



A Personal Perspective on the Unconventional Superconductivity of Sr₂RuO₄

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Abstract

I give a personal account of the unfolding story of the unconventional superconductivity of Sr₂RuO₄. This is a subject of topical importance in light of recent measurements that suggest that its order parameter may be even parity, contrary to the picture that had built up over the course of over two decades of research. With an eye on the past, I stress the generous encouragement that I received from Ted Geballe in the early years of my Sr₂RuO₄ research. Looking to the future, I give my opinion about why the Sr₂RuO₄ problem is of major significance to the development of the field of unconventional superconductivity, whatever order parameter symmetry is finally established to be the correct one.

Keywords Sr₂RuO₄ · Unconventional superconductivity

I should start by saying that it is a pleasure and an honour to contribute to this most special of volumes celebrating the career of a man for whom I have the utmost personal respect. For me, Ted Geballe embodies all that is good in the world. His brilliant scientific insight is accompanied by immense and completely spontaneous generosity and kindness, whether in the professional or personal arena.

As my contribution, I think that it is appropriate that I write a short perspective on the unconventional superconductivity of Sr₂RuO₄. There is no shortage of review articles on Sr₂RuO₄ [1–5]. I certainly do not intend to repeat the content of those here, but for several reasons that I hope will become clear, I think that the time is right for me to write a short personal article. The first reason for this choice is that, like Ted, Sr₂RuO₄ superconductivity is celebrating a milestone birthday. It is a trivial one compared to his, but 2019 marks a quarter of a century since Yoshi Maeno and colleagues' landmark discovery in Hiroshima [6] of superconductivity in

high-quality single crystals grown a few years earlier by Frank Lichtenberg in Georg Bednorz's group at IBM Zürich.

I had met Yoshi at a conference in 1993, and he knew from those discussions that I had an interest in low temperature measurements. He therefore sent me a preprint of his 1994 paper (in those days still a paper item brought to me by a colleague who met him in Japan) and I was fortunate to establish a collaboration with him to study the normal state properties of Sr₂RuO₄ even before his paper appeared in print. Those were truly exciting times, with the de Haas-van Alphen (dHvA) effect oscillations that had stubbornly eluded observation in the cuprates rising high above the background noise in Sr₂RuO₄. My own work, carried out in Cambridge in close collaboration with Stephen Julian [7] and, later, Christoph Bergemann [8, 9], was mainly on dHvA and other normal state properties, but I did have the chance to work on one aspect of the superconductivity.

On a visit to Hiroshima, one of Yoshi's graduate students, Koji Yoshida, casually mentioned to me that they did not always manage to obtain superconductivity in the crystals that they grew. By that time, the field was abuzz discussing the brilliant suggestion by Maurice Rice and Manfred Sigrist that the superconductivity of Sr₂RuO₄, though low- T_c with transitions at approximately 1 K, might be unconventional in nature. Specifically, based on analysis of the normal state data, they proposed that Sr₂RuO₄ might be a spin triplet superconductor, in analogy with the pairing in superfluid ³He [10]. I knew little—very little indeed!—about triplet superconductivity

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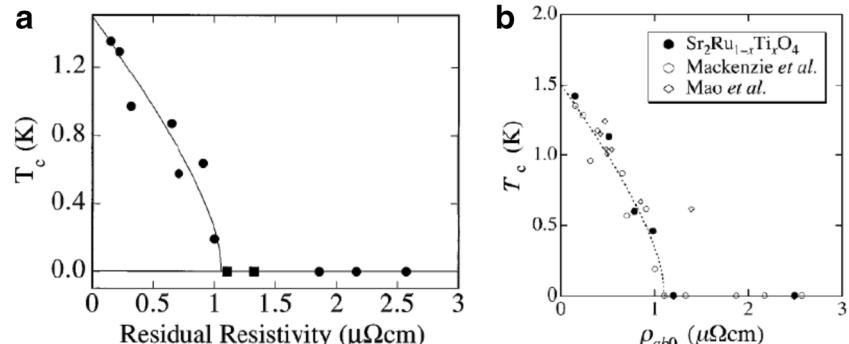
and vector order parameters, but I had picked up from discussions with my PhD supervisor Gil Lonzarich that if that were really the case, one expected the superconducting transition temperature to be extremely sensitive to non-magnetic disorder. My ears therefore pricked up when Koji told me about the sample-sample variations, and I returned to Cambridge with samples from across the range of growth batches. In one of the great coincidences that often shape scientific progress, I had exactly the right expertise to investigate a problem like this, because in the absence of cuprate dHvA oscillations, my PhD had ended up as a highly technical one on using electron probe microanalysis to study the chemical composition of difficult oxides. The Cambridge Earth Sciences department had a state-of-the-art instrument for these purposes, which featured Bragg spectrometer x-ray detectors that enabled excellent signal-to-noise detection of trace elements. It was in the charge of one of the founders of that field, Stephen Reed, from whom I learned a lesson that I still pay heed to today—if at all possible, do not place a technician between a sophisticated instrument and its users. Instead, give advice and tuition, but allow them the freedom to really use it themselves. Because of Stephen's enlightened attitude, I had been given the time to learn how to programme it to perform long, bespoke data taking runs. A quick bit of coding and about 2 weeks of data acquisition time later, we had good evidence that although the crystals were all nominally being prepared in the same way, they had significant variations in the levels of non-magnetic Al and Si impurities. This fired us up to perform measurements correlating the resistive mean free path with the superconducting transition temperature, on a dilution refrigerator set up for other reasons by Kurt Haselwimmer in Cambridge. The result, shown in Fig. 1a, provided what I believe is conclusive evidence that Sr_2RuO_4 has an unconventional order parameter. It is important to mention that this does not prove that Sr_2RuO_4 is a triplet superconductor, just that it is unconventional. Any odd parity spin triplet state can be destroyed by non-magnetic scattering, because randomizing the phase of the order parameter averages it to zero, but the same logic applies to even parity d - (or g - ...) order parameters as well.

I had enjoyed that experiment and was vaguely aware of its potential significance, but I was not rushing to write it up.

My reluctance was partly due to pressure of work on the normal state properties and partly because I was concerned that, with only seven data points for superconducting samples, its apparent conclusions might simply be wrong! At that stage of the project I had the privilege of discussing it with Ted, and his enthusiasm and encouragement were absolutely key to driving me on to writing the paper [11]. As a young scientist, I learned so much from those early discussions with Ted, and I have tried hard to remember them as I grow older (though still a spring chicken in his terms!). Not coming from the US community, and being badly ignorant about scientific history, I really had not known his incredible pedigree until talking with him. He liked the work because, to him, it was a good example of real life materials physics. We all know that he is extremely strong in terms of chemical intuition, but he was so knowledgeable about superconductivity theory as well, and he instantly recognized the significance of the data seeming to follow the Abrikosov-Gorko's pair breaking function. The thing that really came through to me from the conversations was his ability to inspire and the simple way he achieved that. He is always tough on scientific matters, but he is also unfailingly kind and constructive and has the precious ability to make young people feel good about themselves and the work they are doing. I try to act in a similar way when I can, and often cross-refer to him as my example.

Yoshi Maeno is a consummate materials physicist (also influenced by Ted of course!) and he reacted to our findings by leading his group's refinement of the sample preparation techniques to the level where it became possible to grow crystals with $T_c \sim 1.5$ K as predicted by extrapolation of the pair breaking function to the high purity limit [15]. That refinement continues to this day [16], and means that single crystals of Sr_2RuO_4 with mean free paths of microns are available for the world-wide collaborative research that Yoshi has stimulated by his wonderfully unselfish attitude to sharing the crystals with other groups. The refinement in crystal quality in turn made it possible to deliberately disorder pristine crystals with controlled levels of impurities introduced during the growth process, and hence for a number of groups to check the validity of the data we had published [12–14]. To my relief, our

Fig. 1 **a** Original data from our group on the purity dependence of T_c in Sr_2RuO_4 [11]. **b** The results of some of the follow-up confirmatory work by other groups [12–14].



work stood up to that scrutiny, as evidenced by the data shown in Fig. 1b.

‘Living’ an experiment the way we lived the impurity one on Sr₂RuO₄ is a good way of maximizing what one can learn from it, and it helped me form the opinion that dirty samples are hiding legions of unconventional superconductors that we would be able to discover in the clean limit. The disorder-sensitivity of other unconventional superconductivity has been noted or studied before and since our work, but to the best of my knowledge Sr₂RuO₄ still holds the record as the superconductor with the most stringent and proven mean-free-path criterion ($>\sim 10^3$ Å) for the existence of the superconductivity. That that should be the case comes as something of a surprise, and I look forward to the record being broken soon!

Our experiment showing that Sr₂RuO₄ must have an unconventional order parameter was followed very quickly by two highly influential pieces of work that have largely defined the field until the last year. In the first, Luke and collaborators presented muon spin rotation evidence that a spontaneous magnetic field was generated in the superconducting state of Sr₂RuO₄, implying an order parameter in which time reversal symmetry is broken [17]. Shortly afterwards, Ishida and collaborators reported that the NMR Knight shift remained unchanged as the sample is cooled through T_c [18]. This strongly implied that the spin susceptibility was the same in the superconducting and normal states, something that is impossible in the presence of spin singlet pairing for which the formation of non-magnetic singlets suppresses the spin susceptibility. Both these observations were consistent with one of the triplet order parameters suggested in the classic prediction of Rice and Sigrist, and both were subsequently confirmed in other experiments [19, 20]. Combined with evidence from tunnelling experiments for two-component order parameters [21] with odd parity [22], this all seemed to add up to compelling evidence for a real ‘feel-good’ situation—a superconductor whose order parameter was a two-dimensional version of that of the A phase of superfluid ³He. This excitement of this possibility gained further momentum with the publication of a second beautiful theoretical idea, namely that such a superconductor can host topologically protected Majorana zero modes in its vortex cores [23]. It was clear that the superconducting gap contained nodes or very deep gap minima which would severely restrict any practical use of the Majorana modes [24], but as long as the nodes were accidental they did not rule out a topological state.

When a beautiful hypothesis exists in some physical situation, it is always tempting to use it as the reference interpretation for subsequent work. This certainly happened in the case of Sr₂RuO₄, for which many experiments over the next decade were shown to give results consistent with the prevailing received wisdom, without, however, providing clear proof in favour of it. Balancing these results were another set of experiments that gave reasons for doubt. For example, neither the

critical field splitting expected near T_c nor the internal fields or edge currents predicted naively expected for a time reversal symmetry breaking superconductor were observed [25, 26]. Some ingenious theoretical work [27, 28] showed that the edge currents need not be a firm expectation in a multi-band superconductor like Sr₂RuO₄, so it was difficult to judge whether these counter-experiments were fatal to the received wisdom—was the cup half full or half empty?

Probably my biggest personal concern about the situation was another discrepancy concerning the anisotropy of the critical magnetic fields. In a strongly two-dimensional superconductor, the critical field anisotropy should be calculable from knowledge of the anisotropy of the Fermi surface. I was pretty confident that our de Haas-van Alphen experiments had established the Fermi surface anisotropy, and it suggested that the critical field anisotropy should be very large—at least a factor of 100. What was measured was approximately a factor of 40 [1]. One would not normally worry about a factor of 2.5 numerical discrepancy, but it was also established that there is a first-order phase transition out of the superconducting state at high magnetic fields [29, 30]. Taken together, these suggested that the critical fields were behaving as if they were limited by the Zeeman energy of breaking Cooper pairs. This would be fine in a standard superconductor but was in direct contradiction to the Knight shift results which suggested that there should be no Zeeman energy gain from breaking Cooper pairs.

Throughout the decade or more during which the above-described situation was developing, my own group was doing little or no work on Sr₂RuO₄ superconductivity, and I played the role of an interested observer. In late 2012, however, we started to work in the field again, but from a different angle. Following a workshop discussion with Jim Sauls, my then post-doc Clifford Hicks proposed a new way of applying uniaxial pressure that would in principle enable a number of desirable things, namely application of both compressive and tensile pressure, pressure actuation at low temperatures based on piezo-electric stacks, and, in ultimately favourable circumstances, the generation of large strains of order 1% or more. It seemed to me that so many things could go wrong that the apparatus was pretty unlikely to work, but I agreed with Clifford that we should put some time and resource into it, in the hope that my pessimism would be unfounded. Happily it was [31].

The original motivation for the uniaxial pressure project that emerged from the conversation with Jim Sauls was to search for one of the expectations for a time reversal symmetry breaking order parameter, namely a cusp in T_c at zero uniaxial pressure. As is so often the case with experiments designed to search for some specific phenomenon, we did not observe it, either in the original experiment [32] or a more sophisticated follow-up in collaboration with the group of Ted’s colleague Kathryn Moler at Stanford [33]. However,

the experiment reaped rewards that we had not thought of when we conceived it. It was possible to increase T_c , not by a few mK but by a large factor of 2.4, up to approximately 3.5 K [34]. We have subsequently performed follow-up measurements showing that this increase of T_c is due to pressure tuning through a Lifshitz transition [35], but even without this microscopic understanding, the experiment enabled a re-examination of the critical field limiting. What had been a worrying but numerically fairly small discrepancy in the as-grown material would be a major contradiction in $T_c = 3.5$ K material if it had a spin triplet order parameter [34].

Disagreement between experiment and theory is both common and healthy in correlated electron physics for which developing appropriate many-body theory is one of the big challenges for twenty-first century physics. Strong apparent disagreement between the results of two complementary experiments is a different level of discrepancy and merits careful investigation. Working with Thomas Scaffidi, Clifford and Yoshi Maeno, we stressed this and other puzzles in a 2017 review article that emphasized the importance of re-examining the received wisdom about the order parameter of Sr_2RuO_4 [5]. This challenge was taken up by Stuart Brown at UCLA, whose group set out to study the Knight shift of Sr_2RuO_4 under uniaxial pressure, in collaboration with our group in Dresden. The striking results obtained in that project are my main motivation for writing this short perspective. Stuart and colleagues began by demonstrating that, in the pressurized $T_c = 3.5$ K material, the Knight shift does not remain constant below T_c , but shows a significant drop that is qualitatively consistent with expectation for singlet rather than triplet pairing. This observation naturally raised the question of what was happening at ambient pressure. In a series of careful experiments, they showed that the famous previous result from Ishida's group was due to transient heating and that in fact the

Knight shift changes strongly below T_c at all pressures [36]. At a stroke, the discrepancy between the NMR and critical field limiting experiments has been removed!

One of the real pleasures of working on Sr_2RuO_4 has always been the interaction with the world-wide community of colleagues who find the material as fascinating as I do. We do not always agree on the science or interpretation, but we work in an atmosphere of constructive discussion and mutual respect. In other fields that I have observed or worked in, a result like Stuart's that overturns a received wisdom can be the prelude to years of unconstructive dispute that obstructs scientific advance. For me, Ted is a guardian of the standards of scientific behaviour, so I am sure that he will approve of what happened in this case. We made sure, before posting our results on the arXiv, that we had described Stuart's experiments in detail to Kenji Ishida. He freely admitted that they came as an unwelcome shock to him, but he reacted in the most admirable way. He went back into the laboratory personally, re-checked his experiments in light of Stuart's findings, and contacted us within weeks to confirm his agreement with the new results. At time of writing he has placed a detailed paper on the arXiv describing what he has done [37].

In my opinion, these very recent developments leave us at a key stage in the quest to understand the unconventional superconductivity of Sr_2RuO_4 . They make it likely that Sr_2RuO_4 is not an odd parity, spin triplet superconductor but an even parity, spin singlet one. It is interesting to observe the reaction of colleagues to this new development. At one level, it comes as a disappointment; a solid-state superconducting analogue to the A phase of superfluid ^3He is such a nice idea! Understandably, people who have worked intensively on that idea, and/or on experiments whose results aligned well with it, are reluctant to change their world view immediately. That is entirely appropriate, and a re-examination of some of the other

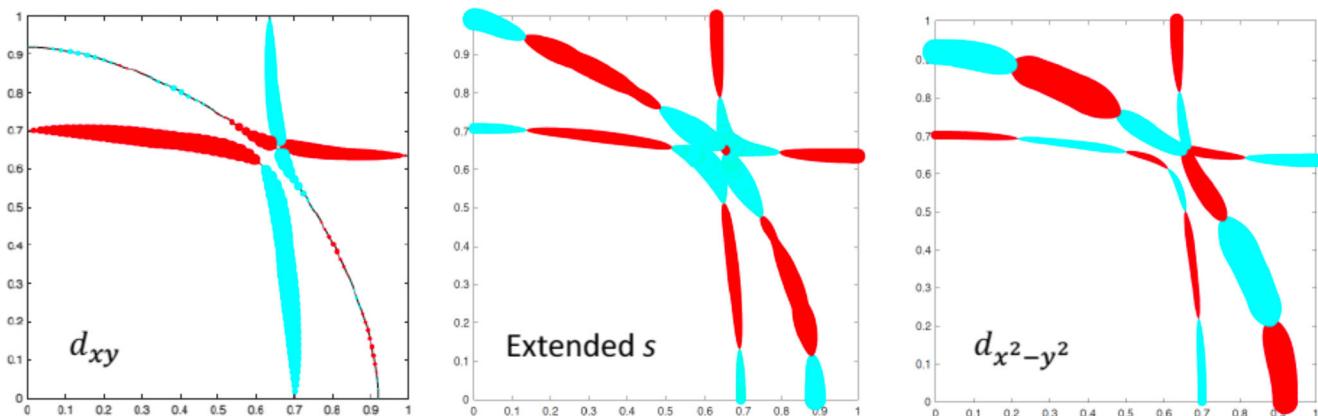


Fig. 2 Illustrative calculation of possible gap structures in Sr_2RuO_4 by Thomas Scaffidi, based on the weak-coupling model of ref. [38]. In each case, one quadrant of the 2D square Brillouin zone is shown, with sign reversals of the order parameter denoted by changes from red to blue.

While not identical, the nodal structures of the three candidate even parity states some striking similarities that run counter to the intuition that one would have based on single-band thinking.

existing results should be a high priority for the future. However, it is important that this is all done with cool heads and open minds. The extreme viewpoints of either clinging on to the triplet interpretation or throwing it out of the window without trying to revisit the outstanding mysteries would be equally unhelpful as the field moves forward. It seems to me that the two biggest issues will be establishing whether time reversal symmetry is really broken at T_c , and increasing efforts to perform measurements sensitive to parity.

Arguably, the biggest issue of all moving forward is whether the effort of doing so is merited. A number of people question whether Sr_2RuO_4 warrants further study at all if it does not turn out to be the ${}^3\text{He}$ analogue of our dreams. This is a perfectly valid issue to raise, but it is one on which I hold a very strong opinion: actually, it is vital that this work be done. For several reasons, it goes to the very heart of how unconventional superconductivity condenses from correlated electron metallic states. Sr_2RuO_4 is the cleanest of all known unconventional superconductors and has one of the simplest and best understood normal states. It is an ideal test-bed for theoretical ideas and for establishing the validity of information coming from the key experiments that are widely applied to unconventional superconductors. If these experiments give contradictory results in a material like Sr_2RuO_4 , how can we just assume that they are reliable when applied to other materials? Although it is sometimes convenient to assume that reliability and use it to assign order parameter classifications based on sparse information, that approach will not lead to progress in the long run. The second reason that Sr_2RuO_4 should remain as an archetype for the field is as a guide to the development of theory. Its low T_c makes the use of weak-coupling theory more justifiable on Sr_2RuO_4 than on most other unconventional superconductors, and the application of that theory within a two-dimensional approximation has already been very revealing. The free energy differences between odd- and even-parity superconducting states is small, almost certainly below the true accuracy (including the validity of assumptions made) of the calculations. In other words, the Sr_2RuO_4 problem is a hard one, and establishing the true experimental picture beyond doubt is therefore a vital guide to the development of theories of the mechanism of unconventional superconductivity as a whole.

To frame that quest I will make unusual move of closing this perspective with a second figure, courtesy of my colleague Thomas Scaffidi. Based on the weak-coupling approach of ref. [38] which includes spin-orbit coupling, it shows typical calculated gap functions for three distinct even parity order parameters, d_{xy} , extended s and $d_{x^2-y^2}$. In simple single-band thinking, these order parameters would have completely different nodal structures, but in three-band Sr_2RuO_4 the distinction is much weaker. Symmetry-imposed nodes are of course present, but each state also has a rich

structure of accidental nodes. The overall message to be taken from inspection of Fig. 2 is that, in a model unconventional superconductor in the weak-coupling limit, the nodal structure is determined not by symmetry alone but by its combination with a complicated spin fluctuation spectrum, with the latter being arguably more influential on the overall nodal structure than the former. Even if the particular theory that led to Fig. 2 is not the perfect theory of Sr_2RuO_4 , this is a sobering observation, because it warns us that the relationship between standard physical observables and order parameter symmetries may not be nearly as simple as we usually assume, either in Sr_2RuO_4 or many other unconventional superconductors. For this and other reasons, unravelling the Sr_2RuO_4 problem is, in my opinion, more important than ever. However, I am optimistic. The new NMR results have resulted in a paradigm shift in the field, but a welcome one that should be viewed as opening previously closed paths to an overall understanding.

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