Strong anisotropic electron-impurity scattering in dilute *Al*Li alloys

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(Received 27 January 1992; revised manuscript received 13 April 1992)

The Hall coefficient and the transverse magnetoresistance of dilute *Al*Li alloys at 4.2 K is reported. A large positive low-field Hall coefficient R_H^0 is found, in comparison to the free-electron Hall coefficient-value R_{FE} of aluminum. The cause of the large R_H^0 can be attributed to the strong anisotropic scattering of the electrons in the third zone of Fermi surface on the substitutional Li impurities.

I. INTRODUCTION

Theoretical calculations of orbitally averaged electron-impurity scattering rates in dilute *Al*Li alloys[1,2] have shown that there is a large anisotropy of the Dingle temperature X_D for α [100], β [100], γ [100], and ξ [100] external orbits of the third-zone electron sheet.

On the other hand, de Haas–van Alphen (dHvA) experiments in dilute *Al*Li alloys[3] indicate that the Dingle temperatures, and hence the relaxation times, for α, β, and γ orbits for the electrons on the third-zone surface, are approximately isotropic.

It should be noted that dHvA experiments on the nearly-free-electron polyvalent metals, like Al, encounter difficulties due to the simultaneous involvement of different orbits, beat effects of neighboring frequencies,[4] and magnetic breakdown.[5]

In our opinion the discrepancy between theoretical calculations and experimental data can be accounted for by these difficulties. Therefore, for nearly-free-electron metals, it appears to be better to use the transport properties in order to obtain information about the anisotropy of the corresponding relaxation time $\tau(\mathbf{k})$. The best candidates for such measurements are the low-field Hall coefficient and the low-field magnetoresistance.

Aluminum, being trivalent, has a roughly spherical Fermi surface (FS) with corresponding slightly negative curvature, but there are strong deviations from the sphere where it intersects Brillouin-zone boundaries. Thus the aluminum FS can be divided into three characteristically different regions. These are (i) a free-electron-like portion in the second zone, which covers most of the FS and is assumed to be characterized by a mean free path l_-; (ii) holelike cylinders, also in the second zone just below the Brillouin-zone boundaries characterized by a mean free path l_{++}; and (iii) electron-like cylinders in the third zone characterized by a mean free path l_{--}.

Under the assumption that the mean free paths l_-, l_{++}, and l_{--}, are constant over their FS areas, Kesternich,[6,7] using this three-group model, has derived the following expressions for the low-field magnetoresistance and Hall effect, i.e.,

$$R_H^0 = R_{\mathrm{FE}}\left[1+1.87\left[\frac{l_{--}^2-l_{++}^2}{l_-^2}\right]\right], \tag{1}$$

$$\frac{\Delta\rho}{\rho_0} = 50\left[R_{\mathrm{FE}}\frac{B}{\rho_0}\right]^2\left[\frac{l_{--}^3+l_{++}^3}{l_-^3}\right], \tag{2}$$

where $R_{\mathrm{FE}} = -3.47\times10^{-11}$ m^{3}C^{-1}, ρ_0 is the residual resistivity at zero magnetic field, and H is the applied magnetic field.

Using these two equations, one can determine the ratios l_{--}/l_- and l_{++}/l_- and, hence, the anisotropy factor l_{--}/l_{++}.

The purpose of the present investigation is to determine the anisotropy factor l_{--}/l_{++} from simultaneous low-field magnetoresistivity and Hall-effect measurements in a series of *Al*Li alloys and to compare it with the theoretical calculations.

II. EXPERIMENTAL PROCEDURE

The master *Al*Li alloy containing 5 at. % Li was prepared by melting and casting 99.9999% pure Al and 99.9% pure Li in a glove box under an argon atmosphere and using a crucible made of boron nitride (BN). The ingot was homogenized near its solidus temperature. From the master alloy the dilute alloys were made in an induction furnace also using a BN crucible. The new ingots were cold rolled to a thickness of 100 μm and the samples were cut with a special punching tool to the appropriate shape. After cutting, the samples were annealed at 450 °C for 4 h. The same instrumentation and d.c. techniques were used as before.[8]

The concentration of Li ranged from 0.011 to 0.54 at. % and was determined by atomic absorption spectroscopy. Figure 1 shows the residual resistivity $\Delta\rho=\rho_{\mathrm{alloy}}-\rho_{\mathrm{Al}}$ as a function of lithium concentration at 4.2 and 77 K, respectively. The residual resistivity per atomic percent of Li at 4.2 K is $33\times10^{-8}\,\Omega$ cm, while at 77 K it is $71\times10^{-8}\,\Omega$ cm. Shown for comparison also in Fig. 1 are the measurements performed at 77 K, of Ceresara *et al.*,[9] who found a value of $80\times10^{-8}\,\Omega$ cm. The difference between the values of the residual resistivities at 4.2 and 77 K is due to deviations from the Mathiessen rule,[10] while the discrepancy between the value of the present study at 77 K and that of Ceresara is probably due to inaccuracies in the determination of Li concentration.

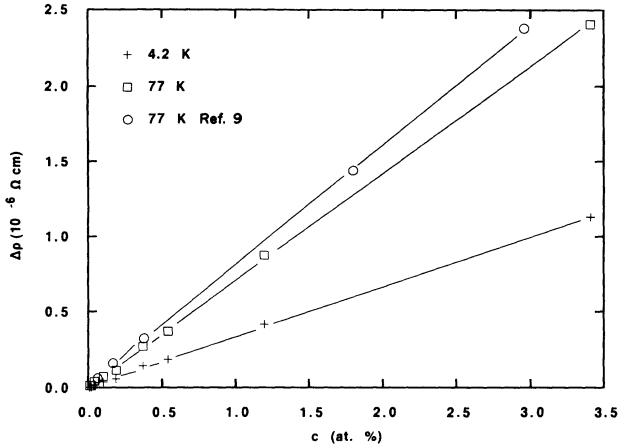

FIG. 1. The residual resistivity $\Delta\rho$ of *Al*Li alloys as a function of Li concentration.

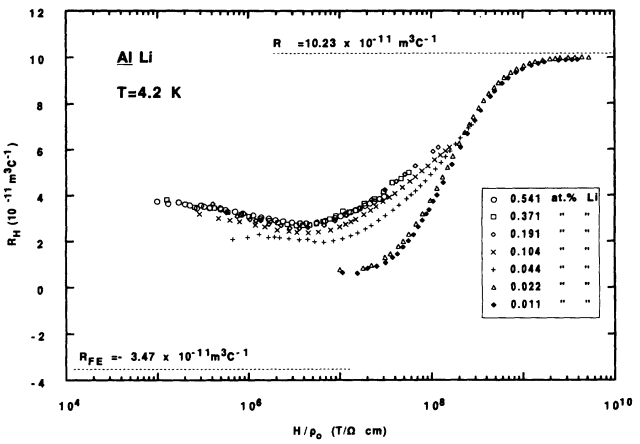

FIG. 3. The Hall coefficient R_H of *Al*Li alloys as a function of magnetic field in the Köhler representation.

III. RESULTS AND DISCUSSION

Figures 2 and 3 show the relative magnetoresistivity $\Delta\rho/\rho_0$ and the Hall coefficient R_H and 4.2 K, respectively, in the Köhler representation, i.e., plotted as a function of "effective" magnetic field H/ρ_0, where ρ_0 is the residual resistivity at zero magnetic field. The figures show that (i) the Köhler rule holds as well as the magnetoresistivity and the Hall coefficient for the alloys with a Li concentration higher than 0.1 at. %, while for the alloys with a Li content smaller than 0.1 at. % there is a deviation from the Köhler rule; (ii) at low magnetic fields, $\Delta\rho/\rho_0$ depends nearly quadratically on H/ρ_0; and (iii) at low fields the Hall coefficient R_H^0 exhibits very high positive values in comparison to the isotropic free-electron-value Hall coefficient $R_{\rm FE}=-3.47\times10^{-11}$ m^{3}C^{-1}. At high magnetic fields the Hall coefficient agrees with the theoretical value $R_\infty=10.2\times10^{-11}$ m^{3}C^{-1}. At low fields the R_H^0 depends weakly on H and goes through a minimum for alloys with a concentration higher than 0.1 at. % Li and is nearly constant for the alloys with a Li content smaller than 0.1 at. %.

Figure 4 shows the dependence of the low-field Hall coefficient R_H^0 as a function of Li concentration at a constant "effective" field H/ρ_0. R_H^0 increases sharply and saturates rapidly to a nearly constant value, which is independent of the concentration. This initial increase of R_H^0 is due to residual impurities, grain boundaries, or dislocations that were made during mounting and cooling of the samples. The saturation behavior of R_H^0 indicates that the FS can always be regarded as sufficiently constant for our purpose. Namely, if the FS would have been disturbed by the presence of the impurities, then these distortions would cause a monotonic increase of R_H^0 with concentration. Consequently, if the positive values of R_H^0 would be caused by these distortions, then a saturation of R_H0 would be impossible.

Using a fixed value for H/ρ_0, namely 10^6 T/(Ω cm), the corresponding values of $\Delta\rho/\rho_0$ and R_H^0, Eqs. (1) and (2), and the anisotropy of the mean free path expressed by the ratios l_{--}/l_- and l_{++}/l_- can be calculated as a function of Li concentration. The saturation values of l_{++}/l_- and l_{--}/l_- are 1.47 and 1.09, respectively.

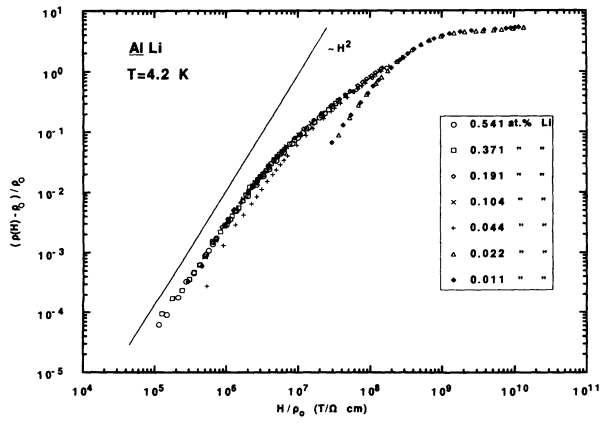

FIG. 2. The relative magnetoresistivity $\Delta\rho/\rho_0$ of *Al*Li alloys as a function of magnetic field in the Köhler representation.

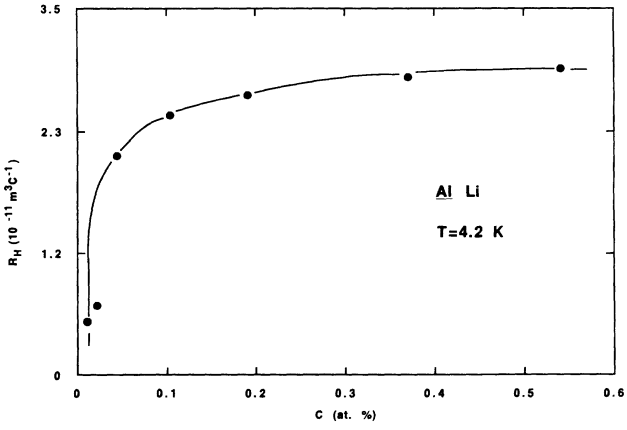

FIG. 4. The low-field Hall coefficient R_H^0 as a function of Li concentration.

From these ratios an anisotropy factor $l_{--}/l_{++}=0.75$ follows. In order to examine the dependence of the above anisotropy factor on the "effective" magnetic field, two additional calculations were performed for $H/\rho_0=4\times10^5$ and 2×10^6 T/(Ω cm). The resulting anisotropy factor l_{--}/l_{++} are 0.76 and 0.75, respectively. The mean value of the above three anisotropy factors is $\langle l_{--}/l_{++}\rangle=0.75$. The ratios l_{--}/l_{-} and l_{++}/l_{-}, as a function of Li concentration for the three values of "effective" fields, are shown in Fig. 5, which shows that $l_{++}/l_{-}>l_{--}/l_{-}$, i.e., $l_{++}>l_{-}$. This analysis shows that the third-zone electrons have a transport relaxation time considerably smaller than that of the second-zone electrons, which means that the Li impurities prefer to scatter the electrons in the third zone rather than those in the second zone.

In order to compare this anisotropy transport factor with the theoretical value,[1-3] we take the mean values of the calculated Dingle temperatures X_D for the α, β, and γ orbits on the third-zone arm of the FS and for the ψ orbits of the second zone. The ratio between $\langle X_D^{2\text{-zone}}\rangle$ and $\langle X_D^{3\text{-zone}}\rangle$ gives

$$\frac{\langle X_D^{2\text{-zone}}\rangle}{\langle X_D^{3\text{-zone}}\rangle}=0.76\ .$$

It can be seen that there is surprising agreement between the transport anisotropy factor and that derived from the mean values of the Dingle temperatures. If this result is not accidental, then it is remarkable that the difference between the ratio $(\tau^{3\text{-zone}}/\tau^{2\text{-zone}})_{\text{tr}}$ and $(\tau^{3\text{-zone}}/\tau^{2\text{-zone}})_{\text{dHvA}}$ is small, even though the definitions of the relaxation times τ_{tr} and τ_{dHvA} are very different.

The physical origin of the observed anisotropy of the transport relaxation time $\tau_{\mathbf{k}}$ arises from the corresponding differences in the electron wave functions $\psi_{\mathbf{k}}(\mathbf{r})$ and the scattering potential of the defects. On the free-electron-like parts of the FS the wave function $\psi_{-}(\mathbf{r})$ is a plane wave. At the edges, the mixing of two plane waves results partly in standing waves, the amplitude of which is greatest at interstitial positions for the edges in the second zone $[|\psi_{++}(\mathbf{r})|^2]$ and at lattice positions for the edges in the third zone $[|\psi_{--}(\mathbf{r})|^2]$. The local charge densities obtained from four orthogonalized-plane-wave calculations[11] confirm this general characterization of ψ_{-}, ψ_{++}, and ψ_{--}. Hence the electrons on the holelike edges in the second zone will be preferentially scattered at defects near interstitial positions, and the electrons on the electronlike edges in the third zone will be preferentially scattered at defects near lattice positions. On the other hand, the electrons on the free-electron-like FS regions will be scattered with similar probability on both types of defects. The Li impurities in the solid solution state (α phase) take substitutional positions in the aluminum lattice and, therefore, according to the above considerations, they will strongly scatter the conduction electrons in the third-zone, while relatively little scattering will occur from electrons in the corners of the second-zone.

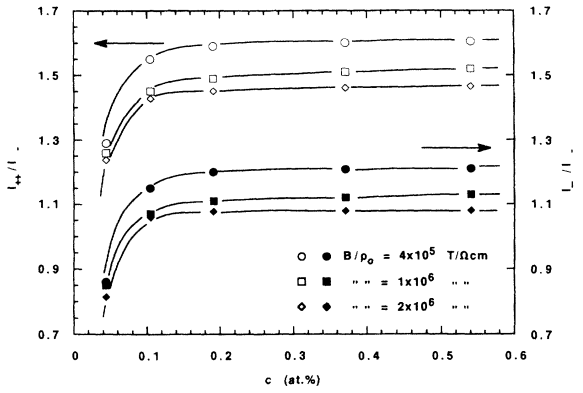

FIG. 5. The anisotropy ratios of the mean free paths, l_{--}/l_{-}, and l_{++}/l_{-}, as a function of Li concentration.

In the present discussion we do not consider the influence of the small-angle scattering, because, as diffuse elastic neutron scattering[12] has shown, the displacement field around the substitutional Li atom is negligibly small. This implies that the magnetoresistance and Hall effect are essentially unaffected by small-angle scattering and so we do not expect a change of l_{--} and l_{++}.

IV. CONCLUSIONS

We have reported measurements of the transverse magnetoresistivity $\Delta\rho/\rho_0$ and the low-field Hall coefficient R_H^0 for a series of dilute AlLi alloys at 4.2 K. A three-group model for the mean free paths has been used to interpret the results. The numerical value of the transport anisotropy factor l_{--}/l_{++}, which is computed from the experimental data, is in very good agreement with the theoretical value calculated from the mean value of the Dingle temperatures.

[1]L. Cole, W. E. Lawrence, R. Monnier, W. Van der Mark, and M. Manninen, Phys. Rev. Lett. **42**, 1174 (1979).
[2]W. Joss, W. Van der Mark, R. Monnier, L. Cole, W. E. Lawrence, and J. Deutz, Helv. Phys. Acta **55**, 736 (1982).
[3]L. Cole and W. E. Lawrence, Phys. Rev. B **30**, 4796 (1984).
[4]J. P. G. Shephard and W. L. Gordon, Phys. Rev. **169**, 541 (1968).
[5]W. Kesternich and C. Papastaikoudis, Phys. Status Solidi B **64**, K41 (1974).
[6]W. Kesternich, Phys. Rev. B **13**, 4227 (1976).
[7]W. Kesternich, H. Ullmaier, and W. Schilling, J. Phys. F **6**, 1867 (1976).
[8]C. Papastaikoudis, D. Papadimitropoulos, and E. Rocofyllou, Phys. Rev. B **22**, 2670 (1980).
[9]S. Ceresara, A. Giarda, and A. Sanchez, Philos. Mag. **35**, 97 (1977).
[10]N. Boukos and C. Papastaikoudis (unpublished).
[11]K. Böning, K. Pföndner, P. Rosner, and M. Schlüter, J. Phys. F **5**, 1176 (1975).
[12]G. Solt and K. Werner, Phys. Rev. B **24**, 817 (1981).