

# Thirty years of puzzling superconductivity in $\text{Sr}_2\text{RuO}_4$

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Superconductivity in  $\text{Sr}_2\text{RuO}_4$  was discovered 30 years ago. Among the many intriguing aspects of this unconventional superconductor is the picture of spin-triplet superconductivity, which could potentially carry both charge and spin supercurrents. This proposal was considered for a long time but was ultimately disproven in 2019. Despite intense research over the past several years, the superconducting symmetry of the archetypal unconventional superconductor  $\text{Sr}_2\text{RuO}_4$  remains unresolved. Here we highlight the recent controversies and give a perspective of how the final resolution may be reached.

Superconductivity in the layered oxide  $\text{Sr}_2\text{RuO}_4$ , which is isostructural to the first-discovered copper oxide high-temperature superconductor<sup>1</sup>, was first reported in 1994<sup>2</sup>. Its superconducting transition temperature  $T_c$  is strongly dependent on sample quality, and it is 1.5 K in the best crystals. Because of its unconventional superconductivity, evidenced by its extreme sensitivity to non-magnetic impurities<sup>3</sup>, and the strong electron correlations observed in the Mott insulating sister compound  $\text{Ca}_2\text{RuO}_4$  (ref. 4),  $\text{Sr}_2\text{RuO}_4$  is considered an archetypal quantum material. Despite extensive research efforts and state-of-the-art experimental and theoretical studies, understanding its puzzling superconducting state remains a challenge<sup>5</sup>. In Box 1, we introduce some key terms used to describe the peculiar features of unconventional superconductivity.

The electronic state of  $\text{Sr}_2\text{RuO}_4$  contains typical complications of quantum materials exhibiting emergent properties, such as exotic superconductivity and magnetism. It features strong electron correlations, moderate spin–orbit coupling and a superconducting state that involves more than one electronic orbital. The normal state of  $\text{Sr}_2\text{RuO}_4$  exhibits textbook-like Fermi-liquid properties at low temperatures<sup>6</sup>, in stark contrast to the exotic normal-state properties of other well-known unconventional superconductors, such as copper oxide, iron-based and heavy fermion superconductors. Its highly correlated, quasi-two-dimensional (2D) electronic structure has been accurately characterized by angle-resolved photoemission spectroscopy and quantitatively understood down to fine details by electronic structure calculations<sup>7</sup>. Importantly, chemically stable, extremely high-quality single crystals have been grown by several groups using the floating-zone method (Fig. 1), and experimental results from different research groups are highly reproducible as long as the same type of probe is used. Therefore, we believe that complete understanding of the basic superconducting properties of  $\text{Sr}_2\text{RuO}_4$  is possible.

## Controversies about the superconducting symmetry

Over the past 30 years,  $\text{Sr}_2\text{RuO}_4$  has challenged key experimental techniques. The angle-resolved photoemission spectroscopy measurements reported by two expert groups were initially believed to be associated with bulk properties. However, years later, an essential revision was made because of the presence of surface states caused by the rotation of the  $\text{RuO}_6$  octahedra on the cleaved surface<sup>8</sup>. For this reason, the  $\Gamma$  band, which hosts a van Hove singularity, was initially reported as a hole band, rather than as an electron band.

In 2019, another key result concerning the spin state of the superconducting electrons was found to be incorrect<sup>9</sup>. Owing to the extremely high in-plane conductivity of  $\text{Sr}_2\text{RuO}_4$ , eddy-current joule heating by radio-frequency pulses during nuclear magnetic resonance (NMR) measurements increased the sample temperature above  $T_c$ . Even if such overheating lasted up to only 10 ms, it was enough to alter the results<sup>10</sup>. For this reason, the NMR pulse energy for  $\text{Sr}_2\text{RuO}_4$  needs to be reduced to an unusually low level to observe the correct shift of the resonance line—the Knight shift—associated with the reduction of spin susceptibility in the superconducting state. This precaution may also apply to other low- $T_c$  superconductors, especially those with extremely low residual resistivity. These findings have encouraged NMR researchers to present their results as a function of pulse energy, thereby demonstrating that overheating does not occur during the measurement process.

With additional NMR results under different in-plane field directions, it was concluded that the Cooper-pair spin state is singlet and thus not compatible with either chiral or helical spin-triplet states<sup>11</sup>. Another experiment involving polarized neutrons that previously concluded in favour of triplet pairing was repeated under lower applied

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## BOX 1

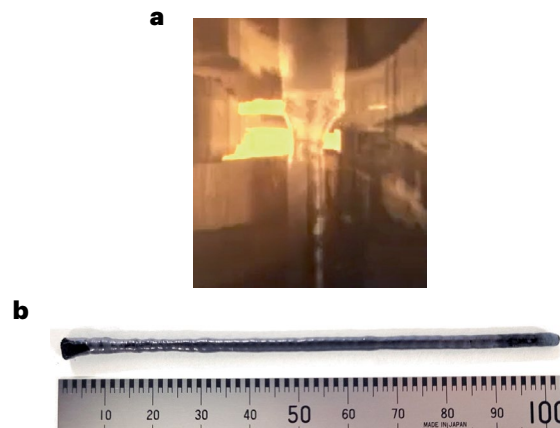
# Unconventional superconductivity

Superconductivity is usually called ‘conventional’ when it is described by the original Bardeen–Cooper–Schrieffer (BCS) theory<sup>48</sup>, which involves spin-singlet s-wave pairing. Superconducting states not described by the BCS theory are usually referred to as ‘unconventional’ and involve other types of pairing.

The fermionic nature of electrons gives rise to antisymmetric states, meaning that the state vector undergoes a minus sign change upon particle exchange. An antisymmetric spin-singlet state can be represented as  $\frac{1}{\sqrt{2}} \{ |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \}$  (in which  $\uparrow$  and  $\downarrow$  represent spin-up and spin-down states, respectively), and it must be accompanied by an orbital wavefunction of even parity that corresponds to orbital angular momentum, such as  $L=0$  (s wave),  $L=2$  (d wave) and  $L=4$  (g wave). Conversely, the spin-triplet states of the form  $|\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle$  or  $\frac{1}{\sqrt{2}} \{ |\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle \}$  must be accompanied by odd-parity orbital states, such as  $L=1$  (p wave) and  $L=3$  (f wave). High-temperature copper oxide superconductors, many of the heavy-electron superconductors and organic superconductors are believed to be in spin-singlet d-wave states. Naively, strong electron correlations with strong Coulomb repulsion in these unconventional superconductors disfavour s-wave pairing, where the maximum amplitude of the order parameter is achieved when the paired electrons are at zero distance between each other. However, such a simple classification may partially break down if the spin–orbit coupling is strong. Among the unconventional superconductors, there are some promising candidates for spin-triplet pairing, but a complete characterization of these superconducting states has not been achieved so far. The superconducting energy gap of unconventional superconductors is often accompanied by the presence of nodes in the momentum space around which the order-parameter phase changes its sign.

Among unconventional superconductors, some exhibit exotic properties, including the breaking of time-reversal symmetry (TRS) and rotational symmetry. TRS is the property under which a physical system is unchanged upon reversal of time, whereas rotational symmetry is the property under which a physical system remains unchanged upon rotation of a certain angle. Systems showing broken TRS for intrinsic reasons are often called ‘chiral’ or ‘non-unitary’, depending on whether the symmetry breaking originates from the orbital or spin part of the order parameter. Systems where the rotational symmetry of the electronic structure is spontaneously broken in the superconducting state are often called ‘nematic’. According to the irreducible representation to which it belongs, the superconducting order parameter can be classified as 1D if it is described by a single function or 2D if it is described by a pair of functions. Chiral TRS breaking can be realized using a 2D order parameter. Topological superconductors have an order parameter characterized by a topological invariant, a quantity that does not change upon deformation of the space, and have specific edge states at the boundary based on bulk–edge correspondence. Spin-singlet and triplet superconducting states with chiral TRS breaking have topological properties.

field, and it manifested the reduction of the magnetic susceptibility as expected for a singlet pairing<sup>12</sup>. Thus, the superconducting symmetry of  $\text{Sr}_2\text{RuO}_4$  took a turn from ‘spin-triplet chiral p-wave pairing’ to ‘spin-singlet pairing’.



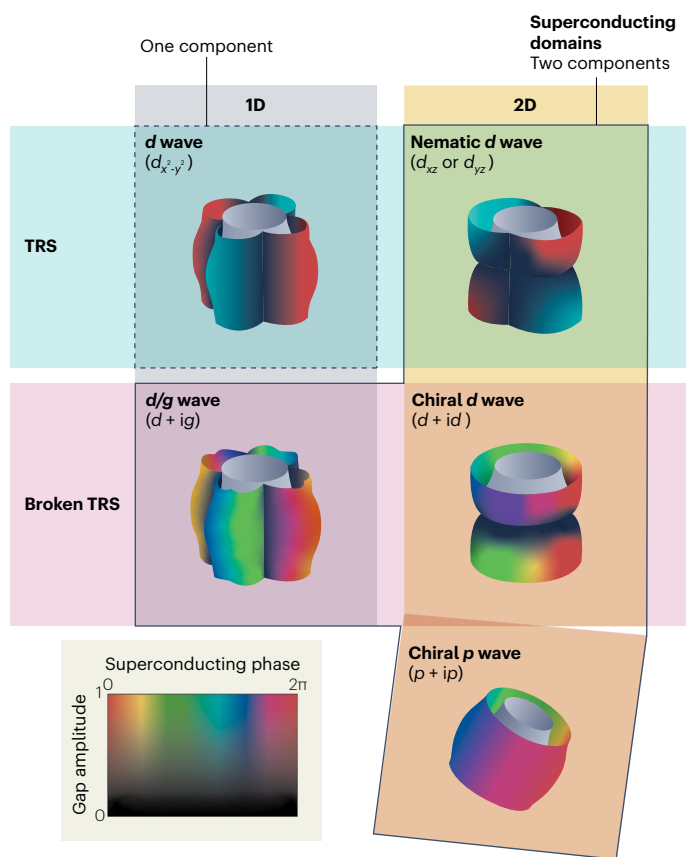
**Fig. 1 | Crystal growth of  $\text{Sr}_2\text{RuO}_4$  using the floating-zone method. a**, Large, high-quality single crystals of  $\text{Sr}_2\text{RuO}_4$  that exhibited  $T_c = 1.50$  K were grown using the floating-zone method. A feed rod (diameter of 6 mm) of sintered powder was used, and the (001) facet parallel to the  $\text{RuO}_2$  plane was clearly visible. **b**, Typical crystal with a cross-section of  $3 \times 4$  mm<sup>2</sup> and length of up to 100 mm (ref. 46).

This development forced us to re-examine other existing results from a new viewpoint. Nevertheless, the symmetry of the superconducting state of  $\text{Sr}_2\text{RuO}_4$  has not yet been unanimously agreed on at present. Meanwhile, a serious controversy among the key results has become evident. Two main issues that currently prevent us from reaching a consensus among the candidate superconducting states are illustrated in Fig. 2: (1) whether the TRS is broken or not and (2) whether there is a horizontal line node in the superconducting gap in addition to the vertical line nodes or gap minima. For the examples shown in Fig. 2, the latter issue also corresponds to whether the order parameter dimensionality is one-dimensional (1D) or 2D. We examine these issues in the following sections.

## Does the superconductivity of $\text{Sr}_2\text{RuO}_4$ break TRS?

Of equal importance to the NMR result, another key experimental result is the TRS breaking of the order parameter, which is based on the observation of the spontaneous internal magnetic field emerging below  $T_c$  in a muon spin rotation ( $\mu\text{SR}$ ) experiment<sup>13</sup>. The internal magnetic field detected at the muon site has been interpreted to be a consequence of the topological edge current around either impurities or the implanted muon itself in a TRS broken superconductor. The observation of TRS breaking signals in  $\text{Sr}_2\text{RuO}_4$  is robust and reproducible among different investigators and muon facilities<sup>14–17</sup>. The broken TRS state was also detected using the magneto-optic Kerr effect<sup>18</sup>. Recently, *ab initio* theories for the normal state of  $\text{Sr}_2\text{RuO}_4$  have been used to identify the muon stopping site and determine the influence of a muon in the electronic states<sup>19,20</sup>. The positive charge of an injected muon with its lifetime of 2.2  $\mu\text{s}$  substantially distorts the lattice, but because of the short Thomas–Fermi screening length of about 0.1 nm, the effect is rather local. In the superconducting state, the charge screening length is expected to be essentially unchanged<sup>21</sup>.

The innovative development of a uniaxial-stress apparatus using piezo stacks introduced new insights into the study of  $\text{Sr}_2\text{RuO}_4$  (ref. 22). Uniaxial stress along the [100] direction enhanced  $T_c$  from 1.5 to 3.5 K (ref. 23), thus answering the long-standing question of the mechanism of  $T_c$  enhancement in the eutectic ‘3-K phase’<sup>24</sup>. Uniaxial stress, which breaks the tetragonal symmetry of the lattice, provides important information concerning the TRS breaking. The superconducting onset  $T_c$  increases with uniaxial stress, whereas the temperature at which muons start to experience an emerging magnetic field  $T_{\text{TRSB}}$  remains almost the same or decreases slightly<sup>16</sup>. As the splitting between  $T_c$  and  $T_{\text{TRSB}}$  was observed only when the tetragonal crystalline symmetry was



**Fig. 2 | Some of the proposed superconducting symmetries of  $\text{Sr}_2\text{RuO}_4$ .** To identify the superconducting state of  $\text{Sr}_2\text{RuO}_4$ , some controversies among the observed results need to be resolved. Left, 1D irreducible representation states:  $d$ -wave (top) and accidentally degenerate  $d + ig$ -wave (bottom) states. Right, 2D irreducible-representation states: nematic  $d$ -wave (top) and chiral  $d + id$ -wave (middle) states. Previously proposed spin-triplet, chiral  $p$ -wave state (bottom right) is not consistent with the recent NMR results. The time-reversal symmetry (TRS) is preserved for only the top two states; superconducting domains under the tetragonal crystal symmetry may form in the three states, except for the  $d$ -wave state on the top left<sup>5,47</sup>. The colours depict the superconducting gap amplitude and phase as shown on the bottom left. Diagrams reproduced with permission from: TRS and broken TRS rows, ref. 5, Physical Society of Japan; chiral  $p$  wave, ref. 47, Association of Asia Pacific Physical Societies.

broken, it was suggested that the degeneracy between  $T_c$  and  $T_{\text{TRSB}}$  was symmetry protected<sup>17</sup>.

The  $\mu\text{SR}$  results, however, have recently been challenged by thermodynamic measurements searching for the second transition at  $T_{\text{TRSB}}$ . The heat capacity  $C_p$  indicates a clear jump in  $C_p/T$  at  $T_c$  as it increased to 3.5 K under [100] strain<sup>25</sup>. However, there was no sign of any additional jump anticipated at  $T_{\text{TRSB}}$  (ref. 24). Furthermore, elastocaloric measurements did not probe any sign of the anticipated second transition<sup>26</sup>. A possible explanation is that the entropy release at the second transition is small or broadened in the thermal measurements<sup>27</sup>. An increasing number of superconductors seem to demonstrate a signature of broken TRS in  $\mu\text{SR}$  measurements<sup>28</sup>, including some with simple  $s$ -wave character, raising some doubts about whether all  $\mu\text{SR}$  data are being correctly interpreted as detections of TRS breaking. Another possibility for  $\text{Sr}_2\text{RuO}_4$  is that the presence of surface magnetism in the normal state, observed with ultraslow muons<sup>29</sup>, may lead to spontaneous vortices in the bulk below  $T_c$ .

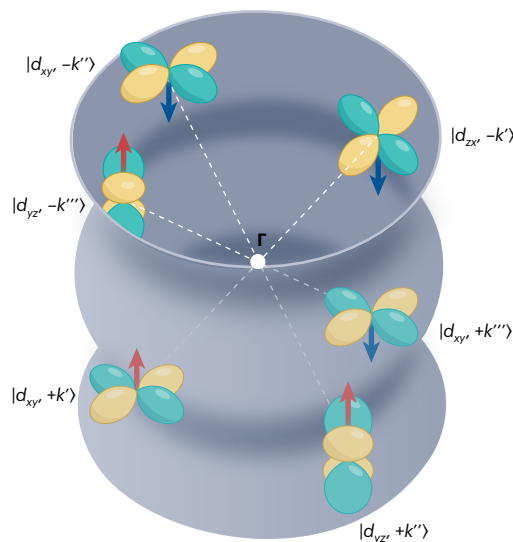
As a related development on the multicomponent order parameter, two groups have recently reported an unusual jump in a  $c_{66}$  shear-mode ultrasound velocity at  $T_c$ , in addition to the ordinary jumps

in the compressional modes<sup>30,31</sup>. This implies that the superconducting order parameter is in specific irreducible representations:  $E_g$ ,  $B_{1g} \oplus A_{2g}$  or  $B_{2g} \oplus A_{1g}$ . Considering this requirement and depending on whether the TRS is considered broken, the two groups proposed different superconducting order parameters: nematic  $E_g$  state with the gap structure  $d_{xz}$  or  $d_{yz}$  that does not break TRS and accidental TRS breaking  $B_{1g} \oplus A_{2g}$  with the gap  $d + ig$ .

As shown in Fig. 2, for 2D ( $E_g$ ) and two-component 1D order parameters, a superconducting domain structure may form in the crystallographic single-domain material:  $d_{xz} + id_{yz}$  and  $d_{xz} - id_{yz}$  domains for TRS breaking chiral superconductors and  $d_{xz}$  and  $d_{yz}$  for nematic superconductors. Various phenomena observed only in undistorted  $\text{Sr}_2\text{RuO}_4$  are attributed to the phase interference between such coexisting domains<sup>32</sup>.

## Horizontal line node and interorbital pairing

Determination of the superconducting gap anisotropy, especially the characterization of the location of the gap nodes, is essential for identifying the superconducting symmetry and mechanisms. The temperature and field dependence of the specific heat suggests the presence of line nodes or strongly suppressed gaps. The location of the node in the superconducting gap was investigated using the specific heat under a magnetic field applied in various directions<sup>33,34</sup>. The experimental data of the two groups were consistent with each other. If interpreted in terms of vertical line nodes or vertical gap minima, the nodal direction is along the  $\Gamma$ -M direction<sup>33</sup>. This interpretation is consistent with either  $p + ip$  or  $d_{xy}$  symmetry but inconsistent with  $d_{x^2-y^2}$  symmetry. Extending the measurements down to temperatures as low as 0.04  $T_c$ , Kittaka et al. did not observe the anticipated reversal of the oscillatory phase of the four-fold specific heat oscillations with respect to the in-plane field direction<sup>34</sup>. These results led the authors to conclude that there was a horizontal line node in the superconducting order parameter. In this case, the origin of the oscillation in the specific heat is the four-fold anisotropy of the quasiparticle Fermi velocity around the horizontal line node. Thermal conductivity measurements



**Fig. 3 | Interorbital pairing state proposed for  $\text{Sr}_2\text{RuO}_4$ .** In the orbital basis, a Cooper pair on the same band in the reciprocal space consists of electrons of the orbitals  $d_{xy}$  and either  $d_{yz}$  or  $d_{xz}$  with opposite momenta  $k'$  ( $k''$ ,  $k'''$ ) and  $-k'$  ( $-k''$ ,  $-k'''$ ), forming a spin-triplet ( $|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$ ), orbital-antisymmetric, even parity state. When transformed into the band basis, it is a spin-singlet,  $d$ -wave state with a symmetry-protected horizontal line node and gap minima along the vertical direction. Although rather exotic, such a state can explain many of the observed facts. The different colours of the orbital wave-function envelopes represent different phases.

## BOX 2

# Bridging pressure and ultrasound experiments

Here we discuss how available results obtained from different techniques can be compared, leading to the two controversies introduced in the main text regarding the kink and slope of the variations in  $T_c$  with strain. On the one hand, Hicks et al. applied uniaxial pressure reporting that the change in  $T_c$  with respect to the strain  $\varepsilon$  induced by the stress  $\sigma$  in the [110] direction is smooth and does not exhibit any kink at  $\varepsilon_{<110>}=0$  (ref. 22). As illustrated in panel **a**,  $\varepsilon_{<110>}$  can be decomposed into two components: a volume-conserving shear strain  $\varepsilon_6$  (also written as  $\varepsilon_{12}$  or  $\varepsilon_{xy}$ ) and an in-plane compression  $\varepsilon_{A_{1g,1}}=\varepsilon_1+\varepsilon_2$  (also written as  $\varepsilon_{11}+\varepsilon_{22}$  or  $\varepsilon_{xx}+\varepsilon_{yy}$ ). For the first component, we note that the effect of a positive  $\varepsilon_6$  is equivalent to a negative  $\varepsilon_6$  because of the four-fold rotational symmetry of the lattice. Therefore, if  $T_c$  changes with a positive  $\varepsilon_6$ , it should also change by the same amount with a negative  $\varepsilon_6$ , or  $\Delta T_c \propto |\varepsilon_6|$ , leading to a kink in  $T_c$  versus  $\varepsilon_{<110>}$  at  $\varepsilon_{<110>}=0$ . For the second component,  $\varepsilon_{A_{1g,1}}$  does not lead to a kink because positive and negative  $\varepsilon_{A_{1g,1}}$  should result in changes in  $\Delta T_c$  of the opposite sign. Thus, the experimental absence of a kink in  $T_c$  versus  $\varepsilon_{<110>}$  indicates that  $T_c$  is insensitive to  $\varepsilon_6$ . On the other hand, ultrasound experiments revealed a jump in  $c_{66}$  across  $T_c$  (refs. 30,31). The ultrasound results, in combination with the Ehrenfest relation described below, indicate that  $T_c$  depends on  $\varepsilon_6$ , contradicting the uniaxial pressure result. A detailed quantitative analysis of this controversy was conducted by Jerzembeck et al.<sup>49</sup>.

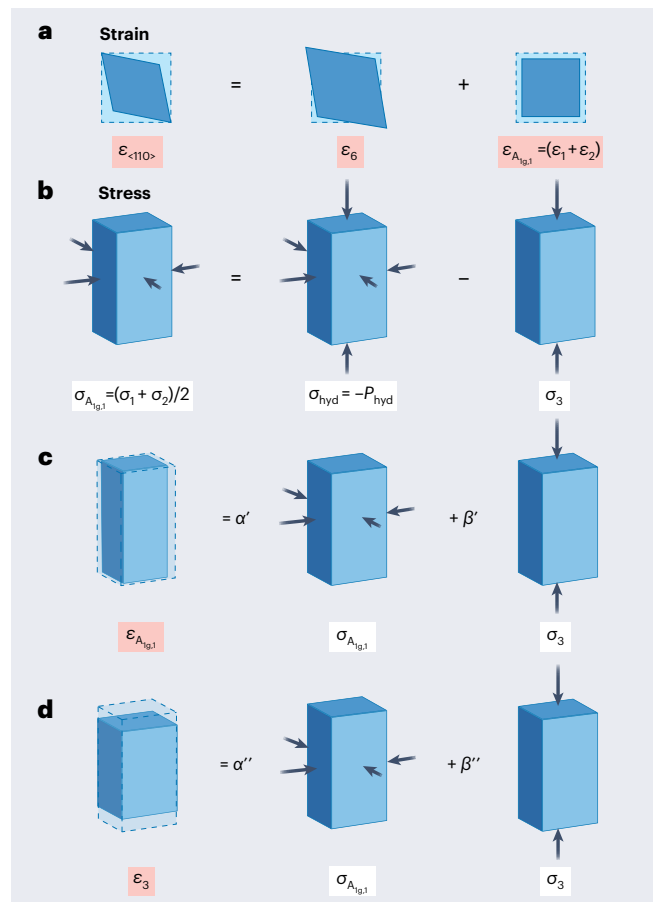
As a second issue, we examined the initial slope of the  $T_c$  variations with strains. The change in  $T_c$  with uniaxial stress in the  $c$ -direction  $\sigma_3$  (also written as  $\sigma_{33}$  or  $\sigma_{zz}$ ) was reported as  $\partial T_c / \partial \sigma_3 = +76 \text{ mK GPa}^{-1}$  (ref. 41). In addition, the change in  $T_c$  with hydrostatic pressure  $P_{\text{hyd}}$  was reported as  $dT_c/dP_{\text{hyd}} \approx -0.21 \text{ K GPa}^{-1}$  (ref. 42). We note that  $P$  has the opposite sign to both  $\sigma$  and  $\varepsilon$ ; positive  $P$  corresponds to lattice compression, whereas positive  $\sigma$  and  $\varepsilon$  correspond to lattice expansion. As the hydrostatic pressure is a combination of the in-plane compression  $\sigma_{A_{1g,1}}=(\sigma_1+\sigma_2)/2$  (also written as  $(\sigma_{11}+\sigma_{22})/2$  or  $(\sigma_{xx}+\sigma_{yy})/2$ ) and the  $c$ -axis compression  $\sigma_3$  as illustrated in panel **b**, we calculated from the experimental results

$$\frac{\partial T_c}{\partial \sigma_{A_{1g,1}}} = -\frac{dT_c}{dP_{\text{hyd}}} - \frac{\partial T_c}{\partial \sigma_3} \approx 0.13 \text{ K GPa}^{-1}.$$

As shown in panels **c** and **d**, we used this expression along with elastic moduli  $c_{11}$ ,  $c_{12}$ ,  $c_{13}$  and  $c_{33}$  (reported in ref. 31) to calculate the strain dependence of  $T_c$ , as shown in the left column of the table below. These results are compared with the changes in  $T_c$  predicted by compressional ultrasound in the right column of the table below, in which the initial slope  $\partial T_c / \partial \sigma_i$  ( $i$  represents  $A_{1g,1}$  or 3) is linked to the jump of the elastic modulus  $c_i$  by the thermodynamic Ehrenfest relation<sup>30,31,50,51</sup>, with  $\Delta C_p$  the jump at  $T_c$  of the specific heat per volume. We note that there is a prefactor on the left-hand side of this equation depending on mode  $i$  and the coefficients in the Ginzburg–Landau free energy, which, for the cases discussed here, is equal to unity<sup>51</sup>. Using the values  $T_c=1.42 \text{ K}$ ,  $\Delta C_p=33 \text{ mJ mol}^{-1} \text{ K}^{-1}=570 \text{ J m}^{-3} \text{ K}^{-1}$  and  $\Delta c_i$  from ref. 31, we can estimate the strain dependence of  $T_c$  again. These values obtained from ultrasound in the right column are about a factor of 5 larger than those calculated using the pressure experiments in the left column.

As an additional final comparison, we note that the jump in  $c_{11}$  in the pulse–echo ultrasound experiment<sup>30</sup> implies  $|\partial T_c / \partial \varepsilon_1|=1.08 \text{ K \%}^{-1}$  from the Ehrenfest relation combined with the fact that little jump is observed in the shear mode  $(c_{11}-c_{12})/2$ .

This large variation in  $T_c$  is in sharp contrast to the absence of an initial slope under [100] uniaxial stress<sup>22,23,44</sup>.



**Relationships between strain and stress. a**, Uniaxial strain in the [110] direction can be described as the sum of a volume-preserving shear deformation and a biaxial strain. **b**, In-plane biaxial stress can be described as the hydrostatic pressure minus the out-of-plane stress. **c,d**, Biaxial strain (**c**) and out-of-plane strain (**d**) can be described as combinations of stresses with different coefficients  $\alpha$  and  $\beta$ .

**Changes in  $T_c$  by strain calculated from the pressure experiments via the elasticity tensor (left) and compressional ultrasound experiments via the Ehrenfest relation (right)**

Pressure experiments	Sound velocity
$\frac{\partial T_c}{\partial \varepsilon_i} = \sum_j \frac{\partial \sigma_j}{\partial \varepsilon_i} \frac{\partial T_c}{\partial \sigma_j} = \sum_j c_{ji} \frac{\partial T_c}{\partial \sigma_j}$	$\left( \frac{\partial T_c}{\partial \varepsilon_i} \right)^2 = \frac{-T_c \Delta c_i}{\Delta C_p}$ : Ehrenfest relation
$\frac{\partial T_c}{\partial \varepsilon_{A_{1g,1}}} = \frac{c_{11}+c_{12}}{2} \frac{\partial T_c}{\partial \sigma_{A_{1g,1}}} + c_{13} \frac{\partial T_c}{\partial \sigma_3} \approx 0.32 \text{ K \%}^{-1}$	$\left  \frac{\partial T_c}{\partial \varepsilon_{A_{1g,1}}} \right  = \sqrt{\frac{-T_c \Delta c_{A_{1g,1}}}{\Delta C_p}} = 1.59 \text{ K \%}^{-1}$
(panel <b>c</b> )	
$\frac{\partial T_c}{\partial \varepsilon_3} = c_{13} \frac{\partial T_c}{\partial \sigma_{A_{1g,1}}} + c_{33} \frac{\partial T_c}{\partial \sigma_3} \approx 0.31 \text{ K \%}^{-1}$	$\left  \frac{\partial T_c}{\partial \varepsilon_3} \right  = \sqrt{\frac{-T_c \Delta c_3}{\Delta C_p}} = 1.56 \text{ K \%}^{-1}$
(panel <b>d</b> )	



under a rotating in-plane magnetic field also concluded in favour of a horizontal line node<sup>35</sup>.

Several experiments have been conducted to probe the low-energy quasiparticle excitations in this system. Thermal conductivity measurements indicate that low-energy quasiparticles have a Fermi velocity component along the  $c$  axis<sup>36</sup>. Measurements of the penetration depth by  $\mu$ SR<sup>37</sup>, scanning-probe superconducting interference devices<sup>38</sup> and a.c. susceptibility<sup>39</sup> suggest line nodes along the interlayer  $c$  direction (vertical line nodes). It should be noted that gap models with only vertical line nodes are often compared with models with only a horizontal line node<sup>5</sup>. These analyses conclude the presence of vertical line nodes but do not necessarily exclude the presence of an additional horizontal line node.

The horizontal line node, if it is at the interlayer wave number  $k_z = 0$ , is not natural for quasi-2D materials, such as  $\text{Sr}_2\text{RuO}_4$ , because it implies that intralayer pairing is not allowed. An interorbital pairing model was proposed as a microscopic mechanism that allows a horizontal line node<sup>40</sup>. It first examines the pairing in the orbital basis and then transforms it into the band basis using a realistic three-dimensional orbital-characterized three-band electronic structure. As a promising candidate, a spin-triplet ( $|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle$ ),  $s$ -wave, orbital-antisymmetric (orbital-singlet) pairing state has been proposed. This state is depicted in Fig. 3. This state belongs to the 2D irreducible representation  $E_g$  and transforms into a spin-singlet,  $d$ -wave superconducting state on the band basis. It has an approximate vertical line node in addition to the horizontal line node in the gap, and the TRS can be either broken (chiral) or not (nematic). These interorbital  $E_g$  states have some similarities to the traditional intra-orbital  $E_g$  states shown in Fig. 2. The order parameters with  $E_g$  symmetry must change sign by reflection on the  $x$ - $y$  plane and lead to a horizontal line node at  $k_z = 0$ . The interorbital model further explains its origin on a microscopic basis. In essence, if the pairing is between electrons with  $d_{yz}$  ( $d_{xz}$ ) and  $d_{xy}$  orbitals, the symmetry information encoded in the first orbital captures the sign change by  $x$ - $y$ -mirror reflection<sup>5</sup>. Whether such an exotic pairing state can be realized remains an open question to be resolved by further experimental investigation. However, the interorbital pairing model can explain the following unusual observations under strains<sup>5,27</sup>:  $T_c$  decreases under uniaxial strain along the  $c$  axis<sup>41</sup>, despite the increase in the electronic density of states;  $T_c$  decreases under hydrostatic pressure<sup>42</sup>, under which the Fermi surface becomes more 2D, thus weakening the interlayer coupling. Furthermore, a theory predicts an anisotropic reduction of the Knight shift in such a spin-triplet interorbital pairing, distinct from the spin-singlet pairing<sup>43</sup>.

## Controversies among various lattice-distortion experiments

As described previously, the application of uniaxial strains has provided some fresh insights into  $\text{Sr}_2\text{RuO}_4$ , which can be compared with the strain susceptibility measured by ultrasound. Two controversies that need to be resolved concerning the changes in  $T_c$  under strain are presented in detail in Box 2. One is the absence of a kink in the  $T_c$  versus strain, as anticipated from ultrasound. The other is the slope of the  $T_c$  versus strain curves for in-plane and out-of-plane compressional strains.

First, the absence of a kink in the variation of  $T_c$  with the  $[110]$  strain<sup>22</sup> is qualitatively incompatible with the observed jump in the ultrasound velocity of the shear mode  $c_{66}$  (refs. 30,31). The absence of a kink with the  $[100]$  strain is also not consistent with the  $\mu$ SR results, which suggests a two-component order parameter that contains a linear  $T_c$  increase and naturally leads to a kink. Watson et al. did not observe such a kink under compressional and tensile  $[100]$  strains<sup>44</sup>. Second, aside from the kink, the magnitude of the slopes of  $T_c$  versus strain with in-plane and out-of-plane compressional strains<sup>22,41</sup> is quantitatively different by a factor of about five from the observed ultrasound velocity jumps. Considering the precision of the strain and

ultrasound experiments, these qualitative and quantitative disagreements pose a serious issue that requires attention.

In all these cases, the variations in  $T_c$  deduced from the ultrasound velocity jumps were substantially greater than those observed under the corresponding compressional strains. Concerning the difference in the magnitude of the ultrasound velocity jumps between the two groups<sup>30,31</sup>, the role of the superconducting domains has been discussed<sup>45</sup>. We note that typical magnitudes of strain are as small as  $10^{-4}$  in uniaxial strain experiments,  $10^{-6}$  to  $10^{-8}$  in pulse-echo ultrasound and  $10^{-10}$  in resonant ultrasound spectroscopy. The difference in magnitude, in conjunction with the differences in the measurement frequency and configuration of the superconducting domains, may be crucial for resolving these controversies.

If the observed jump in  $c_{66}$  is reliable, this result provides strong evidence that the order parameter consists of two components. This is because the symmetry of the square of the order parameter contains the same representation as  $\varepsilon_6$  ( $B_{2g}$ ) to induce such a coupling. This is the case for  $d_{yz} + d_{zx}$  (2D) and  $d_{x^2-y^2} + g_{xy}(x^2-y^2)$  (accidentally degenerated). If such coupling with shear strain is absent, these candidate order parameters are unlikely.

## Conclusion

We selected some promising candidates for the superconducting state of  $\text{Sr}_2\text{RuO}_4$ , as shown in Fig. 2. They are classified by the irreducible representations allowed in the point symmetry of  $D_{4h}$  applied to  $\text{Sr}_2\text{RuO}_4$  without distortions. Except for the  $B_{1g}$   $d$ -wave state, which is well known for high- $T_c$  copper oxide superconductors, superconducting domains may form in a tetragonal lattice.

The  $E_g$  candidates are further divided into two types. One is the traditional unconventional pairing, for example, a chiral  $d$ -wave state with gap anisotropy  $\Delta(\mathbf{k}) = \Delta_0 k_z (k_x + ik_y)$ , and the other is the exotic interorbital pairing. The results of  $\mu$ SR support the TRS breaking 2D  $E_g$  candidates and two-component 1D order parameters. The results of shear ultrasound additionally support the TRS-preserving nematic  $E_g$  candidates. If the additional horizontal line node does not exist, the promising state is  $B_{1g}$   $d_{x^2-y^2}$  with a gap function  $\Delta(\mathbf{k}) = \Delta_0 (k_x^2 - k_y^2)$ . In this case, the TRS is preserved, the second transition does not exist under uniaxial strain, the field angle-resolved specific heat needs reinterpretation and the superconducting domains do not form.

The 30-year history of the superconductivity of  $\text{Sr}_2\text{RuO}_4$  has taught us how challenging it can be to thoroughly understand an unconventional superconductor. More than once, we have seen how an indispensable technique, although giving reproducible results among specialists, had to be revised. In this sense,  $\text{Sr}_2\text{RuO}_4$  grew with the progress of modern-day condensed-matter physics. As in the comic, the community may still have to sacrifice some of the traditional knowhows to finally solve the  $\text{Sr}_2\text{RuO}_4$  puzzle and fully open the gate of truth.

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### Author contributions

Y.M. drafted the main part of the article, A.I. drafted Box 2 and G.M. devised and prepared most of the artwork. All authors contributed to discussing the content, editing and revising the article.

### Competing interests

The authors declare no competing interests.

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