Very-Low-Temperature Resistivity Anomaly in *K*Rb, *K*Na, and *Li*Mg Alloys

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High-precision measurements of the electrical resistivities of *K*Rb, *K*Na, and *Li*Mg alloys reveal large anomalous departures from the expected T^2 dependence below ~ 0.5 K. The dependences of the anomaly in *K*Rb upon temperature, residual resistivity, and magnetic field have been studied and, taken together, appear to rule out localization, electron-electron interaction, or Kondo effects as explanations for the anomaly. Two-level–system and charge-density-wave explanations are also considered.

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Potassium (K) is usually, but not universally,[1] assumed to have the simplest electronic structure of any metal at low temperatures, with a nearly spherical Fermi surface entirely within the first Brillouin zone, and no unfilled d or f shells. Rubidium (Rb) has the same valence as K and is 100% soluble in K, and the Rb ion is nearly the same size as K. *K*Rb should thus be the ideal "simple" alloy for making precision studies of very-low-temperature electronic transport.

A few years ago, we showed[2] that below 1 K the temperature-dependent resistivity $\rho(T)$ of *dilute* *K*Rb alloys could be understood as the sum of two terms:

$$\rho(T) = AT^2 + B\rho_0 T^2, \tag{1}$$

where the term AT^2 was attributed to umklapp electron-electron scattering, and the term $B\rho_0 T^2$ was attributed to inelastic electron-impurity scattering. In Eq. (1), ρ_0 is the residual resistivity of the sample.

Since then, we have improved the absolute accuracy of our temperature scale, and also our measuring precision by nearly an order of magnitude, so that we can now detect changes in $\rho(T)$ of ~ 1 part in 10^8. These improved conditions allow us to examine in more detail the form of $\rho(T)$ in alloys at very low temperatures. A reexamination of low-concentration *K*Rb alloys confirmed the general form of Eq. (1), but at the same time uncovered a new resistivity anomaly at the lowest temperatures. In this Letter, we describe the characteristics of this anomaly, and consider whether it can arise from localization, electron-electron interaction, two-level, charge-density-wave, or Kondo effects. We also briefly describe observations of apparently similar anomalies in *K* (1 at.% Na) and *Li* (1 at.% Mg) alloys.

The general procedure for our measurements, involving the use of a SQUID null detector and a high-precision current comparator, is given elsewhere.[3] Details specific to the present measurements will be described in a more complete description of our results.[4] The quantities we measure are the temperature T and $(1/\rho)\Delta\rho/\Delta T = \Delta \ln\rho/\Delta T$, where ΔT is a small temperature difference. We choose to present our results in the form of $(\rho_0/T)d\ln\rho/dT$ vs T, where the finite differences ΔT are chosen to be small enough to allow us to go to the derivative limit. If Eq. (1) were exact, then

$$\frac{\rho_0}{T}\frac{d\ln\rho}{dT} = \frac{1}{T}\frac{d\rho}{dT} = 2A + 2B\rho_0, \tag{2}$$

where we have made use of the fact that $\rho(T) \ll \rho_0$ over the entire temperature range. Plotted in this manner, the data will lie along a horizontal straight line if Eq. (1) is obeyed. We call any departure from a horizontal straight line an anomaly. In Fig. 1 we show plots of our data for several *K*Rb alloys (for greater detail of low-concentration alloys, see Bass *et al.*[5]). For the 0.38 at.% Rb sample, there is an extensive region over which Eq. (1) holds, with a small hint of a down turn at the lowest temperatures. This down turn becomes more pronounced in the 1.3 at.% Rb sample, and in the 9.4 at.% Rb alloy there is no doubt that a substantial low-temperature anomaly exists. In the 23.6 at.% Rb and higher-concentration alloys the low-temperature anomaly becomes still larger, and there is no part of the curve for which we can say that Eq. (1) is obeyed. In the vicinity of 1 K, we also begin to see, in the most concentrated alloys, an electron-phonon resistivity contribution due to a decreasing Debye temperature, details of which will be discussed elsewhere.[4]

In this paper we concentrate on the low-temperature anomaly. We wish to determine how it varies with T, ρ_0, and magnetic field. Suppose

$$\rho = \rho_0 + AT^2 + B\rho_0 T^2 + f(\rho, T), \tag{3}$$

where $f(\rho,T)$ is the anomalous contribution to ρ. Then

$$\frac{\rho_0}{T}\frac{d\ln\rho}{dT} = 2A + 2B\rho_0 + f'(\rho,T)/T,$$

with the assumption again that, apart from terms containing the derivative of ρ with respect to T, $\rho = \rho_0$ to a very good approximation. We now suppose that we can separate the ρ_0 and T dependences of f, i.e.,

$$f(\rho,T) = \rho_0^N f(T).$$

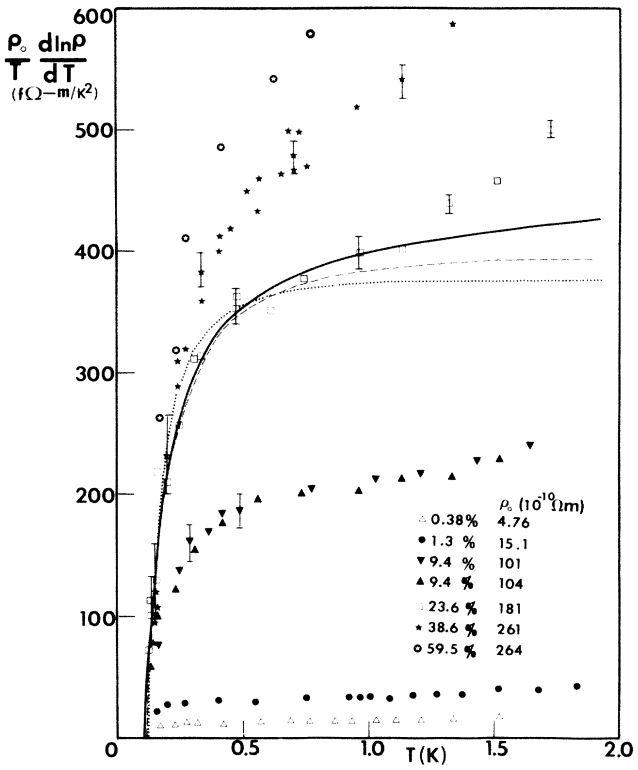

FIG. 1. $(\rho_0/T)d\ln\rho/dT$ as a function of T for K-Rb alloys. The solid curve is a fit of Eq. (7) to the data for the 23.6 at.% alloy. The dashed and dotted curves are fits by the same equation, except that $C\rho_0 T$ is replaced by $D\rho_0 T^{1/2}$ or $E\rho_0 \ln T$, respectively.

We then have

$$\frac{\rho_0}{T}\frac{d\ln\rho}{dT} = 2A + 2B\rho_0 + \rho_0^N f'(T)/T. \tag{4}$$

We first consider the three temperature dependences expected for localization,[6] electron-electron interactions,[6] and the Kondo effect[7]:

$$f(T) = -CT \quad \text{(localization)}, \tag{5a}$$

$$f(T) = -DT^{1/2} \quad \text{(electron interaction)}, \tag{5b}$$

$$f(T) = -E\ln T \quad \text{(Kondo effect)}, \tag{5c}$$

for which $(\rho_0/T)d\ln\rho/dT$ should be proportional to T^{-1}, $T^{-3/2}$, and T^{-2}, respectively. When we plot this quantity as a function of these three powers of T for various samples, we find[4] that the graphs with the T^{-1} abscissa give slightly better fits than those with $T^{-3/2}$, and that the T^{-2} graphs give the worst fits. The solid, dashed, and dotted curves in Fig. 1 illustrate the three fits to the K (23.6 at.% Rb) alloy data. We note that all three curves fall below the data at sufficiently high temperatures. This is due to the onset of the electron-phonon contribution to $\rho(T)$, which is expected to contain both T^5 and exponential terms.

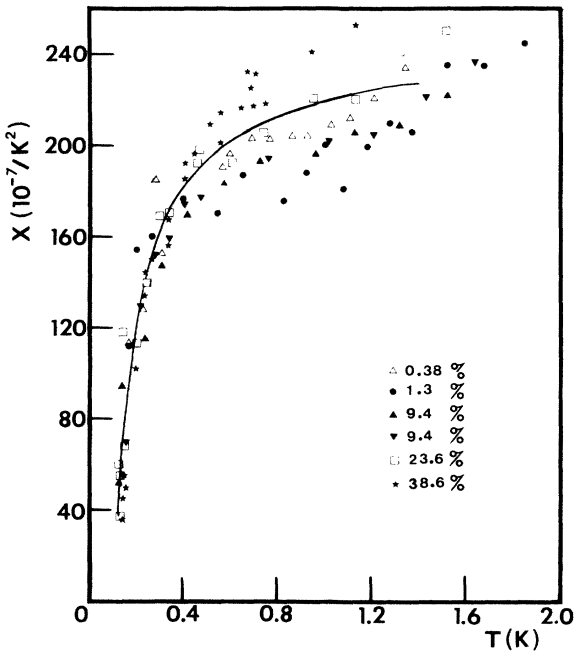

FIG. 2. X vs T for KRb alloys [see Eq. (6)]. The solid curve is a fit of Eq. (7) to the data for the 23.6 at.% alloy.

With two high-temperature terms to adjust, we can fit the entire range of data for each alloy—to within experimental uncertainty—using each of the three forms given in Eq. (5). In the absence of detailed knowledge of the magnitudes of these two electron-phonon terms, we choose to parametrize our results by means of the low-temperature form which, by itself, fits the data over the widest temperature range. This is $f(T) = -CT$.

To examine the ρ_0 dependence of the anomaly, we use Eq. (4) to define

$$X = \frac{d\ln\rho}{TdT} - \frac{2A}{\rho_0} = 2B + \rho_0^{(N-1)} f'(T)/T. \tag{6}$$

In Eq. (6), $2A/\rho_0$ is small for the concentrated alloys, so that we will make little error in X if we take A to have the value derived from the low-concentration data alone. With this value of A, we plot X as a function of T in Fig. 2, and see that the data are nearly independent of ρ_0. Such behavior corresponds to $N = 1$, and means that the anomalous contribution to $(\rho_0/T)d\ln\rho/dT$ is approximately proportional to ρ_0. We therefore offer as an approximate empirical equation for our data

$$\rho = \rho_0 + AT^2 + B\rho_0 T^2 - C\rho_0 T. \tag{7}$$

The solid line in Fig. 2 is a fit by this expression for the 23.6 at.% alloy with $C = 2.6\times10^{-6}$/K, $B = 1.23\times10^{-5}$/K², and $A = 2.4$ fΩ·m/K². The uncertainty in N is difficult to estimate reliably, but several alternative analyses suggest that a value as large as 2,

or as small as $\frac{1}{2}$, is unlikely.

We also examined the effect of applying an inhomogeneous, longitudinal magnetic field of maximum strength 0.2 T to a 9.7 at.% Rb sample. As shown by the open and filled circles in Fig. 3, this field produced no significant change in $d\rho/dT$. In contrast, application of the same field to a Kondo-type sample consisting of K in contact with polyethylene[8] produced changes in $d\rho/dT$ more than an order of magnitude larger than the measuring uncertainty.

To see if the anomalous behavior is unique to *K*Rb alloys, we also measured $(\rho_0/T)d\ln\rho/dT$ below 1.5 K for *K* (1 at.% Na) and *Li* (1 at.% Mg) alloys. We chose the alloy concentration (1 at.%) to be large enough to show effects, if they existed, but small enough that the solubility limits of Na in K and Mg in Li were not exceeded. For both *K*Na and *Li*Mg, we found anomalous behavior qualitatively similar to that for *K*Rb. The triangles in Fig. 3 illustrate for *Li*Mg both the presence of an anomaly and the absence of any effect of a magnetic field upon $d\rho/dT$. We note that Na ions are much smaller than K ions, and that Na has limited solubility in K. *Li*Mg undergoes a phase transition to a different crystal structure upon cooling. From these facts, we conclude that the anomaly is not limited to alloys with (a) ions of about the same size; (b) a wide range of solubility; or (c) the bcc crystal structure of *K*Rb. Finally, we note that in two of the three *Li* (1 at.% Mg) samples studied, $d\rho/dT$ became negative at the lowest temperatures. Such behavior corresponds to a resistivity minimum in these samples.

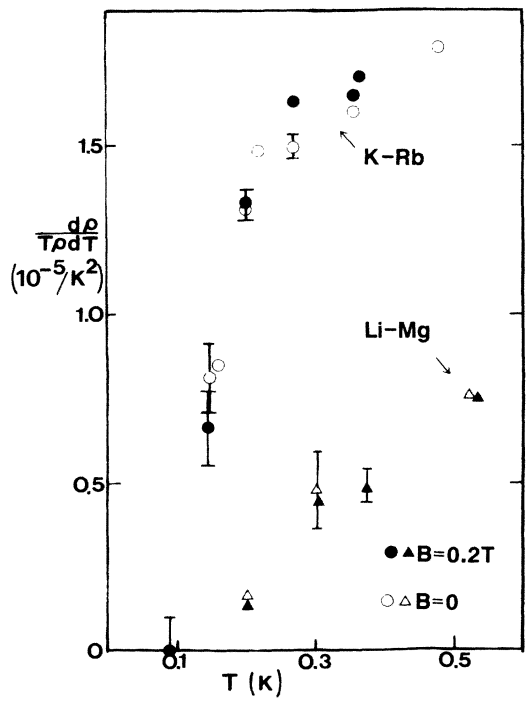

FIG. 3. $(1/T)d\ln\rho/dT$ vs T for a *K* (9.7 at.% Rb) alloy (circles) and a *Li* (1 at.% Mg) alloy (triangles) for magnetic fields of $B=0$ (open symbols) and $B_{max}=0.2$ T (filled symbols).

In the light of these experimental observations we now briefly consider the theoretical models mentioned above. More details will be given elsewhere.[4]

Our data should be in the "weak" localization and interaction regimes.[6] Assuming that the inelastic-scattering time is dominated by inelastic electron-impurity scattering,[2] we would then expect ρ_0^3 and $\rho_0^{5/2}$ dependences for localization[6] or electron-interaction[6] effects, respectively. Since the anomalies in our *K*Rb alloys (1) are at least 2 orders of magnitude larger than predicted, and (2) have a ρ_0 dependence which is close to linear, localization or interaction effects seem to be ruled out. We can obtain predicted magnitudes which cross our data, by extrapolating to the 10^{-6}–10^{-7}-Ω-cm range a phenomenological scaling law[9] that has been used to fit data on samples with ρ_0 ranging from 1 to 10^{-4} Ω cm.[10] In this model, localization and correlations effects are treated equally in the "strong" regime, and $\rho(T)$ is found to be proportional to $T^{1/2}$ and ρ_0^2. Since, however, we are *not* in the "strong" regime, and our *K*Rb data vary more nearly as ρ_0 than as ρ_0^2, this model does not appear to explain our data.

The Kondo effect seems to be ruled out by the facts that a magnetic field causes no change in $d\rho/dT$, and that we find no Kondo-type anomalies in the thermoelectric properties.[4]

Two-level systems were, for a while, proposed to explain similar anomalies in structurally disordered metals.[11] For a two-level system, the simplest expected T dependence of $\rho(T)$ is $\ln(T^2+T_0^2)^{1/2}$. The extra parameter T_0 permits an improvement in the logarithmic fit to $\rho(T)$. Moreover, the anomaly for a two-level system would be expected to be independent of magnetic field. However, it is difficult to see how isolated Rb impurities in K could have two different, nearly equivalent, energy states, and pairs of impurities would be expected to give an anomaly proportional to ρ_0^2.

Finally, Hu and Overhauser[12] have suggested that this anomaly could result from the failure of Migdal's theorem for low-momentum transfer, coupled with the presence of a charge-density wave. Their calculation is, however, still too preliminary for detailed comparison with the data.

In view of the simplicity of the *K*Rb alloy system, and the apparently similar anomalous behavior of $d\rho/dT$ in *K*Rb, *K*Na, and *Li*Mg, we believe that the source of the anomalies we report in this Letter is likely to be fundamental, and thus important to elucidate.

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