Properties of strongly correlated metals

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A study is made of the properties of strongly correlated metals (containing heavy fermions) for which the degeneracy temperature is much less than the value corresponding to the plasma frequency. It is shown that a semiquantum liquid model proposed for ^{3}He by Andreev and Kosevich [JETP Lett. **28**, 556 (1978); Sov. Phys. JETP **50**, 1218 (1979)] accounts for the temperature dependences of the thermodynamic and transport parameters above the degeneracy temperature. The conductivity is then of the order of the Mott minimum metallic value and the spin susceptibility behaves in accordance with the Curie-Weiss law. At low temperatures the cross section for the scattering of quasiparticles by impurities is much larger than atomic. The feasibility of applying these results to doped semiconductors near the metal-insulator transition is considered.

1. There are metals (containing heavy fermions) for which the effective mass m^* deduced from the low-temperature specific heat is one or two orders of magnitude greater than the mass m of a "bare" electron,[1)] whereas the quantum degeneracy temperature defined by

$$T_d = p_F^2/2m^*, \quad p_F = (3\pi^2\hbar^3 N)^{1/3}$$

(p_F is the Fermi momentum and N is the electron density) is anomalously low.[1–4] For example, in the case of UPt_3, UBe_{13} (Refs. 1 and 2), and $CeAl_3$ (Refs. 1 and 3), the degeneracy temperature is $T_d \approx 10$ K, whereas in the case of V_2O_3 at pressures $p > 18$ kbar, we have $T_d \approx 60$–600 K (Refs. 4–6). A review of experiments on heavy fermions is given in Ref. 1.

The analogy with a different Fermi liquid (^{3}He) has been invoked frequently (see, for example, Ref. 7) to account for the thermodynamic properties of heavy fermions at temperatures $T \ll T_d$. Our aim will be to show that this analogy helps to understand the universal transport and thermodynamic properties of heavy fermions at temperatures $T \gg T_d$, as well as their anomalous transport properties at $T \ll T_d$.

The energy $U = e^2 N^{1/3}$ of the Coulomb interaction of electrons at interatomic distances and the plasma frequency $\Omega_p = (4\pi e^2 N/m)^{1/2}$ defined in terms of the electron density $N \approx 10^{22}$ cm^{-3} and the mass of bare electrons, differ severalfold for these compounds and are of the order of 1 eV.

It therefore follows that the electron liquid in these compounds has two characteristic energies, T_d and $\hbar\Omega_p$, where

$$\hbar\Omega_p \gg T_d. \tag{1}$$

The inequality (1) allows us to consider such systems as close to electron crystallization.

According to Frenkel,[8] at moderately high temperatures the properties of liquids are close to those of solids. Castaing and Noziéres[9] pointed out that an inequality similar to Eq. (1) and the consequent proximity to crystallization apply also to liquid ^{3}He. (The role of Ω_p in the case of ^{3}He is played by the Debye frequency.)

If the temperature T of a metal is low compared with the characteristic energy U of the interaction between neighboring electrons, the motion of such electrons is mainly in the form of small oscillations about some equilibrium positions and such motion has a characteristic frequency Ω_p much higher than the reciprocal of the lifetime τ^{-1} near this equilibrium position. If $T \ll \hbar\Omega_p$, these are basically zero-point oscillations and the delocalization time

$$\tau \approx \Omega_p^{-1} \exp[(a/\lambda)^2] \gg \Omega_p^{-1}, \quad \lambda \approx a(\hbar\Omega_p/U)^{1/4}$$

is governed by quantum tunneling. Here, λ is the amplitude of zero-point electron oscillations and $a = N^{-1/3}$.

Over a time scale longer than τ we can expect electrons to interchange places and the effects of indistinguishability of electrons, i.e., the exchange effects, to become important.

In view of the finite value of τ the energy indeterminacy $\hbar/\tau$ governs the order of magnitude of T_d, since T_d can be regarded as the width of an allowed band associated with translational tunneling of a particle. The inequality (1) corresponds to the condition $\Omega_p \tau \gg 1$ of validity of the Frenkel liquid model.

If the strong inequality

$$\lambda/a \approx (\hbar/mUa^2)^{1/4} \ll 1$$

is satisfied, electron crystallization takes place. The phase transition from an electron crystal to a Fermi liquid occurs when

$$\lambda(a_c)/a_c \approx [\hbar\Omega_p(a_c)/U(a_c)]^{1/4} \lesssim 1$$

and the question whether the inequality (1) is satisfied by the Fermi-liquid phase is related to a large numerical parameter $\exp[a_c/\lambda(a_c)] \gg 1$ (a_c is the critical distance between electrons at which the transition to a Winger crystal takes place).

There are therefore two characteristic temperature intervals: $T \ll T_d$ and $T_d \ll T \ll \hbar\Omega_p$.

2. If $T \ll T_d$, we can expect the usual Fermi-liquid behav-

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ior with an effective mass $m^* \approx \hbar\tau/a^2 \gg m$. The spin susceptibility in this temperature range is also high:

$$\chi \approx \frac{\mu^2 N}{\hbar\tau^{-1}} \approx \frac{\mu^2 N}{T_d},$$

where μ is the Bohr magneton. This is due to the fact that indistinguishability of electrons is important at times longer than τ and, therefore, the spin exchange energy is of the order of $|\Theta| \approx \hbar/\tau \approx T_d$.

It is important to note that a strong rise of m^* and χ is associated with one large parameter $\tau\Omega_p$ so that the densities of states calculated from the low-temperature measurements of these quantities should be of the same order of magnitude.[2)]

Finally, we shall estimate the residual conductivity of a Fermi liquid of heavy fermions in the limit $T \to 0$. We shall assume that impurities have a potential with a small radius of the order of a. The proximity of an electron liquid to a Wigner crystal results in a giant enhancement of the scattering cross section α of a quasiparticle with an effective mass m^* compared with a^2 (in the approximation which is linear with respect to the electric field intensity the attainment of the Wigner crystal state is hindered by one impurity and we have $\alpha = \infty$).

We shall estimate α using reasoning similar to that in Ref. 8. A Fermi liquid moving relative to impurities at an average hydrodynamic velocity v experiences a force due to impurities and this force is of the order of the stokes force $F = na\eta v$, where n is the impurity concentration and η is the dynamic viscosity.

Applying the Drude-Lorentz formula $\sigma = e^2 N/p_F n\alpha$, we find that $\alpha = \eta a/Np_F$. By analogy with Ref. 7, we can estimate $\eta = (a\varkappa)^{-1}$, where

$$\varkappa = \frac{a}{eE}[\tau^{-1}(U - eEa) - \tau^{-1}(U + eEa)]|_{E\to 0} \approx \frac{a^2}{\hbar\Omega_p\tau}$$

is the electron mobility which in this case is related to the processes of electron tunneling to neighboring equilibrium positions. As a result, we obtain

$$\alpha = a^2 m^*/m. \tag{2}$$

Therefore, the cross section is much larger than atomic and the distance at which an electron liquid differs from a Wigner crystal is of the order of $a(m^*/m)^{1/2}$. This behavior has been observed experimentally on many occasions (see, for example, Ref. 5).

3. We shall now consider the temperature range $T_d \ll T \ll \hbar\Omega_p$. The properties of heavy fermions in this temperature range can be explained in a natural manner by the model of a semiquantum liquid proposed by Andreev and Kosevich[11,12] to account for the high-temperature properties of ^{3}He and ^{4}He.

The inequality $T \gg T_d$ means that an electron liquid is nondegenerate and the effects associated with the indistinguishability of electrons are unimportant. The electron spins are then disordered. On the other hand, the inequality $T \ll \hbar\Omega_p$ implies that at times shorter than τ the electrons are still localized near certain equilibrium positions. As a result we obtain in a natural manner the Curie-Weiss susceptibility of localized moments ($C_1 \approx 1$):

$$\chi \approx \mu^2 N(T^{-1} + C_1\Theta T^{-2}), \quad |\Theta| \approx T_d. \tag{3}$$

The contribution to the specific heat from the spin degrees of freedom in the case when $\mu H \ll T$ is of the order of

$$N[(T_d/T)^2 - C_2(T_d\mu H/T^2)^2],$$

where $C_2 \approx 1$. In view of the condition $T \ll \hbar\Omega_p$ the orbital degrees of freedom are hardly excited and the main contribution to the specific heat from these degrees of freedom is made by the mechanism proposed by Halperin, Anderson, and Varma[13] to account for the low-temperature properties of glasses and used later by Andreev[11] to account for the properties of ^{3}He and ^{4}He. A more detailed analysis of the thermodynamic properties of ^{3}He can be found in Ref. 14. Similar results have been recently obtained using the variational method for solving the Hubbard model.[15]

Since there is no long range-order in an electron liquid and the equilibrium positions of electrons are distributed irregularly, over time intervals shorter than τ such a liquid behaves like a glass. In the electron liquid under consideration all the barriers are permeable and the low concentration of excitations is ensured by the fact that the temperature is low compared with this characteristic scatter of energy at neighboring equilibrium positions, which is of the order of U. Consequently, the contribution to the specific heat made by the orbital degrees of freedom is of the order of νT (Ref. 11), where $\nu \approx zN/U$ is the density of states of two-level systems which is assumed to be independent of energy at low excitation energies, and $z \gtrsim 1$.

Consequently, the temperature dependence of the electron specific heat C at temperatures $T \gg T_d$ is of the form

$$C = \beta N[(T_d/T)^2 - C_2(T_d(\mu H)/T^2)] + zNT/U, \quad \beta \lesssim 1. \tag{4}$$

Bearing in mind that at temperatures $T < T_d$ the specific heat is $C \approx NT/T_d$, we reach the conclusion that C has a singularity at $T \approx T_d$ in agreement with the experimental results.[1] The existence of such a singularity is associated with a significant contribution to the specific heat of the spin degrees of freedom at $T \approx T_d$ and a reduction in this contribution on increase in temperature. The electron contribution to the specific heat at temperatures $T \gg T_d$ is difficult to observe because of the growing contribution of the phonon specific heat.

At relatively low temperatures the temperature dependence of the electrical conductivity is governed by electron-electron collisions.[4] At temperatures $T < T_d$ the mean free path in the case of electron-electron collisions is long in terms of the parameter $T_d/T \gg 1$ and it decreases on increase in temperature.

However, at $T \approx T_d$, on the one hand, there is no small parameter which can ensure that the frequency of the e-e collisions is low and, on the other, the e-e collisions are as frequent as the normal e-e collisions. A natural estimate of the mean free path of an excitation at $T \approx T_d$ is then provided by the electron-electron distance a, which gives the minimum metallic conductivity[3)] $\sigma_{\min} \approx e^2/\hbar a$.

A further increase in temperature alters little the degree

of disorder in the electron subsystem because the spin subsystem is completely disordered at $T > T_d$ and the orbital degrees of freedom are weakly excited in terms of the parameter $T/\hbar\Omega_p \ll 1$. This is the reason for the weak temperature dependence $\sigma(T)$ in the range $T \gg T_d$. A strong frequency dependence of the conductivity begins at $\hbar\omega > T_d$ (ω is the frequency of an electromagnetic field).

Considerations similar to those given above lead to an estimate of the thermoelectric power amounting to $|\beta| \approx (\sigma/e)\ln 2$.

We shall now consider the dependence of the specific heat C on a magnetic field. At temperatures $T > T_d$ this dependence is given by Eq. (4). The specific heat decreases on increase in H and we have $[C(0) - C(H)]/C(0) \approx 1$ for $\mu H \approx T \approx T_d$.

At temperatures $T \ll T_d$ we must recall first of all that the model of an ideal Fermi gas with $\mu H \gtrsim T_d$ predicts a reduction in the specific heat by a factor $2^{2/3}$. The Kondo effect (Kondo lattice) also reduces the specific heat on increase in H. The above model predicts an increase in C on rise of H. In a situation when Eq. (1) is obeyed, the effective mass m^* depends weakly on H, since τ is governed primarily by single-particle subbarrier tunneling. The dependence of τ on H is due to the fact that interchange of places by electrons with antiparallel spins becomes easier and this increases m^* on increase in H. Similar behavior is exhibited also by the Hubbard model near the metal-insulator transition.[10]

It therefore follows from the above model that

$$\Delta C = C(H) - C(0) > 0, \quad T < T_d,$$
$$\Delta C < 0, \quad T > T_d.$$

This is exactly the situation in UPt_3. However, UBe_{13} and $CeCu_6$ behave in the opposite manner: ΔC also changes its sign at $T \sim T_d$, but $\Delta C < 0$ when $T < T_d$ and $\Delta C > 0$ when $T > T_d$. The reason for this behavior is not clear.

4. We can see that the above model accounts for the following properties of strongly correlated metals.

At low temperatures $T \ll T_d$ these compounds exhibit the Fermi-liquid behavior. The specific heat depends linearly on temperature: $C = \gamma T$, where $\gamma = m^* p_F/3\hbar^3$ is large. The Pauli magnetic susceptibility is also large and the densities of states calculated from the specific heat and magnetic susceptibility are of the same order of magnitude. The conductivity is metallic, but the residual resistance remains high in the limit $T \to 0$, because the cross section for the scattering of quasiparticles by impurities is one or two orders of magnitude greater than the atomic cross section.

At high temperatures $T \gg T_d$ the susceptibility behaves in accordance with the Curie-Weiss law and the electrical conductivity is of the order of the minimum metallic value $\sigma_{\min}$, varies slowly with temperature, and is independent of impurity concentration. Therefore, at temperatures $T > T_d$ these compounds demonstrate properties which distinguish them qualitatively from conventional metals.

The electron specific heat rises on increase in T in the range $T < T_d$ but falls at $T > T_d$. At high temperatures the specific heat is clearly governed by phonons. This behavior is not in conflict with the experimental results obtained on heavy fermions.[1]

However, we cannot exclude the possibility that the semiquantum liquid model can describe also doped (extrinsic) semiconductors at relatively high temperatures near the metal-insulator transition.

Recent experiments indicate[16] that near the metal-insulator transition in semiconductors at temperatures close to the limit $T \to 0$ the conductivity does not exhibit a discontinuity but vanishes smoothly, i.e., there is no minimum metallic conductivity effect.

However, at relatively high temperatures $T > T_0$ (T_0 is usually of the order of 1 K) there is frequently a temperature range where the conductivity is of the order of $\sigma_{\min}$ and depends weakly both on temperature and on the proximity to the metal-insulator transition. In this temperature range the spin susceptibility exhibits the Curie-Weiss behavior.[17] Therefore, at the metal-insulator transition a doped semiconductor may exhibit a temperature range where all the transport and thermodynamic properties behave in the universal manner discussed above.

We shall conclude by noting that there is another class of compounds in which a strong electron-electron correlation results in a basically different type of behavior. These are substances of the Fe_3O_4 and V_3O_5 type (Ref. 6) which at low temperatures are electron Wigner crystals,[4] i.e., which satisfy the strong inequality $\lambda \ll a$. When temperature is increased, the electron subsystem melts and electrons become delocalized. Substances of this type clearly behave similarly to weak solutions of ^{3}He in crystalline ^{4}He when the energy of the interaction between electrons at interatomic distances is much greater than the width of an allowed band. Such behavior was investigated by Kagan and Maksimov.[18] All the transport and thermodynamic properties then behave quite differently from those discussed in the present paper. The question of a transition between the two regimes is still to be considered.

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[1)]The mass of a bare electron is the mass associated with the single-particle band motion without allowance for the electron-electron interaction.
[2)]The model proposed here is in many respects similar to the model of Brinkman and Rice[10] who used the variational method of Gutzwiller in the Hubbard model near the metal-insulator transition, and for this reason this model is valid only at the limit $T \to 0$.
[3)]The author is aware that similar ideas have been put forward by A. M. Dyugaev in respect of the viscosity and thermal conductivity of ^{3}He.

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