# Odd-frequency superconductivity in Sr<sub>2</sub>RuO<sub>4</sub> measured by Kerr effect

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## Outline



- Introduction to odd-frequency (ω) superconductivity
   SF, SN junctions
- Odd- $\omega$  bulk superconductivity in multiband systems
  - Simple two-band superconductors
- Odd- $\omega$  superconductivity and Kerr effect in Sr<sub>2</sub>RuO<sub>4</sub>

## **Different Phases of Matter**



Ordered states  $\rightarrow$  symmetry of order parameter,  $\Delta$ 

Superconductivity:  $\Delta \sim F = \langle \psi_{\alpha} \psi_{\beta} \rangle$ 



## **Different Phases of Matter**



Ordered states  $\rightarrow$  symmetry of order parameter,  $\Delta$ 

Superconductivity:  $\Delta \sim F = \langle \psi_lpha(t)\psi_eta(0)
angle$  ( $t\leftrightarrow\omega$ , frequency)



## $\textbf{Odd-}\boldsymbol{\omega} \text{ Pairing}$



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BCS order parameter: 
$$F(\mathbf{r}, t; \mathbf{r}', t' \rightarrow t) = \langle \psi(\mathbf{r}, t)\psi(\mathbf{r}', t' \rightarrow t) \rangle$$

vanishes for odd- $\omega$  pairing





### Spin-singlet *s*-wave SC $\rightarrow$ odd- $\omega$ spin-triplet *s*-wave pairing

- Long-range superconducting proximity effect in F
- *s*-wave = disorder robust

[1]: Eschrig, Phys. Today 64, 43 (2011)



### Spin-singlet *s*-wave SC $\rightarrow$ odd- $\omega$ spin-singlet *p*-wave pairing

- Only high-transparency junctions
- *p*-wave = only ballistic systems

[1]: Tanaka et al, PRL 99, 037005 (2007)



## Odd-ω Bulk Superconductivity in Multiband Systems

- A clue from Bi<sub>2</sub>Se<sub>3</sub>
- Simple two-band superconductors



## Superconducting Symmetries in Bi<sub>2</sub>Se<sub>3</sub>



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Singlet/triplet, spatial (s/p/d), even/odd- $\omega$ , even/odd orbital

	Superconductor		Even	-frequency	Odd-frequency		
Г	Basis function	$J_z$	Even-orbital	Odd-orbital	Even-orbital	Odd-orbital	
$A_{1g}$	$\psi = 1$	0	$A_{1g}$ singlet,	-	-	$A_{1g}$ singlet,	
			$A_{2u}$ triplet (m <sub>s</sub> = ±1)			$A_{2u}$ triplet (m <sub>s</sub> = ±1)	
$B_{1g}$	$\psi = k_x^2 - k_y^2$	$\pm 2$	$B_{1g}$ singlet,	-	-	$B_{1g}$ singlet,	
			$B_{2u}$ triplet (m <sub>s</sub> = ±1)			$B_{2u}$ triplet (m <sub>s</sub> = ±1)	
$B_{2g}$	$\psi = 2k_xk_y$	$\pm 2$	$B_{2g}$ singlet,	-	-	$B_{2g}$ singlet,	
			$B_{1u}$ triplet (m <sub>s</sub> = ±1)			$B_{1u}$ triplet (m <sub>s</sub> = ±1)	
$A_{1u}$	$\mathbf{d} = (k_x, k_y, 0)$	0	$A_{1u}$ triplet (m <sub>s</sub> = ±1)	$A_{1g}$ triplet (m <sub>s</sub> = 0)	$A_{1g}$ triplet (m <sub>s</sub> = 0)	A <sub>1u</sub> triplet (m <sub>s</sub> = $\pm 1$ )	
$A_{2u}$	$\mathbf{d} = (k_y, -k_x, 0)$	0	$A_{2u}$ triplet (m <sub>s</sub> = ±1),	-	-	A <sub>2u</sub> triplet (m <sub>s</sub> = $\pm 1$ ),	
			$A_{1g}$ singlet			$A_{1g}$ singlet	
$B_{1u}$	$\mathbf{d} = (k_x, -k_y, 0)$	$\pm 2$	$B_{1u}$ triplet (m <sub>s</sub> = ±1),	${ m B_{1g}} { m triplet} ({ m m}_s=0)$	$B_{1g}$ triplet (m <sub>s</sub> = 0)	$B_{1u}$ triplet (m <sub>s</sub> = ±1),	
			$B_{2g}$ singlet			$B_{2g}$ singlet	
$B_{2u}$	$\mathbf{d} = (k_y, k_x, 0)$	$\pm 2$	$B_{2u}$ triplet (m <sub>s</sub> = ±1),	$B_{2g}$ triplet (m <sub>s</sub> = 0)	$B_{2g}$ triplet (m <sub>s</sub> = 0)	$B_{2u}$ triplet (m <sub>s</sub> = ±1),	
			$B_{1g}$ singlet			$B_{1g}$ singlet	
$E_{2u}^+$	$\mathbf{d} = (0, 0, k_x + ik_y)$	1	$E_{2u}^+$ triplet (m <sub>s</sub> = 0)	$A_{1g}$ triplet (m <sub>s</sub> = 1),	$A_{1g}$ triplet (m <sub>s</sub> = 1),	$E_{2u}^+$ triplet (m <sub>s</sub> = 0)	
				$B_{1g}+iB_{2g}$ triplet (m <sub>s</sub> = -1)	$B_{1g}+iB_{2g}$ triplet (m <sub>s</sub> = -1)		
$E_{2u}^-$	$\mathbf{d} = (0, 0, \overline{k_x - ik_y})$	-1	$E_{2u}^{-}$ triplet (m <sub>s</sub> = 0)	$A_{1g}$ triplet (m <sub>s</sub> = -1),	$A_{1g}$ triplet (m <sub>s</sub> = -1),	$E_{2u}^{-}$ triplet (m <sub>s</sub> = 0)	
				$B_{1g} - iB_{2g}$ triplet (m <sub>s</sub> = 1)	$B_{1g} - iB_{2g}$ triplet (m <sub>s</sub> = 1)		

ABS and Balatsky, PRB 87, 220506(R) (2013)

### **Frequency and Orbital**



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#### Complete reciprocity in oddness in frequency and orbital index

Superconductor			Ever	n-frequency	Odd-frequency		
Г	Basis function	$J_z$	Even-orbital	Odd-orbital	Even-orbital	Odd-orbital	
A <sub>1g</sub>	$\psi = 1$	0	$A_{1g}$ singlet,	-	-	$A_{1g}$ singlet,	
			$A_{2u}$ triplet (m <sub>s</sub> = ±1)			$A_{2u}$ triplet (m <sub>s</sub> = ±1)	
$B_{1g}$	$\psi = k_x^2 - k_y^2$	$\pm 2$	$B_{1g}$ singlet,	-	-	$B_{1g}$ singlet,	
	Jan		$B_{2u}$ triplet (m <sub>s</sub> = ±1)			$B_{2u}$ triplet (m <sub>s</sub> = ±1)	
B <sub>2g</sub>	$\psi=2k_xk_y$	$\pm 2$	$B_{2g}$ singlet,	-	-	$B_{2g}$ singlet,	
			$B_{1u}$ triplet (m <sub>s</sub> = ±1)			$B_{1u}$ triplet (m <sub>s</sub> = ±1)	
$A_{1u}$	$\mathbf{d}=(k_x,k_y,0)$	0	A <sub>1u</sub> triplet (m <sub>s</sub> = $\pm 1$ )	$A_{1g}$ triplet (m <sub>s</sub> = 0)	$A_{1g}$ triplet (m <sub>s</sub> = 0)	$A_{1u}$ triplet (m <sub>s</sub> = ±1)	
$A_{2u}$	$\mathbf{d} = (k_y, -k_x, 0)$	0	$A_{2u}$ triplet (m <sub>s</sub> = ±1).	-	-	$A_{2u}$ triplet (m <sub>s</sub> = ±1),	
			$A_{1g}$ singlet			$A_{1g}$ singlet	
$B_{1u}$	$\mathbf{d} = (k_x, -k_y, 0)$	$\pm 2$	$B_{1u}$ triplet (m <sub>s</sub> = ±1),	$B_{1g}$ triplet (m <sub>s</sub> = 0)	$B_{1g}$ triplet (m <sub>s</sub> = 0)	$B_{1u}$ triplet (m <sub>s</sub> = ±1),	
			$B_{2g}$ singlet			$B_{2g}$ singlet	
$B_{2u}$	$\mathbf{d}=(k_y,k_x,0)$	$\pm 2$	$B_{2u}$ triplet (m <sub>s</sub> = ±1),	$B_{2g}$ triplet (m <sub>s</sub> = 0)	$B_{2g}$ triplet (m <sub>s</sub> = 0)	$B_{2u}$ triplet (m <sub>s</sub> = ±1),	
			$B_{1g}$ singlet			$B_{1g}$ singlet	
$E_{2u}^{+}$	$\mathbf{d} = (0, 0, k_x + ik_y)$	1	$E_{2u}^+$ triplet (m <sub>s</sub> = 0)	$A_{1g}$ triplet (m <sub>s</sub> = 1),	$A_{1g}$ triplet (m <sub>s</sub> = 1),	$E_{2u}^+$ triplet (m <sub>s</sub> = 0)	
				$B_{1g} + i B_{2g}$ triplet (m <sub>s</sub> = -1)	$B_{1g} + i B_{2g}$ triplet (m <sub>s</sub> = -1)		
$E_{2u}^{-}$	$\mathbf{d} = (0, 0, k_x - ik_y)$	-1	$E_{2u}^-$ triplet (m <sub>s</sub> = 0)	A <sub>1g</sub> triplet $(m_s = -1)$ ,	A <sub>1g</sub> triplet $(m_s = -1)$ ,	$E_{2u}^{-}$ triplet (m <sub>s</sub> = 0)	
				$B_{1g} - iB_{2g}$ triplet (m <sub>s</sub> = 1)	$B_{1g} - iB_{2g}$ triplet (m <sub>s</sub> = 1)		

ABS and Balatsky, PRB 87, 220506(R) (2013)

### **Multiband Superconductors**

- S: Spin (even: spin-triplet; odd: spin-singlet)
- P: Spatial parity (even: *s,d*-wave; odd: *p,f*-wave)
- O: Orbital or band parity (even; odd orbital)
- T: Time (even; odd-frequency)

S = 0	P	Т	Ο	S = 1	P	Т	0
even- $\omega$	+	H	A	even- $\omega$			Ð
even- $\omega$	—	+	_	even- $\omega$	+	+	—
odd- $\omega$	+		$\overline{\bigcirc}$	odd- $\omega$	+	—	+
odd- $\omega$			+	odd- $\omega$			$ \rightarrow $

ABS and Balatsky, PRB 88, 104514 (2013)





Simple Two-Band Superconductor  

$$H_{ab} = \sum_{k\sigma} \varepsilon_{a}(k)a_{k\sigma}^{\dagger}a_{k\sigma} + \varepsilon_{b}(k)b_{k\sigma}^{\dagger}b_{k\sigma}$$

$$+ \sum_{k} \Delta_{a}(k)a_{k\uparrow}^{\dagger}a_{-k\downarrow}^{\dagger} + \Delta_{b}(k)b_{k\uparrow}^{\dagger}b_{-k\downarrow}^{\dagger} + \text{H.c.} \qquad \text{Interband}$$

$$H_{cd} = \sum_{k\sigma} \varepsilon_{c}(k)c_{k\sigma}^{\dagger}c_{k\sigma} + \varepsilon_{d}(k)d_{k\sigma}^{\dagger}d_{k\sigma} \quad \text{Diagonal bands}$$

$$+ \sum_{k} \Delta_{c}(k)c_{k\uparrow}^{\dagger}c_{-k\downarrow}^{\dagger} + \Delta_{d}(k)d_{k\uparrow}^{\dagger}d_{-k\downarrow}^{\dagger} + \text{H.c.} \quad \text{Intraband pairing}$$

$$+ \sum_{k} \Delta_{cd}(k)[c_{k\uparrow}^{\dagger}d_{-k\downarrow}^{\dagger} + d_{k\uparrow}^{\dagger}c_{-k\downarrow}^{\dagger}] + \text{H.c.} \quad \text{Interband pairing}$$

$$\Delta_{cd} = \frac{(\Delta_{b} - \Delta_{a})|\Gamma|}{\sqrt{(\varepsilon_{a} - \varepsilon_{b})^{2} + 4|\Gamma|^{2}}}$$
ABS and Balatsky, PRB 88, 104514 (2013)

## **Time-Dependent Pairing**



Time-ordered s-wave interband pairing:

$$F^{\pm}(\tau) = \frac{1}{2N_{\mathbf{k}}} \sum_{\mathbf{k}} \mathcal{T}_{\tau} \langle c_{-\mathbf{k}\downarrow}(\tau) d_{\mathbf{k}\uparrow}(0) \pm d_{-\mathbf{k}\downarrow}(\tau) c_{\mathbf{k}\uparrow}(0) \rangle$$

 $F^e = F^+(\tau 
ightarrow 0^+)$  Even- $\omega$ , even-interband pairing



ABS and Balatsky, PRB 88, 104514 (2013)





Komendova, Balatsky, and ABS, PRB 92, 04517 (2015)

### **Interband Pairing**



Perturbation theory to infinite order in  $\Gamma$ 

Odd-interband: 
$$F_{12}^{\text{odd}}(k, i\omega) = \frac{F_{12} - F_{21}}{2} = i\omega \Gamma(\Delta_1 - \Delta_2)/D$$
  
Even-interband:  $F_{12}^{\text{even}}(k, i\omega) = \frac{F_{12} + F_{21}}{2} = \Gamma(\Delta_1 \varepsilon_2 - \Delta_2 \varepsilon_1)/D$   
 $\begin{pmatrix} D = (\omega^2 + E_1^2)(\omega^2 + E_2^2) - \Gamma^2[2(\varepsilon_1 \varepsilon_2 - \omega^2) - \Delta_2^* \Delta_1 - \Delta_1^* \Delta_2] + \Gamma^4 \\ E_j^2 = \varepsilon_j^2 + |\Delta_j|^2 \end{pmatrix}$ 

**Odd-frequency** pairing:  $\Gamma \neq 0$ ,  $\Delta_1 \neq \Delta_2$ 

Komendova, Balatsky, and ABS, PRB 92, 04517 (2015)







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# **Odd-** $\omega$ **Superconductivity in Sr**<sub>2</sub>**RuO**<sub>4</sub>

- Superconductivity in Sr<sub>2</sub>RuO<sub>4</sub>
- Two- and three-orbital models
- Odd- $\omega$  pairing measured by Kerr effect

### Strontium Ruthenate, Sr<sub>2</sub>RuO<sub>4</sub>





# Superconductivity in a layered perovskite without copper

- Y. Maeno<sup>\*</sup>, H. Hashimoto<sup>\*</sup>, K. Yoshida<sup>\*</sup>, S. Nishizaki<sup>\*</sup>, T. Fujita<sup>\*</sup>, J. G. Bednorz<sup>†</sup> & F. Lichtenberg<sup>†</sup><sup>‡</sup> Nature 372, 532 (1994)
- T<sub>c</sub> = 1.5K
- Non-s-wave (disorder sensitive)
- Spin-triplet (neutron scattering, Knight shift)
- Breaks time-reversal symmetry (Kerr effect)

→ Spin-triplet chiral  $p_x$ +i $p_y$ -wave symmetry

## **Properties of Sr<sub>2</sub>RuO<sub>4</sub>**

Three Fermi surfaces (FSs)





Three Ru 4d orbitals:

- $xy \rightarrow \gamma$  (electron-like)
- *xz*, *yz*  $\rightarrow \beta$  (electron-like) and  $\alpha$  (hole-like)

[1]: Damascelli et al, PRL 85, 5194 (2000), [2]: Phys. Today 54 (2001), [3]: Mackenzie and Maeno, RMP 75, 657 (2003)



### Superconducting state

 $\begin{pmatrix} \Delta_{\uparrow\uparrow}(\mathbf{k}) & \Delta_{\uparrow\downarrow}(\mathbf{k}) \\ \Delta_{\downarrow\uparrow}(\mathbf{k}) & \Delta_{\downarrow\downarrow}(\mathbf{k}) \end{pmatrix} = \begin{pmatrix} -d_x(\mathbf{k}) + id_y(\mathbf{k}) & \psi(\mathbf{k}) + d_z(\mathbf{k}) \\ -\psi(\mathbf{k}) + d_z(\mathbf{k}) & d_x(\mathbf{k}) + id_y(\mathbf{k}) \end{pmatrix}$ 

#### Triplet **d**-vector for chiral state

 $\mathbf{d}(\mathbf{k}) = (0, 0, k_x \pm ik_y)$ 



# Kerr Effect in Sr<sub>2</sub>RuO<sub>4</sub>

Reflected light has a slightly rotated plane of polarization if material breaks time-reversal symmetry (TRS)



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SC state in Sr<sub>2</sub>RuO<sub>4</sub> breaks TRS But ...

Clean single-band chiral SC has zero Kerr effect

→ Interband pairing with relative superconducting phases [2]

Electric-field driven interband transitions with relative SC phases  $\rightarrow$  finite transverse Hall current response  $\rightarrow$  finite Kerr effect

[1]: Xia et al, PRL 97, 167002 (2006), [2]: Taylor and Kallin, PRL 108, 157001 (2012); Wysokinski et al, PRL 108, 077004 (2012)



# **Two-Orbital Model for Sr<sub>2</sub>RuO<sub>4</sub>**

Hamiltonian  $\sum_{\mathbf{k}} \Psi_{\mathbf{k}}^{\dagger} \hat{H}_{\mathbf{k}} \Psi_{\mathbf{k}}$  with  $\hat{H}_{\mathbf{k}} = \begin{pmatrix} \hat{H}_{0}(\mathbf{k}) & \dot{\Delta}(\mathbf{k}) \\ \dot{\Delta}^{\dagger}(\mathbf{k}) & -\hat{H}_{0}(-\mathbf{k}) \end{pmatrix}$ 

 $\mathbf{h} \quad H_{\mathbf{k}} = \begin{pmatrix} \Pi_0(\mathbf{k}) & \Delta(\mathbf{k}) \\ \check{\Delta}^{\dagger}(\mathbf{k}) & -\hat{H}_0(-\mathbf{k}) \end{pmatrix}$  $\Psi_{\mathbf{k}}^{\dagger} = (c_{\mathbf{k}\uparrow 1}^{\dagger} c_{\mathbf{k}\uparrow 2}^{\dagger} c_{-\mathbf{k}\downarrow 1} c_{-\mathbf{k}\downarrow 2})$ 

• Spin-triplet  $\mathbf{d} \parallel \hat{\mathbf{z}}$ 

 $\rightarrow \alpha, \beta$  bands

•

*xz* (1),*yz* (2) orbitals



## **Odd-** $\omega$ **Pairing and Kerr Effect**



### Odd- $\omega$ , odd-interorbital pairing:

$$F_{12} - F_{21} = i\omega [(\Delta_2 - \Delta_1)\epsilon_{12} + \Delta_{12}(\xi_1 - \xi_2)]/D \qquad (D \sim \omega^2)$$

Interorbital hybridization + gap asymmetry

Interorbital pairing + dispersion asymmetry

#### Kerr effect: [1]

$$\sigma_H \propto \epsilon_{12} \operatorname{Im}(\Delta_1^* \Delta_2) + \xi_1 \operatorname{Im}(\Delta_2^* \Delta_{12}) - \xi_2 \operatorname{Im}(\Delta_1^* \Delta_{12})$$

Interorbital hybridization + gap asymmetry

Interorbital pairing + dispersion asymmetry

### $\rightarrow$ Intrinsic Kerr effect evidence of odd- $\omega$ superconductivity

Komendova and ABS, PRL 119, 087001 (2017), [1]: Taylor and Kallin, PRL 108, 157001 (2012)

### **Generic Three-Orbital Model**

$$\hat{H}_{0}(\mathbf{k}) = \begin{pmatrix} \xi_{1} & \epsilon_{12} & \epsilon_{13} \\ \epsilon_{12} & \xi_{2} & \epsilon_{23} \\ \epsilon_{13} & \epsilon_{23} & \xi_{3} \end{pmatrix}, \quad \check{\Delta}(\mathbf{k}) = \begin{pmatrix} \Delta_{1} & \Delta_{12} & \Delta_{13} \\ \Delta_{12} & \Delta_{2} & \Delta_{23} \\ \Delta_{13} & \Delta_{23} & \Delta_{3} \end{pmatrix}$$

*xy* (1), *xz* (2),*yz* (3) orbitals  $\rightarrow$   $\gamma$ ,  $\alpha$ ,  $\beta$  bands

### General interorbital pairing:

 $F_{AS} = \sum_{i,j,k=1,\dots,N} \epsilon_{ijk} F_{ij}$  $F_{AS}(-\omega) = -F_{AS}(\omega)$ 

odd- $\omega$ , odd-interorbital

Komendova and ABS, PRL 119, 087001 (2017)

$$F_S = \sum_{i \neq j=1,\dots,N} F_{ij}$$
  
$$F_S(-\omega) = F_S(\omega)$$

#### even- $\omega$ , even-interorbital



## **Examples Odd-**ω Pairing



• No interorbital pairing and  $\epsilon_{ij} = \Gamma$ :

 $F_{\text{odd}} = 2\Gamma i\omega [\Delta_1(\epsilon_2 - \epsilon_3)(\epsilon_2 + \epsilon_3 + \Gamma) + |\Delta_1|^2 (\Delta_3 - \Delta_2) + \text{two cyclic permutations}]/D_3$ 

- Only  $\epsilon_{23} \neq 0, \Delta_{23} \neq 0$  (xz, yz  $\Rightarrow \alpha, \beta$  bands with hybridization, xy  $\Rightarrow \gamma$  band)  $F_{\text{odd}} = 2i\omega[\Delta_{23}(\epsilon_3 - \epsilon_2) + \epsilon_{23}(\Delta_3 - \Delta_2)]/D'_3$
- → Odd-ω pairing requires only limited interorbital processes, exactly same as finite Kerr rotation [1]

Komendova and ABS, PRL 119, 087001 (2017), [1]: Gradhand et al, PRB 88, 094504 (2013)



### Pair Amplitudes in Sr<sub>2</sub>RuO<sub>4</sub>





# Odd- $\omega$ Pairing in k-Space



- Chiral *p*-wave symmetry
- Peaked at  $\alpha$ ,  $\beta$  hybridization
- Nodal structure dependent on exact parameters

Komendova and ABS, PRL 119, 087001 (2017)

### Summary



- Bulk odd- $\omega$  superconductivity in multiband systems
  - Finite interband/orbital pairing or hybridization
- Odd- $\omega$  pairing in Sr<sub>2</sub>RuO<sub>4</sub> measured by Kerr effect
- Future directions:
  - Other odd- $\omega$  bulk superconductors (UPt<sub>3</sub>, ...)
  - Consequences of odd- $\omega$  pairing?
  - Band/orbital  $\rightarrow$  layer, wires, ...

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