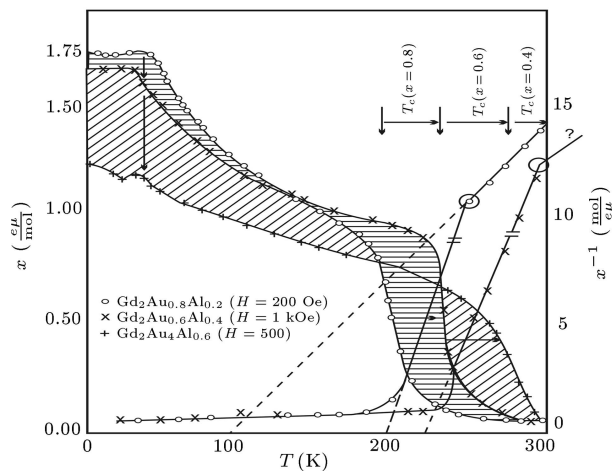


Figure 2. Temperature dependence of the susceptibility $\chi(T)$ and χ^{-1} for (o) $x = 0.8$, (x) $x = 0.6$, (+) $x = 0.4$, and () $x = 0.4$ in high field.

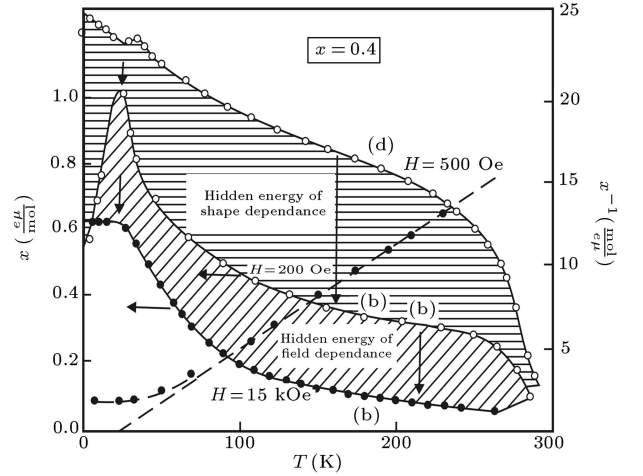


Figure 3a. Temperature dependence of D.C susceptibility for $x = 0.4$ in different shapes and fields.

fields, two closed magnetic transitions, followed by a broad maximum, are observed (Figure 3a). The small plateau, which is observed at intermediate temperature, is typical of a re-entrant, near the F.M-SG transition temperature.

These two transitions, with a small intermediate plateau temperature, are developed by increasing the c.e.c, which is associated with the depression of $\chi(T)$, as mentioned before, and are distributed among each other in such a way as to become one transition with a broad range of ordering temperature at certain c.e.c ($x = 0.4$). This transition is found to be a too smeared mixture of two transitions, so, it is difficult to estimate the transitions magnetic order from $\chi(T)$, due to the great dispersion of exchange near double FM-AF.M percolation in a low field. The system almost reaches P. M. around $x = 0.4$, defined by the low value of $\chi(T)$.

However, this behavior could be due to a canted system or by absorbing conduction electrons that reduce the FM moments; the latter can even result in moment compensation, which is expected when leaving the stable moment regime, which is the main goal of this paper.

The low value of μ_{eff} , defined by θ_p , which corresponds to a high T_c , is also a confirmation of the above suggestion.

Moreover, the dependence of the distortion of the crystal structure and the sign and strength of exchange, J_{ij} , on the c.e.c, as well as the hidden energy of the magnetic shape dependence (Figure 3a) [5], bring up the question of if, really, there are any strong correlations between the magnetic phase transition and c.e.c. This correlation is more pronounced by the continued lowering of the magnetic transition with a low value of $\chi(T)$, which is due to the energetic moment compensation. That is, the magnetic structure of $x = 0.4$ should be expected to follow the Kondo

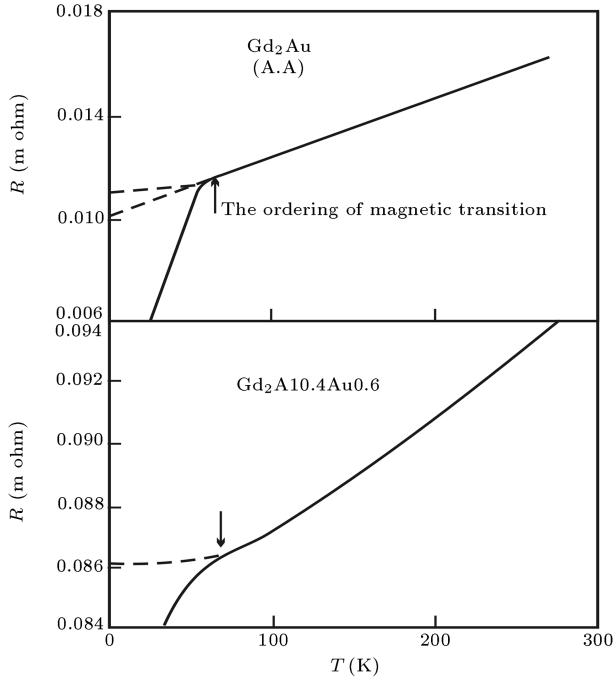


Figure 3b. Temperature dependence of the resistivity for $x = 0.4$, $x = 0.6$ and $x = 0.2$.

behavior, by which the experimental measurement on $\rho(T)$ supports the authors suggestion (Figure 3b). Where, as the density of conduction electrons increases, the beautiful sharpness of $\rho(T)$ at $T = 60$ K for Gd_2Au is spread out for $x = 0.6$ and completed at $x = 0.4$ in the Kondo regime, where it is rounded and a minimum is formed around $T = 60$ K (Figure 3b).

Such virtual fluctuation in the valence, thus, results in spin fluctuations, which quench the magnetic moment at low temperatures.

Consequently, if $x = 0.4$ is close to the critical value, near the leaving of the stable moment regime, then, it should behave extremely sensitively to the physical parameters, which take the sample to its stable position, by which the following external parameters are applied:

- (i) The shape of the sample at $x = 0.2$ [5];
- (ii) The critical external magnetic field, at $x = 0.4$, in the field of $H_{\text{ex}} = 15$ kOe (as in Figure 3a) and finally;
- (iii) The critical electron concentration at $x = 0.3$. An important dramatic phenomena, named superparamagnet, is observed at $x = 0.3$ which is a stable magnetic moment region. Here, the system is field and shape independent and a complete P. M. order (Figures 4a and 4b), defined by the linear field dependence of $\sigma(H)$, $\chi^{-1}(T)$, in the range of 0-20 kOe and temperature range of 0-300, is exhibited.

Finally, the completeness of the Kondo lattice-like

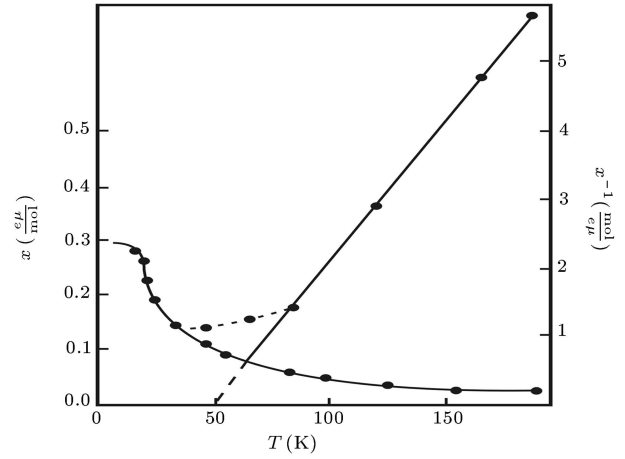


Figure 4a. Temperature dependence of the D.C susceptibility for various isotherms for $x = 0.3$.

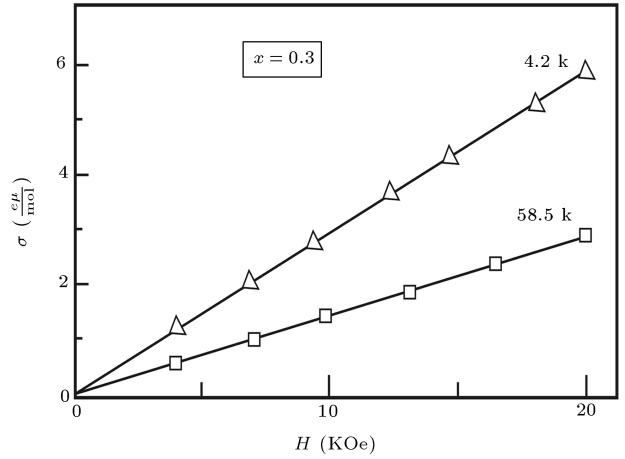


Figure 4b. Field dependence of magnetization for various isotherms for $x = 0.3$.

behavior, named the HF formation, is suggested for this composition, which is confirmed by $\rho(T)$ in Figure 4c and is curved to a maximum around 80 K.

Consequently, the following corresponding results could be concluded:

- a) Local electron behavior, which is manifested by a depression of the magnetic susceptibility, the hidden energetic moment caused by continuous lowering of the magnetic transition temperature, with a low value of $\chi(T)$;
- b) Itinerant electron behavior, which is shown by electrical resistivity, as well as the structure distortion of the topological position of the magnetic ions, is presumed to be due to the short range ordering effect, as the thermal fluctuation of intra-cluster exchange extends through the long range of inter-layer interaction. The fluctuation is around the internal magnetic field. The internal and field fluctuation are originated from the spin-spin ex-

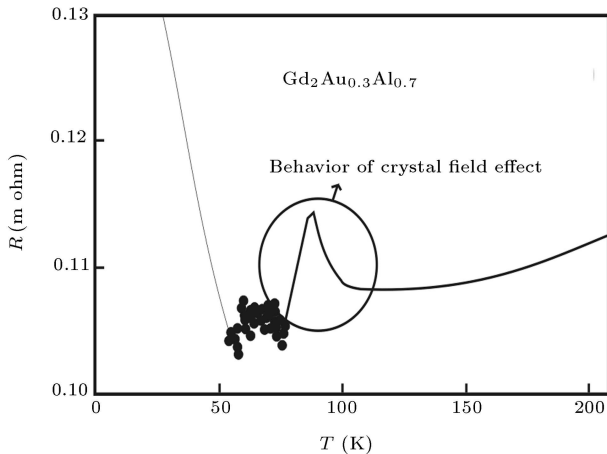


Figure 4c. Temperature dependence of the resistivity for $x = 0.3$.

change, which is due to the interatomic space and the number of nearest neighbors (n.n) (included in the clustering region of $3.39\text{\AA} < R_C < 3.6\text{\AA}$) [16].

However, the understandings from this experimental observation are the following: while the “ c ” parameter is almost expanded linearly, the “ a ” and “ b ” parameters are drastically being nonlinearly contracted and expanded, respectively (Figure 1). This means that distortion in the “ $a - b$ ” plane depends on the interatomic space in the clustering region, R_c , and the other factors, such as pair potential [17]. The created distortion energy can be either removed or minimized by changing the heat treatment, defined by the annealing processes. The change of unit cell then affects the two non equivalent lattice sites of 8 n.n [16]. This can be considered as a kind of topological distortion (via Fermi sphere “ K_F ”). The strong distortion in the “ $a - b$ ” plane is due to the strong intra-plane exchange interaction of the magnetic ions ($J_{sn} \rightarrow J_{in}$)^($a-b$) [5].

The distortion depends on the energy of a band and some part of the exchange. The crystal will strain itself, such that to lower either, or both, the energy of the band that contains the conduction electrons (c.e), and exchange energy, will result in lowering the free energy of the crystal.

This distortion can change the sign and strength of the phase transition and, also, the phonon dispersion, due to the magnetoelastic behavior, because the magnetic phase transition strongly affects $\rho(T)$ around $x = 0.4$ (Figure 3b). A phonon with a well defined wave-number, thereby, couples coherently with the spin polarized conduction electron (c.e).

Consequently, a deformed potential, for these spin polarized particles (heavy quasi particles) that are participating in the crystal structure, increasing T_c , with lowering the corresponding $\chi(T)$, and nonlinearity of $\rho(T)$, is obtained.

From the above discussion, it can be suggested that the coexistence of fine-particles (ions) and quasi-like particles (polarized c.e. with inter band mixing in the clustering region) will stabilize the system in certain external magnetic fields (e.g. $x = 0.4$, $H = 15$ kOe), as well as certain values of “c.e.c” ($x = 0.3$). This will make a balance between the topological distortion and exchange, to minimize the free energy, as well as lowering the entropy (Figure 4b).

The dramatic difference in the magnetic structure, at low and high fields, at $x = 0.4$, is almost due to the equality of the external and inter local field, “ $\mu \cdot H_{in} = \mu \cdot H_{ex}$ ”. The broad magnetic transition, which may also be due to the domination of the magneto energy coupling, (J_{sI}), completely breaks down. This will minimize the exchange energy of the magnetic ions indicated by the low ordering temperature, T_{of} , the characteristic temperature above which the system is almost PM (in high field (straight line $\chi^{-1}(T)$, in high magnetic field (Figure 3)). This also indicates the coincidence of θ_p with T_{of} .

At temperatures below $T_{of} > \theta_p$ (below which a deviation from linearity, of $\chi^{-1}(T)$ can be observed) where $H_{in} \geq H_{ex}$, the exchange fluctuation may start becoming frozen antiferromagnetically by short range intracluster interaction (associated with cluster). The clustering region starts tunnelling, on which intercluster interaction competes with intracluster exchange through the strong polarized “c.e”.

This condition is the crossover of a different kind of magnetic instability, which is related to the exchange splitting of the HF-band. The coincidence of magnetic ordering and increase in resistivity, as shown in Figure 3, for $x = 0.4$, may also indicate the existence of an antiferromagnetic gap energy [18], which is due to interband mixing developed by intracluster exchange.

However, the Kondo lattice-like behavior is suggested for this composition ($x = 0.4$), as is confirmed by $\rho(T)$ for $30 < T_k < 90$, Figure 3, (T_f corresponds to $T_{max} = T_N = 30$ K). Consequently, the changes of $\rho(T)$ can be explained by two phenomena; long range magnetic ordering, developed by the spin polarization of the conduction electrons, “ J_{cf} (interplaner), and scattering of c.e.s (spin flip intracluster exchange).

The Kondo lattice behavior may be developed by the peculiar dispersion of the, so called, band quasi particle, through the $s \rightarrow d$ transformation ($J_{fd} > 0$). Thus, one will have $\sum \varepsilon_{ij} J_{ij} = 0$, in the clustering region, so, the strong electron phonon coupling is also suggested. In this case, it has been observed that the system is field independent and in PM order, defined by the linear dependency of $\sigma(H)$ and $\chi^{-1}(T)$ (Figures 4a and 4b). The magnetization measurement, $\sigma(H)$, on polycrystalline samples with two different shapes (needle and powder), so far seem to imply that the F.M moment can hardly be saturated in finite magnetic

fields up to 20 kOe. Any experiment on the magnetic anisotropy of a single crystal of these samples (based on Gd, which is in an s -state) has not yet been performed, since, for these metallic systems, a simple interpretation of the Crystal Field Effect (CFE), via the ionic point charge of the neighboring ions, does not work. CF- schemes are essentially due to the effect of the instability of a $5d$ -intermediate character (originating from both the atomic RE configuration ($4f\ 5d\ 6s^2$) and strong spin polarization of c.e as interband mixing, in terms of the coupling of anisotropic $4f$ charge clouds to “semic-c.e” and phonon), developed by an intercluster interaction, which forms a V.B.S. The produced CFE, which is masked in D.C susceptibility in low field ($J = 500$ Oe), can be revealed in A.C. susceptibility in low field ($H = 10$ Oe), at temperature $T > 100^\circ\text{K}$ (Figure 5).

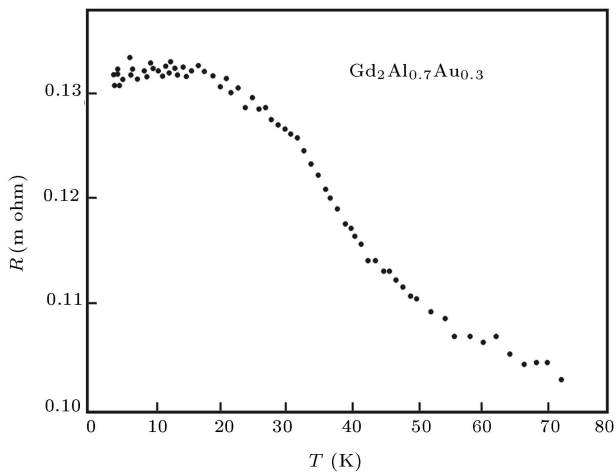


Figure 4d. Temperature dependence of the resistivity for $x = 0.3$ in superconducting magnet cryostat (cryogenic liquid He).

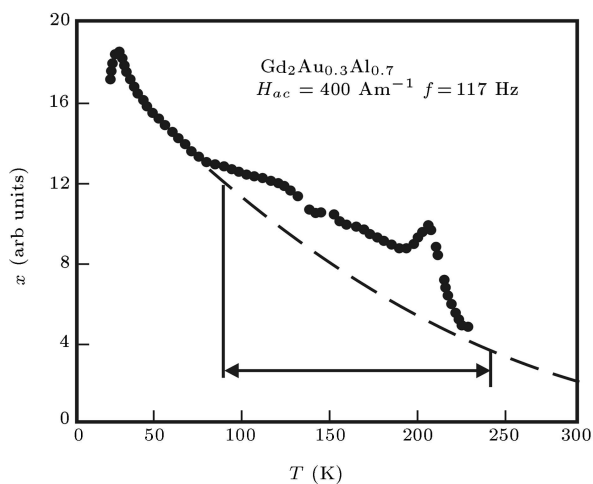


Figure 5. Temperature dependence of the A.C. susceptibility for $x = 0.3$ in low magnetic field ($H = 10\text{Oe}$).

Thus, the system can lead to HF-formation, developed by the completeness of the Kondo lattice (Kondo - behavior at any T , Figures 4c and 4d) defined by the virtual band state (V.B.S.).

The completeness of strong short range intracluster produces a potential barrier attributed to both (i) The transition from coherent to incoherent scattering, where the Mean Free Path (MFP) of conduction electron λ_m is much shorter than the size of clustering region “ R_c ” (ii) Distortion in the ($a - b$) plan and expansion in c -directions (Figure 1).

Under these circumstances, complete quenching of local moments, in the clustering region of the lattice, will occur where $\sum \varepsilon_{ij} J_{ij} = 0$. The quenching is supported by the AF.M coupling developed by a partial overlap of up spin and down spin semielectrons and of the balancing of quasi particles (strong polarized electrons located on atomic space) and fine particles (magnetic - ions) developed by intercluster interaction (Figures 4a and 4b).

Thus, the magneto elastic coupling can drastically alter the character of the phase transitions and, hence, strong correlation between $\rho(T)$ and $\chi(T)$ will exist. The characteristic, in this case, lies well below the CF-energy, where it appears to be at $100 < T_{cf} \leq 200$, developed by the interband mixing of d -like electrons in low field.

CONCLUSION

Based on the s -state of “Gd” and experimental measurements in two different limiting boundaries: Resistivity $\rho(T)$ and D.C/AC susceptibility $\chi(T)$, the following assumptions can be suggested.

- The ground state of “ $4f$ ” is assumed to be completely stable with well defined local moments, as Gd is in an s -state.
- The mixed f -valence fluctuation can be avoided, while the exchange fluctuation originating from spin/spin interaction (local field) plays an important role and is manifested by short range interaction of the instability of the magnetic structure at high temperature. This behavior is related to the number of magnetic ions and interatomic spaces in the clustering related to the number of magnetic ions and inter - atomic space in the clustering region “RC” [15].
- The interband mixing, as in opposite directions (valence local fluctuation), or the existence of an AF.M energy gap [18] through the intra-cluster interaction, is possible, where the formation of a new Kondo lattice for the “Gd” intermetallic compound with the magnetic ordering is observable.
- Finally, HF formation takes place by the completeness of the Kondo lattice through the intra

planner short range exchange ($J_{ij} - J_{sh}$), which is developed by intracuster exchange, via a virtual bound state (V.B.S) at $x = 0.3$ (Figure 4). The form of V.B.S can be considered by the effect of $5d$ -electrons originating from the atomic Gd-configuration [$4f^n - 5d - 6s^2$]. This can cause severe anisotropy in RKKY conduction electrons, which is masked in D.C magnetic measurements ($H = 5000$ Oe) and manifested in A.C. magnetic susceptibility ($H = 10$ Oe) at $100 < T < 200$ (Figure 4c).

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