Electron-Phonon Umklapp Interaction in the Low-Temperature Electrical Resistivity of Potassium*

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It has long been postulated that, in those metals where the Fermi surface does not touch the Brillouin zone boundary, the electron-phonon umklapp scattering contribution to the electrical resistivity should rapidly disappear at sufficiently low temperatures. New experimental and theoretical evidence for such an effect is reported in the low-temperature electrical resistivity of potassium; the contribution from umklapp processes prevails over that of normal processes above ~2.5°K, but decreases rapidly for temperatures below this.

In those metals where the Fermi surface does not touch the Brillouin zone boundary, there is a lower limit on the wave vector of phonons which can participate in electron-phonon umklapp scattering processes. At very low temperatures, phonons of this sort are frozen from the Bose-Einstein distribution. Thus, it was postulated[1] some time ago that the umklapp contribution to the electrical resistivity of such metals should disappear rather rapidly at sufficiently low temperatures. Various estimates at higher temperatures indicate that the umklapp contribution exceeds the normal electron-phonon scattering contribution. Therefore such a rapid decrease in the umklapp component should be readily observable. This Letter reports the first conclusive evidence for such an effect in the low-temperature electrical resistivity of potassium.[2]

The important role played by electron-phonon umklapp processes in the low-temperature electrical resistivity of the monovalent metals was not realized at the time of the Bloch theory. The results of later calculations[3] have indicated that the electrical resistivity is dominated by umklapp processes and that any freezing out of such processes should occur at much lower temperatures than previously thought, indeed possibly as low as $\sim\frac{1}{30}\Theta_{\text{Debye}}$. To be able to observe such an effect, it was consequently necessary to make very careful resistivity measurements on a pure ideal monovalent metal at quite low temperatures. Potassium was chosen since other investigations had indicated its Fermi surface to be reasonably spherical, facilitating comparison with theoretical calculations, and because it did not have the complicating effects of a martensitic transformation present in some of the other alkalis.

Data were obtained in the 1-25°K temperature range from measurements on polycrystalline samples[4] having residual resistance ratios (RRR $\equiv \rho_{20°C}/\rho_{0°K}$) from ~1000 to ~8000. No significant change in the temperature-dependent part of the resistivity with RRR was observed for RRR values above about 4500, indicating the absence of serious impurity deviations from Matthiessen's rule in this regime.[5] For such samples, the ideal (temperature-dependent) resistivity was obtained from the experimental data by subtracting the observed value of the residual resistivity. The uncertainty in the residual value contributed negligible error to the resultant ideal resistivity at temperatures above 2.5°K. The results, obtained from samples having resistance ratios of 7880, 7440, and 4480, are presented in Fig. 1(a) (the difference between the ideal resistivity values from these samples was less than the width of the solid line in the figure, and hence no attempt has been made to plot individual data points). From this figure it is obvious that the temperature dependence of the resistivity in the 2-4°K range becomes rapidly steeper than the usual T^5 temperature dependence predicted by the Bloch theory (which ignores umklapp processes). As seen in Fig. 1(a), the fall-off in resistivity with temperature exceeds even that of a T^7 dependence near 3°K.

To see if the steep temperature dependence in this regime is indeed a manifestation of the rapid disappearance of the electron-phonon umklapp component it was necessary to perform an accurate theoretical calculation of such effects for comparison with the data. Hasegawa[6] has calculated the temperature dependence of the normal and umklapp contribution to the resistivity of potassium above 8°K. More recently, Rice and Sham[7] have determined the total (umklapp plus normal) resistivity of potassium using a number of different pseudopotentials[8] and the phonon frequencies of Cowley, Woods, and Dolling[9] obtained from neutron inelastic-scattering studies.

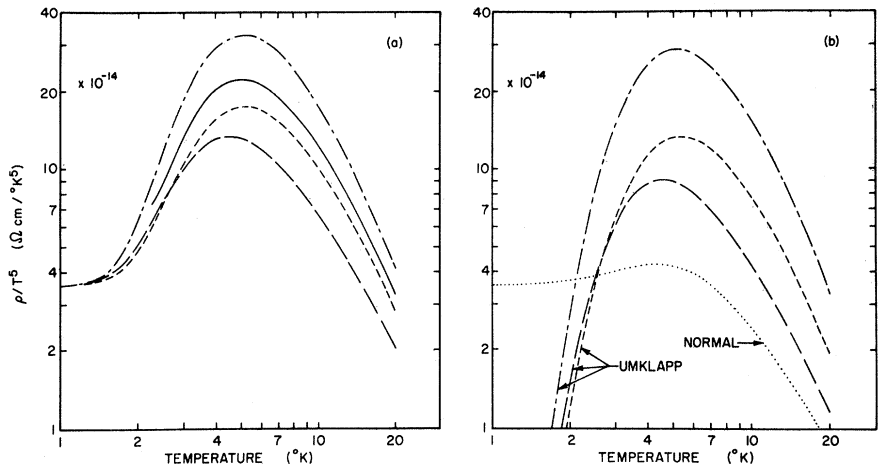

FIG. 1 Comparison of experimental and theoretical results. The resistivity ρ has been divided by T^5 in order to facilitate comparison with a pure T^5 temperature dependence (straight horizontal line). (a) The experimental ideal resistivity (solid line) obtained from samples having residual resistivities $\rho(0)$ of (0.902, 0.954, and 5.531) $\times 10^{-9}$ Ω cm along with the theoretical results for the total (umklapp plus normal) resistivity: long and short dashes, Bardeen pseudopotential; short dashes, lower Lee-Falicov pseudopotential; and long dashes, Ashcroft pseudopotential. (b) The theoretical total-resistivity results explicitly separated into umklapp (dashed lines) and normal (dotted line) components. The normal components from the three pseudopotentials were so nearly identical that they could not be resolved on this scale.

Following the treatment by Rice and Sham, we have calculated the resistivity in potassium due to electron-phonon interaction down to temperatures less than 1°K and have explicitly separated the total resistivity into normal and umklapp contributions.[10] A Kohler variational approach was used in conjunction with the standard trial function proportional to the scalar product of the electron wave vector with the electric field vector. The other principal assumptions were those of an equilibrium phonon distribution, spherical Fermi surface, and states of single, plane-wave electrons.[11]

The theoretical results are compared with the experimental data in Fig. 1. No normalization between experimental and theoretical curves has been made, and, where comparisons were possible, both are independently in good agreement with the work of other investigators.[5] Above ~2° K, the slight variation between the theoretical results for different pseudopotentials is due to the extreme sensitivity of the umklapp component to the form of each pseudopotential near $|\vec{k}_f - \vec{k}_i| = 2k_F$ ($\vec{k}_f$ and $\vec{k}_i$ being respectively the final and initial scattered-electron wave vectors, and k_F the Fermi wave vector). The normal component, on the other hand, varies little since each pseudopotential approaches $-\frac{2}{3}E_F$ as $|\vec{k}_f - \vec{k}_i| \to 0$ (E_F being the Fermi energy). This sensitivity of the umklapp component makes the low-temperature electrical resistivity an ideal experimental test for various pseudopotential models.

Most importantly, a comparison of the theoretical and experimental curves in Fig. 1 shows clearly that the steep temperature dependence in the data below about 4°K is a result of the rapid disappearance of the electron-phonon umklapp component. This conclusion is independent of which pseudopotential was used. The umklapp contribution prevails over that of normal processes for temperatures greater than about 2.5° K, but decreases rapidly for temperatures below this.

It is interesting that further confirmation of this rapid disappearance of umklapp processes at these temperatures results when our data is compared with that of MacDonald, Pearson, and Templeton[12] obtained from low-temperature studies of the *thermoelectric effect* in potassium. They fitted the umklapp component of their data by a decaying exponential function of the form $\exp(-\theta^*/T)$ in the temperature range ~3 to ~1°K and found a characteristic temperature θ^*, of 21°K. When phonons which can participate in umklapp processes are frozen from the Bose-Einstein distribution, they occur only with an exponentially decreasing density. When such is the case, one might expect the umklapp components of different transport coefficients to decrease approximately exponentially in temperature with the *same* characteristic dependence.[13] With this in mind, we have fitted the umklapp component

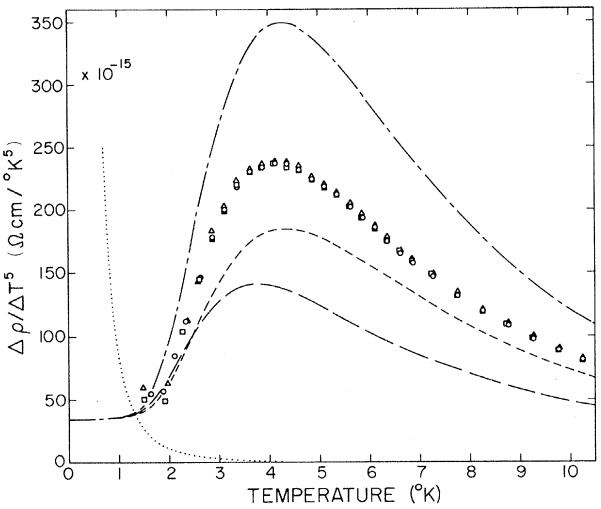

FIG. 2. Slope of the resistivity as a function of temperature, where $\Delta\rho/\Delta T^5 \equiv (\rho_i - \rho_j)/(T_i^5 - T_j^5)$ with i and j any two adjacent data points. The low-temperature experimental data are compared with the results for the total (umklapp plus normal) electron-phonon contribution to the resistivity as calculated from the same pseudopotentials as in Fig. 1 (same curve codes as Fig. 1). Experimental data: circles, $\rho(0) = 5.531 \times 10^{-9}$ Ω cm, wire diam = 1.2 mm; squares, $\rho(0) = 0.954 \times 10^{-9}$ Ω cm, diam = 2.2 mm; and triangles, $\rho(0) = 0.902 \times 10^{-9}$ Ω cm, diam = 2.2 mm. The theoretical estimate of the electron-electron scattering contribution (dotted line) calculated by Lawrence, Ref. 14, has been included for comparative purposes.

of our electrical resistivity data by a decaying exponential function of the above form. To within 5%, the same characteristic temperature θ^* was found in the same temperature range as with the thermoelectric studies.

Finally, in order to eliminate any ambiguity about separating the residual and ideal resistivities, especially at very low temperatures, we plot in Fig. 2 the *slope* of the experimental data. The slope was taken in such a way that the graph represents deviations from a T^5 temperature dependence (a horizontal line corresponding to a pure T^5 dependence). The theoretical total-resistivity results of Fig. 1(a) have also been replotted in this manner and are included for comparitive purposes. Although experimental scatter is significantly magnified on such a plot, the extension of the steep temperature dependence (greater than T^5) of the experimental data down to ~1.5°K and the agreement with theory can still be seen clearly. It is emphasized that in this form the significance of the low temperature ($T < 2.5$°K) experimental results is independent of extrapolation errors in determining the residual resistivity.

What happens at temperatures below ~1.5°K is still open to question. The data lose significance at such temperatures and theoretically there are a number of uncertainties including phonon-drag effects, deviations from Matthiessen's rule, and current-degrading mechanisms other than electron-phonon scattering. For example, it is interesting to compare these results with the theoretical estimate of the *electron-electron* scattering contribution calculated by Lawrence[14] (dotted line in Fig. 2). A contribution of this type could easily become important at such low temperatures.[15] Nevertheless, where we have data available (above ~1.5°K), electron-phonon scattering *alone* appears to account quite well for their complicated temperature dependence and absolute value.

We thus conclude that the electron-phonon umklapp contribution to the electrical resistivity of potassium does indeed disappear at low temperatures. This is supported by detailed agreement between theory and experiment over a wide range of low temperatures and by agreement in form with the results of previous thermoelectric studies at liquid-helium temperatures. The relatively large and rapidly changing magnitude of umklapp scattering effects seen here could have a direct bearing on the interpretation of many other low-temperature measurements involving electron-phonon interaction in the alkali metals.

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[1]R. Peierls, Ann. Phys. (Paris) 12, 154 (1932). Further details may be found in a number of textbooks: for example, J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, England, 1960), pp. 369-370, or F. J. Blatt, *Physics of Electronic Conduction in Solids* (McGraw-Hill, New York, 1968), p. 193.

[2]In a few other cases, deviations from the predictions of the Bloch theory in the low-temperature electrical resistivity of various metals have been ascribed to this effect, some correctly {for example: S. B. Woods, Can. J. Phys. 34, 223 (1956); J. C. Garland and R. Bowers, Phys. Rev. Lett. 21, 1007 (1968); V. S. Tsoĭ

and V. F. Gantmakher, Zh. Eksp. Teor. Fiz. 56, 1232 (1969) [Sov. Phys. JETP 29, 663 (1969)]}, some incorrectly {for example: Yu. N. Chiang, V. V. Eremenko, and O. G. Shevchenko, Zh. Eksp. Teor. Fiz. 54, 1321 (1968) [Sov. Phys. JETP 27, 706 (1968)]; for a further treatment of this problem see J. W. Ekin and B. W. Maxfield, Phys. Rev. B 2, 4805 (1970)}. In each case, however, only a speculative interpretation was possible.

[3]See, for example, J. M. Ziman, Proc. Roy. Soc., Ser. A 226, 436 (1954); M. Bailyn and H. Brooks, Bull. Amer. Phys. Soc. 1, 300 (1956); M. Bailyn, Phys. Rev. 120, 381 (1960).

[4]The samples were free-mounted polycrystalline wires extruded from potassium obtained from Mine Safety Appliances Ltd., Callery, Pa. All were annealed one week at room temperature and then slowly cooled to liquid-helium temperatures. A dc four-terminal resistance-measurement technique was employed; separation between current and potential contacts was greater than 5 sample diameters. The error in the absolute resistivity values, which were determined assuming a resistivity at 22°C of 7.19 $\mu\Omega$ cm, is about 2%. The relative error is about 0.02%. Temperatures were measured with an absolute accuracy of 0.01°K.

[5]Of course, at lower RRR values, significant deviations from Matthiessen's rule were observed. A more detailed treatment of this and other aspects of the data, as well as a tabulation of the complete experimental and theoretical results, will be given in a later article: J. W. Ekin and B. W. Maxfield, to be published.

[6]A. Hasegawa, J. Phys. Soc. Jap. 19, 504 (1964).

[7]T. M. Rice and L. J. Sham, Phys. Rev. B 1, 4546 (1970).

[8]The pseudopotentials used were those of J. Bardeen, Phys. Rev. 52, 688 (1937); N. W. Ashcroft, Phys. Lett. 23, 48 (1966); M. J. G. Lee and L. M. Falicov, Proc. Roy. Soc., Ser. A 304, 319 (1968). In the latter case, two sets of values were used corresponding to the extremal values reported by these authors; for details see Rice and Sham, Ref. 7.

[9]R. A. Cowley, A. D. B. Woods, and G. Dolling, Phys. Rev. 150, 487 (1966).

[10]We have defined an umklapp process to be one in which $\vec{k}_f - \vec{k}_i = \vec{q} + \vec{G}$, with $\vec{G} \neq 0$, where $\vec{k}_i$ and $\vec{k}_f$ are, respectively, the initial and final electron wave vectors, $\vec{q}$ the phonon wave vector, and $\vec{G}$ a reciprocal lattice vector; for a normal process, $\vec{G} = 0$.

[11]Not all of these assumptions have been justified theoretically; in particular, phonon-drag effects may become important at those very low temperatures where umklapp processes are frozen out. The agreement with experiment, however, seems not to be affected appreciably by these assumptions, at least at temperatures above 1.5°K. For further details of the calculation as well as a theoretical treatment of the role of electron-phonon umklapp processes in ultrasonic attenuation see P. N. Trofimenkoff and J. W. Ekin, to be published.

[12]D. K. C. MacDonald, W. B. Pearson, and I. M. Templeton, Proc. Roy. Soc., Ser. A 256, 334 (1960).

[13]It is understood that this simplified exponential form is only an approximation valid over limited temperature ranges since it neglects other slower-varying functions of temperature as well as the fact that different phonon polarizations decay at different rates.

[14]W. E. Lawrence, thesis, Cornell University, 1970 (unpublished).

[15]Vice versa, the large and rapidly changing electron-phonon umklapp component at higher temperatures might readily explain the previous lack of experimental evidence of any electron-electron scattering in the alkalis; see, for example, the Letter by Garland and Bowers, Ref. 2.

Short-Wavelength Cyclotron Waves and Electron Correlations in Potassium

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Observations of the Azbel'-Kaner cyclotron resonance spectra in thick slabs of high-purity potassium metal have revealed additional oscillations on the high-field side of the fundamental resonance. These anomalies in the surface impedance correspond to turning points of the cyclotron-wave dispersion curve at wavelengths which are short compared to orbit radii. The behavior of the signals agrees quite well with free-electron theory and confirms the extremely oscillatory character of the extraordinary mode. The absence of deviations due to Fermi-liquid effects in our experiments is investigated, and in particular we show under what circumstances the Fermi-liquid parameter A_1 can be determined from these signals.

Weak surface-resistance anomalies associated with turning points of the dispersion curves[1] for cyclotron or high-frequency waves have been studied under Azbel'-Kaner cyclotron-resonance (AKCR) conditions in the case of metallic potassium where the absence of band-structure effects makes comparison with theory particularly interesting. As in Henningsen's recent discovery of