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Anomalous electronic transport behavior, including a Kondo-like effect, for potassium in contact with hydro- and halocarbons

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We report measurements below 1.5 K of the residual resistivity ρ_0, the temperature derivative of the electrical resistivity, $d\rho/dT$, and the thermoelectric ratio G, for potassium (K) samples encased in polyethylene and Teflon tubing or in contact with either polyethylene or the halocarbons Teflon and Kel-*F*, and for a dilute K-Rb alloy encased in polyethylene tubing. Pure-K samples encased in any type of tubing show anomalous behavior in ρ_0, in which ρ_0 is unusually large upon initial cooling of the sample to 4.2 K and then decreases upon "annealing" at room temperature to the lowest values characteristic of bare samples. We attribute this behavior to constraints placed upon the samples by the tubing. Both pure-K and alloy samples in contact with polyethylene show anomalous behaviors in $d\rho/dT$ and G below 1 K that have all of the characteristics of a Kondo effect, including a resistivity minimum, a thermoelectric anomaly, and disappearance of both effects upon application of a small ($\simeq 0.1$ T) magnetic field. No such anomalies are seen in bare unstrained samples or in samples in contact with Teflon or Kel-*F*. We argue that these ρ_0 and Kondo-like anomalies provide plausible mechanisms for a substantial portion of the unusual behaviors that were reported in the first high-precision measurements of $d\rho/dT$ in K in the vicinity of 1 K, which had previously been attributed to the presence of either defects or a charge-density-wave ground state in these samples. We have not been able to discover the physical source of the Kondo-like anomalies.

I. INTRODUCTION

There has been great interest in the temperature-dependent electrical resistivity $\rho(T)$ of K at temperatures below 1.5 K ever since the late 1970s, when van Kempen *et al.*[1] (VK), Rowlands *et al.*[2] (RO), and Levy *et al.*[3] (LY) reported that high-precision measurements[1-3] at such low temperatures showed anomalous behavior involving time dependences of both the residual resistivity ρ_0 and the magnitude of $d\rho/dT$ in the vicinity of 1 K. Theorists quickly developed models[4-6] to explain the anomalies based on the assumption that they were intrinsic to K samples free from any perturbations, except for the presence of unavoidable point or extended defects. In this paper we argue that this assumption is probably not correct. We argue that the anomalous changes in $d\rho/dT$ and some of the changes in ρ_0 reported in the first studies of K were more likely due to physical constraints upon the samples, coupled with either a Kondo-like anomaly for the two cases—VK (Ref. 1) and LY (Ref. 3) —where the samples were encased in polyethylene tubing[1,3] to protect them from contamination, or to an anomalous size effect in thin wires in the case—RO (Ref. 2)—where the samples were bare, but unusually thin ($d=0.8$ mm) and wound around a grooved Teflon cylinder. We have argued elsewhere[7,8] the case for the anomalous "size effect" in explaining both the form and magnitude of RO's data for $d\rho/dT$ below 1.5 K. In this paper we will show that constraining a K sample inside a plastic tube can produce exactly the time-dependent changes in the residual resistivity ρ_0 reported by VK and changes very similar to those reported by RO. We show also that contact of K samples with the hydrocarbon polyethylene produces a Kondo-like behavior that results in changes in $d\rho/dT$ in the vicinity of 1 K that reproduce most of the anomalous behaviors reported by VK and LY.

The paper is organized as follows. In Sec. II we provide necessary background for understanding the issues addressed in this paper: we discuss the behavior expected for $\rho(T)$ and G in high-purity K; review the anomalies seen by the first investigators of $\rho(T)$ in K below 1.5 K and the models developed to explain them, and describe the characteristic behaviors expected for a Kondo system. In Sec. III we briefly describe our samples and experimental procedures. In Sec. IV we present our experimental data and use them to address four issues. (1) How physical constraint of K samples can lead to anomalous behavior of ρ_0. (2) How physical contact of K samples with polyethylene leads to anomalous behavior of $d\rho/dT$ in the vicinity of 1 K. (3) How the behavior below 1 K of both $\rho(T)$ and the thermoelectric ratio $G(T)$ for K samples in contact with polyethylene accords with expectations for a Kondo system. (4) What conditions lead to a Kondo-like effect in K below 1 K. Section V contains a summary and conclusions. Preliminary results from this study have previously been reported.[9,10]

II. BACKGROUND

A. Low-temperature behavior expected for a free-electron metal like K

It is generally believed (e.g., from measurements of the de Haas–van Alphen effect[11]) that K has one of the simplest electronic structures of any metal, with a nearly spherical Fermi surface that does not contact Brillouin-zone boundaries, and with completely empty *d* and *f* shells. There are, however, various anomalies in the behavior of K, especially in the presence of a magnetic field, which are not easily understood in terms of this simple free-electron model, and these anomalies led to the proposal that the ground state of K is a charge-density-wave (CDW) state.[12] The discussion in the present paper will be in the context of a free-electron ground state.

The low-temperature transport properties of a free-electron metal such as K should be well described by standard theory.[13] Above 1.5 K the electrical resistivity of K is dominated by electron-phonon scattering, which is now well understood in this metal.[14] Below about 1.2 K such scattering should be unimportant[1,2] and we expect the temperature-dependent resistivity $\rho(T)$ to have the simple form

$$\rho(T)=AT^2+B\rho_0T^2 \tag{1a}$$

$$=A'T^2\,. \tag{1b}$$

In Eq. (1a) the term AT^2 is due to electron-electron scattering[13] and the term $B\rho_0T^2$ is due to inelastic-impurity scattering.[15] If Eqs. (1) correctly described $\rho(T)$ in K, then a plot of $d\rho/dT$ versus T should yield a straight line that passes through the origin and has slope A'. For high-purity samples this slope should be determined mainly by A, with only a small contribution from $B\rho_0$. Alternatively, a plot of $(1/T)(d\rho/dT)$ versus T should yield a horizontal straight line; this form has the advantages that anomalies are easily seen as deviations from a horizontal line and that the relative visibility of low-temperature anomalies is enhanced.

In our previous studies of free-hanging thick (diameter $d>1$ mm) high-purity K samples,[16] and dilute K-Rb alloys,[17] we found exactly the form predicted by Eqs. (1) from 1.3 K down to approximately 0.3 K, as illustrated in Fig. 1. In addition, as would be expected if the first term in Eq. (1a) is dominant, for values of ρ_0 ranging from 9.4 to 16.9 pΩ m [or, alternatively, residual resistivity ratios $R=\rho(295\text{ K})/\rho(0\text{ K})$ ranging from 4250 to 7600] the values of A' in Fig. 1 varied by only ±12% after correction for the $B\rho_0$ term in Eq. (1a). Finally, as expected for samples prepared from similar high-purity stock, all of the K samples had high initial R's (≥ 4250) when slowly cooled to 4.2 K, and both R and A' for a given sample remained stable if the sample was remeasured after being held at room temperature for days or weeks. As also shown in Fig. 1, we found, below about 0.3 K, an anomalous upturn in $(1/T)(d\rho/dT)$, the size of which varied from sample to sample. We have tentatively ascribed this behavior to the presence of residual defects such as dislocations. It is the subject of another publication[10] and will not be considered further in this paper.

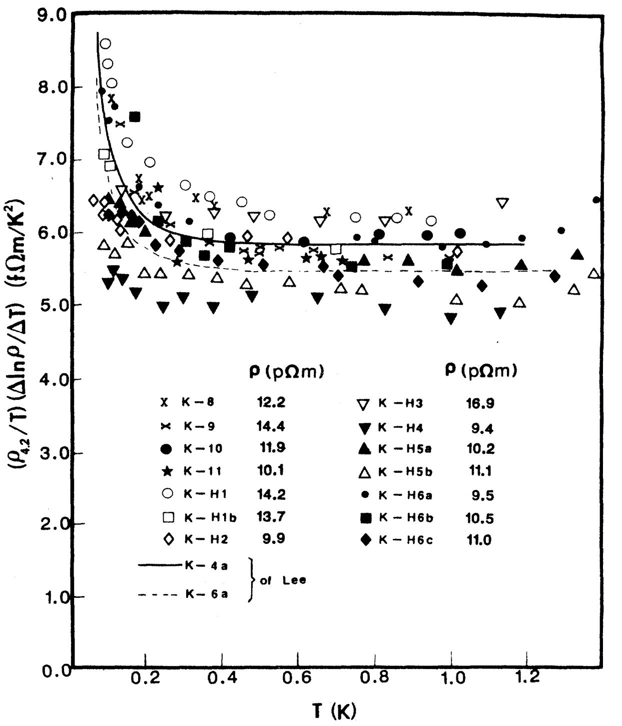

FIG. 1. $(\rho_{4.2\text{ K}}/T)(\Delta\ln\rho/\Delta T)\simeq(1/T)(d\rho/dT)$ vs T for a series of thick, bare, free-hanging K samples. For a combination of just electron-electron scattering plus inelastic-impurity scattering, the data should lie on horizontal lines.

The thermoelectric ratio G is defined by[18]

$$G=(J/Q)|_{E=0}\,, \tag{2}$$

where J and Q are, respectively, the electrical and thermal current densities in the sample when the electric field E in the sample is zero. The thermopower S is related to G by[18]

$$S=GLT\,, \tag{3}$$

where L is the Lorenz ratio.[13] At low temperatures ($T\leq 1.1$ K for K),[19] where impurity scattering of electrons predominated, $L\doteqdot L_0$, the Sommerfeld value of the Lorenz ratio. Under these circumstances, G for a free-electron metal like K should have the simple form[18]

$$G=G_0-FT^2\,. \tag{4}$$

Here, G_0 is due to elastic impurity scattering and FT^2 is due to normal phonon drag (possibly with a contribution from phony phonon drag[20]). At higher temperatures, we expect from previous studies[18] to see a phonon-drag minimum in G due to competition between a negative contribution from normal phonon drag and a positive contribution from umklapp phonon drag, with the latter winning out above about 3.5 K. This behavior will be illustrated below.

B. Results of previous studies of $\rho(T)$ in K below 1.3 K

Four groups of investigators have previously studied K samples in contact with plastics at low temperatures. Three involved contact with polyethylene: VK;[1] LY;[3] and Haerle *et al.*[10] (HE) in our laboratory. Two involved contact with Teflon: RO (Ref. 2) and HE (Ref. 10).

VK studied K samples encased in polyethylene tubes of diameter $d = 0.9$ mm. Since their measurements extended down only to 1.1 K, they were not able to see direct evidence of the anomalous form of $d\rho/dT$ below 1 K that we describe below for samples in contact with polyethylene. They did, however, find deviations from the simple behavior described above, in that both ρ_0 and A' decreased substantially when a sample was held at room temperature for days to months after being fabricated. In one sample, ρ_0 decreased from 24 to 9 pΩ m, and at the same time A' decreased from 1.6 to 0.8 $f\Omega\,\text{m}/K^2$. These decreases in ρ_0 corresponded to increases in R from 3100 to 8100. The behavior VK found is shown in Fig. 2(b). For comparison, Fig. 2(b) also contains typical data for a free-hanging sample of K (see Fig. 1) and for two stages of room temperature "annealing" of a sample (K-*PH*2a and K-*PH*2d) encased in polyethylene that we discuss later. Note from Fig. 2 that measurements to temperatures well below 1 K were essential to recognize the more complex behavior displayed by our samples encased in polyethylene.

LY measured K samples encased in $d = 1.0$ mm polyethylene tubes at temperatures from 4.2 K down to 1.1 K. In the vicinity of 1.1 K they also found data apparently consistent in form with Eqs. (1). However, when they deliberately contaminated their samples—which initially had very high R's ($R \simeq 14\,000$)—they found large, simultaneous decreases in both the R's and the values of

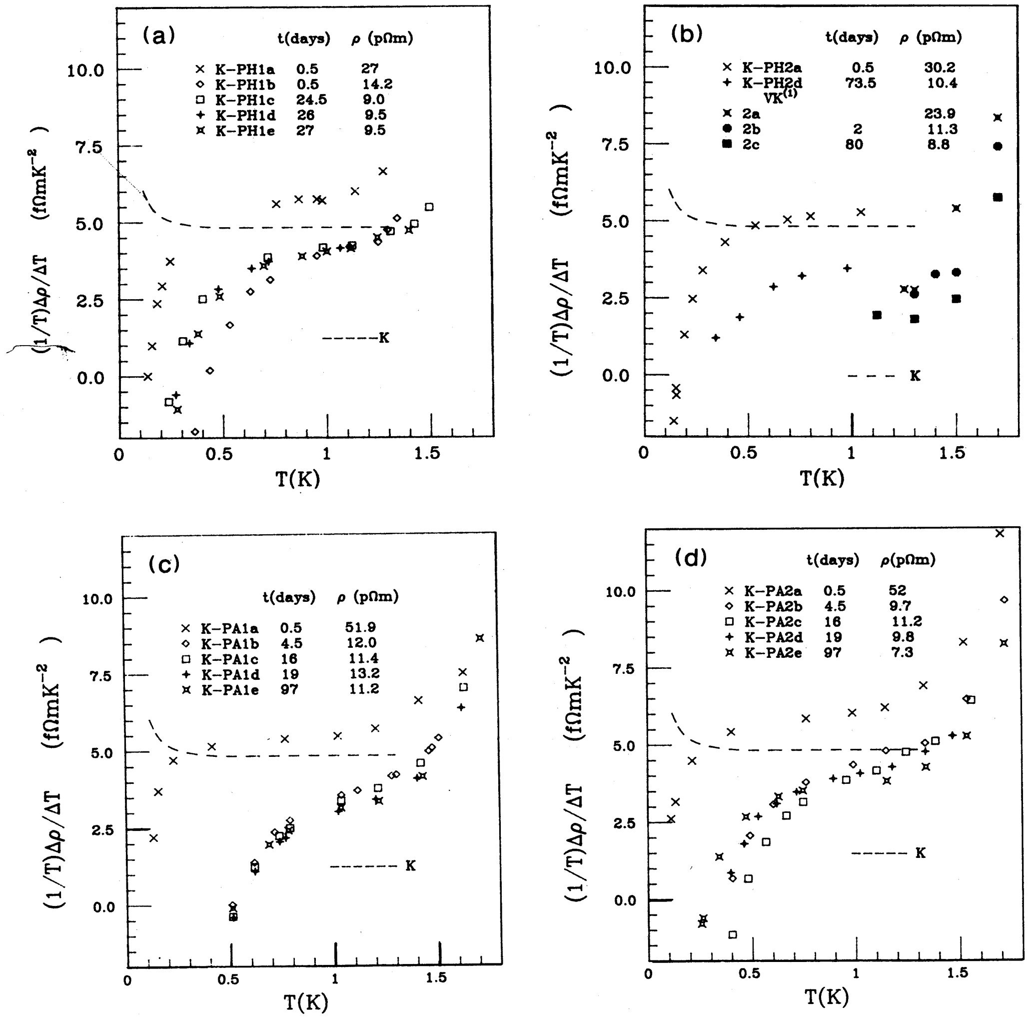

FIG. 2. $(1/T)(\Delta\rho/\Delta T)$ vs T for four polyethylene-clad K samples subjected to annealing treatments. Panel (b) also contains data for one of VK's (Ref. 1) polyethylene-clad samples.

A'. Contamination was introduced (a) by holding the samples at room temperature for extended time periods, (b) by heating the samples to $T \geq 323$ K, or (c) by cold-working surface contamination into the samples by rolling a cylinder over the polyethylene tubes. All of these procedures took place under circumstances where the sample surfaces could be contaminated by oxygen.

RO measured, down to 0.5 K the resistivity of $d=0.8$ mm K wires wound in grooves around a Teflon holder. Their samples also showed decreases in both ρ_0 and in the magnitude of $\rho(T)$ in the vicinity of 1 K with annealing time. The decreases they saw in ρ_0 were similar to those seen by VK. However, for $T \leq 1.2$ K they reported an apparent $T^{3/2}$ variation of $\rho(T)$, rather than the T^2 behavior expected from Eqs. (1). As already noted, we believe that this deviation from T^2 behavior is associated with a "size effect" discussed elsewhere;[7,8] we consider in this paper only the ρ_0 annealing behavior that RO observed.

The first evidence that contact with polyethylene could produce a Kondo-like anomaly was found by HE in our laboratory in the process of studying effects of plastic deformation upon both $\rho(T)$ and G in K to temperatures below 0.1 K. They used insulating films to isolate the samples electrically from two metal blocks between which the samples were squashed. When various forms of polyethylene were used as the insulating films, the sample surfaces in contact with the film remained shiny, but below about 0.2 K a resistance minimum appeared [Fig. 10 in Ref. 10; more recent data are shown in Figs. 2(a)–2(d)]. This resistance minimum manifests itself as a change in sign of $d\rho/dT$. In addition, a low-temperature anomaly appeared in G. In contrast, when Teflon was used for the films, the sample surfaces in contact with the Teflon became black, but no anomalous behavior occurred in either $d\rho/dT$ or in G (we will see later that ρ_0 does decrease with room-temperature annealing in samples enclosed in Teflon tubes). The $d\rho/dT$ and G anomalies found for samples in contact with polyethylene both had forms consistent with a Kondo effect, which led HE to propose the Kondo effect as a likely source of these anomalies. Our current measurements were undertaken both to elucidate the nature and characteristics of this Kondo-like effect and to try to understand the differences between the rather simple behavior we had found in free-hanging pure-K samples (Fig. 1), and the more complex behaviors reported by VK, LY, RO, and HE for samples in contact with plastics.

C. Theories stimulated by the data of VK, LY, and RO

The theories developed to explain the data of VK, LY, and RO all started from two premises: (1) that the observed behaviors were representative of pure bulk K free from significant perturbations except for unavoidable defects in the samples, and (2) that all of the data had to be explained with a single mechanism.

The first theory published started with the presumption of a CDW ground state for K. Bishop and Overhauser[4] (BO) argued that the approximately $T^{3/2}$ variation of $\rho(T)$ reported by RO should be interpreted as evidence that the scattering process which dominates $\rho(T)$ in K below 1.3 K is not scattering of electrons by other electrons, as assumed in Eqs. (1), but rather scattering of electrons by phasons—excitations of the CDW ground state. They showed that the form of the RO data could be fitted by a model of electron-phason scattering rather similar to the Bloch-Grüneisen model of electron-phonon scattering, but with a characteristic temperature of about 6 K instead of the Debye temperature of ~ 100 K for K.[13] The variation in magnitude of the RO data was attributed to changes in the orientation of CDW domains within the samples upon annealing, since the magnitudes of both ρ_0 and $\rho(T)$ were expected to vary with domain orientation. This model was generalized by Bishop and Lawrence,[5] who argued that the presence of a CDW ground state had two effects on $\rho(T)$ in K at low temperatures: (1) it added the electron-phason scattering term of BO to Eq. 1(a), and (2) it produced strong anisotropy in the coefficient A as a function of CDW-domain orientation. They argued that the observed changes in ρ_0 and $d\rho/dT$ in the vicinity of 1 K seen by VK and LY were due to changes in the orientations of the CDW domains produced by changes in the defect structures of their samples.

KW (Ref. 6, Kaveh and Wiser) developed an alternative model which assumed that the data of VK, RO, and LY were all dominated by electron-electron scattering below 1.2 K, and that the primary issue was to explain the different systematic changes of A' with changes in ρ_0 that these authors had seen. KW noted that the T^2 coefficient of electron-electron scattering in Eq. 1(a) consists of two components, one due to normal electron-electron scattering (NEES), in which a reciprocal-lattice vector is not involved, and the other to umklapp electron-electron scattering (UEES), in which a reciprocal-lattice vector participates. We can thus write A in Eq. (1a) as $A = A_N + A_U$, where A_N is due to NEES and A_U to UEES. For a metal with a perfectly spherical Fermi surface, and completely isotropic impurity scattering, both A_N and A_U are zero. Deviations of the Fermi surface from sphericity cause A_U to become nonzero, but in the usual approximation[21] A_N remains zero so long as the dominant scattering process for electrons is isotropic. KW noted that the nearly spherical Fermi surface of K should make A_U fairly small, so that if highly anisotropic scattering should occur, it was plausible that A_N could become much larger than A_U. In such a case, a change in the dominant scattering process from isotropic to highly anisotropic would produce a large increase in A (and thus in A'). They ascribed the large decreases in A' seen with decreasing ρ_0 by VK and by RO, and with increasing ρ_0 by LY, to a common source—reductions in the anisotropy of the dominant scattering mechanism. For the VK and RO data, they postulated that the observed decreases in both ρ_0 and A' were due to the annealing out of dislocations and other extended defects which they asserted produced highly anisotropic scattering. They argue that the annealing process thus produced a decrease both in ρ_0 and in the fraction of anisotropic scattering in the system. For the LY data, in contrast, they postulated that contamination led to an increase in isotropic impurity scattering, thus increasing ρ_0 but decreasing both the fraction of anisotropic scattering and A'.

D. Characteristics of a Kondo effect

When a dilute concentration of a magnetic impurity is present in a nonmagnetic host metal, it is often found that the resulting alloy displays a resistivity minimum and a thermoelectric anomaly, both of which are strongly affected by application of a magnetic field. These properties characterize the Kondo effect.[22] The Kondo effect arises from an exchange interaction between the spins of conduction electrons and the localized moments associated with the magnetic impurities.

In the simplest case the Kondo effect contributes a term to $\rho(T)$ that rises with decreasing temperature as $-\ln(T)$ for $T = T_K$ (T_K is the Kondo temperature) and eventually becomes temperature independent for $T \ll T_K$. Adding this term to Eqs. (1), one expects at temperatures where the logarithmic form is appropriate, a resistivity of the form

$$\rho(T) = A'T^2 - C\ln(T) . \tag{5}$$

Concurrently, the negative thermopower S exhibits a very broad minimum near T_K.[18] Thus in this temperature region S is essentially independent of T. Using Eq. (3) we anticipate that this behavior of S will introduce a term in the thermoelectric ratio G of the form DT^{-1}. That is,

$$G = G_0 - FT^2 - DT^{-1} . \tag{6}$$

According to this relation, the Kondo anomaly in G should manifest itself as a negative divergence of G at low temperatures.

If a large enough magnetic field B is applied, both the $\ln T$ term in $\rho(T)$ and the T^{-1} term in G should be significantly reduced. The field required is $B > k_B T_K / \mu_B$ or $k_B T / \mu_B$, whichever is larger.[23] k_B is Boltzmann's constant and μ_B is the Bohr magneton.

Traditionally, the anomaly in S or G has been more obvious than that in ρ. However, with our very high measuring precision for ρ (less than 1 part in 10^7), we may expect, and do find, that the anomaly in ρ is more clearly discerned.

III. EXPERIMENTAL PROCEDURES AND SAMPLES

We have measured a variety of samples, including free-hanging, bulk bare K and dilute K-Rb alloys; K samples encased in polyethylene and Teflon tubes; K samples in contact with various polyethylene-containing plastics, Teflon, Kel-*F*, and paraffin oil; and a dilute K-Rb alloy in a polyethylene tube.

As in our previous studies,[16] two samples were always measured together, each serving as the reference for the other. Details of the measuring techniques are given elsewhere.[16,24]

To test for effects of different atmospheres, samples were prepared in both He and Ar atmospheres; as we shall show, no significant differences were found. The samples for which effects of room-temperature annealing were studied in detail were melted in a glove box, drawn up with a syringe into $d = 0.9$ or 1.6 mm plastic tubing while molten, and then allowed to solidify in the tubes. Potential leads of the same material as the samples were attached to the samples either by slicing holes through the tubes and touching the sticky leads to the sample body, or by pressing the leads onto the samples just outside the ends of the plastic tubes; again, both techniques yielded similar results. Detailed information about our samples prepared in contact with various plastics are given in Table I. The labels H and A indicate that the ambient atmosphere was helium or argon, respectively, during preparation of the samples in the glove box. P or T indicates that the sample was clad in polyethylene or Teflon tubing, respectively.

A special superconducting solenoid was constructed to apply a magnetic field to a polyethylene-clad sample. Because the superconducting quantum-interference device (SQUID) used as a detector in these measurements can detect very small currents, the sample had to be rigidly attached to the magnet so that mechanical vibrations would not induce a changing magnetic flux in the SQUID circuit. Copper-clad, 0.1-mm-diam NbTi superconducting wire was wound on a copper coil form of 0.3 cm i.d. and 4 cm length. The sample inside its polyethylene tube was pushed through the 0.3-cm hole in the form, and potassium voltage and current leads were cold-welded to the ends of the sample where it protruded from the form. The form was thermally anchored to one end of the sample. Since the magnetic field applied to the sample was not uniform, we quote its average value.

IV. EXPERIMENTAL DATA AND ANALYSIS OF THE ISSUES DEFINED IN SEC. I

A. Decreases in ρ_0 with room-temperature annealing

We see from Table I and Figs. 2 and 3 that the behavior of ρ_0 for our samples in both Teflon and polyethylene

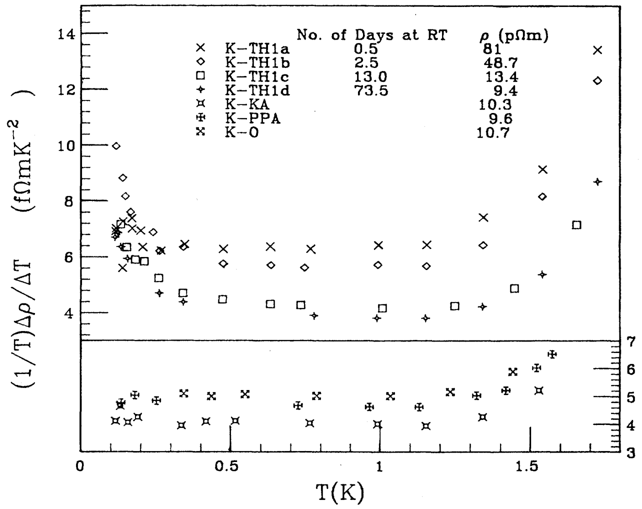

FIG. 3. $(1/t)(\Delta\rho/\Delta T)$ vs T for various K samples that did not show Kondo-like anomalies, including Teflon-clad (K-*TH*) samples.

tubes is very similar to that found by both VK and RO. The values of ρ_0 were large (i.e., 27–81 pΩ m) when the samples were initially prepared, and decreased to values representative of unconstrained samples (i.e., 7–11 pΩ m) as the samples "annealed" at room temperature. For the samples in polyethylene, most of the decrease occurred within the first 2–4 d after sample preparation, just as found by VK for their samples in polyethylene. For the samples in Teflon, in contrast, the decrease was more gradual, as also found by RO for samples wound on a Teflon cylinder. No comparable annealing effects on ρ_0 were seen in either free-hanging K samples (Fig. 1) or in samples simply pressed against plastic (Table I). In these cases, ρ_0 reached ultimate values of ≈ 10 pΩ m the first time the samples were cooled.

We note that both Teflon and polyethylene contract

TABLE I. Important sample parameters. The fifth column refers to the total time that the samples were allowed to "anneal" at room temperature.

	Encapsulated in	ρ_0 (pΩ m)	d (mm)	t(RT) (d)	Kondo effect
K-*PH*1a	polyethylene	27.0	1.6	0.5	yes
K-*PH*1b	polyethylene	14.2	1.6	11.0	yes
K-*PH*1c	polyethylene	9.0	1.6	24.5	yes
K-*PH*1d	polyethylene	9.5	1.6	26	yes
K-*PH*1e	polyethylene	9.5	1.6	27	yes
K-*PH*2a	polyethylene	30.2	0.9	0.5	yes
K-*PH*2b	polyethylene	11.0	0.9	2.5	yes
K-*PH*2c	polyethylene	11.7	0.9	13	yes
K-*PH*2d	polyethylene	10.4	0.9	73.5	yes
K-*PA*1a	polyethylene	51.9	0.9	0.5	yes
K-*PA*1b	polyethylene	12.0	0.9	0.5	yes
K-*PA*1c	polyethylene	11.4	0.9	16.0	yes
K-*PA*1d	polyethylene	13.2	0.9	19.0	yes
K-*PA*1e	polyethylene	11.2	0.9	(97.0)	yes
K-*PA*2a	polyethylene	52.0	1.6	0.5	yes
K-*PA*2b	polyethylene	9.7	1.6	4.5	yes
K-*PA*2c	polyethylene	11.2	1.6	16	yes
K-*PA*2d	polyethylene	9.8	1.6	19	yes
K-*PA*2e	polyethylene	7.3	1.6	97	yes
K-*TH*1a	Teflon	81.0	1.5	0.5	no
K-*TH*1b	Teflon	48.7	1.5	2.5	no
K-*TH*1c	Teflon	13.4	1.5	13	no
K-*TH*1d	Teflon	9.4	1.5	73.5	no
VK (Ref. 1)					
K-2a	polyethylene	23.9	0.9		?
K-2b	polyethylene	11.3	0.9	2	?
K-2c	polyethylene	8.8	0.9	80	?
	In contact with				
K-2*he*[a]	Handiwrap®	19.5	0.9	3	yes
K-3*h*[a]	Handiwrap®	10.4	0.9	0.5	yes
K-4*h*[a]	parafilm	10.5	0.9	4	yes
K-7*h*[a]	polyethylene	12.4	0.9	1	yes
K-*KA*	Kel-*F*	10.3	1.5		no
K-*PPA*	potential leads polyethylene	9.6	1.5		no
K-*O*	oil dripped on	10.7	1.5		no
K-*S* (single crystal)	grown in oil	9.2	1.5		yes
K-*PA*Rb (K-Rb)	polyethylene	86.0	1.6	3	yes

[a] Reference 10.

more than K upon cooling. Since we see no time-dependent changes in ρ_0 in free-hanging K samples, we must conclude that the behavior of ρ_0 in our samples in both Teflon and polyethylene was due to constraints imposed by the surrounding tubing. Similar constraints were certainly present for VK's samples, and produced very similar results. In other cases, however, the situation was not so simple. Thus while RO saw similar behavior in ρ_0 to those we report for samples in Teflon tubes, it is not clear how the grooved Teflon cylinders onto which RO's samples were wound constrained their samples, since we have just noted that Teflon contracts more than K on cooling. Furthermore, Woods *et al.*[25] have found even more dramatic changes in ρ_0 for K samples ≈ 6 cm long laid loosely in straight grooves cut in a Teflon cylinder and positively supported only at the current and voltage terminals. On the other hand, LY reported no anomalously large values of ρ_0 in their K samples constrained in polyethylene. These samples had values of $\rho_0 \approx 3-5$ pΩ m, about half as large as achieved by anyone else.

With these contradictory results in hand, we can only make the following comments about our data and the comparable data of VK. First, we believe that the production of dislocations and other debris by stress can only play a very minor role. We know from our experiments on stressed K (Ref. 10) that strains of 50% only increase ρ_0 by ≈ 10 pΩ m. In cooling samples encapsulated in either polyethylene or Teflon the strain is $\approx 1\%$, which should cause ρ_0 to increase by less than 1 pΩ m. This value is 2 orders of magnitude too small to explain the value of $\rho_0 \approx 81$ pΩ m seen in our Teflon-enclosed sample upon its first cooling.

If unusually high values of ρ_0 are not caused by dislocations and related defects, then they must be caused by impurities. These must either diffuse into the K from the plastic or already be in the K. The former alternative is difficult to accept, since it would require the diffusion process to reverse itself in order to make ρ_0 subsequently decrease with extended annealing at room temperature. We are thus left with the latter, which is plausible as we now show.

According to the manufacturer's analyses, the typical impurity content of K is at least 50 ppm—sufficiently large to account for the highest values of ρ_0 we have observed if the impurities were in solution. It has been known for some time that the R for "pure" K is much higher than the impurity content would lead one to expect.[26] The assumption is usually made that the impurities precipitate out during the cooling process, if it is not too rapid.[26,27] A possible explanation for unusually high values of ρ_0 is that enclosing our samples and VK's in plastic inhibited this precipitation. Alternatively, the high values of ρ_0 could be due to dissolved gases held in solution by the surrounding plastic, but our experience tends to suggest that this contribution is too small.

B. Decreases in $d\rho/dT$ in the vicinity of 1 K with room-temperature annealing and cold-working

Figures 2–4 show the behavior of $(1/T)(d\rho/dT)$ below 1.5 K as a function of annealing time at room temperature for samples constrained in polyethylene and Teflon tubes. The samples studied and the circumstances of their measurements were as follows.

Sample K-*PH*1, enclosed in a $d=1.6$ mm polyethylene tube, was measured five times with anneals under He gas and partial vacuum. Its reference sample was a bare K sample which showed no anomalies. After the second measuring run, sample K-*PH*1 was taken out of the sample can and stored in the glove box. It was remounted just before the third run, along with a new bare K sample as reference. The $(1/T)(d\rho/dT)$ data for sample K-*PH*1 are shown in Figs. 2(a) and 4(a).

Sample K-*PH*2, in a $d=0.9$ mm polyethylene tube, was measured four times with anneals under He gas. Its reference sample was K-*TH*1, which was inside a $d=1.5$ mm Teflon tube. After the third run, one of the connections to K-*PH*2 broke, and the sample can was opened and the sample repaired. It was remounted just before the fourth run using the same reference sample. The $(1/T)(d\rho/dT)$ data for sample K-*PH*2 are shown in Figs. 2(b) and 4(b) along with the data of VK (Ref. 1) in Fig. 2(b). The $(1/T)(d\rho/dT)$ data for sample K-*TH*1 are shown in Fig. 3.

Samples K-*PA*1 and K-*PA*2, in $d=0.9$ and 1.6 mm polyethylene tubes, respectively, were measured together five times with anneals under Ar. To test for the rolling effect reported by LY, both of these samples were taken out of the sample can after the third run, rolled with a metal cylinder to simulate the cold-working procedure used by LY, left at room temperature for about 20 h, and then remounted just before the fourth run. After the fourth run, both samples were again taken out of the sample can, and later remounted for the fifth run. The $(1/T)(d\rho/dT)$ data for these two samples are shown in Figs. 2(c), 2(d) and 4(c), 4(d).

We see from Figs. 2 and 3 that the magnitudes of $d\rho/dT$ in the vicinity of 1 K decreased with annealing time at room temperature for samples in both Teflon and polyethylene, and that there were no significant systematic differences between the behaviors seen with $d=0.9$ and 1.6 mm samples in polyethylene or for samples prepared and cooled in He gas or in Ar.

With these data in hand, we now turn to consideration of the anomalous behaviors reported by RO,[2] VK,[1] and LY,[3] in the order listed.

(1) The data of RO. For our sample K-*TH*1 in Teflon, both the decreases in ρ_0 and in $(1/T)(d\rho/dT)$ occurred over many days and were consistent with decreases in A' due simply to the $B\rho_0 T^2$ term in Eq. 1(a). For their samples wound on Teflon holders, RO also saw decreases in both ρ_0 and $d\rho/dT$ in the vicinity of 1 K which took place over several days. As indicated in Sec. II B, the decreases in ρ_0 that they saw were consistent with what we see. On the other hand, the decreases in $d\rho/dT$ that they saw were larger than those to be expected simply from the $B\rho_0 T^2$ term in Eq. 1(a). We have argued elsewhere[7,8] that the behavior of RO's $d\rho/dT$ data can be understood in terms of a "size-effect" anomaly in bare thin wires subject to surface contamination that causes $d\rho/dT$ in the vicinity of 1 K to be reduced from its value in bulk samples. We note here only that RO's thin wires were sub-

ject to contamination in a fashion similar to ours,[7,8] and that the magnitude of such an anomaly is expected[8] to increase as the material of the wire becomes purer. This latter behavior could explain the substantial reduction in $d\rho/dT$ in RO's samples as ρ_0 decreased upon room temperature annealing.

(2) The data of VK. In contrast to its more leisurely decrease in Teflon, ρ_0 in both our samples in polyethylene and VK's decreased rather quickly ($\sim$2–4 d) to its ultimate value. However, large decreases in $d\rho/dT$ continued for weeks. These extended-time decreases in $d\rho/dT$ are much too large to be explained by the small $B\rho_0T^2$ contribution to $\rho(T)$ due to the very small concurrent changes in ρ_0. We conclude that the behavior of $d\rho/dT$ for VK's samples in polyethylene must involve, in addition to the $B\rho_0T$ term, another effect, the form of which is shown by our data below 1 K in Figs. 2 and 4. We see in these two figures, that below 1 K, $(1/T)(d\rho/dT)$ becomes rapidly more negative with decreasing temperature, and that the magnitude of the decrease in $(1/T)(d\rho/dT)$ at any given temperature becomes larger the longer the samples remain in contact with polyethylene at room temperature. We argue that the decrease in $d\rho/dT$ in the vicinity of 1 K seen by VK upon extended annealing at room temperature is due primarily to systematic growth of the "high-temperature tail" of this larger lower-temperature anomaly.

(3) The data of LY. LY reported finding decreases in A' with increasing ρ_0 produced either by contaminating their samples or by cold-working the samples by rolling metal rods over the outsides of the polyethylene tubes. These procedures both produced white contamination on the surfaces of their samples and moved this contamination from the surface of the sample into its body. In an attempt to stimulate the behavior they observed after cold-working, we also rolled a cylindrical rod over two of

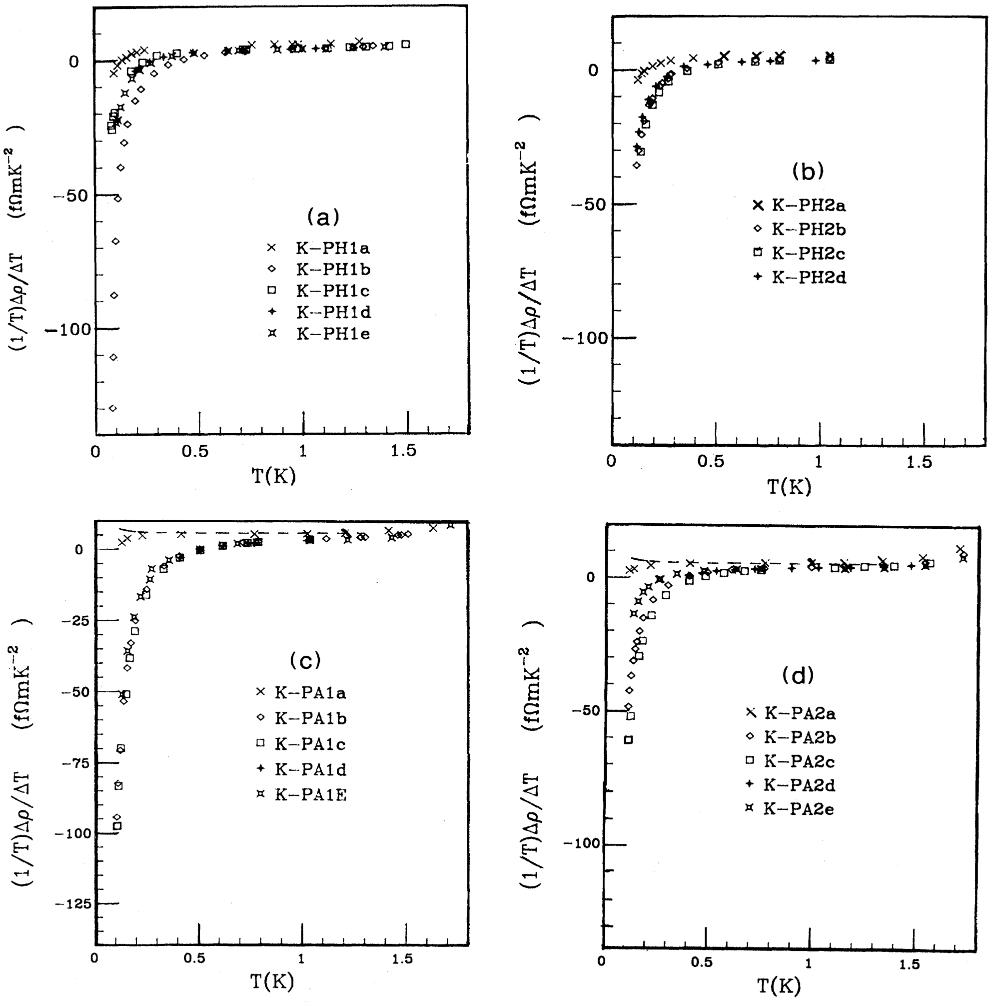

FIG. 4. The lowest-temperature behavior of $(1/T)(d\rho/dT)$ vs T for the polyethylene-clad K samples of Fig. 2, showing the full extent of the low-temperature anomaly.

our polyethylene-enclosed samples at room temperature [K-*PA*1c and K-*PA*2c in Figs. 2(c) and 2(d), respectively]. However, we did not obtain consistent results for the two samples. In one case [Fig. 2(c)] ρ_0 increased and $d\rho/dT$ decreased between 1 and 1.3 K (where our measurements overlapped those of LY), similar to what LY saw. However, in the other case [Fig. 2(d)], ρ_0 decreased and, in the temperature range of overlap, $d\rho/dT$ remained essentially unchanged. We presume that the differences in behavior between our results and LY's were due primarily to the fact that the surfaces of our samples remained shiny at all times, whereas theirs were white. It was exactly the moving of white surface contamination into the body of the sample which they put forward as evidence of increasing contamination after the cold-work.

LY attributed both the increases in ρ_0 and the decreases in A' that they observed to the introduction of contamination by both the heating and the cold-work, and described their results in terms of the model of KW. We feel that our data shown in Figs. 2 and 4 indicate that the situation of samples enclosed in polyethylene is more complex. When our results are combined with data of HE,[10] which show that dislocations produce a much more complex effect upon $d\rho/dT$ than predicted by KW, it seems very unlikely that the behavior of LY's data is due to the KW mechanism. Rather, we propose that the major portion of the decreases in A' found by LY was due to effects of the "high-temperature tail" of the lower-temperature anomaly shown in Figs. 2 and 4, which grew in magnitude as LY's samples spent increasing time in contact with polyethylene. It is plausible that LY's heating of their samples in polyethylene tubes to $T \geq 323$ K enhanced the growth of the Kondo effect, and that additional reductions in $d\rho/dT$ with annealing time were also produced by "size effects" in the presence of surface contamination, as we have described elsewhere.[7,8]

C. Tests for a Kondo effect

We argued in Sec. II that there are three qualitative tests for a Kondo anomaly: (1) a resistivity anomaly associated with a resistivity minimum, (2) a thermoelectric anomaly, and (3) a strong reduction in both anomalies (1) and (2) upon application of a magnetic field. In this subsection we show not only that our data satisfy all three of these qualitative tests, but also that the forms we find for $d\rho/dT$ and $G(T)$ are completely compatible with Eqs. (5) and (6), the equations expected for Kondo behavior.

The fact that $d\rho/dT$ becomes negative in Fig. 2 below about 0.5 K corresponds to the presence of a resistivity minimum. Figure 4 shows that as T is lowered to well below 1 K, $d\rho/dT$ becomes increasingly negative, and seems to diverge as $T \rightarrow 0$ K. If $\rho(T)$ is given by Eq. (5), then $(1/T)(d\rho/dT)$ should have the form

$$(1/T)(d\rho/dT) = A' - CT^{-2} . \tag{7}$$

If Eq. (7) applies to our data for samples in polyethylene tubes, then a plot of $(1/T)(d\rho/dT)$ versus T^{-2} should yield a straight line. Similarly, if Eq. (6) applies to our G data, then a plot of G versus T^{-1} should yield a straight line at temperatures low enough so that the FT^2 phonon-drag term is small. In Fig. 5 we present both of these plots for pure-K sample K-*PAB*1. A different cryostat was used here so that a magnetic field could be applied to the sample. This cryostat permitted this sample to be cooled to below 50 mK, almost twice as

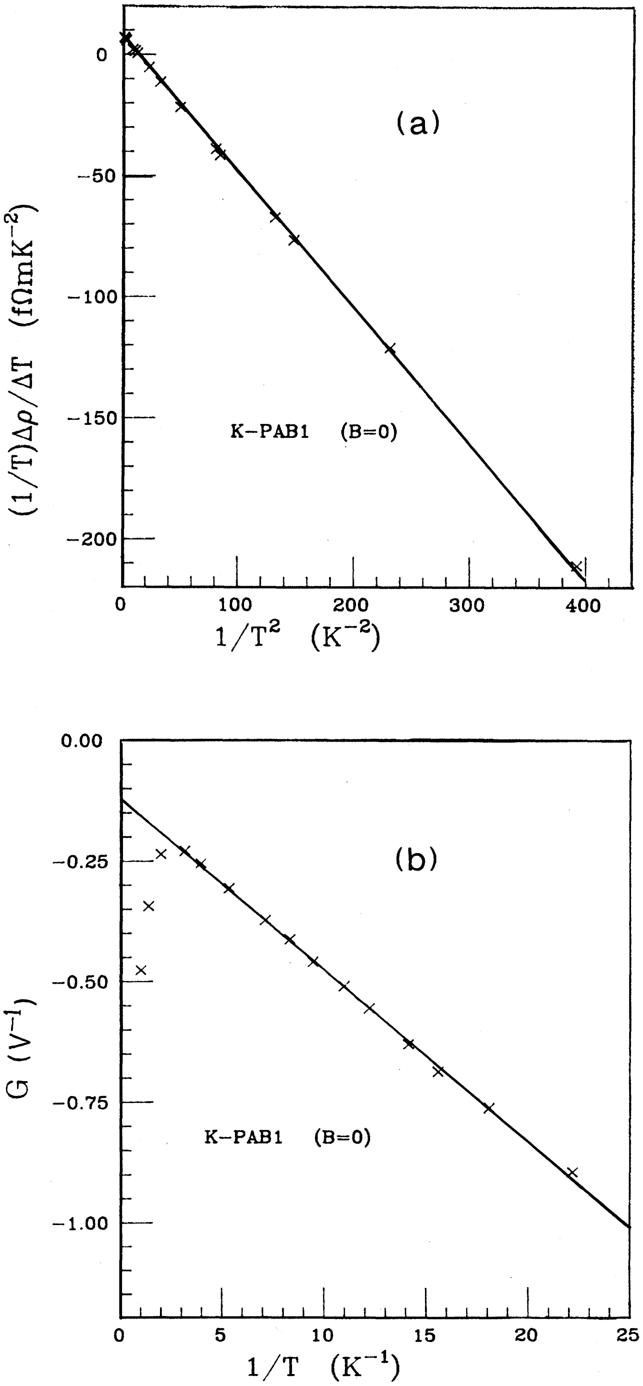

FIG. 5. (a) $(1/T)(\Delta\rho/\Delta T)$ vs T^{-2} and (b) G vs T^{-1} for polyethylene-clad sample K-*PAB*1, which was measured down to 50 mK.

low as the minimum temperature reached with the other samples discussed in this paper. Thus Fig. 5 represents our best test of Eqs. (6) and (7). These zero-magnetic-field data are nicely consistent with the expected straight-line behaviors. From these results we estimate that $T_K \leq 0.05$ K.

Figure 6 shows a plot to test Eq. (7) for all four of the pure-K samples in polyethylene on which we studied the effects of annealing, and Fig. 7 shows a similar plot for a K–0.053 at. % Rb alloy in polyethylene. In both figures the lowest-temperature data are consistent with straight lines.

Figure 8 contains G data for all of our pure-K samples in polyethylene, and Fig. 9 shows that these data follow the expected T^{-1} divergence of Eq. (6) pretty well, especially when the data are viewed from the perspective of the lower-temperature data of Fig. 5.

As further confirmation of Kondo-like behavior, we present in Fig. 10 plots of $(1/T)(d\rho/dT)$ and G versus T for sample K-*PAB* both in zero field and in an average field of $B \simeq 0.1$ T. As expected, this modest field severly attenuates both the $-\ln T$ behavior in $\rho(T)$ and the T^{-1} divergence in G. This strong effect of such a small field is consistent with the estimate given above of $T_K \leq 0.05$ K.

Finally, we note the coefficients of the Kondo terms, D and C in Eqs. (6) and (7), respectively, should each be proportional to the concentration of magnetic impurities. This is obvious for the coefficient C in Eq. (7). To see that it is also true for D in Eq. (6), one must invoke the Gorter-Nordheim rule.[18]

Let ρ_K be the resistivity component associated with the predominant (temperature-independent) scattering of electrons by the magnetic impurities. Thus ρ_K will be proportional to the number of these Kondo impurities. Since we anticipate that the number of these impurities is quite low, we expect to have $\rho_K \ll \rho_0$. The Gorter-Nordheim rule for G then becomes

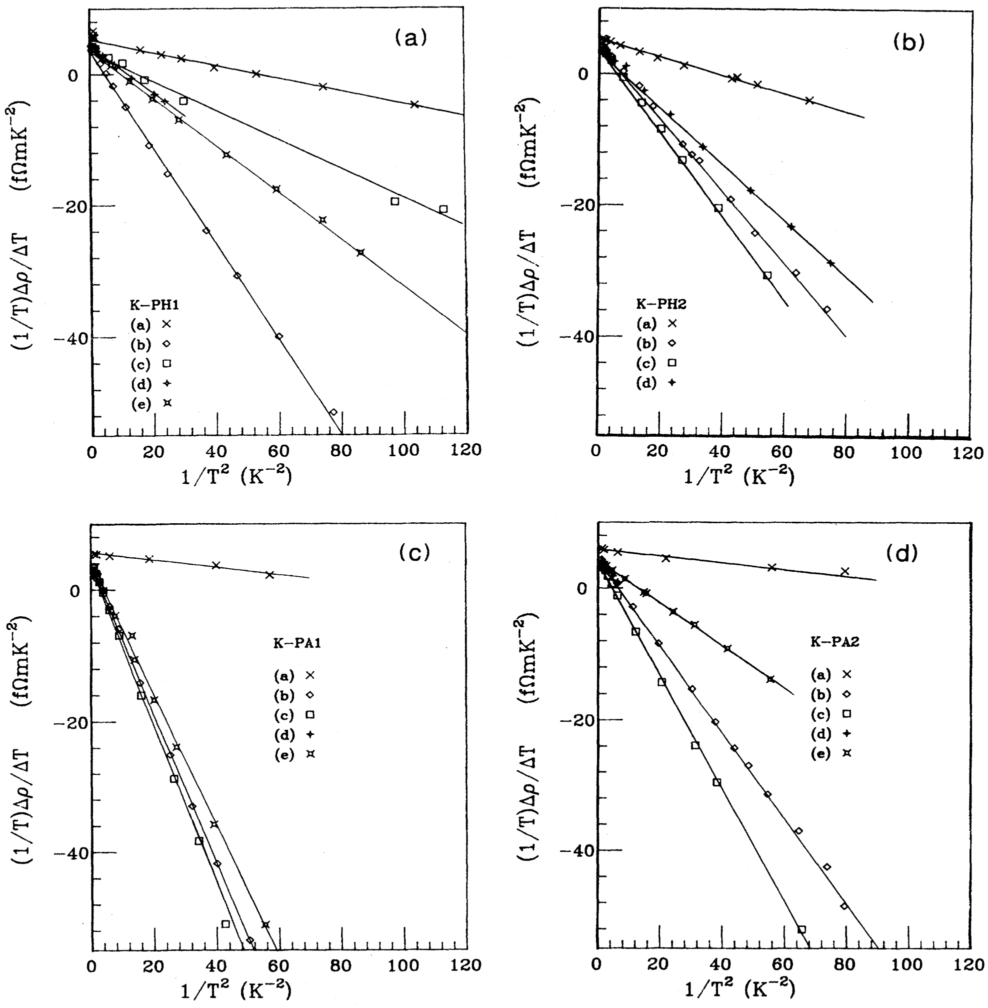

FIG. 6. $(1/T)(\Delta\rho/\Delta T)$ vs T^{-2} for polyethylene-clad K samples of Fig. 2.

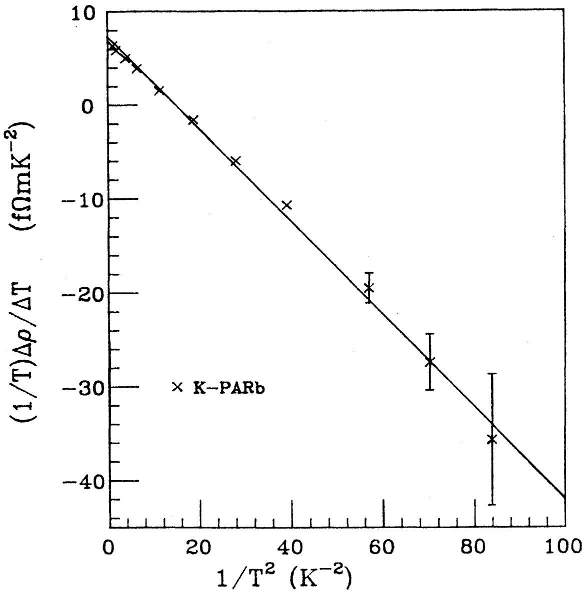

FIG. 7. $(1/T)(\Delta\rho/\Delta T)$ vs T^{-2} for a polyethylene-clad K-0.053 at. % Rb alloy.

$$G = G_0 + (\rho_K/\rho_0)G_K \ , \tag{8}$$

where G_K is the intrinsic (impurity-concentration-independent) Kondo term that diverges as T^{-1}. Comparing Eq. (8) with Eq. (6) under the condition $F \simeq 0$, we obtain $DT^{-1} = (\rho_K/\rho_0)G_K$, so that D should be proportional to ρ_K and, thereby, to the magnetic impurity concentration. Thus, if all of our samples had the same value of ρ_0, we would expect the coefficients C in Eq. (7) and D in Eq. (6) to be linearly related. However, since most of our samples have different values of ρ_0, we must instead plot C versus $\rho_0 D$ to obtain a straight line. Figure 11 shows such a plot. The data are consistent with the expected linear behavior, although the scatter is large. The nonzero intercept of the least-squares line is not statistically significant.

Our data are thus consistent with all of the standard tests for a Kondo effect. It is for this reason that we call this new anomaly "Kondo-like."

D. Conditions for appearance of the Kondo-like anomaly

We have shown in Secs. IV A–IV C that K samples constrained in polyethylene tubes manifest a Kondo-like anomaly below about 1 K, and that the presence of this anomaly is able to account for most of the anomalous behaviors reported by previous observers for $d\rho/dT$ in the vicinity of 1 K. We now describe attempts we have made to establish the conditions under which this Kondo-like anomaly appears and does not appear, and to discover its source.

1. Samples which did not show Kondo-like anomalies

Neither we nor HE observed Kondo-like $d\rho/dT$ or G anomalies in free-hanging bulk samples (Fig. 1). HE (Ref. 10) and, more recently, Yin[28] have observed small G anomalies after application of strain, but the absence of associated Kondo-like $d\rho/dT$ anomalies leads us to believe that their G anomalies are due to a different cause. With one exception to be described in Sec. IV D 2, anomalies were only seen in samples that were in direct physical contact with hydrogen-containing material. Thus, as illustrated in Fig. 3, no Kondo-like $d\rho/dT$ anomalies were seen in a He-cooled sample (K-*TH*1) encased in Teflon tubing. Since both Teflon and polyethylene contract somewhat more than K upon cooling to 4.2 K, and polyethylene contracts more than Teflon, one would expect to see the effect of strain-induced defects such as dislocations more easily in polyethylene-clad K than in Teflon-clad K. This fact argues against the possibility that a Kondo anomaly is present in the Teflon, but is simply swamped by a large upturn in $(1/T)(d\rho/dT)$ due to the presence of dislocations.

No anomalies were seen in an Ar-cooled sample (K-

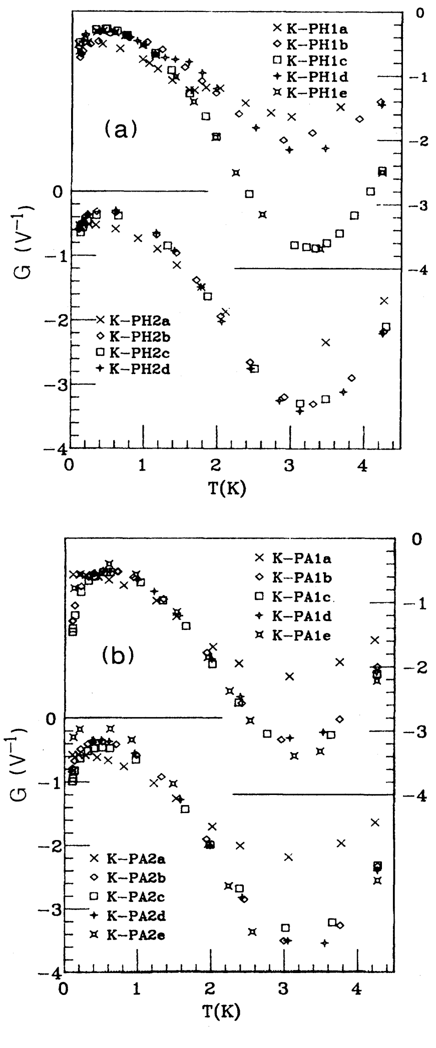

FIG. 8. G vs T for polyethylene-clad K samples of Fig. 2.

KA —Fig. 3) against which pieces of Kel-*F* were pressed; an Ar-cooled $d=1.5$ mm sample (K-*PPA* —Fig. 3) only connected to polyethylene via its potential leads; nor were there any anomalies seen in a K sample that was drawn up into a $d=1.6$ mm polyethylene tube, left there for 3 d, and then reextruded in the solid state as a free-hanging, bare sample. The behavior of this last sample suggests that the Kondo-like scattering may occur at the surface of the polyethylene. Finally, no anomaly was seen in an Ar-cooled sample (*K-O* — Fig. 3) coated at a room temperature with cleaned paraffin oil after mounting. As shown in Figs. 1 and 3, $(1/T)$ $(d\rho/dT)$ is essentially flat from 1.3 K down to at least 0.3 K for all the samples without anomalies. We note that both the Teflon-clad sample K-*TH*1 and the sample in contact with Kel-*F* showed very low values of A' for the lowest values of ρ_0. Except for the strained samples noted above, none of the samples that were free from Kondo-like $d\rho/dT$ anomalies showed any anomalous behavior in G.

2. *Samples which showed Kondo-like anomalies*

To try to establish the general conditions under which the Kondo-like anomaly occurs, and how its size varies

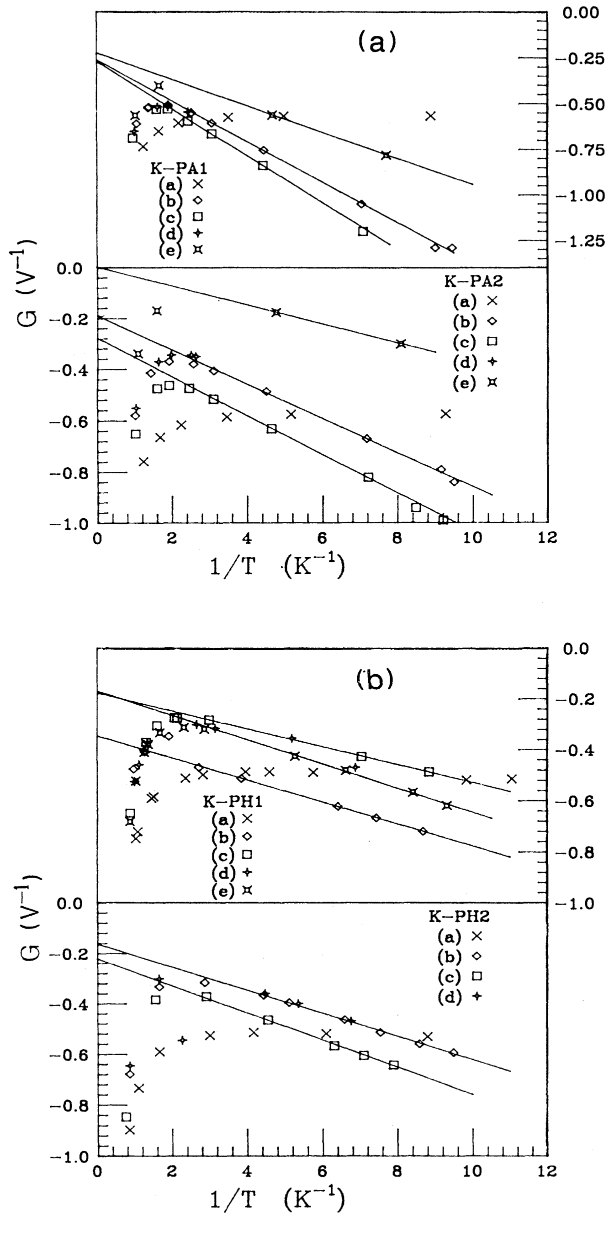

FIG. 9. G vs T^{-1} for polyethylene-clad K samples of Fit. 2.

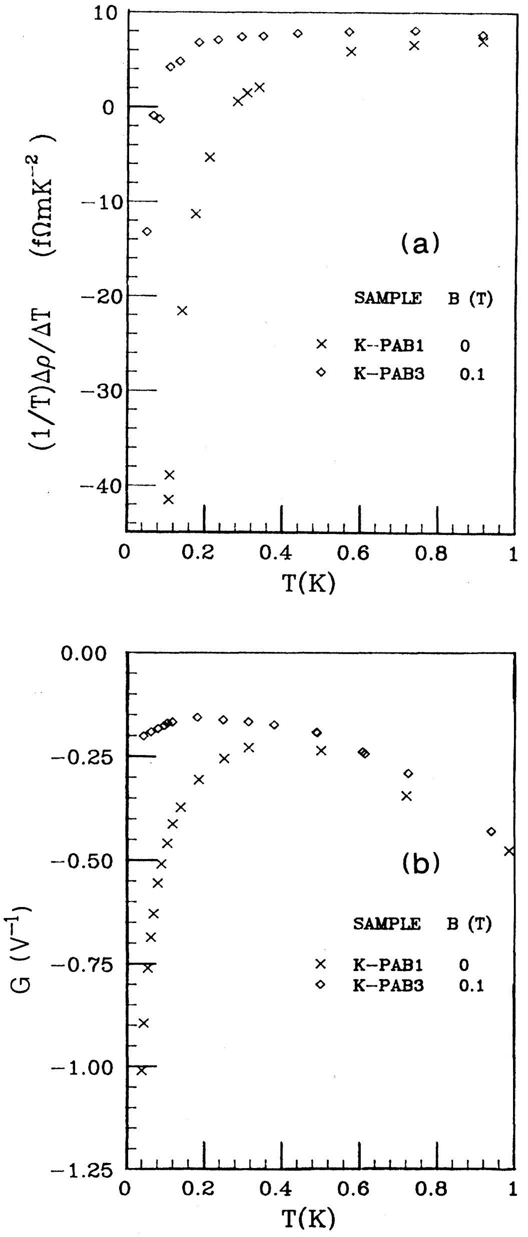

FIG. 10. The effect of a magnetic field on (a) $(1/T)(\Delta\rho/\Delta T)$ and (b) G for polyethylene-clad sample K-*PAB*1.

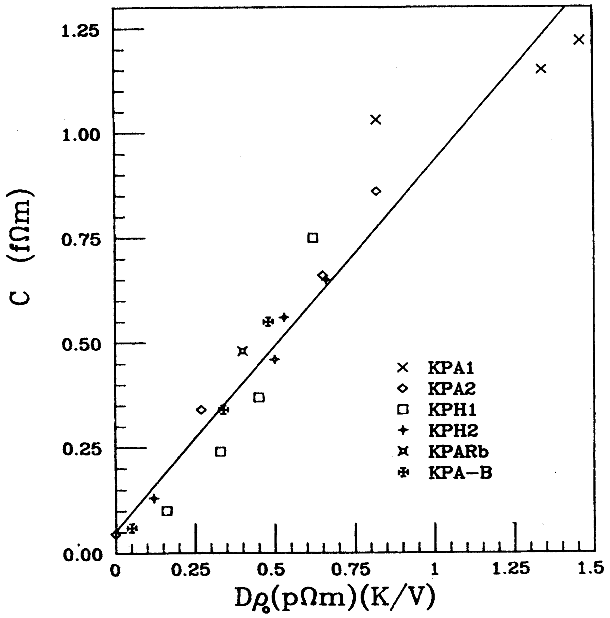

FIG. 11. C vs $D\rho_0$ for all samples that showed Kondo-like anomalies in both $(1/T)(\Delta\rho/\Delta T)$ and G. C determines the magnitude of the Kondo-like anomaly in $(1/T)$ $(\Delta\rho/\Delta T)$, and $D\rho_0$ determines its magnitude in G.

under different conditions, we made a wide variety of tests on samples in contact with polyethylene and one on a sample melted and resolidified under cleaned paraffin oil. The data for samples in polyethylene tubes have already been shown in Figs. 2, 4, 8, and 9.

$(1/T)(d\rho/dT)$ and G data for samples pressed against polyethylene films are shown in Figs. 12(a) and 12(b), respectively. Figure 12 also shows that a small Kondo-like $d\rho/dT$ anomaly was found in a $d=1.5$ mm K wire (sample K-S) that had been melted and resolidified slowly under paraffin oil. No G anomaly was seen in this sample.

Kondo-like $d\rho/dT$ anomalies, but no G anomalies, were also seen (Fig. 12) in one pair of very thin ($d=0.1$ mm) K wires that were not directly touching polyethylene. These two samples were each hung in vacuum between two $d=1$ mm wires that were wrapped with polyethylene. No anomalies were seen in equivalent samples prepared and hung in the same way when Teflon was used as the wrapping material instead of polyethylene.

To see whether the electron mean free path λ for elastic scattering was a critical parameter for the existence of Kondo-like behavior, we drew up into a $d=1.6$ mm polyethylene tube a dilute K–0.053 at. % Rb alloy for which λ was $\simeq 0.02$ mm, and let the sample (K-PARb) sit at room temperature for 3 d before measuring it. We see in Fig. 12 that a large anomaly was found in $(1/T)(d\rho/dT)$ and a small one in G. In Fig. 7 the $(1/T)(d\rho/dT)$ data for K-PARb are shown to agree in form with Eq. (7). The resulting coefficient C of the Kondo term is about 75% of that for sample K-PA2b, a pure-K sample in a $d=1.6$ mm polyethylene tube that had been held at room temperature for the slightly longer time of 4.5 d. Clearly, the size of the Kondo-like anomaly in $(1/T)(d\rho/dT)$ was not significantly changed by reducing λ. The much smaller G anomaly exhibited by K-PARb is simply a consequence of the Gorter-Nordheim rule, Eq. (8), since ρ_0 for the K-PARb sample was about nine times larger than that for sample K-PA2b. Indeed, the ratio $C/(D\rho_0)$ for the K-Rb alloy is in good agreement with the values for the pure-K samples, as shown in Fig. 11. We conclude that λ does not appear to be an important parameter for Kondo-like behavior, at least so long as $\lambda \ll d$.

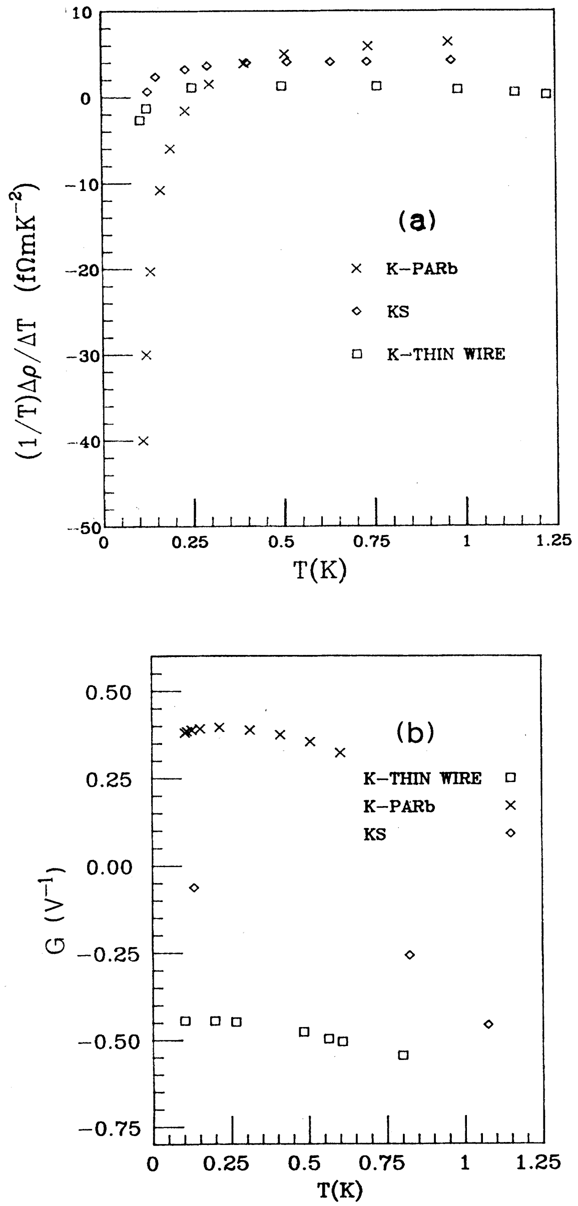

FIG. 12. (a) $(1/T)(\Delta\rho/\Delta T)$ and (b) G for miscellaneous K samples that showed Kondo-like behavior in $(1/T)(\Delta\rho/\Delta T)$ but not always in G.

Since the only obvious difference between polyethylene and other plastics such as Teflon and Kel-*F* is that the former contain hydrogen and the latter do not, we tried explicitly to introduce hydrogen into pure K by bubbling H_2 through molten potassium, both with and without an electrical discharge, and also by heating K to a temperature of 450 K in a H_2 atmosphere. None of these three procedures produced any evidence of anomalies. The last procedure produced a large quantity of white KH, but samples made from the remaining K showed no anomalous behavior in their low-temperature transport properties.

VI. SUMMARY AND CONCLUSIONS

We have observed anomalous behavior in both ρ_0 and $d\rho/dT$ below 1 K in K and K-Rb samples in contact with plastics.

(1) Behavior of ρ_0. We have found that constraining K samples in either polyethylene or Teflon tubing leads to unusally large values of ρ_0 upon initial cooling to 4.2 K, followed by gradual reduction in these values upon room-temperature annealing for days to weeks. We attribute this behavior to the initial retention in solution of impurities due to physical constraint produced by the plastic tubing, followed by slow annealing away of these defects. We argue that similar behavior observed by VK almost surely resulted from the same source. Similar behavior observed by RO and by Woods *et al.*[25] is not easily explainable solely on the basis of constraints.

(2) Behavior of $d\rho/dT$ and G below about 1 K. We have found that physical contact of K or K-Rb with polyethylene leads to anomalous behaviors of $d\rho/dT$ and G below about 1 K which manifest all of the traditional characteristics of a Kondo effect. These include a resistivity minimum, a thermoelectric anomaly, and great sensitivity of both the resistivity minimum and the thermoelectric anomaly to application of a small ($\simeq 0.1$ T) magnetic field. In addition, both $d\rho/dT$ and the thermoelectric ratio G display the temperature dependences expected for a Kondo effect.

We have also found that the magnitudes of the Kondo-like anomalies in both $d\rho/dT$ and G increase with time in contact with the polyethylene at room temperature. We argue that this Kondo-like effect was probably responsible for most of the anomalous behavior in $d\rho/dT$ reported by VK and by LY. We attribute the anomalous behavior of $d\rho/dT$ seen by RO primarily to a "size-effect" anomaly in thin wires which has been discussed elsewhere.

(3) We have attempted to isolate the physical source of the Kondo-like anomaly due to contact between K and polyethylene, without success.

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