Pressure Dependence of T_c for $(Au_{1-x}Pd_x)Ga_2$ Alloys

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The superconducting transition temperature for the alloys $(Au_{1-x}Pd_x)Ga_2$ with $x = 0$, 0.012, and 0.024 has been measured as a function of hydrostatic pressure to 24 kbar. The very marked discontinuity in T_c, which occurs at ~ 6 kbar for $x = 0$ is displaced to lower pressure and becomes considerably broader as x is increased. This behavior is discussed in terms of the Fermi-surface transition previously proposed to explain de Haas–van Alphen and NMR measurements.

An almost discontinuous increase of the superconducting transition temperature T_c for the compound $AuGa_2$ at a pressure of ~ 6 kbar was recently reported[1] by one of us. An abrupt change in the Fermi-surface topology was suggested as the cause of this behavior in support of the interpretation placed upon some earlier[1,2] de Haas–van Alphen (dHvA) measurements. The present investigation extends the pressure range of the T_c measurement from the ~ 8-kbar maximum available in the previous work to 24 kbar and also included an examination of the effect upon the pressure dependence of T_c of changing the electron concentration by substituting 1.2 and 2.4-at.% Pd (nominal) for the Au.

Samples were cut from the same single-crystal ingots as those for the dHvA measurements. A clamp arrangement, in which pressure was generated in a fluid-pressure medium at room temperature, provided pressure up to 24 kbar. The pressure at the low temperature was determined with a superconducting-tin manometer.[3] Pressures up to ~ 5 kbar were also applied to the Pd-doped samples by the isobaric freezing of helium, as in the initial measurements[1] on $AuGa_2$. The transition to the superconducting state was detected by a standard ac inductance technique in both pressure systems.

The variation of the superconducting transition temperature with pressure for the $AuGa_2$ and the Pd-substituted samples is shown in Fig. 1. The data from the earlier measurements on $AuGa_2$ have also been included and the two sets of measurements are in excellent agreement. With the exception of the values close to 6 kbar using the clamp technique, T_c is taken to be the midpoint of the central linear portion of the superconducting transition curve. The curves obtained for $AuGa_2$ in the clamp broadened considerably in the vicinity of the sudden increase in T_c, with the spread of the transition curves being consistent with a continuum of transition temperatures distributed over the range of the discontinuity in T_c. The deviation from a homogeneous pressure distribution within the transmitting medium could possibly be as much as 0.3 kbar at low temperature, which would be sufficient to account for the broadening of the transition curves near the abrupt change in T_c. However, the onsent of the superconducting transition remained sharp and agrees well with the T_c values obtained by pressuring with helium, where there was much less broadening ($\leq$ 40 mK), presumably because of superior hydrostatic pressure conditions at the low temperature. At higher pressure the transition curves sharpened again, ultimately attaining widths[4] of ~ 5 mK, making them sharper than the initial zero-pressure and low-pressure curves which had widths of 10–15 mK. This sharpening of the transition curves presumably reflects the decrease in the sensitivity of the transition temperature to pressure above the discontinuity.

Further, notable features of the transitions for

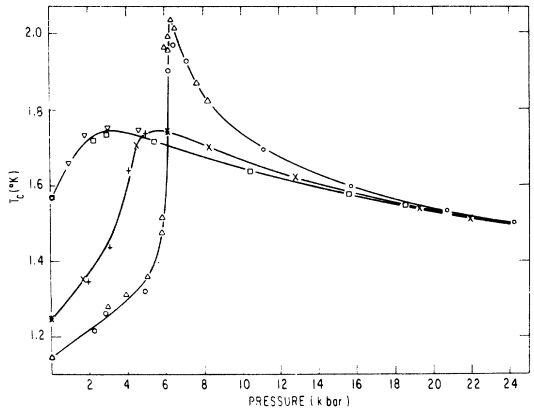

FIG. 1. Variation of T_c with pressure for $Au_{1-x}Pd_xGa_2$ alloys. The different symbols distinguish the value of x and the pressure system in which the measurements were made as follows; $x = 0$, △—helium, ○—clamp; $x = 0.012$, +—helium, ×—clamp; $x = 0.024$, ▽—helium, □—clamp. The solid lines are smooth curves drawn through the data.

$AuGa_2$ observed at pressures above 15 kbar were a well resolved and reproducible supercooling of ~5 mK for decreasing temperature and a marked differential paramagnetic effect (DPE) upon warming back through the transition. The DPE is characteristic of samples which are magnetically reversible.[5] The supercooling would indicate that $AuGa_2$ is a type-I superconductor, consistent with its ratio of Debye temperature to T_c of ~170. Thus, in spite of any inhomogeneity in strain distribution introduced into the sample as a consequence of the application of pressures in excess of 15 kbar, it would appear that the magnetic reversibility of the sample has been improved relative to its behavior at zero and low pressures.

The transitions for the Pd-doped samples, where the change in T_c with pressure is much less abrupt than for the pure $AuGa_2$, did not exhibit any pressure broadening, but did become sharper at higher pressure, particularly in the case of the 2.4-at.% sample where the zero-pressure transition width of ~50 mK decreased to ~5 mK at 19 kbar. Supercooling was not observed for the Pd-doped samples, but a DPE, less pronounced than that for the pure $AuGa_2$, was seen at pressures in excess of 10 kbar.

The T_c measurements for the Pd-substituted samples made in the helium system were taken in conjunction with a NMR study[6] and a small adjustment to the measured T_c value (~60 mK) was necessary to correct for the residual field (~10 G) of the electromagnet. The corrections required to bring the zero-pressure T_c values, determined in the two pressure systems, into agreement are consistent with estimates based upon rough values[7] for the zero-temperature critical field.

The rapid change in the zero pressure T_c and an accompanying change in the coefficient of the electronic specific heat, γ, when Pd is substituted for Au in $AuGa_2$, was reported by Wernick *et al.*[7] who suggested a change in the relative position of the energy bands as a possible explanation of this behavior. A change in the character of the electrons at the Fermi surface from p-like to s-like in going from low temperature to room temperature had been proposed[8] earlier to account for the anomalous temperature dependence of the Ga^{71} Knight shift in $AuGa_2$. These suggestions were subsequently substantiated by the band-structure calculation of Switendick and Narath[9] which predicted a very flat portion of the second-zone energy band, of strong Ga 4s character, lying just below the Fermi level. Thus, even a small reduction in electron concentration would place the Fermi energy in a region of high density of states and would account for the observed increases in T_c and γ. A thermal depopulation of this band, rather than its movement with temperature, provides the most consistent description of the NMR results.[8,9] Recent dHvA[1,2] and NMR[6] measurements, taken as a function of pressure, as well as the band-structure calculations as a function of pressure,[2] show that this band moves up relative to the Fermi energy as the volume is decreased. The almost discontinuous change of T_c at ~6 kbar was then associated with the passage of the band through the Fermi level.[1]

It can be seen from Fig. 1 that a small substitution of Pd for Au significantly displaces the peak in T_c towards lower pressure and broadens it considerably. This displacement is consistent with the above picture of a lowering of the Fermi level by Pd substitution and thus a reduction of the relative shift of the second-zone band required to cross it. The sharpness of the peak in T_c for the $AuGa_2$ and its subsequent broadening with addition of Pd, points to a very strong singularity in the density of states, which is smeared out by electron scattering.

Changes in the Fermi-surface topology, much less dramatic than that considered here, have been held responsible for relatively small contributions (typically $\Delta T_c \sim 20$ mk) to the pressure dependence of T_c in several elements and alloys.[10] These small changes are observable because of the exponential dependence of T_c on the product of the density of states and the electron-phonon interaction. Since the Knight Shift for $AuGa_2$, which can be considered in this context to be proportional to the density of states alone, is observed[6] to

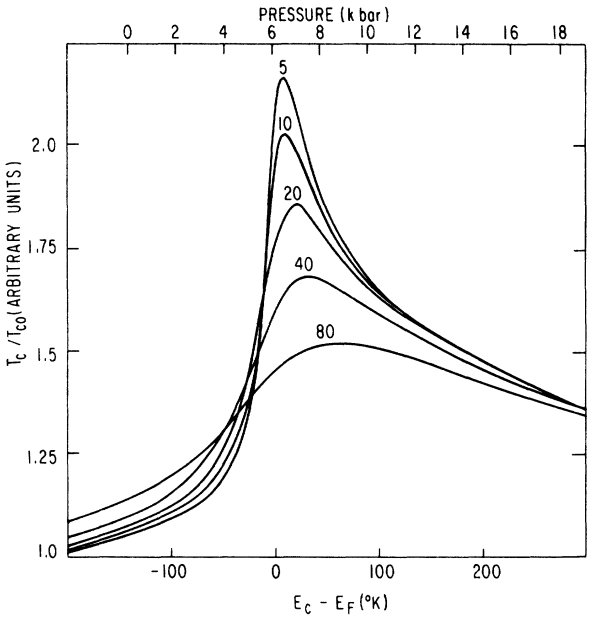

FIG. 2. Calculated variation of T_c/T_{c0} as a function of the energy difference $E_c - E_F$ for the indicated values, in K, of the electron-scattering parameter Γ. The upper pressure scale is based upon the band structure calculation as a function of volume for $AuGa_2$.

also undergo a striking increase with pressure, concurrent with T_c, we have chosen to assume that the bulk of the change in T_c is associated with an abrupt increase in the density of states and neglect any change in the electron-phonon interaction. By considering the density of states as a function of energy about the singularity of E_c to be composed of an essentially constant background $N_0(E_F)$ and a strongly energy-dependent part $\Delta N(E_F)$, the appropriate expressions for the variation of T_c are[11]

$$T_c = T_{c0} \exp\left[F(\Delta N(E_F), T_c)/2N_0(E_F)\right], \quad (1)$$

where

$$F(\Delta N(E_F), T_c) = \int_{-k\theta D}^{k\theta D} \frac{\tanh(|E|/2kT_c)}{|E|} \times \Delta N(E_F - E)\, dE \,. \quad (2)$$

Various simple functional forms for $\Delta N(E_f)$ have been substituted in (2) and (1) solved numerically in an attempt to reproduce the measured variation of T_c for $AuGa_2$. The nearest approach to the correct shape was found for $\Delta N(E_F) \propto E^{-1/2}$. The inclusion of electron scattering may be achieved most readily by considering the Green's-function method[12] for obtaining the density of states which leads to the expression[13]

$$\Delta N(E_f) = \frac{\Gamma}{\pi} \int_{\infty}^{\infty} \frac{dk}{(E-\epsilon)^2 + \Gamma^2} \,, \quad \epsilon = \frac{\hbar^2 k^2}{2m} \quad (3)$$

$$\Delta N(E_f) = (A\Gamma)\left\{(E^2+\Gamma^2)\left[(E^2+\Gamma^2)^{1/2} - E\right]\right\}^{-1/2} \quad (4)$$

where Γ is the imaginary part of the electron energy, i.e., $1/\Gamma$ represents the lifetime of an electron in a given energy state and A is a renormalization constant. Figure 2 shows the resulting curves of T_c as a function of $E_c - E_F$ ($\propto P$) for different values of Γ, obtained by substituting (4) into (2). The pressure scale as determined from the band calculation as a function of volume[1,2] for pure $AuGa_2$ is also shown to facilitate comparison with the experimental curves in Fig. 1. The correspondence between the general characteristics of these curves with the observed behavior is quite striking. However, any attempt to make a much more quantitative fit to the data in view of the assumptions involved or to attach significance to the details of the energy dependence of the density of states does not seem to be warranted.

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