Specular Boundary Scattering and Electrical Transport in Single-Crystal Thin Films of $CoSi_2$

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Resistivity measurements on epitaxial, single-crystal films of $CoSi_2$ grown on Si show little dependence on film thickness down to ~60 Å. This dimension is much less than the bulk transport scattering length of ~1000 Å as determined from magnetoresistance measurements at liquid-He temperatures. Boundary scattering of the carriers is therefore essentially specular in this system.

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The question of what happens to the resistivity in metallic films as they become thinner was first discussed nearly a century ago by Thomson.[1] It was predicted that when the thickness d becomes comparable to or less than the electrons' bulk scattering length l the conductivity will be dominated by surface scattering and there will be a "size effect." This behavior is only true if the scattering is diffuse; if the scattering were specular the influence of the boundaries would be correspondingly lessened or eliminated. These ideas were first incorporated in a unified fashion in a transport theory by Fuchs[2] (in the 1930's). As simple as the concepts may be, experimental progress in this subject has been slow. The problem has mainly to do with the films' growth and characterization as stressed in a recent critique by Sambles.[3] It is difficult to grow thin, continuous, and defect-free metallic films with good interfaces. Moreover, even in the best films grain-boundary scattering is a serious stumbling block in the determination[3] of size effects and specularity. In this Letter we present results of transport studies in a new system, epitaxial *single-crystal* films of $CoSi_2$ (a metal) grown on Si, which largely eliminate these difficulties and reveal a size effect that is strikingly small. This demonstrates unambiguously a high degree of specularity and therefore a high degree of phase coherence in transport in this unique system.

The experiments are made possible by recent developments[4] in the growth of single-crystal thin films of $CoSi_2$ and $NiSi_2$ on Si. Among the class of metal silicides these two are especially amenable to epitaxial growth on Si because of their (cubic) fluorite structure and close lattice match to Si. We have previously established[5] that the scattering lengths in electrical transport in $CoSi_2$ are considerably larger than in $NiSi_2$ making $CoSi_2$ more suitable for size-effect determinations. The $CoSi_2$ samples used were of two kinds: (1) thin samples, ~60 to ~500 Å, prepared by UHV deposition of Co on atomically clean, n-type Si(111) wafers followed by an anneal; (2) thick samples, ~1000 Å, prepared under somewhat less stringent conditions [Co deposition at $\sim 10^{-6}$ Torr on chemically cleaned (111) and (100) wafers followed by an anneal at 10^{-7} Torr]. In the former case the films are single-crystal having the type-B orientation[4]; in the latter the films were polycrystalline with grain sizes > 1000 Å. Film thicknesses and stoichiometry were determined to an accuracy of ~5% by Rutherford backscattering of 2-MeV ^{4}He ions. Transmission electron microscopy[4] reveals the silicide/Si interface in the UHV samples to be nearly atomically perfect. The free surface is extremely smooth but not perfect on the same scale.

Transport measurements were made on specimens in the form of a standard six-leg bridge prepared by photolithographic patterning and chemical etching, ensuring thereby a precisely defined geometry for Hall and conductivity measurements.[5] Low values of currents were employed (less than a few millivolts voltage drop across the sample) to obviate nonlinear effects such as heating.

The low-temperature bulk scattering length l_e, which sets the length scale for the problem, was determined in an independent experiment from magnetoresistance measurements on "thick" samples (> 1000 Å). In a metal the band structure is usually of sufficient complexity to preclude the determination of l_e by measurements of Hall effect and resistivity ρ. However, in the case of $CoSi_2$, measurements of the "low-field" magnetoresistance $\Delta\rho/\rho_0$ permit a reasonably direct estimate of l_e to be made. [A single determination is sufficient as nearly identical values of residual resistivity $\rho_0 \approx 2.6\ \mu\Omega$ cm are seen for all our samples (of thickness > 200 Å) thus implying that l_e is essentially sample independent.] The existence of a magnetoresistance as Fig. 1 shows requires at least two bands. In general $\Delta\rho/\rho$ is an even function of the field H, and so at the lowest fields it is quadratic ($\Delta\rho_2/\rho$). With increasing H higher-order contributions appear —fourth-order, $\Delta\rho_4\rho$ (see Fig. 1), and so on—until $\Delta\rho/\rho$ eventually saturates, barring compensation (equal numbers of holes and electrons) and/or open Fermi surfaces. Adopting two bands as a "minimum" model[6] and assuming that they can be characterized simply in terms of isotropic effective masses[7] m_1 and

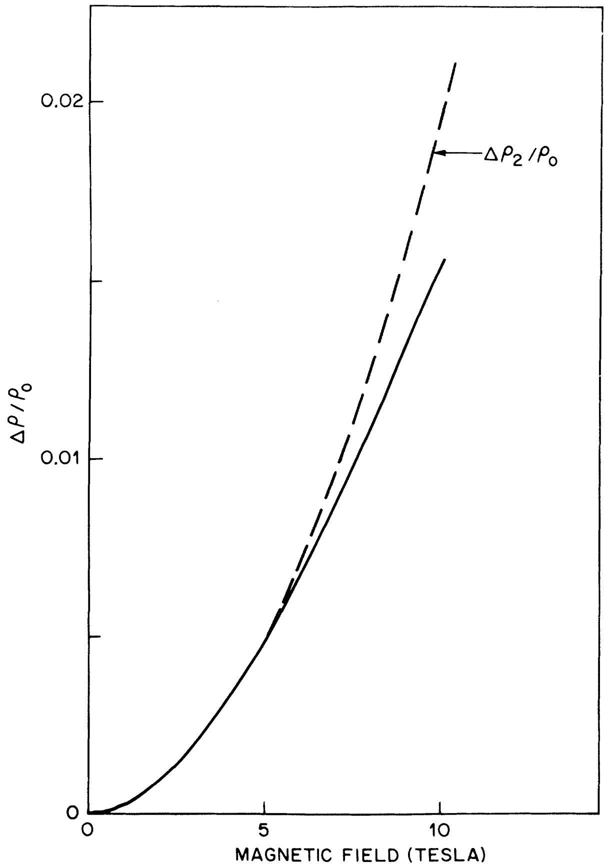

FIG. 1. Experimental trace (solid curve) of the magnetoresistance $\Delta\rho/\rho_0$ for a thick film (1100 Å) of $CoSi_2$ ($\rho_0 = 2.6$ $\mu\Omega$ cm) measured at 4.2 K. Also shown in the component second order in H, $\Delta\rho_2/\rho_0$ (dashed curve), a *negative* fourth-order contribution $\Delta\rho_4/\rho_0$ mainly accounting for the difference.

m_2, we can express the second-order contribution to magnetoresistance in the following fashion:

$$\Delta\rho_2/\rho = (\omega_{c1}\tau_1)(\omega_{c2}\tau_2) f(x,y) = (\omega_{c1}\tau_1)^2 y f(x,y), \tag{1}$$

where $f(x,y) = x(sy-1)^2/(xy+1)^2$ is a slowly varying function of order unity magnitude for reasonable values of the variables which are the ratios of carrier densities ($x = n_2/n_1$) and mobilities ($y = \mu_2/\mu_1$). In (1) the ω_c's are cyclotron frequencies ($\omega_c = eH/mc$), and the τ's are scattering times. [In $f(x,y)$ $s = \mathrm{sgn}(e_2/e_1)$ where e_2 and e_1 are the carrier charges.] Additional relations can be written in terms of x and y for $\Delta\rho_4/\rho$ and for the linear and third-order Hall effects. A self-consistent analysis of these data yields $x \sim 0.05$ and $y \sim 2$ whence it follows that $yf(x,y) \approx 0.7$ [Hall results show the two carrier species to be of opposite signs ($s = -1$) but predominantly holes[6] (95%)]. Thus, with the value $\Delta\rho_2/\rho_0 = 2.1\%$ (an average from four runs, cf. Fig. 1) we find

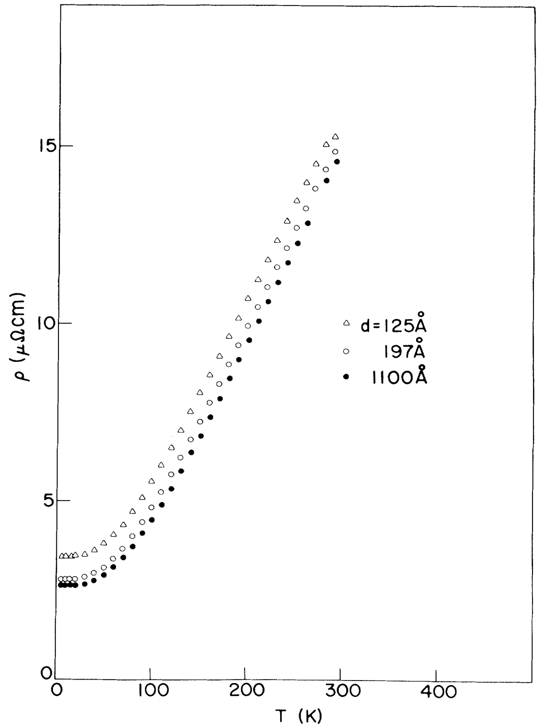

FIG. 2. Temperature dependence of the resistivity of thin, UHV-grown, single-crystal films of $CoSi_2$. Data for a 1100-Å-thick, polycrystalline (001) film are shown for comparison.

from (1) that for the majority carriers $\omega_{c1}\tau_1 \sim 0.17$ at 10 T. Finally, noting the relation

$$l_e = k_F l_c^2 (\omega_c \tau), \tag{2}$$

where k_F is the Fermi wave vector[8] and l_c is the magnetic length ($l_c^2 = \hbar c/eH$) we obtain $l_e = 970$ Å for the holes at 4.2 K and approximately the same for the electrons. This value of scattering length is surprisingly large for a metallic compound. It should be noted that this result does not depend on the effective masses. Estimates[9] from the saturation value of resistivity of $CoSi_2$ in the limit of strong disorder also give a value of $l_e \sim 1000$ Å.

Figure 2 shows measurements of the temperautre dependence of ρ for several films of $CoSi_2$ whose thickness is much less than the liquid-He value of l_e and, for comparison, a "thick" film, $d \sim 1100$ Å. The resistivity in the latter case behaves conventionally, i.e., as the sum $\rho(T) = \rho_0 + \rho_L(T)$, of additive contributions according to Matthiessen's rule of ρ_0, the residual resistivity, and $\rho_L(T)$, the phonon (Bloch-Grüneisen) resistivity. The remarkable thing is that the thin-film data differ almost not at all from the thick-film data. The most sensitive measure of the size effect is the residual resistivity, and even at $d \sim \frac{1}{8} l_e$ the residual resistivity is only increased by

25%.

In Fig. 3 we plot the ratio of the film residual resistivity ρ_0 to the bulk residual resistivity ($\rho_{0\infty} \approx 2.6\ \mu\Omega$ cm) for various samples of thickness ranging from $d = 60$ to 1100 Å as a function of the parameter $\kappa = d/l_e$. Again the data show little change from bulk behavior down to $\kappa \sim 0.1$; below this value there is an accelerated upturn in $\rho_0/\rho_{0\infty}$ indicating deterioration of quality in the thinnest films. It must be emphasized that the data in Fig. 3 are the *raw* data, representing an upper limit to the actual resistivity. Corrections for pinholes and other defects in the films would tend to depress the points towards the specular limit, $\rho_0/\rho_{0\infty} = 1$. Size effects in these films are very small indeed. No other data in *raw* form, to our knowledge,[10] exhibit such small size effects for a given κ and so directly indicate nearly total specularity.

The Fuchs theory[2] gives the following expression for the ratio of the resistivity of a film of finite thickness to the bulk resistivity ρ_∞:

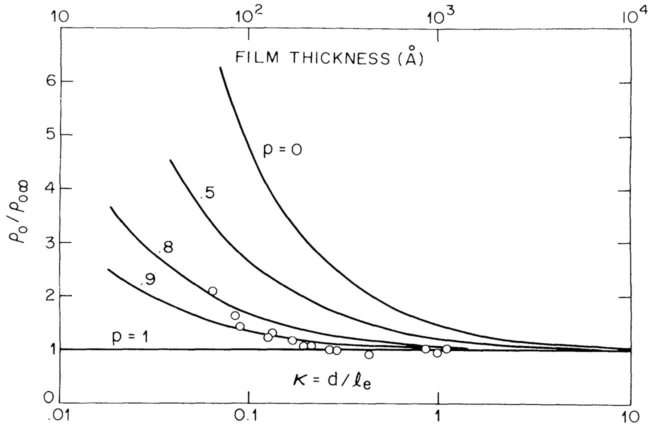

FIG. 3. Size effect in thin films of $CoSi_2$: A plot of residual resistivity (relative to $\rho_{0\infty}$) for fourteen specimens vs their thickness (relative to the bulk scattering length $l_e \approx 1000$ Å). Curves representing the Fuchs theory (see text) are shown for several values of the specularity parameter p.

$$\frac{\rho_\infty}{\rho} = 1 - \frac{3}{2\kappa}\int_0^1 du\, \frac{(u-u^3)(1-p)[1-\exp(-\kappa/u)]}{1-p\exp(-\kappa/u)}. \tag{3}$$

The curves in Fig. 3 are calculated from (3) for a range in values of the specularity parameter p ($p = 0$ is purely diffuse scattering; $p = 1$ is purely specular). A comparison of curves and data indicates that p lies between 80% and 90%. We reiterate that there are no corrections to either data or theory, especially as regards grain boundary scattering which tends to be a dominant factor in previous work.[10] If any such effects were present, the true specularity would even be closer to 100%.

The fact that the specularity is as high as $\sim 90\%$ indicates that the free surface, even though rougher than the solid/solid interface, acts as nearly specular reflector. Moreover, the specularity of the solid/solid interface is presumably even closer to 100%.

The Fuchs theory oversimplifies the problem by specifying the scattering in terms of a fixed specularity parameter. In fact, specularity depends upon the angle of incidence θ to the plane. Soffer[11] has introduced this refinement by replacing the fixed parameter p by the expression,

$$p(\cos\theta) = \exp\left[-\left(\frac{4\pi h}{\lambda_e}\cos\theta\right)^2\right] \tag{4}$$

adapted from the optics, where h is the rms roughness of the boundary and λ_e is the de Broglie wavelength, i.e., $\lambda_e = 2\pi/k_F$ which is ≈ 8 Å for $CoSi_2$. This produces curves, when (4) is substituted into (3) ($\cos\theta = u$), not very different from the Fuchs theory and leads to the result that $h \approx \frac{1}{20}\lambda_e \approx 0.4$ Å.

In conclusion, resistivity measurements on $CoSi_2$ thin films show little dependence on film thickness. This lack of size effect results from the bulk and interface perfection of epitaxial $CoSi_2$ grown on single-crystal Si. In terms of specularity, this would represent a degree of specularity of $\sim 90\%$ as an average for scattering from the free surface and solid/solid interface. These experiments demonstrate that in longitudinal transport in the present system the carriers reflect from the boundaries with little loss of phase coherence.

We are grateful to J. M. Gibson for providing invaluable transmission electron-microscope analysis of the silicides and their interfaces.

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[6]Very recent band calculations (L. F. Mattheiss, unpublished) suggest that there may be *three* hole bands (no electrons) with simple, closed Fermi surfaces. An extension of the theory [Eq. (1)] to the three-band case shows that the main conclusion, i.e., that $\Delta\rho_2/\rho_0 = (\omega_c\tau)^2 \times$ (a quantity of order unity), is not basically altered.

[7]The fact that the bands near the Fermi energy are free-electron-like (s and p character) affords some hope that such a simple analysis might work. See, e.g., Y. J. Chabal, D. R. Hamann, J. E. Rowe, and M. Schluter, Phys. Rev. B **25**, 7598 (1982); also J. Tersoff and D. R. Hamann, Phys. Rev. B **28**, 1168 (1983).

[8]Even a rough value for the density $n_1 \approx 2\times 10^{22}$ cm^{-3} provided by the above analysis is adequate to estimate k_F in view of the weak dependence, $k_F = (3\pi^2 n)^{1/3}$.

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