Electronic Raman Spectra of Superconducting Borocarbides

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Abstract. – Recently we have proposed a $s + g$-wave model for the superconductivity in borocarbides, YNi$_2$B$_2$C and LuNi$_2$B$_2$C [1, 2, 3]. In the present paper we first summarize thermodynamic properties of $s + g$-wave model. Then we shall analyse the recent Raman spectra data of RNi$_2$B$_2$C (with R=Lu and Y) by Yang et al. [4]. The present model appears to describe salient features of the Raman spectra.

Introduction. – The Superconductivity in rare earth borocarbides is of great interest [5,6]. In particular its interplay with magnetism and superconductivity is very fascinating [7]. However in the following we limit ourselves to superconducting borocarbides LuNi$_2$B$_2$C and YNi$_2$B$_2$C. They have relatively high superconducting transition temperature $T_c = 16.5$ K and 15.5 K, respectively. Although the dominance of $s$-wave component in $\Delta(k)$ has been established by substituting Ni by Pt and subsequent opening of the energy gap [8, 9], the superconductivity exhibits a number of peculiarities common to nodal superconductors [10,11]. For example both the $\sqrt{H}$ dependence of the specific heat and the $H$ linear dependence of the thermal conductivity indicate that the superconductivity has the nodal excitations [12,13,14,15,16,17,18] similar to $d$-wave superconductors in high $T_c$ cuprate superconductors. Further the presence of de Haas van Alphen oscillation in the vortex state of LuNi$_2$B$_2$C down to $H = 0.2H_c$ indicates again the nodal superconductors [19]. In conventional $s$-wave superconductor de Haas van Alphen oscillation would disappear for $H < 0.8H_c$ [20]. Further the upper critical field determined for LuNi$_2$B$_2$C and YNi$_2$B$_2$C [21] in a magnetic field within the a-b plane exhibits clear fourfold symmetry reminiscent to $d$-wave superconductors [22].

These experiments indicate clearly $\Delta(k)$ in borocarbides has to have an anisotropic $s$-wave order parameter. Further a) $\Delta(k)$ has to have the nodal structure or the quasiparticle density of states, $N(E) \sim |E|$ for $|E| \ll \Delta$ where $\Delta$ is the superconducting order parameter (i.e. the maximum of $\Delta(k)$), which gives both the $\sqrt{H}$dependence of the specific heat and the $H$
linear thermal conductivity in the vortex state. b) the nodal structure has to have the fourfold symmetry within the $a$-$b$ plane and to be consistent with the tetragonal symmetry. These two constraints appear to suggest almost uniquely $s+g$-wave superconductors. Here $\theta$ and $\phi$ are the polar and azimuthal angles describing $\mathbf{k}$. Contrary to Ref. [1] we have minus(-) sign in front of the $g$-wave term. This corresponds to point nodes at [100], [010], etc. Those positions of the nodal points are consistent with the magnetothermal conductivity data [2]. More recently the point node at the same positions have been seen by the magnetospecific heat measurement in the vortex state of YNi$_2$B$_2$C by Park et al. [23]. We shall see later the Raman spectra data from both YNi$_2$B$_2$C and LuNi$_2$B$_2$C are consistent with Eq.(1). Recently the above Raman data have been analysed in terms of 2D $s+g$-wave model by Lee and Choi [24]. Unlike the present model their model has line nodes. Therefore their model cannot describe the magnetothermal conductivity data [2]. Further the description of the Raman spectra within this model is unsatisfactory.
Thermodynamics. — First we determine the temperature dependent energy gap $\Delta(T)$ within the weak coupling theory. Here $\Delta(T)$ is the maximum value of the energy gap $\Delta(k)$.

$$\lambda^{-1} = \langle f^2 \rangle^{-1} \int_0^{E_c} dE \left( \frac{f^2}{\sqrt{E^2 - \Delta^2 f^2}} \right) \tanh(\frac{E}{2T})$$

(2)

where $f = \frac{1}{2}(1 - \sin^4 \theta \cos(4\phi))$, $\lambda$ and $E_c$ are the dimensionless coupling constant and the cut-off energy ($\gg \Delta$), respectively. Further $\langle \ldots \rangle = \int d\Omega/4\pi$.

In the vicinity of $T = T_c$, Eq.(2) gives

$$\Delta^2(T) \simeq \frac{2(2\pi T)^2}{7\zeta(3)} \langle f^2 \rangle^{-1} \langle -\ln \frac{T}{T_c} \rangle$$

(3)

where $T_c = \frac{2\gamma}{\pi} E_c e^{-1/\lambda}$ and $\gamma = 1.78107 \ldots$ the Euler constant. On the other hand for $T \ll \Delta_0$

$$-\ln \left( \frac{\Delta(T)}{\Delta_0} \right) = \langle f^2 \rangle^{-1} \left\{ \frac{3\pi}{16} \zeta(3) \left( \frac{T}{\Delta} \right)^3 + \frac{7\pi^4}{100} \left( \frac{T}{\Delta} \right)^4 + \ldots \right\}$$

(4)

$$= 1.9635 \left( \frac{T}{\Delta} \right)^3 + 14.2 \left( \frac{T}{\Delta} \right)^4 \ldots$$

(5)

with $\Delta_0 = \Delta(0) = 2\gamma T_c \exp[\langle f^2 \rangle^{-1} \langle -\ln |f| \rangle] \simeq 2.76 T_c$

(6)

Then the specific heat $C_s$ is given by

$$C_s = T^{-2} N_0 \int_0^\infty d\xi < \text{sech}^2 \left( \frac{E}{2T} \right) (E^2 - \frac{T}{2} \frac{d\Delta^2}{dT} f^2) >$$

(7)

$$\simeq \frac{2\pi^2}{3} N_0 \left\{ \frac{27}{4\pi} \zeta(3) \left( \frac{T}{\Delta} \right) + \frac{63}{80} \left( \frac{T}{\Delta} \right)^2 + \ldots \right\}$$

(8)

where $E = \sqrt{\xi^2 + \Delta^2 f^2}$ and $N_0$ is the density of states in the normal state. In Fig.1 we show $C_s/(\frac{2\pi^2}{3} N_0 T)$ versus $T/T_c$ for $s + g$-wave, $s$-wave and $d$-wave superconductors. As is readily seen the specific heat of $s + g$-wave superconductor is very similar to the one in $d$-wave superconductors [25]. Also the spin susceptibility of $s + g$-wave superconductor is shown in Fig.2, which is very similar to the one in $d$-wave superconductors.

The superfluid density, on the other hand, has the axial symmetry. The superfluid density in the $a$-$b$ plane is given by

$$\rho_{s,ab}(T) = 1 - \frac{3}{4T} \int_0^\infty d\xi < \sin^2 \theta \ \text{sech}^2 \left( \frac{E}{2T} \right) >$$

(9)

$$\simeq 1 - \frac{3\pi}{4} (\ln 2) \left( \frac{T}{\Delta} \right) - \frac{5\pi^2}{32} \left( \frac{T}{\Delta} \right)^2 + \ldots$$

(10)

while the superfluid density parallel to the $c$-axis is given by

$$\rho_{s,c}(T) = 1 - \frac{3}{2T} \int_0^\infty d\xi < \cos^2 \theta \ \text{sech}^2 \left( \frac{E}{2T} \right) >$$

(11)

$$\simeq 1 - \frac{\pi^2}{4} \left( \frac{T}{\Delta} \right)^2 - \frac{783\pi}{256} \left( \frac{T}{\Delta} \right)^3 + \ldots$$

(12)
Fig. 3 – the superfluid density in the a-b plane for s + g- and d-wave superconductors.

Fig. 4 – the superfluid density parallel to the c-axis for for s + g- and d-wave superconductors.

In Fig.3 and Fig.4 we show the superfluid density in the a-b plane and parallel to the c-axis, respectively. The superfluid density in the a-b plane is very similar to the one in d-wave superconductor. Further the superfluid density parallel to the c-axis is somewhat similar to the one in the d-wave superconductor which is due the coherent Josephson tunneling [26]. We note also $T^{-1} \sim T^3$ behavior has been already observed in Ref. [27].

Further the thermal conductivity within the a-b plane exhibits the universal heat conduction [28, 29]

$$\kappa(T)/\kappa_n = \frac{\pi^2}{8} \frac{n}{\Delta_0 m}$$

in the limit $T \to 0$ K. Also the thermal conductivity for $\mathbf{H} \parallel \mathbf{c}$ gives

$$\kappa(H)/\kappa_n = \frac{3}{\pi} \frac{v_a^2 (eH)}{\Delta^2}$$

for $T, \sqrt{\Gamma/\Delta} \ll v_a \sqrt{eH}$, where $\Gamma$ and $v_a$ are the electron scattering rate and the Fermi velocity within the a-b plane, respectively. $\kappa_n$ is the one in the normal state and $v_a$ is the Fermi velocity within the a-b plane. Indeed the $H$-linear thermal conductivity is observed recently by Boaknin et al. [15].

Also very recently the specific heat of the vortex state in YNi$_2$B$_2$C in a magnetic field within the a-b plane is observed by Park et al. [23]. It exhibits cusps at $\mathbf{H} \parallel \mathbf{a}$ and $\mathbf{H} \parallel \mathbf{b}$ typical to the point nodes in $s + g$-wave superconductors.

**Electronic Raman Spectra.** – We consider the case the polarization vector of photons lie in the a-b plane. Then the Raman spectra are given by [30, 31]

$$S_i(\omega/2\Delta) = \text{Im} \left[ \frac{< \gamma_i \lambda >}{< \lambda >} - \frac{< \gamma_i \lambda >^2}{< \lambda >} \right]$$

where $\gamma_{A1g} = \sqrt{2} \cos(4\phi_k)$, $\gamma_{B1g} = \sqrt{2} \cos(2\phi_k)$ and $\gamma_{B2g} = \sqrt{2} \sin(2\phi_k)$ and

$$\lambda = \lambda' + i\lambda''$$
Fig. 5 – The theoretical Raman spectra in $A_{1g}$, $B_{1g}$ and $B_{2g}$ modes using $s + g$ model at $T = 0$ K (left panel) and the experimental Raman spectra for YNi$_2$B$_2$C taken at $T = 6$ K (right panel) are shown.

\[
\lambda' = \frac{f^2}{x \sqrt{f^2 - x^2}} \tan^{-1} \left( \frac{x}{\sqrt{f^2 - x^2}} \right) \theta(f^2 - x^2) \\
- \frac{f^2}{x \sqrt{x^2 - f^2}} \tan^{-1} \left( \frac{\sqrt{x^2 - f^2}}{x} \right) \theta(x^2 - f^2) \\
\lambda'' = \frac{\pi}{2x} \frac{f^2}{\sqrt{x^2 - f^2}} \theta(x^2 - f^2)
\]  

\[(17)\]

\[(18)\]

where $x = \omega/2\Delta$ and $f = \Delta(k)/\Delta$ and $\langle \ldots \rangle$ means the $k$(angle) average for the Fermi surface. $\theta(x)$ is the step function, i.e., $\theta(x) = 1$ for $x > 0$ and $\theta(x) = 0$ for $x < 0$. We note for the present model the second term in Eq.(10) does not contribute for $B_{1g}$ and $B_{2g}$ modes due to the symmetry constraints. The electronic Raman spectra from these 3 modes are obtained numerically and shown in Fig.3 (the left-side panel). In parallel to the theoretical result we show the experimental data for YNi$_2$B$_2$C taken at $T = 6$ K.

We note $A_{1g}$ mode is very consistent with the observed spectra. Also the low frequency parts ($\omega \leq \Delta(T)$) of both the $B_{1g}$ mode and the $B_{2g}$ mode are very consistent. On the other hand the theoretical curve for the $B_{2g}$ mode exhibits a cusp at $\omega = 2\Delta(T)$, which is not seen experimentally. We don’t know if the cusp-like feature will disappear with increasing temperature or not. Also the peak position of the $B_{1g}$ mode is somewhat in the higher energy
than that of the $B_{2g}$ mode. Again we don’t know if this is the effect of the temperature. In any event we may conclude that $s + g$-wave model captures the main feature of the Raman spectra of YNi$_2$B$_2$C. Also the peak position in the $B_{2g}$ mode gives $\Delta(T)$.

The weak coupling theory gives $\Delta_0$ for YNi$_2$B$_2$C ($T_c \simeq 15.3$ K) and LuNi$_2$B$_2$C ($T_c \simeq 15.7$ K) is 42.2 K(3.64 meV) and 43.3 K(3.73 meV), respectively. On the other hand the data at $T/T_c \simeq 1/2$ indicate $\Delta_0 = 50.4$ K and 64.7 K for YNi$_2$B$_2$C and LuNi$_2$B$_2$C, respectively. Therefore we may conclude that the borocarbides superconductors are in the intermediate coupling region. However, clearly a further experiment at low temperatures is highly desirable.

Summary. – We have analysed further the $s + g$-wave superconductors proposed in [12]. This model appears to describe the recent specific heat data [23] as well. We have also studied the Raman spectra reported in Ref. [4]. The present model appears to capture the main feature of the observed spectra. This further confirms the presence of the order parameter $\Delta(k)$ with point nodes at $k = (100), (010), (001)$, and $(010)$.

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