High precision heat capacity measurements on Sr$_2$RuO$_4$ under uniaxial pressure

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The heat capacity is one of the most fundamental properties that can be studied in any material, vital in the search for exotic phase transitions. Given the rapid developments in tuning the physical properties of solids by the application of external stimuli, it is particularly important to extend the circumstances over which heat capacity measurements can be made, to include tuned systems. Developments in recent years have highlighted the potential of uniaxial pressure to become a major tuning technique in the field, and its compatibility with transport, magnetic, spectroscopic and scattering experiments has been demonstrated $^1$–$^9$. Until now, the small pressure range and the coupling of the samples to their environment have strongly limited heat capacity measurements under uniaxial pressure $^{10}$–$^{14}$. Here, we demonstrate that they can be done up to large pressures. We use a highly non-adiabatic ac technique at frequencies of several kilohertz to study Sr$_2$RuO$_4$. In doing so we both provide a stringent test of our new technique by applying it to a system whose phase transitions are among the most disorder-sensitive ever discovered, and obtain thermodynamic information from a material whose unconventional superconductivity is a topic of considerable current interest.

Understanding the superconductivity of Sr$_2$RuO$_4$ has reached a pivotal stage. It has long been known to be an unconventional superconductor, as evidenced by the extreme sensitivity of its superconducting state to non-magnetic disorder $^{15}$. For nearly two decades, a nuclear magnetic resonance (NMR) Knight shift measurement gave what was assumed to be irrefutable evidence for the spin susceptibility remaining constant on entering the superconducting state $^{16}$. This observation could only be consistent with odd parity, spin triplet order parameters whose vector order parameter was unidirectional $^{17}$–$^{21}$. A natural proposal was the two-component $d=\hat{z}(k_x \pm ik_y)$ state, in which time reversal symmetry (TRS) is broken. The past few months have seen a seismic shift, however, because new NMR Knight
shift studies identified a previously overlooked problem with sample heating in the original work, and showed that the spin susceptibility actually drops substantially on entry to the superconducting state, ruling out any vector order parameter of the $\hat{z}$ form and quite possibly any odd parity state at all. In contrast, the issue of TRS breaking remains wide open. Evidence for a two-component order parameter has come most notably from measurements of the Kerr effect \(^2,\) muon spin rotation \(^4\), Josephson interferometry \(^5,\) and ultrasound. How this evidence might be reconciled with the strong possibility that the order parameter is spin-singlet and even-parity is a mystery. The most obvious possibility for chiral, even-parity order on a tetragonal lattice, $\Delta \sim d_{xz} \pm id_{yz}$, has a horizontal line node at $k_z = 0$. This implies that there are no pairing correlations within each RuO$_2$ layer, which would be a surprise for a metal as anisotropic as Sr$_2$RuO$_4$. Because the evidence for two-component order comes from multiple experiments, this issue should be investigated further.

The deductive path to the reversal of the received wisdom regarding the superconducting state of Sr$_2$RuO$_4$ was strongly linked to the introduction of uniaxial pressure as a tool for its study. Pressure applied along the [100] direction of Sr$_2$RuO$_4$ increases the superconducting transition temperature $T_c$ by a factor of 2.3, from 1.5 K in unstrained material to a maximum of 3.5 K at a strain of approximately 0.5\% \(^3\). This increases the superconducting critical fields, which made the critical field limiting for in-plane fields more clear, and increased the robustness of the superconductivity against the radio-frequency pulses for NMR measurements. Now that the field of Sr$_2$RuO$_4$ superconductivity has been thrown open, it is particularly important to have accurate knowledge of the uniaxial pressure dependence of foundational thermodynamic properties, to establish a baseline for the re-examination of other candidate order parameters. In this regard, one issue has assumed special significance. A key feature of uniaxial pressure is that it is expected to break the degeneracy that would lead to the formation of any two-component order parameter thought to be possible in a tetragonal material such as Sr$_2$RuO$_4$ \(^6\). One predicted consequence of this is a cusp in $T_c$ as the pressure is swept through zero. This has not been seen in measurements of $T_c$ through magnetic susceptibility \(^2,\) but there is always the concern that this is an issue of experimental resolution. A more direct route would be to observe the second transition. This cannot be done in the magnetic susceptibility but can be achieved in principle by studying the heat capacity, which is sensitive to transitions within the superconducting state and was pivotal in establishing the existence of such a transition in UPt$_3$ \(^7\). A further possibility raised by the work reported in \(^3\) is that of a strain-dependent transition between two order parameters. These might both be superconducting \(^2\), but the possibility of competing magnetic phases also exists \(^8\). Again, the heat capacity is an ideal way to search for such strain-dependent phase transitions. However, performing heat capacity measurements in a uniaxial pressure cell is extremely challenging, because samples tend to break at relatively small uniaxial pressures and because of the generally large thermal conductance between the sample and its environment.

To address this fundamental experimental challenge, we employ a variation of known a.c. heat capacity measurement techniques \(^9\) on a sample in a uniaxial pressure rig \(^1\) to obtain bulk thermodynamic data in this most extreme environment. In Fig. 1a we show a photograph of a bar of single crystal Sr$_2$RuO$_4$ that has been carefully cut along its [100] direction, polished, and then mounted with in the jaws of the uniaxial pressure rig using Stycast 2850FT epoxy (Henkel Loctide) \(^1\). A resistive thin film resistor chip (State of the Art, Inc.) as heater and a calibrated Au-AuFe(0.07\%) thermocouple are fixed to opposite sides of the sample using...
Dupont 6838 silver epoxy. Four independent experimental runs using different samples (#1 – #4) proved the reliability of the results.

**Fig. 1 Sample configuration and characteristics.** **a,** A photograph of the setup of the heat capacity measurements under strain including heater and thermometer. The sample is glued between the jaws of the uniaxial pressure device. It allows the application of compressive and tensile strains. The red, yellow and white dashed lines indicate the homogeneous, inhomogeneous and unstrained regions schematically. **b,** Frequency sweeps for sample #3 at $T = 1$ K and 4.23 K. **c,** Heat capacity measurements under a strain -0.19% for sample #3 at various excitation frequencies. The dashed (dotted) line indicates the $T_c$ from strained (unstrained) part of the sample.

The governing relationship for measurements of the a.c. heat capacity $C_{ac}$ is

$$C_{ac} = \frac{P \omega}{\omega T_{ac}} F(\omega).$$

The power $P$, angular frequency $\omega$ and oscillatory temperature $T_{ac}$ are known or can be precisely measured, but the frequency response curve $F(\omega)$ differs for each sample, because it depends on time constants determined by the geometry, thermal conductances, and heat capacities of the system. A measurement of $F(\omega)$ for one of our $\text{Sr}_2\text{RuO}_4$ samples is shown in Fig. 1b, and is seen to follow the expected form for measurement under highly non-adiabatic conditions: $F(\omega)$ is reduced at low frequencies due to dissipation of temperature oscillations into the environment (here the body of the uniaxial apparatus), and at high frequencies because the system does not thermalise. The plateau region between these limits is, in general, the region in which a heat capacity measurement can be made successfully.
For the current experiment, however, the conditions are still more demanding. The nature of the apparatus means that only the central portion of the Sr$_2$RuO$_4$ crystal is homogeneously strained. Force is transferred to the sample through the epoxy layer around the sample; the sample ends are unstrained, and there are intermediate regions where the strain is built up. In the lower frequency part of the plateau in Fig. 1b, temperature oscillations extend throughout the sample and all three regions are probed. This is shown in the 313 and 613 Hz data in Fig. 1c: a higher-temperature peak is visible, corresponding to $T_c$ of the central portion, and a lower-frequency peak, corresponding to $T_c$ of the sample ends. To avoid this, one has to work at the high end of the feasible range of frequencies, where temperature oscillations do not extend into the end regions. For this particular sample, a measurement frequency above ~1.5 kHz was required to satisfy this more demanding criterion. The main results that we will present were taken at frequencies in the range 3.5 – 4 kHz, and with low power input to minimize sample heating. Working at those high frequencies gives a very low signal, with an r.m.s. thermocouple voltage of only 1 – 2 nV, so low temperature passive amplification was employed to achieve an r.m.s. noise level of 20 pVHz$^{-1/2}$, ensuring a signal to noise ratio in excess of 50. A consequence of having to operate the experiment in this mode is that the probed volume depends on the thermal conductivity of the sample as well as the frequency and its heat capacity, so the directly measured quantity $T_{ac}$ is only an approximation to the true volume specific heat capacity $c_v$. As we show below, it can be used to calculate $c_v$ if the temperature dependence of the thermal conductivity is known, but we stress that the raw $T_{ac}$ data are already enough to address some key physics questions, so we begin for simplicity by showing the unprocessed experimental signal.

In Fig. 2 we show representative raw data demonstrating our capability of resolving the a.c. specific heat signal of the superconducting phase transition at strains below (Fig. 2a) and above (Fig. 2b) that at which $T_c$ is maximized. The raw signal contains a background, which we illustrate for an unstrained sample by applying a small 0.1 T magnetic field to suppress the superconductivity (dotted line in Fig. 2a). The breadth of the transition leading edge is due to residual strain inhomogeneity, due to bending and sample defects, and so is approximately proportional to the strain derivative of $T_c$ (Fig. 2c). We estimate this residual inhomogeneity to be 10% of the applied strain; see Supplementary Information. Over most of the range of measurement the transition is well resolved, and the results for the strain dependence of the transition temperature (estimated as the mid-point of the leading edge of the heat capacity anomaly) are in excellent agreement with previous studies of the diamagnetic response (Fig. 2c and ref. 3).

Since a key motivation for our experiment was to look for transition splitting and / or strain-dependent transitions between differing superconducting order parameters, we worked at higher excitation powers to maximize our signal-to-noise ratio, and studied the superconducting transition in fine strain steps. The results are shown in Fig. 3. The above-mentioned transition broadening is again evident at intermediate strains but the data reveal no convincing evidence for transition splitting. Qualitatively, they show a smooth growth in the height of the anomaly, but no sudden changes that would be indicative of a transition between two different order parameters.
**Fig. 2 Heat capacity measurements for sample #4 under various strains.**

- **a**, Measurements before the peak in $T_c$. Measurements after the peak in $T_c$. The dashed line in panel (a) is the heat capacity measurement at $\mu_0H_{\parallel c} = 0.1$ T and $\varepsilon_{xx} = 0\%$. The dashed line in panel (b) repeats the data at maximum $T_c$.
- **b**, Superconducting transition temperature against strain for three experimental runs using different samples taken with the similar frequencies. $T_c$ of sample #4 has been probed for compressive and tensile strains. Solid points are $T_c$ taken as midpoints of the leading edge of the transitions ($50\%$ level). The coloured area represents the width of the transitions from $20\%$ to $80\%$ levels.
Fig. 3 Evolution of the heat capacity anomaly. \((\frac{T_{ac}}{T})^{-1}\) against temperature for sample #4 at a series of small increments of compressive strains up to the peak in \(T_c\). Smaller increments of strain and a larger power were used to reduce the noise and to allow more precise inspection of the presence of a transition between different order parameters inside the superconducting state.

In Figs. 2 and 3 we deliberately presented unprocessed data based on our calibrated measurement of the a.c. temperature change measured by the thermocouple, because those raw data already enable us to prove the existence of bulk superconductivity and search for transitions between different superconducting order parameters. Going further requires calculation, and in Fig. 4 we show two examples of what can be done.

Under an assumption that the heater is narrow and the sample width is small in comparison with the thermal diffusion length, the signal is related to the true volume heat capacity \(c_v\) by

\[
\frac{1}{T_{ac} T} = \frac{P + F(\omega)}{2A\sqrt{\omega \kappa(T)c_v(T)}} ,
\]

in which \(A\) is the cross-sectional area of a long bar and \(\kappa\) is the thermal conductivity. In principle, a full temperature dependence of \(c_v\) could be calculated by inverting this expression if the thermal conductivities were known at all temperatures and strains. In practice this is not the case, but the strain evolution of the size of jump in true heat capacity \(\Delta c_v\) as a function of strain, normalized to the normal state value, can be tracked by plotting the ratio \((\frac{T_{ac}}{T_c})^2\).

This is depicted in Fig. 4a. The data well below the transition contain systematic uncertainty because of the unknown change in \(\kappa(T)\) but since \(\kappa\) remains continuous at the transition itself, the information about \(\Delta c_v/c_v^n\) is reliable. This ratio is observed to grow slightly as \(T_c\) is increased.
Fig. 4 The square of the normalized heat capacity curves and the calculations for the specific heat. 

(a) $(T_{ac,n}/T_{ac,s})^2$ against temperature for sample #4 at a series of compressive strains up to the peak in $T_c$. The calculations of normalized specific heat curves at zero strain from our ac calorimetry by inverting Eq. 2 and using the thermal conductivity from ref.30. The red and blue curves are the calculations for sample #4 with $f_{exc} = 2333$ Hz and 3913 Hz, respectively. The black curve is the published data from Deguchi et al. 31.

Although we do not have sufficient information to invert Eq. 2 at all strains, enough is known about $\kappa(T)$ at zero strain to allow this to be attempted. The results, shown in Fig. 4b, demonstrate the level of accuracy that can be achieved using the present set-up (for which Eq. 2 is not a perfect description, as discussed in the Supplementary Information). In future, improved experimental design including in situ measurement of thermal conductivity should in principle enable fully quantitative measurements of $c_v(T)$ under strain.

The results presented in this paper demonstrate several important advances. First and foremost, they show that the heat capacity of a superconductor can be measured in an extended pressure range in a uniaxial pressure device providing access to compressive as well as tensile strains. The thermodynamic information that we have obtained will form an
important part of the on-going quest to understand the superconductivity of Sr$_2$RuO$_4$. Our results show conclusively that the increase of $T_c$ demonstrated in refs. 2 and 3 is a bulk phenomenon; a surface effect cannot be ruled out by resistivity and magnetic susceptibility measurements alone. Inspection of the data shown in Figs. 2 and 3 demonstrates qualitatively that we resolve no splitting of the superconducting transition as a function of temperature at any sample strain. A more quantitative analysis based on data averaged across many runs (Supplementary Information) shows that if there is a smaller second transition that we do not resolve because of a signal to noise issue, it must be less than a few per cent of the primary one, and that if, alternatively, it is hidden because it stays very close in temperature to the primary transition, that splitting at low or high strains must be less than approximately 50 mK. A second piece of physics that can be inferred directly from Fig. 3 is the lack of any obvious transition within the superconducting state as the strain is altered. Such a transition might be expected to lead to a sharp change in the size of the heat capacity anomaly; none is resolved in our data. Finally, we draw attention to the significance of the $\Delta c_v/c_v^n$ data of Fig. 4a. Qualitatively, an increase in this ratio, as we observe in our measurements, indicates that the superconducting gap shifts its weight towards regions of the Brillouin zone in which the density of states is large. This seems inconsistent with the expected situation for an odd parity order parameter, which, because it must vanish at the Van Hove point, is expected to shift its weight away from the high-density-of-states region around this point.

Taken at face value and in isolation, our data look more consistent with a single component even parity order parameter than with a two-component one, and are even more deeply in conflict with a two-component odd parity state. Their broader significance will come as a fundamental baseline against which to reference the results of other experiments. The experiments that indicate a two-component order parameter in Sr$_2$RuO$_4$ do not provide information on the energy scale associated with TRS breaking. It is therefore still possible that they are consistent with our findings here, with the second transition below our resolution. However, we have been able to place crucial and quantitatively testable thermodynamic constraints on such transitions. As the physics of Sr$_2$RuO$_4$ enters an exciting new phase, we also foresee a fruitful combination of the kind of information presented here with data from ongoing ultrasound studies in which anomalies observed at $T_c$ also suggest the existence of more than one order parameter component in the superconducting state. For the longer term, there is no need to restrict investigation to Sr$_2$RuO$_4$. Our work invites equivalent thermodynamic uniaxial pressure studies on other unconventional superconductors and, possibly, materials with other exotic correlated electron ground states.
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Methods

High-quality single-crystal Sr$_2$RuO$_4$ samples were grown in a floating zone furnace (Canon Machinery) using techniques refined over many years to those described recently in ref. They were aligned using a bespoke Laue x-ray camera, and cut using a wire saw into thin bars with whose long axis aligned with the [100] direction of the crystal. For the best results these bars were polished using home-made apparatus based on diamond impregnated paper with a minimum grit size of 1 µm. Special care was taken when epoxying to the pressure cell to minimize tilt and ensure as homogeneous a strain field as possible. During the course of the experiments we discovered that differential thermal contraction due to the coupling between the heater and sample can itself be a significant source of strain imhomogeneity so we settled on a flexible coupling made of thin silver strips, making multiple contacts to the central part of the sample as shown in Fig. 1a.

The uniaxial pressure apparatus was mounted on a dilution refrigerator, with thermal coupling to the mixing chamber via a high purity silver wire. The data shown in the paper were acquired between 500 mK and 4.2 K, with operation above 1.5 K achieved by circulating a small fraction of the mixture. The thermocouple was spot-welded in-house and its calibration fixed by reference to that of a calibrated RuO$_2$ thermometer. The extremely low noise level of 20 pV/√Hz on the thermocouple readout was achieved by the combination of a low temperature transformer mounted on the 1K pot of the dilution refrigerator, operating at a gain of 300, and an EG&G 7265 lock-in amplifier. A Keithley 6221 low-noise current source was used to drive the heater. The piezo electric actuators were driven at up to ± 400 V using a bespoke high-voltage amplifier.

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Author contributions

The idea to try this programme of research came from M.N., and the appropriate experimental apparatus was conceived by M.N. and C.W.H. and developed by Y.-S.L.. The large majority of the experiments and data analysis was performed by Y.-S.L. with contributions from M.N., C.W.H. and F.J., on crystals grown and characterized by N.K., A.S.G., D.A.S.. F.J. and Y.M.. A.P.M. oversaw the complete project and drafted the paper, with input from Y.-S.L., M.N., C.W.H. and Y.M..
SUPPLEMENTARY INFORMATION

Heat capacity measurements in the temperature range between 0.5 and 4 K

In the main text, we have shown the heat capacity measurements between 1 and 4 K taken at a frequency of 3913 Hz. In Fig. S1 we present additional data at 2333 Hz in an extended temperature range down to 0.5 K. For each strain two data sets recorded in different temperature regions, from 0.5 K to 1.3 K and from 1 K to 4 K, have been combined. The data sets of the two different measurement runs coincide in an excellent way in the overlap region. The increase in \( T_c \) with strain is consistent with the result at \( f_{\text{exc}} = 3913 \) Hz. Within our experimental resolution, we do not find any indication of an additional anomaly below the superconducting transition at any given strain.

![Diagram of 1\(/
T_{ac}\) against temperature for different strains up to the peak in \( T_c \). The dashed line is a heat capacity measurement at \( \mu_0 H |_{\epsilon} = 0.1 \) T and \( \epsilon_{xx} = 0\% \).]

**Fig. S1** Heat capacity measurements for sample #4 at \( f_{\text{exc}} = 2333 \) Hz under various strains. \( 1/T_{ac} \) against temperature for different strains up to the peak in \( T_c \). The dashed line is a heat capacity measurement at \( \mu_0 H |_{\epsilon} = 0.1 \) T and \( \epsilon_{xx} = 0\% \).

The relation between the heat capacity and the specific heat

The conversion between the heat capacity and specific heat in a conventional setup is trivial, since the volume (or mass) of the sample is constant. In our measurements, the probed sample volume varies since the thermal diffusion length changes as a function of temperature. Therefore, it is nontrivial to convert our heat capacity data to specific heat. We start with an ideal case to demonstrate the relation between heat capacity and specific heat in case of our experimental setup.
Suppose that the heater contact is point-like and the sample is very narrow such that the heat flow is one-dimensional. The probed volume $V$ is equal to the cross-sectional area $A$ times twice, heat propagating on both sides, the diffusion length $l_d$, which is a function of the frequency $\omega$, the volume specific heat $c_v$ and the thermal conductivity $\kappa$.

$$l_d = \sqrt{\frac{\kappa(T)}{\omega c_v(T)}}$$  \hspace{1cm} (S1)

The experimentally obtained heat capacity $C_{ac}$ can be expressed as follows:

$$C_{ac} = c_v \times V = c_v \times A \times 2 \sqrt{\frac{\kappa}{\omega c_v}} = \frac{2A}{\sqrt{\omega}} \sqrt{\kappa(T)c_v(T)}$$  \hspace{1cm} (S2)

By inserting equation (S2) into equation (1), the temperature readout from the thermocouple is:

$$T_{ac} = \frac{P * F(\omega)}{2A\sqrt{\omega} * \sqrt{\kappa(T)c_v(T)}}$$  \hspace{1cm} (S3)

The excitation frequencies in our current measurements are not far away from the upper cut-off frequency, which describes the time scale for the heat propagating from the heater to the thermocouple, so the frequency response at the excitation frequencies $F(\omega) < 1$ and depends on temperature. As a consequence, it is not possible to obtain the specific heat in the current sample configuration. However, we can normalize the data in the superconducting state by the normal state data to eliminate $F(\omega)$:

$$\frac{C_{ac}^s}{C_{ac}^n} = \frac{T_{ac}^n}{T_{ac}^s} = \frac{\kappa_sc_v^s}{\kappa_nc_v^n}$$  \hspace{1cm} (S4)

The validity of the equations (S2) to (S4) is based on the above-mentioned assumptions that the heater contact is point-like and the heat flow is one-dimensional. In reality, both the sample width and the heater contact size are finite. This implies for the experimental setup to satisfy the assumptions of the examined model system, the exposed sample length ($l_{sample}$) must be far longer than the heater length ($l_h$) and the sample width ($w_{sample}$), $l_{sample} \gg l_h, w_{sample}$. For sample #4 we have $l_{sample}(2\text{ mm}) > l_h(0.5\text{ mm}), w_{sample}(0.2\text{ mm})$. This shows that our current setup already is a good proof of principle, although, the described conditions are not perfectly met. Future designs will aim at $F(\omega) = 1$ and $l_{sample} \gg l_h, w_{sample}$ to enable quantitative measurements of $c_v(T)$ under strain.
Experimental limits on the detection of a potential second superconducting transition

The signal-to-noise ratio determines one of the experimental limits on detecting a potential second superconducting transition in Sr$_2$RuO$_4$. The noise level remains almost the same for all strains; therefore, we used the heat capacity data at zero strain to analyze the experimental resolution limit given by the noise of the thermometer readout. Fig. S2a shows the raw signal $V_{ac}$ of the thermocouple as function of temperature, after 300x amplification by a transformer mounted on the 1 K pot of the dilution refrigerator and using time constants of 20 and 50 s for 2333 and 3913 Hz, respectively. To determine the noise we fit a polynomial to the normal state data and subtracted it. Fig. S2b shows the result as function of temperature. The noise is temperature-independent and its standard deviation $\sigma$ is only 0.63 nV, corresponding to 2.1 pV at the thermocouple.

![Fig. S2 Raw signal and noise. a, The thermocouple voltage readout $V_{ac}$ against temperature at zero strain for sample #4. The red line is a fit to the normal state data with a 5$^{th}$ degree polynomial. b, The difference between the measured signal $V_{ac}$ and the fitted curve. The standard deviation is 0.63 nV.](image)

| $f_{exc}$  | TC | $V_{ac}$ (1.6 K) | $V_{ac}$ (3.6 K) | Noise$^A$ | limits$^B$ (1.6 K, 3.6 K) |
|-----------|----|-----------------|-----------------|----------|------------------------|
| 2333 Hz   | 20 s | 370 nV          | 230 nV          | 0.75 nV  | 3%, 5%                 |
| 3913 Hz   | 50 s | 210 nV          | 130 nV          | 0.63 nV  | 4%, 7%                 |

$A$: Standard deviation; $B$: The limit with respect to the first transition.

**Table S1 Detection limits for a potential second transition.** Estimates of the sizes of the experimental limits on the potential second transition for sample #4 at different temperatures and frequencies.
Since 95% of data points fall within twice the standard deviation in a normal distribution, we use $4\sigma \approx 2.5 \text{nV}$ as the detection limit for a step size. The size of the signal at $T = 1.6 \text{K}$ is about 210 nV so the experimental limit on detecting a small jump in $\Delta c/c$ is $\Delta V_{ac}/V_{ac} = 2.5 \text{nV}/210 \text{nV} \approx 0.012$. The visible transition in the data has a jump size of $\Delta c/c \approx 0.3$. That implies that a potential second transition has to be more than $0.012/0.3 = 4\%$ of the size of the visible transition to be resolved. Therefore, the experimental limit on detecting the second transition is about 4% of the size of the visible transition for sample #4 with $f_{exc} = 3913 \text{Hz}$ in the temperature region around 1.6 K. Table S1 provides estimates of the experimental limits on detecting the second transition at 1.6 K and 3.6 K for sample #4 for the excitation frequencies and time constants (TC) used in our standard measurements.

The limit on determining the separation of two potential transitions

It will be not possible to resolve two transitions if they are too close together. Therefore, in Sr$_2$RuO$_4$ the breadth of the visible transition determines the detection limit for the separation of two transitions. Fig. S3a and S3b show the first derivative of the measured $1/T_{ac}(T)$ data with $f_{exc} = 3913 \text{Hz}$ at strains before and after the peak in $T_c$ for sample #4, respectively. The full width at half maximum (FWHM) is used as a conservative criterion for the limit on determining the separation of two potential transitions. The results are shown in Fig. S3c. The FWHM increases from 50 mK at zero strain to 300 mK at $\varepsilon_{xx} = -0.53\%$.

Strain inhomogeneity

Strain inhomogeneity causes a distribution of $T_c$’s and leads to a rounded heat capacity anomaly. From the breadth of the transition, the size of the inhomogeneity can be estimated as follows:

$$\Delta T_c \cong \frac{dT_c(\varepsilon)}{d\varepsilon} \times \Delta \varepsilon = \frac{dT_c(\varepsilon)}{d\varepsilon} \times \frac{\Delta \varepsilon}{\varepsilon} \times \varepsilon. \quad (S5)$$

$\Delta T_{c,FWHM}$ can be determined from the $d(1/T_{ac})/dT$ curves as shown in Fig. S3 and, therefore,

$$\Delta T_{c,FWHM} = \left| \frac{dT_c(\varepsilon)}{d\varepsilon} \right| \times \frac{\Delta \varepsilon_{FWHM}}{|\varepsilon|} \times |\varepsilon|. \quad (S6)$$

The distribution in $T_c$, $\Delta T_{c,FWHM}$, is related to the tangent slope on the $T_c(\varepsilon)$ curve $|dT_c(\varepsilon)/d\varepsilon|$, the strain inhomogeneity $\Delta \varepsilon_{FWHM}/|\varepsilon|$ and the applied strain $|\varepsilon|$. It is inevitable to have a certain strain inhomogeneity in a sample and, therefore, the higher the applied strain, the wider the distribution in $T_c$. Note, the distribution is even larger when the applied strain goes beyond $\varepsilon_{peak in T_c}$ because the tangent slope is steeper. The $T_c(\varepsilon)$ curve determined by the midpoints of the leading edge of the transitions is used to simulate $\Delta T_{c,FWHM}$ with different
sizes of the inhomogeneity as shown in Fig. S3c. The values of $\Delta T_{c,\text{FWHM}}$ near zero strain are different from the simulations because there is a finite transition width intrinsic to the sample. The distribution of $T_c$ scales with the tangent slope on $T_c(\varepsilon)$. Hence, the inhomogeneity determined primarily by matching to the simulations around the peak in $T_c$ is approximately 8% in sample #4.

![Graph](image_url)

**Fig. S3** Temperature resolution and strain inhomogeneity. The first derivative of $1/T_{ac}$ with respect to temperature for sample #4 at different strains a, before the peak in $T_c$ and b, after the peak. The curve at -0.57% in panel a is reduced by a factor of 2 for clarity. The transition breadth against strain. The solid points are the FWHM derived from the results in a and b. Three simulation curves (see text) with different strain inhomogeneities are shown for comparison. The arrow marks the position of the peak in $T_c$. 

$\Delta T_{c,\text{FWHM}}$
A stricter limit for the second transition

![Graph showing heat capacity measurements](image)

**Fig. S4 Higher resolution heat capacity measurements.** a, Measurements at several low strains. $P_{\text{exc}}$ was doubled and the measurements were repeated several times at each strain—20, 34, 18 and 20 times for the 0%, -0.07%, -0.1% and -0.13% curves, respectively. The arrow indicates a tiny discontinuity near 1.15 K, caused by the switch between two thermometer calibration ranges b, The averaged voltage readout from the thermocouple at zero strain. The red line is a linear fit to the data below the discontinuity. c, The difference between the measured curve and the linear fit. The standard deviation is only about 0.11 nV.

In the previous discussions, we already put a relatively stringent limit on the experimental detection of a potential second transition on the example of sample #4. Here, we present an analysis using a set of heat capacity recorded with a doubled excitation power also on sample #4. Additionally, the measurement runs were repeated and averaged up to 34 times providing higher quality heat capacity data with a much better signal-to-noise ratio. We restricted the measurements on the temperature range between 1 K and 1.5 K since a potential second transition is expected at temperatures lower than the visible transition anomaly. **Fig. S4a**
shows higher quality heat capacity measurements for sample #4 at different strains after the averaging. The averaged curves are smooth below $T_c$. There is a small discontinuity at 1.15 K, indicated by an arrow, which is an artifact due to the switch between two different calibration ranges of the thermometer used in the experiments. We emphasize the existence of the discontinuity, since it demonstrates that the signal-to-noise ratio in our measurements is high enough to resolve this very small feature in the data. Fig. S4b and S4c show the averaged voltage response and the noise. Following the same analysis scheme as above, the detection limit in voltage is 0.44 nV and the detection limit relative to the primary transition is 0.3%. A self-consistent check is the discontinuity. It is a 0.4% change in the signal and about a 1.3% change relative to the visible anomaly. Since the detection limit is 0.3%, the 1.3% artifact near 1.15 K is well resolved as shown in Fig. S4a. We note, however, that this discussion concerns the ability to resolve a sharp discontinuity. If, as seems reasonable, a second transition had a similar width to the primary one, our detection limits would fall to a few per cent. This still places very strong constraints on any theory proposing that strain splits the transition temperature of a two-component superconducting order parameter.