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# Supplementary information

# Ultrasound evidence for a two-component superconducting order parameter in $Sr_2RuO_4$

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# Supplementary Information for “Ultrasound evidence for a two-component superconducting order parameter in $Sr_2RuO_4$”

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## 1. ECHO PATTERN

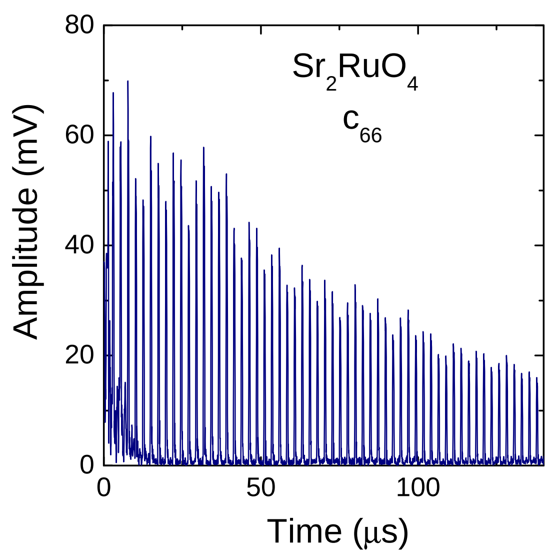


FIG. S1: Echo pattern for the transverse sound mode $c_{66}$ measured in Toronto, in the superconducting state ($H = 0$), corresponding to the data shown in Fig. 2a (red circles). The frequency was $f = 169$ MHz and the temperature $T = 40$ mK. The sample length was 4.0 mm and 58 echoes were recorded.

## 2. REPRODUCIBILITY

Fig. S2 shows data taken in Toulouse for the transverse mode $c_{66}$ in the superconducting state, at $H = 0$ (red dots). The same sample was used but the faces were re-polished and a different bonding agent was used to attach the transducer. As for the Toronto data [1], we again observe a precipitous drop immediately below $T_c$, but we now also see a gradual decrease, apparent below $\sim 1.3$ K, not present in the Toronto data (Fig. 2). We attribute this, and the somewhat larger total change in $c_{66}$ (1.0 ppm vs 0.2 ppm), to a slight contribution coming from other modes mixed in. Indeed, in the Toulouse experiment, the echo pattern was not as clean as in the Toronto experiment (Fig. S1). This may be due to ringing of the transducer, a spurious effect that leads to a non-zero background of the echo amplitude. Moreover, we saw the mixing of another acoustic mode in the echo pattern, whose sound velocity was close to the $c_{66}$ mode. This could be the $c_{44}$ mode, for example, if the polarization of the transducer was not exactly aligned in the $RuO_2$ plane. This effect could explain the softening below $T_c$ but not the discontinuity since no discontinuity is expected in the shear $c_{44}$ mode at $T_c$ by symmetry. As shown in Fig. S2, a softening alone, such as seen in the $(c_{11} - c_{12})/2$ mode (Fig. 1d), yields a much more gradual decrease below $T_c$ than that seen in our $c_{66}$ data.

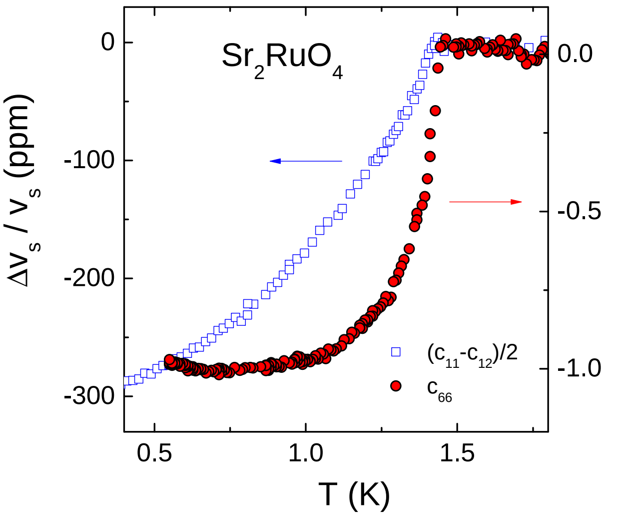


FIG. S2: Relative change in sound velocity for the transverse mode $c_{66}$, measured in Toulouse at a frequency $f = 201$ MHz, at $H = 0$ (red dots, right axis). The open blue diamonds (left axis) show the corresponding data for the mode $(c_{11} - c_{12})/2$ (from Fig. 1d). The drop in $c_{66}$ below $T_c$ is much more abrupt than the softening seen in $(c_{11} - c_{12})/2$, for example.

### 3. EHRENFEST RELATION

The Ehrenfest relation is a general and thermodynamic relation that links the jump of the sound velocity with the jump of the specific heat and the strain dependence of $T_c$ [2],

$$\frac{\Delta c_{nm}}{c_{nm}} = -\frac{\Delta C_p}{T_c}\left(\frac{1}{v_s}\frac{\partial T_c}{\partial u_n}\right)\left(\frac{1}{v_s}\frac{\partial T_c}{\partial u_m}\right) \quad (1)$$

where $C_p$ is the heat capacity jump (by mass) between the normal and the superconducting states. $v_s$ is the sound velocity of the $c_{nm}$ mode and $u_n$ is the strain, both using the Voigt notation.
First we estimate the jump in $c_{11}$. Since we don't know experimentally the strain dependence of $T_c$ along [100] in the linear regime, we will rely on the hydrostatic pressure effect on $T_c$ [3]. In order to use equation 1 to evaluate $\Delta c_{11}$, we need to estimate $\frac{\partial T_c}{\partial u_1}$. The effect of pressure on $T_c$ can be decompose as the effect of $u_i$ $(i = 1...6)$ on $T_c$,

$$\frac{\partial T_c}{\partial P} = \frac{\partial T_c}{\partial u_i}\frac{\partial u_i}{\partial P}$$

where each deformation $u_i$ could contribute differently. Unfortunately, we don't readily have access to each $\frac{\partial T_c}{\partial u_i}$ individually. Nevertheless, we can estimate the contribution of $u_1$ as,

$$\frac{\partial T_c}{\partial P} = \frac{1}{w_1}\frac{\partial T_c}{\partial u_1}\frac{\partial u_1}{\partial P}$$

where $w_1$ is the weight of the $u_1$ contribution. If all three normal deformations $(i = 1, 2, 3)$ have an equal effect, $w_1 = \frac{1}{3}$.

Using this weighted contribution we can write an estimation of the effect of $u_1$ on $T_c$,

$$\frac{\partial T_c}{\partial u_1} = w_1\frac{\partial T_c}{\partial P}\left(\frac{\partial u_1}{\partial P}\right)^{-1} \quad (2)$$

We are only missing the last term, say how $u_1$ is affected by an hydrostatic pressure. To express that, we need to use Hooke's law,

$$\sigma_i = c_{ij}u_j$$

and remember that an hydrostatic pressure applies an isotropic strain,

$$\sigma_1 = \sigma_2 = \sigma_3 = P \qquad \sigma_4 = \sigma_5 = \sigma_6 = 0.$$

Let's rewrite Hooke's law for this case as,

$$P\eta_i = c_{ij}u_j$$

where $\eta_i$ are the component of the vector (1,1,1,0,0,0). Inverting this equation and taking the derivative with respect to $P$, we can explicitly write,

$$\frac{\partial u_i}{\partial P} = \left[c^{-1}\right]_{ij}\eta_j$$

For a tetragonal system we get,

$$\frac{\partial u_i}{\partial P} = \frac{1}{(c_{11}+c_{12})\,c_{33} - 2c_{13}^2}\begin{pmatrix} c_{33}-c_{13} \\ c_{33}-c_{13} \\ c_{11}+c_{12}-2c_{13} \\ 0 \\ 0 \\ 0 \end{pmatrix}$$

Usually, diagonal elastic constants are larger then off-diagonals one. If one neglect $c_{13}$ in front of $c_{11}$ and $c_{33}$, we get a simplified estimation for the jump on $c_{11}$,

$$\frac{\Delta c_{11}}{c_{11}} \approx -\frac{\Delta C_{NS}}{T_c}\left(\frac{w_1}{v_s}\frac{\partial T_c}{\partial P}(c_{11}+c_{12})\right)^2$$

Using $\frac{\Delta C_{NS}}{T_c} = 0.1$ J K$^{-2}$ kg$^{-1}$ [4], $v = 6$ km s$^{-1}$, $\frac{\partial T_c}{\partial P} = 0.2$ K GPa$^{-1}$ [3], $c_{11} = 230$ GPa, $c_{12} = 130$ GPa and $w_1 = \frac{1}{3}$, we get,

$$\frac{\Delta c_{11}}{c_{11}} = 1.6 \text{ ppm}$$

This estimate depends significantly on the value of $\frac{\partial T_c}{\partial P}$.

Next, to estimate the jump in the shear modulus $c_{66}$ we need to deduce the value of $\frac{\partial T_c}{\partial u_6}$. We rely on the dependence of $T_c$ on strain along [110], $\epsilon_{(110)}$, that has been reported in [5]. A strain $\epsilon_{(110)} = u$ implies $u_{xx} = u_{yy} = u_{xy} = u_{yx} = u/2$. From the definition

$$\frac{\partial T_c}{\partial \epsilon_{(110)}} = \frac{\partial T_c}{\partial u_1}\frac{\partial u_1}{\partial \epsilon_{(110)}} + \frac{\partial T_c}{\partial u_2}\frac{\partial u_2}{\partial \epsilon_{(110)}} + \frac{\partial T_c}{\partial u_6}\frac{\partial u_6}{\partial \epsilon_{(110)}},$$

where $u_1 \equiv u_{xx}$, $u_2 \equiv u_{yy}$ and $u_6 \equiv u_{xy} + u_{yx}$, we get

$$\frac{\partial T_c}{\partial u_6} = \frac{\partial T_c}{\partial \epsilon_{(110)}} - \frac{\partial T_c}{\partial u_1}. \quad (3)$$

Hicks *et al.* have found $(\partial T_c/\partial \epsilon_{(110)}) = 10$ K [5], while in the above we estimated $(\partial T_c/\partial u_1) \approx 1/3(c_{11} + c_{12})(\partial T_c/\partial P) = 24$ K. This implies $(\partial T_c/\partial u_6) = -14$ K. Furthermore the transverse acoustic sound velocity corresponding to the $B_{2g}$ shear is $v_{66} = 3.3$ km/s. Thus, we estimate

$$\frac{\Delta c_{66}}{c_{66}} = \left(-\frac{\Delta C_{NS}}{T_c}\right)\frac{1}{v_{66}^2}\left(\frac{\partial T_c}{\partial u_6}\right)^2 = 1.8 \text{ ppm}. \quad (4)$$

This estimate depends significantly on the previous estimation of the jump in $c_{11}$ and on the dependence of $T_c$ on strain along [110].

## 4. FINITE FREQUENCY EFFECT

The estimated jump is about an order of magnitude larger than the measured jump $\Delta c_{66}/c_{66} \approx 0.2$ ppm in our experiments. One reason for this difference can be due to the fact that an elastic constant determination from ultrasound velocity is not a pure thermodynamic measurement, and it involves effects due to finite frequency $\omega$ of the sound wave. Below we look at how finite frequency affects the mean field jump of $c_{66}$ in the superconducting phase $(\Delta_A, \Delta_B) = \Delta_0(1, 0)$.

To model the ultrasound experiment we consider a perturbation in the form of a transverse acoustic wave described by the atomic displacement $\mathbf{u}(\mathbf{r}, t) = \mathbf{u}_0\exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$, with $\mathbf{u}_0 = u_0(1, 0, 0)$ and $\mathbf{k} = (0, k, 0)$. Such a perturbation triggers only the $c_{66}$ mode, with $u_{xy}(\mathbf{r}, t) = \partial_y u_x(\mathbf{r}, t)$, while the remaining strains are zero.

The above acoustic fluctuation will lead to fluctuations of the superconducting order parameters. We write $\Delta_A = \Delta_0 + d_A(\mathbf{r}, t)$, and $\Delta_B = d_{B1}(\mathbf{r}, t) + id_{B2}(\mathbf{r}, t)$, where $d_A$ is a complex function, and $(d_{B1}, d_{B2})$ are real functions. Our goal is to expand the free energy to quadratic order in the fluctuations. We get

$$F_{\text{fluc}} = \int d\mathbf{r}\left[l\Delta_0^2 d_{B1}^2 + 2c_{66}(\partial_y u_x)^2 + \alpha_4\Delta_0(\partial_y u_x)d_{B1}\right], \quad (5)$$

where $l = \beta_2 + \beta_3 + \alpha_4^2/(2c_{66})$, and the renormalized $\beta_i$'s are given later in equation (26). Thus, to quadratic order the displacement fluctuation couples only to $d_{B1}(\mathbf{r}, t)$. From Newton's law the equation of motion for the displacement is $\rho\partial^2\mathbf{u}/\partial t^2 = -\delta F_{\text{fluc}}/\delta\mathbf{u}$, where $\rho$ is the density. This gives

$$\rho\frac{\partial^2 u_x}{\partial t^2} = 4c_{66}\partial_y^2 u_x + \alpha_4\Delta_0\partial_y d_{B1}. \quad (6)$$

For superconducting fluctuation we postulate a damped dynamics given by $\tau_0\partial d_{B1}/\partial t = -\delta F_{\text{fluc}}/\delta d_{B1}$, where $\tau_0$ is a microscopic timescale [6]. This gives

$$\tau_0\frac{\partial d_{B1}}{\partial t} = -(l/2)\Delta_0^2 d_{B1} - \alpha_4\Delta_0\partial_y u_x. \quad (7)$$

Solving the above two equations we get

$$\rho\omega^2 = k^2\left[4c_{66} + \frac{\alpha_4^2\Delta_0^2}{i\omega\tau_0 - \Delta_0^2 l/2}\right]. \quad (8)$$

From the above the frequency dependence of the jump in $c_{66}$ can be read off as

$$\delta c_{66} = \text{Re}\frac{-\alpha_4^2/(2l)}{1 - i\omega\tau_1} = \frac{-\alpha_4^2/(2l)}{1 + \omega^2\tau_1^2}, \quad (9)$$

where $\tau_1 = 2\tau_0/\Delta_0^2$. The above result is to be compared with the jump measured in a purely thermodynamic measurement (see equation (44)). Thus, at finite frequency the jump reduces by a factor $1/(1 + \omega^2\tau_1^2)$ [6], where $\tau_1$ formally diverges at $T_c$ in the thermodynamic limit. In our experiment, we used sound frequency $f \equiv \omega/(2\pi) = 200$ MHz, from which we estimate $\tau_1 \sim 2$ ns. Note that such effect has also been observed in $La_{2-x}Sr_xCuO_4$, where the jump of the longitudinal elastic constant $c_{11}$ at $T_c$ has been measured at different frequencies [7]. In this case, the estimated $\tau_1 \sim 1$ ns, i.e. the same order of magnitude as in $Sr_2RuO_4$.

## 5. THERMAL CONDUCTIVITY

The results of Ref. [8] reveal that $Sr_2RuO_4$has vertical line nodes, i.e. lines of zeros that are parallel to the $c$-axis. All aspects of the data are consistent with a $d$-wave state, with vertical line nodes either along the $a$-axis or along the diagonal. The thermal conductivity study cannot distinguish between these two variations. Now the $(1, 0)$ state we proposed goes as $\Delta_0(k_x k_z, k_y k_z)$, so in addition to have vertical line nodes (at $k_x = 0$ or $k_y = 0$), it also has horizontal line node ($k_z = 0$). The latter line will introduce extra $a - c$ anisotropy in the thermal conductivity. It is difficult to say whether this extra anisotropy is quantitatively compatible or not with the data of Ref. [8].

## 6. PRODUCT TABLE FOR THE $D_{4h}$ POINT GROUP

| $\Gamma$ | $A_{1g}$ | $A_{2g}$ | $B_{1g}$ | $B_{2g}$ | $E_g$ |
|---|---|---|---|---|---|
| $A_{1g}$ | $A_{1g}$ | $A_{2g}$ | $B_{1g}$ | $B_{2g}$ | $E_g$ |
| $A_{2g}$ | $A_{2g}$ | $A_{1g}$ | $B_{2g}$ | $B_{1g}$ | $E_g$ |
| $B_{1g}$ | $B_{1g}$ | $B_{2g}$ | $A_{1g}$ | $A_{2g}$ | $E_g$ |
| $B_{2g}$ | $B_{2g}$ | $B_{1g}$ | $A_{2g}$ | $A_{1g}$ | $E_g$ |
| $E_g$ | $E_g$ | $E_g$ | $E_g$ | $E_g$ | $A_{1g}$+$A_{2g}$+$B_{1g}$+$B_{2g}$ |

TABLE I: Product table for the $D_{4h}$ point group.

## 7. DETAILS OF THE THEORETICAL COMPUTATIONS

The unit cell of $Sr_2RuO_4$ is tetragonal with $D_{4h}$ symmetry. The only irreducible representation of this group which has dimensionality more than one is the two-dimensional representation $E$. In this representation the superconducting order parameter is a two-component $(\Delta_A, \Delta_B)$ complex variable. At this point there are two distinct possibilities based on inversion symmetry of the unit cell. (i) First, the order parameter is odd under parity transformation. In this case the $E_u$ irreducible representation is relevant and $(\Delta_A, \Delta_B)$, transform as $(x, y)$ under point group operations. (ii) The second possibility is that the order parameter is even under parity. In this case the $E_g$ irreducible representation is relevant and $(\Delta_A, \Delta_B)$, transform as $(xz, yz)$.

Beyond the even/odd classification it is difficult to make definite statements about the orbital and spin contents of the Cooper pairs since the system is multi-orbital (and multi-band) and also spin-orbit coupling is strong. It is a priori not clear whether the pairing is best viewed in spin and orbital basis, or in the Bloch diagonal band basis, where the bands are doubly degenerate (pseudospin). In case the pairing is essentially intraband, then overall antisymmetry of the wavefunction will impose that in case (i) the Cooper pairs are singlets in pseudospin basis, and in case (ii) they are pseudospin triplets (the pseudospin content will be described by a **d** vector). If interband pairing is important, then pseudospin singlet-triplet mixing is possible. Similar considerations will hold if the problem is analyzed in orbital and spin basis.

The form of the Landau-Ginzburg free energy describing the transition is the same for the above two possibilities. The order parameter dependence to fourth order is given by

$$F_\Delta = a\left(\Delta_A^*\Delta_A + \Delta_B^*\Delta_B\right) + \beta_1^0\left(\Delta_A^*\Delta_A + \Delta_B^*\Delta_B\right)^2 + \frac{\beta_2^0}{2}\left((\Delta_A^*)^2\Delta_B^2 + \text{h.c.}\right) + \beta_3^0\Delta_A^*\Delta_A\Delta_B^*\Delta_B. \quad (10)$$

The elastic energy associated with the relevant strains is

$$F_u = \frac{1}{2}c_{11}\left(u_{xx}^2 + u_{yy}^2\right) + c_{12}u_{xx}u_{yy} + 2c_{66}u_{xy}^2 + \frac{1}{2}c_{33}u_{zz}^2 + c_{13}\left(u_{xx} + u_{yy}\right)u_{zz}, \quad (11)$$

where $c$'s are the elastic constants. The symmetry allowed terms to linear order in strain are

$$F_{\Delta-u} = \left[\alpha_1(u_{xx} + u_{yy}) + \alpha_2 u_{zz}\right]\left(\Delta_A^*\Delta_A + \Delta_B^*\Delta_B\right) + \alpha_3(u_{xx} - u_{yy})(\Delta_A^*\Delta_A - \Delta_B^*\Delta_B) + \alpha_4 u_{xy}(\Delta_A^*\Delta_B + \Delta_B^*\Delta_A). \quad (12)$$

The overall free energy of the system is

$$F = F_\Delta + F_u + F_{\Delta-u}. \quad (13)$$

We define $u_1 \equiv u_{xx} + u_{yy}$ (strain describing changes to basal plane area of the unit cell), $u_2 \equiv u_{zz}$ (strain related to $c$-axis length changes), $u_3 = u_{xx} - u_{yy}$ (orthorhombic shear) and $u_4 \equiv u_{xy}$ (monoclinic shear). We also define $c_A \equiv (c_{11} + c_{12})/2$ and $c_O \equiv (c_{11} - c_{12})/2$. It is convenient to rewrite the part of the free energy involving the longitudinal strains in a diagonal form by means of a unitary transformation as

$$(F_u)_{\text{long}} \equiv \frac{1}{2}c_A u_1^2 + \frac{1}{2}c_{33}u_2^2 + c_{13}u_1u_2 = \frac{1}{2}D_1v_1^2 + \frac{1}{2}D_2v_2^2. \quad (14)$$

In the above

$$D_{1,2} = \frac{1}{2}\left[c_A + c_{33} \pm \sqrt{(c_A - c_{33})^2 + 4c_{13}^2}\right] \quad (15)$$

are the eigenvalues of the $2 \times 2$ matrix $((c_A, c_{13}), (c_{13}, c_{33}))$, and $(v_1, v_2)$ are the longitudinal eigenmodes given by

$$v_1 = e_1u_1 + e_2u_2, \qquad v_2 = -e_2u_1 + e_1u_2, \quad (16)$$

with $e_1 \equiv c_{13}/N$, $e_2 \equiv (D_1 - c_A)/N$, $N = [c_{13}^2 + (D_1 - c_A)^2]^{1/2}$. Also, the couplings $(\alpha_1, \alpha_2)$ need to be transformed as $(\alpha_1, \alpha_2) \to (r_1, r_2)$ with

$$r_1 = e_1\alpha_1 + e_2\alpha_2, \qquad r_2 = -e_2\alpha_1 + e_1\alpha_2. \quad (17)$$

The two complex valued order parameters can be written as $(\Delta_A, \Delta_B) = \Delta(\cos\theta, e^{i\gamma}\sin\theta)$. The total free energy now has the form

$$F(\Delta,\theta,\gamma,v_1,v_2,u_3,u_4) = a\Delta^2 + \left[4\beta_1^0 + \sin^2 2\theta\left(\beta_2^0\cos 2\gamma + \beta_3^0\right)\right]\frac{\Delta^4}{4} + \frac{1}{2}D_1 v_1^2 + \frac{1}{2}D_2 v_2^2 + \frac{1}{2}c_O u_3^2 + 2c_{66}u_4^2 + \left(r_1 v_1 + r_2 v_2\right)\Delta^2 + \alpha_3 u_3\Delta^2\cos 2\theta + \alpha_4 u_4\Delta^2\sin 2\theta\cos\gamma. \quad (18)$$

As usual, we take $a = a'(T - T_c)$, and the remaining parameters are $T$-independent. The above free energy is to be minimized with respect to the variables $(\Delta,\theta,\gamma,v_1,v_2,u_3,u_4)$. This results in the following equations.

$$2\Delta\left[a + 2\beta_1^0\Delta^2 + (\beta_2^0/2)\Delta^2\sin^2 2\theta\cos 2\gamma + (\beta_3^0/2)\Delta^2\sin^2 2\theta + r_1 v_1 + r_2 v_2 + \alpha_3 u_3\cos 2\theta + \alpha_4 u_4\sin 2\theta\cos\gamma\right] = 0, \quad (19)$$

$$\Delta^2\left[\Delta^2\sin 4\theta\left(\beta_2^0\cos 2\gamma + \beta_3^0\right)/2 - 2\alpha_3 u_3\sin 2\theta + 2\alpha_4 u_4\cos 2\theta\cos\gamma\right] = 0, \quad (20)$$

$$\left(\beta_2^0\Delta^2\sin 2\theta\cos\gamma + \alpha_4 u_4\right)\Delta^2\sin 2\theta\sin\gamma = 0, \quad (21)$$

$$\frac{\partial F}{\partial v_1} = D_1 v_1 + r_1\Delta^2 = 0, \quad (22)$$

$$\frac{\partial F}{\partial v_2} = D_2 v_2 + r_2\Delta^2 = 0, \quad (23)$$

$$\frac{\partial F}{\partial u_3} = c_O u_3 + \alpha_3\Delta^2\cos 2\theta = 0, \quad (24)$$

$$\frac{\partial F}{\partial u_4} = 4c_{66}u_4 + \alpha_4\Delta^2\sin 2\theta\cos\gamma = 0. \quad (25)$$

### 7.1. Phase diagram

From Eqs. (22) - (25) we get

$$v_1 = r_1\Delta^2/D_1, \quad v_2 = r_2\Delta^2/D_2,$$
$$u_3 = \alpha_3\Delta^2\cos 2\theta/c_O,$$
$$u_4 = \alpha_4\Delta^2\sin 2\theta\cos\gamma/(4c_{66}).$$

This leads to a renormalization of the fourth order coefficients $\beta_i^0 \to \beta_i$ with

$$\beta_1 = \beta_1^0 - (r_1^2/D_1 + r_2^2/D_2 + \alpha_3^2/c_O)/2, \quad (26a)$$
$$\beta_2 = \beta_2^0 - \alpha_4^2/(4c_{66}), \quad (26b)$$
$$\beta_3 = \beta_3^0 - \alpha_4^2/(4c_{66}) + 2\alpha_3^3/c_O. \quad (26c)$$

Note, the combination

$$r_1^2/D_1 + r_2^2/D_2 = \frac{\alpha_1^2 c_{33} + \alpha_2^2 c_A - 2\alpha_1\alpha_2 c_{13}}{c_A c_{33} - c_{13}^2}. \quad (27)$$

In terms of the renormalized fourth order coefficients Eqs. (19), (20) and (21) can be rewritten as

$$2\Delta\left[a + 2\beta_1\Delta^2 + \frac{1}{2}\beta_2\Delta^2\sin^2 2\theta\cos 2\gamma + \frac{1}{2}\beta_3\Delta^2\sin^2 2\theta\right] = 0, \quad (28)$$

$$\left(\beta_2\cos 2\gamma + \beta_3\right)\Delta^4\sin 2\theta\cos 2\theta = 0, \quad (29)$$

$$\beta_2\Delta^4\sin^2 2\theta\sin 2\gamma = 0. \quad (30)$$

For the stability of the system we need $\beta_1 > 0$, and $4\beta_1 \pm \beta_2 + \beta_3 > 0$. Within this range the following three superconducting phases are possible.

(1) In the region $\beta_2 > (0, \beta_3)$ we get $\Delta = \Delta_0 \equiv [-2a/(4\beta_1 - \beta_2 + \beta_3)]^{1/2}$ , $\theta = \theta_0 \equiv \pi/4$ and $\gamma = \gamma_0 \equiv \pm\pi/2$. Thus, $(\Delta_A, \Delta_B) = \Delta_0(1, \pm i)$, and it is the time reversal symmetry broken chiral state. The phase transition is accompanied by finite longitudinal strains $v_1^0 = -r_1\Delta_0^2/D_1$ and $v_2^0 = -r_2\Delta_0^2/D_2$, while the shear strains are zero. Thus, the tetragonal symmetry is preserved.

(2) In the region $\beta_2 < (0, -\beta_3)$ we get $\Delta = \Delta_0 \equiv [-2a/(4\beta_1 + \beta_2 + \beta_3)]^{1/2}$ , $\theta = \theta_0 \equiv \pi/4$ and $\gamma = \gamma_0 \equiv (0, \pi)$. Thus, $(\Delta_A, \Delta_B) = \Delta_0(1, \pm 1)$, and it is a phase that preserves time reversal symmetry. As in case (1), the phase transition is accompanied by finite longitudinal strains $v_1^0 = -r_1\Delta_0^2/D_1$ and $v_2^0 = -r_2\Delta_0^2/D_2$. But, unlike in case (1), now the transition is accompanied by a spontaneous monoclinic distortion $u_4^0 = -\alpha_4\Delta_0^2/(4c_{66})$. Thus the state breaks the tetragonal symmetry spontaneously. On the other hand, there is no spontaneous orthorhombic distortion, i.e., $u_3^0 = 0$.

(3) In the region $\beta_3 > (0, |\beta_2|)$ we get $\Delta = \Delta_0 \equiv [-a/(2\beta_1)]^{1/2}$ , $\theta = \theta_0 \equiv (0, \pi/2)$ and $\gamma = \gamma_0$, where $\gamma_0 \equiv 0$ for $\beta_2 < 0$ and $\gamma_0 \equiv \pi/2$ for $\beta_2 > 0$. Note, $\gamma$ is a meaningful variable only if $\theta$ is non-zero (say, in the presence of external strain, or if nonzero fluctuations of $\theta$ are relevant. Thus, $(\Delta_A, \Delta_B) = \Delta_0(0, 1)$ or equivalently $\Delta_0(1, 0)$, and it is a phase that preserves time reversal symmetry as well. The spontaneous strains generated in this phase are $v_1^0 = -r_1\Delta_0^2/D_1$, $v_2^0 = -r_2\Delta_0^2/D_2$, $u_3^0 = -\alpha_3\Delta_0^2/c_O$, and $u_4^0 = 0$. Thus, this state also breaks tetragonal symmetry spontaneously and the transition is accompanied by finite orthorhombic distortion.

### 7.2. Jumps in elastic constants in the phase $(\Delta_A, \Delta_B) = \Delta_0(1, \pm i)$

(a) In order calculate the jump in $c_{66}$ we consider a finite external monoclinic stress $\sigma_4$ such that Eqn (25) is replaced by

$$\frac{\partial F}{\partial u_4} = 4c_{66}u_4 + \alpha_4\Delta^2 \sin 2\theta \cos\gamma = \sigma_4, \tag{31}$$

while all the other Eqns from minimizing $F$ remain the same as before. From Eqn (21) we get

$$\beta_2^0\Delta^2 \sin 2\theta \cos\gamma + \alpha_4 u_4 = 0.$$

Using the above two eqns we deduce that $u_4 = \sigma_4/(4c_{66} - \alpha_4^2/\beta_2^0)$. On the other hand in the metallic phase ($\Delta = 0$) we would have obtained $u_4 = \sigma_4/(4c_{66})$. Thus, the jump in $c_{66}$ is given by

$$\delta c_{66} = \frac{-\alpha_4^2}{4\beta_2^0} = \frac{-\alpha_4^2}{4\beta_2 + \alpha_4^2/c_{66}}. \tag{32}$$

(b) To calculate the jump in $c_O$ we consider a finite external orthorhombic stress $\sigma_3$ such that Eq. (24) is replaced by

$$\frac{\partial F}{\partial u_3} = c_O u_3 + \alpha_3\Delta^2 \cos 2\theta = \sigma_3, \tag{33}$$

while the remaining equations are unchanged. From Eq. (21) we deduce that $\gamma = \pi/2$, and that $u_4 = 0$. Putting this back in Eq. (20) we get

$$\Delta^2 \cos 2\theta = \frac{-2\alpha_3 u_3}{\beta_2^0 - \beta_3^0}. \tag{34}$$

From the above two equations we get $u_3 = \sigma_3/[c_O - 2\alpha_3^2/(\beta_2^0 - \beta_3^0)]$. This implies that the jump is

$$\delta c_O = \frac{-2\alpha_3^2}{\beta_2^0 - \beta_3^0} = \frac{-2\alpha_3^2}{\beta_2 - \beta_3 + 2\alpha_3^2/c_O}. \tag{35}$$

(c) To calculate the jump in $D_1$ we consider an external longitudinal stress $\sigma_1$ that couples to $v_1$. Eq. (22) is modified to

$$\frac{\partial F}{\partial v_1} = D_1 v_1 + r_1\Delta^2 = \sigma_1. \tag{36}$$

From Eq. (21) we get that $\gamma = \pi/2$ and from Eq. (20) we get that $\theta = \pi/4$. These also imply that $(u_3, u_4) = 0$. Using these values in Eq. (19) we get $\Delta^2(T_c^-) = -2r_1v_1/[4\beta_1^0 - \beta_2^0 + \beta_3^0 - 2r_2^2/D_2]$. Here $T_c^-$ implies approaching $T_c$ from below, and for which $a = 0$. using this in the above equation we deduce that the jump in $D_1$ is

$$\begin{aligned}\delta D_1 &= -2r_1^2/(4\beta_1^0 - \beta_2^0 + \beta_3^0 - 2r_2^2/D_2) \\ &= -2r_1^2/(4\beta_1 - \beta_2 + \beta_3 + 2r_1^2/D_1).\end{aligned} \tag{37}$$

(d) From a very similar calculation we get that the jump in $D_2$ is

$$\begin{aligned}\delta D_2 &= -2r_2^2/(4\beta_1^0 - \beta_2^0 + \beta_3^0 - 2r_1^2/D_1) \\ &= -2r_2^2/(4\beta_1 - \beta_2 + \beta_3 + 2r_2^2/D_2).\end{aligned} \tag{38}$$

From the relations

$$c_A = e_1^2 D_1 + e_2^2 D_2, \qquad c_{33} = e_2^2 D_1 + e_1^2 D_2, \tag{39}$$

we can calculate $\delta c_A = e_1^2\delta D_1 + e_2^2\delta D_2$, and $\delta c_{33} = e_2^2\delta D_1 + e_1^2\delta D_2$.

### 7.3. Jumps in elastic constants in the phase $(\Delta_A, \Delta_B) = \Delta_0(1, \pm 1)$

(a) To calculate the jump in $c_{66}$ we consider a finite external monoclinic stress $\sigma_4$ such that Eq. (25) is replaced by Eq. (31). From Eq. (20) we deduce that $\theta = \pi/4$, from Eq. (21) $\gamma = 0$, and from Eq. (24) $u_3 = 0$. Using these values in Eq. (19) we get $\Delta^2(T_c^-) = -2\alpha_4 u_4/[4\beta_1^0 + \beta_2^0 + \beta_3^0 - 2r_1^2/D_1 - 2r_2^2/D_2]$. Using this in Eq. (31) we get

$$\begin{aligned}\delta c_{66} &= \frac{-\alpha_4^2/2}{4\beta_1^0 + \beta_2^0 + \beta_3^0 - 2r_1^2/D_1 - 2r_2^2/D_2} \\ &= \frac{-\alpha_4^2/2}{4\beta_1 + \beta_2 + \beta_3 + \alpha_4^2/(2c_{66})}.\end{aligned} \tag{40}$$

(b) To calculate the jump in $c_O$ we consider a finite external orthorhombic stress $\sigma_3$ such that Eq. (24) is replaced by Eq. (33). From Eq. (21) we get $\gamma = 0$, while Eq. (20) gives

$$\Delta^2 \sin 2\theta \left[\Delta^2 \cos 2\theta \left(\beta_2^0 + \beta_3^0 - \alpha_4^2/(2c_{66}\right) - 2\alpha_3 u_3\right] = 0.$$

Since $\Delta \neq 0$, and $\sin 2\theta \neq 0$, we get $\Delta^2 \cos 2\theta = -2\alpha_3 u_3/(|\beta_2^0| - \beta_3^0 + \alpha_4^2/(2c_{66}))$. Using this in Eq. (33) we get the jump to be

$$\delta c_O = \frac{-2\alpha_3^2}{|\beta_2^0| - \beta_3^0 + \alpha_4^2/(2c_{66})} = \frac{-2\alpha_3^2}{|\beta_2| - \beta_3 + 2\alpha_3^2/c_O}. \tag{41}$$

(c) To calculate the jump in $D_1$ we consider an external longitudinal stress $\sigma_1$ that couples to $v_1$. Eq. (22) is modified to Eq. (36). From Eq. (20) we deduce that $\theta = \pi/4$, from Eq. (21) $\gamma = 0$, and from Eq. (24) $u_3 = 0$. Using these values in Eq. (19) we get $\Delta^2(T_c^-) = -2r_1v_1/[4\beta_1^0 + \beta_2^0 + \beta_3^0 - 2r_2^2/D_2 - \alpha_4^2/(2c_{66})]$. Thus, the jump is

$$\begin{aligned}\delta D_1 &= -2r_1^2/(4\beta_1^0 + \beta_2^0 + \beta_3^0 - 2r_2^2/D_2 - \alpha_4^2/(2c_{66})) \\ &= -2r_1^2/(4\beta_1 + \beta_2 + \beta_3 + 2r_1^2/D_1).\end{aligned} \tag{42}$$

(d) A similar calculation gives

$$\begin{aligned}\delta D_2 &= -2r_2^2/(4\beta_1^0 + \beta_2^0 + \beta_3^0 - 2r_1^2/D_1 - \alpha_4^2/(2c_{66})) \\ &= -2r_2^2/(4\beta_1 + \beta_2 + \beta_3 + 2r_2^2/D_2).\end{aligned} \tag{43}$$

As before, the jumps $(\delta c_A, \delta c_{33})$ can be evaluated using Eq.(39).

### 7.4. Jumps in elastic constants in the phase $(\Delta_A, \Delta_B) = \Delta_0(1,0)$ or $\Delta_0(0,1)$

(a) To calculate the jump in $c_{66}$ we consider a finite external monoclinic stress $\sigma_4$ such that Eq. (25) is replaced by Eq. (31). Note, a priori, Eq. (21) has three possible solutions. It is simple to check that the solution $\beta_2^0\Delta^2\sin 2\theta\cos\gamma + \alpha_4 u_4 = 0$ leads to unphysical solution. Then, either (i) $\theta = 0$, which also leads to $\gamma = \pi/2$, or (ii) $\gamma = 0$ and $\theta \neq 0$. A bit of algebra shows that the solution (ii) has lower free energy, and therefore is the correct choice. From Eq. (20) we get $\Delta^2\sin 2\theta = -2\alpha_4 u_4/(\beta_2^0 + \beta_3^0 + 2\alpha_3^2/c_O)$. This, along with Eq. (31) implies that the jump is

$$\delta c_{66} = \frac{-\alpha_4^2/2}{\beta_2^0 + \beta_3^0 + 2\alpha_3^2/c_O} = \frac{-\alpha_4^2/2}{\beta_2 + \beta_3 + \alpha_4^2/(2c_{66})}. \quad (44)$$

(b) To calculate the jump in $c_O$ we consider a finite external orthorhombic stress $\sigma_3$ such that Eq. (24) is replaced by Eq. (33). From Eq. (25) we get $\theta = 0$, while Eq. (25) gives $u_4 = 0$. From Eq. (19) we conclude that $\Delta^2(T_c^-) = -\alpha_3 u_3/(2\beta_1^0 - r_1^2/D_1 - r_2^2/D_2)$. Thus, the jump is

$$\delta c_O = \frac{-\alpha_3^2}{2\beta_1^0 - r_1^2/D_1 - r_2^2/D_2} = \frac{-\alpha_3^2}{2\beta_1 + \alpha_3^2/c_O}. \quad (45)$$

(c) A similar calculation gives the jump

$$\delta D_1 = \frac{-r_1^2}{2\beta_1^0 - r_2^2/D_2 - \alpha_3^2/c_O} = \frac{-r_1^2}{2\beta_1 + r_1^2/D_1}. \quad (46)$$

(d) Likewise, the jump in $D_2$ is given by

$$\delta D_2 = \frac{-r_2^2}{2\beta_1^0 - r_1^2/D_1 - \alpha_3^2/c_O} = \frac{-r_2^2}{2\beta_1 + r_2^2/D_2}. \quad (47)$$

As before, the jumps $(\delta c_A, \delta c_{33})$ can be evaluated using Eq.(39).

## 8. EFFECT OF UNIAXIAL STRAIN AT QUADRATIC ORDER ON $T_c$

In this section we study the effect of uniaxial strain $\epsilon_{(100)}$ along the $(1,0,0)$ direction and how it modifies $T_c$ at order $\epsilon_{(100)}^2$ within the scenario of a two-component order parameter belonging to the $E$ irreducible representation.

We consider the external uniaxial stress $\sigma_{xx} = \sigma$, which couples to the strain $u_{xx} = (u_1 + u_3)/2$. Following the notation of the last Section, $u_1 \equiv (u_{xx} + u_{yy})$ is the in-plane $A_{1g}$ longitudinal strain, and $u_3 \equiv (u_{xx} - u_{yy})$ is the in-plane $B_{1g}$ shear strain. To simplify the discussion we ignore the elastic constant $c_{13}$ in Eq. (11), and write the elastic free energy of the above two in-plane modes as

$$F_{u,plane} = \frac{1}{2}c_A u_1^2 + \frac{1}{2}c_O u_3^2 - \frac{\sigma}{2}(u_1 + u_3).$$

As defined in the last Section, $c_A = (c_{11} + c_{12})/2$ and $c_O = (c_{11} + c_{12})/2$. Minimizing $F_{u,plane}$ we get $u_1 = \sigma/(2c_A)$ and $u_3 = \sigma/(2c_O)$ For $Sr_2RuO_4$ the relevant elastic constants are $c_{11} = 233$ GPa and $c_O = 51$ GPa [1], which implies $c_A \approx 3.5c_O$. Using this estimate we get

$$u_1 = \frac{4}{9}\epsilon_{(100)}, \quad u_3 = \frac{14}{9}\epsilon_{(100)}. \quad (48)$$

In other words, the uniaxial strain is not a pure $B_{1g}$ shear, but has a non-negligible $A_{1g}$ component.

The coupling of the superconducting variables to quadratic order in the strains $(u_1, u_3)$ can be written as

$$F_{\Delta-u^2} = \frac{1}{2}(\lambda_{11}u_1^2 + \lambda_{33}u_3^2)(\Delta_A^*\Delta_A + \Delta_B^*\Delta_B) + \lambda_{13}u_1u_3(\Delta_A^*\Delta_A - \Delta_B^*\Delta_B). \quad (49)$$

In the above, the first line describes an $A_{1g}$ perturbation proportional to $\epsilon_{(100)}^2$, and the second line describes a $B_{1g}$ perturbation also proportional to $\epsilon_{(100)}^2$. To simplify the discussion we take $\lambda_{11} = \lambda_{33} = \lambda_{13} = -\lambda$, with $\lambda > 0$. Note, taking a negative $\lambda$ leads to decrease in $T_c$ as a function of $\epsilon_{(100)}^2$, which is opposite to what is observed. For convenience we define

$$p \equiv \lambda u_1 u_3 = (56/81)\lambda\epsilon_{(100)}^2. \quad (50)$$

Then, $\lambda(u_1^2 + u_3^2)/2 \approx 2p$. Since linear strain variation of $T_c$ has not been observed, we can ignore $F_{\Delta-u}$. Writing $F = F_\Delta + F_{\Delta-u^2}$ we get

$$F(\Delta,\theta,\gamma) = (a - 2p - p\cos 2\theta)\Delta^2 + \left[4\beta_1 + \sin^2 2\theta\,(\beta_2\cos 2\gamma + \beta_3)\right]\frac{\Delta^4}{4}. \quad (51)$$

We take $a = a'(T - T_c^0)$, where $T_c^0$ is the transition temperature in the absence of external strain. The above free energy is to be minimized with respect to $(\Delta, \theta, \gamma)$.

To be concrete we first assume that $\beta_i$ are such that the ground state is the $(\Delta_A, \Delta_B) = \Delta_0(1, \pm i)$ phase. In this case one can show that there are two split transitions at temperatures $(T_{c1}, T_{c2})$. Lowering $T$ the system first undergoes the $U(1)$ symmetry breaking superconducting transition at

$$T_{c1} = T_c^0 + 3(p/a'). \quad (52)$$

For $T_{c1} > T > T_{c2}$ the $B_{1g}$ component of the $\epsilon_{(100)}^2$ perturbation stabilizes the $(1,0)$ state characterized by $\theta = 0$. The second transition, where time reversal symmetry is broken, occurs at

$$T_{c2} = T_c^0 + (2 - \eta)(p/a'), \quad (53)$$

where $\eta \equiv 4\beta_1/(\beta_2 - \beta_3) - 1 > 0$. Below $T_{c2}$ the phase is characterized by $\theta \neq 0$ and $\gamma = \pi/2$.

Thus, both the increase of the superconducting transition $(T_{c1} - T_c^0)$ and the split between the two transitions $(T_{c1} - T_{c2})$ are of comparable magnitudes if we assume that the ground state is the $(1, \pm i)$ phase. Analogously, the same is true also if we assume that the ground state is the $(1, \pm 1)$ phase. On the other hand, if the ground state is the $(1, 0)$ [or equivalently the $(0, 1)$] phase, then there is a single transition at the enhanced temperature $T_{c1}$. Experimentally, only the $T_c$ enhancement proportional to $\epsilon^2_{(100)}$ has been reported [5], but the splitting between the two transitions has not been seen in thermodynamic measurements. This observation is consistent with the $(1, 0)$ phase.

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