Evidence for the Coexistence of an Anisotropic Superconducting Gap and Nonlocal Effects in the Nonmagnetic Superconductor LuNi$_2$B$_2$C

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labeled $A$, $C$, and $N$, were studied and revealed an anomalous disorder effect. The heat capacity of the samples with higher $T_c$'s shows a fourfold pattern as a function of magnetic field angle, confirming the result from the other nonmagnetic borocarbide YNi$_2$B$_2$C that the superconducting gap is highly anisotropic with nodes along $\langle100\rangle$ [8]. 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 $\langle110\rangle$ 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 $H_d$. The eightfold pattern and the deviation from $H^{1/2}$ dependence in sample $A$ are consistent with the coexistence of an anisotropic gap and non-local effects.

Figure 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 $C_0 + b(H-H_0)^\beta$, where $C_0$ is zero-field heat capacity and $\mu_0H_0 = 0.1$ 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 [20,21], and is consistent with previous reports on the nonmagnetic borocarbides [4]. Figures 1(b)–1(d) show the heat capacity of sample $A$ at 2.5, 4, 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 a 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.

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 [22]. At $T = 0$ K, the DOS has a simple form:

$$N(E, H, \alpha)/N_0 \approx D_4(1 + \Gamma|\sin2\alpha|),$$

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 the field is along gap maxima and DOS minima when the field is along nodes. A 3D superconductor has a much reduced oscillation amplitude ($=6\%$) compared to that of a 2D system ($=40\%$) due to contributions from the out-of-plane component. We note that a similar effect is predicted for an $(s + g)$-wave superconductor [23].

Figures 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 the flux pinning effect is negligible in our measurement. Background contributions ($C_{\text{bkg}}$) from lattice vibrations and thermometry were subtracted in the usual manner [8], and the remaining field-induced heat capacity $\Delta C = C_{\text{total}} - C_{\text{bkg}}$ was analyzed in terms of $\Delta C(\alpha) = c(1 + \Gamma|\sin2\alpha|)$ for pure samples. At low fields, there is a clear fourfold oscillation with minima along $\langle100\rangle$ for both samples, indicating that the zeros of the gap are located along those directions, consistent with
The two different transition fields can be characterized by the field-angle heat capacity measurement. At 1 T, surprisingly, the heat capacity of sample A develops minima along \( \langle 110 \rangle \), producing two sets of fourfold patterns or eightfold, 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 transition field may differ with different field direction (see Fig. 1). With increasing field, the splitting in sample A gradually disappears and the field-angle heat capacity recovers its fourfold pattern above 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 have a shape similar to the 2 T data at 2 K. The crossover field \( H_{c1} \) increases with increasing temperature.

We focus on the fact that the anomalous eightfold pattern occurs only at sample A which has half the electronic mean-free path of 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 a pure matrix for \( H \parallel [001] \). Because the FLL transition field for the 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 the \( ab \) 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 \( H_{c1} \) and \( H_{c2} \). As a magnetic field rotates within the \( ab \) plane for \( H_{c1} \leq H \leq H_{c2} \), 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 \( H_{c1} \) in the heat capacity of sample A [see Fig. 1(b)] 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 fourfold 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 fourfold 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 in 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 \( H_{c1} \) and the high field to \( H_{c2} \).

Figure 4 summarizes the \( H – T \) phase diagram of the disordered sample A. Unlike pure samples, it has additional phase lines in the superconducting state where the field-angle heat capacity shows a crossover from the fourfold to the \( (4 + 4) \) pattern (\( H_{c1} \)) or vice versa (\( H_{c2} \)). The increase in the crossover fields with increasing temperature is consistent with the nonlocal effects [12], adding strength to our viewpoint that the anomalous eightfold pattern is due to the coexistence of nonlocal effects and gap anisotropy. We note, however, that the difference between the two transition fields \( H_{c1} \) and \( H_{c2} \) is larger than that expected within the nonlocal theory [12]. For discussion, we assume that the anisotropy in the FLL transition field between \( \langle 001 \rangle \) and \( \langle 100 \rangle \) is similar to that between \( \langle 110 \rangle \) and \( \langle 100 \rangle \) because the \( H_{c2} \) anisotropy is similar to the 3D nodal quasiparticle theory with \( v_F = 1.5 \times 10^7 \) cm/s [8].
is almost the same for the two configurations. The observed ratio $H_{c2}/H_{c1} = 4$ is larger than the predicted ratio 2 [12], but is smaller than the reported ratio of 10 in $YNi_2B_2C$ [26]. The difference between experiments and theory may attest that we need to account for both nonlocality and the anisotropic gap nature of the borocarbides.

In summary, we have studied the nonmagnetic superconductor $LuNi_2B_2C$ via the dependence of heat capacity on a magnetic field angle and a magnetic field. Unlike pure samples, a slightly disordered sample $A$ shows a deviation from $H^{1/2}$ and a $(4 + 4)$-fold pattern in $C_p(\alpha)$ 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 nonmagnetic borocarbides.

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