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Stability of Time-Reversal Symmetry Breaking State by applying Magnetic Field in Inhomogeneous Superconductivity

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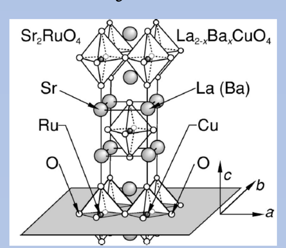
We show a superconducting gap structure with horizontal minimum lines and temperature dependences of spin susceptibility for the non-unitary chiral superconductivity of Eu: $(k_z\boldsymbol{z}+\epsilon k_z\boldsymbol{x}, k_y\boldsymbol{z}+\epsilon k_z\boldsymbol{y})$ mixed through a coefficient ϵ, in the Sr_2RuO_4 model. Assuming the non-unitary chiral state of a bulk phase, the analysis of the Ginzburg-Landau equation clarifies a field-induced chiral transition and paramagnetic chiral supercurrent in an inhomogeneous interface phase of a eutectic Sr_2RuO_4-Ru. Although the field-induced chiral phenomena change by increasing ϵ in the nonunitary state, the features are qualitatively similar to those in the unitary state for $\epsilon=0$

Superconductivity in Sr_2RuO_4 (SRO)

Sr_2RuO_4
Perovskite structure
$T_c = 1.5K$



Y. Maeno, et al., Nature 372, 532 (1994).
A. P. Mackenzie and Y. Maeno, Rev. Mod. Phys. 75, 657 (2003).

SC gap structure

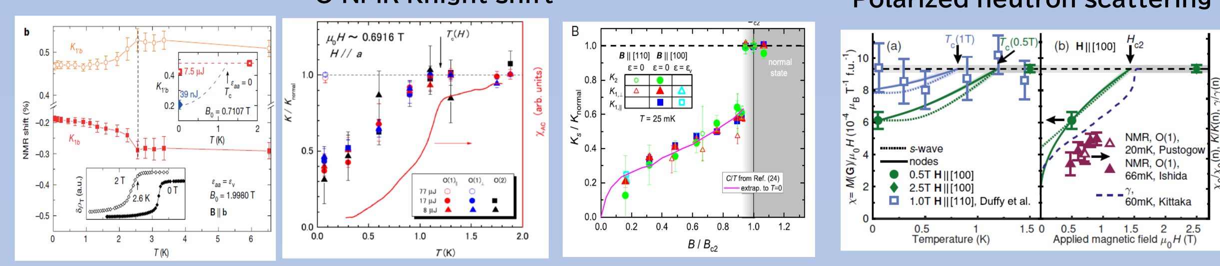
Field-angle-dependent specific heat
Horizontal line node
Dependence on k_z
S. Kittaka, et al, JPSJ. 87, 093703 (2018)

Field-dependence thermal conductivity
Vertical line node
Dependence on k_x, k_y
E. Hassinger, et al, PRX 7, 011032 (2017)

Anisotropic gap structure

Spin susceptibility

^{17}O NMR Knight shift — Polarized neutron scattering



A.Pustogow, et al, Nature, 74, 72(2019) — K. Ishida et al., J. Phys.Soc. Jpn. 89, 034712 (2020) — A. Chronister et al., 118 (25) e202531311 (2020) — A.N. Petsch, et al. Phys. Rev. Lett. 125, 217004 (2020).

Reduction of spin susceptibility in in-plane field

Spontaneous magnetization

μSR measurement in Sr_2RuO_4
G. M. Luke et al , Nature 394, 558 (1998)

μSR in eutectic Sr_2RuO_4-Ru
Split of T_{TRSB} and T_c
T. Shiroka, et al, PRB 85, 134527 (2012)

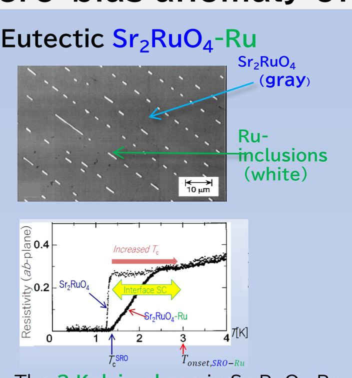
μSR in uni-axial stressed Sr_2RuO_4
Sprit of T_{TRSB} and T_c
V. Grinenko, Nature Phys., 17, 748–754 (2021)

Time reversal symmetry breaking

Two-component order parameter

Zero-bias anomaly of tunneling conductance in Sr_2RuO_4-Ru

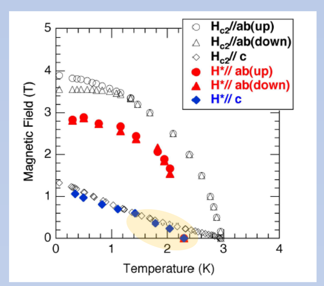
Eutectic Sr_2RuO_4-Ru



The 3 Kelvin phase in Sr_2RuO_4-Ru

Y. Maeno et al., PRL 81, 3765 (1998)

Field-dependence of zero-bias anomaly of differential conductance in tunnel spectroscopy to Ru/SRO-interface in the 3 Kelvin phase of the eutectic SRO-Ru



H. Yaguchi, et al. , AIP Conf. Proc. 850, 543 (2006)

M. Kawamura, et al., JPSJ 74, 531 (2005)

Field-induced order-parameter component

Half-quantum vortex

Feature of spin-triplet pairing, favoring a c-axis oriented d-vector,

X. Cai, et al., Phys. Rev. B 105, 224510 (2022).

Non-unitary chiral state of E_u : $(k_x\boldsymbol{z} + \epsilon k_z\boldsymbol{x}, k_y\boldsymbol{z} + \epsilon k_z\boldsymbol{y})$

non-unitary E_u: $(k_x\boldsymbol{z} + \epsilon k_z\boldsymbol{x}, k_y\boldsymbol{z} + \epsilon k_z\boldsymbol{y})$

Point group D_{4h},
E_u: $k_x\boldsymbol{z}, k_y\boldsymbol{z}$; $k_z\boldsymbol{x}, k_z\boldsymbol{y}$

Two component order parameter (η_1, η_2)
The combination by ϵ is possible through a three-dimensional character originating from the spin-orbit and inter-orbital interlayer couplings. W. Huang and H. Yao, PRL 121, 157002 (2018)

chiral state of non-unitary : E_u

non-unitary state
$$\boldsymbol{d} = \eta_1\boldsymbol{d_1} + \eta_2\boldsymbol{d_2}$$
Two-component order parameter
$$\begin{cases}\boldsymbol{d_1} = k_x\boldsymbol{z} + \epsilon k_z\boldsymbol{x} \\ \boldsymbol{d_2} = k_y\boldsymbol{z} + \epsilon k_z\boldsymbol{y}\end{cases} \quad \epsilon \in Re$$

$\epsilon \neq 0$: non-unitary
$\epsilon = 0$: unitary

chiral state
$$\begin{cases}\eta_1 = \eta_1^{Re} \\ \eta_2 = i\eta_2^{Re}\end{cases}$$
$\eta_1^{Re}, \eta_2^{Re} \in Re$

$$\begin{cases}\boldsymbol{d_1} = \sin k_x\boldsymbol{z} + \epsilon\sin k_z\boldsymbol{x} \\ \boldsymbol{d_2} = \sin k_y\boldsymbol{z} + \epsilon\sin k_z\boldsymbol{y}\end{cases}$$

$$\boldsymbol{d} = \eta_p\boldsymbol{d_1} + \eta_t\boldsymbol{d_2} = \begin{pmatrix}\eta_1\epsilon\sin k_z \\ \eta_2\epsilon\sin k_z \\ \eta_1\sin k_x + \eta_2\sin k_y\end{pmatrix},\ \boldsymbol{d_1} = \begin{pmatrix}\epsilon\sin k_z \\ 0 \\ \sin k_x\end{pmatrix}\ \ \boldsymbol{d_2} = \begin{pmatrix}0 \\ \epsilon\sin k_z \\ \sin k_y\end{pmatrix}$$

$$\boldsymbol{d}^*\times\boldsymbol{d} = -2i\eta_1^{Re}\eta_2^{Im}(\boldsymbol{d_1}\times\boldsymbol{d_2}) = -2i\eta_1^{Re}\eta_2^{Im}\left\{\epsilon k_z\begin{pmatrix}-\sin^2 k_x \\ -\sin^2 k_y \\ \epsilon\sin^2 k_z\end{pmatrix}\right\} \neq 0 \quad \text{at } \epsilon \neq 0$$

non-unitary

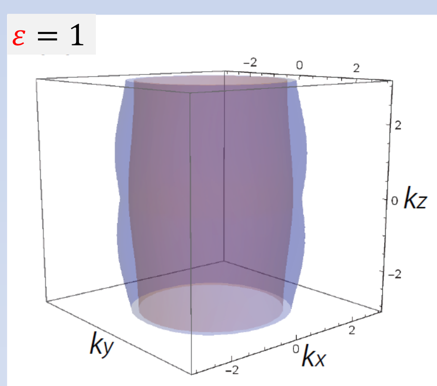
Superconducting Gap and Spin Susceptibility

Energy of Superconducting state in-non-unitary chiral state

$$E_{\boldsymbol{k}}^2 = \varepsilon_k{}^2 + |\boldsymbol{d}|^2 \pm |\boldsymbol{d}^*\times\boldsymbol{d}|$$

Energy dispersion: ε_k
$$\varepsilon_k = (1-\delta\cos k_z)(k_x^2+k_y^2) - \mu$$
$$\delta = 0.11 \quad \mu = 2.0$$
Two-dimensional Fermi Surface weakly warped along to the k_z-axis

$$|\boldsymbol{d}|^2 = (|\eta_1|^2\sin^2 k_x + |\eta_2|^2\sin^2 k_y) + \epsilon^2\sin^2 k_z(|\eta_1|^2+|\eta_2|^2)$$
$$|\boldsymbol{d}^*\times\boldsymbol{d}| = 2\epsilon|\eta_1||\eta_2|\sin k_z\sqrt{\sin^2 k_x + \sin^2 k_y + \varepsilon^2\sin^2 k_z}$$



non-nodal horizontal-line minimum gaps at $k_z = 0, \pi$

Spin susceptivity $\chi_{ij}/\chi_{i,j,n}$

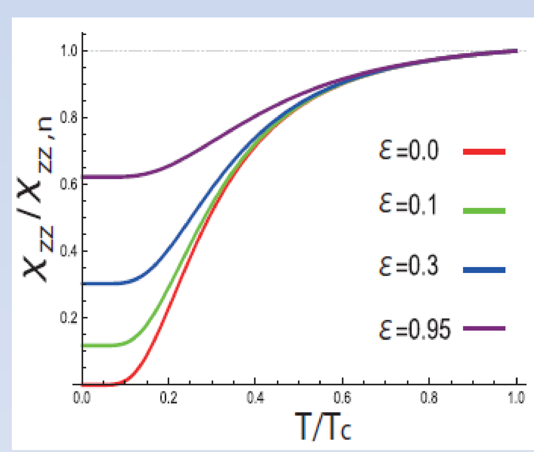
ref. A. J. Leggett, Rev. Mod. Phys. 47, 331 (1975)

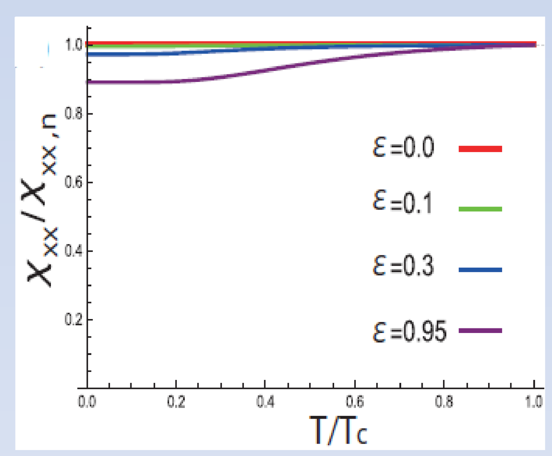
$$\chi_{ij}/\chi_n = \left\{\delta_{ij} - \int dk_x dk_y dk_z\,[1 - Y(\boldsymbol{k}_{FS}, T)][d_i^* d_j/(|\boldsymbol{d}|^2 + |\boldsymbol{d}^*\times\boldsymbol{d}|)]\right\}$$

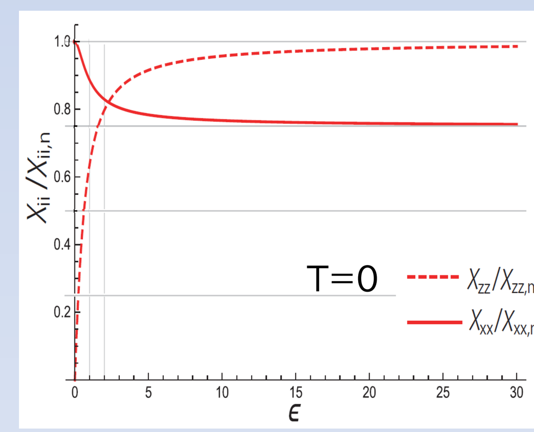
$\boldsymbol{k}_{FS}$ on the cylindrical Fermi Surface (δ=0)

Yosida function: $Y(\boldsymbol{k}:T) \equiv \int_0^\infty d\varepsilon_k \frac{1}{2}\beta\, sech^2\frac{1}{2}\beta\, E_{\boldsymbol{k}\pm}$

$\eta_1 = -i\eta_2 = \eta_0\sqrt{1 - T/T_c}$







The reduction rates χ_{xx} and χ_{zz} at zero temperature change depending on ϵ.
In the non-unitary state at a large value of ϵ=1~2, χ_{xx} reduces by approximately 10~15% and the reduction in χ_{zz} weakens.

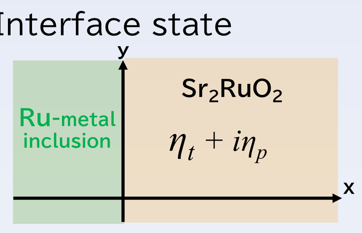
Field-induced Chiral Transition by applying $H//z$ in Inhomogeneous Interface State; The 3 Kelvin Phase of Eutectic Sr_2RuO_4-Ru

Assuming the *chiral non-unitary* state in bulk phase in eutectic Sr_2RuO_4-Ru,

non-unitary state
$$\boldsymbol{d} = \eta_p\boldsymbol{d_p} + \eta_t\boldsymbol{d_t}$$
Two-component order parameter
$$\begin{cases}\boldsymbol{d_p} = k_z\boldsymbol{z} + \epsilon k_z\boldsymbol{x} \\ \boldsymbol{d_t} = k_y\boldsymbol{z} + \epsilon k_z\boldsymbol{y}\end{cases}$$

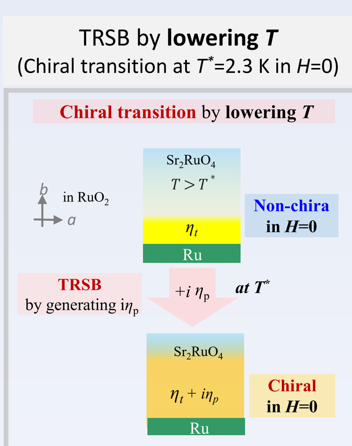
Chiral state
$$\begin{cases}\eta_t = \eta_t^{Re} & \textit{tangential} \text{ component} \\ \eta_p = i\eta_p^{Re} & \textit{papendicular} \text{ component}\end{cases}$$

Interface state

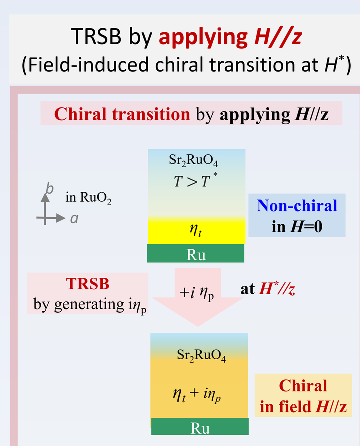


η_t^{Re} : *tangential*
$i\eta_p^{Re}$: *papendicular*

$$\begin{cases}\boldsymbol{d_p} = k_z\boldsymbol{z} + \epsilon k_z\boldsymbol{x} \\ \boldsymbol{d_t} = k_y\boldsymbol{z} + \epsilon k_z\boldsymbol{y}\end{cases}$$



The 3-Kelvin phase model
M. Sigrist and H. Monien, J. Phys. Soc. Jpn. 70, 2409 (2001)



Field-induced chiral transition
H. Kaneyasu et al., Phys. Rev. B 100, 214501 (2019)

Ginzburg-Landau free energy

$$F = \int_0^\infty dx\,[\alpha(T,T_c)\{1+\epsilon^2 a_{zz}\}\left(|\eta_p|^2 + |\eta_t|^2\right)$$
$$+\frac{1}{4}b\{1+\epsilon^2 b_{zz}\}\left\{\frac{3}{2}\left(|\eta_p|^2+|\eta_t|^2\right)^2 - |\eta_p|^2|\eta_t|^2 + \frac{1}{2}\left(\eta_p{}^{*2}\eta_t{}^2 + \eta_p{}^2\eta_t{}^{*2}\right)\right\}$$
$$+K_1\{1+\epsilon^2\mathcal{K}_{1,zz}\}\left(|D_x\eta_p|^2 + |D_y\eta_t|^2\right) + K_2\{1+\epsilon^2\mathcal{K}_{2,zz}\}\left(|D_y\eta_p|^2 + |D_x\eta_t|^2\right)$$
$$+\left\{K_3(D_x\eta_p)^*(D_y\eta_t) + K_4(D_y\eta_p)^*(D_x\eta_t) + c.c\right\}$$
$$+K_5\{1+\epsilon^2\mathcal{K}_{5,zz}\}\left(|D_z\eta_p|^2 + |D_z\eta_t|^2\right) + \frac{1}{8\pi}|\nabla\times\boldsymbol{A} - \boldsymbol{H}|^2]$$

$\boldsymbol{D} = \nabla - iy\boldsymbol{A}$
$\alpha = a'(T - T_c(x))$

Interface energy and Enhancement of T_c at Ru/SRO interface

$F_{int} = 1/l_p|\eta_p(x=0)|^2$ $\quad l_p$: Extrapolation length into the Ru-metal interface

$T_c(x) = T_c + T_0/\cosh(x/w)$ $\quad T_c(x)$ enhances at the Ru/SRO interface

For a cylindrical FS, the relations of coefficients in the weak coupling limit, $a' = 15b/4$ and $K_1 = 3K_{2,3,4} = 150K_5$.
ref. For unitary state of E_u: D. F. Agterberg, et al., Phys. Rev. B 58, 14484 (1998)

We introduces a_{zz}, b_{zz} and $\mathcal{K}_{1,2,5,zz}$ to represent the non-unitary state which is set with values of $a'/10$, $b/10$ and $K_{1,2,5}/10$.

Gradient terms in $H//z$

$$f_K = f_1 + f_3 + f_4$$
$$f_1 = K_1[1+\epsilon^2\mathcal{K}_{1,zz}]|\partial_x\eta_p|^2 + K_2[1+\epsilon^2\mathcal{K}_{2,zz}]|\partial_x\eta_t|^2$$
$$f_3 = (\gamma A_y)^2\{K_1[1+\epsilon^2\mathcal{K}_{1,zz}]\eta_t{}^2 + K_2[1+\epsilon^2\mathcal{K}_{2,zz}]\eta_p{}^2\}$$
$$f_4 = -i\gamma A_y\{K_3\{-(\partial_x\eta_p)^*\eta_t - K_4\eta_p{}^*(\partial_x\eta_t)\} - c.c.\}$$

Magnetization B//z parallel to chiral axis

$$\boldsymbol{B} = \begin{pmatrix}0\\0\\B_z\end{pmatrix} = \begin{pmatrix}\partial_y A_z - \partial_z A_y \\ \partial_z A_x - \partial_x A_z \\ \partial_x A_y - \partial_y A_x\end{pmatrix} \qquad \boldsymbol{A} = \begin{pmatrix}A_x(x,y,z) \\ A_y(x) \\ A_z(x,y,z)\end{pmatrix}$$

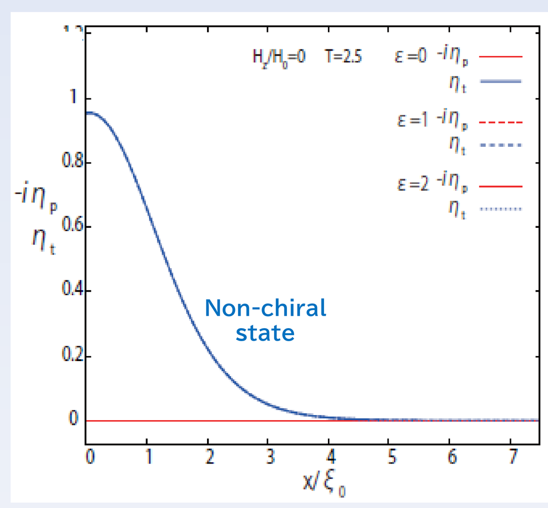
Boundary condition

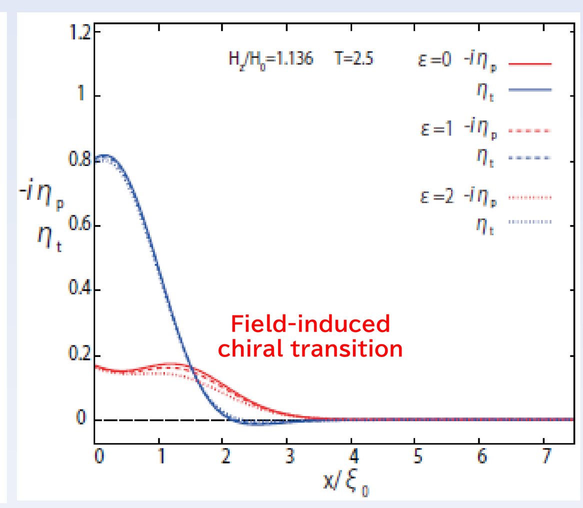
$$K_1[1+\epsilon^2\mathcal{K}_{1,zz}](\partial_x\eta_p)|_{x=0} = 1/l_p\eta_p(0) + i\gamma A_y(0)K_3\eta_t(0)$$
$$K_2[1+\epsilon^2\mathcal{K}_{2,zz}](\partial_x\eta_t)|_{x=0} = i\gamma A_y(0)K_4\eta_p(0)$$
$$\partial_x A_y\Big|_{x=0,L} = H_z$$

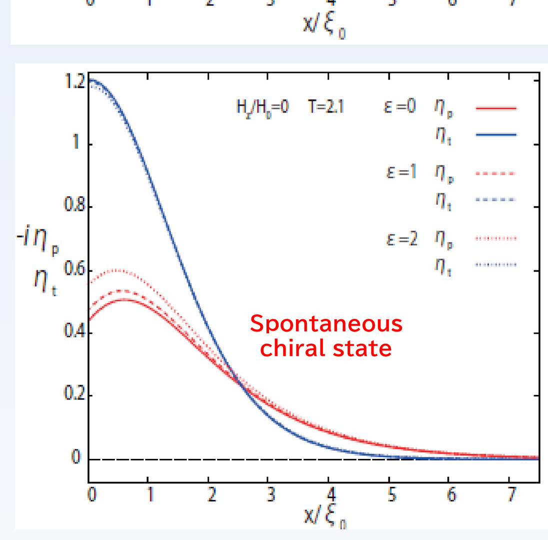
ref. GL-theory in D_{4h}: M. Sigrist and K. Ueda, Rev. Mod. Phys. 63, 239 (1991).
ref. The 3 Kelvin phase model : M. Sigrist and H. Monien, J. Phys. Soc. Jpn. **70**, 2409 (2001).
cf. Field-induced chiral phenomena in the unitary state of E_u, analogical to E_g,
H. Kaneyasu et al. Phys. Rev. B Rev.B 100, 214501 (2019), JPS Conf. Proc. 30, 011039 (2020)

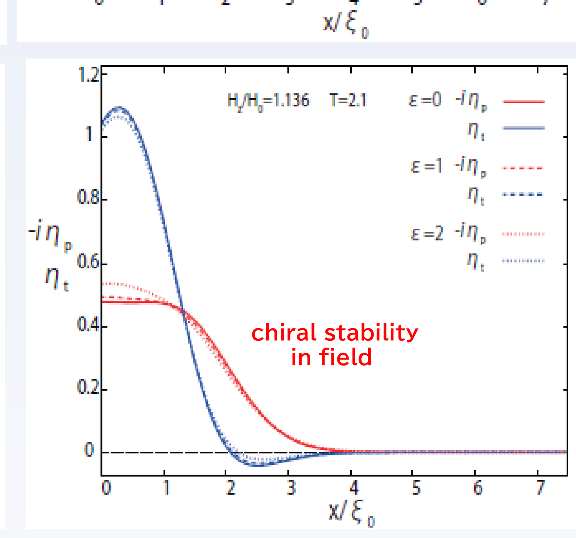
Field-induced chiral transition and paramagnetic chiral current in inhomogeneous interface state in $H//z$

Two-components order parameter (η_p, η_t)









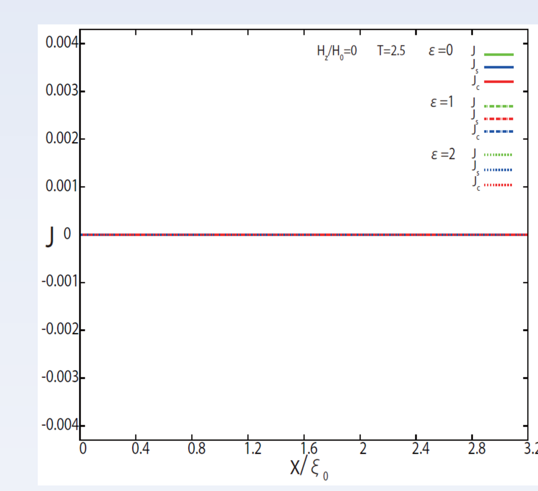
The application of $H//z$ derives second order parameter η_t .
It is the field-induced chiral transition to the two-component state as breaking of TRS. The features are of field-induced chiral transition qualitatively similar to those in the unitary state.

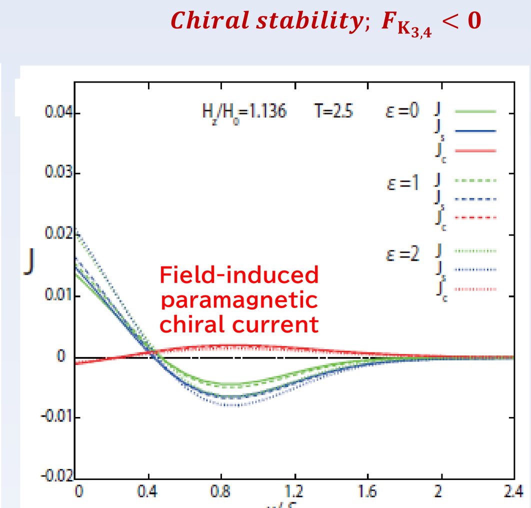
η_p near the interface increases by strengthening ϵ, relative to the unitary state. This behavior is obtained through the effect of ϵ in boundary conditions which contain ϵ introduced from f_1.

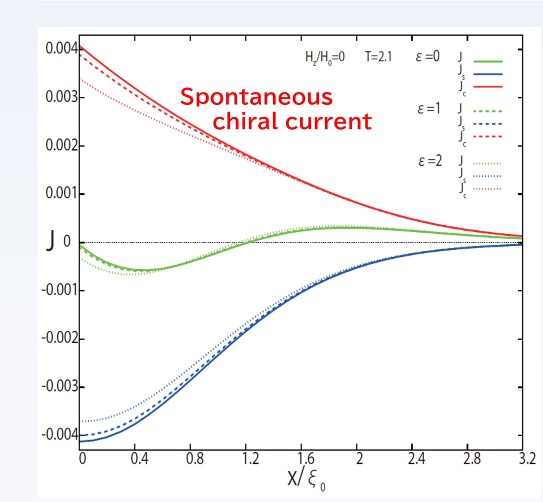
Chiral current j_c and Screening current j_s
Total supercurrent density $j = j_c + j_s$

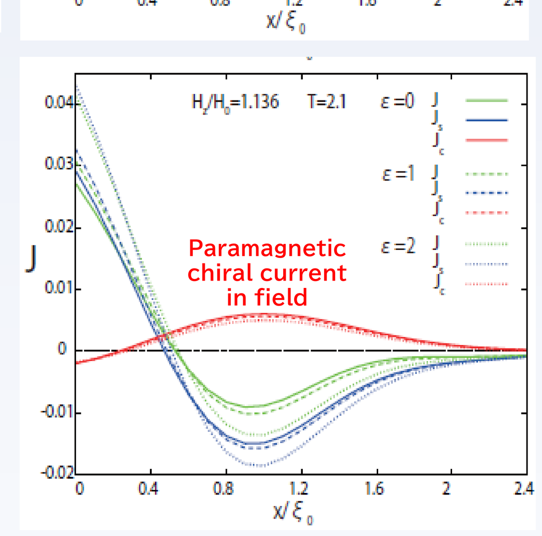
$$\frac{j_y(x)}{8\pi} = \left[-\gamma^2 A_y\left(K_1[1+\epsilon^2 K_{1,zz}]|\eta_1|^2 + K_2[1+\epsilon^2 K_{2,zz}]|\eta_2|^2\right) - i\gamma\{K_3\eta_2^*(\partial_x\eta_1) - K_4\eta_1^*(\partial_x\eta_2) - c.c.\}\right]$$

Screening current; j_s — *Chiral current*; j_c
Pair breating; $F_{K_{1,2}} > 0$ — *Chiral stability*; $F_{K_{3,4}} < 0$









Applying the fields generates the paramagnetic chiral current j_c, and it occurs with the field-induced chiral stability.
By increasing ϵ, the amplitudes of j_s slightly decreases, and j_c increases, compared to that of the unitary state.

cf.: Compared to the field-induced chiral phenomena in the unitary state for $\epsilon = 0$
H. Kaneyasu et al. Phys. Rev. B Rev.B 100, 214501 (2019), JPS Conf. Proc. 30, 011039 (2020).

summary

- Assuming the **non-unitary** chiral p-wave state in E_u of the bulk phase of a pure Sr_2RuO_4, the d-vector described by the second order-parameter components leads the gap structure with **non-nodal horizontal minimum lines at k_z=0, π**. The spin susceptibility **χ_{xx} has a maximum reduction of about 10~15%. This disagrees with the drop less than half in the Knight shift in the in-plane field.**

- Assuming the non-unitary chiral bulk phase in Sr_2RuO_4-Ru, the application of $H/\!/ z$ causes **the field-induced chiral stability and the paramagnetic chiral current** in the 3 Kelvin phase of Sr_2RuO_4-Ru.
Although the order-parameter components have weak changes on varying ϵ,
the behaviors of **the field-induced chiral transition are qualitatively similar between nonunitary and unitary states.**
The Field-induced chiral transition is qualitatively consistent with the field-dependent behaviors of the zero-bias anomaly, observed in the tunnelling spectroscopy, and the consistency can support even the non-unitary chiral state as the bulk state of Sr_2RuO_4.

The consistency can support even the non-unitary chiral state as the bulk state of Sr_2RuO_4.
However, the non-unitary chiral state of E_u is an unlikely candidate for Sr_2RuO_4,
since the reduction rate of χ_{xx} disagrees with that of the experiments.