Phonon Drag in a layered Transition Metal Compound

Quantum oscillations and high carrier mobility in the delafossite PdCoO2

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Recommended and a Commentary by Chandra Varma, University of California, Riverside

The low temperature resistivity of a pure enough nearly free-electron metal due to scattering by acoustic phonons is $\propto T^5$, the factor T^3 coming from the density of thermally excited phonons and a factor T^2 from the phase space required for backward scattering when the momentum transfer to phonons is small. If, as in a typical transition metal, there is more than one Fermi-surface with different Fermi-velocities in nearby regions near the Fermi-vectors or a single band with curvature of the Fermi-surface varying sharply, velocity loss occurs for momentum transfer large compared to the scale of curvature and the T^2 factor is missing. As the temperature is decreased, the phonons scattered have smaller and smaller momentum. So only intra-Fermi-surface scattering between points of increasingly uniform curvature dominates. Therefore the dependence for such metals also must change to $\propto T^5$ at sufficiently low temperatures. However, hardly any case of the crossover from T^3 to T^5 is known, possibly due to the fact that in transition metals, where such a crossover is to be expected, superconductivity intervenes while the metallic resistivity is still in the T^3 regime. T^5 resistivity alone is observed in one band nearly spherical Fermi-surface metals like Potassium and Copper.

All of the above assumes that the momentum transferred from the electrons to the phonons is lost by the phonons through scattering with impurities or at sample boundaries (or what is very unlikely phonon-phonon scattering) at a rate faster than the electron-phonon scattering rate. What if this conditions is not satisfied? The total momentum of the electron-phonon system is then conserved and resistivity with temperature dependence discussed above cannot be realized. This phenomena has been termed Phonon-Drag. Ultimately however, even with a single almost spherical (or in very anisotropic materials almost cylindrical) Fermi-surface, the phonon can scatter to the next Brillouin zone by exciting a phonon of energy $\omega_0 = vk_U$, where k_U is twice the smallest wave-vector from a point in the Fermi-surface to the Brillouin zone boundary, as shown in the Figure, and v is the appropriate sound velocity. Then the resistivity will be proportional to the density

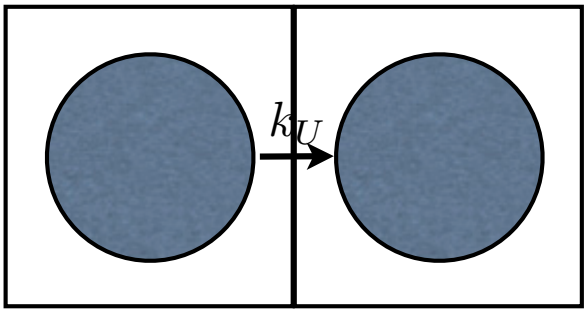

FIG. 1: A circular Fermi-surface in two adjacent Brillouin zones; k_U is the smallest Umklapp vector.

of such excitations and so $\propto \exp(-\omega_0/T)$. All this is ancient knowledge. But the realization of this phenomena even in the purest free-electron metals, such as Potassium, has been uncertain. For an amusing back and forth on this issue, please see the review article[1].

It comes as a real surprise then to find a resistivity with an exponential temperature dependence in the unlikely complicated compound $PdCoO_2$, in the family with the charming name delafossite. The experimental results are unambiguous and so is Δk because the Fermi-surface has been determined in a careful de Haas van Alphen effect measurement and the sound velocity deduced from the low temperature specific heat measurements. The material of-course had to have very low impurity resistivity; the extrapolated $T \to 0$ resitivity is less than 8×10^{-9} Ohm-cm, similar to Cu or K but in fact better signifying lower scattering rate because the density of carriers is about a factor of 2 smaller. The mean-free path is estimated to be 20μm.

How does such a complicated transition metal compound realize the conditions for phonon drag? It is an amusing quirk. delafossite are layered compounds with the composition ABO_2. The A atoms, Pd in the present case from triangular sheets separated by CoO_2 layers. The atomic d-levels of Co split into two crystal field states with a wide splitting and the Co^{+3} state on it occupies bands narrow compared to the splitting. The d-levels of Pd are completely filled and the s-band of Pd lies in between the occupied and the unoccupied bands formed of Co and the chemical potential is such that we can forget about everything at low energy except the s-band whose Fermi-surface is depicted in the figure. The very low residual resistivity is also due probably to the layered structure with defects almost only in the CoO_2 layers, thus providing only forward scattering for electrons near the Fermi-surface.

Another material which has shown enormously low resistivity is Bromine intercalated Graphite[2]. It is worthy of further attention for phonon drag and related phenomena.

An amusing question that arises is, are the phase space factors for electron-phonon interactions in anisotropic materials with nearly cylindrical fermi-surfaces quite different than for three-dimensional nearly spherical fermi-surfaces. The answer in one limit for a purely cylindrical fermi-surface appears[3] to be that the electron and phonon distributions in the presence of an applied electric field cannot come to equilibrium in the two-dimensional case. The limit is $\theta_D/E_f \rightarrow 0$, where equilibrium is attained separately between any point $\mathbf{k}_F$ on the Fermi-surface and the point $-\mathbf{k}_F$ and the phonon emitted and absorbed. In that case, there is no question of the equivalent of the process which leads to T^5 resistivity in 3d. The only choice is the through use of the umklapp vector k_U. Further investigation around this peculiarity may be worthwhile.

One of the results expected from calculation done long ago in a situation of phonon drag is a large thermopower with an interesting temperature dependence. One may also look, not surprisingly, for a quantized Hall effect. But a more set of amusing experiments are Poiseulle flow of heat in the combined electron-phonon system and second sound. In the solid state, these phenomena have been observed clearly only in the He solids.

1. J. Bass, W.P. Pratt, and P.A. Schroeder, Rev. Mod. Phys 62, 645 (1990).

2. S. Tongay, J. Hwang, D. B. Tanner, H. K. Pal, D. Maslov, A. F. Hebard, Physical Review B 81, 115428, (2010).

3. R.N. Gurzhi, A.I. Kopeliovich and S.B. Rutkevich, JETP Letters, 32, 336 (1980).