On Quartet Superfluidity of Fermionic Atomic Gas

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The possibility of a quartet superfluidity in fermionic systems is studied as a new aspect of atomic gas at ultralow temperatures. The fourfold degeneracy of a hyperfine state and moderate coupling is indispensable for the quartet superfluidity to occur. Possible superconductivity with quartet condensation in electron systems is discussed.

KEYWORDS: quartet Fermi superfluidity, laser-cooled fermionic atomic gas, plasmon mechanism superconductivity

The superfluidity is so far known to be sustained by the Bose-Einstein condensate or the Cooper-pair condensate. The former is realized in liquid $^4$He and some atomic gas of an alkali element,\(^1\)–\(^4\) while the latter is realized in liquid $^3$He and a variety of superconductors. Recently, it has been found that Cooper pairs are formed in the fermionic atomic gas of alkali element\(^5\) and the crossover to the Bose-Einstein condensation of a diatomic molecule is possible with the help of Feshbach resonance.\(^6\)

In principle, there exists the other possibility that superfluidity is sustained by a condensate based on four fermions (quartet) as in the $\alpha$-particle correlation in a light nucleus.\(^7\) An $\alpha$-particle consists of two protons and two neutrons which have approximately 4-fold degeneracy corresponding to $2 \times 2$ degeneracy of the real spin and isotopic spin states. Such 4-fold degeneracy is possible in a fermionic atomic gas with total angular momentum $F = S + I = 3/2$, $S$ and $I$ being the electron spin and nuclear spin, respectively. For example, a Be atom has $F = 3/2$ with $S = 0$ and $I = 3/2$.

The purpose of this Letter is to assess the possibility of quartet superfluidity by analyzing an extended Cooper problem,\(^8\) i.e., a four-fermion problem outside a rigid Fermi surface. This is regarded as a natural extension of the Cooper problem to the present case. We expect that the same tendency as in the “Cooper problem” is taken over to the many-body problem in which the deformation of the Fermi surface should be taken into account self-consistently. One might think that there is another way of extending the Cooper problem to the present case, i.e., by analyzing a stability condition of the BCS state against an addition of four extra fermions to the BCS state. However, we do not take such an approach here because not only mathematical treatments but also the concept of a theoretical framework seems to be much more complicated than ours. The result of this study might also give us a clue to the possibility
of quartet superfluidity in the electronic system with a nearly degenerate Fermi surface.

A theorem by Nagaoka and Usui\textsuperscript{9} states that the ground-state wave function of a many-particle system is symmetric with respect to an interchange of position between any pair of particles if one could neglect the possible spin dependence. Therefore, the ground state of four fermions with “spin” $F = 3/2$ should be

$$\Psi_Q(1, 2, 3, 4) = \Phi(\vec{r}_1, \vec{r}_2, \vec{r}_3, \vec{r}_4)\chi(1, 2, 3, 4),$$

where $\Phi$ is a fully symmetric function with respect to the interchange of $\vec{r}_i \leftrightarrow \vec{r}_j$, and the wave function for the spin is given by the fully antisymmetric one:

$$\chi(1, 2, 3, 4) = \frac{1}{\sqrt{4!}} \begin{vmatrix} \alpha(1) & \beta(1) & \gamma(1) & \delta(1) \\ \alpha(2) & \beta(2) & \gamma(2) & \delta(2) \\ \alpha(3) & \beta(3) & \gamma(3) & \delta(3) \\ \alpha(4) & \beta(4) & \gamma(4) & \delta(4) \end{vmatrix}.$$ (2)

Here, $\alpha, \beta, \gamma, \text{ and } \delta$ denote the “spin” states $F_z = 3/2, 1/2, -1/2, \text{ and } -3/2$, respectively. Due to the Pauli principle, the orbital part $\Phi(\vec{r}_1, \vec{r}_2, \vec{r}_3, \vec{r}_4)$ is a fully symmetric function of $\vec{r}_i$. State (1) represents that of a tetra-atomic molecule (quartet) if the bound-state solution is possible, and is more stable than that of two diatomic molecules whose orbital wave function is not fully symmetric.

Now, we discuss a four-body problem outside the rigid Fermi surface, namely, the Cooper problem. Here, we compare the energy of the quartet and two Cooper pairs. Because it is difficult in general to obtain the exact solution of a four-body problem, we try to solve the problem variationally. The variational solution for $\Phi$ is assumed to be in the Hartree form as

$$\Phi(\vec{r}_1, \vec{r}_2, \vec{r}_3, \vec{r}_4) = \prod_{i=1}^{4} f(\vec{r}_i - \vec{R}),$$

where $\vec{R} \equiv \frac{1}{4} \sum_{i=1}^{4} \vec{r}_i/4$ is the center of gravity. The single-particle state $f(\vec{r})$ is expressed as

$$f(\vec{r}) = \sum_{|\vec{k}| > k_F} f_{\vec{k}} e^{i\vec{k} \cdot \vec{r}}.$$ (4)

It is noted here that only the states outside the Fermi surface $|\vec{k}| > k_F$ are available.

The energy for state (3) is given as

$$E_Q = \int d\vec{r}_1 \cdots d\vec{r}_4 \prod_{i=1}^{4} f(\vec{r}_i - \vec{R}) \left[ -\sum_{i=1}^{4} \frac{\hbar^2 \nabla^2}{2m} + \sum_{i>j} V(\vec{r}_i - \vec{r}_j) \right] \prod_{i=1}^{4} f(\vec{r}_i - \vec{R})$$

$$= 4 \int d\vec{r} f(\vec{r}) \left[ -\frac{\hbar^2 \nabla^2}{2m} + \frac{3}{2} \int d\vec{r}' V(\vec{r} - \vec{r}') |f(\vec{r}')|^2 \right] f(\vec{r}),$$

where $V(\vec{r}_i - \vec{r}_j)$ denotes the two-body interaction between fermions. Optimization of $E_Q$ with
respect to \( f(\vec{r}) \) leads to the equation
\[
\left[ -\frac{\hbar^2 \nabla^2}{2m} + 3 \int d\vec{r}' V(\vec{r} - \vec{r}') |f(\vec{r}')|^2 \right] f(\vec{r}) = \lambda f(\vec{r}),
\] (6)
where \( \lambda \) is the eigenvalue. This equation should be solved under the normalization constraint
\[
\int d\vec{r} |f(\vec{r})|^2 = 1.
\] (7)

The resultant expression of optimized energy is given by
\[
(E_Q)_{\text{min}} = 2 \left[ \lambda - \int d\vec{r} f(\vec{r}) \left( -\frac{\hbar^2 \nabla^2}{2m} \right) f(\vec{r}) \right] + 4 \int d\vec{r} f(\vec{r}) \left( -\frac{\hbar^2 \nabla^2}{2m} \right) f(\vec{r}).
\] (8)

In order to discuss the "Cooper-type problem", let us introduce the \( q \)-representation for the interactions as
\[
V(\vec{r}) = \sum_{\vec{k}} V_{\vec{k}} e^{i\vec{q} \cdot \vec{r}},
\] (9)
and
\[
U(\vec{r}) \equiv 3 \int d\vec{r} V(\vec{r} - \vec{r}') |f(\vec{r}')|^2 = \sum_{\vec{k}} U_{\vec{k}} e^{i\vec{q} \cdot \vec{r}},
\] (10)
where \( U_{\vec{q}} \) is expressed in terms of \( V_{\vec{q}} \) and \( f_{\vec{k}} \) as
\[
U_{\vec{q}} = 3 V_{\vec{q}} \sum_{\vec{p}} f_{\vec{q} - \vec{p}} f_{\vec{p}}.
\] (11)

Then, the effective Schrödinger equation for the "Cooper problem" is given as
\[
\epsilon_k f_{\vec{k}} + \sum_{\vec{k}'} U_{\vec{k} - \vec{k}'} f_{\vec{k}'} = \lambda f_{\vec{k}},
\] (12)
where \( \epsilon \equiv \hbar^2 k^2 / 2m \). It is noted that the wave vector in the wave function \( f_{\vec{k}} \) is restricted in a way such that \( k_F < |\vec{k}| \), \( k_F \) being the Fermi wave number.

In order to solve eq. (12), following BCS, we introduce a model attractive interaction as follows:
\[
V_{\vec{k},\vec{k}'} = \begin{cases} 
-V, & (\epsilon_F < \epsilon_k, \epsilon_{k'} < \epsilon_c); \\
0, & \text{(otherwise)}.
\end{cases}
\] (13)

Then, the second term of eq. (12) is expressed as
\[
\sum_{\vec{k}'} U_{\vec{k} - \vec{k}'} f_{\vec{k}'} = \frac{1}{4\pi^2} \int_{k_F}^{k_c} d\vec{k}' \int_{-1}^1 dt U(|\vec{k} - \vec{k}'|)
\equiv \int_{k_F}^{k_c} dk' U^*(k,k') k'^2 f(k'),
\] (14)
where \( |\vec{k} - \vec{k}'| = (k^2 + k'^2 - 2kk't)^{1/2} \), i.e., \( t \equiv (\vec{k} \cdot \vec{k}') / kk' \), and the kernel \( U^* \) is defined as
\[
U^*(k,k') = \frac{1}{4\pi^2} \int_{-1}^1 dt U(|\vec{k} - \vec{k}'|).
\] (15)
Here, the function $U(|\vec{k} - \vec{k}'|)$, (11), is expressed as

$$U(|\vec{k} - \vec{k}'|) = 3V_{k-k'} \frac{1}{4\pi^2} \int_{-1}^{1} \frac{k}{k_F} \, dp \, f((|\vec{k} - \vec{k}'| - p)), \quad \text{(16)}$$

where $t' \equiv (\vec{p} \cdot (\vec{k} - \vec{k}'))/p|\vec{k} - \vec{k}'|$. 

Finally, by introducing the function $F(k) \equiv kf(k)$, eigenvalue equation (12) is transformed to the following form:

$$\epsilon_k F(k) + \int_{k_F}^{k_c} \, dk' kU^*(k, k')k'F(k') = \lambda F(k). \quad \text{(17)}$$

Now, the kernel $kU^*(k, k')k'$ is symmetric with respect to the interchange $k \leftrightarrow k'$. Then, the eigenvalue problem of eq. (17) is easily solved numerically if the "interaction kernel" $U^*(k, k')$ is given. We solve numerically the self-consistent set of equations, (15)~(17). Then, the energy of the quartet (eq. (8)) is given as

$$\langle E_Q \rangle_{\text{min}} = 2 \left( \lambda + \int_{k_F}^{k_c} \frac{dk}{2\pi^2} \epsilon_k |F(k)|^2 \right). \quad \text{(18)}$$

This energy should be compared with that in the original Cooper problem. The energy of one Cooper pair $E_C$ is given by

$$1 = \frac{V}{4\pi^2} \int_{k_F}^{k_c} \frac{k^2}{\epsilon_k - E_C/2}. \quad \text{(19)}$$

The phase diagram in the $\rho_F V/(k_c/k_F)$ plane, $\rho_F \equiv mk_F/2\pi^2\hbar^2$, is given in Fig. 1 in which equi-energy surface of $(E_Q - 2E_C)/\epsilon_F$ is shown. It is seen that the quartet state is stabilized in the intermediate and strong coupling region with $k_c > 3k_F$. This is consistent with the Nagaoka-Usui theorem which indicates the quartet state to be more stable than the two-Cooper-pair state in the dilute limit, i.e., without the Fermi sea. It is noted that there exists a threshold for eq. (17) to have a bound-state solution, while there exists no threshold for the original Cooper problem (eq. (19)). Therefore, in the limit of weak coupling, the fully symmetric solution in the orbital space will not be the ground state of the “Cooper problem”, in contrast to the 4-body problem in vacuum. It is noted that the quantitativeness of the phase boundary in Fig. 1 may not be very good because we adopted the Hartree approximation to solve the 4-body problem and neglected the interaction between two Cooper pairs as in the BCS approximation. Nevertheless, this gives us a guideline to understand the crossover between the Cooper-pair and the quartet condensations.

Although we adopted here the model interaction (13), it is not so difficult to discuss on the basis of a more realistic interaction similar to the 12-6 Lennard-Jones potential, as we will discuss elsewhere. Since the diatomic bound state is barely possible for the $^4$He system, the weakest interaction and lightest mass, it is possible for a neutral atom with a stronger attraction and a heavier mass, such as Be atom, to form the diatomic bound state. Then, due to the Nagaoka-Usui theorem, the quartet bound state is also expected to be possible. In
addition, the tuning of attractive interaction may be possible with the use of a mechanism similar to the Feshbach resonance for two-body interaction.\textsuperscript{11–13} This would make it possible to search a crossover phenomenon between the Bose-Einstein condensation of tetra-atomic molecules and the quartet Fermi superfluidity.

In order to discuss the relative stability between the Cooper-pair and quartet condensed state, we of course must develop a self-consistent treatment that relaxes the rigidity of the Fermi surface. Let us briefly sketch how to describe the many-body quartet condensed state. A creation operator of a 4-atom molecule with zero total momentum is given as

\begin{equation}
    b^\dagger \equiv \sum_{k_1} \cdots \sum_{k_4} \delta(\vec{k}_1 + \vec{k}_2 + \vec{k}_3 + \vec{k}_4) \Phi(\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{k}_4) a^\dagger_{k_1 \alpha} a^\dagger_{k_2 \beta} a^\dagger_{k_3 \gamma} a^\dagger_{k_4 \delta}.
\end{equation}

It is natural to assume that the wave function $\Phi$ is given by a product of a one-particle state as in eq. (3):

\begin{equation}
    \Phi(\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{k}_4) = \prod_{i=1}^{4} \phi(\vec{k}_i),
\end{equation}

Fig. 1. Phase diagram in $\rho_{F}V-(k_{c}/k_{F})$ plane. Numbers attached to the lines denote the energy difference $(E_{Q} - 2E_{C})$ in the unit of $\epsilon_{F}S$. 

Then the many-body quartet condensed state $|\Psi_{QC}\rangle$ may be written in a manner analogous to the BCS state as

$$|\Psi_{QC}\rangle = \mathcal{P}_{N/4} \exp \left( \sqrt{\frac{N}{4}} b^\dagger \right) |\text{vac}\rangle,$$

where $N$ is the total number of fermions, the projection operator $\mathcal{P}_{N/4}$ works to project out the $N/4$-quartet state, and $|\text{vac}\rangle$ denotes the vacuum. State (22) is equivalent to

$$|\Psi_{QC}\rangle = \mathcal{P}_{N/4} \prod_{\{\vec{k}_1 \sim \vec{k}_4\}} \left( \prod_{i=1}^{4} u(\vec{k}_i) + \prod_{i=1}^{4} v(\vec{k}_i) \times a^\dagger_{\vec{k}_1\alpha} a^\dagger_{\vec{k}_2\beta} a^\dagger_{\vec{k}_3\gamma} a^\dagger_{\vec{k}_4\delta} \right) |\text{vac}\rangle,$$

where the factor $\delta(\vec{k}_1 + \vec{k}_2 + \vec{k}_3 + \vec{k}_4)$ has been abbreviated in the product $\prod_{i=1}^{4}$, and $\phi(\vec{k}) = v(\vec{k})/u(\vec{k})$ and the normalization condition $|u(\vec{k})|^2 + |v(\vec{k})|^2 = 1$ should be fulfilled. The variational functions $u(\vec{k})$ and $v(\vec{k})$ should be determined so as to minimize the free energy, as in the BCS theory.\(^{14}\)

Finally, let us discuss the possibility of observing quartet superconductivity in solids. The most promising case is in metals with two almost degenerate bands, e.g., the $a$- and $b$-bands, and an extremely low carrier density. In such a system, quasi-4-fold degeneracy is maintained for each $k$-point by spin $\sigma = \pm$ and band index $m = a, b$. Diluteness of carriers makes it possible for the plasmon mechanism to work,\(^{15,16}\) and the energy range of attraction $\hbar \omega_{pl}$ extends over the Fermi energy $\epsilon_F$:

$$\frac{\hbar \omega_{pl}}{\epsilon_F} = \frac{4}{\sqrt{3\pi}} \frac{r_0}{a_B} >> 1,$$

where $r_0$ and $a_B$ denote the mean distance between electrons and the Bohr radius, respectively.

In conclusion, the possibility of quartet Fermi superfluidity and superconductivity have been investigated on a model interaction of BCS type.

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Note added:

After the present paper had been accepted for publication, we became aware of the previous works which discussed the physics similar to the present one. Transition temperature of the quartet condensation in nuclear matter has been discussed in G. Röpke, A. Schnell, P. Schuck and P. Nozières: Phys. Rev. Lett. 80 (1998) 3177. Model system in the large spin limit has been analyzed in A. S. Stepanenko and J. M. F. Gunn: cond-mat/9901317. The problem of one-dimensional models has also been discussed in P. Schlotmann: J. Phys.: Condens. Matter 6 (1994) 1359; and C. J. Wu: cond-mat/0409247. We thank Congjun Wu who directed our attention to those works.