# **Two types of odd-frequency superconductivity:** odd-frequency-diamagnetic vs. odd-frequency-paramagnetic superconductivity

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**25 December 2014** 

# Superconductivity

$$\begin{split} G &\sim \left\langle \psi \psi^{\dagger} \right\rangle &\quad - \text{ ordinary Green function} \\ F &\sim \left\langle \psi \psi \right\rangle &\quad - \text{ anomalous Green function (wave function of Cooper pairs)} \\ F^{+} &\sim \left\langle \psi^{\dagger} \psi^{\dagger} \right\rangle \\ \hat{G} &= \begin{pmatrix} G & F \\ F^{+} & G' \end{pmatrix} - \text{Nambu space} \end{split}$$

Superconductivity = non-diagonal elements in the Nambu space (  $\hat{\tau}_1, \hat{\tau}_2$  components)

Simplest case: singlet and s-wave (isotropic)

More complicated: triplet and/or anisotropic (p-wave, d-wave, etc.)

# **Odd-frequency superconductivity**

F(1; 2) – anomalous Green function (Matsubara technique)

F(2; 1) = -F(1; 2) – fermionic antisymmetry (the Pauli principle)

Standard classification of superconducting phases: even coordinate dependence (s-wave, d-wave, etc.), odd spin dependence (singlet:  $|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$ ) OR odd coordinate dependence (p-wave, etc.), even spin dependence (triplet:  $|\uparrow\uparrow\rangle$ ,  $|\downarrow\downarrow\rangle$ ,  $|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$ )

Berezinskii (1974):

if the imaginary time dependence is odd, then there is a possibility of even coordinate dependence and even spin dependence

 $\tau = \tau_1 - \tau_2 \quad \rightarrow \quad \omega$ 

Odd-  $\omega$  superconductivity! (Example: odd- $\omega$ , s-wave, spin-triplet)

# **Odd-** $\omega$ **superconductivity**

• Berezinskii (1974) [JETP Lett. **20**, 287]: Possibility of odd- $\omega$  pairing in <sup>3</sup>He due to retarded paramagnon exchange. Not realized.

• Abrahams, Balatsky, Scalapino, Schrieffer (1995) [PRB **52**, 1271]: Studies of bulk properties of hypothetical Berezinskii superconductor.

• Bergeret, Volkov, Efetov (2001) [review – RMP **77**, 1321 (2005)]: Generation of the Berezinskii component in proximity structures conventional superconductor / ferromagnet.



• Tanaka, Golubov (2007) [PRL **98**, 037003]: Generation of the Berezinskii component in proximity structures triplet superconductor / diffusive normal metal.

# $\textbf{Odd-}\omega \textbf{ superconductivity: dia and para}$

• Negative superfluid density ("paramagnetic" Meissner effect):

$$\mathbf{j}_S(q) = -\frac{e^2 n_S}{mc} \mathbf{A}(q), \qquad n_S < 0$$

Ok in inhomogeneous proximity systems (except for some special thin-film structures). But instability in the bulk...

#### Recent proposals:

- Solenov, Martin, Mozyrsky, Phys. Rev. B 79, 132502 (2009);
- Kusunose, Fuseya, Miyake, J. Phys. Soc. Jpn. 80, 054702 (2011); 044711 (2011);
- Matsumoto, Koga, Kusunose, J. Phys. Soc. Jpn. 81, 033702 (2012);
- Kusunose, Matsumoto, Koga, Phys. Rev. B 85, 174528 (2012);
- and more:

Model with a retarded interaction, mean-field analysis.

Possibility of a bulk odd- $\omega$ -dia phase with  $n_s > 0$  and regular diamagnetic Meissner effect in the case when  $\Delta$  is odd- $\omega$ .

## **General derivation**

$$\begin{aligned} \mathcal{Z} &= \int D\psi_{\uparrow}^* D\psi_{\downarrow}^* D\psi_{\uparrow} D\psi_{\downarrow} \exp(-\mathcal{S}_0 - \mathcal{S}_{\text{int}}) \\ \mathcal{S}_0 &= \int_1 \psi_{\alpha}^*(1) \left(\partial_{\tau_1} + \xi\right) \psi_{\alpha}(1) \\ \mathcal{S}_{\text{int}} &= \frac{1}{2} \int_1 \int_2 V_{\alpha\beta;\gamma\delta} (1-2) \psi_{\beta}^*(2) \psi_{\alpha}^*(1) \psi_{\gamma}(1) \psi_{\delta}(2) \end{aligned}$$

where 1 and 2 are 4-vectors:  $1 \equiv (\mathbf{r}_1, \tau_1), \quad 2 \equiv (\mathbf{r}_2, \tau_2)$ 

Interaction is even with respect to interchanging of two electrons:

$$V_{\alpha\beta;\gamma\delta}(1-2) = V_{\beta\alpha;\delta\gamma}(2-1)$$

Hubbard-Stratonovich transformation:

$$\exp(-\mathcal{S}_{int}) \times \int D\Delta^* D\Delta \exp\left\{\frac{1}{2} \int_1 \int_2 \left[V^{-1}(1-2)\right]_{\alpha\beta;\gamma\delta} \Delta^*_{\alpha\beta}(1,2) \Delta_{\gamma\delta}(1,2)\right\}$$
$$\mapsto \int D\Delta^* D\Delta \exp(-\mathcal{S}_{aux} - \mathcal{S}_{\Delta})$$
with  $\mathcal{S}_{aux} = -\frac{1}{2} \int_1 \int_2 \left[\Delta_{\alpha\beta}(1,2) \psi^*_{\beta}(2) \psi^*_{\alpha}(1) + \Delta^*_{\alpha\beta}(1,2) \psi_{\alpha}(1) \psi_{\beta}(2)\right]$  $\mathcal{S}_{\Delta} = -\frac{1}{2} \int_1 \int_2 \left[V^{-1}(1-2)\right]_{\alpha\beta;\gamma\delta} \Delta^*_{\alpha\beta}(1,2) \Delta_{\gamma\delta}(1,2)$ 

V – attraction, the integral must be convergent (so the sign is fixed)

## **Mean-field**

Mean-field: take a trial path for  $\Delta^*$  and  $\Delta$ , minimize the free energy

$$\mathcal{F}_{\rm MF}[\Delta^*,\Delta] = -T\ln\mathcal{Z}_{\rm MF} = -T\ln\int D\psi^*D\psi\exp\left(-\mathcal{S}_{\rm MF}[\psi^*,\psi,\Delta^*,\Delta]\right)$$

with respect to the trial path.

Define anomalous averages:

$$F_{\alpha\beta}(1,2) = \left\langle \psi_{\alpha}(1)\psi_{\beta}(2)\right\rangle_{\mathrm{MF}}$$
$$F_{\alpha\beta}^{+}(1,2) = \left\langle \psi_{\alpha}^{*}(1)\psi_{\beta}^{*}(2)\right\rangle_{\mathrm{MF}}$$

From definition:

$$F^*_{\alpha\beta}(\mathbf{q},\omega) = s_{\Delta}F^+_{\beta\alpha}(\mathbf{q},-\omega)$$

where  $s_{\Delta} = \pm 1$  for the even-/odd- $\omega$  dependence of  $\Delta$ 

$$F^*_{\alpha\beta}(\mathbf{q},\omega) = +F^+_{\beta\alpha}(\mathbf{q},-\omega) - \text{induced odd-}\omega\text{---para state}$$
$$F^*_{\alpha\beta}(\mathbf{q},\omega) = -F^+_{\beta\alpha}(\mathbf{q},-\omega) - \text{principal odd-}\omega\text{---dia state}$$

Induced odd- $\omega$ —para state: frequency symmetries of *F* and  $\Delta$  are different (ok since the odd- $\omega$  component drops out from the self-consistency equation.

## **General homogeneous solution**

Self-consistency equation: 
$$\Delta_{\alpha\beta}(k) = -\int (dk') V_{\alpha\beta;\gamma\delta}(k-k') F_{\gamma\delta}(k')$$
  
Components:  $\Delta_{\alpha\beta}(k) = d_0(k)(i\sigma_2)_{\alpha\beta} + \mathbf{d}(k)(i\sigma\sigma_2)_{\alpha\beta}$   
 $G_{\alpha\beta}(k) = G_0(k)\delta_{\alpha\beta} + \mathbf{G}(k)\sigma_{\alpha\beta}$   
 $F_{\alpha\beta}(k) = F_0(k)(i\sigma_2)_{\alpha\beta} + \mathbf{F}(k)(i\sigma\sigma_2)_{\alpha\beta}$   
where  $G_0(k) = -\frac{(i\omega + \xi_k)[\omega^2 + \xi_k^2 + D_0(k)]}{[\omega^2 + E_+^2(k)]}$  with  
 $\mathbf{G}(k) = \frac{(i\omega + \xi_k)\mathbf{D}(k)}{[\omega^2 + E_+^2(k)]}$   $\omega^2 + E_-^2(k)]$   
 $F_0(k) = \frac{(\omega^2 + \xi_k^2)\mathbf{d}_0(k) + [d_0^2(k) - \mathbf{d}^2(k)]d_0^*(k)}{[\omega^2 + E_+^2(k)]}$   $\mathbf{F}(k) = \frac{(\omega^2 + \xi_k^2)\mathbf{d}(k) - [d_0^2(k) - \mathbf{d}^2(k)]\mathbf{d}^*(k)}{[\omega^2 + E_+^2(k)]}$   $D(k) = d_0(k)\mathbf{d}^*(k) + d_0^*(k)\mathbf{d}(k)$   
 $F_0(k) = \frac{(\omega^2 + \xi_k^2)\mathbf{d}(k) - [d_0^2(k) - \mathbf{d}^2(k)]\mathbf{d}^*(k)}{[\omega^2 + E_-^2(k)]}$   $D(k) = \sqrt{\mathbf{D}^2(k)}$ 

### Simplified examples:

- singlet state,  $\mathbf{d} = 0$
- triplet state,  $d_0 = 0$
- unitary triplet state,  $d_0 = 0$ ,  $[\mathbf{d} \times \mathbf{d}^*] = 0$

# Superconducting components induced due to spatial inhomogeneity

A single superconducting component. Then Eilenberger equations:  $v_F \hat{k} \cdot \nabla g = 2\Delta f_S$ 

$$v_F \hat{k} \cdot \nabla f_B = -2\omega_n f_S$$
  
 $v_F \hat{k} \cdot \nabla f_S = 2(\Delta g - \omega_n f_B)$ 



 $s_p$  - (spatial) parity of the superconducting state

 $f_{\rm B}$  has the same symmetries as  $\Delta$ .  $f_{\rm S}$  – opposite frequency symmetry and parity with respect to  $\Delta$ .

# Classification

	Frequency	Spin	Parity	Magnetic response	
ix due to	(a) Even	Singlet	Even	Dia	Bulk
	Odd	Singlet	Odd	Para	Induced
in-symm. –	(b) Even	Triplet	Odd	Dia	Bulk
eaking	Odd	Triplet	Even	Para	Induced
x due to	(c) Odd	Singlet	Odd	Dia	Bulk
	Even	Singlet	Even	Para	Induced
pin-symm. –	(d) Odd	Triplet	Even	Dia	Bulk
reaking	Even	Triplet	Odd	Para	Induced

Asano, Fominov, Tanaka (2014) [PRB 90, 094512]

The classification of Cooper pairs in inhomogeneous superconductors. In the absence of spin-dependent potentials, the spin state in the bulk and near an inhomogeneity is the same. At the same time, due to broken translational invariance, the spatial parity can change. This leads to changing of the frequency symmetry, in order to conform with the Pauli principle. ESED states realized in metallic superconductors and high- $T_c$  cuprates in (a) have OSOP states as the subdominant component. ETOD states realized in  $Sr_2RuO_4$  and  $UPt_3$  in (b) have OTEP states as the subdominant component. OSOD states in (c) and OTED states in (d) have never been confirmed in real materials. The subdominant component of OSOD states in (c) is ESEP states. ETOP states appears as a subdominant component of OTED states in (d).

## **Odd-** $\omega$ **--dia and --para: problem of coexistence**

Coexistence of dia and para states:  $G_{\alpha\beta} = (G_d)_{\alpha\beta} + (G_p)_{\alpha\beta}$  $F_{\alpha\beta} = (F_d)_{\alpha\beta} + (F_p)_{\alpha\beta}$  $F_{\alpha\beta}^+ = (F_d^+)_{\alpha\beta} + (F_p^+)_{\alpha\beta}$ 

Unitary pairing: 
$$G_{0d} = -\frac{i\omega + \xi}{\omega^2 + \xi^2 + D_{0d}}, \quad G_{0p} = -\frac{i\omega + \xi}{\omega^2 + \xi^2 - D_{0p}}$$
  
 $\mathbf{F}_d = \frac{\mathbf{d}_d}{\omega^2 + \xi^2 + D_{0d}}, \quad \mathbf{F}_p = \frac{\mathbf{d}_p}{\omega^2 + \xi^2 - D_{0p}}$ 

Cross-term in the superfluid density:

$$\frac{\delta n_S}{n} = \pi T \sum_{\omega} \frac{1}{D_{0d} + D_{0p}} \left[ \frac{4\omega^2 + 2D_{0d} + \mathbf{d}_d \mathbf{d}_p^* - \mathbf{d}_d^* \mathbf{d}_p}{\sqrt{\omega^2 + D_{0d}}} - \frac{4\omega^2 - 2D_{0p} + \mathbf{d}_d \mathbf{d}_p^* - \mathbf{d}_d^* \mathbf{d}_p}{\sqrt{\omega^2 - D_{0p}}} \right]$$

#### Complex!!!

## **Odd-\omega-dia and** -**para: Josephson junction**

#### Tunneling action (the case of unitary pairing):

$$S_T = \frac{\pi G}{2} \sum_{\omega} \frac{-\omega^2 + \tilde{\mathbf{d}}_L \tilde{\mathbf{d}}_R (s_L e^{i\varphi} + s_R e^{-i\varphi})/2}{\sqrt{\omega^2 + s_L \tilde{\mathbf{d}}_L^2} \sqrt{\omega^2 + s_R \tilde{\mathbf{d}}_R^2}}$$
$$\mathbf{d}_{L(R)} = \tilde{\mathbf{d}}_{L(R)} e^{i\varphi_{L(R)}}, \quad \varphi = \varphi_R - \varphi_L$$

Phase-dependent part:  

$$\frac{s_L e^{i\varphi} + s_R e^{-i\varphi}}{2} = \begin{cases} -\cos\varphi, & \text{odd-}\omega\text{-para/para} \\ \cos\varphi, & \text{odd-}\omega\text{-dia/dia} \\ -i\sin\varphi, & \text{odd-}\omega\text{-para/dia} \end{cases}$$

 $s_{L(R)} = \pm 1$  for odd- $\omega$ -dia/-para

#### Odd- $\omega$ -para/dia junction – imaginary Josephson current!!!

## **Can odd-ω-dia state really exist?**

The Green functions in the Hamiltonian formulation:

$$F_{\alpha\beta}(\mathbf{r}_1, \mathbf{r}_2; \tau_1, \tau_2) = \left\langle T_\tau \hat{\psi}_\alpha(\mathbf{r}_1, \tau_1) \hat{\psi}_\beta(\mathbf{r}_2, \tau_2) \right\rangle$$
$$F_{\alpha\beta}^+(\mathbf{r}_1, \mathbf{r}_2; \tau_1, \tau_2) = \left\langle T_\tau \hat{\psi}_\alpha^+(\mathbf{r}_1, \tau_1) \hat{\psi}_\beta^+(\mathbf{r}_2, \tau_2) \right\rangle$$

where  $\hat{\psi}_{\alpha}(\mathbf{r},\tau) = e^{\hat{H}\tau}\hat{\psi}_{\alpha}(\mathbf{r})e^{-\hat{H}\tau}, \quad \hat{\psi}^{+}_{\alpha}(\mathbf{r},\tau) = e^{\hat{H}\tau}\hat{\psi}^{\dagger}_{\alpha}(\mathbf{r})e^{-\hat{H}\tau}$ 

Immediate consequence:  $F^*_{\alpha\beta}(\mathbf{q},\omega) = +F^+_{\beta\alpha}(\mathbf{q},-\omega)$ 

if Hamiltonian description exists at any level (in particular, initial many-body  $\hat{H}$ )

Hence: odd- $\omega$  state must be paramagnetic.

## **Can odd-ω-dia state really exist?**

So, what is wrong in the Lagrangian description? How could it lead to the opposite sign?

#### **Our guess:**

No spontaneous gauge symmetry breaking (that could fix the superconducting phase), due to absence of the corresponding physical perturbation [note that in the conventional case we could write the hermitian perturbation  $\int d\mathbf{r} [\delta_0 \hat{\psi}^{\dagger}_{\uparrow}(\mathbf{r}) \hat{\psi}^{\dagger}_{\downarrow}(\mathbf{r}) + \delta_0^* \hat{\psi}_{\downarrow}(\mathbf{r}) \hat{\psi}_{\uparrow}(\mathbf{r})]$  with  $|\delta_0| \to 0$ ].

Then no mean-field description.

Phase is not fixed, integration over phase, anomalous averages vanish.

# Conclusion

### Summary:

- Two closed superconducting classes, containing odd- $\omega$ -para or odd- $\omega$ -dia states.
- Everything is consistent within each class.
- Unphysical results if their coexistence is assumed.

Our conclusion:

• No spontaneous symmetry breaking for the superconducting class containing odd- $\omega$ -dia states, so that the anomalous averages vanish.