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FROM SUPERFLUID ^{3}HE TO TRIPLET SUPERCONDUCTOR SR_2RUO_4

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The post BCS development of the field of superconductivity and its implication to physics are briefly reviewed. After superfluid ^{3}He, heavy fermion superconductors, organic superconductors, and high-T_c superconductors, the garden of superconductivity is inhabited by unconventional (i.e. non-s-wave) superconductors. In particular, aspects of d-wave superconductivity are tested semi-quantitatively on high quality single crystals of YBCO and Bi2212.

1 Prologue

> *Per correr miglior acque alza le vele omai la navicella del mio ingegno, ...*
>
> Dante, *Purgatorio*

One of us (K.M.) met Hagen Kleinert for the first time in the spring of 1978. A young guy jumped into my office unannounced and started talking about his new ideas on topological defects in the superfluid ^{3}He. Superfluid ^{3}He was discovered in 1972 by Doug Osheroff, Bob Richardson, and Dave Lee, at Cornell University, Ithaca [1,2]. After the success of the BCS theory [3] in describing the superconductivity phenomenon in metals, many authors [4–6] considered possible superfluid ^{3}He based on the similar pairing of ^{3}He atoms.

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However, there was almost insurmountable difficulty in predicting the superfluid transition temperature and the symmetry of the pairing, though the possibility of s-wave pairing had been rejected early in the game as very unlikely due to the strong repulsive interaction between two ^{3}He atoms. In the summer of 1972 at the 13th Low Temperature Conference (LT13) at Boulder, Colorado, a special session on this subject was announced. I was in euphoria as if a brave new world was unfolding in front of me and I realized that I was witnessing the almost unique event in my lifetime. After ingenious NMR experiments at Cornell [2] and a brilliant theory by Tony Leggett [7], it became clear that the superfluid ^{3}He is of spin triplet p-wave pairing and consists of at least two distinct phases: an A phase and a B phase. Unlike s-wave superconductors in metals, the superfluid ^{3}He possesses a large internal degree of freedom which manifests itself as several Nambu-Goldstone modes (zero sound, spin waves, orbital waves) and a large class of topological defects [8,9]. In order to catch up this rapid development I moved in 1974 from the Tohoku University, Sendai, Japan to the University of Southern California, which is situated at two hours driving distance from the University of California, San Diego (UCSD) in La Jolla. After the discovery of superfluid ^{3}He in 1972, John Wheatley at La Jolla had done a number of ingenious experiments on superfluid ^{3}He including magnetic ringing, fourth sound, and zero sound in uniform and non-uniform textures [10]. At that time, Tsuneto and I just got a striking confirmation of our theory of magnetic ringing in the superfluid ^{3}He-A [11,12]. Actually Hagen was visiting UCSD at La Jolla in 1978 on his sabbatical. When he tried to discuss his ideas with Wheatley, it was then very natural that Wheatley suggested that Hagen should talk with me.

For us many-body theoreticians, the superfluid ^{3}He provides a wonderful playground where exotic topological objects abound. Just before Hagen came to my office, Kumar and I had succeeded in interpreting a strange NMR satellite, first reported [13] at LT14 in Helsinki in 1975 and later explored in more detail by Gould and Lee [14] in terms of a "soliton" or a "domain wall" [15,16]. This was the first topological defect observed and identified in superfluid ^{3}He. In 1978, Yu Ren Lin-Liu and I were puzzling about the instability of the uniform texture (i.e. $\mathbf{l} \parallel \mathbf{d}$) and $\mathbf{l}$ and $\mathbf{d}$ constant all over the space, where $\mathbf{l}$ is the quantization axis of the orbital angular momentum and $\mathbf{d}$ is the spin vector in superfluid ^{3}He-A. At that time, P. Bhattacharya *et al.* [17] had just published an elegant and intriguing theory about the critical point where the uniform texture becomes unstable in the presence of superflow. The question was what happens to the system after this? We could write down a set of

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coupled differential equations for **l** and **d**. But the usual solution method led us to nowhere. This was where Hagen entered the discussion with a bright idea how to cook the equations, which enabled us to avoid unphysical singularities. Indeed we found the helical texture [18] which was reported at LT15 in Grenoble, France in the summer of 1978. The helical texture is characterized as the static texture where both **l** and $\mathbf{d} \parallel \mathbf{l}$ are winding around the superfluid velocity $\mathbf{v}_s$ [8]. A part of the result and the phase diagram [18,19] was confirmed through the measurement of the abrupt change in the sound attenuation [20]. After this successful collaboration, Hagen invited me to spend one month at the Freie Universität Berlin at Dahlem in the summer of 1980. At that time, the blue phases in liquid crystal were one of the hot topics in this field. Together, we set up a Ginzburg-Landau equation appropriate to the system and tried to find a stable crystalline-like solution. Besides usual tetrahedral and octahedral solutions, we considered the possibility of a dodecahedral phase and described in great detail the complicated icosahedral phase. This was before icosahedral phases were discovered in sputtered aluminium. Our paper [21] would have been completely forgotten if Wright and Mermin [22] had not kindly mentioned our work in their concise review on the blue phases.

Since then, our paths have diverged almost completely. While Hagen developed a disorder field theory for statistical mechanics of line-like vortices and defects [23], worked on membranes and strings [24–26], and found a variational perturbation theory of critical phenomena [27], I focussed attention on the exciting completely new classes of superconductors which appeared on the scene.

2 Day of Unconventional Superconductors

> *How beauteous mankind is! O brave new world that has such people in't!*
>
> Shakespeare, *The Tempest*

The superconducting $CeCu_2Si_2$ was the first heavy fermion superconductor discovered in 1979 [28]. Heavy fermion superconductors are found mostly in intermetallic compounds based on Ce or U [29,30]. We call them the heavy fermions since the mass of the quasi-particles involved in superconductivity is 100 to 1 000 times larger than the bare electron mass. First of all, the presence of superconductivity is very surprising since most of these systems are

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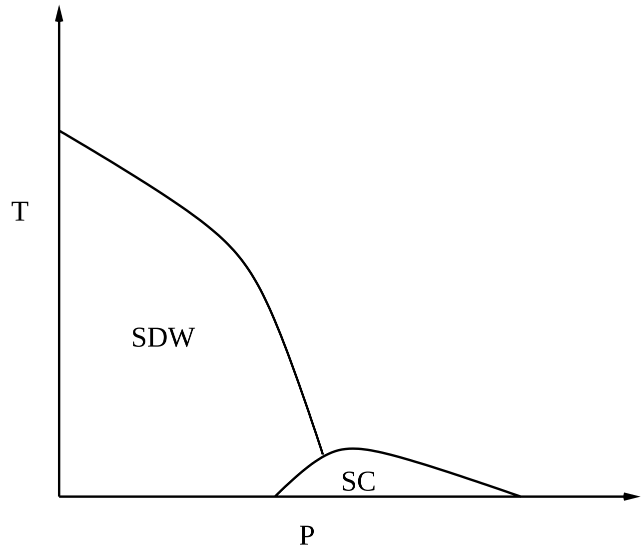

Figure 1. The schematic phase diagram of $(TMTSF)_2PF_6$ where T and P are the temperature and the pressure, respectively. The spin density wave (SDW) is destroyed as the pressure increases and is replaced by the superconducting state (SC).

magnetic, implying the Coulomb dominance [31]; the Coulomb interaction is stronger than the phonon exchange interaction. Indeed a later experiment shows the nodal lines in the superconducting order parameter $\Delta(\mathbf{k})$ [29]; $\Delta(\mathbf{k}) = 0$ along lines on the Fermi surface, which is rather common in unconventional superconductors (non-s-wave). Here $\mathbf{k}$ is the quasi-particle wave vector.

Almost at the same time the superconductivity in an organic conductor $(TMTSF)_2PF_6$, also called Bechgaard salts, was discovered by Jerome *et al.* [32] at Orsay. The Bechgaard salts is a quasi one-dimensional system with strong anisotropy in the electric conductivity. It has the particular phase diagram where the spin density wave (SDW) exists next to the superconducting state [32] and the absence of Hebel-Slichiter peak in T_1^{-1} in NMR [33,34] indicates unconventional superconductors (see Fig. 1). After that, a variety of organic superconductors have been discovered [35,36]. It appears that most of them are unconventional. In particular there is evidence indicating that the superconductivity in Bechgaard salts is of p-wave while the one in κ-$(ET)_2$ salts is of d-wave [37–39]. This development culminated in 1986 in the discovery of the high-T_c cuprate superconductor $La_{2-x}Ba_xCuO_4$ by Bednorz and

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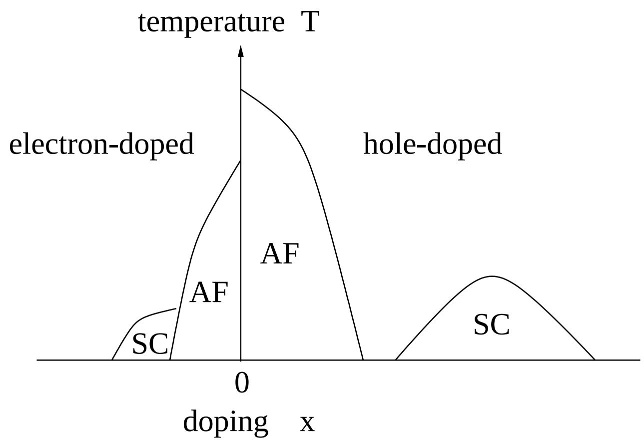

Figure 2. The schematic diagram of the hole-doped ($x > 0$) and the electron-doped ($x < 0$) high-T_c cuprates. Here AF means the antiferromagnetic state and SC the d-wave superconducting state.

Müller [40]. Within a few years the superconducting transition temperature T_c increased from 35 K to 165 K, though these temperatures are still much lower than room temperature. All of these high-T_c cuprates have a layered structure and have the Cu-O_2 planes as basic elements.

These cuprates are also insulating and in an antiferromagnetic state in the absence of electron or hole doping. As the carrier density increases, the antiferromagnetic state is destroyed such that the superconductivity arises with further increase in the carrier density. This behavior is sketched in Fig. 2. For $x > 0$ we present a typical phase diagram of the hole doped cuprates. For the hole concentration around 0.15–0.2, the superconducting transition temperature reaches the maximum value. Further the dependence of the superconducting transition temperature is well approximated by

$$T_c(x) = T_c^0 - a(x - x_0)^2, \tag{1}$$

where T_c^0 is the superconducting transition temperature at the optimal doping (i.e. $x = x_0$) and $a = T_c^0/x_0^2$. On the other hand, T_c of the electron-doped cuprates decreases monotonically with increasing electron density. In order to understand this phase diagram, P.W. Anderson [41] proposed his famous dogma, stating that

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a) all the actions take place in the Cu-O_2 plane;
b) the phase diagram should be understood in terms of a single Hamiltonian. Here, the presence of the antiferromagnetic phase implies the Coulomb dominance;
c) one should propose a two-dimensional one-band Hubbard model for high-T_c cuprate superconductor which is the consequence of a) and b).

Of course the Hubbard model is considered as the simplest model to describe magnetism. It may be surprising that a similar Hamiltonian can describe the high-T_c superconductors. Unfortunately the complete solution of the 2D one-band Hubbard model is still not available. However, it appears that the following is certain:
a) The normal state is the Fermi liquid, though unlike the usual Fermi liquid some of the nesting channels (i.e. $\mathbf{q} = (\pi,\pi)$) play an important role. This point was clarified by Shankar [42] and others [43,44].
b) There is strong spin fluctuation (antiparamagnon) in this model, which gives rise to d-wave pairing (i.e. $\Delta(\mathbf{k}) \sim \cos(2\phi)$ where $\phi = \tan^{-1}(k_y/k_x)$) [45–47].
c) The superconductivity is well described by the BCS theory of d-wave superconductor. In general, the mean-field theory appears to apply for all unconventional superconductors [48].

We consider the period 1993-1994 to be the most important time for understanding high-T_c cuprate superconductors, where a number of ingenious methods of exploring d-wave order parameter were developed. Also the availability of high quality single crystals of high-T_c cuprate superconductors provides indispensable support for this success. Perhaps one of the most important experiments is the phase sensitive test using the Josephson interference effect. First Wollman *et al.* [49] constructed a SQUID configuration between YBCO and Pb and studied the interference pattern. They saw the shift, i.e. the peak position shifted by π [see Fig. 3(b)] and current I versus Φ/Φ_0, where $\Phi_0 = hc/2e$ is the quantum flux. This shift reflects the fact that the order parameter $\Delta(\mathbf{k})$ has the opposite sign for $\mathbf{k} \parallel \mathbf{a}$ and $\mathbf{k} \parallel \mathbf{b}$. In another experiment the whole corner of YBCO was covered by Pb. There instead of a usual Frauenhofer pattern, they observed the anti-Frauenhofer pattern (see Fig. 3(d)) [50].

Also the tricrystal geometry was exploited by Tsuei and Kirtley [51]. They grew three crystals mutually oriented by 60° to each other epitaxially. If we are here dealing with d-wave superconductors, the order parameter has to

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change the sign three times when coming back to the starting crystal. But this is unacceptable. In order to resolve this frustration, there appears a half quantum flux at the center of these three crystals, which is detected by an extremely sensitive micromagnet-meter with the diameter ∼10 microns. With this special technique they have established d-wave superconductivity in YBCO, GdBCO, Bi2212, Tl2201, and more recently in two electron-doped high-T_c cuprates NCCO and PrCCO [51].

Also the $\mathbf{k}$ dependence of $\Delta(\mathbf{k})$ became accessible through the angular resolved photoemission spectrum(ARPES) done by Shen *et al.* [52,53].

Of course, the nodal lines in $\Delta(\mathbf{k})$ imply that the low temperature thermodynamic and transport properties are completely different from the ones in s-wave superconductors. For example, the magnetic penetration depth increases linearly in T while the specific heat like T^2 at low temperatures, which is observed in YBCO [54,55] and in LSCO [56]. Also the quasi-particle density of states, as seen by STM [57] and the electronic Raman scattering [58], exhibits clear d-wave signatures.

Further it is known that the impurity provides a fine proof for unconventional superconductors. Let us just indicate some papers on this subject [59,60].

Perhaps the vortex state will provide the better test of the BCS theory of d-wave superconductivity [61]. For $\mathbf{H} \parallel \mathbf{c}$ the most striking prediction is that the square vortex lattice tilted by 45° from the a-b axis is more stable than the usual hexagonal vortex lattice [62,63].

Though originally the prediction was made in the vicinity of the upper critical field, the square vortex lattice is seen in single crystals of YBCO at low temperatures ($T \simeq 4$ K) and at a low magnetic field ($H \simeq$ a few Tesla) by small angle neutron scattering [64] and in a scanning tunneling microscope [65]. Strictly speaking, the apex angle of the observed vortex lattice is 77° and not 90°. But this difference is easily understood in terms of the a-b anisotropy in YBCO. Due to the orthorhombic distortion in YBCO, the coherence lengths ξ_a and ξ_b are not equal but $\xi_b/\xi_a \simeq 1.5$, where the subscripts a and b mean the component parallel to the a- and b-axis, respectively.

Still controversial are also the thermodynamic and the transport properties of the vortex state. Concerning the weak magnetic field (i.e. $H/H_{c2} \ll 1$, where H_{c2} is the upper critical field), Volovik [66] has written the seminal paper that the effect of the magnetic field can be treated quasi-classically. This approach was generalized by a number of studies, including one by us [67–69]. More exciting is that the careful measurement of thermodynamics, NMR, and

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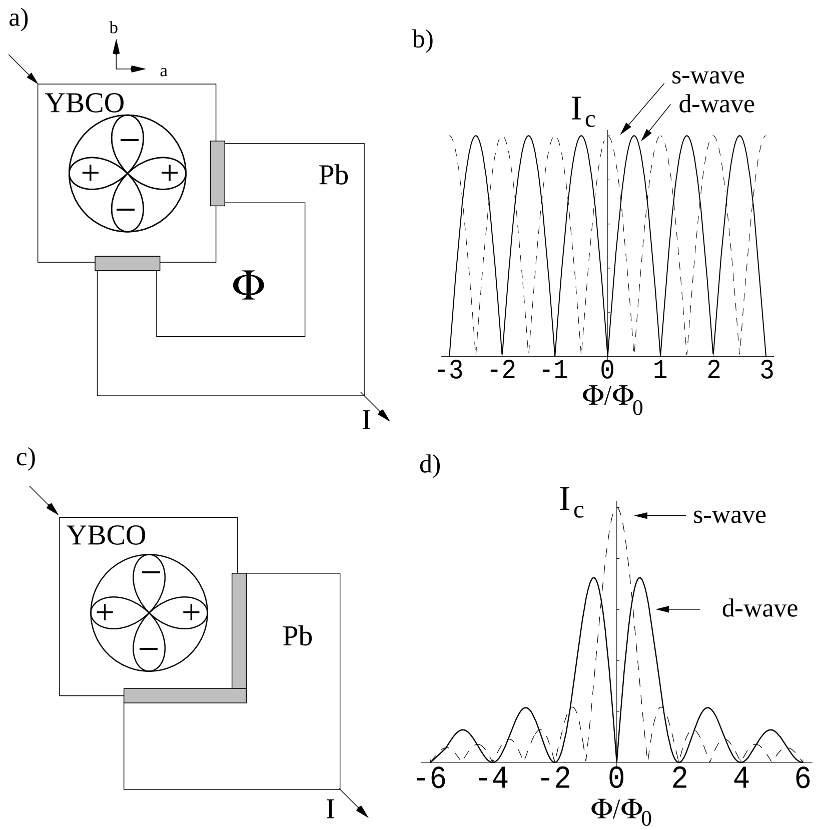

Figure 3. a) The Josephson interferometry between d-wave superconductor (YBCO) and s-wave superconductor (Pb). The critical current I_c depends on the magnetic flux Φ as shown in b). Therefore the observed π shift provides the test of the d-wave superconductor. c) The corner junction where the corner of YBCO is covered by Pb. Here the magnetic flux Φ is distributed along the boundary between YBCO and Pb. The d-wave superconductor then exhibits the anti-Frauenhofer pattern as shown in d).

thermal conductivity in the vortex state in YBCO and Bi2212 at low temperature have been reported, which confirms in general the predicted $\sqrt{H}$-dependence of the specific heat [55] and the spin susceptibility seen through the Knight shift and the linear H-dependence of T_1^{-1} (the nuclear spin lattice relaxation rate) in NMR. In the future, the single crystals of YBCO and Bi2212 will provide an extremely useful testing ground for new ideas and concepts in unconventional superconductors.

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Among heavy fermion superconductors, UPt_3 is the only system where the nature of the symmetry is well established [30]. UPt_3 is the hexagonal crystal with an axis parallel to the c-axis. At low temperatures ($T \ll T_c$ =0.55 K), both the electronic thermal conductivity κ_c and κ_b behave linear in T, where the subscripts c and b mean parallel to the c-axis and the b-axis [70]. The simplest possibility consistent with this is $\Delta(\mathbf{k}) \sim Y_{3,\pm2}(\theta,\phi)$ or E_{2u}, where $Y_{3,\pm2}(\theta,\phi)$ is the spherical harmonics [71,72]. The Knight shift seen by NMR almost at the same time confirmed the triplet spin pairing [73]. Later the details of the spin configuration in the A, B, and C phases were identified [74]. In 1999, a paper confirmed that both the upper critical field and the ultrasonic attenuation data of UPt_3 are consistent with E_{2u} [75]. Therefore, UPt_3 appears to provide another nice system to test new ideas on unconventional superconductors.

3 Story of Sr_2RuO_4

Ch'ebbe l'origine nell'Alemagna, che poi sì celebre là in Francia fu.
Mozart/da Ponte, *Così fan tutte*

The superconductivity in Sr_2RuO_4 was discovered in 1994 by Maeno *et al.* [76]. This is an isocrystal to La_2CuO_4, a mother system to $La_{2-x}Sr_xCuO_4$, the high temperature cuprates. But unlike HTSC it is already metallic while La_2CuO_4 is insulating and antiferromagnetic. Also, at low temperatures ($T <$ 20 K) the electron in Sr_2RuO_4 behaves as the Fermi liquid. Further, the system becomes superconducting below $T \sim 1$ K.

From the analogy to superfluid ^{3}He it was then proposed [77] that the superconductor is of triplet p-wave with $\mathbf{\Delta}(\mathbf{k}) \sim \hat{d}(k_1 \pm ik_2) = \hat{d}e^{i\phi}$ and the spin vector $\mathbf{d}$ parallel to the c-axis. Both the spontaneous spin polarization observed by the muon spin rotation experiment [78] and a flat Knight shift [79] in the superconducting state of Sr_2RuO_4 confirm the triplet nature of the pairing and also the sensitivity to the disorder points to the unconventional superconductivity [80].

But recently the situation changed dramatically by the availability of the high quality single crystals of Sr_2RuO_4 with $T \sim 1.5$ K. As shown in Figs. 4 and 5, both the specific heat [81] and the superfluid density [82] suggest there is no energy gap. Indeed, the overall behavior is much more consistent with d-wave superconductors [83]. So many people proposed possible f-wave superconductivity in Sr_2RuO_4 ($\Delta(\mathbf{k}) \sim \cos(2\phi)e^{\pm i\phi}$, $\Delta(\mathbf{k}) \sim \sin(2\phi)e^{\pm i\phi}$,

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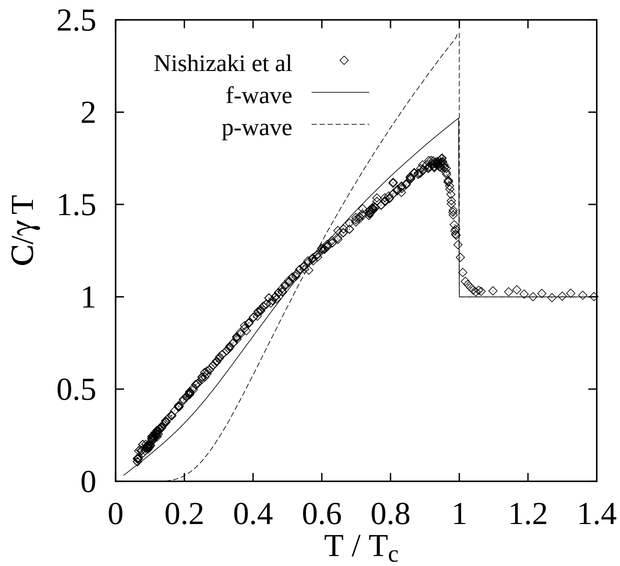

Figure 4. The specific-heat data [76] divided by γT, where γ is the Sommerfeld constant, is compared with the theoretical results for the p-wave [72] and f-wave [81] superconductors.

and $\cos(ck_3)e^{\pm i\phi}$) [83–87]. In particular, within the weak-coupling theory, these three f-states have the same thermodynamics as the one in d-wave superconductors [88]. Therefore the thermodynamic data cannot discriminate one from the other.

Another result is that the specific heat in the vortex state for $\mathbf{H} \parallel \mathbf{c}$ at $T =$ 0.1 K exhibits clearly the $\sqrt{H}$-behavior (see Fig. 6) [81,85]. The deviation from the $\sqrt{H}$-behavior for $H < 0.01$ T is most likely due to the fact that the system is in the Meissner state. Additionally, the thermal conductivity in the vortex state at low temperature exhibits the H-linear dependence [85,89,90], which indicates not only the nodal structure in $\Delta(\mathbf{k})$ but also that the system is in the superclean limit ($\Gamma/\Delta \ll H/H_{c2} \ll 1$, where Γ is the quasi-particle scattering rate). Indeed, the quasi-particle mean free path of these systems is much longer than a few micrometers.

To choose the correct order parameter, we need experiments which are sensitive to the anisotropy within the a-b plane. A first experiment of such a kind was provided by the upper critical field in a planar magnetic field [91]. Indeed, they found a fourfold term in the upper critical field or $H_{c2}(\theta, T)$

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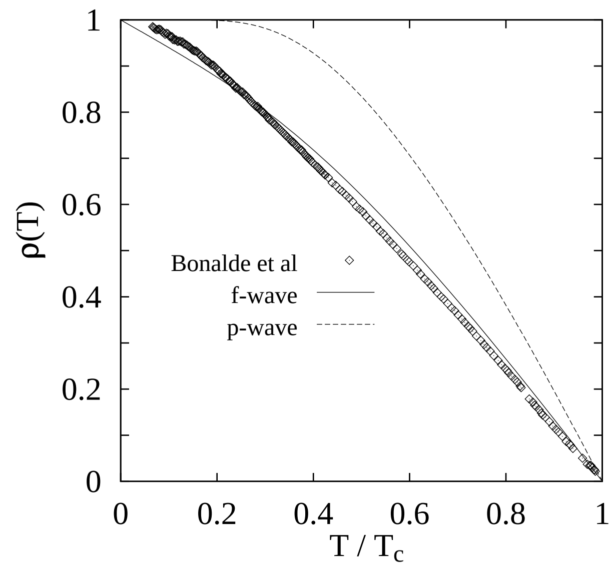

Figure 5. The superfluid density data [77] is compared with the p-wave and f-wave models.

which is approximated by

$$H_{c2}(\theta, T) = H^0_{c2}(T) - H^1_{c2}(T)\cos(4\theta), \tag{2}$$

where θ is the angle which the magnetic field makes with the a-axis. However, $H^1_{c2}(T)$ is rather small ($H^1_{c2}(T)/H^0_{c2}(T) \sim 3$ %). This suggests strongly that this anisotropy does not reflect the symmetry of $\Delta(\mathbf{k})$ but rather the band structure effect [92,93]. Indeed both $\cos(2\phi)e^{\pm i\phi}$ and $\sin(2\phi)e^{\pm i\phi}$ should exhibit large anisotropy ($\sim$ 30 %) and therefore they are incompatible with the experiment. The thermal conductivity in the vortex state in a planar magnetic field has also been measured recently: it shows extremely small anisotropy [90,94]. Therefore, the thermal conductivity data are also incompatible with $\cos(2\phi)e^{\pm i\phi}$, and $\sin(2\phi)e^{\pm i\phi}$. This leaves us only with $\cos(ck_3)e^{\pm i\phi}$, though this state requires a strong interlayer spin coupling or interlayer Coulomb interaction [95].

Since the discovery of unconventional superconductors, the collective modes and the possible topological defects have been considered [29]. However, so far there is no evidence for the collective modes or topological defects, except for the usual Abrikosov vortex. Perhaps this situation may change

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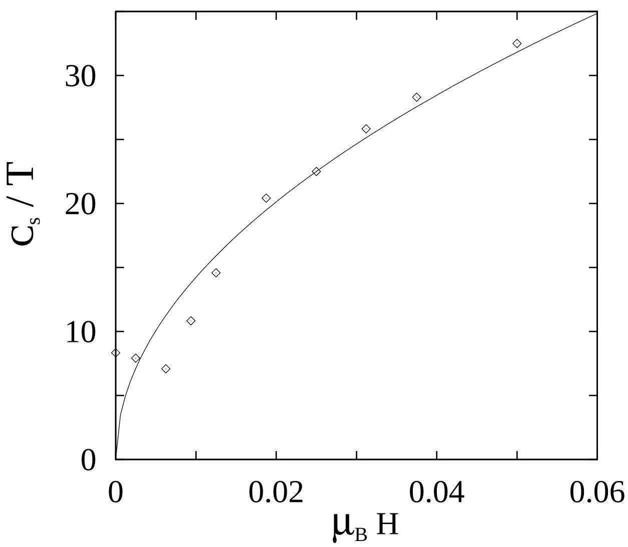

Figure 6. The specific-heat data of Sr_2RuO_4 in $\mathbf{H} \parallel \mathbf{c}$ and $T = 0.05$ K [76] is compared with the square-root of H. The deviation below $H \simeq 0.01$ T is due to the fact that $H_{c1} \simeq 0.01$ T.

drastically by the appearance of the triplet superconductors.

First of all, the triplet superconductors should have spin waves as collective modes [96,97]. Due to the two-dimensional representation there should also be the clapping mode [98,99], which couples to both the sound wave and the Raman photon. Unfortunately the coupling to the sound wave appears to be discouragingly small [99]. On the other hand, we believe that the Raman scattering is more promising if one can do the Raman scattering experiment below 100 mK. For this we clearly need the development of a new technology. The topological defect-like $\hat{\mathbf{l}}$-soliton [100] and $\hat{\mathbf{d}}$-soliton with half-quantum vortex [101] have also been predicted. So perhaps the single crystal of Sr_2RuO_4 will provide the unique laboratory to test these new concepts.

4 Outlook

O glücklich, wer noch hoffen kann, aus diesem Meer des Irrtums aufzutauchen!

Goethe, *Faust*

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We have seen that the field of superconductivity expanded enormously since 1979. Actually most of the new superconductors in heavy fermion systems, charge conjugated organic conductors and high-T_c cuprates are unconventional. Therefore, unconventional superconductors will play a central role in the 21st century. Compared with conventional superconductors, unconventional superconductors are more sensitive, subtle and delicate to the environment. This will require much more delicate control of the sample preparation and the crystal formation. Their response to the external perturbation is more subtle and delicate. In spite of this, it is surprising that the mean-field theory as embodied in the BCS theory and the Landau theory of Fermi liquid works very fine in describing a manifold of phenomena. Can we trust in this approach for a long time? Of course, there are now many people claiming that the mean-field theory is unreliable. But if we limit ourselves to unconventional superconductors, we have not seen any failure or sign of failure of the mean-field theory. Quite parallel to this development we may have now unconventional charge density wave and spin density wave as well [102,103]. We believe that the understanding of all subtleties of these new superconductors is also crucial for the real application of unconventional superconductors, including high-T_c cuprate superconductors. So we may approach to soft matter physics from solid state physics through a different passage.

5 Acknowledgments

We are very happy to dedicate this article to the 60th birthday of Hagen Kleinert, our friend and colleague. We wish him fruitful work in coming years. On our trajectory from superfluid ^{3}He to triplet superconductor Sr_2RuO_4 we have enjoyed many collaborations and support. We would like to thank in particular Thomas Dahm, Balazs Dora, Peter Fulde, Stephan Haas, Takehiko Ishiguro, Koichi Izawa, Hae-Young Kee, Yong-Baek Kim, Mahito Kohmoto, Yoshiteru Maeno, Yuji Matsuda, Yoshifumi Morita, Jun'ichi Shiraishi, Makaryi Tanatar, Silvia Tomić, and Attila Virosztek for helpful collaborations. H.W. acknowledges the support from the Korea Research Foundation under the Professor Dispatching Scheme.

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