Evidence for the Coexistence of Anisotropic Superconducting Gap and Nonlocal Effects in the Non-magnetic Superconductor LuNi$_2$B$_2$C

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A study of the dependence of the heat capacity $C_p(\alpha)$ on field angle in LuNi$_2$B$_2$C reveals an anomalously disorder effect. For pure samples, $C_p(\alpha)$ exhibits a fourfold variation as the field $H < H_{c2}$ is rotated in the [001] plane, with minima along $< 100 > (\alpha = 0)$. A slightly disordered sample, however, develops anomalous secondary minima along $< 110 >$ for $\mu_0H > 1$ T, leading to an 8-fold pattern at 2 K and 1.5 T. The anomalous pattern is discussed in terms of coexisting superconducting gap anisotropy and non-local effects.

Exotic superconductors, defined as those that follow the Uemura relation 1 between the superconducting transition temperature $T_c$ and the magnetic penetration depth $\lambda$, $T_c \propto \lambda^{-2}$, have physical properties that differ from conventional (BCS) superconductors. High-$T_c$ cuprates, bismuthates, Chevrel-phases, organic, and heavy-fermion superconductors were suggested to constitute this class. For most of the members, the superconducting gap function is highly anisotropic or has gap zeros on the Fermi surface 2. The rare-earth nickel borocarbides RNi$_2$B$_2$C (R=Y, Lu, Tm, Er, Ho, and Dy) remain a challenge because they share many features in common with the exotic superconductors but, like elemental BCS superconductors, fall below the Uemura trend 3.

Unlike BCS superconductors, which exhibit exponential temperature dependence in density-of-states (DOS)-dependent quantities below $T_c$, the borocarbides have power-law temperature dependences in their low temperature specific heat $\gamma$, NMR $1/T_1$ 4, and thermal conductivity $\kappa$, indicating that electronic excitations persist even well below $T_c$. Recently, compelling evidence for the presence of nodes along $< 100 >$ directions has been reported both from field-angle thermal conductivity $\kappa$ and field-angle heat capacity measurements 5 of YNi$_2$B$_2$C.

Another interesting feature of the borocarbides is that the flux-line lattice (FLL) undergoes a field-driven transition from hexagonal to square with increasing magnetic field $H \parallel [001]$ 4, 9. Kogan et al. successfully incorporated nonlocal corrections to the London model and Fermi surface anisotropy in these materials 12 to describe the transition within a BCS scheme, i.e., isotropic $s$-wave gap with electron-phonon coupling. Nonlocal effects were also used to explain, without resorting to any exotic order parameter, the modulation of the upper critical field $H_{c2}(\alpha)$ 13 and magnetization $M(\alpha)$ 14 with the angle $\alpha$ between the magnetic field and the crystal axes. The two seemingly irreconcilable viewpoints have only increased the confusion about the nature of the order parameter of the borocarbides. Recently, Nakai et al. considered the coexistence of an anisotropic gap and nonlocal effects in the borocarbides 15 and thereby explained the reentrant FLL transformation 16 in terms of the interplay between the two effects. In this report, we present further evidence for the coexistence of the gap anisotropy and nonlocal effects. In clean systems, the gap anisotropy effects dominate while both effects are comparable in less clean samples.

Single crystals of LuNi$_2$B$_2$C were grown in a Ni$_2$B flux as described elsewhere 17. Samples A and C were not annealed while sample N was annealed at $T = 1000$ C for 100 hours under high vacuum. Typical sample dimensions were $1 \times 1 \times 0.1$ mm$^3$. The crystal axes of the sample were determined by two independent methods, x-ray and upper critical field measurements as a function of magnetic field angle 13, which were consistent with each other. The $T_c$’s, determined by the point where the steepest drop occurs in the resistive superconducting transition, are 15.5, 15.9, and 16.1 K for sample A, C and N respectively (not shown). The resistivity at $T_c$ is 2.34 and 1.44 $\mu\Omega$-cm for sample A and sample N, corresponding to mean free paths of 144.5 and 234 Å respectively. Assuming that 16.1 K is the transition temperature for a pure sample, sample A with $T_c$ of 15.5 K is equivalent to 0.8 % of Co doping on the Ni site, i.e., Lu(Ni$_{1-x}$Co$_x$)$_2$B$_2$C with $x = 0.008$ 18. The disorder in sample A may be associated with defects which can be removed by judicious post growth annealing 19.

Using an ac technique 8, we have measured the low temperature specific heat of non-magnetic LuNi$_2$B$_2$C as a function of magnetic field intensity and magnetic field angle. Three samples of LuNi$_2$B$_2$C with different $T_c$’s, labeled A, C, and N, were studied and revealed an anomalous disorder effect. The heat capacity of the samples...
The best fit is obtained when the deviation at 8 K. Above the deviation field, the data fall below the $H^{1/2}$ line.

Field-angle heat capacity directly measures the change in the DOS with magnetic field direction. The DOS of a d-wave superconductor has been shown to exhibit a fourfold oscillation with field angle against crystal axes. At $T = 0$ K, the DOS has a simple form:

$$N(E, H, \alpha)/N_0 \approx D_4(1 + \Gamma \sin 2\alpha),$$

where $D_4$ is a Doppler-shift induced coefficient and $\Gamma$ describes the oscillation amplitude. The field-angle sensitive Doppler effect, arising from the supercurrent flows circulating around the vortices, leads to maxima in the DOS when field is along gap maxima and DOS minima when field is along nodes. A 3D superconductor has a much reduced oscillation amplitude ($\approx 6\%$) compared to that of a 2D system ($\approx 40\%$) due to contributions from the out-of-plane component. We note that a similar effect is predicted for an (s+g)-wave superconductor.

Fig 2(a) and 2(b) show the low-temperature field-angle heat capacity of sample $C$ at 2.5 K and sample $A$ at 2 K, respectively. The samples were field-cooled to 2 K (or 2.5 K) and were rotated within the $ab$-plane by a computer controlled stepping motor at increments of $3^\circ$. The heat capacity with increasing and decreasing field angle showed reversible behavior for all measured fields, indicating that flux pinning effect is negligible in our measurement. Background contributions ($C_{bkg}$) from lattice vibrations and thermometry were subtracted in the usual manner and the remaining field-induced heat capacity $\Delta C = C_{total} - C_{bkg}$ was analyzed in terms of $\Delta C(\alpha) = c(1 + \Gamma \sin 2\alpha)$ for pure samples. At low fields, there is a clear fourfold oscillation with minima along $<100>$ for both samples, indicating that the zeros of the gap are located along those directions, consistent with those of YNi$_2$B$_2$C.

At 1 T, surprisingly, the heat capacity of sample $A$ develops minima along $<110>$, producing two sets of fourfold patterns or 8-fold, an effect not observed in either sample $C$ or sample $N$ with higher $T_c$'s. The crossover field from the fourfold to the $(4 + 4)$ pattern of sample $A$ lies between 0.6 and 1 T, which is also the point where the heat capacity of sample $A$ deviates from the square-root field dependence (see Fig. 1). With increasing field, the splitting in sample $A$ gradually disappears and the field-angle heat capacity recovers its fourfold pattern above $800 \%$ of $C_{total}$.

with higher $T_c$'s shows a fourfold pattern as a function of magnetic field angle, confirming the result from the other non-magnetic borocarbide YNi$_2$B$_2$C that the superconducting gap is highly anisotropic with nodes along $<100>$. In contrast, the heat capacity of the disordered sample with the lowest $T_c$ (sample $A$) shows a dramatic change in the field-angle heat capacity above 0.8 T at 2 K; the maxima along $<110>$ split, giving rise to minima separated by $\pi/4$ (eightfold pattern) and, further, $C_p$ deviates from a square-root field dependence at the same field, labeled $H_{c1}$. The eightfold pattern and the deviation from $H^{1/2}$ dependence in sample $A$ are consistent with the coexistence of an anisotropic gap and nonlocal effects.

Fig. 1(a) shows the magnetic field dependence of the heat capacity of sample $C$ at 2.5 K with increasing (circles) and decreasing (crosses) magnetic field. The dashed line represents the least-square fit of $C_0 + b(H - H_0)^2$, where $C_0$ is zero-field heat capacity and $\mu_0 H_0 = 0.1$ T is a fitting parameter that takes account of the Meissner effect. The best fit is obtained when $\beta = 0.46$, namely the Volovik effect for nodal superconductors, and is consistent with previous reports on the non-magnetic borocarbides [4]. Fig. 1(b), (c), and (d) shows the heat capacity of sample $A$ at 2.5 K, 4 K, and 8 K, respectively. The dashed line is the square-root field dependence and the arrows indicate where the data deviate from the fit. The deviation field shows systematic increase with temperature, i.e., 0.8 T at 2.5 K, 1.8 T at 4 K, and no clear deviation at 8 K. Above the deviation field, the data fall below the $H^{1/2}$ line.
4 T. We also measured the field-angle heat capacity of sample A at 4 K to check if the anomalous peak splitting persists at higher temperatures (not shown). The fourfold pattern now persists to 1 T, evolving into two sets of fourfold patterns above 2 T. The 4 T data at 4 K has a shape similar to the 2 T data at 2 K. The crossover field $H_{s1}$ increases with increasing temperature.

We focus on the fact that the anomalous 8-fold pattern occurs only at sample A which has half the electronic mean free path of the sample N while the $T_c$ is slightly decreased. According to the nonlocal theory by Kogan et al. [12], the hexagonal-to-square FLL transition depends on the electronic mean free path $l$ and the superconducting coherence length $\xi$ of the sample. Gammel et al. found that a mere 9% of Co doping onto the Ni site in Lu1221 can make the FLL transition field at least 20 times higher than that of pure matrix for $H \parallel [001]$ [25]. Because the FLL transition field for pure sample is expected to be small [12]—possibly below our measurement range—the nonlocal effects would not influence the field-angle heat capacity of sample N (or C). In contrast, the disorder in sample A is expected to increase the transition to a higher field, i.e. to a field relevant in the field-angle heat capacity measurement.

When the magnetic field is rotated within ab-plane, the transition field may differ with different field directions because of the different nonlocal range, i.e., $\xi/l$. The two different transition fields can be characterized by $H_{s1}$ and $H_{s2}$. As a magnetic field rotates within the $ab$-plane for $H_{s1} \leq H \leq H_{s2}$, the FLL will experience a structural change (or distortion), i.e. hexagonal for $H \parallel [100]$ and square for $H \parallel [110]$. Since the borocarbides have nodes on the Fermi surface, the DOS will differ depending on the FLL structure [21]. The negative deviation from the $H^{1/2}$ above $H_{s1}$ in the heat capacity of sample A (see Fig. 1b) also indicates that the DOS of the hexagonal FLL is larger than that of the square FLL. The additional FLL anisotropy in the DOS will modulate the gap-anisotropy oscillation and leads to a (4+4)-fold pattern.

We hypothesize that the two effects are independent of each other and have a form of two cusped 4-fold oscillations in the DOS:

$$\Delta C(\alpha) = p_1 + p_2(1+\Gamma|\sin 2\alpha|)(1+\gamma|\sin 2(\alpha-45)|),$$

(2)

where $p_1$ and $p_2$ are fitting parameters. The value $\Gamma$ represents the oscillation due to gap anisotropy in pure samples (see Fig. 3). The nonlocal effects give rise to a 45°-shifted 4-fold pattern and are accounted for by $\gamma$. The solid line in Fig. 2(b) is the least square fit of Eq. (2) and represents the data very well. The oscillations due to the nonlocal effects ($\gamma$) and the gap anisotropy ($\Gamma$) at 2 K are compared as a function of magnetic field at the bottom panel of Fig. 3. The FLL effect $\gamma$ increases sharply above 0.6 T and decreases gradually to zero at 4 T, indicating that the low field corresponds to $H_{s1}$ and the high field to $H_{s2}$.

Fig. 4 summarizes the $H-T$ phase diagram of the disordered sample A. Unlike pure samples, it has additional
FIG. 4: $H - T$ phase diagram of sample $A$ with lowest $T_c$. $H_{s1}$ is the FLL transition for $H ||[110]$ and $H_{s2}$ for $H ||[100]$. The dotted lines are guide to the eye. Triangles and squares were sketched to show the corresponding FLL.

In summary, we have studied the non-magnetic superconductor LuNi$_2$B$_2$C via the dependence of heat capacity on magnetic field angle and magnetic field. Unlike pure samples, a slightly disordered sample $A$ shows a deviation from $H^{1/2}$ and a (4+4)-fold pattern in $C_p(\alpha)$ at above 1 T. The anomalous properties were explained in terms of the coexistence of gap anisotropy and nonlocal effects. These experiments resolve the apparently irreconcilably different views on the nature of the order parameter in the non-magnetic borocarbides.

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[1] Y. J. Uemura, L. P. Le, G. M. Luke, B. J. Sternlieb, W. D. Wu, J. H. Brewer, T. M. Riseman, C. L. Seaman, M. B. Maple, M. Ishikawa, et al., Phys. Rev. Lett. 66, 2665 (1991).
[2] J. F. Annett, Physica C 317, 1 (1999).
[3] B. H. Brandow, Phil. Mag. 83, 2487 (2003), references therein.
[4] M. Nohara, M. Ishiki, H. Takagi, and R. J. Cava, J. Phys. Soc. Jpn. 66, 1888 (1997).
[5] G.-Q. Zheng, Y. Wada, K. Hashimoto, Y. Kitaoka, K. Asayama, H. Takeya, and K. Kadowaki, J. Phys. Chem. Solids 59, 2169 (1998).
[6] E. Boaknin, R. W. Hill, C. Proust, C. Lupien, L. Taillefer, and P. C. Canfield, Phys. Rev. Lett. 87, 237001 (2001).
[7] K. Izawa, K. Kamata, Y. Nakajima, Y. Matsuda, T. Watanabe, M. Nohara, H. Takagi, P. Thalmeier, and K. Maki, Phys. Rev. Lett. 89, 137006 (2002).
[8] T. Park, M. B. Salamon, E. M. Choi, H. J. Kim, and S.-I. Lee, Phys. Rev. Lett. 90, 177001 (2003).
[9] Y. DeWilde, M. Javaroni, U. Welp, V. Metlushko, A. E. Koshelev, I. Aranson, G. W. Crabtree, and P. C. Canfield, Phys. Rev. Lett. 78, 4273 (1997).
[10] M. R. Eskildsen, P. L. Gammel, B. P. Barber, A. P. Ramirez, D. J. Bishop, N. H. Andersen, K. Mortensen, A. C. Bolle, C. M. Lieber, and P. C. Canfield, Phys. Rev. Lett. 79, 487 (1997).
[11] M. Yethiraj, D. M. Paul, C. V. Tomy, and E. M. Forgan, Phys. Rev. Lett. 78, 4849 (1997).
[12] V. G. Kogan, M. Bullock, B. Harmon, P. Miranovic, L. Dobrosavljevic-Grujic, P. L. Gammel, and D. J. Bishop, Phys. Rev. B 55, R8693 (1997).
[13] V. Metlushko, U. Welp, A. Koshelev, I. Aranson, G. W. Crabtree, and P. C. Canfield, Phys. Rev. Lett. 79, 1738 (1997).
[14] L. Civale, A. V. Silhanek, J. R. Thompson, K. J. Song, C. V. Tomy, and D. M. Paul, Phys. Rev. Lett. 83, 3920 (1999).
[15] N. Nakai, P. Miranovic, M. Ichio, and K. Machida, Phys. Rev. Lett. 89, 237004 (2002).
[16] M. R. Eskildsen, A. B. Abrahamsen, V. G. Kogan, P. L. Gammel, K. Mortensen, N. H. Andersen, and P. C. Canfield, Phys. Rev. Lett. 86, 5148 (2001).
[17] B. K. Cho, P. C. Canfield, L. L. Miller, D. C. Johnston, W. P. Beyermann, and A. Yatskar, Phys. Rev. B 52, 3864 (1995).
[18] K. O. Cheon, I. R. Fisher, V. G. Kogan, P. C. Canfield,
P. Miranovic, and P. L. Gammel, Phys. Rev. B 58, 6463 (1998).

[19] X. Y. Miao, S. L. Bud’ko, and P. C. Canfield, J. Alloys Comp. 338, 13 (2002).

[20] G. E. Volovik, JETP Lett. 58, 469 (1993).

[21] M. Ichioka, A. Hasegawa, and K. Machida, Phys. Rev. B 59, 184 (1999).

[22] I. Vekhter, P. J. Hirschfeld, J. P. Carbotte, and E. J. Nicol, Phys. Rev. B 59, R9023 (1999).

[23] K. Maki, H. Won, P. Thalmeier, Q. Yuan, K. Izawa, and Y. Matsuda, Europhys. Lett. 64, 496 (2003).

[24] L. F. Mattheiss, Phys. Rev. B 49, 13279 (1994).

[25] P. L. Gammel, D. J. Bishop, M. R. Eskildsen, K. Mortensen, N. H. Andersen, I. R. Fisher, K. O. Cheon, P. C. Canfield, and V. G. Kogan, Phys. Rev. Lett. 82, 4082 (1999).

[26] H. Sakata, M. Oosawa, K. Matsuba, N. Nishida, H. Takeya, and K. Hirata, Phys. Rev. Lett. 84, 1583 (2000).