Relativistic Theory of Superconductivity

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Abstract

A relativistic generalization of the Bogolubov-de Gennes equations is derived where the particle and hole amplitudes are Dirac spinors. In the weakly relativistic limit one obtains, besides the usual spin-orbit, Darwin and kinetic energy corrections, a new "spin-orbit" and a new "Darwin" term involving the pair potential in place of the electrostatic potential. The results indicate significance of the relativistic corrections for superconductors with a high Fermi velocity, a small coherence length or heavy elements in the lattice.

Relativistic effects play an important role at several places in the theory of superconductivity : First of all, it is a well-known fact that the traditional (non-relativistic) theory of Bardeen, Cooper and Schrieffer (BCS) [1] gives a rather poor description of the spin susceptibility found experimentally in the superconducting phase [2]. Inclusion of the spin-orbit coupling, which is a relativistic effect of order $(v/c)^2$, provides a partial explanation of the discrepancy [3, 4, 5]. Second, theoretical attempts to describe magnetic impurities in superconductors and the coexistence of magnetism and superconductivity have taken the spin-orbit coupling into account [6, 7, 8]. Third, the spin-orbit coupling is known to affect the symmetry of the order parameter. This is of particular importance for the heavy-fermion superconductors, where currently much experimental and theoretical effort is put in a determination of this symmetry [9, 10, 11, 12, 13, 14]. Finally, the Meissner effect can be regarded as an intrinsically relativistic effect [15] since it is associated with an energy contribution of order $(v/c)^2$.

The non-relativistic standard theory of inhomogeneous superconductors [16, 17, 18, 19, 20] is based on the Bogolubov-de Gennes Hamiltonian

$$H_{non-rel} = \int d^3r \sum_\sigma \psi_\sigma^\dagger(\mathbf{r}) \left[\frac{\pi^2}{2m} + w(\mathbf{r}) - \mu \right] \psi_\sigma(\mathbf{r}) - \int d^3r \left[D^*(\mathbf{r}) \psi_\uparrow(\mathbf{r}) \psi_\downarrow(\mathbf{r}) + H.c. \right] \quad (1)$$

where $\psi_\sigma^\dagger(\mathbf{r})$ and $\psi_\sigma(\mathbf{r})$ are the usual (non-relativistic) field operators. σ is a spin index and $H.c.$ denotes the Hermitian conjugate. The first term of (1) is a single-particle Schrödinger Hamiltonian containing an effective electrostatic potential $w(\mathbf{r})$ which consists of the Coulomb potential of the ionic lattice and, in the most general case, an additional external voltage applied to the system. μ is the chemical potential and π is the generalized momentum $\mathbf{p} - \frac{q}{c}\mathbf{A}$ where $\mathbf{A}$ is an external vector potential and q is the charge of the particles involved. The second term of (1) contains the pair potential $D(\mathbf{r})$ that couples to the superconducting order parameter $\Delta(\mathbf{r}) = \psi_\uparrow(\mathbf{r})\psi_\downarrow(\mathbf{r})$. For the time being, the three potentials $w(\mathbf{r})$, $\mathbf{A}(\mathbf{r})$ and $D(\mathbf{r})$ are regarded as given fields.

All the work on relativistic effects mentioned above is based on the Hamiltonian (1) with the additional inclusion of the spin-orbit term in the potential $w(\mathbf{r})$. This procedure, however, does not treat relativistic effects in a consistent way because it includes only relativistic corrections arising from $w(\mathbf{r})$ but neglects possible relativistic corrections of the same order in (v/c) associated with the pair potential $D(\mathbf{r})$.

It is the purpose of the present work to provide a relativistically consistent theory of superconductivity on the single-particle level that treats all relativistic effects on the same footing. We shall first present a fully relativistic treatment leading to a set of Dirac type Bogolubov-de Gennes equations. Then we take the weakly relativistic limit of these equations to order $(v/c)^2$. One obtains, in addition to the usual spin-orbit, Darwin and kinetic energy corrections, a new "spin-orbit" and a new "Darwin" term involving the pair potential $D(\mathbf{r})$.

Obviously the relativistic generalization of the first term on the right-hand side of Eq. (1) is the Dirac Hamiltonian

$$\int d^3r\, \bar{\Psi}(\mathbf{r}) \left[c\gamma \cdot \mathbf{p} + mc^2 + q\gamma_\nu A^\nu\right] \Psi(\mathbf{r}) \tag{2}$$

with the four potential $A^\nu = (\frac{w(\mathbf{r})-\mu}{q}, \mathbf{A}(\mathbf{r}))$ and the four vector of gamma matrices γ_ν [21]. $\Psi(\mathbf{r})$ denotes the Dirac spinor field operator and $\bar{\Psi}(\mathbf{r}) = \Psi^\dagger(\mathbf{r})\gamma_0$. In order to find the corresponding relativistic generalization of the second term in Eq. (1) we have to construct a covariant extension of the non-relativistic singlet order parameter $\Delta(\mathbf{r}) = \psi_\uparrow(\mathbf{r})\psi_\downarrow(\mathbf{r})$. This is achieved by requiring the Cooper pair to consist of time-conjugate states, i.e. the relativistic order parameter is required to be a Kramers pair, $\Delta_{rel}(\mathbf{r}) = b\Psi^T(\mathbf{r})\mathcal{T}\Psi(\mathbf{r})$. Here $\Psi^T(\mathbf{r})$ is the transposed Dirac field operator and $\mathcal{T} = i\gamma^1\gamma^3$ is the time-reversal operator. The proportionality constant b is determined in such a way that $\Delta_{rel}(\mathbf{r})$ reduces to $\Delta(\mathbf{r})$ in the non-relativistic limit. This leads to $\Delta_{rel}(\mathbf{r}) = \Psi^T(\mathbf{r})\eta\Psi(\mathbf{r})$ with

$$\hat{\eta} = \frac{1}{2}\begin{pmatrix} 0 & 1 & 0 & 0 \\ -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & -1 & 0 \end{pmatrix}. \tag{3}$$

It follows that $\Delta_{rel}(\mathbf{r})$ has a simple and definite transformation behaviour under Lorentz transformations : it is a Lorentz scalar. The fully relativistic Hamiltonian is then given by

$$H = \int d^3r \bar{\Psi}(\mathbf{r}) \left[c\gamma \cdot \mathbf{p} + mc^2 + q\gamma_\mu A^\mu\right] \Psi(\mathbf{r}) - \int d^3r \left[\Psi^T(\mathbf{r})\, \hat{\eta}\, \Psi(\mathbf{r}) D^*(\mathbf{r}) + H.c.\right]. \tag{4}$$

Next we search for a set of single-particle eigenvalue equations describing the excitation spectrum of this Hamiltonian. H can be diagonalized by means of the unitary and canonical transformation

$$\Psi_i(\mathbf{r}) = \sum_{kj} \left(u_{ikj}(\mathbf{r})\gamma_{kj} + v^*_{ikj}(\mathbf{r})\gamma^\dagger_{kj} \right) \tag{5}$$

which is a straightforward generalization of the well-known Bogolubov transformation [16]. The operators $\gamma^\dagger_{kj}$ and γ_{kj} are the new quasi-particle creation and annihilation operators. The indices i and j refer to components of four-spinors while the index k stands for all quantum numbers that characterize the quasi-particle states. For homogeneous systems k represents the wave vector, in the presence of a periodic lattice potential it additionally contains a band index and for the case of spherical symmetry it includes angular quantum numbers. In analogy to the non-relativistic case [17], the unitarity and canonicity conditions lead to a set of generalized orthonormality and completeness relations for the amplitudes $u_{ikj}(\mathbf{r})$ and $v_{ikj}(\mathbf{r})$, which will be presented elsewhere [22].

The Bogolubov transformation (5) leads to a set of eight coupled differential equations which, in terms of the Dirac spinors $u_{kj} = (u_{1kj}..u_{4kj})^T$ and $v_{kj} = (v_{1kj}..v_{4kj})^T$, can be condensed in two 4×4 equations.

$$\gamma_0[c\gamma \cdot \mathbf{p} + mc^2(I - \gamma_0) + q\gamma_\mu A^\mu]u_{kj}(\mathbf{r}) = E_{kj}u_{kj}(\mathbf{r}) - D(\mathbf{r})\,(2\hat{\eta})\,v_{kj}(\mathbf{r}) \tag{6}$$

$$\gamma_0[c\gamma \cdot \mathbf{p} + mc^2(I - \gamma_0) + q\gamma_\mu A^\mu]^* v_{kj}(\mathbf{r}) = -E_{kj}v_{kj}(\mathbf{r}) + D^*(\mathbf{r})\,(2\hat{\eta}^T)\,u_{kj}(\mathbf{r}) \tag{7}$$

where I represents the 4×4 unit matrix. We have subtracted mc^2 from the Dirac operator to facilitate the transition to the weakly relativistic limit. Eqs. (6) and (7) constitute one of the main results of the present work. They will in the following be called Dirac-Bogolubov-de Gennes equations. Their algebraic structure is very similar to that of the traditional non-relativistic Bogolubov-de Gennes equations [16]. The main difference is that the particle and hole amplitudes $u_{kj}(\mathbf{r})$ and $v_{kj}(\mathbf{r})$ in Eqs. (6) and (7) are Dirac spinors while they are single-component functions in the non-relativistic Bogolubov-de Gennes equations.

Specializing to homogeneous systems ($w(\mathbf{r}) = const \equiv 0$ and $D(\mathbf{r}) = const \equiv D$) without magnetic fields ($\mathbf{A}(\mathbf{r}) \equiv 0$) the energy spectrum of Eqs. (6) and (7) can be determined exactly. A straightforward calculation gives

$$E_k = \pm\sqrt{(\epsilon_k \pm a)^2 + |D|^2} \tag{8}$$

where $a = mc^2 + \mu$ and $\epsilon_k = +\sqrt{(\hbar k)^2 c^2 + m^2 c^4}$. The energy spectrum has four branches corresponding to the four possible choices of the signs (see Fig. 1). We can immediately check two important limiting cases: (i) In the non-superconducting limit ($D \equiv 0$) Eq.(8) reduces to $\pm(\epsilon_k \pm a)$. The two branches $\pm\epsilon_k - a$ are the usual Dirac spectrum, shifted by a. The remaining two branches, $-(\pm\epsilon_k - a)$, being the negative of the first, represent the hole spectrum, as always for Bogolubov-de Gennes-type equations. (ii) In the non-relativistic limit ($v/c \to 0$), Eq.(8) reduces to the well known BCS result $\pm\sqrt{(\hbar^2 k^2/2m - \mu)^2 + |D|^2}$.

In both the relativistic and the non-relativistic case, the superconducting gap (see Fig. 1) is given by $2|D|$. The relativistic theory predicts, however, that the position of the gap is slightly shifted away from the Fermi wave vector k_F. One finds

$$k_{gap}^2 = k_F^2 (1 + \frac{1}{4}(v_F/c)^2) \tag{9}$$

with the Fermi velocity v_F. The predicted shift is of the same order of magnitude as the experimentally confirmed relativistic correction to the Cooper-pair mass [23].

For inhomogeneous systems a complete numerical solution of the Dirac-Bogolubov-de Gennes equations (6) and (7) is required. Since, for ordinary matter, terms of higher than second order in (v/c) are negligibly small it is desirable to have a set of weakly relativistic (Pauli-type rather than Dirac-type) Bogolubov-de Gennes equations. Such equations would be easier to handle and, in addition, they would allow one to identify the superconducting analogues of the spin-orbit and Darwin terms. In the remaining part of this paper we determine the weakly relativistic limit of the Dirac-Bogolubov-de Gennes equations to second order in (v/c). Straightforward application of the Pauli elimination method [24] leads to

$$(H_0 + \tilde{H}_2) \begin{pmatrix} u_{kj} \\ v_{kj} \end{pmatrix} = E_{kj} \begin{pmatrix} u_{kj} \\ v_{kj} \end{pmatrix} \tag{10}$$

where the particle and hole amplitudes u_{kj} and v_{kj} are now Pauli spinors $u_{kj} = (u_{1kj}, u_{2kj})^T$

and $v_{kj} = (v_{1kj}, v_{2kj})^T$. The zero-order term

$$H_0 = \begin{pmatrix} (\sigma\cdot\pi)^2/2m + (w(\mathbf{r}) - \mu) & D(\mathbf{r})\,(i\sigma_y) \\ -(i\sigma_y)D^*(\mathbf{r}) & -(\sigma^*\cdot\pi^*)^2/2m - (w(\mathbf{r}) - \mu) \end{pmatrix} \tag{11}$$

is the well-known [16] non-relativistic spin-Bogolubov-de Gennes Hamiltonian.The second-order term is given by

$$\tilde{H}_2 = \frac{1}{4m^2c^2}\begin{pmatrix} (\sigma\cdot\pi)((w(\mathbf{r}) - \mu) - E_{kj})(\sigma\cdot\pi) & i(\sigma\cdot\pi)\,D\,\sigma_y\,(\sigma^*\cdot\pi^*) \\ -i(\sigma^*\cdot\pi^*)D^*\sigma_y\,(\sigma\cdot\pi) & (\sigma^*\cdot\pi^*)(-(w(\mathbf{r}) - \mu) - E_{kj})(\sigma^*\cdot\pi^*) \end{pmatrix} \tag{12}$$

Eq. (10) is not a standard eigenvalue equation because $\tilde{H}_2$ contains the eigenvalue E_{kj} to be calculated [24]. To underline this fact we write $\tilde{H}_2(E_{kj})$. In the following we determine an *energy-independent* Hamiltonian H_2 such that the spectrum of $(H_0 + H_2)$ is identical to order $(v/c)^2$ with the spectrum of $(H_0 + \tilde{H}_2)$. By first-order perturbation theory, H_2 must satisfy the equation

$$\langle\psi_{kj}^{(0)}|H_2|\psi_{kj}^{(0)}\rangle = \langle\psi_{kj}^{(0)}|\tilde{H}_2(E_{kj}^{(0)})|\psi_{kj}^{(0)}\rangle = (\langle\psi_{kj}^{(0)}|\tilde{H}_2(E_{kj}^{(0)})|\psi_{kj}^{(0)}\rangle + \langle\psi_{kj}^{(0)}|\tilde{H}_2(E_{kj}^{(0)})|\psi_{kj}^{(0)}\rangle^*)/2 \tag{13}$$

where $|\psi_{kj}^{(0)}\rangle$ is a shorthand for the eigenstates $(u_{kj}^{(0)}, v_{kj}^{(0)})^T$ of H_0 and $E_{kj}^{(0)}$ are the corresponding energy eigenvalues. The last equality in (13) follows from the fact that $\tilde{H}_2(E)$ is a Hermitian operator for *any* value of E.

For simplicity we now consider systems without magnetic fields so that

$$H_0 = \begin{pmatrix} p^2/2m + (w(\mathbf{r}) - \mu) & D(\mathbf{r})(i\sigma_y) \\ -(i\sigma_y)D^*(\mathbf{r}) & -p^2/2m - (w(\mathbf{r}) - \mu) \end{pmatrix} \tag{14}$$

and

$$\begin{aligned} \tilde{H}_2 &= \frac{1}{4m^2c^2}\begin{pmatrix} \sigma\cdot(-i\hbar\nabla w)(\sigma\cdot p) & i(\sigma\cdot p)D\sigma_y(\sigma^*\cdot p^*) \\ -i(\sigma^*\cdot p^*)D^*\sigma_y(\sigma\cdot p) & -\sigma^*\cdot(+i\hbar\nabla w)(\sigma^*\cdot p^*) \end{pmatrix} \\ &+ \begin{pmatrix} (w(\mathbf{r}) - \mu) - E_{kj} & 0 \\ 0 & -(w(\mathbf{r}) - \mu) - E_{kj} \end{pmatrix}\frac{p^2}{4m^2c^2} \end{aligned} \tag{15}$$

Inserting this representation of $\tilde{H}_2$ in the r.h.s. of Eq. (13) and using the fact that

$$\left\langle\psi_{kj}^{(0)}\right|\begin{pmatrix} (w(\mathbf{r}) - \mu) - E_{kj}^{(0)} & 0 \\ 0 & -(w(\mathbf{r}) - \mu) - E_{kj}^{(0)} \end{pmatrix} = \left\langle\psi_{kj}^{(0)}\right|\begin{pmatrix} -p^2/2m & -i\sigma_y D(r) \\ i\sigma_y D^*(r) & p^2/2m \end{pmatrix} \tag{16}$$

the Hamiltonian H_2 is readily identified to be

$$H_2 = \begin{pmatrix} h_2 & d_2 \\ d_2^\dagger & -h_2^* \end{pmatrix} \tag{17}$$

with

$$h_2 = \frac{1}{4m^2c^2}\left(\frac{\hbar^2}{2}\nabla^2 w + \hbar\sigma\cdot(\nabla w)\times p - \frac{p^4}{2m}\right)\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \tag{18}$$

and

$$d_2 = \frac{1}{4m^2c^2}\left(\frac{\hbar^2}{2}\nabla^2 D + \hbar\sigma\cdot(\nabla D)\times p\right)(i\sigma_y). \tag{19}$$

The three terms of h_2 are the usual Darwin, spin-orbit and kinetic energy corrections known from atomic physics. The two terms of d_2 are the central result of our analysis. These new terms are of the *same* order in (v/c) as the well-known terms of h_2 and they have the same algebraic structure. For homogeneous systems, d_2 does not contribute and the energy spectrum of $(H_0 + H_2)$ is easily seen to reproduce the spectrum (8) of the Dirac-Bogolubov-de Gennes Hamiltonian to order $(v/c)^2$. For inhomogeneous systems, the off-diagonal terms d_2 and d_2^+ become relevant. To estimate their magnitude we replace the gradients by suitable mean values, i.e., $|\nabla^2 D| \to \bar{D}/\xi^2$, $|\nabla u| \to \bar{u}/\xi$ and $|\nabla v| \to \bar{v}/\xi$. where ξ is the typical length over which the pair potential and the particle and the hole amplitudes can vary. This typical length is the coherence length of the superconductor. As a consequence, d_2 is found to be proportional to $(\lambda_C/\xi)^2$, where λ_C is the Compton wave length. In the high-temperature superconductors the coherence length is several orders of magnitude smaller than in conventional superconductors. The resulting values of d_2, although still small, are expected to produce a clearly measurable effect that might shed some light on the nature of superconductivity in these materials.

Another interesting effect in inhomogeneous systems comes from the term with the vector product in d_2. Concentrating on one unit cell and assuming $D(\mathbf{r})$ to be spherically symmetric in this cell (which is a reasonable approximation for instance for some superconductors with one atom per unit cell like Nb) this term can be written as

$$\frac{1}{2m^2c^2}\frac{1}{r}\frac{dD}{dr}\mathbf{S}\cdot\mathbf{L}(i\sigma_y). \tag{20}$$

This shows, that the eigenstates of the complete Hamiltonian cannot be simultaneously eigenstates of the spin-operator $\mathbf{S}_z$ and the angular momentum operator $\mathbf{L}_z$. Consequently, the symmetry of the order parameter should be classified according to the total angular momentum $\mathbf{J}$. The occurance of an offdiagonal $\mathbf{S}\cdot\mathbf{L}$ term has previously been postulated

[25] on the basis of group theoretical arguments. The relativistic theory of superconductivity presented above establishes, for the first time, the explicit form of the coefficient to be $(2m^2c^2r)^{-1}(dD/dr)$. Previous treatments of the symmetry of the order parameter [10, 13, 26] have focussed on the relativistic corrections due to h_2. It is, however, essential to include the new *offdiagonal* symmetry breaking terms of d_2 in a complete analysis of the order parameter. This is of particular importance for the heavy-fermion superconductors. In these systems the presence of heavy elements favours relativistic effects. The offdiagonal terms d_2 and $d_2^\dagger$, having roughly the magnitude of $(\lambda_C/\xi)^2$, are nevertheless very small in the heavy-fermion superconductors. However, the mere presence of these symmetry breaking terms, no matter how small, affects the possible symmetries of the order parameter. Applications of the theory presented in this letter to these systems are currently under study.

So far the vector potential has been treated as a *given* field. However, in an actual superconducting system placed in a given external field, $\mathbf{A}_{ex}(\mathbf{r})$, supercurrents are set up which act as sources of an induced field, $\mathbf{A}_{in}(\mathbf{r})$, resulting in a total field $\mathbf{A}(\mathbf{r}) = \mathbf{A}_{ex}(\mathbf{r}) + \mathbf{A}_{in}(\mathbf{r})$. The induced field is essential, e.g., for the description of the Meissner effect. $\mathbf{A}_{in}$ has to be determined self-consistently by the following procedure: We first solve the Dirac-Bogolubov-de Gennes equations (6) and (7) or the weakly relativistic Bogolubov-de Gennes equations (10) with the external vector potential $\mathbf{A}_{ex}(\mathbf{r})$. The resulting particle and hole amplitudes $u(\mathbf{r})$ and $v(\mathbf{r})$ determine the current density $\mathbf{j}(\mathbf{r})$. From the current density the induced vector potential $\mathbf{A}_{in}(\mathbf{r})$ is obtained by solving the Maxwell equation $\nabla \times (\nabla \times \mathbf{A}_{in}(\mathbf{r})) = \frac{4\pi}{c}\mathbf{j}(\mathbf{r})$ which leads to a new total field $\mathbf{A}(\mathbf{r}) = \mathbf{A}_{ex}(\mathbf{r}) + \mathbf{A}_{in}(\mathbf{r})$. With this total vector potential the Dirac- or the weakly relativistic Bogolubov-de Gennes equations are solved again, leading to a new current density, etc. The cycle is repeated until selfconsistency is achieved. It should be emphasized that the energy, $(1/c^2) \cdot \int d^3r d^3r' \, \mathbf{j}(\mathbf{r}) \cdot \mathbf{j}(\mathbf{r}')/|\mathbf{r} - \mathbf{r}'|$, associated with the induced currents is of the same relativistic order as the new terms presented in this work. A consistent treatment of the Meissner effect and, in particular, the calculation of the penetration depth should therefore include the new terms as well. Detailed results will be presented elsewhere [22].

In a more complete treatment, the potentials $D(\mathbf{r})$ and $w(\mathbf{r})$ will have to be determined self-consistently as well. This requires a consistent relativistic treatment of the particle-particle interaction, leading, e.g., to the inclusion of the Breit interaction. In this context, variationally stable two-component Hamiltonians such as the Douglas-Kroll-Hess operator [27, 28] will have to be considered as well. To determine $D(\mathbf{r})$ and $w(\mathbf{r})$ self-consistently, a relativistic extension [29] of the density-functional formalism for superconductors [30, 31, 32, 33] is envisaged.

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