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Crystalline Field Effects in Superconducting $La_{1-x}Tb_xAl_2$ under Pressure

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Abstract. The pressure dependence of the superconducting transition temperature of $LaAl_2$ and alloys with Tb and Gd impurities has been measured. The results are compared with a theoretical calculation of the pressure dependence of T_c, which is based on the variation by pressure of the crystalline field experienced by the Tb ions. Measurements on $LaGdAl_2$ where crystal field effects are absent have been used to study the influence of pressure on the scattering rate.

1. Introduction

In the presence of a crystalline field the $(2J+1)$-fold degenerate groundstate of a rare earth impurity with angular momentum J splits into a sequence of crystalline field levels. This has important consequences on the properties of superconductors containing rare earth impurities, expecially in the case when the groundstate in the presence of crystalline field is nonmagnetic. The influence of crystalline fields has been demonstrated by experiments on the depression of the superconducting transition temperature as function of the impurity concentration [1, 2], the jump in the specific heat at the phase transition [3], and the upper critical magnetic field [2]. These experiments can be interpreted by a theory which takes into account the pairbreaking effect of inelastic scattering of conduction electrons on the magnetic impurities [4–7].

In this paper we present measurements and a theoretical analysis of the pressure dependence of the superconducting transition temperature T_c of

$La_{1-x}Tb_xAl_2$ and $La_{1-x}Gd_xAl_2$.

The dependence of T_c on the impurity concentration x measured previously [2] indicates that the groundstate of Tb^{3+} ions is nonmagnetic and separated from the first excited magnetic state by the energy $\delta = 5-7$ K. These results are supported by measurements of the susceptibility [8], the Schottky anomaly in the specific heat [9] and the thermoelectric power [10]. The alloy $La_{1-x}Gd_xAl_2$ does not show any crystal field effects because of the vanishing orbital momentum of Gd^{3+} ions. This is demonstrated by the good agreement of the measurements [11] T_c versus x with the theory of Abrikosov and Gorkov [12].

By applying pressure, the strength of the crystalline field is changed because of its dependence on the lattice parameter. The resulting change of the level splitting leads to a variation of the pairbreaking effect and hence to a change in T_c. This effect has been measured recently on $La_{1-x}Tb_xAl_2$ by Guertin *et al.* [13]. Our experiments on $La_{1-x}Tb_xAl_2$ give similar results.

We compare our experimental results with a theoretical calculation of the pressure dependence of T_c using the complete level scheme of Tb ions as calculated by Lea, Leask and Wolf (LLW) [14] for different values of the crystalline field parameters. Though it is not possible to determine the crystalline field parameters from these measurements uniquely, we can show that the pressure dependence of T_c is consistent with the depression of T_c as function of the impurity concentration. Thus, pressure experiments are an additional test of the model underlying the theory of superconductors containing magnetic impurities with crystalline field split energy levels.

Our experiments on $La_{1-x}Gd_xAl_2$ show that contrary to previous assumptions [15] the change of the scattering rate of conduction electrons under pressure cannot be neglected. This effect can be ascribed to a variation of the exchange interaction or the density of states at the Fermi surface.

2. Influence of Pressure on the Pairbreaking Effect of Magnetic Impurities

The strength of the effective crystal field acting on a rare earth impurity at a lattice site of cubic symmetry can be characterized by two expansion coefficients B_4 and B_6. The level system of rare earth ions has been calculated by LLW using the parameters, W, X_L which are related to B_4 and B_6 by

$$B_4 F(4) = W \cdot X_L, \tag{1a}$$

$$B_6 F(6) = W(1 - |X_L|) \tag{1b}$$

$F(4)$ and $F(6)$ are numerical factors depending on the total angular momentum J. The energy separation of any two levels μ, ν is given by:

$$\delta_{\mu,\nu} = W \cdot E_{\mu,\nu} \tag{2}$$

where $E_{\mu\nu} = E_\mu(X_L) - E_\nu(X_L)$ is the difference of the corresponding eigenvalues of the reduced Hamiltonian, tabulated by LLW. The strength of the crystal field depends on the lattice parameter R. On applying a hydrostatic pressure p the relative change of R is given by:

$$\Delta R/R = -\tfrac{1}{3}\kappa p \tag{3}$$

where κ is the compressibility of the material. Assuming

$$B_4 \sim R^{-5}; \quad B_6 \sim R^{-7} \tag{4}$$

which is valid under rather general conditions (especially for the point charge model), the variation of the parameters W and X_L is given by

$$\Delta W/W = -(7 - 2|X_L|)\Delta R/R, \tag{5a}$$

$$\Delta X_L/X_L = 2(1 - |X_L|)\Delta R/R. \tag{5b}$$

The relative change of the energy separation of the impurity levels contains two contributions:

$$\frac{\Delta\delta}{\delta} = \frac{\Delta W}{W} + \frac{\Delta E}{E}. \tag{6}$$

The first term reflects the general increase of the strength of the crystal field due to pressure and is always positive. The second term is proportional to ΔX_L and is related to the change of the level scheme by the change of the relative magnitude of B_4 and B_6. This term is important near crossing points of energy levels.

Concerning the pairbreaking of magnetic impurities with crystalline field split impurities the principal effect of pressure is the change of the ratio δ/T_{c0}, where δ is the energy separation between the ground state and the first excited state with a transition, allowed by the exchange interaction, and T_{c0} is the transition temperature of the host material. Thus a decrease of T_{c0} under pressure enhances the variation of the pairbreaking effect of the magnetic impurities.

There are secondary effects resulting from a change of the crystal field, which are proportional to ΔX_L: 1. the change of the relative position of higher energy levels compared to the ground state separation, 2. the change of transition matrix elements.

Furthermore we have to consider the influence of pressure on the exchange interaction J_{ex} between conduction electrons and magnetic impurities and on the density of states $N(0)$ at the Fermi surface, which both enter the scattering rate of conduction electrons on magnetic impurities:

$$\tau^{-1} = x \cdot 2\pi \cdot J_{ex}^2 N(0)(\Lambda - 1)^2. \tag{7}$$

Here x and Λ are the concentration and the Landé factor of the magnetic impurities. As in Ref. 5 the transition matrix elements have been separated from the definition of τ^{-1}, because they depend on the particular level system. In the following discussion of the data, the change of the scattering rate due to pressure will be determined from experiments on $\underline{La}GdAl_2$.