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PAIR-BREAKING AND MAGNETIC ORDERING IN THE PSEUDO-QUATERNARY INTERMETALLIC SERIES (Y,Gd)Ni_2B_2C

M.El Massalami, S.L.Bud'ko, B.Giordanengo, M.B.Fontes, J.C.Mondragon and E.M.Baggio-Saitovitch.
CBPF, R.Xavier Sigaud 150, 22290-180, Rio de Janeiro, Brazil
A. Sulpice
CNRS CRTBT BP 166 38042 Grenoble Cedex 9

ABSTRACT

The alloying influences on the superconducting and the magnetic properties of the $(Gd_xY_{1-x})Ni_2B_2C$ series were studied. For $x < 0.25$, T_c is degraded linearly with x as expected from Abrikosov-Gor'kov theory. Magnetization and magnetoresistivity measurements on x=0.22 show that the magnetic saturation is not attained for field as high as 80 kOe implying that even for such a low Gd concentration, antiferromagnetic couplings are quite strong. On the other hand, the magnetic dilution in the Heisenberg antiferromagnet $GdNi_2B_2C$ causes a linear depression of the magnetic ordering temperatures as expected from Molecular Field Theory. The abrupt quench of superconductivity for $x > 0.25$ and the coexistence of superconductivity with the long range antiferromagnetic order for $0.1 < x < 0.25$ are discussed.

Key Words: $(Gd_xY_{1-x})Ni_2B_2C$, superconductivity, magnetic interactions

1. INTRODUCTION

The 4f-magnetism in the newly discovered RNi_2B_2C (R=Y, rare earth) [1-3] has a decisive influence on their superconducting properties [3-15]. As an example, in the antiferromagnetic (AFM) superconductors R = Tm [3,4,7], Er [3,4, 6, 10] and Ho [3-5,12], the s-f interaction depresses (but not quenches) T_c through the de Gennes factor, $(g-1)^2J(J+1)$, and gives rise to a commensurate AFM order that coexists with superconductivity. On the other hand, the abrupt quenching of superconductivity for R =Dy, Tb, Gd is caused by the adverse influence of the prevailing magnetic order[3,4,11,13-15].

In all of the above mentioned compounds, the R-ions concentration is constant. Then the variation in the strength of the interplay between magnetism and superconductivity through this series is due to the variation in the de Gennes factor and in the strength of the magnetic interactions [4,13]. It is interesting to study the competition between the 4f-magnetism and superconductivity as a function of the 4f-moment concentration. The $(Gd_xY_{1-x})Ni_2B_2C$ ($0<x<1$) solid solution is a good candidate for such a study, since the Gd-ion is in a S-state and so no complications due to crystal field effects are expected in the temperature range of interest. Our results show that in the low Gd-concentration, T_c is depressed linearly with x. Moreover, for $0.1<x<0.25$, the RKKY interactions among the Gd-moments are strong enough to establish a long range AFM order in the superconducting state. For $x>0.25$, superconductivity is completely suppressed and the magnetic transition temperatures are linearly related to the magnetic dilution.

2. EXPERIMENTAL

Pellets from the stoichiometric mixtures of high purity elements were arc-melted under argon in a conventional Argon arc-melt furnace [1-6]. Room-temperature structural characterization was carried out in standard Cu-Kα diffractometer. Magnetization were measured on SQUID magnetometer (up to 70 kOe) and on ac susceptometer (500Hz, ~1Oe).

The longitudinal magnetoresistivity (j // H) was measured on a bar of a polycrystalline x=0.22 sample in fields up to 80 KOe using the four-point dc method. The sample temperature was monitored by Carbon glass thermometer with its long axis set perpendicular to the field direction. Temperatures were corrected for the magnetic field influence following the procedure described in Ref.16.

3. RESULTS

The X-ray-diffractograms (Fig.1) confirmed the expected structural character [2] of the polycrystalline samples. Traces of impurities were detected and two of them were identified as RNi_5 and RNiBC. The Rietveld analysis was carried out assuming the structural data as reported in Ref.[2]. The lattice parameters (inset of Fig.1) vary monotonically with the Gd concentration: while the c-parameter decreases, the a-parameter increases. It is worth to recall that all atomic positions (except the z-parameter of boron) are fixed by symmetry requirements. Then, such a concentration-dependent cell lengths variation (which is also observed in the RNi_2B_2C series[2]) can be attributed to the opening of the tetrahedral angle due to the insertion of the large-sized Gd-ion. This entails a compression along the c-axis and an elongation along the basal plane.

For all the studied samples, the resistivity ρ shows a linear metallic behavior down to 30K and then ρ saturates with $\rho(300K)/\rho_{sat} = 3 \sim 10$ (Fig.2) On cooling further, the superconductivity was detected only for $x \leq 0.25$ and T_c degrades linearly with the Gd concentration. Fig.2 demonstrates also that the Gd-rich samples show a remarkable anomalous drop in ρ, indicating a decrease in the incoherent electronic scattering due to the onset of the magnetic phase transition.

The χ_{ac}-T curves (Fig.3) show that Gd-substitution degrades the superconductivity monotonically and with an eventual quenching for $x>0.25$. For the Gd-rich samples, the Curie-Weiss behavior was observed (as an example see Fig.4) with an effective moment $\mu_{eff} = (7.9\pm0.1)\mu_B$.

The sample with $x=0.22$ is situated in the concentration region that separates the superconducting and the normal-state solid solutions. Qualitatively, it has some similarity with the $HoNi_2B_2C$ compound in the RNi_2B_2C series [4]. For that reason, we studied its magnetic and superconducting features in somewhat more details and the results are displayed in Figs.4-6. Fig.4 shows its M-T curves which manifest characteristic Zero-Field Cooled (ZFC) and Field-Cooled (FC) features. For a better understanding of Fig.4, the low-field magnetization loop for this sample, at T = 1.7K, was carried out and it is shown in the inset of Fig.5. It is clear that a small magnetic field of the order of few Oe is enough to quench superconductivity. This explains the pronounced differences in ZFC and FC behavior of Fig.4 because the employed dc field (11.593 Oe) was greater than the critical field.

On the other hand, the magnetization isotherm for $x = 0.22$ sample at $T = 1.7K$ (Fig.5), shows that a relatively large field (~70 kOe) is needed to align appreciably the magnetic moments along the field direction. Since the Gd single-ion anisotropy is negligible, then this implies that even for such a low concentration, AFM exchange coupling are quite strong. In fact, for $x<0.25$, these strong couplings are giving rise to a magnetic ordering even in the superconducting state (see Fig.3). With the addition of more Gd moments superconductivity will be quenched as manifested by Figs.2-3 for $x>0.25$.

The longitudinal magnetoresistivity $\Delta\rho(H)=\{\rho(H)-\rho(H=0)\}/\rho(H=0)$ of $x = 0.22$ sample at 4.7K (Fig.6) shows that the superconductivity is quenched with a small external field and that the $\Delta\rho(H)$ rises fast to its normal-state resistivity value. On the further application of the field; $\Delta\rho(H)$ decreases monotonically. Molecular field Approximation relates $\Delta\rho(H)$ to the thermally averaged Gd moment $<\mu>$ along the field direction by the following relation [17]:

$$\Delta\rho(H)=\{\rho(H)-\rho(H=0)\}/\rho(H=0) \propto -<\mu>^2 \qquad (1)$$

Let us assume that the magnetization curve at $T=4.7K$ is similar to the curve at $T=1.7K$(Fig.5). Then we can substitute [17] the experimentally determined $<\mu>$ of Fig.5 into Eq.1 and compare it to the measured $\Delta\rho(H)$. This is shown in Fig.6, where the solid line is the scalling of the $<\mu>^2$ to the $\Delta\rho(H)$ values. Although, the sample is a polycrystal and is contaminated with a minority impurity phase, nevertheless, the agreement between $\Delta\rho(H)$ and $<\mu>$ according to Eq.1 confirms once more that the AFM couplings in samples with these low Gd-concentrations are quite strong.

In Fig.7, we show the characteristic temperatures versus Gd-concentration as obtained from the results of ρ, χ_{ac} and magnetization measurements. As evident, the T_c is linearly related to the concentration of the pair-breaking centers: the $\Delta T_c/T_c$ is fitted successfully to the Abrikosov-Gor'kov relation. On the other hand, the magnetic transition temperatures are linearly related to the Gd-concentration. It is worth emphasizing that the magnetic boundary line crosses the superconductivity boundary line at the same Gd-concentration above which superconductivity disappears. This behavior was also observed in the RNi_2B_2C series [13].

4. DISCUSSION

YNi_2B_2C is a superconductor with $T_c = 15.6K$ [3] while no superconductivity is detected in $GdNi_2B_2C$ down to 1.5K [9,14,15]. In this series, superconductivity degrades with the addition of the paramagnetic centers. As a consequence, some of the destroyed Cooper-pair are available for the mediation of an RKKY interactions among the Gd-moments. For $0.1 < x < 0.25$, this eventually brings about a long range AFM order that coexists with superconductivity. The strength of these interactions is demonstrated by the facts that for $x=0.22$ a relatively strong field ~80 kOe is needed to saturate the magnetic moment and to completely overcome these couplings. The prevailing zero-field magnetic ground state enforced by these coupling is the one which brings about the quench of superconductivity for $x>0.25$. With our present data, we are unable to rule out the possibility that this state is spin-glass-like state.

We now consider the nature of the magnetic ground state for Y-rich samples. Let us recall that for, e.g., the $x=0.22$ solid solution, the Gd-ions are substituting randomly some of the Y-ions. Then, it is not easy to visualize a long range magnetic order with an oscillatory arrangement of the Gd-moments. This excludes the possibility of establishing a helical ground state. Moreover, our structural analysis (Fig.1) excludes a phase segregation in these compounds. Then the randomly-distributed Gd moments may be involved in a non-collinear non-periodic AFM spin structure (see Figs.2,4-5). This structure (in contrast to the conventional collinear AFM ground state) can give rise to a non-canceling exchange field at the Ni-sheets and so quenches the superconductivity. Some faint traces of quasi-reentrant behavior can also be observed for $0.1 <x<0.25$ (see Fig.2) and can be attributed to the interplay between this magnetic state and the superconductivity.

The $GdNi_2B_2C$ is a weakly-anisotropic Heisenberg antiferromagnetic [15]. It orders antiferromagneically at $T = (19.5\pm0.5)$K. Moreover, a magnetic field of 133 kOe is needed to align the spin against the interplanar couplings. Molecular Field Theory predicts that magnetic dilution will weaken these exchange couplings and linearly depress the magnetic transition temperatures. As shown in Fig.7, this is the case here. However, as mentioned above the interplanar RKKY interactions are still strong even at $x=0.22$.

The overall features of this series showed a strong similarity with the magnetic and superconducting features of the pseudo-ternary compounds $(Lu_{1-x},Ho_x)Rh_4B_4$ [18], $(Tm_{1-x},Ho_x)Rh_4B_4$ [19] and $(Er_{1-x},Ho_x)Rh_4B_4$ [20]. Qualitatively similar concentration-dependent superconducting and magnetic features were reported for the solid solutions $(Y_{1-x}Er_x)Ni_2B_2C$ [10], $(Y_{1-x}A_x)Ni_2B_2C$ (A=Sm,Dy) [11] and $(Er_{1-x}Ho_x)Ni_2B_2C$ [11]. In these (pseudo-) quaternary and (pseudo-) ternary compounds [21] the depression of T_c can be scaled to the de Gennes factor and the structure can accommodate quite a large percentage of 4f-magnetic moments without a complete loss of superconductivity.

5. CONCLUSION

The Gd substitution on the non magnetic superconductor YNi_2B_2C introduces pair-breaking centers and consequently leads to a linear depression of T_c with x. The available normal conducting electrons around the Fermi surface will be ready to mediates an RKKY interaction that eventually materialize a long range AFM order. Depending on the Gd-concentration, a quasi-reentrant behavior or a complete loss of superconductivity will result. For higher Gd-concentration, the magnetic ordering temperatures depends linearly on the Gd-concentration.

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REFERENCES

[1] R. Nagarajan, C. Mazumdar, Z. Hossein, S.K. Dhar, K.V. Golpakrishna, L.C. Gupta, C. Godart, B.D. Padalia and R.Vijayaragharan, Phys.Rev.Lett. 72(1994)274.

[2] T. Siegrist, H.W. Zandbergen, R.J. Cava, J.J. Krajewski and W.F.Peck, Nature 367 (1994) 252; H.W. Zandbergen, R.J. Cava, J.J. Krajewski, W.F. Peck, Jr., Physica C224(1. 1994)6.

[3] R.J. Cava, H. Takagi, B. Batlogg, H.W. Zandbergrn, J.J. Krajewski, W. F. Peck, Jr, T. Siegrist, B. Batlog, R. B. van Dover, R. J. Felder, K. Mizuhashi, J.O. Lee, H. Eisaki, and S. Uchida, Nature 367(1994)252.

[4] H. Eisaki, T. Takagi, R.C. Cava, K. Mizuhashi, J.O. Lee, B. Batlogg, J.J. Krajewski, W.F. Peck and S.Uchida, Phys.Rev.B 50(1994)647.

[5] T.E. Grigereit, J.W.Lynn, Q. Huang, A. Santoro, R.J. Cava, J.J. Krajewski and W.F. Peck, Jr, preprint., A.J. Goldman, C. Stassis, P.C. Canfield, J. Zarestky, P. Dervenagas, B.K. Cho, D. C. Johnston and B. Sternlieb, Phys. Rev. B 50(1994)9668.

[6] S.K. Sinha, J.W. Lynn, T.E. Grigreit, Z. Hossain, L.C. Gupta, R. Nagarajan and C. Godart, Phy. Rev. 51(1995) in press.

[7] R. Movshovich, M.F. Hundley, J.D. Thompson, P.C. Canfield, B.K. Cho, A.V. Chubukov, Physica C 227(1994)381.

[8] D.W. Cooke et al preprint; L.P. Le, R.H. Heffner, G.J. Niewenhuys, P.C. Canfiled, B.K. Cho, A.Amato, R. Feyerherm, F.N. Gygax, D. E. Maclaughlin and A. Schenck preprint.

[9] M.El Massalami, S.L. Bud'ko, B. Giordanengo, M.B. Fontes, J.C. Mondragon and E.M. Baggio-Saitovitch. to appear in M^2S proceedings, Grenoble (1994).

[10] H. Michor, W. Perthod, T. Holubar, N.M. Hong and G. Hilscher,to appear in M^2S proceedings, Grenoble (1994).

[11] C.V.Tomy, L.J. Chang, G. Balakrishnan and D.McK. Paul, to appear in M^2S proceedings, Grenoble (1994)

[12] H. Schmidt and H.F. Braun Physica C in press.

[13] M.El Massalami, S. L. Bud'ko, B. Giordanengo and E.M. Baggio-Saitovitch, preprint.

[14] F.M. Mulder, J.V.V.J.Brabers, R.C. Thiel, K.H.J.Buschow,F.R.de Bore, to appear in J. Alloys and Compounds.

[15] B. Giordanengo, M.El Massalami, S.L. Bud'ko, E.M. Baggio-Saitovitch, J. Voirn and A. Sulpice preprint.

[16] H. H. Sample, B. L. Brandt and L. G. Rubin, Rev. Sci. Instrum. 53(1982)1129.

[17] B. Coqblin , The Electronic Structure of Rare-Earth Metal and Alloys: the Magnetic Heavy Rare-Earth (Academic Press, London, 1977).

[18] M.B.Maple, H.C. Hamaker, D.C.Johnston, H.B.Mackay and L.D.Woolf, J. Less Common Met. 62(1978) 251.

[19] C.U. Serge and H.F. Braun, Physics Lett 85A(1981)372

[20] D.C.Johnston, W.A. Fertig, M.B. Maple and B.T.Matthias, Solid State Commun. 26(1978) 141.

[21] S.V. Vonsovsky, Yu.A. Izyumov and E.Z. Kurmaev; Superconducitivity of Transition Metals (Springer series in Solid-state Sciences 27, N.Y., 1982; A.V. Narlikar and S.N. Ekbote, Superconductivity and Superconducting Materials, (South Asian Publisher-New Delhi,1983)

Figures Captions

Fig.1. Room temperature Cu-K_α X-ray diffractograms of the pseudo-quaternary system $(Y_{1-x}Gd_x)Ni_2B_2C$. The inset shows the lattice parameters of the tetragonal unit cell as a function of the Gd concentration.

Fig.2 Representative ρ vs. T curves for $(Y_{1-x}Gd_x)Ni_2B_2C$ compounds.$\rho(T)$ is normalized to the room temperature value

Fig.3. Representative χ_{ac}-T curves for $(Y_{1-x}Gd_x)Ni_2B_2C$. The dotted line emphasize the additional magnetic contribution to the χ_{ac} (see text).

Fig.4 The molar χ_{dc} -T curve for $(Gd_xY_{1-x})Ni_2B_2C$ (x=0.22). The right hand side scale shows the Curie-Weiss behavior for this concentration. Due to impurities some deviation of CW is observed at low-T.

Fig.5 The magnetization isotherm for $(Gd_xY_{1-x})Ni_2B_2C$ (x=0.22) at T=1.7K. The magnetic moment saturates to $7\mu_B$ at 7T. The inset shows the low-field magnetization loop at T=1.7K classifying it as a type-II superconductor.

Fig.6 The longitudinal magnetoresistivity versus field of the polycrystalline x=0.22 sample at 4.7(2)K. The solid line represents the scalling of the square of magnetization of Fig.5 to the experimental data above the H_{c2}.

Fig.7 The concentration dependence of the magnetic and superconducting transition temperatures of $(Y_{1-x}Gd_x)Ni_2B_2C$. The solid line on the superconducting boundary represents the Abrikosov.-Gor'kov relation. The solid line lines on the magnetic boundary shows the linear influence of the magnetic dilution linear on the magnetic transitions (T_1 and T_2) (see Ref.15).

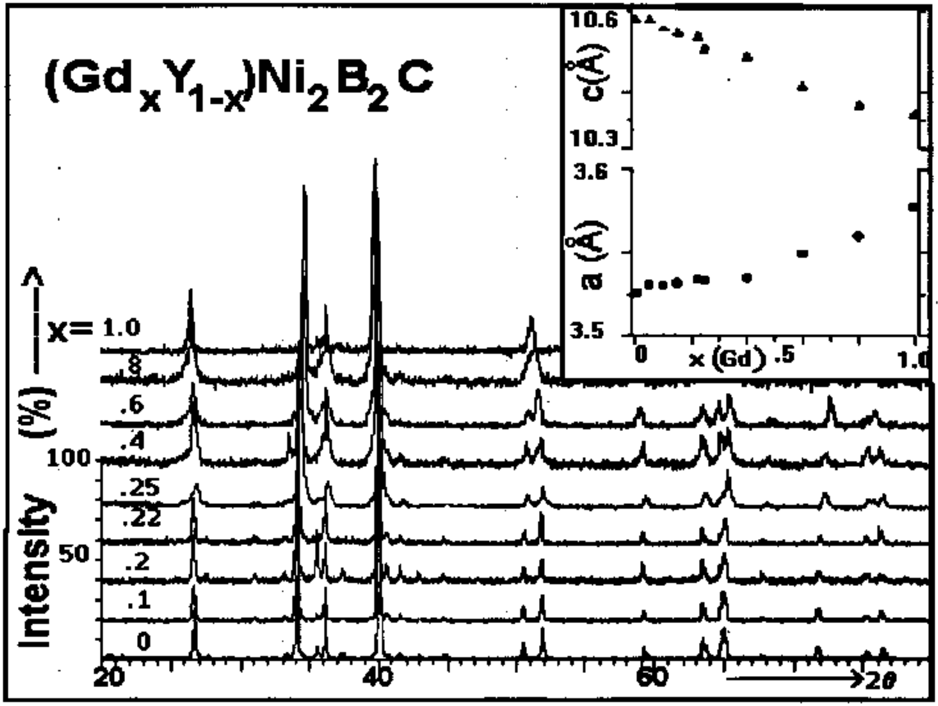

(Gd$_x$Y$_{1-x}$)Ni$_2$B$_2$C
x= 1.0
.8
.6
.4
.25
.22
.2
.1
0
Intensity (%)
100
50
20
40
60
2θ
10.6
c(Å)
10.3
3.6
a (Å)
3.5
0
x (Gd)
.5
1.0

Fig.1.

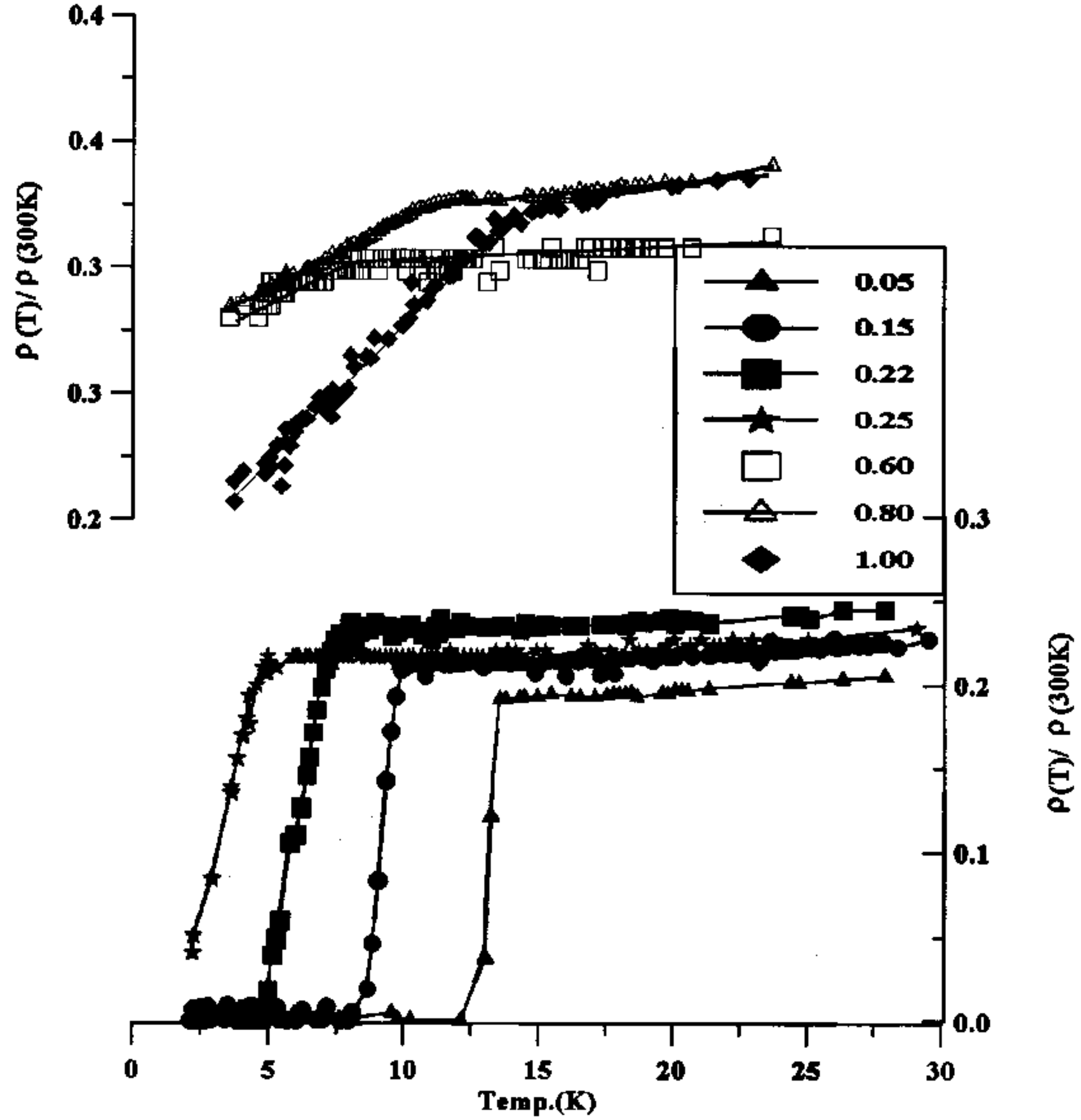

0.4
0.4
0.3
0.3
0.2
ρ (T)/ ρ (300K)
0.05
0.15
0.22
0.25
0.60
0.80
1.00
0.3
0.2
0.1
0.0
ρ (T)/ ρ (300K)
0
5
10
15
20
25
30
Temp.(K)

Fig.2

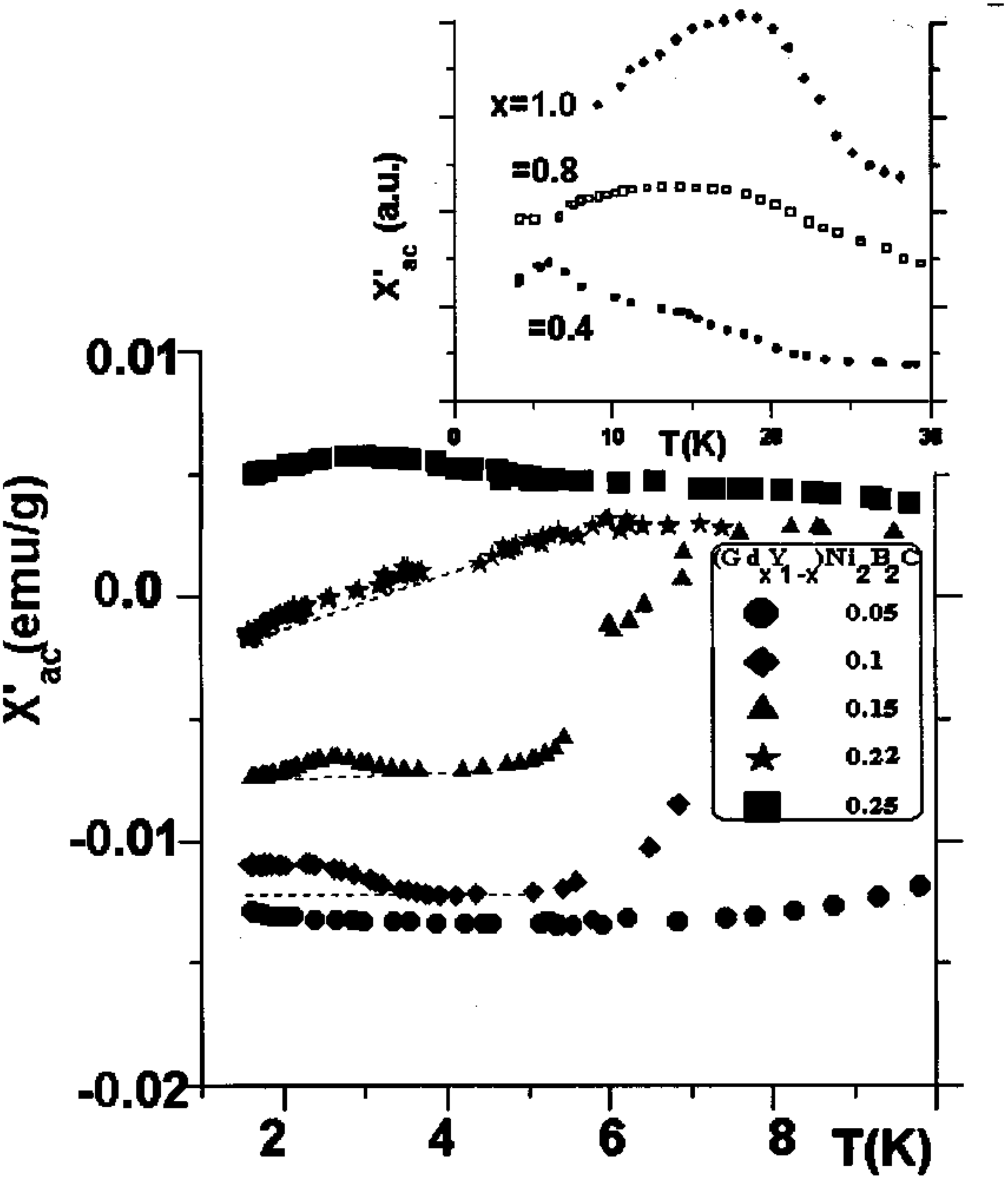

x=1.0
=0.8
=0.4
X'ac (a.u.)
0
10
T(K)
20
30
0.01
0.0
-0.01
-0.02
X'ac(emu/g)
2
4
6
8
T(K)
(GdxY1-x)Ni2B2C
0.05
0.1
0.15
0.22
0.25

Fig.3

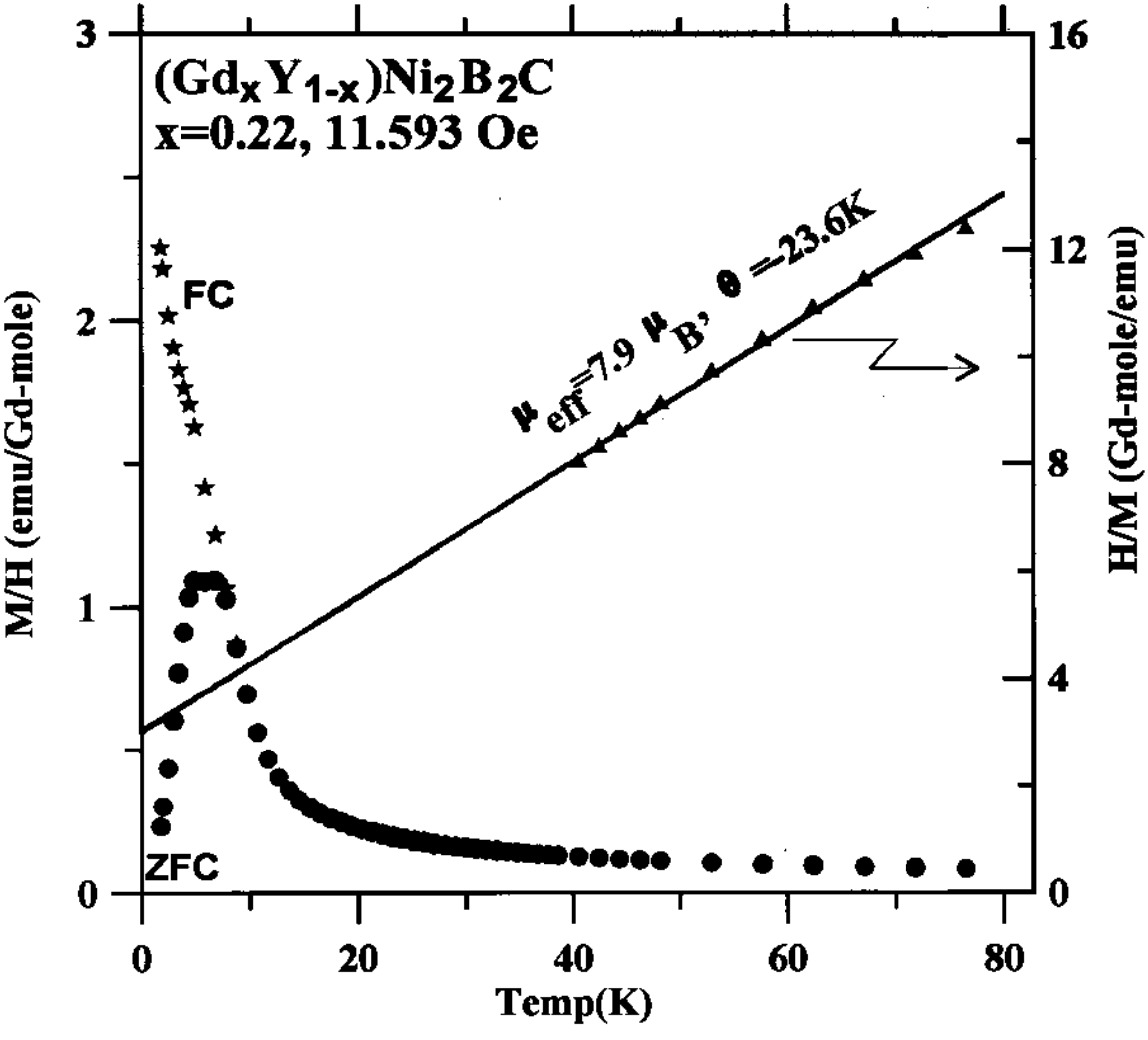
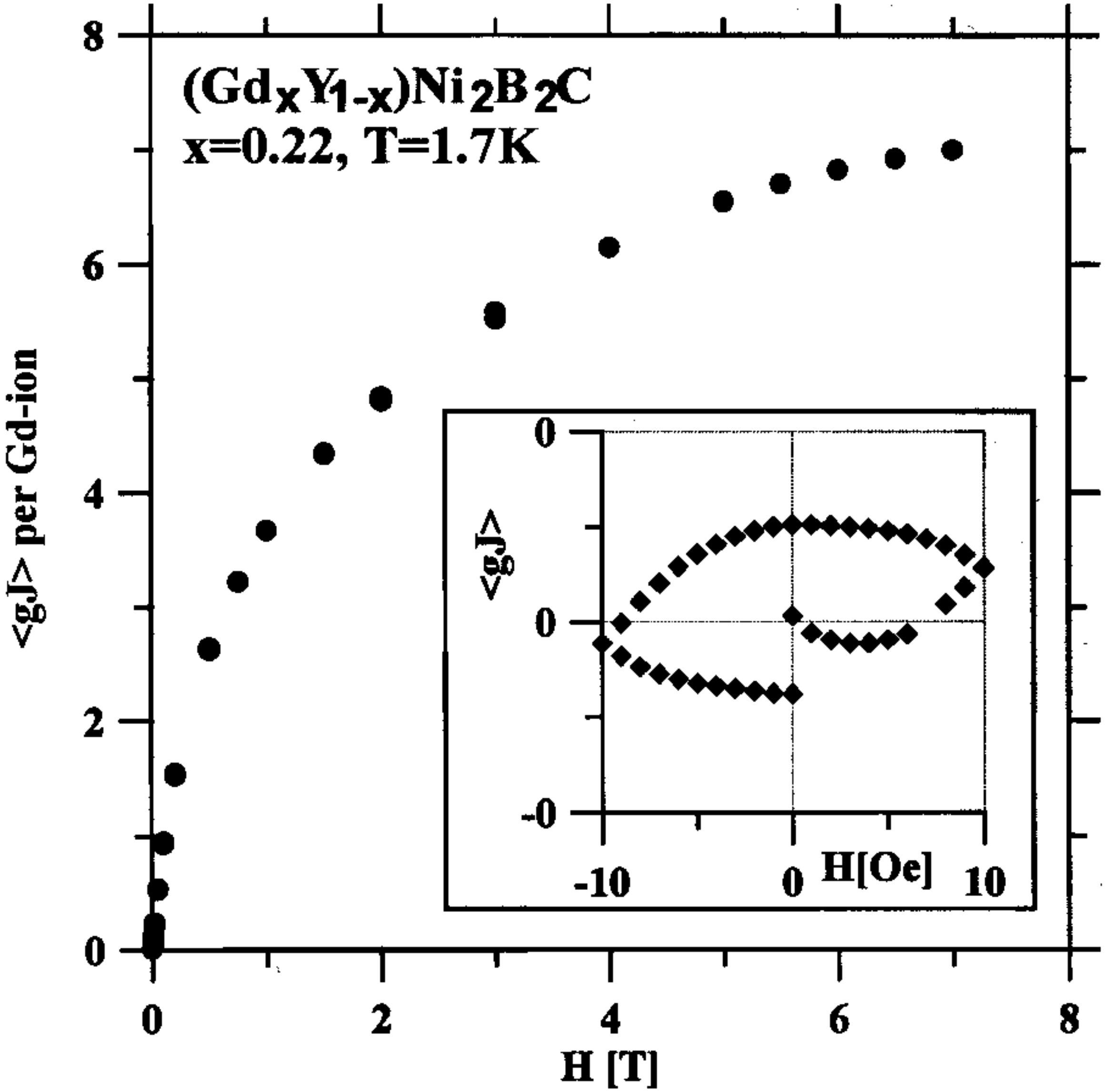

(Gd$_x$Y$_{1-x}$)Ni$_2$B$_2$C
x=0.22, 11.593 Oe
μ_{eff}=7.9 μ_B, θ =-23.6K
FC
ZFC
M/H (emu/Gd-mole)
H/M (Gd-mole/emu)
Temp(K)
0
1
2
3
0
4
8
12
16
0
20
40
60
80

Fig.4



(Gd$_x$Y$_{1-x}$)Ni$_2$B$_2$C
x=0.22, T=1.7K
<gJ> per Gd-ion
H [T]
0
2
4
6
8
0
2
4
6
8
<gJ>
H[Oe]
-10
0
10
0
0
-0

Fig.5

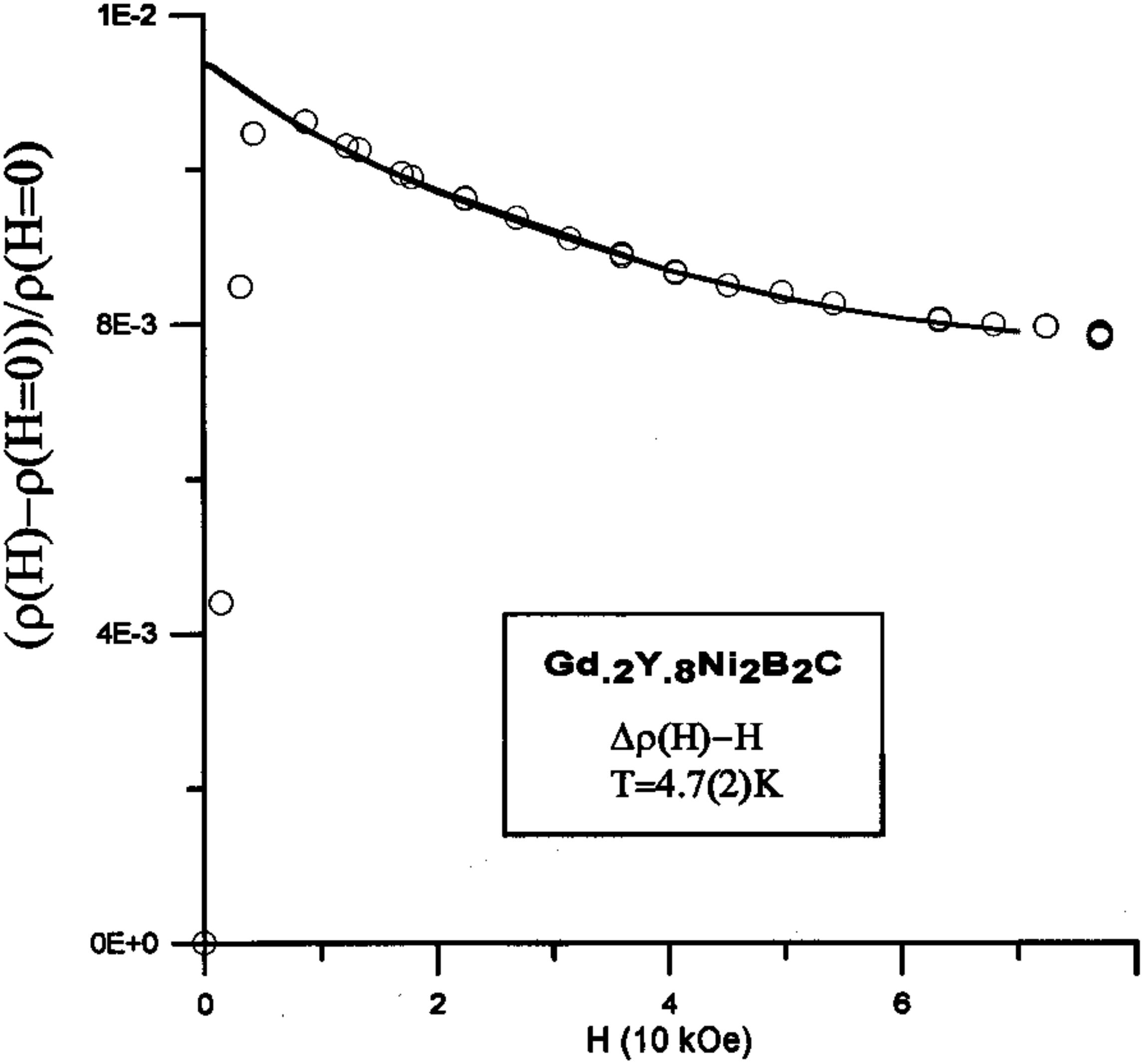

(ρ(H)−ρ(H=0))/ρ(H=0)
1E-2
8E-3
4E-3
0E+0
0
2
4
6
H (10 kOe)
Gd.2Y.8Ni2B2C
Δρ(H)−H
T=4.7(2)K

Fig.6

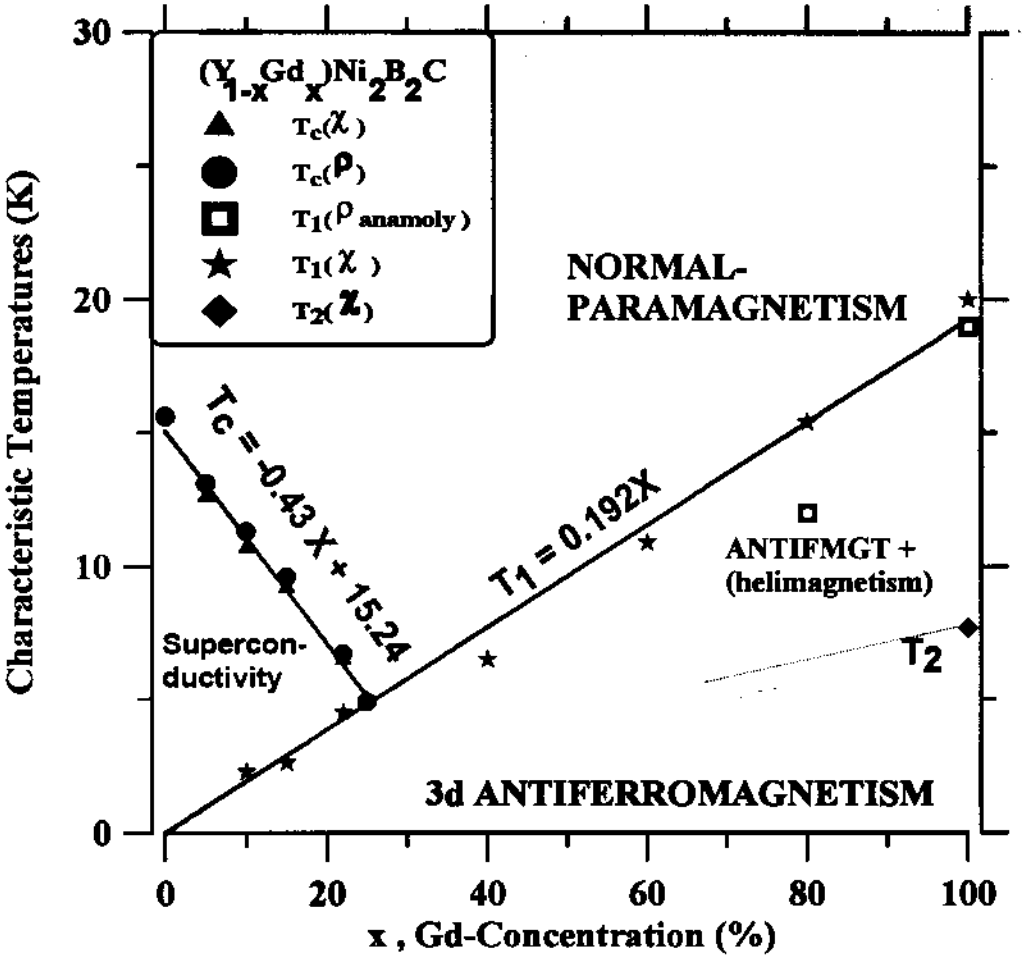

$(Y_{1-x}Gd_x)Ni_2B_2C$
▲ $T_c(\chi)$
● $T_c(\rho)$
□ $T_1(\rho$ anamoly$)$
★ $T_1(\chi)$
◆ $T_2(\chi)$
NORMAL-PARAMAGNETISM
$T_c = -0.43 X + 15.24$
$T_1 = 0.192X$
ANTIFMGT + (helimagnetism)
T_2
Supercon-ductivity
3d ANTIFERROMAGNETISM
Characteristic Temperatures (K)
0
10
20
30
x , Gd-Concentration (%)
0
20
40
60
80
100

Fig.7