Quantum transport in ultrathin $CoSi_2$ epitaxial films

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Magnetotransport measurements have been performed in thin cobalt disilicide films epitaxially grown on Si(111) wafers. Films of thickness between 4.0 and 20.0 nm were studied in order to ascertain the important electron scattering rates. A temperature independent contribution to the phase breaking scattering rate was determined and attributed to spin–spin scattering of the conduction electrons which increases as the film thickness is decreased. The origin of this scattering and its importance to the low-temperature electron transport are discussed.

In recent years there has been considerable interest in the transport properties of very thin cobalt disilicide films. This interest stems from the high epitaxial quality of the crystalline films prepared using ultrahigh vacuum techniques. The lattice mismatch between $CoSi_2$ and Si(111) is only 1.2% at room temperature permitting the growth of pinhole-free films of thickness greater than 3.0 nm and continuous metal films down to 1.0 nm thick.[1–4]

Hensel *et al.* have shown that the resistivity of these $CoSi_2$ films is nearly independent of film thickness down to 10 nm.[5] The mean free path (l) has been measured to be 100 nm and therefore the electron scattering at the film interfaces is essentially specular. They also found that as the film thickness drops below 10 nm, the resistivity increases more rapidly than expected classically from the Fuchs–Sondheimer theory. The strong divergence of the resistivity has been attributed to a quantum size effect due to the discreteness of the momentum eigenstates in the direction perpendicular to the film.[4,6]

In the same thickness range (< 10 nm) Badoz *et al.*[7] found that the superconducting critical temperature (T_c) was abruptly depressed. This measurement was of particular interest since the mechanism for the T_c suppression could not be associated with a transition into a localized regime, the sheet resistance for these metal films was less than 35 $\Omega/\square$.[8] It was suggested (by Badoz *et al.*) that the cause of this T_c depression was the presence of a "perturbed layer of about 5 nm at the $CoSi_2$/Si" boundary due to small departures from $CoSi_2$ stoichiometry or from "ill-coordinated" cobalt atoms at the interface.[7]

In this letter we report on careful magnetotransport measurements designed to increase our understanding of these unusual properties of $CoSi_2$ thin films. The epitaxial crystalline $CoSi_2$ on Si(111) thin films used in this study were prepared and analyzed in the same manner reported by Hensel and co-workers.[4,5] In the low field limit, fits to the magnetoresistance provide information on the important scattering mechanisms.[8,9] We have studied the thickness dependence of the electron phase breaking rate in the interesting range of film thickness between 4.0 and 22.0 nm. An enhanced temperature-independent scattering rate was found in the thinner films.

Temperature sweeps at zero field revealed a resistance minimum at a temperature of a few Kelvin in all films less than 12.0 nm thick. The resistivity at temperatures below this minimum could be fit by the localization and interaction effects model[10]

$$\Delta R/R = -R_{\square}\xi\alpha_T \log(T/T_0),$$

where

$$\xi = e^2/2\pi^2\hbar. \tag{1}$$

For all films our data are consistent with a value of $\alpha_T = 1.0(\pm 0.05)$. Temperature sweeps at all fields up to 4 T reveal an α_T that remains at a value of 1.0. A field-independent α_T implies a large spin scattering rate (τ_s^{-1}) which has significant consequences for the electron transport properties. For τ_s^{-1} larger than the inelastic scattering rates (τ_i^{-1}), $\alpha_T = [1-(3/4)F]$ and is independent of field.[11] Here F is the parameter connected to the Hartree correction to the exchange interaction. Our value of $\alpha_T \approx 1.0$ at all fields points to a small screening parameter $F(\leqslant 0.1)$ in these films. This case is similar to the copper films studied by Gershenzon *et al.*[12]

The magnetic field sweeps showed a positive magnetoresistance (MR) at all fields up to 7 T for all samples studied (including field sweeps around zero field using 0.1 mT increments). The MR for the 8.4 nm films is shown in Fig. 1 for fields up to 0.1 T. In the theory of weak localization and interaction effects there are two mechanisms contributing to a positive MR. The first is a large spin-orbit scattering rate and the second is due to interaction effects. We can write the field-dependent conductivity in the region of field greater than the characteristic field for both effects as

$$\sigma(H) = \alpha_H\xi \log(H/H_0). \tag{2}$$

The term α_H can be written as $\alpha_H = \alpha_{ee} + \alpha_{HL}$, where α_{ee} is the contribution from interaction effects and α_{HL} is from weak localization effects.[10,13] Estimates of α_{ee} for this material at 1 K range from 0.02 to 0.008 depending on whether the calculated or measured values of F are used.[14]

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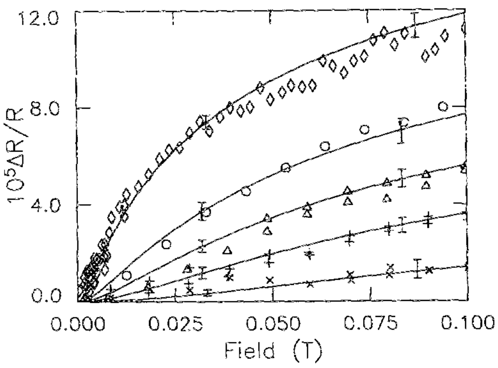

FIG. 1. Magnetoresistance of the 8.4 nm $CoSi_2$ film at temperatures of 0.2(◇), 1.3 (○), 4.2(△), 10.0(+), 20.0 K(×). The lines are the one parameter (τ_ϕ^{-1}) fit to the data with τ_{so}^{-1} chosen from the best fit to the 4.2 K data. The error bars represent the range of the fit with a 10% variation in τ_ϕ^{-1}.

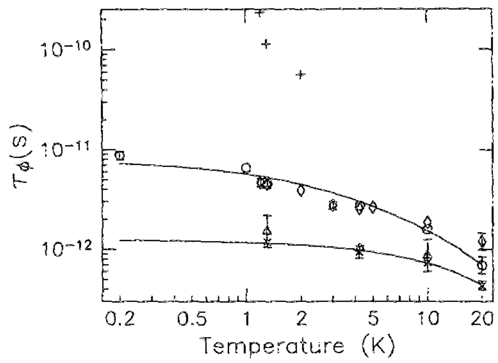

FIG. 2. Phase breaking scattering times for the films of thickness 22.0(+), 12.0(◇), 8.4(○), 6.4(×), 3.9 nm(△) from the fits to the magnetoresistance. The lines are the fits for the 8.4 and 6.4 nm films to the times for a $(A+BT+CT^3)^{-1}$ dependence. The error bars shown represent the uncertainty in the fits due to the scatter in the data.

In the theory of weak localization with strong spin orbit scattering, the value of $\alpha_{HL} \approx 0.5$.[15] Measurements of α_H yielded values dependent on thickness which range between 0.2 and 0.8. Because the value for α_H remains greater than α_{ee} for all films thickness and all temperatures studied we ignore the electron interaction contribution to the MR in the analysis of the data.[16]

The magnetoresistance data discussed above was fit to weak localization theory following the review of Al'tshuler *et al.*,[11] where we have also included the Maki–Thompson (MT) fluctuation terms for the samples showing a superconducting transition. The detailed analysis was similar to that of McGinnis and Chaikin[13] with the inclusion of a MT scattering term.[17-19] In this work $R_\Box$ and T_c were determined experimentally and the diffusion constant ($D = v_f l/2$) was calculated from a free-electron model.[20] A two-parameter fit to the MR at 4.2 K was used to establish the value of the spin orbit scattering rate (τ_{so}^{-1}) for each film. The τ_{so}^{-1} could be found with an accuracy of $\pm 30\%$ and this value was then used at all temperatures since it is expected to be temperature independent. A value for $\tau_{so}^{-1} \approx 5 \times 10^{12}$/s was found to fit well for all films which is also expected since this rate is thought to be a material constant. For measured values of $R_\Box$, T_c, D, and τ_{so}^{-1}, a single parameter fit can be performed to obtain the phase breaking scattering rate τ_ϕ^{-1} to within 10%.[21,22]

The results of this analysis are shown in Fig. 2 for τ_ϕ for the five film thicknesses studied. This clearly shows a decrease in the temperature-independent contribution to the phase breaking scattering time of a factor of 150 from the bulk-like 22.0 nm film to the 3.9 nm film. This term in the scattering rate is associated with a spin flip scattering from paramagnetic defects in the sample. The scattering time of this mechanism can be calculated from the relation $\tau_\phi(T=0) = \tau_s/2$. The values of τ_s^{-1} found from the extrapolated scattering times at zero temperature are shown in Table I.

We do not believe that the origin of this magnetic scattering rate can be associated with a random distribution of magnetic impurities. Such a contribution would not show the thickness dependence seen here. Furthermore, the samples were all prepared in the same ultrahigh vacuum chamber and most of the films (22.0, 12.0, 6.4, 3.9 nm samples) were deposited at the same time on the same substrate and therefore the concentration of these types of impurities should have been a constant from film to film. Instead, we believe that we are observing an interface effect; a spin scattering rate due to magnetic defects at the $Si/CoSi_2$ interface which are believed to be magnetic cobalt atoms. Our data are consistent with the ideal of Badoz *et al.*[7] of a thin layer ≈ 5 nm at the $Si/CoSi_2$ boundary that exhibits either a small departure from $CoSi_2$ stoichiometry or ill-coordinated interface Co atoms. In fact transmission electron microscope (TEM) studies[23,24] of the $Si/CoSi_2$ interface suggest that the Co atoms at the interface are fivefold coordinated instead of the usual eightfold coordination in the bulk.[25] This change of coordination number may create a different electron energy level structure for the *d* shell molecular orbitals, which could result in a nonvanishing magnetic moment on the interfacial Co atoms. In either case the magnetic properties of the interface will have a more dominant effect on the electron transport properties of the thinner films.

Abriksov and Gor'kov showed that magnetic impurities lead to a strong depression of the superconducting critical temperature.[26] The suppression of T_c in our films due to magnetic scattering alone can be estimated[27] from

$$\ln \frac{T_c}{T_{c0}} = \psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \frac{\hbar/2\tau_s}{2\pi k_B T_c}\right), \qquad (3)$$

where ψ is the digamma function and T_{c0} is the superconducting temperature without magnetic scattering (taken to

TABLE I. Parameters for $CoSi_2$ films, $R_\Box$ is for 4.2 K, τ_s is the magnetic scattering time deduced from Fig. 2, and T_c Calc. is the calculated critical temperature from Eq. (3).

Film thick. (nm)	$R_\Box$ ($\Omega/\Box$)	D (cm^2/s)	τ_s (ps)	T_c (K)	T_c Calc. (K)
20.0	0.80	472	454	1.06	1.24
12.0	4.28	150	12.9	0.57	1.013
8.40	8.45	107	16.8	<0.20	1.067
6.40	19.5	61.4	2.5	<0.20	<0.001
3.90	48.9	40.4	3.0	<0.20	<0.001

be T_c of bulk). The superconducting temperatures calculated from the measured spin-spin scattering rates are listed in Table I as well as the measured T_c for each film studied. From this comparison we believe that the magnetic scattering rate measured from the low-field MR is at least partially responsible for the T_c suppression in these films. It is difficult to make a reliable absolute comparison between the calculated and measured T_c depression since our values of τ_s are dependent on the diffusion constant. Our method for determining the diffusion constant from the low-temperature resistivity may be inaccurate and will add a small systematic error to the calculated scattering times. Still it is suggestive that in the region of thickness with appreciable T_c depression, a corresponding increase in the magnetic scattering rate is found. This rate is of the right magnitude to cause the observed suppression of the superconducting transition.

In summary, we have carried out careful measurements of the localization contribution to the resistivity and to the magnetoresistance in the $CoSi_2$/Si(111) thin-film system for the first time. The electron phase breaking times have been accurately determined from the the MR. A magnetic scattering rate which increases with decreasing film thickness has been found. This magnetic scattering rate has been associated with the depression of the superconducting critical temperature and related to results of previous TEM studies.

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[22] It should be noted that the 22 nm film was very clean ($R_\square = 0.8\ \Omega$) resulting in a small weak localization MR and we were unable to perform an accurate fit to the data for this contribution. Therefore τ_ϕ could only be measured in a narrow temperature range above the T_c (1.06 K) of this film.
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