Gap Symmetry of Superconducting Borocarbide YNi_2B_2C and Skutterudite $PrOs_4Sb_{12}$

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Received on 23 May, 2003.

Within the quasiclassical approximation we have studied the thermodynamics and the thermal conductivity in the vortex state in nodal superconductors (sc). Recent angle dependent magnetothermal conductivity results indicate a gap function $\Delta(\mathbf{k})$ corresponding to f- wave sc in Sr_2RuO_4 and d- wave sc in $CeCoIn_5$ and κ-$(ET)_2Cu(NCS)_2$ respectively. More recently it is shown that $\Delta(\mathbf{k})$ in both YNi_2B_2C and $PrOs_4Sb_{12}$ have point nodes described by hybrid s+g wave gap function.

1 Introduction

The gap symmetry of superconductivity is the central issue since the appearance of many unconventional and/or nodal superconductors in heavy Fermion systems, organic superconductors, high T_c cuprate superconductors, Sr_2RuO_4 and borocarbides YNi_2B_2C and $LuNi_2B_2C$. Following the quasiclassical approach introduced by Volovik [2] it is possible to calculate both the thermodynamic and transport properties of the vortex state in nodal superconductors.

Especially in quasi 2D systems like high T_c cuprates, Sr_2RuO_4 , $CeCoIn_5$ and κ-ET_2X with X=$Cu(NCS)_4$, $Cu[N(CN)_2]Br$ and $Cu[N(CN)_2]Cl$ the thermal conductivity in a magnetic field within the conducting plane provides the crucial insight on the nodal structure of $\Delta(\mathbf{k})$ [3, 4]. Indeed Izawa et al have shown f-wave sc in Sr_2RuO_4 [5], d-wave sc in $CeCoIn_5$ [6] and κ-$(ET)_2Cu(NCS)_2$ [7] through the angle dependent magnetothermal conductivity. More recently gap functions $\Delta(\mathbf{k})$ in borocarbide YNi_2B_2C and skutterudite $PrOs_4Sb_{12}$ are shown to have point nodes and described in terms of s+g-wave sc [8-12]. We show $|\Delta(\mathbf{k})|$ of these superconductors in Fig. 1.

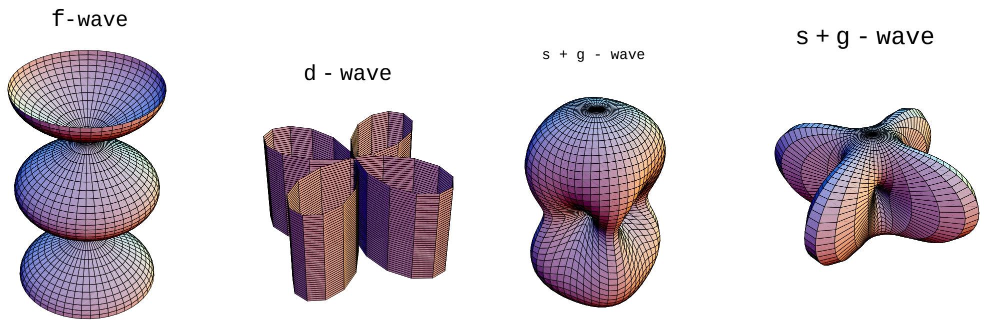

Figure 1. Anisotropic gap functions for Sr_2RuO_4 , $CeCoIn_5$, $PrOs_4Sb_{12}$ (A- phase) and YNi_2B_2C .

2 The Volovik effect

For simplicity we shall consider four examples of nodal $\Delta(\mathbf{k})$ in quasi- two dimensional systems; $\Delta(\mathbf{k}) = \Delta f(\mathbf{k})$ with f = $\cos(2\phi)$, $\sin(2\phi)$, $e^{\pm i\phi}\cos(ck_z)$, $e^{\pm i\phi}\cos(\frac{c}{2}k_z)$. They are $d_{x^2-y^2}$- wave, d_{xy}- wave, f- wave and f'- wave. The last one is introduced by Zhitomirsky and Rice [13] (i.e. the multigap model) in order to account for the nodal superconductor seen in Sr_2RuO_4 through the specific heat [14] and the magnetic penetration depth [15]. Indeed all four states have the same quasiparticle density of states as d-wave superconductors [16]. As we shall see later, however, the multigap model is incompatible with the angle dependent magnetothermal conductivity observed by Izawa et al [5] and the universal heat conduction in κ_{zz} observed by Suzuki et al [17] in Sr_2RuO_4 . On the other hand these thermal conductivity data are fully consistent with f-wave sc shown in Fig. 1.

In the absence of a magnetic field the quasiparticle density of states (DOS) of these four superconductors is given by [16]

$$g(x) = \frac{N(E)}{N_0} = \begin{cases} \frac{2}{\pi} x K(x) & \text{for } x = \frac{|E|}{\Delta} < 1 \\ \frac{2}{\pi} K(x^{-1}) & \text{for } x > 1 \end{cases} \tag{1}$$

Where N_0 is the the normal state DOS and K(x) is the complete elliptic integral of the first kind. In particular for $|x| \ll 1$

$$g(E) \simeq \frac{|E|}{\Delta} \tag{2}$$

Now in a magnetic field H perpendicular to the conducting plane Eq. (2) is replaced by

$$g(E,H) \simeq \left\langle \frac{|E - \mathbf{v}\cdot\mathbf{q}|}{\Delta} \right\rangle \tag{3}$$

Where **v**·**q** is the Doppler shift [18]; with **v** denoting the Fermi velocity and 2**q** the pair momentum describing the circulating superflow around vortices. Here $\langle ... \rangle$ means average over the Fermi surface and over the unit cell of the vortex lattice. In the present configuration this is readily done [2, 19]

$$g(0,H) = \frac{4v}{\pi d^2 \Delta} \int_0^{\frac{\pi}{2}} d\alpha \cos\alpha \int_0^d \frac{r dr}{2r} = \frac{2}{\pi} \frac{v\sqrt{eH}}{\Delta} \tag{4}$$

where we have assumed a square vortex lattice [20]. Here d=$\sqrt{eH}$. For a usual triangle lattice Eq. (4) will be multiplied by a factor $\sqrt{\sqrt{3}/2}$= 0.9306. . Then the specific heat, the spin susceptibility and the superfluid density at low temperature (i.e. T$\ll v\sqrt{eH} \ll \Delta_0$) are given by [21]

$$\begin{aligned} C_s/\gamma_n T &= \chi_s/\chi_n = g(0,H) \\ 1 - \rho_s(H)/\rho_s(H=0) &= g(0,H) \\ T_1^{-1}/T_{1n}^{-1} &= g^2(0,h) \end{aligned} \tag{5}$$

This $\sqrt{H}$ dependence of the specific heat has been seen in YBCO[22, 23], LSCO[24] and Sr_2RuO_4 [21]. Further the $\sqrt{H}$ dependence of χ_s and the H linear dependence of T_1^{-1} have been seen by NMR in a slightly underdoped Bi2212 [25]. Although the above expressions are obtained in the absence of impurity scattering, the Eqs. (4),(5) still hold in the superclean limit where $(\Delta\Gamma)^{\frac{1}{2}} \ll v\sqrt{eH}$ with Γ denoting the quasiparticle scattering rate in the normal state [3, 4]. There are only two energy scales in this limit, namely $v\sqrt{eH}$ and T. Therefore the thermodynamic functions obey the scaling law as proposed by Simon and Lee [26] and shown for a particular case by Kübert and Hirschfeld [19].

The thermal conductivity is treated similarly. In the absence of magnetic field κ_{xx} exhibits the universal heat conduction [27, 28] for all four superconductors.

$$\frac{\kappa_{xx}}{T} = \frac{2}{3}\pi \frac{n}{\Delta m} \tag{6}$$

This is also valid for κ_{zz} except for the f'- wave sc of the multigap model where we have

$$\frac{\kappa_{zz}}{T} = \frac{16}{3}\pi \frac{\Gamma n}{\Delta^2 m} \tag{7}$$

Thus for f'- wave κ_{zz} depends on the scattering rate Γ and never reaches the universality limit contrary to [17].

In a magnetic field **H** $\parallel$ **c** we obtain

$$\kappa_{xx} = \frac{2}{\pi^2} \frac{v^2(eH)}{\Delta^2} \tag{8}$$

in the superclean limit for all four superconductors. The H linear thermal conductivity has been seen in Sr_2RuO_4 [5].

3 Field angle dependent thermal conductivity

When the magnetic field lies in the a-b plane the Doppler shift generates the quasiparticles in the plane perpendicular to the magnetic field. When the plane cuts the nodal directions, there will be strong increase in the quasiparticle density. Therefore the thermal conductivity takes the maximum value when the magnetic field is perpendicular to the nodal directions [3, 4]. In particular in the superclean limit we obtain

$$\frac{\kappa_{xx}}{\kappa_n} = \frac{2}{\pi} \frac{\tilde{v}^2(eH)}{\Delta^2} I_i(\phi) \tag{9}$$

$$\frac{\kappa_{xy}}{\kappa_n} = -\frac{2}{\pi} \frac{\tilde{v}^2(eH)}{\Delta^2} J_i(\phi) \tag{10}$$

for i= 1,2,3,4 which corresponds to $d_{x^2-y^2}$- wave, d_{xy}-wave, f- wave and f'- wave superconductors respectively. Here $\tilde{v}^2 = v_a v_c$ and ϕ is the azimuthal angle of H with respect to the a axis. We obtain the following angular functions $I_i(\phi)$ and $J_i(\phi)$:

$$\begin{aligned} I_1(\phi) &\simeq 0.912 + 0.05\cos(4\phi) \\ I_2(\phi) &\simeq 0.925 - 0.256\cos(2\phi) \\ &\quad -0.042\cos(4\phi) \\ I_3(\phi) &\simeq 1.479 - 0.128\cos(2\phi) \\ I_4(\phi) &\simeq 0.439 - 0.128\cos(2\phi) \\ J_i(\phi) &\simeq -a_i \sin(2\phi) \end{aligned} \tag{11}$$

with a_1 = 0.256, a_2 = 0, a_3 = 0.124 and a_4 = 0.106. Their angular dependence is shown in Fig. 2. As is readily seen the ϕ- dependence of the thermal conductivity is adequate to identify one of four $\Delta(\mathbf{k})$ gap functions we have considered sofar.

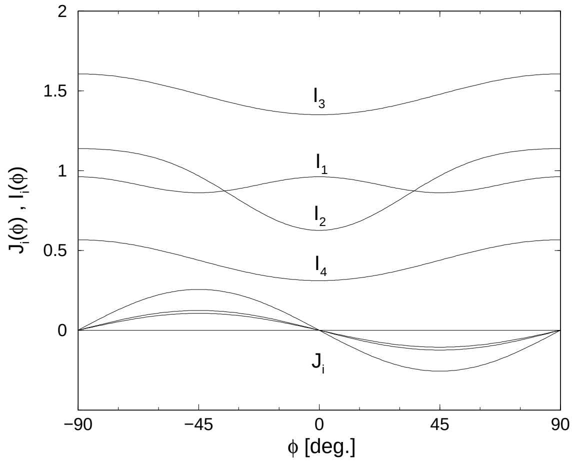

Figure 2. Angular dependence of thermal conductivities κ_{xx} (I_i) and thermal Hall coefficient κ_{xy} (J_i) according to Eq. (11) for the various 2D gap functions i=1,2,3,4 ($d_{x^2-y^2}$-, d_{xy}-, f- and f'- wave respectively).

In this way $\Delta(\mathbf{k})$ in Sr_2RuO_4 [5], $CeCoIn_5$ [6] and κ-$(ET)_2Cu(NCS)_2$ [7] are identified. On the other hand the ϕ- dependence of κ_{xx} and κ_{xy} in YBCO so far determined [30, 32, 33] do not match the one for $d_{x^2-y^2}$- wave. This is because in the above analysis we have limited ourselves for T$\ll \tilde{v}\sqrt{eH}$, while the experiments have been done for T$\gg \tilde{v}\sqrt{eH}$ [29].

4 Superconductivity in Borocarbides YNi_2B_2C and $LuNi_2B_2C$

So far we limit ourselves to the quasi- 2D systems. In 3D systems it is more expedient to consider the thermal conductivity as function of both θ and ϕ, which are the polar coordinates defining the field direction. In this respect the superconductivity in YNi_2B_2C and $LuNi_2B_2C$ is of great interest in part due to their relatively high transition temperature 15.5 K and 16.5 K resepectively [34]. Although the dominance of s-wave component in $\Delta(\mathbf{k})$ has been established by substituting Ni by a small amount of Pt and subsequent opening of the energy gap [35, 36], the superconductivity exhibits many characteristics of nodal superconductors like T^3 behaviour of T_1^{-1} and the $\sqrt{H}$- dependence of the specific heat[38]. Due to their tetragonal space group $I4/mmm$ the borocarbides have fourfold rotational symmetry within the a-b plane. The simplest $\Delta(\mathbf{k})$ which satisfies these constraints appears to be s+g-wave superconductor [8, 9]

$$\Delta(\mathbf{k}) = \frac{1}{2}\Delta(1-\sin^4\vartheta\cos(4\varphi)) \tag{12}$$

where ϑ and φ are polar coordinates describing $\mathbf{k}$. We show the quasiparticle DOS in Fig. 3. For $|E|/\Delta \ll 1$, we obtain

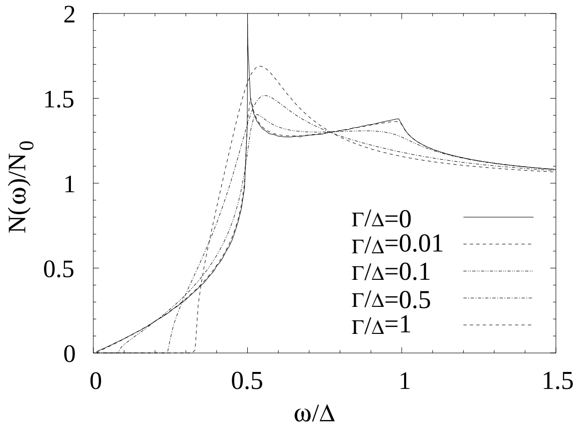

Figure 3. Normalised quasiparticle DOS for s+g wave gap in pure YNi_2B_2C (Γ/Δ=0 or x = 0) and for various doping induced nonzero Γ.

$$g(E) = \frac{N(E)}{N_0} = \frac{\pi}{4}\frac{|E|}{\Delta} \tag{13}$$

Also there are similarity and difference between d-wave superconductors as seen in [39]. In a magnetic field with orientation defined by (θ, ϕ) the quasiparticle DOS is given by [10]

$$\begin{aligned} g(0,\mathbf{H}) &= \frac{\tilde{v}\sqrt{eH}}{2\Delta} I(\theta,\phi) \\ I(\theta,\phi) &= \frac{1}{2}\big(I_s(\theta,\phi)+I_c(\theta,\phi)\big) \\ I_s(\theta,\phi) &= (1-\sin^2\theta\sin^2\phi)^{\frac{1}{2}} \\ I_c(\theta,\phi) &= (1-\sin^2\theta\cos^2\phi)^{\frac{1}{2}} \end{aligned} \tag{14}$$

Then the specific heat, the spin susceptibility and the superfluid density for T $\ll \Delta_0$ are given by

$$\begin{aligned} C_s/\gamma_s T &= g(0,\mathbf{H}) \\ \chi_s/\chi_N &= g(0,\mathbf{H}) \\ 1-\rho_s^{ab}(0,\mathbf{H})/\rho_s^{ab}(0,0) &= \frac{3}{2}g(0,\mathbf{H}) \end{aligned} \tag{15}$$

We show in Fig. 4 the ϕ dependence of I(θ,ϕ) for various polar field angles θ. In particular for $\theta = \frac{\pi}{2}$, I(θ,ϕ) develops cusps at ϕ=0 and $\frac{\pi}{2}$ etc. They are a characteristic signature of the point nodes. We note that a recent specific heat measurement on YNi_2B_2C has excactly found these cusps[40].

In order to calculate the thermal conductivity an analysis of impurity scattering is necessary. Unlike in other nodal superconductors we find the Born limit and the unitarity limit gives practically the same result. There is no resonance scattering in s+g- wave superconductors [41]. Secondly the energy gap opens up immediately in the presence of impurity scattering. The quasiparticle DOS in the presence of impurity scattering is also shown in Fig. 3 for several Γ/Δ. In the absence of magnetic field the related equations are given by

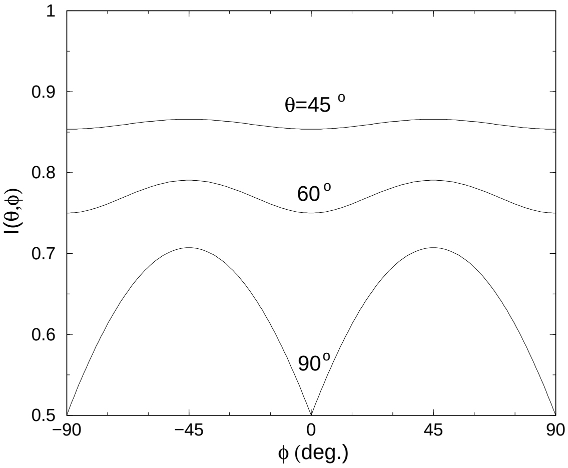

Figure 4. Angular function I(θ, ϕ) which determines $\kappa_{zz}(\theta, \phi)$ for s+g wave gap function in Eq. (18).

$$\tilde{\omega}_n = \omega_n + \Gamma \langle \frac{\tilde{\omega}_n}{\sqrt{\tilde{\omega}_n^2 + (\tilde{\Delta} - \frac{1}{2}\Delta f)^2}} \rangle \quad (16)$$

$$\tilde{\Delta} = \frac{1}{2}\Delta + \Gamma \langle \frac{(\tilde{\Delta} - \frac{1}{2}\Delta f)}{\sqrt{\tilde{\omega}_n^2 + (\tilde{\Delta} - \frac{1}{2}\Delta f)^2}} \rangle \quad (17)$$

where f=$\frac{1}{2}\sin^4\vartheta\cos(4\varphi)$. Then for $\omega \to 0$, we find $\tilde{\omega} \to 0$ and $\tilde{\Delta} \to \frac{1}{2}\Delta + \Gamma$. In other words the energy gap opens up immediately with Γ. The energy gap $\omega_g(\Gamma)$ visible in Fig. 3 is well approximated by $\omega_g(\Gamma) = \Gamma/(1 + \frac{2\Gamma}{\Delta})$.

This immediate opening of the energy gap is very different from the case of s+d- wave superconductors as discussed in [36]. This has immediate consequences: There are no nodal excitations for T $< \Gamma$. In this limit both the specific heat and the thermal conductivity decrease exponentially. Also there will be no universal heat conduction [27, 28] in sharp contrast to usual nodal superconductors. From this we expect that also the thermal conductivity in the vortex state of the s+g wave superconductor will be quite different from the case of usual nodal superconductors. It is now given by

$$\begin{aligned}
\frac{\kappa_{zz}}{\kappa_n} &= \frac{x}{\ln\left(\frac{2}{x}\right)}(1-y^2)^{\frac{3}{2}}\theta_H(1-y) \\
\frac{\kappa_{xx}}{\kappa_n} &= \frac{3}{2\ln\left(\frac{2}{x}\right)}\left(\frac{x'}{x}\right)^2(\cos^{-1}y + y'\sqrt{1-y'^2} \\
&\quad + \frac{1}{6}x(1-y'^2)^{\frac{3}{2}})\theta_H(1-y') \qquad (18) \\
x &= \frac{2}{\pi}\frac{\tilde{v}\sqrt{eH}}{\Delta}I(\theta,\phi) \qquad x' = \frac{1}{\pi}\frac{\tilde{v}\sqrt{eH}}{\Delta}I_c(\theta,\phi) \\
y &= \frac{\Gamma}{\Delta x} \qquad y' = \frac{\Gamma}{\Delta x'}
\end{aligned}$$

Here θ_H is the Heaviside step function. Furthermore κ_n refers to the normal state, $\tilde{v} = \sqrt{v_a v_c}$ and we have assumed Γ,T $\ll \tilde{v}\sqrt{eH} \ll \Delta$. In the other limit $\tilde{v}\sqrt{eH} < \Gamma$ there will be no nodal excitations. We note that first of all the angular dependence of κ_{zz} is given by I(θ, ϕ) which is fully confirmed in [8]. The details of the above expressions are different from those given previously [9, 10] due to the present more realistic treatment of the impurity scattering in the s+g wave case. On the other hand the conclusion that a gap $\omega_g \simeq \Gamma$ opens immediately for s+g wave order parameter makes it now hard to understand the H linear dependence of thermal conductivity at lowest temperatures reported in [42].

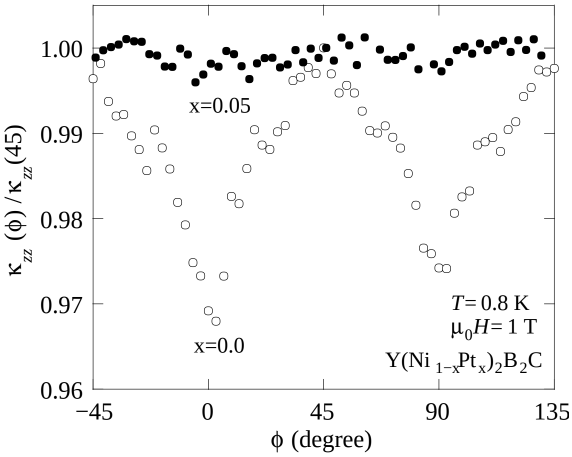

Figure 5. Cusp-like fourfold oscillations in $\kappa_{zz}(\frac{\pi}{2}, \phi)$ of YNi_2B_2C are destroyed by 5% Pt- substitution [43].

As is readily seen from Eq. (18) we predict that κ_{zz} increases like $\sqrt{H}$ while the dominant term in κ_{xx} is independent of H. Indeed the $\sqrt{H}$ dependence of κ_{zz} is consistent with the experimental data in [8]. More recently the anglular dependence of the thermal conductivity in a single crystal of $Y(Ni_{1-x}Pt_x)_2B_2C$ with x = 0.05 has been studied[43]. It is shown in Fig. 5 in comparison to the pure YNi_2B_2C (x=0). Clearly no fourfold oscillation in $\kappa(\frac{\pi}{2}, \phi)$ survives in the 5% Pt- doped compound. From T_c = 13.1 K of the 5% Pt- substituted crystal and T_c = 15.5 K for YNi_2B_2C (x=0) we can estimate Γ using

$$-\ln\left(\frac{T_c}{T_{c0}}\right) = \frac{\langle f^2 \rangle}{1 + \langle f \rangle^2}\left[\psi\left(\frac{1}{2} + \frac{\Gamma}{2\pi T_c}\right) - \psi\left(\frac{1}{2}\right)\right] \quad (19)$$

where ψ(z) is the di-gamma function, f = $\sin^4\theta\cos(4\phi)$ and $\langle f^2 \rangle$ = 0.203. The resulting Γ = 23.8 K $>$ T_c shows that we are already close to the limit l=$\hbar\tilde{v}/\Gamma \simeq \xi_0$. Clearly the large Γ has eliminated the nodal excitations. Therefore it is of great interest to study the thermal conductivity of the Pt-substituted crystals with x $<$ 0.05. It is also necessary to study theoretically the extremely dilute limit where eventually the gap induced by impurity scattering should become inhomogeneous and a crossover to a new regime should set in.

5 Skutterudite $PrOs_4Sb_{12}$

Skutterudites with rare earth atoms may exhibit Heavy Fermion behaviour and in addition magnetic, quadrupolar and superconducting phase transitions. Among them $PrOs_4Sb_{12}$ is a rather unique case [44, 45, 46]. Again the angular dependent thermal conductivity data [12, 11] suggest i) the presence of *two* phases denoted A and B and ii) a gap $\Delta(\mathbf{k})$ with planar symmetry in A while one with axial symmetry in B phase (see Fig. 6). Also, the singlet pairing in this system is very likely though not confirmed sofar. In order to describe the angular dependent thermal conductivity we have proposed [12, 11] the hybrid gap functions

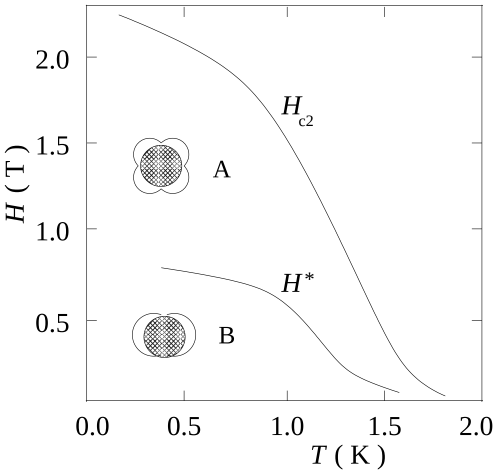

Figure 6. B-T phase diagram of $PrOs_4Sb_{12}$ with different nodal structures of A- and B- phases as obtained from $\kappa_{zz}(\theta,\phi)$ dependence [11].

$$\begin{aligned}\text{(A-phase) } \Delta(\mathbf{k}) &= \Delta(1-k_x^4-k_y^4)\\ \text{(B-phase) } \Delta(\mathbf{k}) &= \Delta(1-k_y^4)\end{aligned} \tag{20}$$

Where the A- phase again corresponds to a (tetragonal) s+g wave gap. A and B- phases have point nodes in x,y and y directions respectively. We assume here the absence of nodes along the z- direction since they have not been confirmed sofar. Their corresponding quasiparticle DOS is shown in Fig. 7 In zero field these phases exhibit characteristics of nodal superconductors like those in YNi_2B_2C . Again impurity scattering immediately opens a gap. Also as before there is no resonance scattering and universal low temperature heat conduction since it vanishes like $\exp(-\Gamma/T)$. In the presence of a magnetic field the residual quasiparticle DOS is given by ($v = v_{a,c}$)

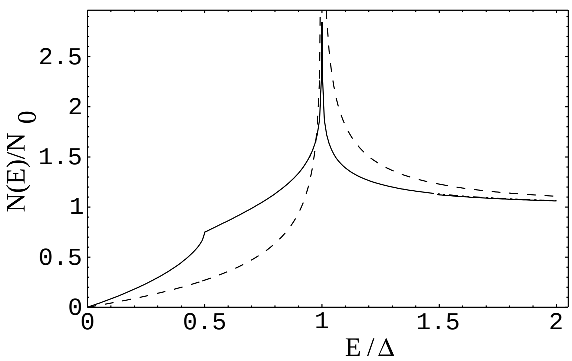

Figure 7. Quasiparticle DOS for A- phase (full line) and B- phase (dashed line) of $PrOs_4Sb_{12}$.

$$g(0,\mathbf{H}) = \frac{2}{\pi}\frac{v\sqrt{eH}}{\Delta} I_i(\theta,\phi) \tag{21}$$

whith angular functions given by

$$\begin{aligned}I_A(\theta,\phi) &= \frac{1}{2}[(1-\sin^2\theta\sin^2\phi)^{\frac{1}{2}} + (1-\sin^2\theta\cos^2\phi)^{\frac{1}{2}}]\\ I_B(\theta,\phi) &= \frac{1}{4}(1-\sin^2\theta\sin^2\phi)^{\frac{1}{2}}\end{aligned} \tag{22}$$

We note that $I_A(\theta,\phi) \equiv I(\theta,\phi)$ given in Eq. (14). Despite the different functional form of $|\Delta(\mathbf{k})|$ these s+g wave sc phases have the same quasiparticle DOS. As discussed in Sect.(4) this expression is valid when $v\sqrt{eH} \gg \Gamma$. Also the effect of a small Γ may be incorporated by changing $v\sqrt{eH}I_i(\theta,\phi) \rightarrow v\sqrt{eH}I_i(\theta,\phi)$-$\Gamma$. The specific heat etc. are expressed in terms of g(0,**H**) as before. Like the residual quasiparticle DOS the thermal conductivity $\kappa_{zz}(\theta,\phi)$ for the A- phase is given by the same expression Eq. (18) as for the YNi_2B_2C s+g- wave gap with $I(\theta,\phi) \rightarrow I_i(\theta,\phi)$ (i=A,B). Its angular dependence is completely determined by $I_i(\theta,\phi)$ and agrees with the experimentally observed fourfold and twofold oscillations observed in the A- and B- phases respectively in $PrOs_4Sb_{12}$ below T = 0.5 K. However experimental results at lower temperatures are highly desirable. At this preliminary stage the microscopic origin of the symmetry breaking into planar and axial symmetry seen in A and B- phases is not yet clear.

6 Concluding remarks

First we have reviewed our earlier work on the magnetothermal conductivity in quasi 2D nodal superconductors. These superconductors are described in the BCS context by Cooper pairs having non- zero angular momomentum. The effect of a magnetic field is incorporated within the quasiclassical approximation in the vortex phase as first done by Volovik[2]. In nodal superconductors it represents a simple and accurate description of quasiparticle excitations. The associated magnetothermal conductivity provides a unique window to investigate nodal structures of $\Delta(\mathbf{k})$. In this way Izawa et al have succeeded in identifying the gap symmetry of Sr_2RuO_4 , $CeCoIn_5$ and κ- $(ET)_2Cu(NCS)_2$ [5, 6, 7]. More recently the magnetothermal conductivity data from YNi_2B_2C and $PrOs_4Sb_{12}$ has revealed the presence of point nodes in $\Delta(\mathbf{k})$ [8, 11]. These order parameters are described in terms of s+g wave gap functions[9, 10, 12]. This is the first time such hybrid gap functions consisting of a superposition of representations have been found. In addition, due to the presence of an s- wave component in $\Delta(\mathbf{k})$, the effect of impurities is very different[41] from that in usual nodal superconductors whose gap functions have sign change and belong to a single nontrivial representation. For example there is no resonance scattering due to nonmagnetic impurities. Also the energy gap opens up immediately due to impurity scattering[41] which is completely different from the p- or d- wave superconductors. Therefore the hybrid nodal superconductors YNi_2B_2C and $PrOs_4Sb_{12}$ appear to open completely new vista in the rich field of unconventional superconductors.

Acknowledgement:
We thank Konomi Kamata, Koichi Izawa and Yuji Matsuda for useful discussions, especially on ongoing experiments on YNi_2B_2C and $PrOs_4Sb_{12}$. We thank also K. Kamata to allow us to use her unpublished data of the single crystal $Y(Ni_{1-x}Pt_x)_2B_2C$ with x = 0.05.

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