Umklapp scattering and electron pairing cutoff in BCS superconductors

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In a superconductor electrons form pairs for which the end states of normal and umklapp scattering may overlap. This cuts electron pairing off at a phonon frequency, \( \omega_c \), low compared with the Debye frequency, \( \omega_D \). Using this insight, correct values of \( 2\Delta/k_B T_c \) (average error 8.9%) for 12 superconductive metals, including Hg and Pb, are achieved from simple BCS formalism with an average \( \omega_c/\omega_D \) of 0.148: Landau’s idea of a Fermi liquid may cover strong-coupling superconductors. The cancellation between normal and umklapp scattering may be more important than a strong electron-phonon interaction in reaching a high critical temperature \( T_c \).

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Despite the enormous effort in nearly 50 years since BCS [1], a fundamental issue at the very heart of the study of superconductivity appears to have escaped attention: in a superconductor the end states of normal and umklapp scattering may intersected by the phonon sphere centered at this surface (the other sphere is centered above the Fermi surface at height \( \hbar \omega_D/2\pi \)). The shaded circles represent two identical spherical phonon sphere centered at \( \mathbf{q} \) and \( -\mathbf{q} \), the Debye momentum. If the phonon momentum \( \mathbf{q} \) exceeds \( q_0 \) then an electron in state \( \mathbf{k} \) may be scattered into \( \mathbf{k} + \mathbf{q} \) to lie on the adjacent Fermi surface. It makes no physical difference if the reciprocal lattice vector \( \mathbf{G} \) (parallel to \( \mathbf{k} \) for simplicity) is added to an electron state: we can replace \( \mathbf{k} + \mathbf{q} \) with \( \mathbf{k} + \mathbf{q} + \mathbf{G} \), which lies on the original Fermi surface, and is known as the end state of umklapp scattering; it is usually far away from the initial state of the electron.

Umklapp scattering often plays a significant role in solids at low temperatures [2], and superconductors may be no exception. Let \( \mathbf{k} \) and \( -\mathbf{k} \) be the states of a Cooper pair, which by nature are on opposite sides of the Fermi sea, like a pair of rotating doors. Correspondingly \( \mathbf{k} + \mathbf{q} + \mathbf{G} \) must be paired with \( -\mathbf{k} - \mathbf{q} - \mathbf{G} \); scattering of the electron pair is synchronized, reminiscent of the synchronized motion of rotating doors. It is evident from Fig. 1 that \( -\mathbf{k} - \mathbf{q} - \mathbf{G} \) lies inside the phonon sphere centered at state \( \mathbf{k} \), where all the electron states are involved in normal scattering: thus the end state of

\[ \mathbf{k} + \mathbf{q} - \mathbf{G} \]

\[ \mathbf{k} - \mathbf{q} - \mathbf{G} \]

\[ \mathbf{k} + \mathbf{q} + \mathbf{G} \]

\[ \mathbf{k} - \mathbf{q} + \mathbf{G} \]

\[ \mathbf{k} \]

\[ \mathbf{-k} \]

\[ q_0 \]

\[ \mathbf{G} \]

\[ \mathbf{q} \]

FIG. 1: A spherical Fermi surface (open circle) and a pair of electrons in states \( \mathbf{k} \) and \( -\mathbf{k} \). This pair is scattered into states \( \mathbf{k} + \mathbf{q} + \mathbf{G} \) and \( -\mathbf{k} - \mathbf{q} - \mathbf{G} \) in the umklapp process, \( \mathbf{q} \) being the phonon wavevector, \( \mathbf{G} \) the reciprocal lattice vector. The shaded circles represent two identical spherical phonon zones, angles \( \alpha \) and \( \beta \) measure the size of the sections of the Fermi surface being intersected by these phonon zones. We have \( \alpha \beta > 1 \) because a larger section of the Fermi surface is intersected by the phonon sphere centered at this surface (the other sphere is centered above the Fermi surface at height \( q_0 \)). As a result, the electron state \( -\mathbf{k} - \mathbf{q} - \mathbf{G} \) must lie inside the phonon sphere centered at \( \mathbf{k} \), where all the states are involved in normal scattering. Furthermore, when \( \mathbf{k} + \mathbf{q} + \mathbf{G} \) runs over the shaded phonon zone on the left, \( -\mathbf{k} - \mathbf{q} - \mathbf{G} \) also runs over a sphere indicated in part by the broken white circle (centered above the Fermi surface at height \( q_0 \) to mirror the phonon sphere on the left).

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unklapp scattering may not be empty, a scattering that may have serious consequences.

In order to incorporate umklapp scattering into the BCS formalism, we must make sure that in this formalism normal and umklapp scattering do not drive electrons into the same end state at the same time. We let

$$\Psi = \prod_{k} ((1-h_k)^{1/2} + \epsilon_k h_k) \prod_{k'} (1-h_{k'})^{1/2}$$

be our ground state wave function, where $h_k$ is the pair generation operator, $h_k$ occupation probability, $|0\rangle$ the vacuum, $N$ and $U$ specify paired end states of normal and umklapp scattering (or normal and umklapp pairs for short) respectively. It is easy to check that wave functions of the normal and umklapp pairs with the same $k$ are always orthogonal to each other: the probability is zero for normal and umklapp scattering to share the same end state at the same time. The ground state energy $W = \langle \Psi | H_{BCS} | \Psi \rangle$ is minimized if $\partial W / \partial h_k = 0$, $H_{BCS}$ being the BCS reduced Hamiltonian, which leads through the constraint $h_k = (1/2)(1 - \epsilon_k/E(k))$ to

$$\Delta(k) = \sqrt{\sum_{k} - \sum_{k'}} \frac{V_{kk'}}{2E(k')} \Delta(k')$$

where $E(k) = [\Delta^2 + \epsilon_k^2]^{1/2}$. $\epsilon$ being the energy relative to the Fermi level, $k$ can be specified by either $N$ or $U$.

According to the above self-consistent equation normal pairs sustain superconductivity but umklapp pairs frustrate it. This is true only when the normal pairs dominate, i.e. some terms in $k'(N)$ survive cancellation by terms in $k'(U)$. Otherwise $\partial W / \partial h_k = 0$ will become the condition to maximize $W$, unless we choose $h_k = (1/2)(1 + \epsilon_k/E(k))$ as the constraint and consequently, we have to exchange $N$ and $U$: then umklapp pairs sustain superconductivity but normal pairs frustrate it. It appears that either normal or umklapp pairs alone may lead to an energy gap ($h_k$ or $1-h_k$ interpreted as the occupation probability respectively), but their coexistence mutually cancels their effect. We are reminded that $(1-h_k)^{1/2} + h_k^{1/2}$ was also used by BCS to generate excited pairs, apparently in order to forbid excited and ground pairs sharing the same state at the same time, so that the entropy of the particle ensemble could be counted properly. No confusion need arise. If the normal pairs dominate, then all the terms in $k'(U)$ will be cancelled: excited pairs will be introduced in the absence of umklapp pairs. If the umklapp pairs dominate, then we use $(1-h_k)^{1/2} + h_k^{1/2}$ to generate excited pairs. To make the discussion concrete we assume dominance of normal pairs.

A note about the Coulomb repulsion, which was dropped from the reduced Hamiltonian by BCS. According to calculations based on the Bogolubov model potential, the Coulomb repulsion has negligible effect on the value of the energy gap in the BCS theory, compared with the effect of the electron-phonon interaction, if the so-called Coulomb cutoff frequency, introduced artificially, is large enough. In fact, an energy gap may arise even from an entirely repulsive interaction, when this interaction is perturbed by an attractive interaction over a narrow range of phonon frequencies. It appears that in a metal Coulomb repulsions between electrons are in a balance which can be topped even by a weak attractive interaction.

We may also justify the reduced Hamiltonian from the field-theoretic point of view. This Hamiltonian arises from the canonical transformation $H_{BCS} = e^{-S} H e^{-S}$, where $H = H_e + H_p + H_{ep}$ with $[H_e + H_p, S] + H_{ep} = 0$, $H_e$, $H_p$ and $H_{ep}$ are Hamiltonians of electron, phonon and electron-phonon interaction, respectively, the brackets represent the operation $[A, B] = AB - BA$. With this canonical transformation we cancel $H_{ep}$ in the first order expansion of $H_{BCS}$. Apparently we can do the same when $H = H_e + H_p + H_{col} + H_{ep}$ (i.e. to use $H_e + H_p + H_{col}$ to replace $H_e + H_p$, in the above canonical transformation, $H_{col}$ being the Hamiltonian of the Coulomb repulsion), provided that we use the eigen-functions of $H_e + H_p + H_{col}$ (instead of the eigen-functions of $H_e + H_p$) as the base functions in the second quantization. Now $H_{BCS}$ measures the second order perturbation to the energy of the particle ensemble with the Coulomb repulsion. This brings little change to the BCS theory, except that in $V_{kk'}$ the electron energy may be shifted slightly.

Let us proceed. In principle we can identify the terms in $k'(N)$ that have survived cancellation by terms in $k'(U)$, or vice versa, and go on to solve the self-consistent equation, a daunting task due to the incompatible symmetries of the Fermi sea and the reciprocal lattice. It is clear from Fig. 1 that we have to find $k'(N)$ and $k'(U)$ all over again whenever the direction of $k$ changes. In order not to do so we follow BCS and assume that phonons are cut off abruptly at $\omega_c$. We also assume an isotropic energy gap function written as $\Delta(\epsilon)$. We will vary $\omega_c$ to let the calculated $\Delta$ match its experimental value. Our theory will be supported if $\omega_c/\omega_D << 1$ holds to reflect the low onset phonon frequency for umklapp scattering.

We adopt the free electron model, on the basis that the BCS theory is based on the principle of variation, which is not sensitive to errors in the trial function. We also adopt the Debye phonon model. Therefore in

$$V_{kk'} = \frac{2\hbar \omega_q |M_q|^2}{(\hbar \omega_q)^2 - (\epsilon_k' - \epsilon_k)^2}$$

where $\omega_q$ is the phonon frequency, $q = k - k'$, we have $\epsilon_k' = \epsilon_k + 4\epsilon_F \xi x$ for states close to the Fermi surface, $\epsilon_F$ being the Fermi energy, $\xi = q/2k_F$, $x = \cos \theta + \xi$, $k_F$ the Fermi wavenumber, $\theta$ angle between $k$ and $q$. Consequently we have $(\hbar \omega_q)^2 - (\epsilon_k' - \epsilon_k)^2 = (4\epsilon_F \xi x)(\xi^2 - x^2)$, $\delta = \hbar \omega_q / 4\epsilon_F \xi = (1/4\epsilon_F)(\omega_q / \omega_D)(\hbar \omega_D / \epsilon_F) = (k_F / 2q)(q/\omega_D)(T_D / T_F) = (Z/16)^{1/3}(T_D / T_F)$, $T_D$ and $T_F$ are Debye and Fermi temperatures. We have $\delta \sim 10^{-3}$ in Table I but in more realistic models $\delta$ may not
tron wave functions (spin absorbed into $k$-dependent $\kappa$ without terms in $k$) depends on the atomic potential $V$, atomic mass, $N$ number of atoms in unit volume, $r$ coordinates in real space and $\Omega$ the Wigner-Seitz cell. Carbotte and Dynes utilized tabulated data of the Heine-Abarenkov pseudopotential to estimate $\delta V$ and calculated the electric resistivity $\rho$ (another effect of the electron-phonon interaction) and energy gap $\Delta$ (using the Eliashberg formalism) separately. We use the Mott-Jones formula to link $\delta V$ with $\rho$ and then use this $\rho$ to express $\delta V$: the Heine-Abarenkov pseudopotential is no longer needed. Letting $k'$ in the self-consistent equation [without terms in $U$] run over a phonon sphere centered at the Fermi surface, we find:

$$\Delta(\epsilon) = \Delta_0 \left[ \frac{\pi}{2} \int_{\Omega} F^2(3.84V^{1/3}) \xi^3 d\xi \right]^{-1} \times \int_{0}^{\Xi} F^2(3.84V^{1/3}) \xi^2 d\xi \times \frac{1}{2} \int_{-1+\xi}^{1+\xi} \frac{\Delta(\epsilon + 4\epsilon_F \xi)}{E(\epsilon + 4\epsilon_F \xi)} \frac{dx}{\delta^2 - x^2}$$

Here $F$ is the overlap integral function and $\Delta_0 = \hbar e^2 n^2 v^2 / 2k_B T$.

| TABLE I: |
|---|---|---|---|---|
| $2\Delta_0^a$ | $2\Delta(0)^a$ | $\delta^b_\omega / \omega_c$ | $2\Delta(0)/k_B T_c^c$ |
| Zn | 2.38 | 2.40 | 1.50 | 1.67 | 3.63 | 3.20 |
| Cd | 0.51 | 1.50 | 1.20 | 0.245 | 3.70 | 3.20 |
| Hg | 1.62 | 16.15 | 0.044 | 0.124 | 4.76 | 4.60 |
| Al | 2.74 | 3.40 | 1.81 | 0.183 | 3.54 | 3.30 |
| Ga | 7.92 | 3.30 | 1.51 | 0.096 | 3.51 | 3.50 |
| In | 0.49 | 10.5 | 0.62 | 0.203 | 3.92 | 3.60 |
| TI | 0.46 | 7.35 | 0.48 | 0.164 | 3.92 | 3.57 |
| Sn | 2.52 | 11.5 | 1.25 | 0.158 | 3.70 | 3.50 |
| Pb | 1.39 | 27.3 | 0.60 | 0.160 | 4.56 | 4.38 |
| V | 27.9 | 16.0 | 1.20 | 0.203 | 3.92 | 3.60 |
| Nb | 9.80 | 30.5 | 1.04 | 0.107 | 3.70 | 3.50 |
| Ta | 6.72 | 14.0 | 0.90 | 0.095 | 3.95 | 3.60 |

*a in 10^{-4} eV
*b in 10^{-3}
*c experimental data bracketed

be a constant. The matrix element

$$\mathcal{M}_q = \left( \frac{\hbar N}{2M\omega_q} \right)^{1/2} \int_{\Omega} \psi^*_k r \delta V(r) \psi_k(r) dr$$

depends on the atomic potential $\delta V$, where $\psi$’s are electron wave functions (spin absorbed into $k$ and $k'$), $M$ atomic mass, $N$ number of atoms in unit volume, $r$ coordinates in real space and $\Omega$ the Wigner-Seitz cell. Carbotte and Dynes utilized tabulated data of the Heine-Abarenkov pseudopotential to estimate $\delta V$ and calculated the electric resistivity $\rho$ (another effect of the electron-phonon interaction) and energy gap $\Delta$ (using the Eliashberg formalism) separately. We use the Mott-Jones formula to link $\delta V$ with $\rho$ and then use this $\rho$ to express $\delta V$: the Heine-Abarenkov pseudopotential is no longer needed. Letting $k'$ in the self-consistent equation [without terms in $U$] run over a phonon sphere centered at the Fermi surface, we find:

$$\Delta(\epsilon) = \Delta_0 \left[ \frac{\pi}{2} \int_{\Omega} F^2(3.84V^{1/3}) \xi^3 d\xi \right]^{-1} \times \int_{0}^{\Xi} F^2(3.84V^{1/3}) \xi^2 d\xi \times \frac{1}{2} \int_{-1+\xi}^{1+\xi} \frac{\Delta(\epsilon + 4\epsilon_F \xi)}{E(\epsilon + 4\epsilon_F \xi)} \frac{dx}{\delta^2 - x^2}$$

Here $F$ is the overlap integral function and $\Delta_0 = \hbar e^2 n^2 v^2 / 2k_B T$.

![FIG. 2: $\Delta(\epsilon)$ curve for Sn at $T = 0$. The temperature curve of $\Delta$ at $\epsilon = 0$ is shown in the insertion.](image)

We find $\Delta(\epsilon)$ through iteration. The above integration with respect to $x$ is of the Cauchy type. In first iteration we let $\Delta = \Delta_0$, so that the Cauchy principal value has an analytical expression:

$$\int \frac{\Delta_0}{[\Delta_0^2 + (\epsilon + 4\epsilon_F \xi)^2]^{1/2}} \frac{dx}{\delta^2 - x^2} = \frac{\Delta_0/2\delta}{[\Delta_0^2 + (\epsilon + 4\epsilon_F \delta)^2]^{1/2}} \ln \frac{2A(\epsilon)}{|\epsilon - \delta|} - \frac{\Delta_0/2\delta}{[\Delta_0^2 + (\epsilon - 4\epsilon_F \delta)^2]^{1/2}} \ln \frac{2B(\epsilon)}{|\epsilon + \delta|} + C$$

where $C$ is the integration constant,

$$A(\epsilon) = \Delta_0^2 + (\epsilon + 4\epsilon_F \delta)(\epsilon + 4\epsilon_F \xi)$$

and

$$B(\epsilon) = \Delta_0^2 + (\epsilon - 4\epsilon_F \delta)(\epsilon + 4\epsilon_F \xi)$$

which is integrated numerically. We vary $\Xi$, the upper limit of the integration, until $\Delta(0)$ matches its observed value. The result in FIG. 2 (dashed curve) is virtually identical to the full numerical solution (where the Cauchy principal value is found numerically) in first iteration. It can be seen that prominent features of $\Delta(\epsilon)$ have already emerged from first iteration: further iterations improve the accuracy of solution but retain the physics.

One such prominent feature is that $\Delta(\epsilon)$ is structured: it has a peak flanked by two negative dips. For Sn the distance between the peak and one dip is $\sim 4 \times 10^{-3}$ eV (FIG. 2), compared with $\hbar \omega_D = 1.7 \times 10^{-3}$ eV; the pairing effect extends fairly deep into the Fermi sea, in spite of the low $\omega_c$. As a result, the curve of $d\Delta/dE$, which represents the tunnelling density of states, also has a structure featuring a dip, similar to the characteristic dip in.
In order to justify the values of $\omega_c/\omega_D$ in Table I we solve the self-consistent equation at $T > 0$, which is identical to its counterpart at $T = 0$, save an additional factor $\tanh(E/2k_BT)$ in the integrand. Now $\Delta = \Delta(\epsilon, T)$ which is a function of $T$ also found through iteration $[\Delta = \Delta(\epsilon)]$ implies $T = 0$. We let $T$ increase in small steps and use converged $\Delta(\epsilon, T)$ to start iteration at the next $T$. We find $T_c$ through a quadratic curve fit once $\Delta(0, T) < 0.01\Delta(0)$. Table II and FIG. 3 show that $2\Delta(0)/k_BT_c$ matches experimental data reasonably well (average error 8.9%). In particular the large values 4.6 and 4.38 for Hg and Pb have been calculated fairly successfully. If we were to cut phonons off at $\omega_D$, then we would have high $T_c$ or low (on average ~2-5% of measured values) with an erroneous $2\Delta(0)/k_BT_c$ (e.g. 3.63 and 3.67 for Hg and Pb).

In conclusion the electron pairing cutoff frequency $\omega_c$ in BCS theory may arise naturally. Since $\omega_c/\omega_D << 1$ the 12 metals in Table I Hg and Pb included, can all be treated as weak-coupling superconductors, where the BCS theory can be applied to yield e.g. correct values of $2\Delta(0)/k_BT_c$: Landau’s idea of a Fermi liquid covers strong-coupling BCS superconductors. Strong electron-phonon interaction may not be necessary for high $T_c$: Hg has a small $\Delta_0$ but large energy gap, whereas V has a large $\Delta_0$ but roughly the same gap (see Table I). Our conclusions are valid for BCS superconductors. They may help us to understand MgB$_2$ or other superconductors with umklapp scattering.

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