# Temperature-dependent part of the resistivity of Al-Cu alloys

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The electrical resistivity of dilute polycrystalline Al-Cu alloys has been investigated in the temperature range 4.2-360 K. The phonon part of the resistivity at low temperatures varies as  $T^n$  where 3 < n < 3.6. The temperature corresponding to the maxima of the deviation from Matthiessen's rule shifts to higher values with the concentration as  $c^{1/5}$ . At high temperatures the deviations have negative slopes.

## I. INTRODUCTION

The old problem of phonon-impurity mixed resistivity of dilute nonmagnetic alloys has become, in the last few years, an interesting subject for the investigation of the scattering mechanisms of conduction electrons. Bass<sup>1</sup> has given a large number of possible scattering mechanisms which can explain the deviations from Matthiessen's rule. Most of them are important only in certain temperature regions, while others apply to specific alloys only.

Measurements of the electrical resistivity of the polycrystalline aluminum-copper alloy system have been made by Caplin and Rizzuto,<sup>2,3</sup> who have investigated the temperature-dependent part of the resistivity of the Al-Cu alloy system containing 0.43-at.% Cu in the temperature range 8-50 K. Their measurements indicate that the phonon resistivity varies as  $T^3$  at low temperatures.

Because they have measured the electrical resistivity only up to 50 K, they have missed the humps of the deviation from Matthiessen's rule and their dependence on the impurity concentration, and also the sign of the deviation at high temperatures.

Our purpose was to investigate the influence of different amounts of Cu impurities on the deviation of Al from Matthiessen's rule in the temperature range 4.2-360 K.

#### **II. EXPERIMENTAL PROCEDURE**

The measurements were performed on polycrystalline cylindrical samples of 0.2-mm diameter and 340-mm length. The samples were pure aluminum (99.9999%) with a residual resistivity ratio of 8500, and aluminum alloyed with 0.025-, 0.050-, 0.075-, 0.10-, 0.20-, 0.30-, 0.40-, 0.50-, and 0.60-at.% Cu. The concentration was determined by chemical analysis with an accuracy of  $\pm 0.005$  wt%. The alloys were in the solid solution region, as evidenced by the residual resistivity which was always proportional to the impurity concentration. The residual resistivity per atomic per cent was 700 n $\Omega$  cm. The calculated value for the Al-Cu system is 810 n $\Omega$  cm/at.%.<sup>4</sup> The specimen wires were annealed for 5–6 h at 500 °C in vacuum (~10<sup>-5</sup> Torr).

The apparatus for the measurements of the electrical resistance has been described previously.<sup>5,6</sup> The main errors in the value of the resistivity can be attributed to the inaccuracy in the determination of the geometry factor  $(\pm 0.2\%)$ .

### **III. EXPERIMENTAL RESULTS**

In order to calculate the deviation  $\Delta(c, T)$ , we must measure the resistivities as a function of temperature for a dilute alloy and for a pure host sample. We have used a pure aluminum sample for which the residual resistivity  $\rho_{p}^{0}$  is so small that no deviation from Matthiessen's rule occurs. The departure from Matthiessen's rule is given by

 $\Delta(c, T) = [\rho_a(T) - \rho_p(T)] - [\rho_a(4.2 \text{ K}) - \rho_p(4.2 \text{ K})],$ 

where  $\rho_a(T)$  and  $\rho_p(T)$  are the phonon-induced resistivities for the alloyed and pure samples, respectively.

In Fig. 1 the relative deviations from Mathiessen's rule,  $\Delta(c, T)/\rho_0$ , for nine Al-Cu alloys are plotted as a function of temperature. At low temperatures,  $\Delta(c, T)/\rho_0$  increases sharply with temperature. At intermediate temperatures the deviations show maxima which shift with the concentration and which become more and more pronounced with decreasing concentration. In the high-temperature region the  $\Delta(c, T)/\rho_0$  are linear in T, but independent of the concentration, and show a weakly negative slope.

The low-temperature behavior of the temperature-dependent part of the electrical resistivities is presented in Fig. 2, where the quantity  $[\rho(T) - \rho_0]/T^3$  is plotted as a function of temperature in a log-log plot.  $\rho(T)$  is the measured resistivity at the temperature *T*. In this figure it

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FIG. 1. Relative deviation from Matthiessen's rule as a function of temperature for Al-Cu alloys.

can be seen that the phonon resistivity cannot be characterized by a single power law. Owing to the experimental errors of the electrical resistance in the temperature region 6–10 K we cannot define with accuracy the exponent of T according to which the quantity  $\rho(T) - \rho_0$  varies. As the temperature increases  $\rho(T) - \rho_0$  goes over to a  $T^3$  plateau which disappears as the concentration of the impurities increases. In the range 24–60 K the phonon part of the resistivity varies approximately as  $T^{3.6}$  for all alloy concentrations. In contradiction to the present measurements Caplin and Rizzuto<sup>2,3</sup> found for their Al-Cu alloy containing 0.43-at.% Cu, in the region 8–50 K, a  $T^3$  dependence.

In Fig. 3 the dependence of  $\rho(T) - \rho_0$  on the residual resistivity  $\rho_0$  is shown for five different



FIG. 2. Variation with temperature of  $[\rho(T) - \rho_0] T^3$  for Al-Cu alloys.

fixed temperatures. The behavior as a function of impurity content is apparently similar to that obtained for other aluminum alloys.<sup>7-9</sup> The isotherms at 75 and 50 K show well-defined "pure" and "dirty" regions, characterized by  $\rho_0$  independent of  $\ln\rho_0$  behavior, respectively. For the three lowest isotherms (30, 20, and 14 K) the samples are always in the "dirty" region and the logarithmic dependence of  $\rho(T) - \rho_0$  on  $\rho_0$  extends over about two decades in  $\rho_0$ .



FIG. 3. Temperature-dependent part of the resistivity,  $\rho(T) - \rho_0$ , as a function of the residual resistivity  $\rho_0$  for five representative temperatures.

A large number of reports have been published on the dependence of the phonon resistivity on the temperature and the residual resistivity. In most of these reports the authors point out the importance of the umklapp scattering processes to the resistivity. The different calculations predict different temperature and concentration dependence at low temperatures. Detailed investigations of the contribution of the umklapp processes to the phonon resistivity have been published by Lawrence and Wilkins,<sup>10</sup> Dosdale and Morgan,<sup>11</sup> Bergman et al.,<sup>12</sup> and Trofimenkoff and Bhatia.<sup>13</sup> Lawrence and Wilkins found that above some characteristic temperature  $\Theta_1$ , which is on the order of  $0.01\Theta_D$  to  $0.05\Theta_D$  (for Al,  $\Theta_D = 395$  K), the temperature-dependent part of the electrical resistivity of the dirty polyvalent metals due to the umklapp electron-phonon scattering cannot be characterized by a specific power of T. The umklapp contribution is broken in two terms which are proportional to  $T^2$  and  $T^4$ . Dosdale and Morgan<sup>11</sup> using a multiband model found that the temperature-dependent resistivity of aluminum in the dirty limit shows a  $T^{3.5}$  dependence. In their calculation they have used transverse umklapp processes which were defined differently from that of the Lawrence and Wilkins calculation.

An extension of the work of Lawrence and Wilkins is the calculation of Bergman *et al.*,<sup>12</sup> who used a new trial function that takes proper account of the anisotropy of the Fermi surface, and also a two-orthogonalized-plane-wave function for both normal and umklapp processes. The calculation of Bergman *et al.* gave nearly a  $T^3$  law for the phonon resistivity of aluminum for temperatures less than about 10 K and a logarithmic variation of  $\rho(T) - \rho_0$  over more than two orders of magnitude in  $\rho_0$ .

Trofimenkoff and Bhatia also attempted to explain the dependence of  $\rho(T) - \rho_0$  on  $\rho_0$  by anisotropic umklapp electron-phonon scattering and isotropic impurity and normal phonon scattering. The result of this calculation shows that  $\rho(T) - \rho_0$ has approximately a  $\ln \rho_0$  dependence of about three orders of magnitude in  $\rho_0$ .

In the temperature range where the maximum of the deviations lies, the position of the maximum shifts towards higher temperatures with increasing concentration (Fig. 1). In Fig. 4 the maximum temperatures  $T_{\rm max}$  are plotted in a log-log plot against the Cu concentration. It can be seen that  $T_{\rm max}$  is proportional to  $c^{1/5}$ , as it was also found for<sup>5</sup> Al-Ge and<sup>6</sup> Al-Ga alloys. This behavior was predicted first by Kagan and Zhernov.<sup>14</sup> The cause for the Kagan-Zhernov  $c^{1/5}$  law is the change in the



FIG. 4.  $T_{max}$  against solute concentration.

anisotropy of the electron distribution function  $f(\mathbf{p}, n)$ . In the case of a spherical Fermi surface, the anisotropy of the distribution function is strongly pronounced. This is due to the anisotropic character of the electron scattering associated mainly with the umklapp processes in the electron-phonon interaction and the anisotropy of the phonon spectrum.

We now confine our discussion to the high-temperature part of the deviation. There are two different calculations which attempt to explain the deviation in this temperature region. The interference effect, proposed by Kagan and Zhernov.14,15 predicts that  $\Delta(c,T)/\rho_0$  will be proportional to T and that the sign of the derivative  $\rho_0^{-1} d\Delta(c,T)/dT$ will depend on the sign of the difference of the valence of the impurity from that the host atoms. Bhatia and Gupta<sup>16</sup> predict that the interference term should lead to a linear dependence of  $\Delta(c,T)/2$  $\rho_0$  on T and that the deviation should change sign when the solvent and solute atoms are interchanged. The present results for Al-Cu alloys are in good agreement with the Kagan and Zhernov calculation.

#### V. CONCLUSION

The present measurements on the Al-Cu alloys show that below 50 K the electrical resistivity varies with a not well-defined power law. The temperature corresponding to the maximum of the deviations varies proportionally to  $c^{1/5}$ . This behavior can be explained with the Kagan and Zhernov calculation. The high-temperature part of the deviation is proportional to T and has a weakly negative slope.

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