Theory of Pairing Assisted Spin Polarization in Spin-Triplet Equal Spin Pairing: Origin of Extra Magnetization in Sr$_2$RuO$_4$ in Superconducting State

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It is shown that an extra magnetization is induced by an onset of the equal-spin-pairing of spin triplet superconductivity if the energy dependence of the density of states of quasi-particles exists in the normal state. It turns out that the effect is observable in Sr$_2$RuO$_4$ due to the existence of van Hove singularity in the density of states near the Fermi level, explaining the extra contribution in the Knight shift reported by Ishida et al. It is also quite non-trivial that this effect exists even without external magnetic field, which implies that the time reversal symmetry is spontaneously broken in the spin space.

Properties of the Fermi superfluidity sustained by the triplet pairing have been discussed extensively since the discovery of superfluid $^3$He in 1972, and its fundamental aspects seem to have been clarified so far. On the other hand, it has recently been measured by the Knight shift that the magnetization of Sr$_2$RuO$_4$, which is considered to be a triplet superconductor in the equal-spin-pairing (ESP) state, exhibits an extra magnetization under the external magnetic field other than that expected in the ESP state. This phenomenon cannot be understood in the framework of spin-singlet pairing state, while some researchers doubt the spin-triplet state because the first-order superconducting transition has been observed under the magnetic field which is characteristic of the paramagnetic effect in the spin-singlet pairing state. In this sense, it is desired to give a theoretical explanation for this phenomenon reported by Ishida. In this Letter we discuss theoretically the mechanism for such an extra magnetization in Sr$_2$RuO$_4$, which is considered to be in the spin-triplet ESP superconducting state, under the external magnetic field. This gives an explanation for the recent observation of extra magnetization in Sr$_2$RuO$_4$.

A physical reason for this extra contribution is rather simple. Under the magnetic field, the density of states (DOS) of the normal state quasiparticles of up-spin, $N_\uparrow(\xi)$, and those of down-spin, $N_\downarrow(\xi)$, are different if the particle-hole symmetry is apparently broken, i.e., $N(\xi)$'s are not constant but have a considerable linear term in the quasiparticle energy $\xi$ measured from the chemical potential. Then, the free energy gains associated with Cooper pair condensation are different in general, resulting in a redistribution of up-spin and down-spin components so as to gain much more condensation energy. Therefore, depending on the...
sign of the linear term of $N(\xi)$, the extra magnetization change arises under the external magnetic field $H$. This mechanism was first predicted almost four decades ago by S. Takagi as a possible effect of discontinuity in the spin susceptibility of superfluid $^3$He at the critical temperature $T_c$ where $^3$He exhibits a second-order phase transition from the normal to the A phase at $H = 0$. The paper by Takagi also predicted that in the A1 phase there exists an extra spin-polarization independent of $H$ other than the BCS-type contribution.

On the other hand, Takagi’s theory predicted that the extra magnetization quickly fades away in the A (or A2) phase where both up- and down-spin components are forming the Cooper pairs. This is because Takagi’s theory did not take into account the redistribution of fermions with up- and down-spin components in the SC state, while it took into account the migration of fermions in the normal down-spin band to the up-spin ESP state in the A1 phase. Here, we reconsider Takagi’s discussion and extend it to the ground state under the magnetic field $H$.

To begin with, we assume that a $\xi$ dependence of the DOS $N(\xi)$ without the magnetic field $H$ are given by

$$N(\xi) \simeq N_F + A\xi.$$  \hspace{1cm} (1)

Then, the DOS of up spin, $N_\uparrow(\xi)$, and down-spin, $N_\downarrow(\xi)$, under the field $H$ are shifted as shown in Fig. 1. Here, we neglect the shift in the chemical potential of the order of $\mathcal{O}(\mu_B H/\epsilon_F^*)^2$, $\epsilon_F^*$ being the effective Fermi energy of the quasiparticles.

**Ground State**

First, we discuss the case of ground state. Let us define the difference of condensation energy for majority down-spin and minority up-spin states in the ground state of EPS pairing as

$$\delta E_{\text{cond}} = \left[-\frac{1}{2} N_{F\downarrow} \Delta_\downarrow^2 - \left(\frac{1}{2} N_{F\uparrow} \Delta_\uparrow^2\right)\right] \times \frac{1}{2},$$  \hspace{1cm} (2)

where $N_{F\downarrow} \equiv N_F + A\mu_B H$ and $N_{F\uparrow} \equiv N_F - A\mu_B H$, and $\Delta_\downarrow$ and $\Delta_\uparrow$ are the superconducting gap of down-spin and up-spin components, respectively. With the use of the weak-coupling expression for the superconducting (SC) gap $\Delta$’s, $\Delta = \epsilon_F^* \exp(-1/VN_F)$, and eq. (1) for the DOS’s, $\delta E_{\text{cond}}$ is expressed as

$$\delta E_{\text{cond}} = -\frac{1}{4}(\epsilon_F^*)^2 \left\{ (N_F + A\mu_B H) \exp\left[-\frac{2}{V(N_F + A\mu_B H)}\right] - (N_F - A\mu_B H) \exp\left[-\frac{2}{V(N_F - A\mu_B H)}\right] \right\},$$  \hspace{1cm} (3)

where we have substituted relations $N_{F\downarrow} = N_F + A\mu_B H$ and $N_{F\uparrow} = N_F - A\mu_B H$. Then, the derivative $\partial \delta E_{\text{cond}} / \partial H$ at $H = 0$ is given as

$$\left(\frac{\partial \delta E_{\text{cond}}}{\partial H}\right)_{H=0} = -\frac{N_F}{2} \Delta^2 A\mu_B \frac{N_F}{N_F} \left(1 + \frac{2}{VN_F}\right).$$  \hspace{1cm} (4)
Fig. 1. Density of states $N(\xi)$ vs energy $\xi$ of quasiparticles measure from the Fermi level. Line passing $\xi = 0$, $\mu_B H$, and $-\mu_B H$ are DOS without magnetic field $H$, for up-spin band, and down-spin band, respectively. Full (dashed) lines indicate the state is occupied (unoccupied). Chemical potential shift due to the magnetic field is neglected as a negligible effect of the order of $O[(\mu_B H/\epsilon_F)^2]$.

Therefore, up to the linear term in $H$, the $\delta E_{\text{cond}}$ is given as

$$\delta E_{\text{cond}} \simeq -\frac{N_F}{2} \Delta \frac{2 A \mu_B}{N_F} \left(1 + \frac{2}{V N_F}\right) H.$$

If $A > 0$ as shown in Fig. 1, $\delta E_{\text{cond}} < 0$, which implies that the $\downarrow$-spin pairs have much lower energy than the $\uparrow$-spin ones. This calculation has been performed on the constraint that the distribution of $\downarrow$-spin and $\uparrow$-spin electrons number is fixed as the same as in the normal state. However, if this constraint were relaxed, electrons forming Cooper pairs should have migrated from $\uparrow$-spin to $\downarrow$-spin band to gain more condensation energy, giving rise to an extra magnetization.

In order to estimate this extra magnetization, we first consider the case without external magnetic field. The estimation leading to eq. (5) is valid also in this case where magnetization $\delta m$ increases virtually (associated with migration of Cooper pairs from $\uparrow$-spin to $\downarrow$-spin band), if $H$ in eq. (5) is replaced by $\delta m/\chi$, with $\chi$ being the magnetic susceptibility in the normal state. Namely, if $A > 0$ as shown in Fig. 1, the virtual magnetization $\delta m$ causes energy gain given by eq. (5) with $H$ replaced by $\delta m/\chi$. On the other hand, the virtual magnetization $\delta m$ is accompanied by energy cost corresponding to the magnetic energy $(\delta m)^2/2\chi$. Then, the
total energy change $\Delta E(\delta m)$, due to this virtual magnetization $\delta m$, is given as

$$\Delta E(\delta m) \simeq -\frac{N_F}{2} \Delta^2 \frac{A\mu_B}{N_F} \left(1 + \frac{2}{V N_F}\right) \frac{\delta m}{\chi} + \frac{(\delta m)^2}{2\chi}.$$  (6)

By minimizing this with respect to $\delta m$, we obtain a spontaneous magnetization $\delta m$ as

$$\delta m \simeq \frac{N_F}{2} \Delta^2 \frac{A\mu_B}{N_F} \left(1 + \frac{2}{V N_F}\right).$$  (7)

Namely, the time reversal symmetry is spontaneously broken even without the magnetic field. Of course, negative magnetization $\delta m$ given by eq. (7) with negative sign is also possible, without the external magnetic field, as in the case of Ising-like ferromagnetic order. In any case, these spontaneously induced magnetizations are caused by the migration of Cooper pairs among opposite spin components to gain the condensation energy.

This induced extra magnetization exists also under the magnetic field $H$. In this case, the sign of $\delta m$ is positive, if $A > 0$ as in Fig. 1. Indeed, the total energy $E(m + \delta m)$, where $m$ is the magnetization in the conventional ESP state under the magnetic field as discussed below and $\delta m$ is the deviation from the conventional one owing to the effect of migration of Cooper pairs, is given as

$$E(m + \delta m) = E(m) - \frac{N_F}{2} \Delta^2 \frac{A\mu_B}{N_F} \left(1 + \frac{2}{V N_F}\right) \frac{\delta m}{\chi} + \left[\frac{(m + \delta m)^2}{2\chi} - \frac{m^2}{2\chi}\right] - \delta m H,$$  (8)

where the first term represents the “conventional” condensation energy under the magnetic field $H$ giving the magnetization $m$, the second term the energy gain due to the migration of Cooper pairs causing the change $m \rightarrow m + \delta m$, the third term the energy loss due to the excess spin polarization, and the last term the excess Zeeman energy under the magnetic field $H$. The explicit form of the first term $E(m)$ of r.h.s. in eq. (8) is given by eq. (10) as shown below, in which the effect of the magnetic field is taken into account only through the difference of the DOS of majority and minority bands in the normal state. The form of the second term of r.h.s. in eq. (8) is derived from the expression eq. (5) for the expression of the energy gain by replacing $H$ by a ”magnetic field” $\delta m/\chi$ corresponding to the excess magnetization $\delta m$. By minimizing $E(m + \delta m)$, eq. (8), with respect to $\delta m$, we easily arrive at the relation (7), considering that the relation $m = \chi H$ holds in the conventional ESP state, except for a small correction given by eq. (11) as shown below. The latter correction is of the order of $O[(\Delta/\epsilon_F^a)^2]$ which gives only a negligibly small correction to $\delta m$, eq. (7), of the relative order of $O(\mu_B H/\epsilon_F^a) \ll 1$.

The size of coefficient $A$ in eq. (1) is parameterized as $A = N_F(a/\epsilon_F^a)$, where $a \sim O(1)$ parameterizes steepness of the slope of $N(\xi)$ around $\xi = 0$. The magnetization $m_n$ in the normal state under the magnetic field $H$ is given by $m_n \simeq 2\mu_B^2 N_F H/(1 + F_0^a)$, $F_0^a$ being the Fermi liquid parameter for the correction of the magnetic susceptibility. Therefore, the ratio

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of $\delta m$ and $m_n$ is given by

$$
\frac{\delta m}{m_n} = \frac{1}{4} \frac{a \Delta^2}{\mu_B H \epsilon_F^*} \left( 1 + \frac{2}{V N_F} \right) (1 + F_0^a).
$$

(9)

There exists other “conventional contribution” to the magnetization through the $H$ dependence of the condensation energy $E_{\text{cond}}$ in the ground state as discussed in ref. 6 in some different context. Here, “conventional contribution” implies that obtained without migration of Cooper pairs of down- and up-spin components. Indeed, the $E_{\text{cond}}$ is given by

$$
E_{\text{cond}} = -\frac{1}{4} (\epsilon_c^*)^2 \left\{ (N_F + A \mu_B H) \exp \left[ -\frac{2}{V(N_F + A \mu_B H)} \right] 
+ (N_F - A \mu_B H) \exp \left[ -\frac{2}{V(N_F - A \mu_B H)} \right] \right\}.
$$

(10)

Then, the magnetization $m_s \equiv -\langle \partial E_{\text{cond}} / \partial H \rangle$ (at $H \neq 0$) is calculated as

$$
m_s = \frac{N_F \Delta^2 A \mu_B}{2 V N_F^2} \frac{4 A \mu_B H}{N_F},
$$

(11)

where the terms of the order of $O[(A \mu_B H/N_F)^2]$ have been discarded. This $m_s$ is smaller than $\delta m$, eq. (7), by a small factor $A \mu_B H/N_F = a \mu_B H/\epsilon_F^* \ll 1$. Therefore, the “conventional contribution”, eq. (11), can be safely neglected.

**GL Region**

Next, we discuss the case in GL region, in which we estimate the free energy gain $\delta F$ due to SC condensation in stead of the ground state energy at $T = 0$ K. In the GL region, the free energy difference $\delta F_{\text{cond}} \equiv F_{\text{cond}}^{(+)} - F_{\text{cond}}^{(-)}$ is given as follows:

$$
\delta F_{\text{cond}} = -\frac{K}{4} \left[ (N_F + A \mu_B H) (T_c^{(+)} - T)^2 - (N_F - A \mu_B H) (T_c^{(-)} - T)^2 \right],
$$

(12)

where the SC transition temperatures are given by $T_c^{(\pm)} = \epsilon_c^* \exp[-1/V(N_F \pm A \mu_B H)]$, and $K \equiv 8\pi^2/7\zeta(3) \simeq 9.38$, with $\zeta(x)$ being the Riemann $\zeta$ function. By calculations similar to the case $T = 0$ K, corresponding to eq. (5), we obtain

$$
\delta F_{\text{cond}} \simeq -\frac{K}{2} \frac{A \mu_B}{N_F} \frac{1}{V N_F} \left( T_c - T \right)^2 + \frac{2 T_c(T_c - T)}{V N_F} H.
$$

(13)

In the GL region, $T \simeq T_c$, the first term in the bracket is neglected compared to the second term. Then, corresponding to eq. (7), the extra magnetization $\delta m$ is given as

$$
\delta m \simeq K \frac{A \mu_B}{N_F} \frac{1}{V N_F} T_c(T_c - T).
$$

(14)

Therefore, corresponding to eq. (9), we obtain the ratio of $\delta m$ and $m_n$ as

$$
\frac{\delta m}{m_n} = \frac{8\pi^2}{7\zeta(3)} \frac{a T_c(T_c - T)}{\mu_B H \epsilon_F^*} \frac{1}{V N_F} (1 + F_0^a).
$$

(15)

The result (14) is consistent with that for the extra magnetization in the A1 phase, eq.
predicted in Takagi’s paper,\(^5\)

\[ M_1 - M_n = N_F T_c \mu_B \eta (t + \eta h) / 2\beta, \] (16)

considering that correspondence of parameters between Takagi’s paper and ours is as follows: 
\[ t = (T_c - T) / T_c, \eta = AT_c / V(N_F)^2, h = \mu_B H / T_c, \] and that our theory has not taken into account the feedback effect; i.e., \((1/\beta) = K\). A difference in overall factor by 2 can be understood from the fact that Takagi’s eq. (4) is for near the A1 transition associated with only up-spin pairing while our result eq. (14) is for both up- and down-spin pairings. The reason why the extra magnetization which is independent of the external magnetic field \(H\) is missing in the A2 phase in Takagi’s expression, eq. (5), seems to be traced back to the fact that he has not taken into account the migration of electrons from down-spin \(^3\)He nuclei in the normal state to the up-spin Cooper pairs in the A1 phase.

The “conventional contribution” to the magnetization through the \(H\) dependence of the free energy \(F_{\text{cond}}\) in GL region is calculated similarly to the case in the ground state. The \(F_{\text{cond}}\) is given as

\[ F_{\text{cond}} = -K \frac{4}{4} \left[ (N_F + A\mu_B H) \left( T_c^+ - T \right)^2 + (N_F - A\mu_B H) \left( T_c^- - T \right)^2 \right], \] (17)

Then, the magnetization \(m_s \equiv - (\partial F_{\text{cond}} / \partial H) \) (at \(H \neq 0\)) is calculated as

\[ m_s \simeq K N_F A\mu_B \frac{1}{N_F} \left[ 2T_c (T_c - T) + T T_c \right] \frac{A\mu_B H}{N_F}, \] (18)

where the terms of the order of \(O(A\mu_B H / N_F)^2\) have been discarded as in the case of ground state above. This \(m_s\) is smaller than \(\delta m\), eq. (14), by a small factor \(A\mu_B H / N_F = a \mu_B H / \epsilon_F^* \ll 1\). Therefore, the “conventional contribution”, eq. (18), can be safely neglected again.

It is remarked that the expression (18) is exactly the same as eq. (5) in Takagi’s paper for the A2 phase to the zeroth order in \((T - T_c)\):\(^5\)

\[ M_{II} - M_n = N_F T_c \mu_B \eta^2 h / \beta, \] (19)

considering again that correspondence of parameters between Takagi’s paper and ours is as follows: 
\[ t = (T_c - T) / T_c, \eta = AT_c / V(N_F)^2, h = \mu_B H / T_c, \] and that our theory has not taken into account the so-called feedback effect due to spin fluctuations; i.e., \((1/\beta) = K\) and \(\delta = 0\).

**Order Estimation**

Here we give a rough order estimation for \(\delta m / m_n\) in Sr\(_2\)RuO\(_4\). With the use of the correlation length at \(T = 0\) K, \(\xi_0 \simeq 1050\) Å,\(^7\) the effective Fermi energy of the quasiparticles \(\epsilon_F^*\) is estimated as

\[ \epsilon_F^* \simeq 2.5 \times 10^3 T_c \simeq 3.8 \times 10^3\) K. \] (20)
Assuming $\tilde{\epsilon}_c \sim \epsilon_F^*$, the coupling constant $VN_F$ is estimated as
\[
\frac{1}{VN_F} \simeq 7. 
\]
(21)
The SC gap at $T = 0$ K is estimated by using the BCS relation:
\[
\Delta \simeq 1.7 \times T_c \simeq 2.6 \text{ K}. 
\]
(22)
The Landau parameter $F_0^a$ is estimated from the Wilson ratio as $F_0^a \simeq -0.5.8$

The magnetic field $H \simeq 1$ T, used in the NMR Knight shift measurements, is equivalent to $H\mu_B \simeq 0.67$ K. Then, the ratio $\delta m/m_n$, eq. (9), at $T = 0$ K is estimated as
\[
\frac{\delta m}{m_n} \simeq 5.0 \times 10^{-3} \times a. 
\]
(23)
Since there exists the van Hove singularity in the DOS of the $\gamma$ band just above the Fermi level, the parameter $a$, parameterizing the steepness of the slope in DOS at the Fermi level, can be much larger than 1/2, the value for free fermions. Indeed, according to Fig. 41 for the DOS of $\gamma$ band in ref. 9, and considering $m^*/m_{\text{band}} \simeq 5.5,10$ the parameter $a$ is estimated as $a \simeq 3.6$. The effect of the $\alpha$ and $\beta$ bands may give some additional contribution. However, since the DOS of the $\gamma$ band dominates those of $\alpha$ and $\beta$ bands, the effect is expected to be limited. Thus, the ratio $\delta m/m_n$ at $T = 0$ K can be a few % in consistent with the Knight shift measurements reported in ref. 3, while the above estimations are rather crude.

**Discussions**

It is noted that the excess magnetization given by eqs. (7) and (14) exists without external magnetic field. This implies that such a magnetization gives a spontaneous magnetic field breaking time reversal symmetry. It is crucial that this effect is not related to the orbital effect of degenerate component of the Cooper pairs, such as $(\sin k_x + i \sin k_y)$ state.$^{11}$ The size of this magnetic field is roughly estimated as follows: By using the relation $N_F = 3N/4\epsilon_F^*$ for a free dispersion, eq. (7) is reduced to
\[
\delta m = \frac{3}{8} \left( \frac{\Delta}{\epsilon_F^*} \right)^2 a \left( 1 + \frac{2}{VN_F} \right) N\mu_B. 
\]
(24)
By assuming that there exists one electron per unit cell ($a = b = 3.9 \times 10^{-10}$ m, and $c = (12.7/2) \times 10^{-10}$ m), the number of electrons $N$ per unit volume is estimated as $N \simeq 1.04 \times 10^{28}$. Then, using the values, eqs. (20), (21), and (22), $\delta m$ at $T = 0$ is estimated as
\[
\delta m \simeq 0.92 \text{ J} \cdot \text{T}^{-1}. 
\]
(25)
This corresponds to the magnetic field $\delta B$ as
\[
\delta B = \mu_0 \delta m \simeq 1.1 \times 10^{-6} \text{ T} = 1.1 \times 10^{-2} \text{ G}, 
\]
(26)
where $\mu_0 = 4\pi \times 10^{-7}$ H·m$^{-1}$ is the magnetic permeability of vacuum. This is far smaller than the lower critical field $H_{cl}^{ab} = 10$ G and $H_{cl}^c = 50$ G,$^{12}$ so that it would be fully screened.
out by the Meissner effect. Therefore, it seems technically impossible to observe this small spontaneous magnetic field if the domain size is larger than the penetration depth of magnetic field.

It is interesting that the effect similar to that observed in \( \text{Sr}_2\text{RuO}_4 \) seems to have been observed in \( \text{UPt}_3 \) although the effect is smaller than that in \( \text{Sr}_2\text{RuO}_4 \) by one order of magnitude.\(^{13}\) It is also interesting that an upper bound of spontaneous magnetic filed of the order of 1 mG, one order smaller than a value given by eq. (25), was reported in \( \text{UPt}_3 \) on a measurement by using a SQUID magnetometer.\(^{14}\) This is consistent with the fact that \( \mu \text{SR} \) measurement of high quality single crystal has given estimations of upper bound of the spontaneous magnetization as \( \sim 30 \text{mG}^{15} \) or \( \sim 80 \text{mG}^{16} \).

The pairing assisted spin polarization should exist also in the A-phase of superfluid \( ^3\text{He} \). Indeed, \( \delta m/m_n \), eq. (9), is estimated under a hypothetical situation, i.e., \( T = 0 \text{ K} \) and \( H = 1 \text{ Tesla} \), as follows: With the use of a parameter set for \( ^3\text{He} \) at \( p = 27 \text{ bar} \) (\( e^*_{F} \simeq 1.09 \text{ K} \), \( \Delta = 1.7 T_c \simeq 4.3 \text{ mK} \), \( \mu_N \simeq 1.1 \times 10^{-26} \text{ J/T} \), \( 1/VN_F \simeq 6 \), \( F^a_0 \simeq -0.755 \) and \( a = 1/2 \)),\(^{17}\) the ratio \( \delta m/m_n \) is estimated as

\[
\frac{\delta m}{m_n} \simeq 7.7 \times 10^{-3}. \tag{27}
\]

Thus, the extra magnetization in the A-phase of superfluid \( ^3\text{He} \) is nearly the same order as that expected in \( \text{Sr}_2\text{RuO}_4 \).

\textit{Conclusion}

It has been shown that the extra magnetization (or spin polarization) is induced in the ESP state due to the migration of the Cooper pairs from minority to majority pairing state to gain the condensation energy (free energy). This effect seems to have been overlooked for four decades, and to give a semi-quantitative explanation for the effect which was discovered quite recently by the Knight shift measurements in \( \text{Sr}_2\text{RuO}_4 \) by Ishida and coworkers. This extra magnetization is induced spontaneously even without the external magnetic field.

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