

## Anisotropy of the Superconducting Gap of the Borocarbide Superconductor $\text{YNi}_2\text{B}_2\text{C}$ with Ultrasonic Attenuation

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The ultrasonic attenuation  $\alpha$  of the highly anisotropic  $s$ -wave superconductor  $\text{YNi}_2\text{B}_2\text{C}$  has been measured for all the symmetrically independent elastic modes to explore the location of the zero superconducting gap region on the Fermi surface. The attenuation of the longitudinal mode shows a pronounced anisotropy in the superconducting state: While  $\alpha$  shows a thermally activated behavior along  $[110]$  and  $[001]$  directions, it shows  $T$ -linear dependence along  $[100]$ . These results together with those for the transverse modes demonstrate the presence of point nodes or zero-gap regions along  $[100]$  and  $[010]$  directions. This is a clear demonstration of ultrasonic attenuation as a powerful probe for the structure of the anisotropic superconducting gap.

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The borocarbide superconductors  $\text{YNi}_2\text{B}_2\text{C}$  and  $\text{LuNi}_2\text{B}_2\text{C}$  have been attracting renewed interest because of the growing evidence for a highly anisotropic superconducting gap. The presence of practically a zero gap has been demonstrated from a number of experiments, including specific heat [1,2], photoemission spectroscopy [3], nuclear relaxation rate  $1/T_1$  [4,5], thermal conductivity [6], microwave impedance [7], and Raman scattering [8]. Usually, a zero gap has been taken as evidence for  $p$ -wave or  $d$ -wave superconductivity where the sign change of the order parameter inherently gives rise to gap nodes. There is a consensus, however, that superconductivity in borocarbides is predominantly of  $s$ -wave character, as expected from strong electron-phonon interactions [9–11]. The most convincing evidence is the robustness of superconductivity against nonmagnetic impurities. Even in the dirty limit, superconductivity can survive [2]. This controversy has been raising one of the most challenging issues in borocarbide physics: How does the zero gap emerge in such a predominantly  $s$ -wave pairing state?

To address this issue, it is critically important to identify the position of the zero gap on the Fermi surface. Ultrasonic attenuation  $\alpha(T)$ , with its directional property, can be a powerful technique to determine the zero-gap position. It had been, however, suffering from the lack of a theoretical basis, because theories, except for a recent one, assume an extremely long mean free path,  $ql \gg 1$ . Here,  $q$  is the wave number of sound and  $l$  is the electron mean free path. This, in fact, is unrealistic in most systems. Recently, a theory was developed to analyze  $\alpha$  for anisotropic superconductors under experimentally accessible conditions,  $ql \ll 1$ . This enabled us to explore the position of zero gap using ultrasonic attenuation. Figures 1(a) and 1(b) show directional distribution of quasiparticles that couple with longitudinal and trans-

verse ultrasonic waves, respectively [12–15]. A longitudinal wave couples dominantly with the quasiparticles having momentum parallel to sound propagation direction  $q$ , while a transverse wave couples with quasiparticles having momentum between propagation  $q$  and polarization  $u$  directions. By measuring sound attenuation for all of the crystallographically independent elastic modes, the  $k$ -selective couplings should allow us to identify the zero-gap position where thermally activated quasiparticles remain even at low temperatures.

Another technique to explore the location of gap node is the magnetic field orientation dependence of thermal conductivity  $\kappa(H, \theta)$ . This has been successfully applied to exotic superconductors such as  $\text{Sr}_2\text{RuO}_4$  [16], organic superconductor [17], and  $\text{CeCoIn}_5$  [18]. For  $\text{YNi}_2\text{B}_2\text{C}$ , Izawa *et al.* have suggested that point nodes exist along  $[100]$  and  $[010]$  directions from  $\kappa(H, \theta)$  measurements [19]. The thermal conductivity and the ultrasonic attenuation techniques were applied to the same system,  $\text{Sr}_2\text{RuO}_4$  but, indeed, had given contradictory conclusions. While the thermal conductivity  $\kappa(H, \theta)$  [16] indicated the presence of horizontal line nodes,  $\alpha(T)$  for the

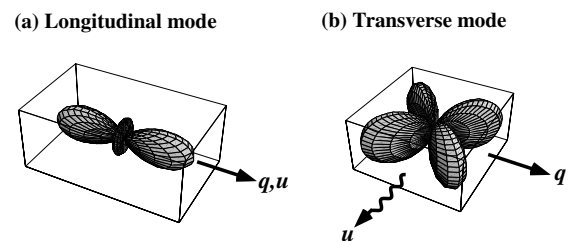


FIG. 1. Directional distribution of the quasiparticles on the spherical Fermi surface, which couple strongly with (a) longitudinal sound propagating along  $q$ , and (b) transverse sound propagating along  $q$  with polarization  $u$  in the  $ql \ll 1$  limit [12–15].

in-plane elastic modes implied vertical line nodes [20]. We believe that it is because of the lack of data on the out-of-plane elastic mode  $C_{33}$  in the ultrasonic measurement, mainly because of the size limitation of  $\text{Sr}_2\text{RuO}_4$  crystals along the  $c$  axis. This made it rather difficult to distinguish the horizontal and the vertical nodes. The thermal conductivity, on the other hand, has a drawback in that magnetic field has to be applied to probe quasiparticle states, which may modify the symmetry of gap. It is tempting to investigate whether gap structure can be determined consistently between ultrasonic attenuation and thermal conductivity measurements, by measuring  $\alpha(T)$  for all the symmetrically independent elastic modes. The availability of high quality single crystals with large enough dimensions in borocarbides provides a unique opportunity to establish firmly the validity of the two techniques, particularly the ultrasonic attenuation as a probe for the location of zero superconducting gap. In this Letter, we demonstrate that ultrasonic attenuation is, indeed, quite powerful to determine the zero-gap position for anisotropic superconductors, and establish that there exist point nodes along [100] and [010] for  $\text{YNi}_2\text{B}_2\text{C}$  inconsistent with previous thermal conductivity measurement.

A large single crystal of  $\text{YNi}_2\text{B}_2\text{C}$  was grown by the floating zone method [21]. The dimensions of grown single crystalline rod were about 60 mm long, and about 3.5 mm diameter. The crystal was compositionally homogeneous without any trace of secondary phase in the powder X-ray diffraction pattern. The superconducting transition temperature of  $T_c = 15.1$  K with a transition width of less than 0.5 K was magnetically determined. The residual resistivity ratio of 37 indicates that the sample is in the clean limit [2]. For the ultrasonic measurements, the crystal was cut into parallelepipeds with (100) and (001) planes, and with (110) and (001) planes. The dimensions of the sample were approximately 3.7, 4.6, and 3.1 mm along the [100], [110], and [001] axes, respectively, which was large enough to measure all of the independent elastic modes summarized in Table I. L and T denote longitudinal and transverse modes, respectively, and the indices denote the propagation direction. Ultrasonic attenuation was measured by a pulse-echo method. Ultrasound in the frequency range between 40 and 120 MHz was generated and detected by  $\text{LiNbO}_3$

TABLE I. Elastic modes for  $\text{YNi}_2\text{B}_2\text{C}$  and the corresponding sound propagation direction  $q$ , polarization  $u$ , and symmetry.

Mode	$q$	$u$	Symmetry
L100	[100]	[100]	$A_{1g} + B_{1g}$
L110	[110]	[110]	$A_{1g} + B_{2g}$
L001	[001]	[001]	$A_{1g}$
T100	[100]	[010]	$B_{2g}$
T110	[110]	[110]	$B_{1g}$
T001	[001]	[110]	$E_g$

transducers glued onto parallel surfaces of sample. The measurements were performed in a temperature range 2–20 K without magnetic field and under magnetic field of 9 T, which is higher than the upper critical fields of  $H_{c2}(2\text{ K}, H \parallel c) = 6.5$  T and  $H_{c2}(2\text{ K}, H \perp c) = 8.5$  T.

Temperature dependences of  $\alpha(T)$  for longitudinal and transverse modes are shown in Figs. 2 and 3, respectively. Since the phonon contribution to  $\alpha(T)$  is small in this temperature range, this can be attributed to the normal electron contribution. In the normal state,  $\alpha(T)$  shows a gradual increase on cooling. In contrast,  $\alpha(T)$  exhibits a rapid decrease in the superconducting state below  $T_c = 15.1$  K, which is due to the opening of gap and the resultant decrease of quasiparticle densities. The magnitude  $\alpha(T)$  increases rapidly with the increase of frequency. We found that  $\alpha(T)$  shows quadratic behavior for both in the normal and the superconducting states [22]. For the L100 mode,  $\alpha(T)$  data with a frequency of only 43.4 MHz is shown since large attenuation of this mode prevented accurate measurements for higher frequencies.

For longitudinal modes, a pronounced anisotropy of  $\alpha(T)$  can be seen in the superconducting state.  $\alpha_S(T)$  for L110 and L001 shows a rapid decrease below  $T_c$  and is almost zero below about  $0.3T_c$  ( $\sim 5$  K), which is analogous to those of fully gapped superconductors. In marked

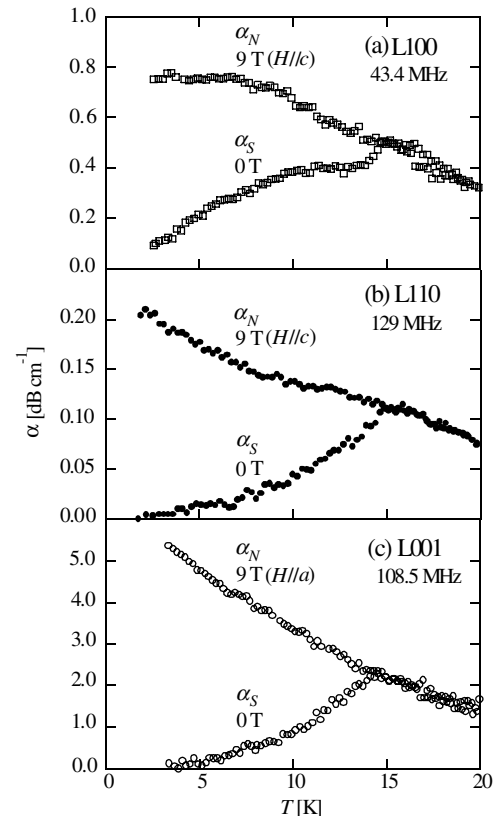


FIG. 2. Temperature dependence of ultrasonic attenuation  $\alpha(T)$  of longitudinal modes in the superconducting and normal states of  $\text{YNi}_2\text{B}_2\text{C}$ .

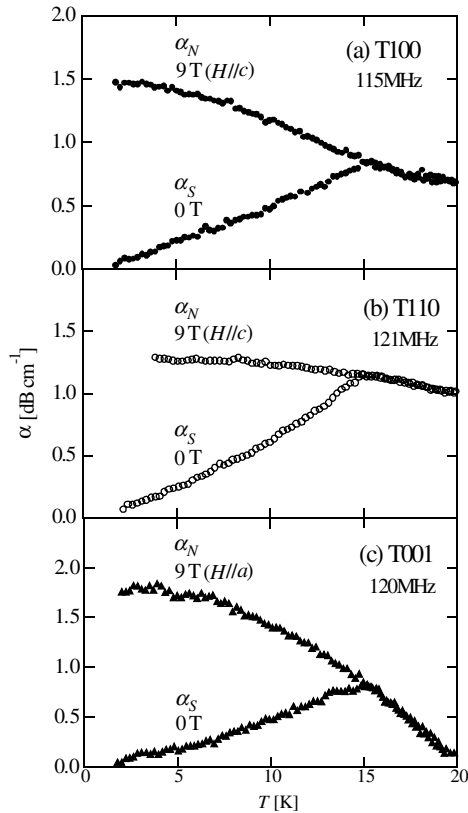


FIG. 3. Temperature dependence of ultrasonic attenuation  $\alpha(T)$  of transverse modes in the superconducting and normal states of  $\text{YNi}_2\text{B}_2\text{C}$ .

contrast,  $\alpha_S(T)$  for L100 shows a rather gradual decrease to the lowest temperature. The anisotropy of transverse modes is less pronounced than those of longitudinal modes. To inspect detailed temperature dependence in the superconducting state, we have measured  $\alpha_N(T)$  in the normal state below zero field  $T_c$  by applying a magnetic field of 9 T ( $> H_{c2}$ ) and have normalized  $\alpha_S(T)$  in the superconducting state by  $\alpha_N(T)$ , as shown in Fig. 4. Temperature dependence of  $\alpha_S/\alpha_N$  for L110 and L001 is, indeed, close to the exponential temperature dependence expected for fully gapped BCS superconductors, indicated by the solid line. It is clear, on the other hand, that  $\alpha_S/\alpha_N$  for L100 shows almost  $T$ -linear behavior down to the lowest temperature and is, therefore, distinct from the BCS curve, indicating the presence of almost gapless quasiparticle excitations.

The observed anisotropy can now be compared with the theoretical results shown in Fig. 1 [22], which, indeed, allows us to identify the locations of the zero-gap positions or nodes. For longitudinal modes,  $T$ -linear  $\alpha_S/\alpha_N$ , observed only for L100, indicates that low-lying quasiparticle excitations are localized along the [100] direction rather than the [110] and [001] directions, namely, along [100] and crystallographically equivalent [010]. The results for transverse modes are consistent with the implication from the longitudinal modes. Among the

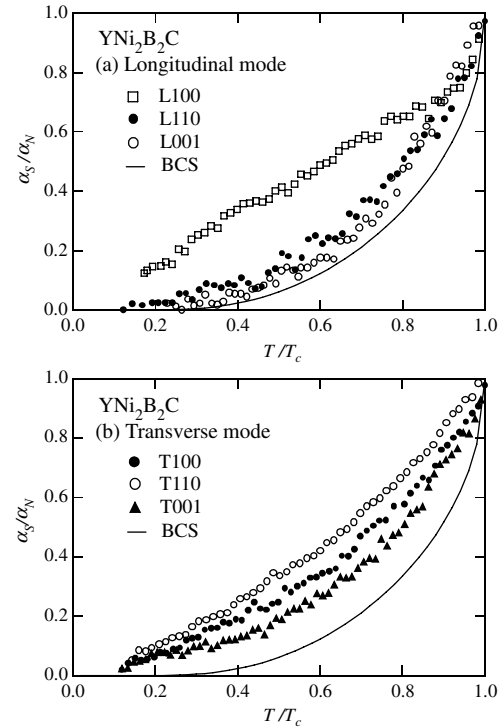


FIG. 4. Ultrasonic attenuation divided by normal state attenuation  $\alpha_S/\alpha_N$  as a function of reduced temperature  $T/T_c$  for (a) longitudinal modes and (b) transverse modes. The solid lines represent  $\alpha_S/\alpha_N$  for the BCS model.

three transverse modes, the most gradual decay of  $\alpha_S/\alpha_N$  on cooling is observed for T110. T110 probes quasiparticles along [100] and [010] (see Fig. 1) and therefore implies the presence of smaller gap along [100] and [010]. T001 mode probes quasiparticles along [111] as well as [101]. Note that T001 is rotationally symmetric around the [001] axis for the tetragonal crystal and that  $u \parallel [110]$  is equivalent to any  $u \parallel ab$  planes. The more pronounced decay of  $\alpha_S/\alpha_N$  for T001 than T100 (and T110) likely indicates that the low-lying quasiparticles are well localized within the  $ab$  planes, and rules out a vertical node like  $k_x, k_y$ . From these considerations, we conclude that a zero-gap region is present along the [100] and [010] directions in  $\text{YNi}_2\text{B}_2\text{C}$ .

The anisotropy of the transverse mode is not as significant as those of longitudinal modes. We ascribe this to the nonspherical Fermi surface topology of  $\text{YNi}_2\text{B}_2\text{C}$ , which modifies the directional distribution of quasiparticle velocities probed by an ultrasonic wave from those expected for the spherical Fermi surface. These effects should be pronounced for the transverse modes because of the perpendicular geometry of  $q$  and  $u$ . For further analysis on a quantitative level, the difference of quasiparticle phase velocity and group velocity should be invoked [12–15].

The conclusion drawn here agrees well with that implied from a thermal conductivity measurement [19,23]. This firmly establishes the anisotropic gap structure of  $\text{YNi}_2\text{B}_2\text{C}$  and also ultrasonic attenuation as a powerful

probe for the location of nodes in anisotropic superconductors, when the  $\alpha(T)$  for all crystallographically independent elastic modes are measured. It is true that the size and quality of crystals available often limits the measurements of all the crystallographically independent elastic modes. Nevertheless, ultrasonic attenuation has a substantial advantage against thermal conductivity  $\kappa(H, \theta)$ . The magnetic field in the ultrasonic attenuation  $\alpha(T)$  is used simply to normalize the quasiparticle contribution and does not play any fundamental role. The gap structure can be determined without magnetic fields, which will help to determine the complex  $H$ - $T$  phase diagram of superconductors with multiple degrees of freedom, in particular, at low fields.

The observation of zero gap in  $\text{YNi}_2\text{B}_2\text{C}$  has been casting a problem deep in physics, since it is incompatible with  $s$ -wave superconductivity in its simplest form. There exists a gap anisotropy even in conventional superconductors because of the momentum dependence of pair potential  $V_{kk'}$ . However, this effect usually is not very significant, and, indeed, the observed gap anisotropy in conventional  $s$ -wave superconductor Sn, for example, is at most 25% [24]. The presence of zero gap for  $\text{YNi}_2\text{B}_2\text{C}$  indicates a substantial mixture of the higher order term to the  $s$ -wave order parameter. Maki *et al.* proposed that the gap symmetry of  $\text{YNi}_2\text{B}_2\text{C}$  can be described by a mixture of  $s$  waves and  $g$  waves [25], which is in reasonable agreement with a number of experimental results including the present result. They argue that the origin of  $g$ -wave order-parameter mixing is a Fermi surface nesting with a nesting vector along the [100] direction. The nesting had been experimentally confirmed by electron positron annihilation measurements for  $\text{YNi}_2\text{B}_2\text{C}$  and  $\text{LuNi}_2\text{B}_2\text{C}$  [26]. The presence of Fermi surface nesting leads to a charge-density-wave instability, which generally interferes destructively with phonon mediated superconductivity [27]. A substantial reduction of pair potential is expected on the nested Fermi surfaces, which may give rise to a zero-gap structure along the [100] direction. It is worth noting that correlation between the gap anisotropy and the nesting is also suggested in the  $s$ -wave superconductor Pb, in which large gap anisotropy of  $\Delta_{\text{max}}/\Delta_{\text{min}} > 10$  has been observed by phonon imaging experiment [28].

In summary, with the recent progress in the theoretical treatment of the classical limit  $ql \ll 1$ , we were able to demonstrate ultrasonic attenuation as a practical probe for the location of nodes in anisotropic superconductors. The measurements of ultrasonic attenuation for all the symmetrically independent elastic modes, which are crucial to deduce the nodal structure, have been successfully conducted in  $\text{YNi}_2\text{B}_2\text{C}$ . We found a pronounced anisotropy of the temperature dependent attenuation in the superconducting state, which can be interpreted consistently as the presence of zero superconducting gap or point nodes along [100] and [010]. The [100] nodes appear to suggest an intimate link between the Fermi

surface nesting and the zero gap, which warrants for further exploration.

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