Pressure-Induced Electron Transition in $AuGa_2$

J. E. Schirber
Sandia Laboratories, Albuquerque, New Mexico 87115

(Received 13 March 1972)

We report measurements of a large, nearly discontinuous change in the superconducting transition temperature of $AuGa_2$ near 6 kbar as predicted earlier from pressure measurements of cross-sectional areas of the Fermi surface. This is interpreted as strong evidence for a pressure-induced electron transition near 6 kbar at liquid-He temperatures.

The anomalous temperature dependence of several of the magnetic properties[1,2] of $AuGa_2$ has prompted a considerable body of recent experimental work including studies of the superconducting and magnetic properties as a function of Pd doping,[3] alloying studies[4] with $AuAl_2$ and $AuIn_2$, elastic-constant measurements,[5] optical measurements,[6] and Fermi-surface studies as a function of pressure.[7] Most of the results to date are in qualitative agreement with the band model of Switendick and Narath.[2] Here the extremely flat second band possessing strong Ga 4*s* character is found to be completely below the Fermi level at zero temperature. Depopulation of this band as the temperature is increased was then proposed[2] as the explanation for the change in properties of $AuGa_2$.

Measurements of the pressure dependence of Fermi surface cross sections in $AuGa_2$ and in the isomorphous compounds $AuAl_2$ and $AuIn_2$ indicated that the depopulation of the second was *not* due to the volume effect associated with the thermal expansion.[7] This experimental work and the results of the band calculation as a function of interatomic spacing both predicted that, with sufficient hydrostatic pressure, the second band would pass through the Fermi energy from below. In a recent paper[8] we reported an unusual discontinuity in the pressure dependence of the L-centered necks of the third-band hole sheet. We speculated that this was a reflection of the creation of the second-band hole sheet—as the large number of states in the second band is uncovered, states must be filled elsewhere. We predicted that if this "electron transition"[9] were occurring, the superconducting transition temperature T_c and the Knight shift would increase dramatically at about 6 kbar.

In this Letter we report measurements of the superconducting transition temperature of $AuGa_2$ as a function of hydrostatic pressure to ~8 kbar. These measurements (i) lend strong support to the band picture for $AuGa_2$ and give a direct confirmation of a high density-of-states peak located at normal volume immediately below the Fermi energy, and (ii) provide an excellent situation for a detailed study of the illusive "electron transition."[9] While such transitions are thought to be operative in several instances such as Tl,[10] InCd,[11] and Re,[12] the effects on T_c are extremely small and no direct correspondence with the electronic band structure has yet been demonstrated. Those materials, such as Bi-Sb[13] and As,[14] where the transition has been demonstrated, do not lend themselves to such studies because of the low symmetry of the lattice, the attendant, somewhat complicated, band structures, and the fact that they are nonsuperconducting.

Samples were cut by spark erosion from the same single-crystal boule as were the de Haas-van Alphen (dHvA) specimens. The boule had a residual resistance ratio of ~250 as determined by the eddy-current decay method. Superconducting transitions were determined by measurement of the change in inductance detected with a General Radio inductance bridge in conjunction with a lock-in detector operated from 20–70 Hz. The detection coils were outside the BeCu pressure vessel. We defined T_c as the midpoint of the change in inductance, which may account in part for the fact that our normal volume for T_c of 1.13–1.16 K is somewhat higher than that reported by Wernick *et al.*[3] (1.05–1.12 K). Pressures were generated by careful isobaric freezing of He as described in detail earlier.[15]

In Fig. 1(a) we show our results for T_c as a function of pressure. These are compared with the above-mentioned change of the third-band Fermi-surface cross section shown in Fig. 1(b). We see that T_c increases very abruptly (within 0.2 kbar) near 6 kbar, the same pressure where the third-band cross section begins to decrease at an accelerated rate. The cross-sectional areas were obtained by measurement[14] of dHvA frequencies using the now familiar field-modulation technique. Our reproducibility in pressure

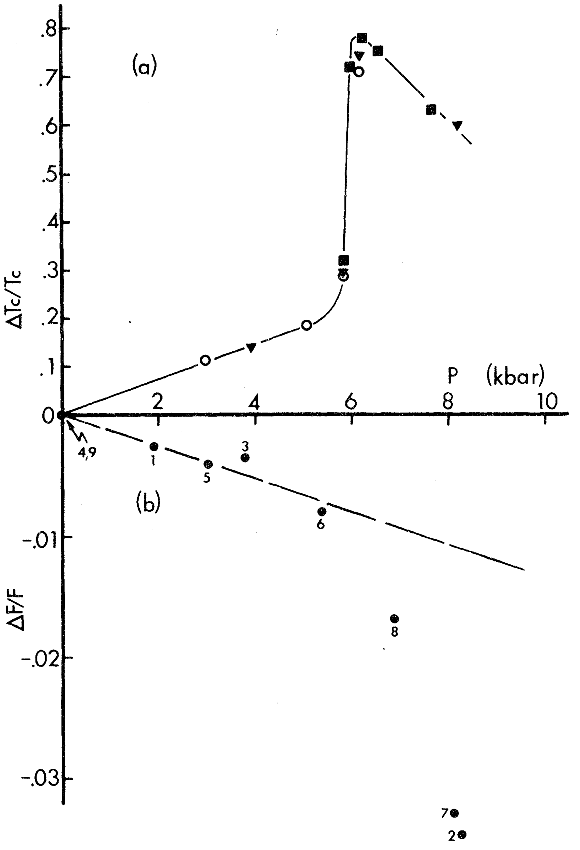

FIG. 1. (a) Pressure dependence of the superconducting transition temperature T_c versus pressure. $\Delta T_c/T_c$ is defined as $[T_c(P)-T_c(P=0)]/T_c(P=0)$. $T_c(P=0)=1.13$–1.16. The solid line is a smooth curve through the data. (b) Pressure dependence of dHvA frequency F associated with the third-zone arms for $\vec{H}\parallel[111]$. $\Delta F/F$ is defined in the same way as is $\Delta T_c/T_c$. The dashed line is the slope of the fluid-He results (≤ 25 bars). Points are numbered in the sequence in which they were taken. In (a), a similar sequence was employed on each of the three runs shown.

with the present experimental setup is estimated to be 0.1 kbar, so the jump in T_c may be even sharper than indicated. The points in Fig. 1(b) are numbered in the sequence they were taken. The T_c data were obtained in similar sequences indicating that the effects are both reproducible and reversible. Both the dHvA and T_c measurements were made on at least two samples, and in one case the same sample was used in both experiments.

The substantial increase in T_c before the abrupt change near 6 kbar is worthy of comment. We found that T_c at zero pressure was quite sensitive to sample history. As-spark-cut crystals had broad transitions (up to 0.150 K) and showed high values of T_c (up to 1.38 K). Subsequent annealing at about 375°C for ~18 h lowered the T_c value below 1.2 K and sharpened the transition to ≤ 0.040 K. The high-pressure (>6 kbar) transitions were sharper (≤ 0.020) and were unchanged by the annealing. We attribute this behavior to internal strains which force portions of the crystal into the high density-of-states region, thus raising and broadening the bulk transition.

The change in T_c observed corresponds, within the BCS framework,[16] to a change in the total density of states at the Fermi level of about 10% (assuming the electron-phonon interaction is not changing). This is of the same order as observed by Wernick *et al.*[3] who (in the spirit of a rigid-band picture) uncovered this band by lowering E_F by Pd doping and saw large changes in the electronic specific heat coefficient and the susceptibility as well as in T_c.

The small volume change involved with 6 kbar (0.7%) and the marked sensitivity to strain of T_c are suggestive that the band lies very near the Fermi energy, unless there is a change in the crystal structure with pressure. We cite the fact that we are able to observe the higher frequency third- and fourth-band Fermi-surface cross sections above 8 kbar as a strong indication that there is no structural change involved, and that we are observing a purely electronic transition as the second band is uncovered. Therefore, we would expect that the Knight shift will prove to be an equally sharp function of pressure at liquid-He temperatures as is the superconducting transition temperature. Our model would thus predict that the band actually never crosses the Fermi level at normal volume at any temperature. Rather, the exponential depopulation of the band with increasing temperature overcomes the relative drop of the band with respect to E_F as the temperature rises (due to thermal expansion). Thus the states in the second band are thermally uncovered even though the band is below the Fermi energy. The shape of the Knight shift versus the temperature curve[1] is consistent with this picture. The Knight shift changes slowly initially (0–40 K), then increases rather rapidly between 60 and 100 K, and then continues to increase more slowly up to room temperature. This slowing of the increase of the Knight shift is expected as the thermal expansion of the lattice lowers the band relative to E_F, thus requiring a larger increase in temperature to thermally depopulate the band. The behavior of the compressibility with temperature, as determined from Testardi's elastic-constant data,[5] is very similar.

We can make an estimate for the distance in energy that the second band lies below E_F at absolute zero from Schirber and Switendick's volume-dependent band calculation.[7] This gave a difference between the increase in energy of the second band at Γ and the energy of the other bands at L of 0.067 V with ~30 kbar. Assuming this difference is a measure of the rate at which the second band moves relative to E_F, this equates ~0.002 V to 1 kbar and puts the band ~0.012 V below the Fermi energy at absolute zero. This value is roughly consistent with the thermal behavior of the Knight shift and elastic-constant data which change in a temperature region centered around 80 K or ~0.007 V. This reasoning also indicates that the peak in the density of states is quite sharp (width at half-height of perhaps 0.005 V) since T_c drops appreciably between 6 and 8 kbar.

In conclusion, we feel that the combination of the unusual pressure dependence of the third-zone Fermi-surface cross sections and the almost discontinuous pressure dependence of the superconducting transition temperature argue very strongly for a pressure-induced electron transition in AuGa$_2$. Such a transition supports both the experimental inference from the studies of AuGa$_2$, AuAl$_2$, and AuIn$_2$ series and the theoretical predictions of Schirber and Switendick's volume-dependent band model. We further propose that the anomalous temperature dependence of magnetic properties of AuGa$_2$ stems from a purely thermal depopulation of the second band, thus requiring no explicit phonon effects such as suggested earlier.[7]

The technical assistance of R. L. White is gratefully acknowledged, as are helpful discussions with A. C. Switendick, J. P. Van Dyke, and A. Narath.

[1] V. Jaccarino, M. Weger, J. H. Wernick, and A. Menth, Phys. Rev. Lett. 21, 1811 (1968), and references therein.

[2] A. C. Switendick and A. Narath, Phys. Rev. Lett. 22, 1423 (1969).

[3] J. H. Wernick, A. Menth, T. H. Geballe, G. Hull, and J. P. Maita, J. Phys. Chem. Solids 30, 1949 (1969).

[4] G. C. Carter, I. D. Weisman, and L. H. Bennett, Phys. Rev. B (to be published).

[5] L. R. Testardi, Phys. Rev. B 1, 4851 (1970).

[6] S. Hüfner, G. K. Wertheim, R. L. Cohen, and J. H. Wernick, Bull. Amer. Phys. Soc. 17, 92 (1972).

[7] J. E. Schirber and A. C. Switendick, Solid State Commun. 8, 1383 (1970).

[8] J. E. Schirber, Phys. Rev. B (to be published).

[9] I. M. Lifshitz, Zh. Eksp. Teor. Fiz. 38, 1569 (1960) [Sov. Phys. JETP 11, 1130 (1960)].

[10] L. D. Jennings and C. A. Swenson, Phys. Rev. 112, 31 (1958).

[11] V. I. Makarov and I. Ya. Volynskii, Zh. Eksp. Teor. Fiz., Pis'ma Red. 4, 369 (1966) [JETP Lett. 4, 249 (1966)].

[12] C. W. Chu, T. F. Smith, and W. E. Gardner, Phys. Rev. B 1, 214 (1970).

[13] E. S. Itskevich and L. M. Fisher, Zh. Eksp. Teor. Fiz., Pis'ma Red. 6, 748 (1967) [JETP Lett. 6, 219 (1967)].

[14] J. E. Schirber and J. P. Van Dyke, Phys. Rev. Lett. 26, 246 (1971), and references therein.

[15] J. E. Schirber, Cryogenics 10, 418 (1970).

[16] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

Magnetophonon Resonance of Hot Electrons in *n*-InSb at 77°K

Chihiro Hamaguchi, Tsuguru Shirakawa, Tatsuo Yamashita, and Junkichi Nakai
Department of Electronics, Osaka University, Suita City, Osaka, Japan

(Received 29 February 1972)

Magnetophonon resonances in the hot-electron mobility have been observed in pure *n*-InSb at 77°K in transverse magnetic fields with the application of sufficiently small voltage to produce a slight change in the mobility. The minima in the quantity β in the formula $\mu = \mu_0(1 - \beta E^2)$ are attributed to resonant cooling of hot electrons due to optical-phonon–induced transitions between Landau levels.

We report here a new type of resonance experiment consisting of the observation of magnetophonon resonances in the hot-electron mobility in *n*-InSb at 77°K. Magnetophonon resonances were first predicted by Gurevich and Firsov,[1] and subsequently confirmed experimentally by many workers.[2] Magnetophonon phenomena in the electrical resistance reflect the resonant inelastic scattering of electrons between the high densities of states in Landau levels close to k_z