Thermodynamic Evidence for a Two-Component Superconducting Order Parameter in Sr$_2$RuO$_4$

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Sr$_2$RuO$_4$ has stood as the leading candidate for a spin-triplet superconductor for 26 years. Recent NMR experiments have cast doubt on this candidacy, however, and it is difficult to find a theory of superconductivity that is consistent with all experiments. What is needed are symmetry-based experiments that can rule out broad classes of possible superconducting order parameters. Here we use resonant ultrasound spectroscopy to measure the entire symmetry-resolved elastic tensor of Sr$_2$RuO$_4$ through the superconducting transition. We observe a thermodynamic discontinuity in the shear elastic modulus $c_{66}$, requiring that the superconducting order parameter is two-component. A two-component $p$-wave order parameter, such as $p_x + ip_y$, naturally satisfies this requirement. As this order parameter appears to be precluded by recent NMR experiments, we suggest that two other two-component order parameters, namely $\{d_{xz}, d_{yz}\}$ or $\{d_{x^2-y^2}, g_{xy}(x^2-y^2)\}$, are now the prime candidates for the order parameter of Sr$_2$RuO$_4$.

INTRODUCTION

Nearly all known superconductors are “spin-singlet”, composed of Cooper pairs that pair spin-up electrons with spin-down electrons. Noting that Sr$_2$RuO$_4$ has similar normal-state properties to superfluid $^3$He, Rice and Sigrist[1] and, separately, Baskaran [2], suggested that Sr$_2$RuO$_4$ may be a solid-state “spin-triplet” superconductor. This attracted the attention of the experimental community, and ensured decades of intense research on Sr$_2$RuO$_4$ that resulted in a detailed understanding of its metallic state [3, 4]. From this well-understood starting point one might expect the superconductivity of Sr$_2$RuO$_4$ to be a solved problem [5], but decades after its discovery the symmetry of the superconducting order parameter remains a mystery, largely due to discrepancies between several major pieces of experimental evidence [6].

Formerly, the strongest evidence for spin-triplet pairing in Sr$_2$RuO$_4$ was a Knight shift that was unchanged upon entering the superconducting state [7]. A recently revised version of this experiment, however, shows that the Knight shift is suppressed below $T_c$, ruling against most spin-triplet order parameters [8, 9]. This is consistent with measurements of the upper-critical magnetic field, which appears to be Pauli-limited and thus suggests spin-
singlet pairing [10]. The challenge is to reconcile these data with previous evidence in favour of a spin-triplet order parameter, including time-reversal symmetry breaking below $T_c$ in $\mu$SR[11] and polar Kerr effect experiments[12], and half-quantized vortices [13].

While the spin-triplet versus spin-singlet aspect of the superconductivity in Sr$_2$RuO$_4$ is still under debate, less well studied is the symmetry of the orbital part of the Cooper pair wavefunction. By symmetry, spin-triplet superconductors are required to have an odd-parity orbital wavefunction, i.e. to be an $l = 1$ ‘$p$-wave’ or $l = 3$ ‘$f$-wave’ superconductor, where $l$ is the orbital quantum number. This is in contrast with conventional $l = 0$ $s$-wave superconductors, or the high-$T_c$ $l = 2$ $d$-wave superconductors. While some information about the orbital wavefunction can be inferred by looking for nodes in the superconducting gap, determination of nodal position does not uniquely determine the orbital structure of the Cooper pair.

One way to distinguish different orbital states is by their degeneracy—the number of states with the same energy. $s$-wave and $d_{x^2-}y^2$-wave Cooper pairing states, for example, are both singly degenerate (“one-component”), while the $\{p_x, p_y\}$ state (which can order in the chiral $p_x + ip_y$ configuration) is two-fold degenerate (“two-component”). This difference in orbital degeneracy has an unambiguous signature in an ultrasound experiment: shear elastic moduli are continuous through $T_c$ for a singly-degenerate orbital state, but are discontinuous across $T_c$ for a doubly degenerate state [14, 15]. The observation of a discontinuity in one of the shear elastic moduli of Sr$_2$RuO$_4$ at $T_c$ would therefore constitute strong evidence in favour of either $p$-wave superconductivity, or one of the other two-component superconducting order parameters. These measurements have been attempted in the past, and were suggestive of a shear discontinuity at $T_c$, but a discontinuity was also found in a symmetry-forbidden channel, and the experiment was thus deemed to be inconclusive [16]. Other independent evidence of a shear discontinuity [17] is now being submitted as part of a separate complementary study using an experimental technique different from our own (for a theoretical interpretation of these results, see Walker and Contreras [18]).

**EXPERIMENT**

Elastic moduli are second derivatives with respect to strain of a system’s total free energy. Elastic moduli are therefore thermodynamic coefficients akin to heat capacity or magnetic
FIG. 1. Irreducible strains in Sr$_2$RuO$_4$ and their coupling to superconducting order parameters. The tetragonal crystal structure of Sr$_2$RuO$_4$ and unit cell deformations illustrating the irreducible representations of strain are shown. There is an elastic modulus corresponding to each of these strains, and a sixth modulus $c_{13}$ that arises from coupling between the two $A_{1g}$ strains. Green check marks denote allowed linear-order couplings to strain for one and two-component order parameter bilinears, and red crosses denote that such coupling is forbidden. These couplings are what lead to discontinuities in the elastic moduli at $T_c$. See Table I for a list of relevant possible order parameters in Sr$_2$RuO$_4$.

susceptibility, and are indicative of a system’s ground-state properties. Strain is a second-rank tensor quantity, and thus it can couple to order parameters in ways that lower-rank quantities, such as temperature and electric field, cannot. This, in particular, requires that elastic moduli behave differently in systems with one- or two-component order parameters. Here we provide a brief overview of the connection between crystal symmetry, order parameter symmetry, and ultrasound: the detailed derivations can be found in the ‘Strain-Order Parameter Coupling’ section in S.I., as well as in a number of theoretical papers [14, 18, 19].

The allowed couplings between strains and superconducting order parameters become transparent when both are described in terms of irreducible representations (irreps) of the
FIG. 2. Resonant ultrasound spectroscopy: schematic and spectrum. (a) A single-crystal sample, polished along known crystal axes, is held in weak-coupling contact between two ultrasonic transducers, allowing it to vibrate freely at its resonance frequencies. Panels (b) through (e) show the crystal’s deformation corresponding to four particular experimentally measured resonances, marked in (f). (f) A portion of the ultrasonic spectrum of Sr$_2$RuO$_4$, in the frequency range from 2.4-2.9 MHz, taken at room temperature. Each resonance creates a unique strain pattern in the material that can be decomposed in terms of the five irreducible strains (Figure 1(a)), modulated in phase along the dimensions of the sample.

point-group symmetry. Sr$_2$RuO$_4$ crystallizes in the tetragonal space group $I4/mmm$, along with its associated point group $D_{4h}$. In this crystal field environment, the five-component $l = 2$ d-representation breaks into three one-component irreps: $d_{z^2}$ ($A_{1g}$ irrep), $d_{xy}$ ($B_{2g}$), and
$d_{x^2-y^2}$ ($B_{1g}$—the familiar ‘$d$-wave’ of the cuprates), and one two-component irrep \{\textit{d}_{xz}, \textit{d}_{yz}\} ($E_g$). The three-component $p$-representation breaks into the one-component irrep $p_z$ ($A_{2u}$) and the two-component irrep $\{p_x, p_y\}$ ($E_u$)—it is this latter representation that has been proposed to order into the chiral $p_z + ip_y$ superconducting state.

As illustrated in Figure 1, there are five unique strains ($\epsilon_{\Gamma}$) in Sr$_2$RuO$_4$ (five irreps ($\Gamma$) of strain in $D_{4h}$): two compressive strains transforming as the $A_{1g}$ irrep, and three shear strains transforming as the $B_{1g}$, $B_{2g}$ and $E_g$ irrep. Each strain has a corresponding elastic modulus, $c_{\Gamma} = \partial^2 F / \partial \epsilon_{\Gamma}^2$, where $F$ is the thermodynamic free energy. A sixth modulus, $c_{13}$, defines the coupling between the two $A_{1g}$ strains ($\epsilon_{xx} + \epsilon_{yy}$ and $\epsilon_{zz}$). Sound velocities can be computed from these moduli as $v_{\Gamma} = \sqrt{c_{\Gamma}/\rho}$, where $\rho$ is the density. When composing terms in the free energy, direct (linear) coupling between strain and the superconducting order parameter ($\eta$) is forbidden because superconductivity breaks gauge symmetry. The next relevant coupling is linear in strain and quadratic in order parameter. For one-component superconducting order parameters, including all s-wave states and the $d_{x^2-y^2}$ state, the only quadratic form is $\eta^2$, transforming as $A_{1g}$, and thus the only allowed coupling is $\epsilon_{A_{1g}} \eta^2$. This coupling produces discontinuities in all the $A_{1g}$ (compressional) elastic moduli across $T_c$. Two-component order parameters $\vec{\eta} = \{\eta_x, \eta_y\}$, on the other hand, have three independent quadratic forms: $\eta_x^2 + \eta_y^2$, $\eta_x^2 - \eta_y^2$, and $\eta_x \eta_y$, transforming as $A_{1g}$, $B_{1g}$, and $B_{2g}$, respectively. Thus in addition to coupling to the $A_{1g}$ elastic moduli, two-component order parameters couple to two of the shear moduli through $\epsilon_{B_{1g}} (\eta_x^2 - \eta_y^2)$ and $\epsilon_{B_{2g}} \eta_x \eta_y$. This produces discontinuities in the associated shear elastic moduli ($(c_{11} - c_{12})/2$ and $c_{66}$, respectively), based purely on symmetry considerations, and independent of the microscopic mechanism of superconductivity.

While traditional pulse-echo ultrasound experiments measure a single elastic modulus per experiment, we use resonant ultrasound spectroscopy (RUS) to measure all six elastic moduli of Sr$_2$RuO$_4$ across $T_c$ in a single experiment, greatly reducing systematic uncertainty [20]. Analogous to how a stretched string has standing waves that can be expressed in terms of sinusoidal harmonics, three-dimensional solids have elastic resonances that can be decomposed in terms of the irreps of strain. RUS measures the frequencies of these resonances for a single crystal sample, from which all elastic moduli can be obtained by inverse-solving the elastic wave equation [21]. The relatively low $T_c$ ($\approx 1.43$ K for our sample) of Sr$_2$RuO$_4$, however, poses severe technical challenges to perform RUS experiments across $T_c$. RUS samples are typically large (of the order 1 mm$^3$), and are only in weak-coupling
contact with the transducers (see Figure 2(a)). This ensures nearly-free boundary conditions but prevents good thermal coupling between the apparatus and the sample. Previous RUS implementations either sacrificed uniform cooling by placing the sample in vacuum [21], or sacrificed a slow cooling rate by placing the sample in direct thermal contact with the helium bath. Our new RUS design employs a double vacuum can arrangement (see Methods for details) to allow for slow, uniform cooling of a sample down to approximately 1.25 K. We observe a sharp (40 mK wide) superconducting transition (Figure 3(a)), signifying high sample quality and uniform sample cooling.

We performed RUS measurements on a single-crystal sample of Sr$_2$RuO$_4$ across $T_c$. The full data set includes 18 resonances (see Figure 5)—five representative resonances are shown in Figure 3(a). We decompose all 18 resonances into the three compressional moduli $((c_{11} + c_{12})/2, c_{33},$ and $c_{13})$ and the three shear moduli $((c_{11} - c_{12})/2, c_{44},$ and $c_{66})$. The temperature evolution of these moduli are shown in Figure 3(b) and (c). Discontinuities across $T_c$ are clearly observed in all three compressional moduli, as required by thermodynamics for all superconductors, as well as in the shear modulus $c_{66}$. The discontinuity in $c_{66}$ is forbidden by symmetry for one-component order parameters, but is allowed for two-component order parameters—this discontinuity is our central finding.

Having measured all six elastic moduli across $T_c$, we can perform several consistency checks. First, since there is no bilinear coupling of the order parameter to $E_g$ strain, $c_{44}$ should not have a discontinuity at $T_c$ for any superconducting order parameter. Within our experimental uncertainty, we observe only a change in the slope of $c_{44}$ at $T_c$, which is expected on general grounds and is not constrained by symmetry [14]. Second, thermodynamics dictates that the discontinuities in the three compressional moduli at a second order phase transition should follow a self-consistency relation (see discussion preceding Equation S16 in S.I.),

$$\left(\frac{\Delta c_{11} + c_{12}}{2}\right) \times (\Delta c_{33}) = (\Delta c_{13})^2. \quad (1)$$

From our measurement, we find $(\Delta c_{11} + c_{12})/2 \times (\Delta c_{33}) = (9.9 \pm 1.5) \times 10^{-5}$ GPa$^2$ and $(\Delta c_{13})^2 = (8.3 \pm 1.1) \times 10^{-5}$ GPa$^2$. These consistency checks validate our measurement of the magnitude of the jumps, and our ability to correctly decompose the jumps in frequency into jumps in the irreducible elastic moduli.

A further check on the data is provided by an Ehrenfest relation that relates the derivative of $T_c$ with hydrostatic pressure, $P$, to the discontinuities at $T_c$ in the specific heat, $\Delta C$, and
FIG. 3. Resonant ultrasound spectroscopy across $T_c$ in Sr$_2$RuO$_4$. (a) Temperature evolution of five representative resonances measured through $T_c$—plots are shifted vertically for visual clarity. The dashed line shows $T_c$ determined by resistivity measurements. A step-like discontinuity (“jump”) is observed at $T_c$—the different magnitudes of this jump signify the contributions of different elastic moduli in each resonance. 18 such resonances were tracked through $T_c$ to determine the elastic moduli. (b) Compressional ($A_{1g}$) and (c) shear ($B_{1g}$, $E_g$ and $B_{2g}$) moduli of Sr$_2$RuO$_4$ across $T_c$, along with their experimental errors arising from the uncertainties in sample dimensions. The absolute values of these moduli at 4 kelvin are determined to be $(c_{11} + c_{12})/2 = 190.8$, $c_{33} = 257.2$, $c_{13} = 85.0$, $(c_{11} - c_{12})/2 = 53.1$, $c_{66} = 65.5$ and $c_{44} = 69.5$ GPa. (d) Magnitudes of the elastic moduli jumps at $T_c$, along with their experimental uncertainties (see ‘Uncertainty Analysis’ in S.I. for details).

the bulk modulus, $\Delta B$, via

$$
\left( \frac{dT_c}{dP} \right)^2 = \frac{-\Delta B}{B^2} \left( \frac{\Delta C}{T} \right)^{-1}.
$$

We measure $\Delta B/B \approx 6.3 \times 10^{-5}$ (see ‘Ehrenfest Relations for Compressional Strains’ in S.I.).
Combined with $\Delta C/T$ for this sample (see Figure 4), Equation 2 yields a value for $dT_c/dP$ of 0.90 K/GPa. This is a factor of 3 higher than what is reported for a direct measurement of $T_c$ as a function of pressure [22]. This discrepancy may be evidence for a pair of transitions occurring at or near the superconducting $T_c$, as discovered by recent $\mu$SR experiments [23]. The two transition temperatures split when stress is applied along the $x$ direction—Meissner screening onsets at the higher transition temperature, $T_c$, while time reversal symmetry is broken at the lower transition, $T_{TRSB}$. To perform the correct Ehrenfest analysis one would require $dT_{TRSB}/dP$, which is unknown at present. A similar Ehrenfest relation—derived for the jump in $c_{66}$ rather than the jump in bulk modulus—requires that $T_c$ shift linearly with $B_{2g}$ strain, specifically as $T_c \propto |\epsilon_{xy}|$. Prior measurements of $T_c$ as a function of $\epsilon_{xy}$, however, have not found this linear dependence on strain [24]. In addition, most ordered states of two-component order parameters should exhibit two transition temperatures under finite strain, but this has not been found either by heat capacity or local susceptibility measurements [25, 26], at least for $B_{1g}$ strain. In the S.I. we show that the current experimental resolution on these phenomena is still consistent with the size of the jump that we find in $c_{66}$ (see ‘Reconciling the $c_{66}$ Discontinuity with Experiments Under Finite Strain’).

**DISCUSSION**

A discontinuity in $c_{66}$ at $T_c$ can only result from a two-component superconducting order parameter (see Table I). This is a critical piece of information because evidence for vertical line nodes in the superconducting gap—from ultrasonic attenuation [17], heat capacity [27], thermal conductivity [28], and quasiparticle interference [29]—are most straightforwardly interpreted in terms of a one-component, $d_{x^2-y^2}$, order parameter. With the discovery of a suppression of the Knight shift strongly suggesting that the order parameter cannot be spin-triplet [8], $d_{x^2-y^2}$ would seem a likely contender. The discontinuity in $c_{66}$, however, rules against any one-component order parameter, including $d_{x^2-y^2}$.

Our measurement is consistent with several two-component $p$-wave scenarios, including $(p_x \pm ip_y)\hat{z}$ and $p_z(\hat{x} \pm i\hat{y})$. Taken at face value, however, the suppression of the Knight shift [8] rules out all $p$-wave order parameters, and is consistent only with spin-singlet order parameters. The only “conventional” spin-singlet order parameter that produces a jump in $c_{66}$ at $T_c$ is $\{d_{xz}, d_{yz}\}$. This state can order into the non-magnetic $d_{xz}$, $d_{yz}$, or $d_{xz} \pm d_{yz}$
| Dimensionality | Order Parameter | Irrep. | Moduli Jumps | Ultrasound | NMR |
|----------------|----------------|-------|--------------|------------|-----|
| One-component  | \(s\)          | \(A_{1g}\) | \(A_{1g}\)   | \(\times\) | \(\checkmark\) |
|                | \(d_{x^2-y^2}\) | \(B_{1g}\) | \(A_{1g}\)   | \(\times\) | \(\checkmark\) |
|                | \(d_{xy}\)     | \(B_{2g}\) | \(A_{1g}\)   | \(\times\) | \(\checkmark\) |
| Two-component  | \(\{p_x, p_y\} \hat{z}\) | \(E_u\) | \(A_{1g}, B_{1g}, B_{2g}\) | \(\checkmark\) | \(\times\) |
|                | \(p_z \{\hat{x}, \hat{y}\}\) | \(E_u\) | \(A_{1g}, B_{1g}, B_{2g}\) | \(\checkmark\) | \(\times\) |
|                | \(\{d_{xz}, d_{yz}\}\) | \(E_{2g}\) | \(A_{1g}, B_{1g}, B_{2g}\) | \(\checkmark\) | \(\checkmark\) |
|                | \(\{d_{x^2-y^2}, g_{xy}(x^2-y^2)\}\) | \(B_{1g} \oplus A_{2g}\) | \(A_{1g}, B_{2g}\) | \(\checkmark\) | \(\checkmark\) |

**TABLE I. Some superconducting order parameters and their representations in \(D_{4h}\).**

For the odd-parity spin-triplet order parameters, \(\hat{x}, \hat{y}\) and \(\hat{z}\) represent the pair wavefunction in spin space in the \(d\)-vector notation [5]. Two component order parameters \(\{\eta_x, \eta_y\}\) can order as \(\eta_x, \eta_y, \eta_x \pm \eta_y, \) or \(\eta_x \pm i\eta_y,\) depending on microscopic details. It is this latter combination that forms the time-reversal symmetry breaking state (e.g. \((p_x + ip_y)\hat{z}, \) or \(d_{xz} + id_{yz}\)). The “Ultrasound” column indicates whether an order parameter is consistent with a jump in \(c_{66}\) at \(T_c\), the “NMR” column indicates whether it is consistent with the suppression of the Knight shift at \(T_c\). Note that the \(B_{1g} \oplus A_{2g}\) state does not belong to a single irrep of \(D_{4h}\), and thus transition temperatures of the \(d\) and \(g\) components must be “fine-tuned” if they are to coincide.

states, all of which break the \(C_4\) rotational symmetry of the lattice. It can also order into the chiral magnetic \(d_{xz} \pm id_{yz}\) state. If one considers the possibility of an accidental degeneracy between two order parameters of different representations, producing accidental two-component order parameters, then \(\{d_{xy}, s\}\) and \(\{d_{x^2-y^2}, g_{xy}(x^2-y^2)\}\) are also consistent with our experiment (\(\{d_{x^2-y^2}, s\}\) [30] produces a jump in \((c_{11} - c_{12})/2\) but not in \(c_{66}\)). We set aside \(\{d_{xy}, s\}\) because it thought to be incompatible with the electronic structure of \(\text{Sr}_2\text{RuO}_4\). If one accepts time-reversal symmetry breaking at \(T_c\) as a property of \(\text{Sr}_2\text{RuO}_4\), there are then two remaining order parameters that are compatible with our experiment: \(d_{xz} \pm id_{yz}\), and \(d_{x^2-y^2} \pm ig_{xy}(x^2-y^2)\). The absence of a discontinuity in \((c_{11} - c_{12})/2\) implies that there is no order-parameter-bilinear that transforms as \(B_{1g}\), which would rule out \(d_{xz} \pm id_{yz}\). It is possible, however, that while a jump in \((c_{11} - c_{12})/2\) is required thermodynamically, it is either unobservably small because the coupling coefficient is small (for microscopic reasons), or the jump is smeared-out due to high ultrasonic attenuation in the \(B_{1g}\) channel.
Thus we consider the implications of both the $d_{xz} \pm id_{yz}$ and the $d_{x^2-y^2} \pm ig_{xy(x^2-y^2)}$ superconducting states in Sr$_2$RuO$_4$.

The first of these, $d_{xz} \pm id_{yz}$, is the chiral-ordered state of $\{d_{xz}, d_{yz}\}$—a two-component $E_g$ representation. There are two main arguments against such a state. First, $\{d_{xz}, d_{yz}\}$ has a horizontal line node at $k_z = 0$, whereas most experiments suggest that the nodes lie along the [110] and [110] directions. There is some evidence, however, for a horizontal line node from angle-dependent heat capacity measurements. Second, Sr$_2$RuO$_4$ has very weak interlayer coupling, and in the limit of weak interlayer coupling, the pairing strength for this state goes to zero. A recent weak-coupling analysis shows that an $E_g$ state can be stabilized by including momentum-dependent spin orbit coupling, and such spin-orbit coupling has been quantified by ARPES in Sr$_2$RuO$_4$. This variant of the $E_g$ state has Bogoliubov Fermi surfaces (rather than line nodes) that extend along the $k_z$ direction in a manner that may mimic line-nodes as far as experiment is concerned.

The second possibility, $d_{x^2-y^2} \pm ig_{xy(x^2-y^2)}$, is less natural in that $d_{x^2-y^2}$ is a $B_{1g}$ irrep and $g_{xy(x^2-y^2)}$ is an $A_{2g}$ irrep. Order parameters of different representations have, in general, distinct transition temperatures, and therefore the composite $B_{1g} \oplus A_{2g}$ order parameter requires fine-tuning to produce a single superconducting transition. Fine-tuning aside, this state has two attractive features. First, it produces bilinears only in the $A_{1g}$ and $B_{2g}$ channels ($B_{1g} \otimes A_{2g} = B_{2g}$). This would naturally explain why a jump is seen in $c_{66}$ but not in $(c_{11}-c_{12})/2$. Second, this state has line nodes along the [110] and [110] directions. While the $l = 4$, $g_{xy(x^2-y^2)}$ state may seem exotic, it has been shown (in the weak-coupling regime) to be competitive with $d_{x^2-y^2}$ when nearest-neighbor repulsion is accounted for.

Both of these two-component order parameters produce a discontinuity in $c_{66}$, break time reversal symmetry, are Pauli limited in their upper critical field, exhibit a drop in the Knight shift below $T_c$, and have ungapped quasiparticles. The accidental degeneracy of $d_{x^2-y^2} \pm ig_{xy(x^2-y^2)}$ means that its $T_c$ should split into two transitions under any applied strain, and indeed, the aforementioned $\mu$SR measurements have found evidence for such a splitting. This suggests that Sr$_2$RuO$_4$ may indeed have two nearly degenerate transitions at ambient pressure. Whether or not such a fine-tuned state is tenable to all experiments remains to be seen, but it is now clear that a two-component superconducting order parameter is an essential feature for understanding the unusual superconducting properties of Sr$_2$RuO$_4$. 


METHODS

Sample Preparation

High-quality single crystal Sr$_2$RuO$_4$ were grown by the floating-zone method, details of which can be found in [41]. A single crystal was oriented along the [110], [110] and [001] directions, and polished to dimensions 1.50mm × 1.60mm × 1.44mm, with 1.44mm along the tetragonal c-axis. The [110] orientation of the crystal was accounted for when solving for the elastic moduli.

Sample Characterization

![Graphs of C/Τ vs. T and B vs. T](a) and (b)

FIG. 4. Characterization of the Sr$_2$RuO$_4$ rod. (a) Specific heat and (b) susceptibility measurements of the upper critical field, measured on different parts of the same rod from which the sample for RUS experiment was obtained. $T_c$ varies by about 200 mK between different parts of the rod.

The quality of the Sr$_2$RuO$_4$ rod from which the RUS sample was cut was characterized by heat capacity and AC susceptibility measurements, shown in Figure 4. Heat capacity measurements of a large piece of the rod exhibit a $T_c$ around 1.45 K, which is close to the optimal $T_c$ [42]. In addition to a relatively high $T_c$, a low concentration of ruthenium inclusions was an important criterion for the selection of the RUS sample. Ruthenium inclusions locally strain the crystal lattice and can enhance $T_c$ up to 3 K. In order to check for ruthenium inclusions, we measured AC susceptibility by a mutual inductance method,
and found a sharp onset-Tc of 1.43 K, with no sign of a Tc at 3 K, indicating a very low concentration of ruthenium inclusions. The variation in Tc between the heat capacity and the susceptibility/critical field experiments arises because the samples were taken from different parts of the same Sr2RuO4 rod. The Sr2RuO4 sample for the RUS experiments was also taken from the same rod, and the onset Tc of 1.425 K is in good agreement with the above-mentioned measurements.

**Low-Temperature RUS**

The relatively low Tc of Sr2RuO4 poses several challenges to perform RUS experiments through the superconducting transition. Since RUS samples are typically large (of the order 1 mm3) and not glued to the transducers (to ensure free boundary conditions), there is weak thermal contact between the sample and its surroundings. Hence, when cooled through Tc, the entire sample may not become superconducting at once, leading to broad superconducting transitions rather than sharp jumps at Tc. To cool below 4.2 K, one could introduce liquid helium into the sample space and pump directly on this space. As RUS is extremely sensitive to vibration, however, this introduces artefacts into the data, and does not provide a particularly homogeneous thermal environment.

To solve these problems the RUS probe was sealed inside a copper can with a small amount of exchange gas, providing good thermal equilibration between the sample, thermometer, and the rest of the apparatus. This inner copper can is separated by a weak thermal link (thin-wall stainless) from an outer brass vacuum can, which provided isolation between the walls of the sample can and the bath. The temperature was regulated by pumping on the external helium bath, and the vacuum isolation of the sample chamber from the bath allows the sample space to then cool very slowly once the bath is pumped to base temperature.

The lowest temperature reached was approximately 1.25 K, as read by a CX-1030 thermometer affixed to the RUS probe. The transition temperature and transition width observed by RUS agree extremely well with those determined from independent susceptibility measurements, suggesting that the Sr2RuO4 sample was uniformly thermalized during the experiment.

We measured the temperature dependence of 18 resonances through the superconducting transition—the full data set is shown in Figure 5. The temperature spacing is not identical
FIG. 5. RUS frequency data. Temperature evolution of 18 resonance frequencies of Sr$_2$RuO$_4$ through $T_c$, with panels (a) and (b) each showing 9 frequencies. Plots are vertically shifted for visual clarity.

because the data was acquired over several sweeps through $T_c$, measuring a few resonances each time. These 18 resonances are then decomposed into the elastic moduli that are shown in Figure 3 of the main text. To do this decomposition we interpolate the frequency versus temperature data, and then plot the elastic moduli at what we consider to be a representative set of temperatures—those from the sweep where we measured the resonances at 2495 kHz, 2549 kHz, and 2551 kHz.

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AUTHOR CONTRIBUTIONS

S.G. and B.J.R designed the experiment. F.J., D.A.S., N.K., M.B., C.W.H. and A.P.M. prepared the crystal and performed characterization measurements. S.G. acquired and analyzed the ultrasound data. S.G., A.S., C.W.H. and B.J.R. wrote the manuscript with input from all co-authors.

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Thermal Homogeneity Below $T_c$

While the presence of exchange gas in the sample space ensures that all sample surfaces are at a uniform temperature (which is the same temperature read by the thermometer), we rely on good thermal conduction for the sample interior to equilibrate with the surfaces. Above $T_c$, normal state Sr$_2$RuO$_4$ is a good metal which conducts heat well, and thus the entire volume of the sample should be in thermal equilibrium with surroundings. Below $T_c$, however, temperature gradients may be enhanced due to the loss of heat carriers, as Cooper pairs do not carry heat. We expect this not to be an issue since Sr$_2$RuO$_4$ is a nodal superconductor, which means that there are always normal quasiparticles that carry heat, and the lowest temperature we reach is $\sim 0.8T_c$, which means that the superconducting gap has only partially opened.

To confirm that the thermal gradients in our sample are always small, we perform a simple calculation starting from the 3-dimensional heat flow equation,

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T,$$

where $T(x, y, z, t)$ denotes the temperature profile within the sample, and $\alpha$ is the thermal diffusivity. For tetragonal Sr$_2$RuO$_4$, this takes the form

$$\frac{\partial T}{\partial t} = \alpha_a \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right) + \alpha_c \frac{\partial^2 T}{\partial z^2}.$$

The thermal diffusivity $\alpha$ is related to the thermal conductivity $\kappa$ as $\alpha = \kappa / \rho C$, where $\rho$ is the density of Sr$_2$RuO$_4$ and $C$ is the specific heat. For Sr$_2$RuO$_4$ below $T_c$, we have $C = 87$ mJmol$^{-1}$K$^{-1}$, $\kappa_a = 13.05$ Wm$^{-1}$K$^{-1}$ and $\kappa_c = 0.07$ Wm$^{-1}$K$^{-1}$, giving $\alpha_a = 8.61 \times 10^{-3}$ m$^2$/s and $\alpha_c = 0.05 \times 10^{-3}$ m$^2$/s.

We first consider the effects of a step-change in the exchange-gas temperature as a worst-case scenario. With the boundary condition that all sample surfaces are at $T_0$, the solution of [Equation 4] is

$$T(x, y, z, t) = T_0 + \sum_{n,n,p=1}^{\infty} A_{mnp} \sin \left( \frac{m\pi x}{L_x} \right) \sin \left( \frac{n\pi y}{L_y} \right) \sin \left( \frac{p\pi z}{L_z} \right) \cdot \exp \left( -\pi^2 \left( \frac{\alpha_a m^2}{L_x^2} + \frac{\alpha_a n^2}{L_y^2} + \frac{\alpha_c p^2}{L_z^2} \right) t \right),$$

where $L_x$, $L_y$, and $L_z$ are the lengths of the sample along the $x$, $y$, and $z$ directions, respectively.
where \(m, n, p\) are integers, \(L_x, L_y, L_z\) are the sample dimensions, and the coefficients \(A_{mnp}\) depend on the initial temperature profile \(T(x, y, z, t = 0)\) within the sample. Thus, temperature variations from \(T_0\) within the sample die out exponentially fast and, in the \(t \to \infty\) limit, \(T(x, y, z) \to T_0\). In particular, the slowest equilibration occurs when \(A_{111} \neq 0\) and all other \(A_{mnp} = 0\), since higher harmonic components have a faster exponential decay. In this case, Equation 5 simplifies to

\[
T(x, y, z, t) = T_0 + A_{111} \sin \left( \frac{\pi x}{L_x} \right) \sin \left( \frac{\pi y}{L_y} \right) \sin \left( \frac{\pi z}{L_z} \right) \exp \left( -\pi^2 \left( \frac{\alpha_a}{L_x^2} + \frac{\alpha_a}{L_y^2} + \frac{\alpha_c}{L_z^2} \right) t \right). \tag{6}
\]

The temperature in the middle of the sample \((T_{\text{mid}}(t) = T(L_x/2, L_y/2, L_z/2, t))\) differs most from \(T_0\) at \(t = 0\), hence we can just look at \(T_{\text{mid}}\) to get an upper bound on how long it takes for the entire sample to come to \(T_0\).

\[
T_{\text{mid}}(t) - T_0 = \Delta T \exp \left( -\pi^2 \left( \frac{\alpha_a}{L_x^2} + \frac{\alpha_a}{L_y^2} + \frac{\alpha_c}{L_z^2} \right) t \right) = \Delta T \exp(-\frac{t}{14 \mu s}), \tag{7}
\]

where we have used the dimensions of the sample \((1.50\text{mm} \times 1.60\text{mm} \times 1.44\text{mm})\), and we have used the worst-case diffusivity taken below \(T_c\) from the thermal conductivity data reported in Hassinger et al. [28] and using the specific heat measured on the same rod our sample came from (see Figure 4). Equation 7 shows that if a temperature difference \(\Delta T\) appears within the sample, it reduces by a factor of 1000 in less than \(10^{-4}\) s. For comparison, we cool the sample at \(\approx 0.3\) mK/s, and acquire a data point roughly every 10 mK. Thus the equilibration time is much faster than time scale over which we do our measurements.

Another consideration is the total temperature offset in the center of the sample as we sweep the temperature, which is not captured by Equation 7. The steady-state solution of Equation 4 for a constant cooling rate results in parabolic temperature profile inside the sample. The steady state profile \(T_{\text{eq}}\) is given by,

\[
T_{\text{eq}}(x, y, z) = T_0 + 64 \frac{x(L_x - x)y(L_y - y)z(L_z - z)}{L_x^2 L_y^2 L_z^2} \delta T \tag{8}
\]

Clearly, \(T_{\text{eq}} = T_0\) at the sample surfaces, and the middle of sample is at \(T_0 + \delta T\). The offset \(\delta T\) can be calculated by evaluating Equation 3 at \((x = L_x/2, y = L_y/2, z = L_z/2)\) and using the cooling rate \((\partial T/\partial t = 0.3\) mK/s),

\[
\delta T = \frac{\partial T/\partial t}{8 \left( \frac{\alpha_a}{L_x^2} + \frac{\alpha_a}{L_y^2} + \frac{\alpha_c}{L_z^2} \right)} \approx 6\text{ nK}. \tag{9}
\]
Immediately below $T_c$, the heat capacity increases by approximately 50%. Over our full temperature range, the thermal conductivity $\kappa$ drops by about 10% [28]. The thermal diffusivity therefore drops by $\approx 50\%$ below $T_c$. Even with this relatively large change in diffusivity, our results above demonstrate that our sample is very homogeneous in temperature during the course of the RUS experiment—both above and below $T_c$.

**Uncertainty Analysis**

While visual inspection is usually sufficient to determine whether or not a particular modulus shows a discontinuity at $T_c$, numerical values are needed to perform consistency checks on the data, and to make quantitative predictions for future experiments.

One source of uncertainty comes from the width of the superconducting transition. To extract the jump magnitudes and their associated uncertainties in a consistent fashion for all the moduli, we use fits above and below $T_c$ to extrapolate the data across the transition (see Figure 6). We take the jump as the difference between the two extrapolated fits at the experimentally-obtained temperature points between 1.38 K and 1.43 K, which is the width of the superconducting transition. This gives a (non-Gaussian) distribution of modulus jumps that correspond to different $T_c$ assignments. We take the uncertainty to be half the difference of the minimum and maximum jumps, and assign the jump itself to the mean.

A second source of uncertainty comes from the sample dimensions, which are used to extract the elastic moduli from the resonance frequencies. Slight deviations from parallelism, rounding of the sample corners, and other small imperfections give an upper-bound on the dimensional uncertainty of $\pm 20$ microns in each direction. This dimensional uncertainty is then incorporated into the elastic moduli calculations (described in Ramshaw et al. [21]), yielding an uncertainty in the moduli. This gives us the error bars shown for compressional and shear moduli in Figure 3(b) and (c) of main text.

Assuming that the two sources of uncertainty (dimensional uncertainty, and the uncertainty in assigning $T_c$) are independent, we add them in quadrature to obtain the total uncertainty in each jump, tabulated in Table II.

We have neglected uncertainty due to misalignment of the crystal axes with respect to the sample faces because our crystal was aligned to better than $2^\circ$ for all 3 axes. The effect
FIG. 6. Extraction of the jumps in the elastic moduli and their uncertainties. We extrapolate fits to data (red lines) from above and below \( T_c \) at temperature points within the transition (highlighted in yellow). The average between the minimum and maximum jump is taken to be the jump magnitude; the difference is the uncertainty. This procedure is illustrated for \( c_{66} \) in panel (a), and \( c_{44} \) in panel (b). Note the significantly reduced vertical scale in (b) as compared to (a).

Of misalignment can be calculated finding the rotated elastic tensor \( C' \) via

\[
C'_{mnop} = R_{mi}R_{nj}R_{ok}R_{pl}C_{ijkl} \tag{10}
\]

where \( C \) is the un-rotated elastic tensor and \( R \) is a rotation matrix. For example, a rotation by an angle \( \gamma \) about the \( a \)-axis transforms the shear elastic modulus \( c_{44} \) as

\[
c'_{44} = \frac{1}{4} \left[ c_{44} (3 + \cos 4\gamma) + \frac{1}{2} (c_{11} + c_{33} + 2c_{13} (1 - \cos 4\gamma)) \right]. \tag{11}
\]

For \( \gamma = 2^\circ \), \( c'_{44} \) differs from \( c_{44} \) by four parts in \( 10^3 \). This introduces a jump into \( c'_{44} \) that is 1 part in \( 10^8 \)—two orders of magnitude smaller than the other sources of uncertainty. Similar expressions can be derived for the other moduli and are similarly small.

**Strain-Order Parameter Coupling**

In this section, we calculate expressions for the elastic moduli discontinuities and sound attenuation, starting from a Landau theory. Similar expressions for the chiral state have been calculated by Sigrist \[19\] and for nematic superconducting states by Huang et al. \[43\]. Our expressions match those of Sigrist for the chiral state, and we correct the expressions derived by Huang et al. by including all possible order parameter fluctuations. In particular,
we find that both in-plane shear moduli \((c_{11} - c_{12})/2\) and \(c_{66}\) should show a discontinuity for the nematic states, contrary to what was concluded in \cite{43}. We also show that the three \(A_{1g}\) jumps always follow a consistency relation, similar to what was concluded for non-superconducting order parameters in URu$_2$Si$_2$ \cite{15}, which is also a tetragonal material with point group \(D_{4h}\).

For a two-component superconducting order parameter \(\eta = (\eta_x, \eta_y)\), the Landau free energy expansion reads

\[
\mathcal{F}_{op}(\eta) = a|\eta|^2 + b_1|\eta|^4 + \frac{b_2}{2} (|\eta_x^\ast \eta_y^\ast|^2 + (\eta_x \eta_y^\ast)^2) + b_3|\eta_x|^2|\eta_y|^2 + \ldots \tag{12}
\]

where \(a = a_0(T - T_c)\), with \(a_0 > 0\), and \(b_i\) are phenomenological constants. In a tetragonal crystal, the elastic free energy density is given by

\[
\mathcal{F}_{el} = \frac{1}{2} \left( c_{11} (\epsilon_{xx}^2 + \epsilon_{yy}^2) + 2c_{12} \epsilon_{xx} \epsilon_{yy} + c_{33} \epsilon_{zz}^2 + 2c_{13} (\epsilon_{xx} + \epsilon_{yy}) \epsilon_{zz} + 4c_{44} (\epsilon_{xx}^2 + \epsilon_{yy}^2) + 4c_{66} \epsilon_{xy}^2 \right) \\
= \frac{1}{2} \left( c_{11} + c_{12}/2 \right) (\epsilon_{xx} + \epsilon_{yy})^2 + c_{33} \epsilon_{zz}^2 + 2c_{13} (\epsilon_{xx} + \epsilon_{yy}) \epsilon_{zz} + c_{11} - c_{12}/2 (\epsilon_{xx} - \epsilon_{yy})^2 + 4c_{44} \epsilon_{xx}^2 + 4c_{66} \epsilon_{xy}^2 \\
= \frac{1}{2} \left( c_{A_{1g},1} \epsilon_{A_{1g},1}^2 + c_{A_{1g},2} \epsilon_{A_{1g},2}^2 + 2c_{A_{1g},3} \epsilon_{A_{1g},1} \epsilon_{A_{1g},2} + c_{B_{1g}} \epsilon_{B_{1g}}^2 + c_{E_g} \epsilon_{E_g}^2 + c_{B_{2g}} \epsilon_{B_{2g}}^2 \right) \tag{13}
\]

where the strains are written as the irreducible representations of \(D_{4h}\), \((\epsilon_{xx} + \epsilon_{yy}) \rightarrow \epsilon_{A_{1g},1}\), \(\epsilon_{zz} \rightarrow \epsilon_{A_{1g},2}\), \((\epsilon_{xx} - \epsilon_{yy}) \rightarrow \epsilon_{B_{1g}}, 2\epsilon_{xy} \rightarrow \epsilon_{B_{2g}}\) and \((2\epsilon_{xz}, 2\epsilon_{yz}) \rightarrow \epsilon_{E_g}\).

The lowest order terms that couple strain to the superconducting order parameter must be quadratic in order parameter to preserve gauge symmetry. This coupling gives rise to
FIG. 7. Coupling between strain and one/two component order parameters. (a) Structure of $d_{x^2-y^2}$ gap and (b) $p_y\hat{z}$ gap in k-space, and their couplings to strain. $\hat{z}$ represents the pair wavefunction in spin space. Allowed couplings modify the gap structure in k-space. Only $A_{1g}$ strain couples to one-component order parameters, while $A_{1g}$, $B_{1g}$ and $B_{2g}$ strain all couple to two-component order parameters.

Additional contributions to the free energy

$$F_c = (g_1\epsilon_{A_{1g,1}} + g_2\epsilon_{A_{1g,2}})|\eta|^2 + g_4\epsilon_{B_{1g}}(|\eta_x|^2 - |\eta_y|^2) + g_5\epsilon_{B_{2g}}(\eta_x^*\eta_y + \eta_x\eta_y^*),$$

(14)

where $g_i$ are coupling constants. Coupling between OP and $B_{1g}$, $B_{2g}$ strains are only allowed for two-component OPs; one-component OPs can only couple to compressive strains (shown for example gap structures in Figure 7). These linear-in-strain, quadratic-in-order-parameter coupling lead to elastic moduli jumps at $T_c$. Hence jumps in shear moduli can only occur if the OP is two-component. Since no OP can couple to $E_g$ strain, $c_{44}$ should not show a jump at $T_c$ for any superconducting order parameter.

Following Sigrist[19], we use the parameterization $\eta = (\eta_x, \eta_y) = \eta(\cos \theta, e^{i\gamma} \sin \theta)$. Depending on the relative magnitudes of $b_1, b_2, b_3$, the system can have different equilibrium OPs[43], characterized by different equilibrium values of $(\theta, \gamma) = (\theta_0, \gamma_0)$: $(\pi/4, \pm \pi/2)$ for the chiral state, $(\pi/4, 0)$ for the diagonal nematic state and $(0, 0)$ for the horizontal nematic state. These states also have different equilibrium values of $\eta = \eta_0$, which can be calculated
from $\partial \mathcal{F}_{op}/\partial \eta|_{(\theta,\gamma)\rightarrow(\theta_0,\gamma_0)} = 0$. Fluctuations of the order parameter amplitude $\eta$, orientation $\theta$ or relative phase $\gamma$ can couple to different strains, leading to the jump in corresponding moduli.

To explicitly calculate the moduli discontinuities, we adapt the approach described in [44] to a multi-component OP. For one-component OPs, the discontinuity is

$$c_{mn}^{<} = c_{mn}^{>} - \frac{Z_m Z_n}{Y} \implies \Delta c_{mn} = c_{mn}^{>} - c_{mn}^{<} = \frac{Z_m Z_n}{Y}$$

(15)

where $c_{mn}^{<}(c_{mn}^{>})$ is the elastic modulus below(above) $T_c$. The thermodynamic coefficients $Z_i, Y$ are defined as $Z_i = \partial^2 \mathcal{F}_c/\partial \eta \partial \epsilon_i$ and $Y = \partial^2 \mathcal{F}_{op}/\partial \eta^2$. For a multi-component OP $\eta$, Equation 15 gets modified to

$$\Delta c_{mn} = Z_{m}^{T} Y^{-1} Z_{n}$$

(16)

where $Z_i = \partial^2 \mathcal{F}_c/\partial \eta \partial \epsilon_i$ and $Y = \partial^2 \mathcal{F}_{op}/\partial \eta^2$ are now matrices. Within this formalism, one can find which OP fluctuation mode couples to a particular strain by looking at the $Z_i$ for that strain. For example, for the chiral state,

$$Z_{A_{1g},1(2)} = \begin{pmatrix} g_{1(2)} \sqrt{\frac{-8a}{4b_1-b_2+b_3}} \\ 0 \\ 0 \end{pmatrix}; Z_{B_{1g}} = \begin{pmatrix} 0 \\ g_4 \frac{4a}{4b_1-b_2+b_3} \\ 0 \end{pmatrix}; Z_{B_{2g}} = \begin{pmatrix} 0 \\ 0 \\ g_5 \frac{2a}{4b_1-b_2+b_3} \end{pmatrix}$$

(17)

This shows that $\eta$ fluctuations couple to the $A_{1g}$ strains, $\theta$ fluctuations couple to $B_{1g}$ strain and $\gamma$ fluctuations couple to $B_{2g}$ strain, consistent with the conclusions of Sigrist [19].

The elastic moduli discontinuities for the various OPs is summarized in Table III. In all cases, the three $A_{1g}$ jumps are found to satisfy the relation

$$\Delta c_{A_{1g},1} \times \Delta c_{A_{1g},2} = (\Delta c_{A_{1g},3})^2$$

(18)

Using the measured jumps in $(c_{11} + c_{12})/2$, $c_{33}$, and $c_{13}$ (see Figure 3 of the main text and Equation 13), we obtain $(\Delta c_{11} + c_{12}) \times (\Delta c_{33}) = (9.9 \pm 1.5) \times 10^{-5}$ GPa$^2$, which is in agreement with $(\Delta c_{13})^2 = (8.3 \pm 1.1) \times 10^{-5}$ GPa$^2$.

We now turn to order parameter dynamics near the phase transition, which can cause smearing of the frequency jumps. We start with the idea outlined in [44], and adapt it to a multi-component OP. Below $T_c$, the order parameter relaxation can be modeled as

$$\frac{\partial \tilde{\eta}}{\partial t} = -\xi \frac{\partial \mathcal{F}}{\partial \tilde{\eta}} = -\xi \left( \frac{\partial^2 \mathcal{F}}{\partial \eta^2} \tilde{\eta} + \sum_m \frac{\partial^2 \mathcal{F}}{\partial \eta \partial \epsilon_m} \epsilon_m \right) = -\xi (Y \tilde{\eta} + \sum_m Z_m \epsilon_m)$$

(19)
TABLE III. Different equilibrium order parameters and the discontinuities they produce in various elastic moduli. In parentheses, we note fluctuation of which OP mode couples to ultrasound. Jump in compressional ($A_{1g}$) moduli is always caused by amplitude ($\eta$) fluctuations of the OP, whereas for shear modes, jumps can arise from coupling to amplitude ($\eta$), orientation ($\theta$) or relative phase ($\gamma$) fluctuations.

where $\tilde{\eta}$ are the fluctuations of OP components about equilibrium, and $-\xi \frac{\partial F}{\partial \eta}$ provides the restoring force towards equilibrium. Assuming linear response of OP fluctuations to strain, when strain is modulated at frequency $\omega$, as in a RUS experiment, [Equation 19] becomes

$$-i\omega \tilde{\eta}(\omega) = -\tau^{-1} \tilde{\eta}(\omega) - \xi \sum_m Z_m \epsilon_m \implies \tilde{\eta}(\omega) = (i\omega \tau - \mathbb{I})^{-1} \sum_m Z_m \epsilon_m$$

(20)

where we have defined $\tau^{-1} = \xi Y$ is the matrix of relaxation times for independent OP modes. For the parameterization $\eta = \eta(\cos \theta, e^{i\gamma} \sin \theta)$,

$$\tau = \begin{pmatrix} \tau_\eta & 0 & 0 \\ 0 & \tau_\theta & 0 \\ 0 & 0 & \tau_\gamma \end{pmatrix} \implies (i\omega \tau - \mathbb{I})^{-1} = \begin{pmatrix} (i\omega \tau_\eta - 1)^{-1} & 0 & 0 \\ 0 & (i\omega \tau_\theta - 1)^{-1} & 0 \\ 0 & 0 & (i\omega \tau_\gamma - 1)^{-1} \end{pmatrix}$$

(21)

Using [Equation 20] we calculate the dynamic elastic constant as,

$$c_{mn}(\omega) = c_{mn}^> + \frac{\partial^2 F}{\partial \eta \partial \epsilon_m \partial \epsilon_n} \implies c_{mn}^<(\omega) = c_{mn}^> + Z_m^T (i\omega \tau - \mathbb{I})^{-1} Y^{-1} Z_n$$

(22)

Elastic moduli jumps come from the real part of [Equation 22]. Depending on which OP mode a particular elastic moduli couples to, the modulus dispersion $c_{mn}(\omega)$ picks up a contribution from the corresponding relaxation time. For example, for the chiral OP,

$$\Delta c_{A_{1g},1(2)} = \frac{2g_1^2}{b_1 - b_2 + b_3} \frac{1}{1 + \omega^2 \tau_\eta^2}$$

$$\Delta c_{B_{1g}} = \frac{2g_4^2}{b_2 - b_3} \frac{1}{1 + \omega^2 \tau_\theta^2}$$

$$\Delta c_{B_{2g}} = \frac{g_5^2}{b_2} \frac{1}{1 + \omega^2 \tau_\gamma^2}$$
Thus OP relaxation effects can broaden out the elastic moduli jumps, if particular relaxation times are long compared to the experimental frequencies. This has been observed experimentally, for example, in the cuprate superconductor LSCO\cite{45}—higher frequencies reduce the magnitude of jump measured. Since we measure non-zero discontinuities in all the $A_{1g}$ moduli, and the $B_{2g}$ modulus, but no jump in $B_{1g}$ modulus, it is plausible that the $B_{1g}$ jump gets strongly smeared due to this effect. Specifically, for the diagonal nematic state, only the $B_{1g}$ jump is affected by $\tau_\theta$, while the other 4 jumps are related to $\tau_\eta$. Hence if $\tau_\theta$ is much larger than $\tau_\eta$ for this state, it provides an explanation for the lack of $B_{1g}$ jump. We also note that for this state, close to $T_c$ ($1 - T/T_c \ll 1$), $\tau_\eta^{-1} \propto |a| \propto \eta_0^2 \propto (T_c - T)$ and $\tau_\theta^{-1} \propto \eta_0^4 \propto (T_c - T)^2$. This would indeed make $\tau_\theta$ much longer than $\tau_\eta$ just below $T_c$. We also note that large ultrasonic attenuation is observed experimentally in the $B_{1g}$ channel \cite{17}, which perhaps motivates the presence of such a long relaxation time.

Reconciling Resonant Ultrasound and Pulse Echo Ultrasound Experiments

The $c_{66}$ discontinuity we measure with RUS is about a factor of 50 larger than what was measured with pulse-echo experiments\cite{17}. This apparent discrepancy may be resolved by looking at the frequency scales of the two experiments: $\sim 2.5$ MHz for RUS versus 169 MHz for pulse-echo. As noted in the previous section, higher frequencies are expected to reduce the magnitude of the experimentally measured jump. This has been observed at the superconducting transition in La$_{1.85}$Sr$_{0.15}$CuO$_4$, where the jump in $c_{33}$ decreases by a factor of $\sim 4$ when the measurement frequency is increased from 16 MHz to 214 MHz.

Applying a simple relaxation model, like the one derived in the previous section for order-parameter relaxation near the phase transition (Equation 22), we obtain

$$\frac{\Delta c(2.5 \text{ MHz})}{\Delta c(169 \text{ MHz})} = \frac{(1 + (2.5 \text{ MHz} \cdot \tau)^2)^{-1}}{(1 + (169 \text{ MHz} \cdot \tau)^2)^{-1}} = 50 \implies \tau \sim 6.6 \text{ ns} \quad (23)$$

where $\tau$ is a relaxation timescale. Applying the same model to the La$_{1.85}$Sr$_{0.15}$CuO$_4$ data from Nyhus et al. \cite{45} we obtain 1 ns — a comparable timescale. Whatever the microscopic mechanism underlying this timescale, it is clear that lower-frequency measurements should measure a jump that is closer to the intrinsic thermodynamic jump.
Ehrenfest Relations for Compressional Strains

At the superconducting transition, a jump discontinuity is also measured in the specific heat. Within our formalism, the specific heat jump at $T_c$, $\Delta C/T_c$, is calculated as

$$\frac{\Delta C}{T} = W^T Y^{-1} W$$

(24)

where $W = \partial^2 F_{op}/\partial \eta \partial T$ and $Y = \partial^2 F_{op}/\partial \eta^2$. For all three superconducting states discussed above, the $A_{1g}$ moduli jumps can be related to the specific heat jump through

$$\Delta c_{A_{1g},1(2)} = - \frac{\Delta C}{T} \left( \frac{dT_c}{d\epsilon_{A_{1g},1(2)}} \right)^2 ; \Delta c_{A_{1g},3} = - \frac{\Delta C}{T} \left| \frac{dT_c}{d\epsilon_{A_{1g},1}} \right| \left| \frac{dT_c}{d\epsilon_{A_{1g},2}} \right|$$

(25)

It is important to note that such relations for the shear strains are more complicated, and we derive them in the next section. For a tetragonal material, the bulk modulus $B$ is related to the three $A_{1g}$ moduli as

$$B = \frac{(c_{11}+c_{12})c_{33}-c_{13}^2}{(c_{11}+c_{12})+2c_{33}-2c_{13}}$$

(26)

From Equation 25, the discontinuity in the bulk modulus $\Delta B/B$ at $T_c$ can be related to $\Delta C/T$ through the Ehrenfest relation

$$\frac{\Delta B}{B^2} = - \frac{\Delta C}{T} \left( \frac{dT_c}{dP_{hyd}} \right)^2$$

(27)

where $dT_c/dP_{hyd}$ is the hydrostatic pressure dependence of $T_c$. Our measurements give $\Delta B/B \sim 6.3 \times 10^{-5}$, about 9 times larger than estimated from specific heat jump and $dT_c/dP_{hyd}$ [46] or, alternatively, the measured jump in the bulk modulus predicts $dT_c/dP_{hyd}$ to be a factor of 3 higher than the measured value. This discrepancy can arise due to the formation of order parameter domains [47], which lead to an additional slowing down of ultrasound and therefore a larger drop in the elastic moduli through $T_c$. Since these domains would be related to each other by time-reversal in a superconductor, however, it is unclear whether such a mechanism would couple strongly to ultrasound. Another possible cause could be the value of $dT_c/dP_{hyd}$ estimated from the data in [46]. Since the $T_c$ of Sr$_2$RuO$_4$ shows a strong increase with $B_{1g}$ strain[24], the measured decrease in $T_c$ under $P_{hyd}$ will be less if the pressure applying medium is not completely hydrostatic. This is particularly relevant because the $B_{1g}$ modulus is almost 4 times smaller than $(c_{11} + c_{12})/2$, which makes it easy to induce $B_{1g}$ strain if the pressure medium is not hydrostatic.
Finally, with the possible discovery of two transitions occurring either simultaneously or near-simultaneously at $T_c$, it will be necessary to map out $T_{RSB}$ with pressure to correctly calculate the Ehrenfest relations, which are modified under the presence of two accidentally degenerate order parameters \[39\].

\textbf{Ehrenfest Relations for Shear Strains}

Unlike $A_{1g}$ strains, shear strains ($B_{1g}$ and $B_{2g}$) are expected to split the superconducting transition if the OP is a symmetry-protected multi-component order parameter \[24\]. This happens because shear strains break the tetragonal symmetry of the lattice, and hence the degenerate OP components of the unstrained crystal now have different condensation energies (and temperatures). Within weak coupling, a crystal under shear strain should therefore show two specific heat jumps\[25\], and, for a chiral OP, time-reversal symmetry breaking (TRSB) should set in at a different temperature than Meissner effect. Recent $\mu$SR experiments\[23\] have indeed reported the latter effect. We show that the shear modulus jump can be related to $\Delta C/T$ (at zero strain) through the strain derivatives of these two transition temperatures, $T_c$ and $T_{RSB}$,

\[
\Delta c_s = -\frac{\Delta C}{T} \left| \frac{dT_1}{d\epsilon} \right| \left| \frac{dT_2}{d\epsilon} \right|,
\]

where $s$ is either $B_{1g}$ or $B_{2g}$, $T_1 = T_c$, and $T_2 = T_{RSB}$.

We start from the free energy expressions $F_{op}$ and $F_c$, and consider the case $b_2 > 0, b_3 < b_2$, which leads to a chiral OP. Further, we keep only the coupling to $\epsilon_{B_{1g}}$ to simplify the subsequent algebra. Then, with the phase between $\eta_x$ and $\eta_y$ set to $\pi/2$, $F_{op}$ and $F_c$ are

\[
F_{op} = a_0(T - T_{c,0})(\eta_x^2 + \eta_y^2) + b_1(\eta_x^2 + \eta_y^2)^2 + (b_3 - b_2)\eta_x^2\eta_y^2
\]

\[
F_c = g_4 \epsilon_{B_{1g}} (\eta_x^2 - \eta_y^2),
\]

where $T_{c,0}$ is the unstrained $T_c$. Clearly, $\epsilon_{B_{1g}}$ breaks the $\eta_x \leftrightarrow \eta_y$ symmetry of the quadratic terms in free energy, thereby making the two components condense at different temperatures.

We assume $g_4 \epsilon_{B_{1g}} > 0$, which favors $\eta_y$ condensing before $\eta_x$. The higher transition temperature $T_1 = T_c$ is determined by when the coefficient of $\eta_y^2$ goes to zero (with $\eta_x = 0$), that is, $a_0(T_1 - T_{c,0}) - g_4 \epsilon_{B_{1g}} = 0$. This gives

\[
T_1 = T_{c,0} + \frac{g_4}{a_0} \epsilon_{B_{1g}}.
\]
Then, the $\eta_y$ that minimizes ($F_{op} + F_c$) is
\[
\eta_y^2 = \frac{a_0(T_{c,0} - T)}{2b_1} + g_4 \epsilon_{B_{1y}} = \frac{a_0(T_1 - T)}{2b_1}.
\] (31)

Further, the specific heat jump at this transition, calculated by using the above $\eta_y^2$, is
\[
\left( \frac{\Delta C}{T} \right)_1 = \frac{a_0^2}{2b_1}.
\] (32)

Below $T_1$, the system undergoes TRSB transition when $\eta_x$ condenses. Naively, one might expect this to occur at $T_{c,0} - g_4 \epsilon_{B_{1y}}/a_0$, found by setting the quadratic coefficient of $\eta_x$ to zero. The condensation of $\eta_y$ prevents this, however, through the $\eta_x^2 \eta_y^2$ terms in $F_{op}$. If the coefficient of this term is zero ($2b_1 + b_3 - b_2 = 0$), then there is no competition between $\eta_x$ and $\eta_y$, in which case the second transition does occur at $T_{TRSB} = T_{c,0} - g_4 \epsilon_{B_{1y}}/a_0$.

For the more general case, when $2b_1 + b_3 - b_2 \neq 0$, $T_2 = T_{TRSB}$ is calculated by setting the coefficient of $\eta_x^2$ to zero in the total free energy, with $\eta_y$ given by Equation 31. This gives
\[
a_0(T_2 - T_{c,0}) + (2b_1 + b_3 - b_2) \eta_y^2 + g_4 \epsilon_{B_{1y}} = 0
\implies T_2 = T_{c,0} - \frac{g_4}{a_0} \left( \frac{4b_1 - b_2 + b_3}{b_2 - b_3} \right) \epsilon_{B_{1y}}.
\] (33)

The specific heat jump at this transition can be calculated by subtracting the jump in first transition ($\Delta C/T$)$_1$ from the total jump $\Delta C/T$ in the unstrained case.
\[
\left( \frac{\Delta C}{T} \right)_2 = \frac{2a_0^2}{4b_1 - b_2 + b_3} - \frac{a_0^2}{2b_1} = \frac{a_0^2}{2b_1} \frac{b_2 - b_3}{4b_1 - b_2 + b_3}
\] (34)

The ratio of the two specific heat jumps can then be related by
\[
\left( \frac{\Delta C}{T} \right)_1 / \left( \frac{\Delta C}{T} \right)_2 = \frac{4b_1 - b_2 + b_3}{b_2 - b_3} = \left| \frac{dT_2}{d\epsilon_{B_{1y}}} \right| / \left| \frac{dT_1}{d\epsilon_{B_{1y}}} \right|
\] (35)

Below $T_2$, the order parameter $(\eta_x, \eta_y)$ can be calculated by minimizing ($F_{op} + F_c$) with respect to both $\eta_x$ and $\eta_y$. This gives
\[
\eta_x^2 = \frac{a_0(T_2 - T)}{4b_1 - b_2 + b_3},
\eta_y^2 = \frac{a_0}{2b_1} \left( (T_1 - T) - \frac{2b_1 - b_2 + b_3}{4b_1 - b_2 + b_3}(T_2 - T) \right).
\] (36)

It is interesting to note that the condensation of $\eta_x$ at $T_2$ decreases the rate at which $\eta_y$ was growing below $T_1$, demonstrating the competition between the two components (see Figure 8).
FIG. 8. **Strain-induced splitting of the transition temperature** $T_{c,0}$. Under $B_{1g}$ shear strain, the two components $\eta_y$ and $\eta_x$ condense at different temperatures, $T_c$ and $T_{TRSB}$, respectively. Above $T_c$, both the components are zero, and the sample is not superconducting. At $T_c$, the Meissner effect sets in, and finally, below $T_{TRSB}$, the system becomes a chiral superconductor. Note that the condensation of $\eta_x$ decreases the rate at which $\eta_y$ was growing below $T_c$. Qualitatively similar behavior is expected for $B_{2g}$ shear strain, see text for details.

The jump in the $B_{1g}$ shear modulus for chiral OP can now be expressed as,

$$\Delta c_{B_{1g}} = 2g_1^2 = \frac{2a_0^2}{b_2 - b_3} \cdot \frac{g_1}{a_0} \cdot \frac{g_4}{b_2 - b_3} \cdot \frac{4b_1 - b_2 + b_3}{b_2 - b_3} = -\frac{\Delta C}{T} \left| \frac{dT_1}{d\epsilon_{B_{1g}}} \right| \left| \frac{dT_2}{d\epsilon_{B_{1g}}} \right| \quad (37)$$

A similar derivation can be carried out for $B_{2g}$ strain. This can be performed simply by re-defining the order parameter variables as $\bar{\eta}_x = (\eta_x + \eta_y)/\sqrt{2}$ and $\bar{\eta}_y = (\eta_x - \eta_y)/\sqrt{2}$ and carrying out the same calculation as the $B_{1g}$ case.

The above derivation assumes that the spontaneous strains generated in the crystal below the first transition are small, such that the quartic coefficients in Landau theory are not strongly renormalized below $T_1$. We also assume that the TRS-breaking transition under finite strain is a second order phase transition; whether this holds in Sr$_2$RuO$_4$ is a question for future studies.
Reconciling the $c_{66}$ Discontinuity with Experiments Under Finite Strain

It follows from our measurement of a non-zero discontinuity in $c_{66}$ that two transitions should occur under $\epsilon_{B_{2g}} = \epsilon_{xy}$ strain, each showing a specific heat jump, and $T_c$ must split linearly with $\epsilon_{xy}$ strain (see Equation 30). However, past experiments[24, 25] have not observed either of these effects, and we comment on that here.

Experimental resolution of specific heat measurements under strain [25], and a lack of an observed discontinuity at a lower transition ($T_{\text{TRSB}} \equiv T_2$) gives a bound on the ratio of specific heat jumps at the purported transitions 1 and 2 as,

$$\left(\frac{\Delta C}{T}\right)_1 / \left(\frac{\Delta C}{T}\right)_2 = \left|\frac{dT_2}{d\epsilon_{B_{2g}}}\right| / \left|\frac{dT_1}{d\epsilon_{B_{2g}}}\right| \geq 20. \tag{38}$$

From the jumps in the $B_{2g}$ modulus, $\Delta c_{66} \approx 10^6$ Pa, and in the specific heat, $\Delta C/T = 25$ mJ mol$^{-1}$ K$^{-2} \approx 450$ J m$^3$ K$^{-2}$, we get

$$\left|\frac{dT_1}{d\epsilon_{B_{2g}}} \right| 2000 \text{ K}^2. \tag{39}$$

From these two equations, we can estimate the shifts in the transition temperatures with strain as

$$\left|\frac{dT_1}{d\epsilon_{B_{2g}}}\right| \leq 10 \text{ K} = 0.1 \text{ K/}\%_{\text{strain}}$$

$$\left|\frac{dT_2}{d\epsilon_{B_{2g}}}\right| \geq 200 \text{ K} = 2 \text{ K/}\%_{\text{strain}} \tag{40}$$

We can now compare these estimates to what has been experimentally observed. $T_c$ as a function of $\epsilon_{xy}$ was reported in Hicks et al. [24], and the resolution on a possible cusp was 0.1 K/%; therefore the data of Hicks et al. [24] do not rule out a cusp of the magnitude predicted here. Furthermore, in recent $\mu$SR experiments under applied $B_{1g}$ strain[23], a modest suppression of $T_{\text{TRSB}}$ is reported: $\sim 0.2$ K under a stress of -0.28 GPa, or a strain of $\sim -0.2\%$, for a slope of $\sim 1$ K/%. Although measurements under $B_{2g}$ strain have not yet been reported, we note that this is comparable to the slope indicated above.