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Superconducting and magnetic properties of $(Th_{1-x}Nd_x)Ru_2$ and $(Th_{1-x}La_x)Ru_2$ intermetallic compounds

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The superconducting properties of $(Th_{1-x}La_x)Ru_2$ and the superconducting and magnetic properties of $(Th_{1-x}Nd_x)Ru_2$ are investigated by ac-susceptibility and dc-magnetization measurements as a function of temperature and applied pressure. Both systems exhibit an unusual increase of the superconducting transition temperature T_c for $0 \leq x \leq 0.33$. For the first time we are able to separate the magnetic contribution in $(Th_{1-x}Nd_x)Ru_2$ by subtracting the T_c values of both systems. The difference of the T_c values at the respective concentrations shows an Abrikosov-Gor'kov (AG) type of behavior which we attribute to the weak interaction of the Nd ions with the conducting electrons. The anomalous T_c behavior is also reflected in the concentration dependence of the upper critical field $H_{c2}(0)$. In contrast to T_c and $H_{c2}(0)$, the calculated electron density of states at the Fermi surface decreases with increasing x. With the application of hydrostatic pressure ($p_{max} = 1.2$ GPa) the T_c values in $(Th_{1-x}Nd_x)Ru_2$ show a weak pressure dependence. The reduction of T_c with pressure varies linearly with x for $0 \leq x \leq 0.20$ in accordance with the AG theory. The $(Th_{1-x}La_x)Ru_2$ compounds show no pressure dependence within the experimental accuracy (5 mK) for $0 \leq x \leq 0.3$.

I. INTRODUCTION

During the last few years there has been considerable interest in pseudobinary and ternary intermetallic compounds. In the ternary compounds, superconductivity can coexist with various types of antiferromagnetic order,[1] and in the binary compounds coexistence of superconductivity and spin-glass-like behavior has been observed.[2] For this reason many experimentalists have given their attention to the $(B_{1-x}A_x)Ru_2$ compounds, where BRu_2 represents a superconductor and A, a magnetic rare-earth substituent. Coexistence was found in $(Ce_{0.73}Ho_{0.27})Ru_2$ and in $(Th_{0.65}Nd_{0.35})Ru_2$.[2–4] Outside of the coexistence region at low concentrations of magnetic impurities an anomalous T_c behavior has been observed and attributed to "alloying effects."[5] To investigate the physical origin of these alloying effects, we have studied the superconducting and magnetic properties in more detail by measuring the phase diagram, the upper critical field $H_{c2}(0)$ and the pressure dependence of T_c of the $(Th_{1-x}Nd_x)Ru_2$ and $(Th_{1-x}La_x)Ru_2$ compounds. The alloying effects will be discussed qualitatively in the context of the McMillan theory for strong coupled superconductors[6]

$$T_c = \frac{\Theta_D}{1.45} \exp\left[\frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right], \tag{1}$$

where Θ_D is the Debye temperature, μ^* is a Coulomb repulsion term (usually of magnitude 0.10 to 0.15), and λ is the electron-phonon coupling constant (with $\lambda \approx 1$ for our system). By writing

$$\lambda = \frac{N(0)\langle I^2 \rangle}{m\langle \omega^2 \rangle}, \tag{2}$$

where $N(0)$ is the electron density of states at the Fermi surface, $\langle I^2 \rangle$ the electron-phonon matrix element, M the atomic mass, and $\langle \omega^2 \rangle$ the average of the squared phonon frequency, one can attribute an initial increase in T_c to an increase in $N(0)$, an increase in $\langle I^2 \rangle$, a decrease in M, and/or a decrease in $\langle \omega^2 \rangle$.

II. EXPERIMENT

The superconducting transition temperature T_c is determined by an ac-susceptibility method. Details of the method are described elsewhere.[7] The upper critical field $H_{c2}(T)$ has been measured as a function of temperature by dc magnetization, using a vibrating sample magneto-

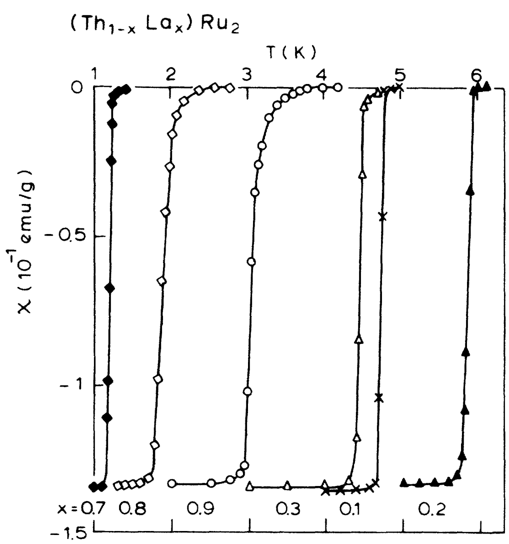

FIG. 1. Superconducting transitions of the various $(Th_{1-x}La_x)Ru_2$ compounds, measured with ac susceptibility in zero field.

meter. For measurements under hydrostatic pressure we used a CuBe pressure clamp where the sample and a superconducting Pb manometer are placed inside a small delrin container filled with an alcohol. The magnetization of the sample is continuously recorded at slowly varying temperatures by means of a commercial superconducting quantum interference device (SQUID).

Both investigated systems crystallize in the $C15$ structure over the whole concentration range. The compounds $ThRu_2$, $NdRu_2$, and $LaRu_2$ have nearly identical lattice constants with a maximal 0.5% difference. The samples were prepared by repeated arc melting in a water-cooled copper crucible under 0.5 atm argon gas, annealed at 900 °C for three days and subsequently quenched into ice water. The sharpness of the superconducting transitions, as shown in Fig. 1, indicates well-ordered intermetallic compounds with a homogeneous distribution of Nd and La over the Th sites.

III. PHASE DIAGRAMS

The superconducting phase diagrams of $(Th_{1-x}A_x)Ru_2$ (A = Nd,La) are shown in Fig. 2. For $0 \le x \le 0.33$, both sets of samples show a comparable T_c behavior: a remarkable increase of T_c for $0 \le x \le 0.20$ and a maximum around $x = 0.20$ followed by a rapid decrease. The Nd compounds show the coexistence of superconductivity and cluster-glass freezing for $0.3 < x < 0.4$. Beyond the percolation limit ($x = 0.42$), long-range ferromagnetism is observed. The T_c values of the La compounds run through a minimum at $x = 0.50$, followed by a monotonic increase of T_c up to $T_c = 4.29$ K for $LaRu_2$ itself. It is observed that the increase of T_c in $(Th_{1-x}La_x)Ru_2$ at low concentrations ($x \le 0.20$), is exactly equal to the increase at high concentrations ($x \ge 0.80$) being 0.12 K/at. % La. We confine ourselves to discuss mainly the Th-rich side of the phase diagram. Here the T_c values of the $(Th_{1-x}La_x)Ru_2$ compounds are systematically larger than those of the $(Th_{1-x}Nd_x)Ru_2$ system. In order to separate the influence on T_c of the magnetic moments in

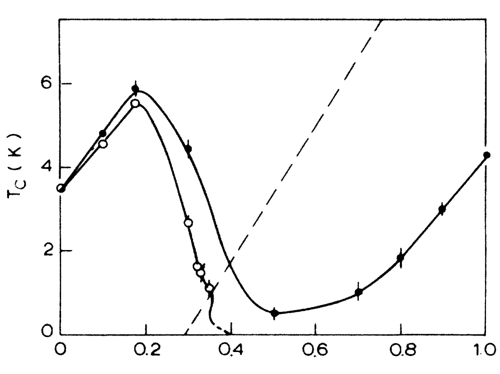

FIG. 2. Superconducting phase diagrams of the $(Th_{1-x}Nd_x)Ru_2$ compounds (open circles) and the $(Th_{1-x}La_x)Ru_2$ compounds (closed circles). For $x > 0.42$ there is long-range ferromagnetic order in the $(Th_{1-x}Nd_x)Ru_2$ compounds (dashed curve). Between $x = 0.33$ and $x = 0.42$ there is coexistence of superconductivity and cluster-glass freezing.

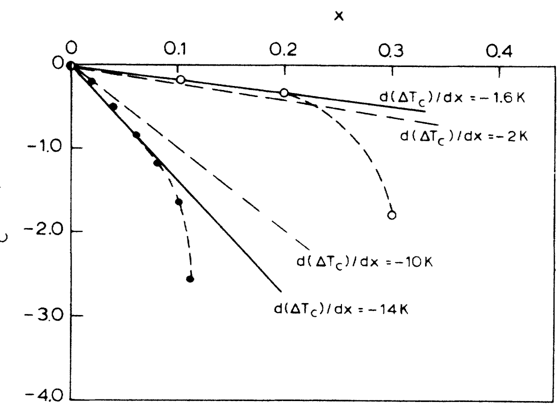

FIG. 3. Plot of $\Delta T_c = T_c(\mathrm{Nd}) - T_c(\mathrm{La})$ versus concentration Nd (open circles) and of $\Delta T_c = T_c(\mathrm{Gd}) - T_c(\mathrm{La})$ versus concentration Gd (closed circles). The dashed curves represent the depression of T_c calculated by Barberis *et al.* (Ref. 8) out of ESR data.

$(Th_{1-x}Nd_x)Ru_2$, we have plotted in Fig. 3 the difference of the T_c values for the respective concentrations, under the assumption that the two systems are otherwise similar enough. For concentrations $0 \le x \le 0.20$, the slope $d(\Delta T_c)/dx$ appears to be -1.6 K, in remarkable agreement with the value of -2 K calculated by Barberis *et al.* from Knight-shift measurements.[8] The initial suppression of T_c at low concentrations of magnetic impurities can be theoretically described by the Abrikosov-Gor'kov (AG) theory[9]

$$\frac{dT_c}{dx} = \frac{-\pi^2}{4k_B} N(0) S(S+1) J^2 , \tag{3}$$

where dT_c/dx is the suppression of T_c by the concentration of magnetic impurities x, k_B is the Boltzmann constant, S is the effective spin ($S = 0.5$ for Nd), $N(0)$ is the density of states at the Fermi surface, and J is the exchange parameter between the conducting electrons and the magnetic moments. Using the observed slope $d(\Delta T_c)/dx$, we can calculate the absolute value for the exchange parameter $|J|$ to be 5.5 meV, in agreement with the observations of Barberis *et al.*[8] From their ESR measurements we know the exchange parameter J of Nd in $ThRu_2$ to be negative. We can apply the same subtraction to the system $(Th_{1-x}Gd_x)Ru_2$ to separate the magnetic contribution of Gd. Using the T_c values given in Ref. 10, we can easily calculate $\Delta T_c = T_c(\mathrm{Gd}) - T_c(\mathrm{La})$. The result is plotted in Fig. 3. Here we find the decrease of ΔT_c having the slope of -14 K, which is again close to the value estimated by ESR measurements ($dT_c/dx = -10$ K).[8]

IV. UPPER CRITICAL FIELD

The density of states $N(0)$ at the Fermi surface usually plays an important role in determining the superconducting transition temperature. Critical field measurements offer a possibility to deduce the density of states since the thermodynamical critical field $H_c(0)$ and $N(0)$ are coupled via the following BCS relation:[11]

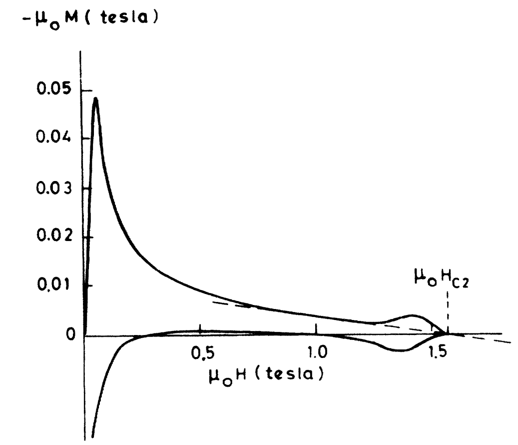

FIG. 4. Typical magnetization curve for the $(Th_{1-x}La_x)Ru_2$ compounds. Note the strong hysteresis and the peak effect near H_{c2}. The dashed line is used to determine H_{c2}.

$$\mu_0 H_c^2(0) = N(0)(1.76 k_B T_c)^2 . \quad (4)$$

The thermodynamical critical field $H_c(0)$ can be calculated using

$$-\left.\frac{dH_c}{dT}\right|_{T_c} = 1.73\frac{H_c(0)}{T_c} , \quad (5)$$

$$\left[\left.\frac{dH_{c2}}{dT} \Big/ \frac{dH_c}{dT}\right]\right|_{T_c} = \sqrt{2}\kappa_{GL} , \quad (6)$$

where κ_{GL} is the Ginzburg-Landau parameter. We have measured the upper critical field $H_{c2}(T)$ of $(Th_{1-x}La_x)Ru_2$ as a function of temperature for concentrations $0 \le x \le 0.3$ and $0.8 \le x \le 1$, and of $(Th_{1-x}Nd_x)Ru_2$ for $0 \le x \le 0.33$. A typical magnetization curve is shown in Fig. 4. With increasing field the negative magnetization increases, passes a maximum, slowly decreases, and reaches zero close to H_{c2}. The curves are not fully reversible and show strong hysteresis effects. In the case of $LaRu_2$, a peak effect occurs near H_{c2} indicating the flux-pinning behavior, probably caused by grain boundaries in the polycrystalline samples serving as pinning centers for the magnetic flux lines. Just above H_{c2} we find a constant slope, and by extrapolating it to zero magnetization we find $H_{c2}(T)$. The resulting $H_{c2}(T)$ values are plotted as a function of the reduced temperature $t = T/T_c$ in Figs. 5 and 6. The theory of Werthamer *et al.*[12] (WHH), developed for the calculation of the upper critical field in type-II superconductors,[12] can be used to extrapolate the measured values to the temperature range not accessible to our experimental equipment (solid curves in Fig. 5). The experimental data for

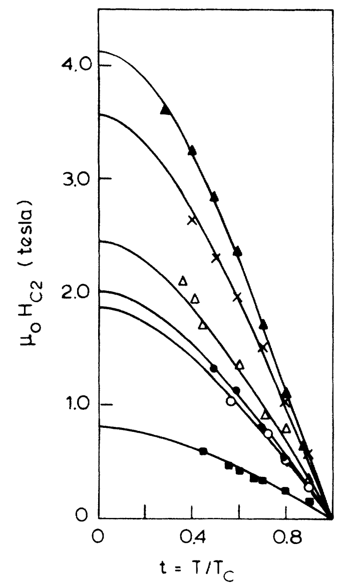

FIG. 5. The upper critical field H_{c2} for the various La compounds as a function of the reduced temperature $t = T/T_c$. (Squares, $x = 0$; crosses, $x = 0.1$; closed triangles, $x = 0.2$; open triangles $x = 0.3$; open circles, $x = 0.9$; and closed circles, $x = 1$.) The lines are extrapolations using the WHH theory for type-II superconductors.

TABLE I. Superconducting transition temperature T_c, calculated values of the Ginzburg-Landau parameter κ_{GL}, the thermodynamical critical field $\mu_0 H_c(0)$, the electron density of states $N(0)$, and the Sommerfeld parameter γ for the $(Th_{1-x}A_x)Ru_2$ samples (A = Nd or La).

x	T_c (K)	κ_{GL}	$\mu_0 H_c(0)$ (T)	$N(0)$ (states/eV atom)	γ (mJ/atom K^2)
			$(Th_{1-x}La_x)Ru_2$		
0	3.47	1.2	0.350	2.6 (2.2[a])	14.6
0.10	4.73	7.0	0.290	0.9	4.0
0.20	5.87	7.0	0.338	0.8	3.6
0.30	4.45	5.6	0.244	0.7	3.1
0.90	3.05	4.0	0.284	2.1	9.0
1.00	4.30	2.8	0.402	2.3 (2.0[a])	10.9 (13.85[b])
			$(Th_{1-x}Nd_x)Ru_2$		
0	3.47	1.2	0.350	2.6	14.6
0.20	4.55	8.9	0.206	0.5	2.4
0.33	1.51	2.5	0.081	0.8	3.1

[a]Values have been taken from Ref. 19.
[b]Value has been taken from Ref. 20.

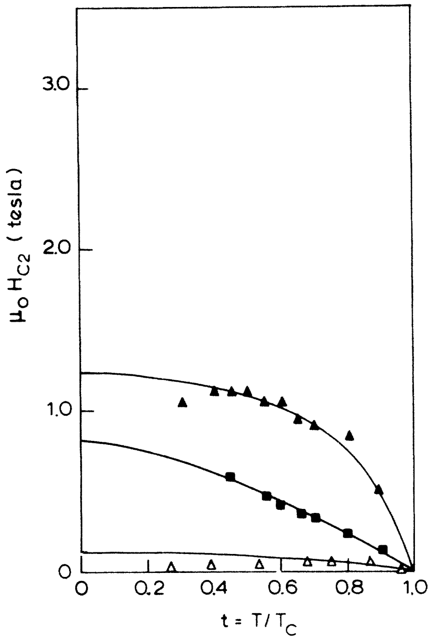

FIG. 6. The upper critical field H_{c2} for the various Nd compounds as a function of the reduced temperature $t=T/T_c$. (Squares, $x=0$; closed triangles, $x=0.2$; and open triangles, $x=0.33$.) The lines are extrapolations using Eq. 3.56 in Ref. 13.

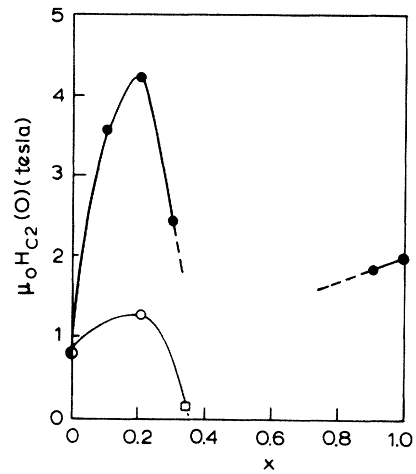

FIG. 7. The upper critical field $H_{c2}(0)$ of $(Th_{1-x}Nd_x)Ru_2$ (open circles) and $(Th_{1-x}La_x)Ru_2$ (closed circles) as a function of concentration. $H_{c2}(0)$ for Nd at $x=0.33$ has been obtained by resistance measurements (open square). All other points are obtained by magnetization measurements.

$(Th_{1-x}La_x)Ru_2$ show a monotonic increase of $H_{c2}(T)$ with decreasing t and are in good agreement with the WHH theory. However, in $(Th_{1-x}Nd_x)Ru_2$, paramagnetic impurities are present, which reduces the upper critical field H_{c2} by an amount proportional to the square of the magnetization. In a magnetically ordered system this fact becomes very important and is one reason for the destruction of superconductivity by ferromagnetic order. We now calculate the upper critical field $H_{c2}(T)$ within the framework of the Ginzburg-Landau theory using Eq. 3.56 in Ref. 13. Owing to the similarity of $(Th_{1-x}La_x)Ru_2$ and $(Th_{1-x}Nd_x)Ru_2$, we use the respective La compounds as nonmagnetic references. For both Nd samples we find good agreement of the data and the calculation, at least close to T_c. The downward deviation for $T \ll T_c$ can be explained by short-range interactions between the magnetic impurities.[2] For both systems investigated, $H_{c2}(0)$ is plotted as a function of concentration in Fig. 7. The overall concentration dependence of $H_{c2}(0)$ reflects the superconducting phase diagram given in Fig. 2; even here the upper critical fields of the $(Th_{1-x}Nd_x)Ru_2$ compounds are systematically lower than the $H_{c2}(0)$ values for $(Th_{1-x}La_x)Ru_2$ compounds at the respective concentrations. Furthermore, we can determine the Ginzburg-Landau parameter κ_{GL} by the magnetization measurements using the following expression:

$$\left.\frac{dM}{dH}\right|_{H=H_{c2}} = \frac{1}{1.16(2\kappa_2^2-1)} , \tag{7}$$

where κ_2 is the second Maki parameter. Since the three Maki parameters κ_1, κ_2, and κ_3 and the Ginzburg-Landau parameter κ_{GL} become identical for $T \rightarrow T_c$, we have extrapolated the obtained κ_2 values to T_c.[14] The thermodynamical critical-field values $H_c(0)$ have been determined from Eqs. (5), (6), and (7). By means of Eq. (4) we can finally calculate the density of states at the Fermi surface $N(0)$, which can further be used to calculate the specific-heat coefficient γ. All these derived quantities κ_{GL}, $H_c(0)$, $N(0)$, and γ are listed in Table I for different concentrations and samples investigated. The $N(0)$ and γ values for $ThRu_2$ and $LaRu_2$ agree nicely with values found by Davidov *et al.*[15] and Joseph *et al.*[16] Surprisingly, the electron density of states at the Fermi surface decreases with increasing La and Nd concentration, indicating that another mechanism must be responsible for the initial increase of T_c and $H_{c2}(0)$, such as an enhancement of the electron-phonon coupling and/or a shift to lower phonon frequencies.

V. PRESSURE DEPENDENCE OF T_c

The high-pressure technique is useful to generate small but well-controlled changes in the solid-state properties. At present there exists no microscopic theory of the pressure dependence of T_c. Therefore it is difficult to arrive at a complete understanding of all the parameters involved. The pressure dependence of T_c can be studied in the framework of the McMillan formalism to describe the pressure dependence of Θ (Ref. 17), μ^*, and λ (Refs. 18–20) and, when magnetic impurities are present, within the framework of the AG theory.[21] The samples $(Th_{1-x}La_x)Ru_2$ showed for $0 \le x \le 0.33$ no pressure dependence at all within the experimental accuracy (5 mK). We would like to note here that $LaRu_2$ itself is pressure dependent with $dT_c/dP = -0.11$ K/GPa. We must assume now the physical quantities determining T_c to be almost independent of pressure, because it is improbable that two such quantities can compensate each other over the complete concentration and pressure range which has been investigated. In Fig. 8 the pressure dependence of T_c of $(Th_{1-x}Nd_x)Ru_2$ is plotted as a function of pres-

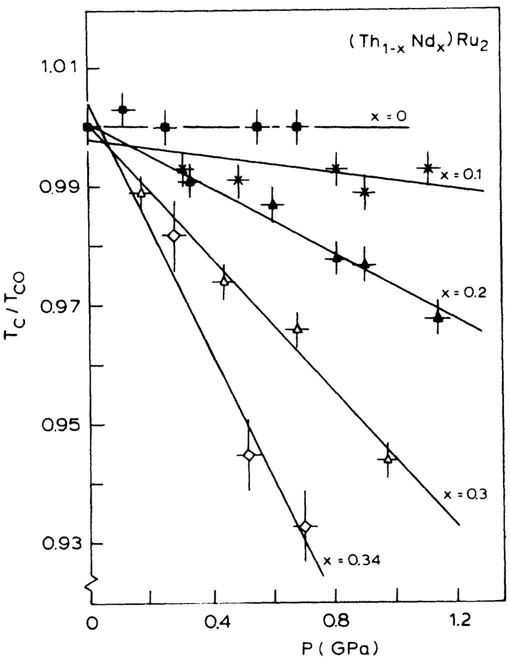

FIG. 8. Plot of the reduced critical temperature $t = T_c/T_{c0}$ for $(Th_{1-x}Nd_x)Ru_2$ versus pressure. The pressure dependence increases with the Nd concentration. T_{c0} is the critical temperature at zero pressure. (Squares, $x=0$; crosses, $x=0.1$; closed triangles, $x=0.2$; open triangles, $x=0.3$; and diamonds $x=0.34$.)

sure (for $0 \le x \le 0.33$), and in Fig. 9 dT_c/dP is plotted as a function of the Nd concentration. The pressure dependence of $(Th_{1-x}Nd_x)Ru_2$ shows an almost linear increase with the concentration of magnetic impurities (Nd) for $0 \le x \le 0.20$ and distinctly deviates from this behavior for higher concentrations.

Restricting ourselves to low concentrations (linear ap-

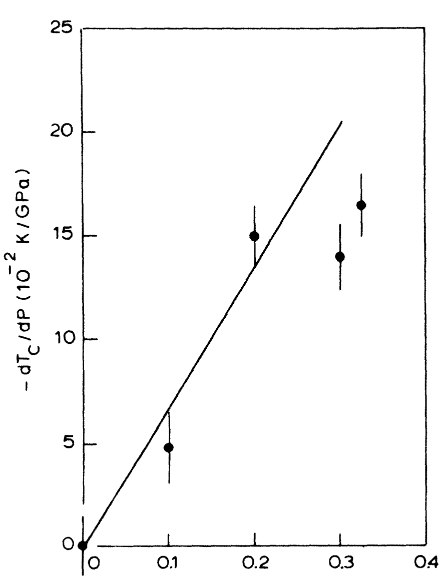

FIG. 9. Pressure dependence of the $(Th_{1-x}Nd_x)Ru_2$ compounds as a function of the Nd concentration. At low concentrations ($x \le 0.2$) a linear increase with the Nd concentration is observed, followed by a deviation at higher concentrations.

proximation of the AG equation) we find, following Smith,[22] by differentiating Eq. (3) with respect to pressure and neglecting the pressure dependence of $N(0)$,

$$\frac{dT_c}{dP} = -x\frac{\pi^2}{2k_B}S(S+1)N(0)J\frac{dJ}{dP} , \quad (8)$$

where x is the concentration of magnetic impurities. This linear dependence of dT_c/dP is in agreement with the experimental results for $x < 0.20$, see Fig. 9. Using the J value found in Sec. III, we can compute $dJ/dP = -0.7$ meV/GPa.

An expression for the pressure dependence of T_c for concentrations up to the critical concentration has been derived by Maple and Kim.[23] Following their paper closely, we may write according to Abrikosov and Gor'kov[9]

$$\ln(T_c/T_{c0}) = \psi(\tfrac{1}{2}) - \psi(\tfrac{1}{2} + 0.140\alpha T_{c0}/\alpha_{cr}T_c) , \quad (9)$$

where T_{c0} corresponds to $\alpha = 0$ and α_{cr} to $T_c = 0$, and ψ is the digamma function. From Eq. (9) it follows that

$$t \equiv T_c/T_{c0} = f(\alpha(x,P)/\alpha_{cr}(P)) , \quad (10)$$

where $\alpha_{cr}(P) = \pi T_{c0}(P)/2\gamma$ ($\ln\gamma$ is Euler's constant). Differentiating Eq. (10) with respect to P yields

$$\left|\frac{dt}{dP}\right|_{P=0} = C[f'(\alpha/\alpha_{cr})(\alpha/\alpha_{cr})]_{P=0} , \quad (11)$$

where

$$C \equiv \left[\frac{\partial}{\partial p}\ln\left[\frac{N(0)J^2S(S+1)}{T_{c0}}\right]\right] .$$

Assuming C to be not dependent on the concentration, Eq. (11) indicates, since α_{cr} is proportional to T_{c0}, a linear increase of $|dT_c/dP|$ at low concentrations, followed by a more rapid increase as α, and thus x, approach their critical value. However, from our experiments we find a deviation of the linear behavior of dT_c/dP in the opposite direction. This disagreement is probably caused by magnetic interactions between the Nd ions.

VI. DISCUSSION

In the preceding sections we have focused our attention on the "alloying effects" in the $(Th_{1-x}A_x)Ru_2$ (A = Nd,La) intermetallic compounds. The unusual T_c behavior with an increasing number of magnetic impurities had previously been observed, but no clear explanation has been given. The similarity of the $(Th_{1-x}Nd_x)Ru_2$ system and the $(Th_{1-x}La_x)Ru_2$ system made it possible to demonstrate the influence of magnetism on the superconductivity by subtracting the T_c values of the La system from those of the Nd system (Figs. 2 and 3). We found a suppression of the differential superconducting transition temperature $d(\Delta T_c)/dx = -1.6$ K for Nd and $d(\Delta T_c)/dx = -14$ K for Gd, in agreement with the values estimated by Barberis *et al.*[8]

Substitution of Th by the much lighter rare-earth elements Nd and La will result in an increase of T_c due to an "isotope effect" [M in Eq. (2)]. But this effect alone cannot account for the unusual sharp increase of T_c for $0 \leq x \leq 0.20$ and for $0.8 \leq x \leq 1$ of $dT_c/dx = +0.12$ K/at. % La. The presence of an appreciable lattice pressure is in both systems very unlikely. The lattice parameters of $ThRu_2$, $LaRu_2$, and $NdRu_2$ differ less than 0.5%, and the T_c of these compounds show no pressure dependence at all (La compounds) or a very small pressure dependence (Nd compounds).

With critical-field measurements we tried to determine the influence of the density of states at the Fermi surface $N(0)$ on the superconducting transition temperature T_c. The concentration dependence of $N(0)$ is in contradiction with the concentration dependence of T_c and H_{c2}. This indicates that some other mechanism is responsible for the unusual T_c behavior. A phonon softening or an enhancement of the electron-phonon coupling induced by the disorder of alloys could possibly explain the increase in T_c. However, these effects can only be verified by either ultrasound attenuation experiments or point-contact spectroscopy.

No pressure dependence of T_c could be observed in $(Th_{1-x}La_x)Ru_2$ for $0 \leq x \leq 0.3$. This is rather surprising because the system is believed to be a d-band superconductor and one should expect a change in T_c with increasing pressure. On the basis of the pressure experiments, we suggest that the Fermi level of these compounds is located mainly in the s band below the d-band peak or in a minimum of the d band, since in these cases $N(0)$ will be only weakly dependent on pressure.[24]

Due to the similarity, except magnetically, of La and Nd, and their AG-type behavior of ΔT_c, we further conclude that the observed pressure dependence of T_c in $(Th_{1-x}Nd_x)Ru_2$ is of magnetic origin. For small concentrations it is approximately linear and we can calculate directly the value for the pressure dependence of the exchange parameter J to be $dJ/dP \approx -0.7$ meV/GPa. This result is consistent with the observation that in systems with negative exchange parameters, J universally is found to increase in magnitude with pressure.[21]

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