3. Experiments

The $La_{1-x}Tb_xAl_2$ samples have been prepared from 4*N* pure La, 3*N* pure Tb (both supplied from Rare Earth Products Limited) and 5*N* pure Al by induction melting the constituents in a water cooled copper crucible in a high purity argon atmosphere. First, $LaAl_2$ and $TbAl_2$ were fabricated. Then a master alloy containing 10% Tb was prepared, portions of which were diluted with $LaAl_2$ in two further steps to get the final concentrations. Each alloy was melted 5 times. The weight loss, controlled after each melting process, was negligible. The samples were heat treated in vacuum of 10^{-7} torr at 850 °C for 16 hours. The Gd alloy was prepared in the same way.

The experiments were carried out with the usual fluid-cell method, the samples and a coil for an inductive measurement of T_c being immersed in the fluid [16]. The given values of T_c are defined by the mid-point of the transition curve. The corresponding transition widths were 40, 22, 180 and 46 mK for the samples $LaAl_2$, $La_{0.998}Tb_{0.002}Al_2$, $La_{0.994}Tb_{0.006}Al_2$ and $La_{0.998}Gd_{0.002}Al_2$, respectively. The pressure was determined from the transition temperature of tin using the relation between T_c and p given in Ref. 17. All samples were placed within the same pressure cell and measured in the same run. Approximately 10 mg were used of each sample.

Experimental values of T_c versus pressure are shown in Fig. 1 for all samples. They have been fitted with parabolic curves. Maximum deviations of the experimental points from the fit curve amount to 5 mK. The initial slope of $LaAl_2$ is 179 mK/kbar.

In Fig. 2a the difference $T_c(p)-T_c(0)$ is plotted with the values taken from the fit. In this plot the different pressure dependences of the transition temperatures of the Tb and Gd-alloys are demonstrated. This becomes clearer in Fig. 2b where the relative change of the pairbreaking effect of a single impurity is plotted on the same pressure scale.

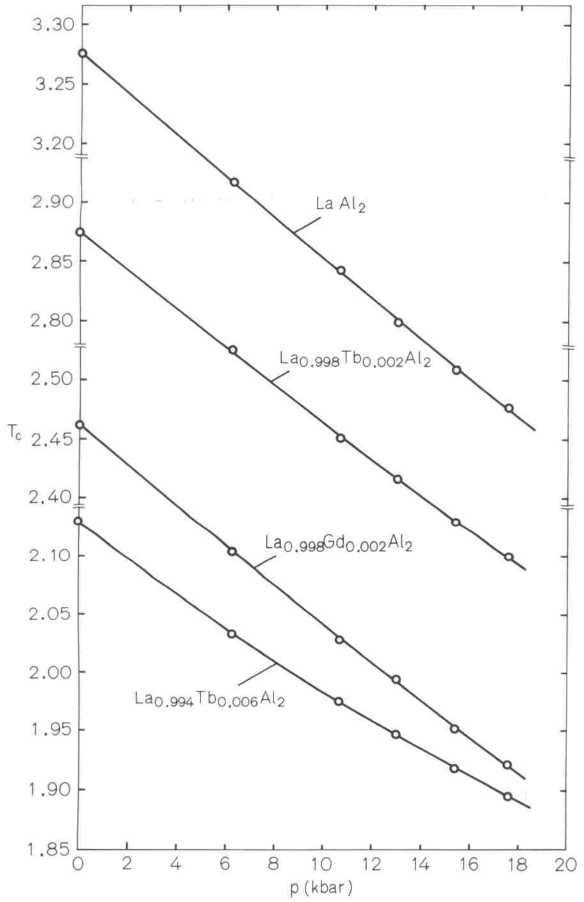

Fig. 1. The transition temperatures T_c of pure $LaAl_2$ and of the alloys <u>La</u>$TbAl_2$ and <u>La</u>$GdAl_2$ as function of the hydrostatic pressure *p*

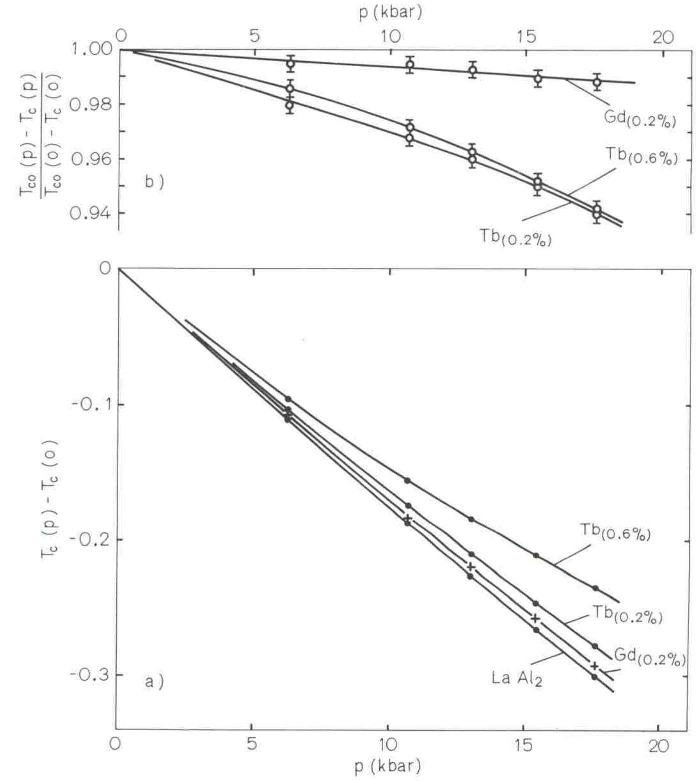

Fig. 2. a) The differences between the transition temperatures under pressure and at zero pressure, $T_c(p)-T_c(0)$, of $LaAl_2$ and the alloys with La substituted by Tb and Gd. b) The relative change of the pair-breaking effect of the Tb- and Gd-impurities $\dfrac{T_c(p)-T_{c0}(p)}{T_c(0)-T_{c0}(0)}$ versus pressure, T_c and T_{c0} denoting the transition temperatures of the magnetic alloys and of pure $LaAl_2$, respectively

Within the uncertainties, the corresponding values of the two Tb alloys coincide and are essentially larger than the small effect shown by the Gd alloy.

It must be noted that the effect of pressure on the pair breaking effect of Gd impurities was not detected in earlier experiments [15]. It is supposed, that in those experiments the superconducting transition of the alloys did not permit a separation of this small effect.

4. Analysis of Experimental Results

The following analysis of the pressure dependence of T_c is based on a theoretical investigation of the dependence of T_c on the level scheme and the concentration of magnetic impurities outlined in Ref. 5, combined with experimental results on the concentration dependence of T_c [2]. The transition temperature T_c of an alloy containing impurities with crystalline field split energy levels can be written quite generally:

$$T_c/T_{c0} = F(Z, Y, X_L) \tag{8}$$

T_{c0} is the transition temperature of the pure sample. $Y = \delta/T_{c0}$, where δ is the energy separation of the two lowest levels (provided that the transition is allowed) and is used as a scale of the whole level scheme. The explicit dependence of F on X_L is used to fix the position of the higher energy levels and the transition matrix elements. $Z = (\tau \cdot T_{c0})^{-1} \cdot f$ is proportional to the impurity concentration, contained in the scattering rate τ^{-1}. It is normalized through the function $f(Y, X_L)$ in such a way that $dT_c/dZ = -1$ for $T_c = T_{c0}$.

If we denote the change of T_c under pressure by $\Delta T_c = T_c(p) - T_c(0)$ and use similar abbreviations for the other pressure dependent quantities, the change of the difference between the transition temperature of the alloy and the pure sample can be written as

$$\Delta T_c - \Delta T_{c0} = (F-1)\Delta T_{c0} + T_{c0}\Delta F. \tag{9}$$

Using the explicit and implicit dependence of F on Z, Y and X_L this quantity can be decomposed into four contributions

$$\begin{aligned}\Delta T_c - \Delta T_{c0} &= \left(F - 1 - Z\frac{\partial F}{\partial Z}\right)\Delta T_{c0}\\ &+ T_{c0} Z \frac{\partial F}{\partial Z}\tau\Delta(1/\tau)\\ &+ T_{c0}\left(Z\frac{\partial F}{\partial Z}\cdot\frac{1}{f}\cdot\frac{\partial f}{\partial Y} + \frac{\partial F}{\partial Y}\right)\Delta Y\\ &+ T_{c0}\left(Z\frac{\partial F}{\partial Z}\cdot\frac{1}{f}\cdot\frac{\partial f}{\partial X_L} + \frac{\partial F}{\partial X_L}\right)\Delta X_L.\end{aligned} \tag{10}$$

The last two terms of the r.h.s. of Eq. (10) contain the influence of pressure on the level system of the magnetic impurities. The derivatives of f and F have to be calculated numerically; they correspond to the change of the initial slope and the curvature of T_c vs x, respectively, for a variation of the crystalline field parameters. The variation of $Y = \delta/T_{c0}$ is given by

$$\Delta Y = Y(\Delta\delta/\delta - \Delta T_{c0}/T_{c0}). \tag{11}$$

The variation of δ can be calculated from Eq. (6) using the results of LLW and the compressibility of $LaAl_2$, given in Ref. 13. As ΔT_{c0} is negative for $LaAl_2$, the total effect of pressure on the pairbreaking effect of magnetic impurities in $LaAl_2$ is enhanced. The contribution of the term proportional to ΔX_L is small in most cases. However, the dependence of δ on X_L (Eq. (6)) is important near crossing points of the two lowest energy levels.

The first term of Eq. (10) is due to a change of the scale of the pairbreaking effect of magnetic impurities for a variation of T_{c0}. It is related to the deviation from linearity of T_c vs x. It can be calculated theoretically for a given sequence of crystalline field levels. But according to the relation

$$F - 1 - Z\frac{\partial F}{\partial Z} = T_c/T_{c0} - 1 - x\frac{\partial(T_c/T_{c0})}{\partial x} \tag{12}$$

this terms can *also* be determined completely from experiments on the concentration dependence of T_c.

The term proportional to $\Delta(1/\tau)$ can be determined from the experiments with <u>La</u>GdAl$_2$ in the following way: As the crystal field has no influence on the magnetic states of Gd ions the pressure dependence is given by the first two terms of Eq. (10). Using the known values of T_c vs x and the experimental result on the pressure dependence of T_c we can evaluate $\tau \cdot \Delta(1/\tau)$. Assuming that the relative change of the scattering rate under pressure is equal for Gd and Tb ions imbedded in the same host material we can use this value in our analysis of Tb alloys.

We concentrate our analysis of the pressure dependence of T_c on the samples

$La_{0.998}Gd_{0.002}Al_2$ and $La_{0.994}Tb_{0.006}Al_2$.

The experimental data for these samples are (variations due to pressure are given for $p = 10$ kbar):

<u>$LaAl_2$</u>

$T_{c0} = 3.276$ K, $\Delta T_{c0} = -0.174$ K,

<u>$La_{0.998}Gd_{0.002}Al_2$</u>

$T_c = 2.451$ K, $Z = 0.24$, $\Delta T_c = -0.170$ K,

$$\left(T_c/T_{c0} - x\frac{\partial(T_c/T_{c0})}{\partial x}\right)\cdot\Delta T_{c0} = -1\,\text{mK}.$$