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1996 J. Phys.: Condens. Matter 8 L399

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LETTER TO THE EDITOR

## The size effect on electrical resistivity in single crystals of dilute Al alloys

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Received 7 May 1996

**Abstract.** In order to study how the scattering anisotropy influences the dc size effect, measurements have been made on the orientation dependence of the size effect in dilute Al–Ag and Al–Si alloys at 4.2 K. The solute Ag decreases the relaxation time near the zone boundaries, while Si has a reverse effect. The surface orientations of the specimens were set parallel to the crystallographic planes {100} and {110} in the light of the anisotropic size effect found in pure Al. The axis orientation of the specimens (direction of current flow) was  $\langle 110 \rangle$ . The results show that the size effect in these specimens can be well described by means of the Fuchs–Sondheimer theory, with the specular parameter  $p = 0$  and the product of bulk resistivity and bulk mean free path  $\rho_b \ell_b = 0.82 \text{ f}\Omega \text{ m}^2$ . This is contrary to the theoretical prediction that the size effect in Al is sensitive to the anisotropy of the relaxation time due to the impurities.

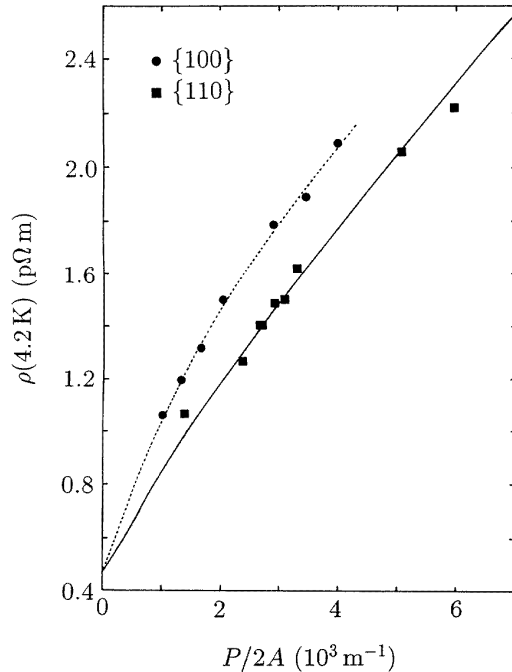
The anisotropy of the dc size effect in Al has drawn a considerable amount of interest since the first measurement made by Risnes and Sollien [1]. However, the origin of the anisotropy still has not been clarified. Figure 1 shows an example of the anisotropic size effect in high-purity Al (RRR  $\simeq 50\,000$ ), obtained in our recent experiments: the size effect due to the {100} surface is significantly larger than that due to the {110} surface.

Sato and Yonemitsu [2], the first to emphasize that the size effect is very sensitive to the scattering anisotropy, calculated that even the anisotropic relaxation time of the isolated Ag in Al brings about stronger anisotropy as shown in figure 1. This is worthy of attention, but not acceptable without experimental confirmation. In this letter, we therefore present an experimental study of the size effect in dilute Al–Ag and Al–Si alloys. The solute Ag in Al decreases the relaxation time near the zone boundaries, while Si has a reverse effect.

Specimens were prepared from two single-crystal rods of dilute Al–Ag and Al–Si alloys, grown by repeating the zone-levelling procedure ten times.

As a material for these alloys, high-purity Al (RRR  $\simeq 15\,000$ ) was used. Two sets of single-crystal plates, about 0.2–1.6 mm thick, were first cut from small parts of each rod by spark erosion: one was parallel to the crystallographic plane {100} and another was parallel to the {110} plane. Then they were cut into straight strips approximately 3 mm by 20 mm as the specimen for the resistance measurement. The axis orientation of the specimens (direction of current flow) was set parallel to  $\langle 110 \rangle$ . After cutting, the specimens were etched with aqua regia and rinsed with distilled water to remove contaminations and damaged layers on the surface.

The crystallographic orientation of the specimens was determined by the transmission Laue method to an accuracy of  $\pm 1^\circ$ . Hereafter the following abbreviations are used: {100} $\langle 110 \rangle$  indicates that the surface orientation of the specimen is {100} and the direction of current flow is  $\langle 110 \rangle$ , and so on.



**Figure 1.** Experimental values of the residual resistivity at 4.2 K,  $\rho(4.2 \text{ K})$ , plotted as a function of the inverse of reduced thickness,  $P/2A$ , for high-purity Al single crystals (RRR  $\simeq 50\,000$ ) with the {100} surface and with the {110} surface. The direction of current flow is  $\langle 110 \rangle$ . The solid curve is the prediction based on the Fuchs–Sondheimer theory with the specular parameter  $p = 0$ , using the product of bulk residual resistivity and bulk mean free path  $\rho_b \ell_b = 0.82 \text{ f}\Omega \text{ m}^2$ . The dotted curve is a spline fitted to the data.

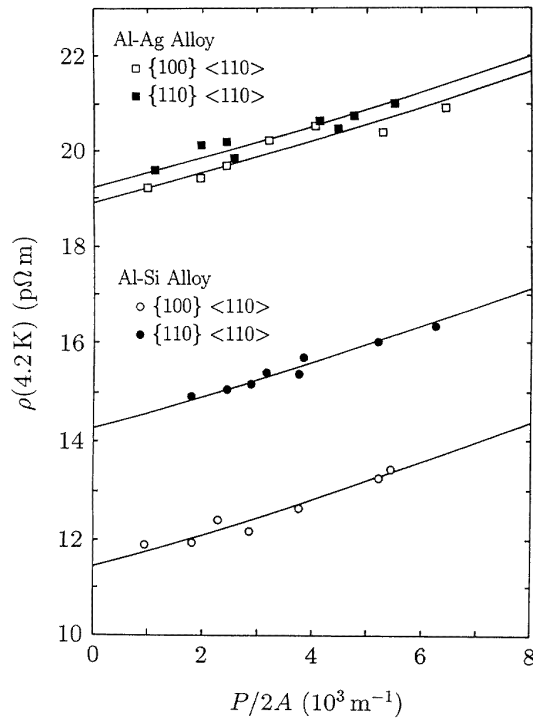
Two pairs of electrodes, 0.3 mm thick wires made of each alloy, were spot-welded on the specimen for the standard dc four-probe method. The potential electrodes were placed far inside to avoid the end effect. All the specimens thus prepared were then annealed in air at 580 °C for 3 h and furnace-cooled to room temperature.

The resistance measurement at 4.2 K was made with a superconducting chopper amplifier with a voltage sensitivity of  $\pm 2 \text{ pV}$  [3]. The resistivity of the specimens at 4.2 K,  $\rho(4.2 \text{ K})$ , was determined from the relation  $\rho(4.2 \text{ K}) = [\rho(300 \text{ K})/R(300 \text{ K})]R(4.2 \text{ K})$ , where  $R(4.2 \text{ K})$  and  $R(300 \text{ K})$  are resistances measured at 4.2 K and 300 K, respectively, and  $\rho(300 \text{ K})$  the resistivity at 300 K:  $\rho(300 \text{ K}) = 27.36 \pm 0.05$  and  $27.38 \pm 0.05 \text{ n}\Omega \text{ m}$  for the Al–Ag and Al–Si alloys, respectively. The resistance at 300 K was measured with a dc comparator potentiometer with a sensitivity of  $\pm 0.5 \text{ nV}$  (Guildline, model 9930).

The thickness  $d$  of the specimens was determined from the relation  $d = (\ell_p/w)[\rho(300 \text{ K})/R(300 \text{ K})]$ . The width  $w$  of the specimen and the distance  $\ell_p$  between the potential electrodes were measured with a travelling microscope with a resolution of 0.01 mm.

Figure 2 shows  $\rho(4.2 \text{ K})$  of Al–Ag and Al–Si alloys, plotted as a function of  $P/2A$  for each of the crystallographic orientations {100}⟨110⟩ and {110}⟨110⟩, where  $P$  is the perimeter of a cross section and  $A$  the sectional area of a specimen. Solid curves in the figure are drawn according to the Fuchs–Sondheimer theory [4], assuming that the specular parameter  $p = 0$ , and that the product of bulk resistivity and bulk mean free path  $\rho_b \ell_b = 0.82 \text{ f}\Omega \text{ m}^2$  [5, 6]. As the specimens have been etched with aqua regia and annealed in air, the specimen surface is rather rough. Therefore, the surface scattering of conduction electrons is reasonably assumed to be diffuse, i.e.,  $p = 0$  [5]. Thus obtained, each of these curves gives a good fit to the data.

The intercept of the curve with the ordinate axis is regarded as the mean bulk resistivity



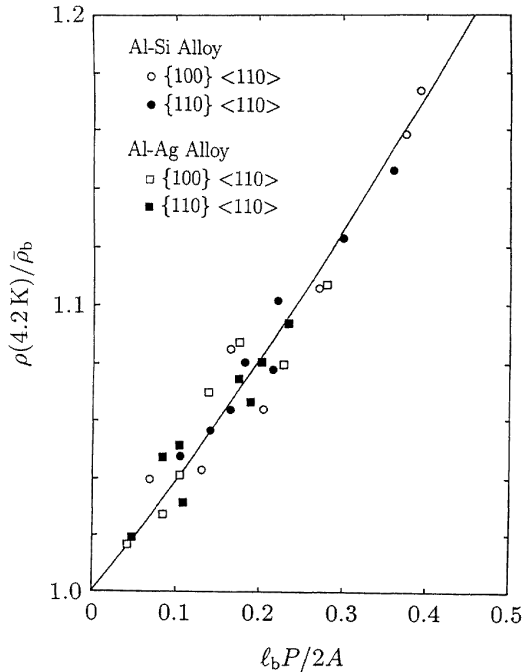
**Figure 2.** Experimental values of the residual resistivity  $\rho(4.2 \text{ K})$  of single crystals of dilute Al–Ag and Al–Si alloys at 4.2 K, plotted as a function of  $P/2A$  for each of the crystallographic orientations  $\{100\}\langle 110\rangle$  and  $\{110\}\langle 110\rangle$ . The curves are predictions based on the Fuchs–Sondheimer theory with  $p = 0$ , using  $\rho_b l_b = 0.82 \text{ f}\Omega \text{ m}^2$ .

$\bar{\rho}_b$  of the specimens prepared into a set of the same crystallographic orientation. The difference in  $\bar{\rho}_b$  in each alloy is attributed to an inhomogeneous distribution of solute atoms along the rod axis, which still remained even after repeating the zone-levelling procedure. Then the possibility exists that the inhomogeneous solute distribution could give rise to systematic errors in the measurements for each specimen set, cut from close parts of the rod. To check this possibility, the thicker specimens were reduced in thickness by spark erosion, and the resistivity was measured again. The results obtained in this manner showed no significant effect of the solute distribution.

In order to compare the experimental results with each other, the plots of  $\rho(4.2 \text{ K})/\bar{\rho}_b$  versus  $l_b P/2A$  are shown in figure 3. Here, the mean free path  $l_b$  was determined from  $\bar{\rho}_b l_b = 0.82 \text{ f}\Omega \text{ m}^2$ . Clearly, all sets of data are well fitted to the Fuchs–Sondheimer theory, with  $p = 0$ , and any orientation dependence of the size effect is less than the experimental error. As already noted, the solute Ag in Al decreases the relaxation time near the zone boundaries, while Si has a reverse effect. Consequently, the anisotropy of the relaxation time due to the impurities is not the intrinsic origin of the anisotropic size effect in Al.

It is generally accepted that scattering of conduction electrons from impurities makes the electron distribution function isotropic [7]. This is invoked as a possible explanation for the disappearance of the anisotropy of the size effect, caused by the addition of the impurities Ag and Si. In such a situation, the Fuchs–Sondheimer theory with a suitable parameter  $\rho_b l_b$  must be a good approximation since the Fermi surface in Al corresponds rather closely to the nearly free electron model.

Conversely, the effect of anisotropy of distribution function will lead to the anisotropic size effect in high-purity Al as shown in figure 1. Important departures from an isotropic distribution are expected around the  $\langle 100\rangle$  direction; in that direction the second-zone Fermi



**Figure 3.** Experimental  $\rho(4.2 \text{ K})/\bar{\rho}_b$  values of single crystals of dilute Al–Ag and Al–Si alloys at 4.2 K, plotted as a function of  $\ell_b P/2A$  for each of the crystallographic orientations  $\{100\}\langle 110\rangle$  and  $\{110\}\langle 110\rangle$ . The solid curve is the prediction based on the Fuchs–Sondheimer theory with  $p = 0$ .

surface is nearest to the zone boundary. In the  $\langle 110\rangle$  direction, on the other hand, it seems reasonable to expect that the effect of anisotropic distribution function is less pronounced since the Fermi surface is more distant from the zone boundary. In fact, the experimental data for the  $\{110\}$  surface are well fitted to the Fuchs–Sondheimer theory with  $p = 0$ , using  $\rho_b \ell_b = 0.82 \text{ f}\Omega \text{ m}^2$ , but for the  $\{100\}$  surface the Fuchs–Sondheimer theory is not applicable any longer.

Finally we briefly mention the parameter  $\rho_b \ell_b$ . As we have found above, our value is  $0.82 \text{ f}\Omega \text{ m}^2$ . This should correspond to the minimum value for  $\rho_b \ell_b$  which can be derived by applying the Fuchs–Sondheimer theory with  $p = 0$ . For instance, the experimental data for the  $\{100\}$  surface in figure 1 shows a tendency that  $\rho_b \ell_b$  decreases towards  $\sim 0.8 \text{ f}\Omega \text{ m}^2$  with decreasing thickness, when the Fuchs–Sondheimer theory is enforced. This may correspond to a situation where the distribution function becomes isotropic due to the increased surface scattering. Our  $\rho_b \ell_b$  value is, however, significantly larger than the free-electron prediction of  $0.48 \text{ f}\Omega \text{ m}^2$ . Unfortunately, this discrepancy still is not clear. To make significant advances in our understanding of the dc size effect, further studies, both experimental and theoretical, are required.

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