Pairing Constraint on the Real Space Formalism of the Theory of Superconductivity†

Yong-Jihn Kim††

Department of Physics, Purdue University, West Lafayette, Indiana 47907

Abstract

We review the new theory of impure superconductors constructed by Kim and Overhauser, and further developed by Kim. It was shown that Gor'kov's self-consistency equation needs a pairing constraint derived from the Anomalous Green's function. Whereas earlier studies have applied a constraint only on the pair potential, we show that the kernel also should be fixed by the ground state. The Bogoliubov-de Gennes equations need a pairing constraint in order to obtain the proper vacuum state by the corresponding unitary transformation. The relation between the pair potential and the gap parameter is clarified. This new study opens up a reinvestigation of inhomogeneous superconductors. We will discuss (i) strong coupling theory of dirty superconductors, (ii) suppression of magnetic impurity effect by ordinary impurities, (iii) weak localization correction to the phonon-mediated interaction, and (iv) other inhomogeneous superconductors.

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†† Present address: Department of Physics, Korea Advanced Institute of Science and Technology, Taejon 305-701, Korea

I. INTRODUCTION

Recently it was shown[1,2] that the Abrikosov-Gor'kov (AG)[3] theory of impure superconductors predicts a large decrease of T_c, linear in the nonmagnetic impurity concentration, which is not consistent with Anderson's theorem.[4] In their response[5] on ref. 1, AG argued that the frequency cutoff makes the AG theory compatible with Anderson's theorem. This argument is based on Tsuneto's application[6] of the AG theory to the Eliashberg equation.

To settle this controversy, we first need to understand the limitation of Anderson's theorem. It was pointed out that Anderson's theorem is valid only up to the first order of the impurity concentration,[1,7] and the phonon-mediated interaction is strongly decreased by Anderson localization.[1,8,9]

However, Tsuneto's theory fails to show the existence of localization correction to the phonon-mediated interaction. The failure comes from the intrinsic pairing problem[10,11] in Gor'kov's formalism.[12] The kernel of the self-consistency equation should be set by the physical constraint of the Anomalous Green's function. The resulting equation is nothing but another form of the BCS gap equation. Using the equation, the localization correction to the phonon-mediated interaction may be calculated.[1,11] A correct strong coupling theory has been reported by Kim.[8]

For magnetic impurity effects, Kim and Overhauser (KO)[7] proposed a BCS type theory with different predictions: (i) The initial slope of T_c decrease depends on the superconductor and is not the universal constant proposed by Abrikosov and Gor'kov(AG).[3] (ii) The T_c reduction by exchange scattering is partially suppressed by potential scattering when the overall mean free path is smaller than the coherence length. This compensation has been confirmed in several experiments.[13–15] The difference comes again from the pairing problem. If we impose a correct pairing condition on the self-consistency equation, or the AG's calculation, we can find KO's result.

II. PHONON-MEDIATED INTERACTION IN BCS THEORY AND GOR'KOV'S FORMALISM

A. BCS Theory

For a homogeneous system, BCS introduced a reduced version of the Fröhlich phonon-mediated interaction,

$$H_{red} = \sum_{\vec{k}\vec{k}'} V_{\vec{k}\vec{k}'} c^{\dagger}_{\vec{k}'} c^{\dagger}_{-\vec{k}'} c_{-\vec{k}} c_{\vec{k}}, \tag{1}$$

where

$$V_{\vec{k}\vec{k}'} = \begin{cases} -V, & \text{if } |\epsilon_{\vec{k}}|, |\epsilon_{\vec{k}'}| \leq \omega_D \\ 0, & \text{otherwise.} \end{cases} \tag{2}$$

This reduction procedure is recognizing in advance which eigenstates will be paired and so contribute to the BCS condensate.

In the presence of impurities, we can derive the phonon-mediated interaction by transforming the Fröhlich interaction using the relation,

$$\psi_{n\sigma} = \sum_{\vec{k}} \phi_{\vec{k}\sigma} < \vec{k}|n> . \tag{3}$$

ψ_n and $\phi_{\vec{k}}$ denote the scattered state and the plane wave state. The reduced version of this interaction anticipates that ψ_n (having spin up) will be paired with its time-reversed counterpart $\psi_{\overline{n}}$ (having spin down). The new reduced Hamiltonian is

$$H'_{red} = \sum_{nn'} V_{nn'} c^{\dagger}_{n'} c^{\dagger}_{\overline{n}'} c_{\overline{n}} c_n, \tag{4}$$

where

$$V_{nn'} = -V \sum_{\vec{k}\vec{k}'\vec{q}} < \vec{k} - \vec{q}|n' >< \vec{k}' + \vec{q}|\overline{n}' >< \vec{k}'|\overline{n} >^{*}< \vec{k}|n >^{*} . \tag{5}$$

Anderson's theorem is valid only when $\vec{k}'$ can be set equal to $-\vec{k}$.

B. Gor'kov's formalism

In Gor'kov's formalism, a point interaction $-V\delta(\mathbf{r}_1-\mathbf{r}_2)$ is used for the pairing interaction between electrons. For a homogeneous system, the pairing interaction is

$$
\begin{aligned}
H_G &= -\frac{1}{2}V\int d\mathbf{r}\sum_{\alpha\beta}\Psi^\dagger(\mathbf{r}\alpha)\Psi^\dagger(\mathbf{r}\beta)\Psi(\mathbf{r}\beta)\Psi(\mathbf{r}\alpha) \\
&= -\frac{1}{2}V\sum_{\vec{k}\vec{k}'\vec{q}\sigma\sigma'} c^\dagger_{\vec{k}-\vec{q},\sigma}c^\dagger_{\vec{k}'+\vec{q},\sigma'}c_{\vec{k}'\sigma'}c_{\vec{k},\sigma},
\end{aligned}
\tag{6}
$$

and

$$
\begin{aligned}
V_{\vec{k}\vec{k}'} &= -V\int \phi^*_{\vec{k}'}(\mathbf{r})\phi^*_{-\vec{k}'}(\mathbf{r})\phi_{-\vec{k}}(\mathbf{r})\phi_{\vec{k}}(\mathbf{r})d\mathbf{r} \\
&= -V.
\end{aligned}
\tag{7}
$$

Eq. (6) is the same as the Fröhlich interaction within the BCS approximation.

Note that the two points are not clear in Gor'kov's formalism, i.e., the BCS reduction procedure and the retardation cutoff. To obtain the same result as that of the BCS theory, these two ingredients should be taken care of in some way. As will be shown later, the negligence of the BCS reduction procedure causes a serious pairing problem especially in impure superconductors.

In the presence of impurities, the matrix element of the pairing interaction is

$$
V_{nn'} = -V\int \psi^*_{n'}(\mathbf{r})\psi^*_{\bar{n}'}(\mathbf{r})\psi_{\bar{n}}(\mathbf{r})\psi_n(\mathbf{r})d\mathbf{r}. \tag{8}
$$

Substituting Eq. (3) into Eq. (8) we find that

$$
V_{nn'} = -V\sum_{\vec{k}\vec{k}'\vec{q}} < \vec{k}-\vec{q}|n' >< \vec{k}'+\vec{q}|\overline{n}' >< \vec{k}'|\overline{n} >^* < \vec{k}|n >^* . \tag{9}
$$

Notice that Eq. (9) is the same as Eq. (5).

III. PAIRING CONSTRAINT ON GOR'KOV'S FORMALISM

A. Inhomogeneous System: Nonmagnetic Impurity Case

Near the transition temperature, the usual self-consistency equation is

$$\Delta(\mathbf{r}) = VT\sum_{\omega}\int \Delta(\mathbf{l})G^{\uparrow}_{\omega}(\mathbf{r},\mathbf{l})G^{\downarrow}_{-\omega}(\mathbf{r},\mathbf{l})d\mathbf{l}$$
$$= \int K(\mathbf{r},\mathbf{l})\Delta(\mathbf{l})d\mathbf{l}. \tag{10}$$

Note that Kernel $K(\mathbf{r},\mathbf{l})$ is not for Anderson's pairing. It includes the extra pairings between $n\uparrow$ and $n'(\neq \bar{n})\downarrow$. The kernel for Anderson's pairing is

$$K^A(\mathbf{r},\mathbf{l}) = VT\sum_{\omega}\{G^{\uparrow}_{\omega}(\mathbf{r},\mathbf{l})G^{\downarrow}_{-\omega}(\mathbf{r}',\mathbf{l})\}_{p.p.}$$
$$= V\sum_{n}\frac{1}{2\epsilon_n}tanh\frac{\epsilon_n}{2T}\psi_n(\mathbf{r})\psi_{\bar{n}}(\mathbf{r})\psi^*_{\bar{n}}(\mathbf{l})\psi^*_n(\mathbf{l}), \tag{11}$$

where p.p. means proper pairing constraint, which dictates pairing between $n\uparrow$ and $\bar{n}\downarrow$.

It can be shown[10,11] that the extra pairings violate the physical constraint of the Anomalous Green's function, i.e.,

$$\overline{F(\mathbf{r},\mathbf{r}',\omega)}^{imp} \sim \overline{\psi_{n\uparrow}(\mathbf{r})\psi_{n'\downarrow}(\mathbf{r}')}^{imp}$$
$$\neq \overline{F(\mathbf{r}-\mathbf{r}',\omega)}^{imp}. \tag{12}$$

These extra pairings should have been eliminated by the BCS reduction procedure in the Hamiltonian. Consequently, the revised self-consistency equation is

$$\Delta(\mathbf{r}) = VT\sum_{\omega}\int \Delta(\mathbf{l})\{G^{\uparrow}_{\omega}(\mathbf{r},\mathbf{l})G^{\downarrow}_{-\omega}(\mathbf{r},\mathbf{l})\}_{p.p.}d\mathbf{l}. \tag{13}$$

Notice that Eq. (13) is nothing but another form of the BCS gap equation,

$$\Delta_n = \sum_{n'} V_{nn'}\frac{\Delta_{n'}}{2E_{n'}}tanh\frac{E_{n'}}{2T}. \tag{14}$$

B. Inhomogeneous System: Magnetic Impurity Case

KO's [7] theory employed degenerate scattered state pairs. It has been claimed that the inclusion of the extra pairing is the origin of the so-called pair-breaking of the magnetic impurities.[16,17] However, the extra pairing terms cause the violation of the physical constraint of the pair potential and the Anomalous Green's function.[11] It can be shown[11] that the homogeneity condition of the Anomalous Green's function, after the impurity average, requires pairing between the degenerate scattered state partners. Then the revised self-consistency equation gives rise to KO's result.[7]

C. Homogeneous System

Near the transition temperature, the Anomalous Green's function is given by

$$F(\mathbf{r}, \mathbf{r}', \omega) = \int \Delta(\mathbf{l}) G^{\uparrow}_{\omega}(\mathbf{r}, \mathbf{l}) G^{\downarrow}_{-\omega}(\mathbf{r}', \mathbf{l}) d\mathbf{l}, \tag{15}$$

Gor'kov[12] pointed out that $F(\mathbf{r}, \mathbf{r}')$ should depend only on $\mathbf{r} - \mathbf{r}'$, i.e.,

$$F(\mathbf{r}, \mathbf{r}') = F(\mathbf{r} - \mathbf{r}'). \tag{16}$$

Note that Eq. (15) includes the extra pairing terms between $\vec{k} \uparrow$ and $\vec{k}' \downarrow$ $(\neq -\vec{k} \downarrow)$, which do not satisfy the homogeneity condition of Eq. (16). In this case, the self-consistency condition of the pair potential happens to eliminate the extra pairing in Eq. (15) because of the orthogonality of the wavefunctions.

However, it is important to eliminate the extra pairing in the Anomalous Green's function from the beginning. Note that the kernel $K(\mathbf{r}, \mathbf{l})$ is not for the pairing between $\vec{k} \uparrow$ and $-\vec{k} \downarrow$, but for the pairing between the states which are the linear combination of the plane wave states $\phi_{\vec{k}}(\mathbf{r})$.[18] The inclusion of the extra pairings hindered our correct understanding of the impure superconductors and the relation between the pair potential and the gap parameter.

IV. PAIRING CONSTRAINT ON THE BOGOLIUBOV-DE GENNES EQUATIONS

A. Inhomogeneous system: Nonmagnetic Impurity Case

By performing a unitary transformation,

$$\Psi(\mathbf{r}\uparrow) = \sum_n (\gamma_{n\uparrow} u_n(\mathbf{r}) - \gamma^\dagger_{n\downarrow} v^*_n(\mathbf{r}))$$
$$\Psi(\mathbf{r}\downarrow) = \sum_n (\gamma_{n\downarrow} u_n(\mathbf{r}) + \gamma^\dagger_{n\uparrow} v^*_n(\mathbf{r})), \tag{17}$$

we obtain the well-known Bogoliubov-de Gennes equations. To understand the physical meaning of the transformation (17), we express $\gamma_{n\uparrow}$ and $\gamma_{n\downarrow}$ by the creation and destruction operators for an electron in the scattered state,[18]

$$\gamma_{n\uparrow} = \sum_{n'} (u^*_{n,n'} c_{n'\uparrow} + v_{n,n'} c^\dagger_{n'\downarrow}),$$
$$\gamma_{n\downarrow} = \sum_{n'} (u^*_{n,n'} c_{n'\downarrow} - v_{n,n'} c^\dagger_{n'\uparrow}), \tag{18}$$

where

$$u_{n,n'} = \int \psi^*_{n'}(\mathbf{r}) u_n(\mathbf{r}) d\mathbf{r}$$
$$v_{n,n'} = \int \psi^*_{n'}(\mathbf{r}) v^*_n(\mathbf{r}) d\mathbf{r}. \tag{19}$$

Accordingly, we obtain a vacuum state where $u_n(\mathbf{r})\uparrow$ and $v^*_n(\mathbf{r})\downarrow$ (instead of $\psi_n(\mathbf{r})\uparrow$ and $\psi_{\bar{n}}(\mathbf{r})\downarrow$) are paired. They are the superpositions of the scattered states. It is clear that the Bogoliubov-de Gennes equations cannot give rise to the correct Anderson's pairing because of the position dependence of the pair potential. We need to supplement a pairing condition. If we assume a constant pair potential,[16,17] Anderson's pairing is obtained. However, then, the impurity effect on the phonon-mediated interaction is gone.

In the presence of magnetic impurities, even the constant pair potential gives a pairing between the states which are the linear superpositions of the scattered states.

B. Homogeneous System

As in Sec. IV. A, the unitary transformation generates a vacuum state and the self-consistency equation for the pairing between $u_n(\mathbf{r})\uparrow$ and $v^*_n(\mathbf{r})\downarrow$, (instead of $\phi_{\vec{k}}(\mathbf{r})\uparrow$ and $\phi_{-\vec{k}}(\mathbf{r})\downarrow$). Note that $u_n(\mathbf{r})\uparrow$ and $v^*_n(\mathbf{r})\downarrow$ are the linear superpositions of the plane wave states until we constrain them. In this case, setting the pair potential gives a pairing between the plane wave states. However, the kernel of the self-consistency equation has not been set accordingly.

V. PAIR POTENTIAL AND GAP PARAMETER

For a homogeneous system, it was shown[19]

$$\Delta(\mathbf{r}-\mathbf{r}') = \int d\vec{k} e^{i\vec{k}\cdot(\mathbf{r}-\mathbf{r}')}\Delta_{\vec{k}}. \tag{20}$$

But this relation is not exact because of the retardation cutoff.

Correct relation may be obtained only after incorporating the pairing constraint into the self-consistency equation. It is given

$$\Delta(\mathbf{r}-\mathbf{r}') = V\sum_{\vec{k}} \frac{\Delta_{\vec{k}}}{2E_{\vec{k}}} tanh\frac{E_{\vec{k}}}{2T}\phi_{\vec{k}}(\mathbf{r})\phi_{-\vec{k}}(\mathbf{r}'). \tag{21}$$

Comparing Eq. (21) with the BCS gap equation and using Eq. (7), we also find

$$\Delta_{\vec{k}} = \int \phi^*_{\vec{k}}(\mathbf{r})\phi^*_{-\vec{k}}(\mathbf{r})\Delta(\mathbf{r})d\mathbf{r}. \tag{22}$$

In the presence of impurities, one finds that

$$\Delta(\mathbf{r}) = V\sum_{n} \frac{\Delta_n}{2E_n} tanh\frac{E_n}{2T}\psi_n(\mathbf{r})\psi_{\bar{n}}(\mathbf{r}), \tag{23}$$

and

$$\Delta_n = \int \psi^*_n(\mathbf{r})\psi^*_{\bar{n}}(\mathbf{r})\Delta(\mathbf{r})d\mathbf{r}. \tag{24}$$

Eq. (24) was obtained first by Ma and Lee.[11]

VI. REINVESTIGATION OF INHOMOGENEOUS SUPERCONDUCTORS

Now we need to reinvestigate the inhomogeneous superconductors studied by Gor'kov's formalism or the Bogoliubov-de Gennes equations. In particular, Gor'kov's microscopic derivation[12] of the Gizburg-Landau equation is not valid. The gradient term may not be derived microscopically. To tackle the inhomogeneous problems, we must choose a correct pairing and calculate the correct kernel for each problem. Then the BCS theory may be more easy to apply. It is not clear whether the pair potential is the more appropriate quantity than the gap parameter in inhomogeneous superconductors.

The following problems are required to restudy:

1. Dirty superconductors and localization
2. Magnetic impurities
3. Proximity effect, Andreev reflection and Josephson effect
4. Mesoscopic superconductivity
5. Type II superconductors, vortex problem
6. Non-equilibrium superconductivity
7. High T_c superconductors, heavy fermion superconductors

A. Theory of Dirty Superconductors

Table I lists the theories of impure superconductors. Notice that Suhl and Matthias[20] and Abrikosov-Gor'kov[3] theories for ΔT_c are essentially equivalent. Both theories over-estimate the change of the density of states caused by impurity scattering, because they apply a retardation cutoff to the energy of the plane wave states and not of the scattered states.

TABLE I. Theories of impure superconductors

	Ordinary impurity	Magnetic impurity
Anderson	$T_c = T_{co}$	
AG	$T_c = T_{co} - \frac{T_{co}}{\pi\omega_D\tau}(\frac{1}{\lambda} + \frac{1}{2})$	$T_c = T_{co} - \frac{\pi}{4}\frac{1}{\tau_s}$
Suhl and Matthias	$T_c \cong T_{co} - \frac{T_{co}}{\lambda\omega_D\tau}$	$T_c = T_{co} - \frac{\pi}{3.5}\frac{1}{\tau_s}$
Baltensperger		$T_c = T_{co} - \frac{\pi}{4}\frac{1}{\tau_s}$
Tsuneto	$T_c = T_{co}$	
KO	$T_c = T_{co} - \frac{T_{co}}{\pi\lambda E_F\tau}$	$T_c = T_{co} - \frac{0.18\pi}{\lambda\tau_s}$
	− localization correction (Kim)	

Now we consider strong coupling theories of dirty superconductors. Tsuneto[6] obtained the gap equation

$$\Sigma_2(\omega) = \frac{i}{(2\pi)^3 p_o} \int dq \int d\epsilon \int d\omega' \frac{qD(q,\omega-\omega')\eta(\omega')\Sigma_2(\omega')}{\epsilon^2 - \eta^2(\omega')\omega'^2}, \tag{25}$$

where $\eta = 1 + \frac{1}{2\tau|\omega|}$, and τ is the collision time. On the other hand, Kim[8] obtained a gap equation

$$\Delta^*(\omega_n, m) = \sum_{n'} \lambda(\omega_n - \omega_{n'}) \sum_{m'} V_{mm'} \frac{\Delta^*(\omega_{n'}, m')}{[-i\omega_{n'} - \epsilon_{m'}][i\omega_{n'} - \epsilon_{m'}]}, \tag{26}$$

where

$$V_{mm'} = g^2 \int |\psi_m(\mathbf{r})|^2 |\psi_{m'}(\mathbf{r})|^2 d\mathbf{r}. \tag{27}$$

Comparing Eqs. (25) and (26), we find that Tsuneto's result misses the most important factor $V_{mm'}$, which gives the change of the phonon-mediated interaction due to impurities. This factor is exponentially small for the localized states.

B. Suppression of Magnetic Impurity Effect by Ordinary Impurities

KO's results[7] can be explained physically. People used to notice that the impurity effect is stronger in the low T_c material than in the high T_c cuprate materials. Because the size of Cooper pair is very much smaller in high T_c material, it sees a very small number of the impurities and so is not much influenced. Consequently, T_c change and its initial slope depend on the material, which is predicted by KO. However, the AG theory predicts the universal slope. Because the AG theory pairs the states, which are the superpositions of the normal states of the material, the very nature of the material does not play an important role.

When the conduction electrons have a mean free path that is smaller than the size of the Cooper pair (for a pure superconductor), the effective size is reduced. Accordingly, if we add ordinary impurities to the superconductors with the magnetic impurities enough to reduce the size of the Cooper pair, the magnetic impurity effect is partially suppressed. This compensation phenomena has been confirmed in several experiments.[13–15]

The notion of gapless superconductor is based on the misunderstanding of the relation between the pair potential and the gap parameter. ¿From Eq. (23), it is clear that the pair potential cannot be finite when the gap parameter is zero. It seems that the long range order between the magnetic impurities or conduction electrons (especially in lead) caused the gaplesslike behavior in the experiments. Then it would rather be called zero gap superconductors or superconductors with nodes.

C. Weak Localization Correction to the Phonon-mediated Interaction

For the strongly localized states, the phonon-mediated interaction is exponentially small like the conductance. It is then expected that the same weak localization correction terms may occur in both quantities. Using the wavefunction obtained by Mott and Kaveh,[21] it can be shown that[11]

$$
\begin{aligned}
V_{nn'}^{3d} &\cong -V[1-\frac{1}{(k_F\ell)^2}(1-\frac{\ell}{L})], \\
V_{nn'}^{2d} &\cong -V[1-\frac{2}{\pi k_F\ell}ln(L/\ell)], \\
V_{nn'}^{1d} &\cong -V[1-\frac{1}{(\pi k_F a)^2}(L/\ell-1)],
\end{aligned} \tag{28}
$$

where a is the radius of the wire.

There are many experimental results which show the reduction of T_c caused by weak localization.[22,23] Previously, it was interpreted by the enhanced Coulomb repulsion. However, Dynes et al.[23] found a decrease of the Coulomb pseudo-potential μ^* with decreasing T_c. We believe that this signals the importance of weak localization correction to the phonon-mediated interaction.

D. Other Inhomogeneous Superconductors

We call attention to a few remarks against the conventional real space formalism.

"By the use of the Gor'kov technique, Abrikosov and Gor'kov have succeeded in $\cdots$, it is *entirely incorrect* as far as any physical results are concerned."[19]

"Certain *inconsistencies* are seen to develop with the use of this approach (the de Gennes-Werthamer model), calling into question previous results obtained."[24]

"Is the discrepancy $\cdots$ indicative of the certain *fundamental inconsistencies* in the Green's function formulation of the theory of nonstationary superconductivity?"[25]

VII. CONCLUSIONS

It is shown that Gor'kov's formalism and the Bogoliubov-de Gennes equations need a pairing constraint. The resulting self-consistency equation is nothing but another form of the BCS gap equation. Most inhomogeneous superconductors should be reinvestigated.

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