Jump in the $c_{66}$ shear modulus at the superconducting transition of Sr$_2$RuO$_4$: Evidence for a two-component order parameter

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The quasi-2D metal Sr$_2$RuO$_4$ is one of the best characterized unconventional superconductors, yet the nature of its superconducting order parameter is still highly debated. Here we use ultrasound velocity to probe the superconducting state of Sr$_2$RuO$_4$. We observe a sharp jump in the shear elastic constant $c_{66}$ as the temperature is raised across the superconducting transition at $T_c$. This directly implies that the superconducting order parameter is of a two-component nature. We discuss what states are compatible with this requirement and propose, given the other known properties of Sr$_2$RuO$_4$, that the most likely candidate is the $(1, 0)$ state of the $E_u$ representation.

I. INTRODUCTION

For 25 years, superconductivity of Sr$_2$RuO$_4$ has been viewed as an electronic analog of superfluid $^3$He [1–3]. The initial report of the temperature independent spin susceptibility through $T_c$ [4] and the indication of time-reversal symmetry breaking [5–8] pointed to a chiral $p$-wave order parameter, $d = z(k_x \pm ik_y)$. However, several experiments are in contradiction with this scenario [7], for example the lack of edge currents [8], the Pauli limiting critical field [9] and the absence of a cusp in the dependence of $T_c$ on uniaxial strain [10]. Importantly, evidence of line nodes in the gap from specific heat [11], ultrasound attenuation [23] and thermal conductivity [12, 13] is not compatible with a chiral $p$-wave order parameter. Recently, measurements of the NMR Knight shift were carefully revisited and a clear drop in the spin susceptibility below $T_c$ was detected [14], pointing to an order parameter with even parity. As a result, the nature of the superconducting state in Sr$_2$RuO$_4$ is now a wide open question but the chiral $p$-wave order parameter and any odd-parity order with an out-of-plane $d$ vector are excluded.

Here we present a study of ultrasound propagation in Sr$_2$RuO$_4$ that will allow us to rule out all the candidate states that are characterized by a one-component order parameter.

II. EXPERIMENTAL

| $T$ | Basis function | Strain component | Elastic constant |
|-----|----------------|-----------------|-----------------|
| $A_{1g}$ | $a(k_x^2 + k_y^2) + bk_x^2$ | $\epsilon_{xx} + \epsilon_{yy}$, $\epsilon_{xx}$ | $(c_{11} + c_{12})/2$, $c_{33}$ |
| $A_{2g}$ | $k_x k_y (k_x^2 - k_y^2)$ | none | none |
| $B_{1g}$ | $k_x^2 - k_y^2$ | $\epsilon_{xx} + \epsilon_{yy}$ | $(c_{11} - c_{12})/2$ |
| $B_{2g}$ | $k_x k_y$ | $\epsilon_{yy}$ | $c_{66}$ |
| $E_u$ | $k_x k_z, k_y k_z$ | $\epsilon_{zz}$, $\epsilon_{xy}$ | $c_{44}$ |

TABLE I. Irreducible representation of the strain tensor for the D$_{4h}$ point group.

Sound velocity is a powerful thermodynamic probe for order parameter. For propagation along high-symmetry directions of the crystal, the sound velocity is $v_s = \sqrt{c_{ij}/\rho}$ where $\rho$ is the density of the material and $c_{ij}$ are the elastic constants defined as the second derivative of the free energy $F$ with respect to the strain $u_{ij}$. In the framework of Landau-Ginzburg theory of phase transition, a discontinuity in the elastic constant at the superconducting transition is a consequence of the symmetry allowed
coupling term between the order parameter $\Delta$ and the strain, $\lambda|\Delta|^2u$, where $\lambda$ is a coupling constant (see S.I.) [14]. As part of the free energy, this coupling term is invariant under all the operations of the point group, i.e. it belongs to the $A_{1g}$ representation. Table I lists the irreducible strains corresponding to the point group $D_{4h}$ for the tetragonal symmetry of Sr$_2$RuO$_4$ (see the corresponding product table in the S.I.). If the superconducting order parameter is one-component, then $|\Delta|^2$ belongs to the $A_{1g}$ representation. Consequently, the strain variable $u$ can only belong to the $A_{1g}$ representation, i.e. it corresponds to a longitudinal sound wave. A jump in the longitudinal elastic constant is observed at $T_c$ in many superconductors and is directly related to the jump in the specific heat at $T_c$ and the strain dependence of $T_c$ via the Ehrenfest relation [10]. If an unusual jump in the elastic constant associated with a shear mode ($B_{1g}$ or $B_{2g}$ representation) is detected at $T_c$, then it necessarily implies that the superconducting order parameter is multi-dimensional [17]. Based on these symmetry arguments that are further developed in this paper, we have performed measurements of longitudinal and transverse sound velocities in Sr$_2$RuO$_4$ across the superconducting transition down to 40 mK. The measurements were performed using a pulse-echo technique with two different home built spectrometers and commercial LiNbO$_3$ transducers, bonded to the crystals (see Methods in the S.I.).

### III. RESULTS

| Elastic constant | $k$  | $p$  | Sound velocity ($\text{km/s}$) | Value (GPa) |
|------------------|------|------|-------------------------------|-------------|
| $c_{11}$         | [100]| [100]| 6.28                          | 233         |
| $c_{44}$         | [100]| [001]| 3.41                          | 68.2        |
| $c_{66}$         | [100]| [010]| 3.3                           | 64.3        |
| ($c_{11}-c_{12}$)/2 | [110]| [100]| 2.94                          | 51          |

**TABLE II.** Definition of the different sound modes measured at $T = 4$ K. $k$ and $p$ stand for the propagation and polarization direction, respectively. Sound velocities were obtained at low temperature using the echo spacing.

Table II shows the different acoustic modes with the directions of sound propagation and polarization of the transducer. The value of the sound velocity is obtained from the echo spacing at low temperature ($T = 4$ K) and can be converted to elastic constants using $\rho = 5.95 \text{ g/cm}^3$. They are in good agreement with resonant ultrasound spectroscopy measurements [18][20]. Fig. 1a and Fig. 1b show the temperature dependence of the sound velocity for the longitudinal mode $c_{11}$ and the transverse mode ($c_{11}-c_{12}$)/2, respectively. Red circles (open squares) correspond to measurement in the superconducting (normal) state. Fig. 1c and Fig. 1d show the difference between the superconducting state and the normal state for the two modes. A discontinuity is expected at $T_c$ for the longitudinal mode, $c_{11}$. From the Ehrenfest relation, we estimate the magnitude of this drop to be $\Delta c_{11}/c_{11} \approx 4$ ppm (see S.I.). This small discontinuity is thus hidden by the strong softening of the longitudinal constant in the superconducting state ($\approx 80$ ppm between $T_c$ and $T \to 0$). A similar, but even stronger softening is observed for the transverse mode ($c_{11}-c_{12}$)/2, below $T_c$ (Fig. 1b). These results are qualitatively in agreement with previous measurements [21][22] (but the absolute value of their elastic constants differs from ours [?])

![Fig. 1](image)

**Fig. 1.** Relative change in sound velocity for a) the longitudinal mode $c_{11}$ measured at $F = 83$ MHz, b) the transverse mode ($c_{11}-c_{12}$)/2 measured at $F = 21.5$ MHz. The normal state data (open squares) are obtained by applying a magnetic field of 1.5 T in the plane, larger than $H_{c2}$. The superconducting state data (red circles) are measured without any applied field. c) Difference between the superconducting state and the normal state, for the $c_{11}$ mode. d) Same, for the ($c_{11}-c_{12}$)/2 mode.

Fig. 2a shows the temperature dependence of the sound velocity for the transverse mode, $c_{66}$. The measurements in the superconducting state ($H = 0$, red circles) display a sharp discontinuity at the superconducting transition. The difference in the shear sound velocity between the normal and superconducting states (Fig. 2b) shows a small but very clear jump at $T_c$, of magnitude $\approx 0.2$ ppm, 10 times larger than our experimental resolution. The exceptional sensitivity of our experiment is due to the very small attenuation of the $c_{66}$ mode [23], which enabled us to detect up to 70 echoes (see SI) and to perform a fit on all of them. Note that we have reproduced the data for the $c_{66}$ mode using another experimental setup (see S.I.). A discontinuity of the sound velocity of the $c_{66}$ mode has also been detected by resonant ultrasound spectroscopy at $T_c$ [20].

This is the key finding of our study: we observe a sharp discontinuity at $T_c$ in the sound velocity for the transverse mode $c_{66}$. This immediately provides unambiguous evidence that the superconducting order parameter must be of a two-component nature, consistent only with the
The superconducting part, expanded to fourth order, is

\[
F_\Delta = a \left( |\Delta_A|^2 + |\Delta_B|^2 \right) + \beta_0 \left( |\Delta_A|^2 + |\Delta_B|^2 \right)^2 \\
+ \frac{\beta_0^2}{2} \left( (\Delta_A^*)^2 \Delta_B^2 + c.c. \right) + \beta_3 |\Delta_A|^2 |\Delta_B|^2.
\]

The relevant elastic energy of the uniform strains is

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

where \(c\)'s are the elastic constants in Voigt notation. The cross-coupling term is

\[
F_{\Delta - u} = [\alpha_1 (u_{xx} + u_{yy}) + \alpha_2 u_{zz}] (|\Delta_A|^2 + |\Delta_B|^2) \\
+ \alpha_3 (u_{xx} - u_{yy}) (|\Delta_A|^2 - |\Delta_B|^2) \\
+ \alpha_4 u_{xy} \Delta_u \Delta_B + c.c.
\]

The analysis of the above free energy is standard, and is described in detail in the SI. Here we quote the main results.

For convenience we define \(c_A \equiv (c_{11} + c_{12})/2\) and \(c_O \equiv (c_{11} - c_{12})/2\). The latter is the orthorhombic elastic constant associated with the shear mode \(u_{xx} - u_{yy}\), while \(c_{66}\) is the elastic constant of the monoclinic shear \(u_{xy}\). Our aim is to calculate the jumps in the shear elastic constants defined by \(\delta c = c(T^-) - c(T^+)\).

The term \(F_{\Delta - u}\) renormalizes the fourth order coefficients \(\beta_i \rightarrow \beta_i^f\) with

\[
\beta_1 = \beta_0^f - \frac{1}{2} \left[ \frac{\alpha_3^2}{c_O} + \frac{\alpha_1^2 c_{33} + \alpha_2^2 c_A - 2 \alpha_1 \alpha_2 c_{13}}{c_A c_{33} - c_{13}} \right],
\]

\[
\beta_2 = \beta_0^f - \frac{\alpha_1}{4 c_{66}}
\]

\[
\beta_3 = \beta_0^f - \frac{\alpha_1^2}{4 c_{66}} + 2 \alpha_3^2/c_O.
\]

For the stability of the system, we need \(\beta_1 > 0\), and \(4 \beta_1 \pm \beta_2 + \beta_3 > 0\). Within these ranges the following three superconducting phases are possible.

(1) **Time reversal symmetry broken superconductor:** In the region \(\beta_2 > (0, \beta_3)\), we get the time reversal symmetry broken state with \((\Delta_A, \Delta_B) = \Delta_0(1, \pm i)\). In this phase, there is no spontaneous shear strain, and the tetragonal symmetry is preserved. The shear moduli jumps are

\[
\delta c_{66} = \frac{-\alpha_3^2}{4 \beta_2 + \alpha_1^2/c_{66}},
\]

\[
\delta c_O = \frac{-2 \alpha_3^2}{\beta_2 - \beta_3 + 2 \alpha_3^2/c_O}.
\]

(2) **Nematic-monoclinic superconductor:** In the region \(\beta_2 < (0, -\beta_3)\), we get a nematic solution, \((\Delta_A, \Delta_B) = \Delta_0(1, \pm 1)\), which breaks the tetragonal symmetry by making the two in-plane diagonal directions inequivalent.
It is accompanied by a spontaneous monoclinic strain, i.e. $u_{xy} \neq 0$. The shear moduli jumps are

$$\delta c_{66} = -\frac{-\alpha_4^2/2}{\beta_1 + \beta_2 + \beta_3 + \alpha_4^2/(2c_{66})},$$  \hspace{1cm} (3a)$$
$$\delta c_{CO} = -\frac{-2\alpha_3^2}{\beta_2 - \beta_3 + 2\alpha_3^2/c_{CO}}. \hspace{1cm} (3b)$$

(3) Nematic-orthorhombic superconductor: In the region $\beta_3 > (0, |\beta_2|)$, we also get a nematic solution, $(\Delta_A, \Delta_B) = \Delta_0(0, 1)$, or equivalently $\Delta_0(1, 0)$, which also breaks the tetragonal symmetry by making the two in-plane crystallographic axes inequivalent. It is accompanied by a spontaneous orthorhombic strain, i.e. $u_{xx} - u_{yy} \neq 0$. The shear moduli jumps are

$$\delta c_{66} = -\frac{-\alpha_4^2/2}{\beta_2 + \beta_3 + \alpha_4^2/(2c_{66})},$$  \hspace{1cm} (4a)$$
$$\delta c_{CO} = -\frac{-\alpha_3^2}{2\beta_1 + \alpha_3^2/c_{CO}}. \hspace{1cm} (4b)$$

Thus, in all three states the two shear elastic constants, $c_{66}$ and $c_{CO}$, jump at the superconducting transition $T = T_c$. In our data, there is a clear jump in $c_{66}$. However, a jump in $c_{CO}$ could not be resolved, most likely because of the strong temperature dependence of $c_{CO}(T)$ below the transition.

V. DISCUSSION

The observed jump in $c_{66}$ at $T_c$ implies that the superconducting order parameter of Sr$_2$RuO$_4$ is of a two-component nature. We now discuss the various implications of this new constraint, in the context of the other known properties of Sr$_2$RuO$_4$.

(i) Discrete symmetry breaking: In a two-component scenario, the $U(1)$ symmetry breaking superconducting transition is necessarily accompanied by a simultaneous discrete symmetry breaking. For case (1) in section IV, this discrete symmetry is time reversal leading to a spontaneous magnetization that can be detected in a $\mu$SR measurement, for example. A non-zero $\mu$SR signal below $T_c$ has indeed been reported [5], but its origin and implications are currently under investigation [24]. For cases (2) and (3) in section IV, the broken symmetry is tetragonal $D_{4h}$ leading to an orthorhombic or a monoclinic distortion of the tetragonal unit cell, which can in principle be detected through x-ray diffraction. At present, no such distortion has been reported.

(ii) Response to uniaxial pressure: $T_c$ as a function of uniaxial strain $u_{xx} - u_{yy}$ should increase linearly. Experimentally, $T_c$ increases, but not in the linear regime since the cusp at zero strain has not yet been resolved [10]. Next, in the presence of a uniaxial strain $u_{xx} - u_{yy}$, there should be two split transitions if the order parameter is the $(1, 1)$ type. But for $(1, 0)$, one expects a single transition, with enhanced $T_c$. Since split transitions have not been observed in thermodynamic measurements [27], it argues in favor of $(1, 0)$ order parameter.

(iii) Spin-wavevector content of Cooper pairs: The drop of the Knight shift below $T_c$ is strongly suggestive of an even parity order parameter [13, 25]. Assuming only intraband pairing, for singlets, the lowest harmonic is a $d$-wave solution $(\Delta_A, \Delta_B) = \Delta_0(k_x, k_y)$.

(iv) Line nodes: All states that we discuss within the $E_g$ and $E_u$ representations necessarily have horizontal line nodes. Experimentally, thermal conductivity measurements show that the gap has vertical line nodes [13]. Recent quasiparticle interference (QPI) experiments corroborate this conclusion [29] with nodes along the diagonal. Concerning the location of the line nodes, the specific-heat under in-plane magnetic fields is interpreted either in terms of vertical line nodes of the $k_xk_y$-type [30] (i.e. rotated by 45 deg compared to the $d_{x^2-y^2}$ orbital) or horizontal line nodes [31]. In the singlet sector ($E_g$), the states $(1, 0)$ and $(1, 1)$, which break tetragonal symmetry, have vertical line nodes. However, the state $(1, 1)$, which breaks time reversal symmetry, typically does not have vertical line nodes, unless the pairing leads to a Bogoliubov Fermi surface [28]. In the triplet sector ($E_u$), the $p$-wave solutions do not have vertical line nodes. In order to have triplet solutions also consistent with vertical line nodes one needs to consider a higher harmonic $f$-wave solution $(\Delta_A, \Delta_B) = \Delta_0k_z(k^2_x - k^2_y)(d_x, d_y)$.

(v) Accidental degeneracy: Until now, we only considered the two-dimensional irreducible representation $E$. In principle, one can also get a two-dimensional $\Delta$ if two one-dimensional representations become accidentally degenerate. The advantage of such a scenario is that it allows the possibility of having a finite jump in $c_{66}$ while having no jump of $c_{CO}$. Thus the $(s + d)$-wave solution $(\Delta_A, \Delta_B) = (1, k_xk_y)$ will have the same free energy structure as in Eq. (1), except with $\alpha_3 = 0$. However, such a state is not guaranteed to have line nodes. Vertical line nodes are present for accidental degeneracy of higher order harmonics such as the $(d + g)$-wave solution $(\Delta_A, \Delta_B) = (k^2_x - k^2_y)/(1, k_xk_y)$ [32].

VI. SUMMARY

In summary, using high-sensitivity measurements of ultrasound propagation in Sr$_2$RuO$_4$, we observe a sharp jump in the transverse sound velocity at $T_c$ for the $c_{66}$ mode. This is only possible if the superconducting order parameter has two components, implying either a two-component representation ($E_g$ or $E_u$) or the accidental combination of two one-dimensional representations (e.g. $B_{1g} \bigoplus A_{2g}$). We can then add the following requirements from previous experiments: 1) the gap has

\[ \Delta_A, \Delta_B = \Delta_0(k_x, k_y) \]

\[ (\Delta_A, \Delta_B) = \Delta_0k_z(k^2_x - k^2_y)(d_x, d_y) \]

\[ (\Delta_A, \Delta_B) = (k^2_x - k^2_y)/(1, k_xk_y) \]
vertical line nodes (from thermal conductivity [13]; 2) the order parameter has even parity (from NMR [14]); 3) uniaxial strain does no split the superconducting transition (from specific heat under strain [27]. If we limit the additional constraints to those three, then the only possible candidate is the (1,0) state in $E_y$ representation, namely the nematic $k_xk_z$ or $k_yk_z$ with both horizontal and vertical line nodes. This is a nematic state, whose onset will be accompanied by an orthorhombic distortion of the lattice and certainly the formation of nematic domains. This implication can be tested by x-ray diffraction measurements.

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