Pressure dependence of the cyclotron mass of antimony

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The de Haas—van Alphen (dHvA) effect of the minimum section of the hole Fermi surface of antimony was observed at pressures between 0 and 4.3 kbar. The cyclotron effective mass at zero pressure is $(0.0637\pm0.0006)m_0$, 6% larger than the band mass calculated by Pospelov and Grachev. The pressure dependence of the cyclotron mass is $(1/m^*)dm^*/dP=(0.02\pm0.4)\times10^{-2}$ kbar^{-1}. The small change in mass is attributed to a change in the band mass that can be described by the ellipsoidal-nonparabolic model. The pressure derivative of the dHvA frequency is negative at low pressures and becomes zero at 3.7 ± 0.3 kbar and then changes sign with further increase in pressure. There appear to be at least two mechanisms determining the pressure dependence of the dHvA frequency.

I. INTRODUCTION

The cyclotron mass of a carrier in a metal is determined by the band-structure mass enhanced by many-body effects that result from electron-phonon and electron-electron interactions. These terms may also be studied through the pressure dependence of the cyclotron mass. The first studies showed that the logarithmic pressure derivative of the mass, $d\ln m^*/dP$, is $(-0.48\pm0.09)\%$ kbar^{-1} and $(-0.71\pm0.12)\%$ kbar^{-1} for sodium and potassium, respectively.[1,2] The dominant contribution to this pressure dependence results from the electron-phonon interaction because phonons stiffen more rapidly with pressure than the plasma frequency and the form factor changes with pressure. The logarithmic pressure dependence of the band mass, $d(\ln m_B)/dP$, calculated from the volume dependence of the mass, is 0.05% and 0.27% for sodium and potassium, respectively.[3] The difference between these calculated values is surprising because the two alkalai metals are very similar. Also, a smaller value for potassium makes the pressure dependence of the cyclotron mass understandable in terms of the band mass and the electron-phonon mass enhancement.[1] For these reasons, it is desirable to measure the pressure dependence of the band mass. However, this is not possible for the alkali metals in which the band mass has considerable enhancement from the electron-phonon and electron-electron interactions. The influence of pressure on the band mass must be studied in a metal with negligible many-body effects, especially negligible electron-phonon interaction. Antimony, which is a semimetal with a low carrier concentration satisfies this criterion. It is, of course, considerably different than the alkali metals but can be used to find out whether the logarithmic pressure dependence of a metal is small or large and whether it can be explained directly by band theory.

The band mass of antimony at zero pressure is also of interest. Recently, it has been calculated[4] directly from integrals that depend on certain parameters determined from the pseudopotential scheme of Falicov and Lin.[5] This value is compared with our measured mass. An explanation is also given for Windmiller's[6] cyclotron mass value that is larger than those obtained in other experiments with the same magnetic field direction.

The Fermi surface of antimony consists of three closed energy surfaces of electrons at point L of the rhombohedral Brillouin zone and six pockets of holes near the point T. The directions of the normal to the minimum cross sections in the bisectrix-trigonal mirror plane are at angles of 87.7° and 53° from the trigonal, threefold axis for electrons and holes, respectively.[6] All measurements of the de Haas—van Alphen (dHvA) effect for this study were taken with the magnetic field perpendicular to the minimum hole cross section.

Previous experiments have established anomalous properties of the pressure dependence of the dHvA frequency of the minimum hole cross section.[7–11] With increasing pressure from zero pressure, the dHvA frequency decreases to a minimum after which it increases with pressure. It attains its zero pressure value at 5 kbar above which it increases linearly with pressure. Previous measurements are in general agreement with the measurements between 0 and 4.3 kbar that are presented in this paper. However, the present results provide considerably more information about the detailed pressure dependence in the interesting region below 5 kbar.

II. EXPERIMENTAL

The field modulation measuring technique[12] modified to achieve frequency discrimination[6] was used to detect the dHvA effect with magnetic fields up to 1.9 T. Two sets of modulation coils were fixed rigidly in an orthogonal configuration on the outside of the pressure cell and the detection coils mounted in series opposition were inside the cell. An antimony sample with a purity of 99.999% was mounted for magnetic field rotation in the trigonal-bisectrix plane. The direction of the magnetic field for the minimum hole frequency was determined to within $\pm0.6°$ from field-rotation diagrams at each pressure.

Hydrostatic pressures up to 4.3 kbar were applied by the helium freezing method using a pressure generating system described previously.[13,14] A correction for the contraction of solid helium between the freezing temperature and the operating temperature was made from the data of Spain and Segall.[15]

The cyclotron mass was determined from the temperature dependence of the dHvA amplitude in the temperature range of 1.2–4.2 K.[16] Data were taken at 10–15 different temperatures at each pressure. The amplitude of the oscillatory magnetization, $A(T)$, is proportional to $T(\sinh u)^{-1}$, where $u=14.69m^*T/m_0H$. This proportionality can be written as

$$\ln[A/T(1-e^{-2u})]=-u+\text{const}\equiv\ln Y \ . \tag{1}$$

Therefore, a plot of $\ln Y$ versus T yields a straight line of slope $-14.69(m^*/m_0H)$, where m^*/m is the cyclotron mass normalized to the free-electron mass. Iterations were performed with improved values of m^*/m_0 until the plot was linear to obtain the best value of m^*/m_0.

The measured hydrostatic pressure dependence of the cyclotron effective mass in the pressure range between 0 and 4 kbar is presented in Fig. 1. The weighted least-squares fit to all data points yields $dm^*/dP=(0.1\pm0.2)\times10^{-3}m_0\ \text{kbar}^{-1}$ and the logarithmic pressure derivative

$$(1/m^*)dm^*/dP=(0.2\pm0.4)\times10^{-2}\ \text{kbar}^{-1} \ .$$

The intercept of m^* versus P corresponds to a standard atmospheric pressure mass of $(0.0635\pm0.0005)m_0$. The cyclotron effective mass at standard atmospheric pressure was measured before applying pressure and after removal of pressure. The measured values were $(0.0635\pm0.0002)m_0$ and $(0.0641\pm0.0009)m_0$. An average of these two values along with the intercept at zero pressure is

$$m^*=(0.0637\pm0.0006)m_0 \ .$$

The hydrostatic pressure dependence of the minimum frequency of the hole Fermi surface is shown in Fig. 2. The data points indicate that the frequency decreases with increasing pressure in a nonlinear fashion up to 3 kbar. The frequency reaches its minimum value at (3.7 ± 0.3) kbar and the pressure derivative changes sign with further increase in pressure.

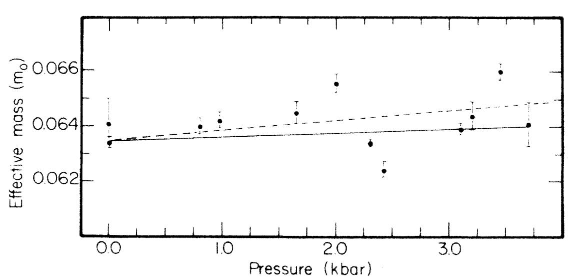

FIG. 1. Pressure dependence of the cyclotron effective mass of antimony for the minimum hole cross section. The solid line is a least-squares linear fit and the dashed line is a prediction from the ellipsoidal-nonparabolic model with $dF/dP=0.6\times10^{-2}\ \text{T kbar}^{-1}$.

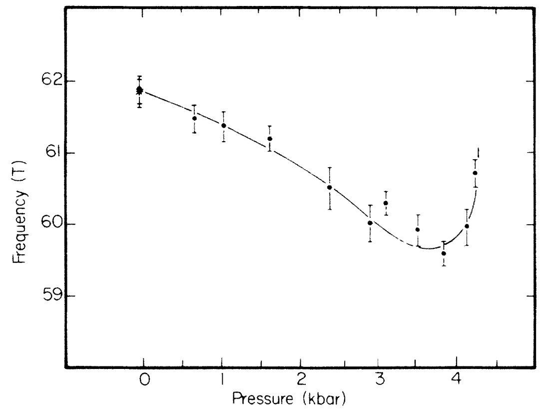

FIG. 2. Pressure dependence of the de Haas–van Alphen frequency of the minimum section of the hole Fermi surface of antimony for pressures between 0 and 4.3 kbar.

III. DISCUSSION

A comparison of experimental and calculated values of the cyclotron effective mass at standard atmospheric pressure determined by various investigators using different methods is presented in Table I. Values by Brandt *et al.*,[17] Windmiller,[6] Dunsworth and Datars,[18] and Altounian and Datars[19] were derived from the temperature dependence of the dHvA effect. The effective mass was determined from cyclotron resonance by Datars and Vanderkooy.[20] The theoretical value of m^*/m_0 was estimated by Pospelov and Grachev[4] who carried out a calculation of cyclotron masses in antimony using the pseudo-potential scheme of Falicov and Lin[5] that does not take into account explicitly many-body effects. The calculated value is 6% less than the measured value. This indicates that many-body mass enhancement is small.

Our present result is in good agreement with other measurements within combined experimental errors, wherever they are quoted, except for the value reported by Windmiller.[6] The difference between our estimated value and Windmiller's result is significantly outside combined experimental error. He derived the value from a plot of $\ln(A/T)$ versus T, which is a first approximation of the expression of the de Haas–van Alphen amplitude as a function of temperature. A least-squares fit of the present experimental data from a plot of $\ln(A/T)$ against T yields $m^*=(0.069\pm0.001)m_0$, which is the value given by Windmiller. Thus, the discrepancy is due to their use of the first approximation rather than Eq. (1) which should have been used.

The pressure dependence of m^* associated with the minimum section of the hole Fermi surface was studied by Brandt *et al.*[11] using the Shubnikov–de Haas effect. In their work, the temperature dependence of the oscillation amplitude was measured only at temperatures of 4.2 and 2.1 K from which m^* was deduced. Their tabulated data of m^* versus pressure showed rather poor precision in comparison with the accuracy achieved in the present measurements. At pressures of 0.4 and 4.5 kbar, they re-

TABLE I. Reported values of the calculated and experimental cyclotron effective mass associated with the minimum section of the hole Fermi surface of pure antimony in the trigonal-bisectrix plane at standard atmospheric pressure.

	m^*/m_0	Investigator
Calculation	0.06	Pospelov and Grachev (Ref. 4)
Experiment	0.062±0.01	Datars and Vanderkooy (Ref. 20)
	0.058	Brandt *et al.* (Ref. 11)
	0.069±0.002	Windmiller (Ref. 6)
	0.063	Dunsworth and Datars (Ref. 18)
	0.063	Altounian and Datars (Ref. 19)
	0.0637±0.0006	Present work

ported m^* to be $(0.075\pm0.004)m_0$ and $(0.081\pm0.006)m_0$, respectively. This large change with pressure is not supported by the present measurements.

The cyclotron mass is independent of the Fermi energy for an ellipsoidal Fermi surface in a parabolic band (the ellipsoidal-parabolic model). Thus, a change in Fermi energy by the application of pressure does not alter the cyclotron mass. However, the Fermi surface and band structure is not as simple as the ellipsoidal-parabolic model and changes in the Fermi energy by adding tin and tellurium cause a change in the cyclotron mass.[18,19] This was described by the ellipsoidal-nonparabolic model although the band structure of Falicov and Lin[5] provides a more complete description. The model is useful for describing the pressure dependence because empirical parameters are available from the work with alloys.

The energy dispersion relation in the ellipsoidal-nonparabolic model has the form

$$\hbar^2k_1^2/m_1+\hbar^2k_2^2/m_2+\hbar^2k_3^2/m_3=E+E^2/E_g\ , \quad (2)$$

where 1, 2, and 3 refer to the principal axes of the ellipsoid, the m's are the band-edge effective mass components along the principal axes, and E_g is the energy gap between bands. The nonparabolic energy band leads to an energy dependence of the cyclotron mass by[18,19]

$$m^*=m_b(1+E/E_g)\ , \quad (3)$$

where m_b is the band-edge mass in units of the free-electron mass. The cross-sectional area S is

$$S=\pi m_b(1+E/E_g)E \quad (4)$$

combining (3) and (4) gives

$$(m^*)^2=m_b^2+(4/\pi)(m_b/E_g)S\ . \quad (5)$$

Since S is directly proportional to the dHvA frequency F,

$$(m^*)^2=m_b^2+0.46(m_b/E_g)F\ , \quad (6)$$

where F is in units of Tesla and E_g is in meV. If we assume that the parameters m_b and m_b/E_g are insensitive to pressure in the pressure region investigated, the derivative of Eq. (6) with respect to pressure yields

$$dm^*/dP=0.23/m^*(m_b/E_F)dF/dP\ . \quad (7)$$

To evaluate Eq. (7) we use the average value of $E_g/m_b=7.2$ meV for Sb-Sn and Sb-Te alloys[18,19] and $m^*=0.0637m_0$. The use of $dF/dP=-0.56$ T kbar^{-1} derived from a linear least-squares fit of data of Fig. 2 for pressures up to 3.89 kbar yields $dm^*/dP=-0.28\times10^{-2}m_0$ kbar^{-1}. This has the opposite sign of that described from the fit in Fig. 1. The Fermi energy should increase with pressure to have dm^*/dP positive according to Eq. (3). This occurs at high pressures for which $dF/dP=(0.6\pm0.1)\times10^{-2}$ T kbar^{-1}.[11] The use of this value in Eq. (7) gives a pressure dependence that is in reasonable agreement as shown in Fig. 1. This indicates that the Fermi energy actually increases with pressure at low pressures although F decreases with increasing pressure.

It is interesting to compare the cyclotron mass of antimony and potassium. The electron-phonon mass enhancement of potassium is approximately 20%. This is in contrast with antimony, where the electron-phonon interaction is expected to be negligible and the measured m^* at standard atmospheric pressure, $(0.0637\pm0.0006)m_0$, is 6% larger than the calculated band-structure mass, $0.06m_0$.[4] The experimentally determined logarithmic pressure derivative of the cyclotron mass of potassium is $-(0.71\pm0.15)\times10^{-2}$ kbar^{-1}, which is approximately 5 times larger than the value for antimony[2] and has a dominant contribution from the electron-phonon interaction. In contrast, the pressure derivative of m^* for antimony is influenced by just the band structure. Thus, investigating the effect of pressure on the cyclotron effective mass of antimony helps to exclude a contribution from the electron-phonon interaction and to identify separately the pressure dependence of the band-structure mass which turns out to be nearly insensitive to pressure as predicted from band theory.

Figure 2 provides a clear picture of the change of the dHvA frequency up to 4.3 kbar. The minimum frequency is at 3.6 kbar. There is an overall downward curvature between 0 and 3 kbar added to an upward curvature which is evident at higher pressures. This indicates that there are at least two mechanisms controlling this pressure dependence. One is the change of Fermi energy with pressure. It controls the change of mass with pressure as shown in Fig. 1. However it is the dominant mechanism only at high pressures for the pressure dependence of the frequency. It is suggested that the second mechanism, which is dominant at low pressures, is a change of shape of the Fermi surface with pressure. This may be possible because the Fermi surface is nonellipsoidal near the minimum section and further deviations from an ellipsoid are possible.

IV. CONCLUSIONS

The cyclotron effective mass of the minimum section of the hole Fermi surface of antimony at zero pressure is $(0.0637\pm0.0006)m_0$. It is 6% larger than the band mass calculated by Pospelov and Grachev.[4] The pressure derivative of the cyclotron mass is small and positive. It is explained in terms of the ellipsoidal-nonparabolic model by a change in Fermi energy with pressure. Detailed data of the dHvA frequency for pressures between 0 and 4.3 kbar show a downward curvature at pressures below that of the minimum frequency. There appear to be at least two mechanisms determining this pressure dependence.

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