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Size effects in the resistivity of thin films of potassium

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Abstract. We have measured size effects in the electrical resistivity of thin films of potassium below 1 K. These size-effect data appear to be highly anomalous when compared with our previous resistivity data for thin wires of potassium. However, we show that both sets of measurements can be explained within a unified theoretical framework in terms of normal electron–electron scattering and normal electron–phonon scattering. Specific predictions of size effects are made for size ranges not covered by the present experimental data.

1. Introduction

About a decade ago, we showed (Lee *et al* 1982) that experiment confirms the theory for the low-temperature electrical resistivity of a pure, dislocation-free, bulk sample of potassium. Below 1 K, the temperature-dependent part of the resistivity $\rho(T)$ was measured to be

$$\rho(T) = AT^2 \tag{1}$$

where the quadratic term is believed to be due to electron–electron scattering. By restricting the data to temperatures below 1 K, one may safely neglect the contribution to $\rho(T)$ due to Umklapp electron–phonon scattering.

We subsequently extended the measurements to thin wires of potassium and found size effects for $\rho(T)$ that exhibited a behaviour complex in both magnitude and temperature dependence (Zhao *et al* 1988). This is illustrated in the lower part of figure 1, where $\rho(T)$ is plotted against temperature for wires with diameters down to 80 μm. In this paper, we report the surprising results we obtained upon extending the measurements of $\rho(T)$ to samples with still smaller dimensions. Since it is difficult to produce potassium wires having smaller diameter, to continue our investigations of size effects, we found it necessary to measure the resistivity of thin films of potassium. Data representative of these new measurements are shown in the upper part of figure 1.

Clearly there is a remarkable contrast between the two sets of data. For the thin wires, $\rho(T)$ is *smaller* than for bulk potassium (the wire having $d = 1.5$ mm) and *decreases* as the thickness d decreases. On the other hand, for the thin films, we found that $\rho(T)$ is *larger* than the bulk values and *increases* as the thickness d decreases.

A theory describing the resistivity of thin wires of potassium has recently been given by Movshovitz and Wiser (1990a), who reference all the recent work on thin wires and films of non-transition metals. Important theoretical contributions have

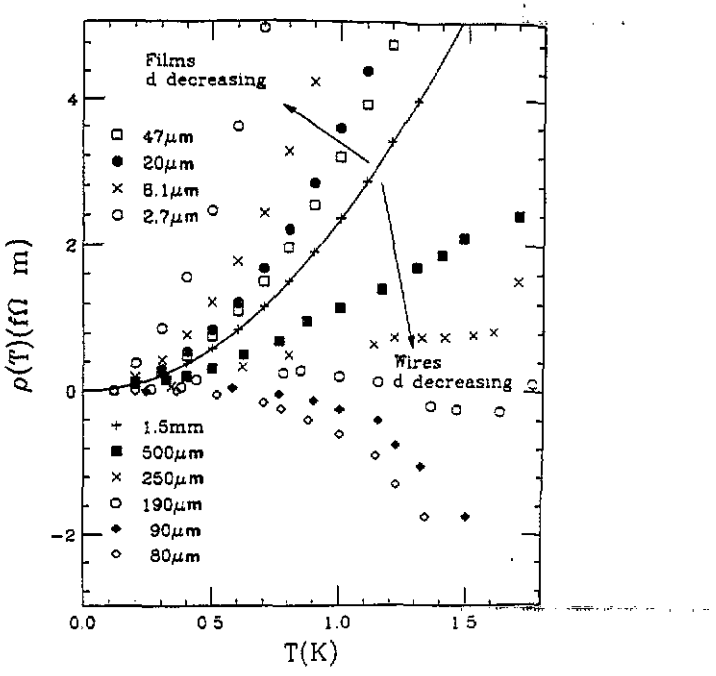

Figure 1. Temperature dependence of $\rho(T)$ for thin wires and films of potassium. The values for a bulk sample (the wire with $d = 1.5$ mm) exhibit the predicted quadratic behaviour shown by the full curve. For the thin films, $\rho(T)$ increases with decreasing thickness d, whereas for the thin wires, $\rho(T)$ decreases with decreasing d.

been made by Sambles *et al* (1980, 1982), De Gennaro and Rettori (1984, 1985), Kaveh and Wiser (1985), and Gurzhi *et al* (1989a,b). We here show that an extension of this theory accounts for the range of film and wire thicknesses discussed in this paper and makes definite predictions for thicknesses for which there are no experimental data available. In particular, the theory explains our principal experimental result: for the thin wires, $\rho(T)$ is much *smaller* than the bulk values, whereas for the much thinner films, $\rho(T)$ is much *larger* than the bulk values.

In section 2, we give some experimental details relating to the measurement of $\rho(T)$ for thin films of potassium. The theory of the non-resistive electron scattering contribution to $\rho(T)$ is reviewed in section 3. In section 4, we discuss the important change in the sign of $\rho(T)$ with decreasing thickness for non-resistive scattering. The comparison between theory and experiment is given in section 5. The summary follows in section 6.

2. Experiment

The films were prepared by evaporating potassium at room temperatures onto glass, silicon and single-crystal KF substrates (Qian 1991). The evaporation was carried out at a pressure of approximately 10^{-6} Torr in an evacuated belljar within a glove box filled with highly purified argon. After the evaporation, the belljar was opened and the samples were transferred to a vacuum-tight sample can, along with a large-area Cu foil coated with potassium. This guaranteed that after several days, when all the measurements were complete, the potassium film would still be bright and

shiny. Measurements of the ratio $(1/\rho)(\mathrm{d}\rho/\mathrm{d}T)$ were carried out down to 0.1 K in a dilution refrigerator using the high-precision techniques described by Edmunds *et al* (1980). We shall concentrate on the results for the samples deposited on Si because the resistivity data for the other substrates (to be presented in a later publication) show large Kondo effects which tend to confuse the interpretation. The results displayed in figure 1 were obtained by performing the necessary integration of the data.

3. Theory

We begin by reviewing the recent theory for $\rho(T)$ for thin wires and films. The major contribution of electron–surface scattering is *not* due to $\rho(T)$ at low temperatures altering the coefficient A in equation (1). Instead, the major effect of electron–surface scattering is to introduce two new contributions to $\rho(T)$ which are completely absent for bulk samples. These are intimately connected with normal electron–electron scattering (NEES) and with normal electron–phonon scattering (NEPS). Neither of these scattering mechanisms contributes to $\rho(T)$ for a bulk sample, and they are therefore designated as *non-resistive*. Regarding NEES, the total electron momentum is conserved upon scattering for a free electron metal, whereas the NEPS contribution to $\rho(T)$ vanishes at low temperatures because of phonon drag (Kaveh and Wiser 1972). However, for thin wires and films, both NEES and NEPS contribute to $\rho(T)$ by altering the direction of the electron trajectory, thereby altering the mean free path for resistive electron–surface scattering.

This phenomenon is illustrated in figure 2. In figure 2(a), the electron begins its trajectory at point O, travelling in the direction θ, ϕ and undergoes a resistive electron–surface collision after traversing a distance $R(\theta, \phi)$. In figure 2(b), the electron again begins its trajectory at point O, travelling in the direction θ, ϕ. However, after traversing a distance $S(\theta, \phi)$ and reaching point P, the electron undergoes a non-resistive collision (either NEES or NEPS) which alters the direction of its trajectory to θ', ϕ'. In this new direction, the distance to the surface of the wire is $T(\theta', \phi')$. Thus, the total distance from the initial point O to the resistive collision at the wire surface is $S + T$, rather than R. The distance $S + T$ may be longer than R, yielding a reduced contribution to $\rho(T)$, or shorter than R, yielding an increased contribution. The net effect on $\rho(T)$ is determined by summing over all possible electron trajectories in which a non-resistive collision takes place.

The method of 'electron dynamics' for calculating $\rho(T)$ for non-resistive electron scattering in thin films and wires has already been given (Movshovitz and Wiser 1990b, Movshovitz 1991). Therefore, we need only summarize here the principal steps of the method.

(i) One calculates Λ_0, the electron mean free path in the presence of surface and impurity scattering, but in the absence of NEES and NEPS.

(ii) One then calculates Λ_{NR}, the electron mean free path in the presence of non-resistive (NR) NEES and NEPS collisions, as well as surface and impurity scattering.

(iii) From the results of (i) and (ii), one obtains the change $\Delta\Lambda_{\mathrm{NR}}$ in the electron mean free path due to NR collisions.

$$\Delta\Lambda_{\mathrm{NR}} = \Delta_{\mathrm{NR}} - \Lambda_0. \tag{2}$$

The contribution of NR scattering to the resistivity is then

$$\rho_{\mathrm{NR}}(T) \propto -\Delta\Lambda_{\mathrm{NR}}/\Lambda_0^2. \tag{3}$$

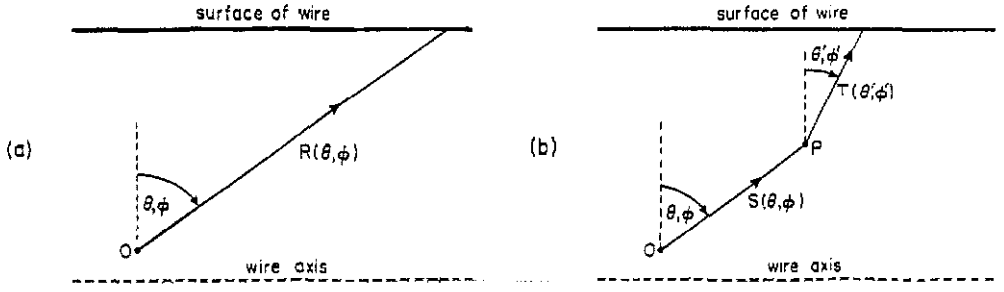

Figure 2. An electron trajectory in the direction θ, ϕ starting at point O. (*a*) No electron scattering occurs except at the surface. The distance along this trajectory to the surface of the wire is $R(\theta, \phi)$. (*b*) Non-resistive scattering occurs. The electron travels a distance $S(\theta, \phi)$ and then undergoes a non-resistive collision at point P, which alters the direction of its trajectory to θ', ϕ'. The distance along this new direction to the surface of the wire is $T(\theta', \phi')$.

(iv) Since the probability of an NR collision taking place is proportional to $1/\lambda_{NR}$, it follows that $\Delta\Lambda_{NR}$ contains this factor, which determines the temperature dependence of $\rho_{NR}(T)$. For NEES and NEPS, this implies that

$$\rho_{NEES}(T) \propto 1/\lambda_{NEES}(T) \propto T^2 \tag{4}$$

$$\rho_{NEPS}(T) \propto T^2/\lambda_{NEPS}(T) \propto T^5. \tag{5}$$

(v) Finally, combining these results with equation (1) yields

$$\rho(T) = \rho_{NEPS}(T) + \rho_{NEES}(T) + \rho_{UEES}(T) = \beta T^5 + \gamma T^2 + \eta A^{\infty}_{UEES} T^2 \tag{6}$$

where η gives the (relatively small) size-effect correction to the bulk value A^{∞}_{UEES}, which is due to Umklapp electron–electron scattering. The complex theoretical task lies in the calculation of the coefficients β, γ, η (Movshovitz and Wiser 1990a,b, Movshovitz 1991).

In carrying out steps (i) and (ii), one must take account of the fact that electron–surface scattering is partially specular. To this end, one introduces the specularity parameter p, whose limiting values $p = 0$ and $p = 1$ denote diffuse and specular electron–surface scattering, respectively. An important feature of the present calculation is that p was not taken to be a constant parameter. Rather, we included the fact that $p(\theta)$ depends on the angle between the electron trajectory and the normal to the surface of the wire (Sambles and Elsom 1980, Sambles *et al* 1982). We used for $p(\theta)$ the expression given by Soffer (1967)

$$p(\theta) = \exp(-(4\pi\alpha \cos\theta)^2) \tag{7}$$

where α is the surface-roughness parameter, defined as the ratio of the root mean square surface roughness to the electron de Broglie wavelength. One expects α to be of order unity (Sambles and Elsom 1980, Sambles *et al* 1982).

4. Sign of the coefficients β and γ

The coefficients β and γ depend on the ratio $x = \lambda_b/d$, where d denotes the diameter of the wire or the thickness of the film, and λ_b denotes the bulk electron mean free

path due primarily to electron–impurity scattering. Since λ_b is comparable for all the thin wires, one may view the variable x as measuring the inverse of the diameter of the thin wire.

The principal result of our calculation is the dependence we found for β and γ as a function of x. This is shown schematically in figure 3, for the range of x extending from the values appropriate to the thin wires to the values appropriate to the films. Of course, the precise values of β and γ are different for films and for wires, but qualitatively, the dependence of β and γ on x is similar in the two cases. Both coefficients are negative for small values of x. As x increases, they decrease to a minimum value, then increase to zero at some value x_0, and thereafter become positive and large.

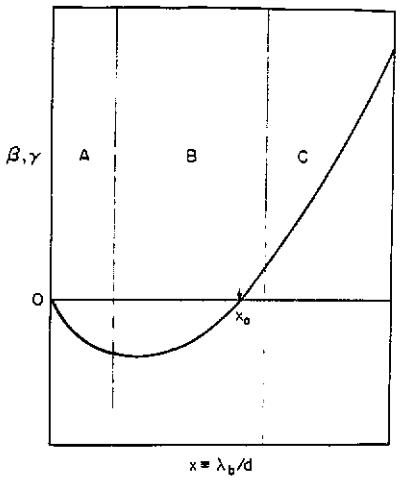

Figure 3. Schematic diagram of the coefficients β and γ as a function of the ratio $x \equiv \lambda_b/d$, showing that these coefficients are negative for $x < x_0$ and positive for $x > x_0$. The value of x_0 (typically between 1 and 5) depends on the surface-roughness parameter α and on whether one is considering β or γ, and a film or a wire. The regimes A and C correspond to the thin-wire and thin-film data, respectively, whereas the regime B corresponds to the intermediate regime for which no low-temperature $\rho(T)$ data are available.

It is convenient to identify three regimes of x in figure 3, which are labelled A, B and C. Our thin wires lie in regime A, where both β and γ are *negative and decreasing* as x increases. At the other extreme, our much thinner films lie in regime C, where both β and γ are *positive and increasing* as x increases. This explains the dramatic contrast between the two sets of $\rho(T)$ data shown in figure 1.

In between regimes A and C lies regime B, in which β and γ pass through zero and change sign. In this intermediate regime B, there are no $\rho(T)$ data available to date. It is this absence of connecting data that produces the apparent contradiction shown in figure 1 between the thin-wire data and the thin-film data.

From this discussion, it follows that the key to our understanding of the low-temperature $\rho(T)$ data is the change in sign of the coefficients β and γ with decreasing thickness (as illustrated in figure 3). Therefore, it is important to emphasize that this change in sign is a general result. For example, any reasonable value of the surface-roughness parameter α yields the change in sign. Similarly, for any purity of the thin

wire or film, the sign of β and γ will change as the thickness becomes sufficiently small.

Finally, it should be noted that the present results for the thickness dependence of β and γ are unrelated to the many previous calculations of size-dependent deviations from Matthiessen's rule (see the comprehensive review by Bass (1972)). The present results deal with non-resistive electron scattering mechanisms, whereas the previous work dealt with resistive electron scattering mechanisms.

5. Comparison between theory and experiment

In the calculation of the coefficients β, γ, η, the value of α has been taken to be an adjustable parameter. The reasonable values of $\alpha = 1$ and $\alpha = 2$ were used for the films and the wires, respectively. The resulting calculated values of the coefficients β, γ, η are listed in table 1, together with the sample characteristics, for each of the measured wires and films.

Table 1. Calculated values of the parameters β, γ and η for the thin films and wires of potassium. The letter d denotes the thickness of the film or the diameter of the wire, ρ_o is the experimental value of the residual resistivity, and λ_b is the bulk value of the electron mean free path.

Sample geometry	d (μm)	ρ_o (pΩ m)	λ_b/d	β (fΩ m K^{-5})	γ (fΩ m K^{-2})	η
Bulk	∞	—	0	0	0	1.0
Wire	900	10	0.21	−0.074	−0.82	1.0
Wire	500	16	0.38	−0.120	−1.38	1.0
Wire	250	20	0.76	−0.152	−1.82	1.0
Film	47	37	1.5	+0.14	+0.45	1.03
Film	10	73	2.05	+0.39	+1.54	1.08
Film	8.1	54	2.45	+0.65	+2.44	1.13
Film	2.6	170	3.2	+1.31	+4.18	1.22

Inserting into equation (6) the values of β, γ, η listed in table 1, together with the experimental bulk value $A^{\infty}_{\mathrm{UEES}} = 2.6$ fΩ m, yields the theoretical curves shown in figure 4. The agreement between theory and experiment is evident from the figure. Nevertheless, we wish to emphasize that we believe that the most significant result of our work is not the detailed agreement between theory and experiment shown in figure 4, but rather the striking variation of β and γ with x that is shown in figure 3.

The good fits to the thin-film data were obtained by adjusting λ_b from film to film, with λ_b decreasing significantly as the thickness of the film decreases. This apparent variation of λ_b with film thickness may be due to the thinnest films having a grain size smaller than in the bulk. Consequently, electron scattering off the grain boundaries may make an additional contribution to λ_b which becomes more important as the film becomes thinner. We note that continuous films of potassium deposited on room-temperature substrates could not be attained for thicknesses much less than 2.7 μm.

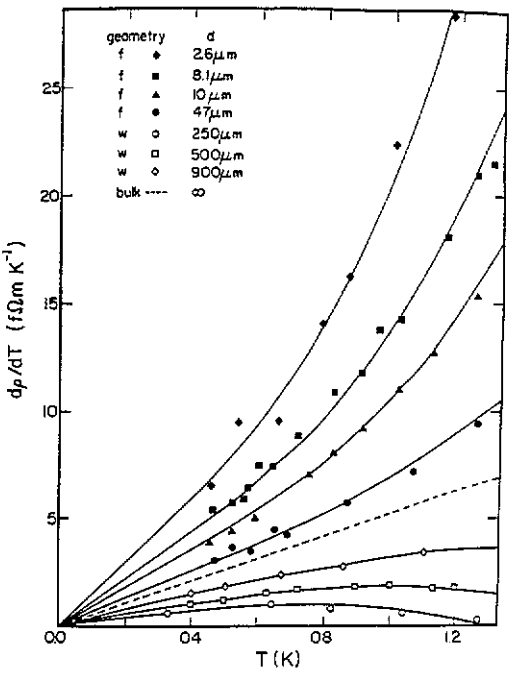

Figure 4. Temperature dependence of $d\rho/dT$ for samples of various thicknesses. For the thinner samples (films having thickness $d < 100$ μm), both the data (full symbols) and the theoretical curves lie above the bulk-sample values (the broken straight line), whereas for the thicker samples (wires having diameter $d > 100$ μm), both the data (open symbols) and the theoretical curves lie below the bulk-sample values.

6. Summary

We have carried out an experimental and theoretical analysis of the thickness dependence of $\rho(T)$ for thin films of potassium, and we have obtained the following results:

(i) A dramatic size effect was measured for $\rho_{\text{expt}}(T)$ for thin films of potassium at low temperatures. This size effect has the opposite sign from that found earlier for thin wires of potassium.

(ii) Both the present results for the films, as well as our previous results for thin wires, can be explained by the same theory in terms of normal electron–electron scattering and normal electron–phonon scattering. This is indeed remarkable because for bulk potassium, these two scattering processes make no measurable contribution at all to $\rho_{\text{expt}}(T)$.

(iii) The theory makes specific predictions for the size dependence of $\rho(T)$ in the intermediate thickness regime between our thin films and the previously measured thin wires. This regime is experimentally accessible, and since its boundaries are broadly defined by our analysis, this suggests some interesting work for future experiments.

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