

AN EXPERIMENTAL DETERMINATION OF THE ELECTRONIC COMPONENT OF THE  
THERMAL EXPANSION COEFFICIENT OF IRON

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In the range 1.5 to 4.2° K the value  $\alpha_e = (3.2 \pm 0.3) \times 10^{-9}$  T/deg is obtained for the electronic component of the thermal expansion coefficient of iron. The Grüneisen constant in this temperature range is found to be close to its high-temperature value. It is therefore concluded that the Debye temperature and electron state density in iron vary in the same way with identical relative changes of volume.

**I**MPORTANT information regarding the properties of conduction electrons has been obtained by measuring the specific heats of metals at low temperatures. It is well-known that the heat capacity of a nonferromagnetic metal can be divided into electronic and phonon terms, represented in the equation

$$C = \gamma T + DT^3, \quad (1)$$

In the case of ferromagnetic metals a term proportional to  $T^{3/2}$  is added to take account of spin waves. The coefficient of the electronic heat capacity,  $\gamma = \frac{2}{3}\pi^2 k^2 N(\epsilon_0)$ , determines the density  $N$  of electron states at the Fermi surface. The quantity  $\gamma$  plays a large part in the study of all electronic properties of metals and has been measured calorimetrically at low temperatures for a large number of metals.

The thermal expansion coefficient of ferromagnetic metals, like the heat capacity, depends on electronic, phonon, and spin mechanisms; the volume thermal expansion coefficient is

$$\beta = AT + BT^3 + ET^{3/2}. \quad (2)$$

The coefficient  $A$  of the electronic term furnishes new information regarding the electron spectrum of metals; its pressure dependence will be discussed here in some detail. Until very recently experimental data for this quantity were lacking because it is extremely difficult to measure very small thermal expansions at liquid helium temperatures. The components of  $\beta$  can be evaluated by using the relation between  $C$  and  $\beta$  given in Grüneisen's formula

$$\beta = \Gamma C \kappa_T / V_m, \quad (3)$$

where  $\Gamma$  is the Grüneisen constant,  $\kappa_T$  is the coefficient of isothermal compressibility, and  $V_m$  is the molar volume. Taking the known heat capacity of iron,<sup>[1]</sup>

$$C = (11.7 \pm 0.1) \cdot 10^{-9} T + 46.44 (T/477)^3 \\ + (2 \pm 1) \cdot 10^{-5} T^{3/2} \text{ cal/mol-deg,}$$

and using the values  $\Gamma \approx 2$ ,  $\kappa_T = 5.95 \times 10^{-13}$  cm<sup>2</sup>/dyne, and  $V_m = 7.1$  cm<sup>3</sup>/mole, we obtain

$$\beta \sim 10^{-9} T + 10^{-11} T^3 + 10^{-10} T^{3/2} \text{ deg}^{-1}.$$

In the first quantitative dilatometric investigations at liquid helium temperatures, the volume jump of tin and lead was measured for the superconducting transition in a magnetic field, as well as the jump of the thermal expansion coefficient of these metals upon passing through the critical temperature.<sup>[2,3]</sup> In very recent years dilatometric techniques have been greatly developed and the electronic component of the thermal expansion coefficient has been determined for several metals.<sup>[4,5]</sup>

The present article presents results obtained from an investigation of the thermal expansion of iron at liquid helium temperatures using an improved version of an earlier technique.<sup>[3]</sup> As previously, the main portion of the apparatus is a bimetallic strip of iron and lead a few meters long which is coiled for the sake of compactness (60 turns, 50 mm in diameter). This helix is very sensitive to relative changes in the length of the constituent metals. The iron is of better than 99.9% purity; the purity of the lead is 99.99%. The lower end of the helix is rigidly fixed, while the

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upper end is suspended permitting free rotation about the helical axis as a result of temperature or pressure changes. The angle of rotation  $\Delta n$  is indicated by a mirror maintained at room temperature in the upper part of the apparatus, and is amplified by a thermal amplifier and galvanometer. The bimetallic helix is enclosed in a sealed case which is filled with helium gas to a pressure of  $\sim 5$  mm Hg and is surrounded by liquid helium.

The described apparatus permitted measurements of the torsion angle of the helix both for temperature changes at constant pressure (thermal expansion) and for pressure changes at constant temperature (isothermal compressibility). The sensitivity of the apparatus had previously been computed from the dimensions of the helix, utilizing the known elastic moduli of iron and lead.<sup>[3,6]</sup> Calibration measurements at room temperature, for which the thermal expansion coefficients  $\alpha$  and the compressibilities  $\kappa$  of the metals are known, yielded the sensitivities

$$S_T = \Delta\alpha(\Delta n / \Delta T)_p^{-1}, \quad S_p = \Delta\kappa(\Delta n / \Delta p)_T^{-1},$$

where  $\Delta\alpha$  and  $\Delta\kappa$  are the differences between the thermal expansion coefficients and the isothermal compressibilities of the metals, and  $\Delta n$  is the scale deflection. The expansion coefficients of the metals have been studied more thoroughly than their compressibilities. In the present case we therefore used the sensitivity derived by calibrating the instrument for thermal expansion:  $S_T = (1.5 \pm 0.1) \times 10^{-9}$ .

In this experiment we measured  $\Delta n$ , characterizing the relative change  $\Delta l/l$  of the sample's length accompanying a temperature change; the derivative with respect to  $T$  gives the thermal expansion coefficient. The use of a bimetallic helix comprises a differential method; the measurements give the difference between the linear thermal expansions of the two metals. We therefore determined the difference between the linear thermal expansion coefficients of lead  $\alpha_{Pb}$  and iron  $\alpha_{Fe}$ :

$$\alpha_{Pb} - \alpha_{Fe} = (\alpha_{Pb}^e - \alpha_{Fe}^e) + (\alpha_{Pb}^l - \alpha_{Fe}^l) - \alpha_{Fe}^s, \quad (4)$$

where the indices  $e$ ,  $l$ , and  $s$  distinguish the electronic, lattice, and spin components of  $\alpha$ .

The measurements were performed in the range 1.5–4.2°K, where there is a very small concentration of normal electrons in lead. Below the critical temperature  $T_C$  of a superconductor the number of normal electrons decreases exponentially with temperature and at 0.5  $T_C$  is only 1/10 of the concentration in the normal state. Therefore in the temperature region of present

interest electrons will make practically no contribution to the thermal expansion of lead. This is the main reason for using a superconductor having a relatively high value of  $T_C$  as one of the metals of the bimetallic helix. In accordance with (2) and (1) the difference between the linear thermal expansion coefficients is given by

$$\alpha_{Pb} - \alpha_{Fe} = -a_{Fe}T + (b_{Pb} - b_{Fe})T^3 - e_{Fe}T^{5/2}. \quad (5)$$

As already noted, at liquid helium temperatures the spin component of the expansion coefficient has the very small value  $\sim 10^{-10}$ , which is considerably below the sensitivity of the instrument. The lattice component of the thermal expansion coefficient of iron is even smaller, being approximately 1/100 of the lattice component for lead, because the Debye temperature of iron is almost five times as large as that of lead. For these experiments we therefore have

$$\alpha_{Pb} - \alpha_{Fe} = -a_{Fe}T + b_{Pb}T^3. \quad (6)$$

In Fig. 1 the measurements of the temperature dependence of  $\Delta l/l$  are given as the number  $n$  of scale divisions, with three experimental runs represented by different symbols. The curve in Fig. 2, derived by differentiating the smooth curve in Fig. 1, with respect to temperature, represents the temperature dependence of the difference between the thermal expansion coefficients of lead and iron in the region 1.5–4.2°K. Since, when treating experimental data on the temperature dependence of the heat capacity of a metal, the small electronic component is isolated by using the coordinates  $C/T$  and  $T^2$ , we follow this procedure in the present case, but only for the smooth curve in Fig. 2.

The same data represented in the coordinates  $(\alpha_{Pb} - \alpha_{Fe})/T$  and  $T^2$  (Fig. 3) yield a straight line, from which the value of  $a_{Fe}$  [the coefficient of  $T$  in (6)] is obtained as the intercept of the extrapolated line on the ordinate axis. In this way we obtain for the electronic component of the

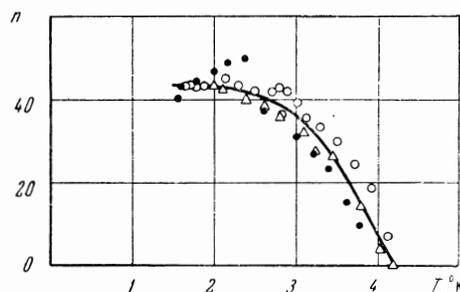


FIG. 1. Relative change of length of iron-lead strip (in scale divisions) vs. temperature. Values obtained in different runs are denoted by different symbols.

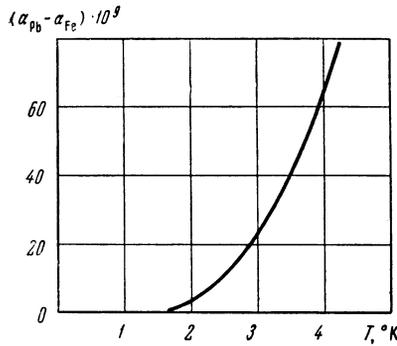


FIG. 2. Difference between the linear thermal expansion coefficients of lead and iron versus temperature.

thermal expansion coefficient of iron:

$$\alpha_{Fe}^e = (3.2 \pm 0.3) \cdot 10^{-9} T \text{ deg}^{-1}.$$

The same value is derived from the fact that the curves in Figs. 2 and 3 cross the temperature axis at 1.62° K, where, according to (6),  $\alpha_{Fe}^e T = b_{Pb} T^3$ ; thus at 1.62° K the electronic component for iron equals the phonon component of the thermal expansion coefficient for lead.

According to the data in<sup>[7,8]</sup> the linear thermal expansion coefficient of superconducting lead is

$$\alpha_{Pb}^1 = (1.3 \pm 0.1) \cdot 10^{-9} T^3 \text{ deg}^{-1}.$$

Therefore at 1.62° K we have

$$\alpha_{Pb}^1 = \alpha_{Fe}^e = 5.53 \cdot 10^{-9} \text{ deg}^{-1},$$

whence

$$\alpha_{Fe}^e / T = \alpha_{Fe} = 3.4 \cdot 10^{-9} \text{ deg}^{-2}.$$

White<sup>[8]</sup> has obtained  $\alpha_{Fe}^e = (2.9 \pm 0.2) \times 10^{-9} T$  for the electronic component of the thermal expansion coefficient of iron. The values of this coefficient obtained by different investigators using different techniques are in good agreement.

The data on the electronic expansion coefficient of iron furnish information regarding the pressure dependence of the electron state density  $\gamma$  at the Fermi surface. The coefficient  $A$  of  $T$  in Eq. (2) for the volume expansion coefficient is

$$3\alpha_{Fe} = A = -V^{-1} \partial \gamma / \partial p. \quad (7)$$

Lifshitz and Kaganov<sup>[9]</sup> have pointed out that  $\gamma$  is related to the shape of the Fermi surface by

$$\gamma = \frac{2\pi^2 k^2}{3(2\pi\hbar)^3} V \oint \frac{ds_F}{v}, \quad (8)$$

where  $ds_F$  is an element of the Fermi surface,  $v$  is the electron velocity on that surface, and  $k$  is Boltzmann's constant. Using (7) and (8), we arrive at

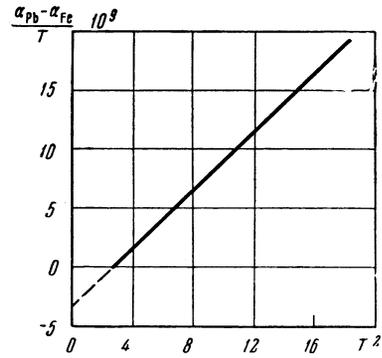


FIG. 3.  $(\alpha_{Pb} - \alpha_{Fe})/T$  versus  $T^2$ . The intercept on the ordinate axis determines the electronic component of the thermal expansion coefficient of iron.

$$L \equiv A \left( \frac{\gamma}{V} \right)^{-1} - \kappa_T = - \left( \oint \frac{ds_F}{v} \right)^{-1} \frac{\partial}{\partial p} \left( \oint \frac{ds_F}{v} \right), \quad (9)$$

where  $\kappa_T = -V^{-1} (\partial V / \partial p)_T$ . The quantity  $L$  has the dimensions of compressibility and can therefore represent the "compressibility" of the Fermi surface. Equation (9) enables us to determine both the magnitude and sign of  $L$  when the magnitude of  $\kappa_T$  is known in this temperature region.

As already stated, the sensitivity of the apparatus enabled us to measure the difference between the compressibilities of lead and iron at liquid helium temperatures as the pressure was varied within the limits 0.1–1.0 atm. The compressibility of lead at these temperatures was calculated using ultrasonic data.<sup>[10]</sup> For the compressibility of iron we obtain  $\kappa_T = 5.7 \times 10^{-13} \text{ cm}^2/\text{dyne}$ . Then  $L = (7.9 \pm 1) \times 10^{-13} \text{ cm}^2/\text{dyne}$ , which is positive, so that the Fermi surface is compressed under increasing pressure.

The Grüneisen constant for an electron gas,<sup>[11]</sup>

$$\Gamma = \beta_e V / C_e \kappa_T,$$

was calculated from our values of  $\beta$  and  $\kappa$  together with the value of  $C_e$  from<sup>[1]</sup>; taking  $V$  at 20° C,<sup>[12]</sup> we obtain for iron  $\Gamma_e = 2.3 \pm 0.3$ , whereas for a phonon gas at high temperatures we have<sup>[13]</sup>

$$\Gamma_p = -d \ln \Theta / d \ln V \approx 2.$$

It is interesting that  $\Gamma_l$  and  $\Gamma_e = -d \ln \gamma / d \ln V$  have close values.

Therefore the Debye temperature, which characterizes the phonon density, and the quantity  $\gamma$ , which determines the electron state density, vary alike for identical relative changes of volume.

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294