Low Energy Quasiparticle Excitation in the Vortex State of Borocarbide Superconductor YNi_2B_2C

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We measured the heat capacity C_p and microwave surface impedance Z_s in the vortex state of YNi_2B_2C. In contrast to conventional s-wave superconductors, C_p shows a $\sqrt{H}$ dependence. This $\sqrt{H}$ dependence persists even after the introduction of the columnar defects which change the electronic structure of the vortex core regime and destroy the regular vortex lattice. On the other hand, flux flow resistivity is nearly proportional to H. These results indicate that the vortex state of YNi_2B_2C is fundamentally different from the conventional s-wave counterparts, in that the delocalized quasiparticle states around the vortex core are important, similar to d-wave superconductors.

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The borocarbide superconductors $L\mathrm{Ni_2B_2C}$, where $L =$ (Y, Lu, Tm, Er, Ho, and Dy) exhibit a rich variety of interesting physical properties. In particular, the occurrence of superconductivity at elevated temperatures [1], the competition and coexistence of the antiferromagnetic ordering and superconductivity [2], and the transition between a triangular and a square vortex lattice [3] have attracted much attention. In spite of extensive studies on these subjects, however, one of the most fundamental properties of the superconducting state, namely, the quasiparticle (QP) structure in the vortex state, is still controversial. In fact, recent measurements of heat capacity C_p on YNi_2B_2C and $LuNi_2B_2C$ in the vortex state have shown that C_p clearly indicates the presence of the $\sqrt{H}$ term [4]. In the conventional s-wave superconductors, C_p should increase linearly with H because all the QPs are trapped within the vortex core with the radius of the coherence length ξ, and hence the QP density of states (DOS) $N(H)$ is proportional to the number of vortices; $N(H) \propto N_F \xi^2 H$, where N_F is the DOS at the Fermi level in the core [5]. Thus, the observed $\sqrt{H}$ dependence of C_p is strikingly in contrast to the ordinary s-wave superconductors. The $\sqrt{H}$ dependence of C_p has been reported in some of the unconventional superconductors with gap nodes in the QP spectrum such as high-T_c $YBa_2Cu_3O_7$ [6] and heavy fermion UPt_3 [7] superconductors. Nonlinear H dependence close to the $\sqrt{H}$ of C_p is also observed in some s-wave clean superconductors such as $CeRu_2$ [8] and $NbSe_2$ [4,9].

Although the unusual H-dependent C_p of borocarbide superconductors has been discussed in terms of several intriguing models, the issue is still far from being settled. For instance, several authors proposed the shrinking of the vortex core with H [4,9–11], but the physical origin behind this phenomenon is unclear. Another group ascribed it to the field-induced gap nodes [8], but this scenario is beyond the applicability of the original argument [12]. Moreover, the extended QP states outside the vortex core owing to a presupposed d-wave symmetry have been invoked [13]. There is, however, no corroborative evidence for d-wave pairing. Therefore, the experimental clarification of the QP structure in the vortex state of YNi_2B_2C is very much needed. Further, it is crucial for understanding the vortex lattice structure. If a substantial portion of the QPs extends in specific directions well outside the core, it should play an important role in determining the superconducting properties, including the vortex lattice structure, magnetization $M(H)$, upper critical field H_{c2}, etc. [14]. Nevertheless, these properties have been discussed in terms of an ordinary s-wave superconductor with an anisotropic Fermi surface with the use of the nonlocal London theory, *without taking into account the effect of the extended QPs* [15].

We stress that we can extract detailed information about the QP spectrum only when we measure both the heat capacity and the surface impedance Z_s for the same sample. In superconductors with gap nodes, C_p is essentially determined by the QPs excited *in the node directions* [16]. Energy dissipation in the flux flow state, on the other hand, occurs mainly in the "normal regions" created by the vortices. Thus, the dissipative response is dominated by Andréev bound states localized within vortex cores; those states have momenta whose directions lie *away from the nodes* [17]. Therefore, the measurement of Z_s is complementary to C_p. In this Letter, we report measurements for both Z_s and C_p in the vortex state of YNi_2B_2C. We also examined the effect of introducing columnar defects (CD) on C_p; since the diameter of CD is comparable to $\xi \sim 70$ Å of YNi_2B_2C and the inside of CD is semiconducting, they should strongly influence the electronic structure of the vortices. As we shall discuss below, these results provide strong evidence that the $\sqrt{H}$ term in C_p originates mainly from the Doppler shift of the delocalized QP spectrum due to the superfluid electrons surrounding the vortex cores [16].

Single crystals of YNi_2B_2C with $T_c = 13.4$ K were grown by the floating zone method. In all measurements, dc magnetic fields $\boldsymbol{H}$ were applied parallel to the c axis. The microwave surface impedance $Z_s = R_s + iX_s$, where R_s and X_s are the surface resistance and the reactance, respectively, was measured by the cavity perturbation technique. We used a cylindrical Cu cavity with $Q \sim 3 \times 10^4$ operated at 28.5 GHz in the TE_{011} mode. In our configuration, the vortex lines along the c axis respond to oscillatory driving currents within the ab plane induced by $\boldsymbol{H}_{\rm ac}$ ($\boldsymbol{H}_{\rm ac} \parallel \boldsymbol{H} \parallel c$). Several single crystals $\sim$100 μm in thickness along the c axis were irradiated at GANIL (Caen, France) with a 6.0-GeV Pb-ion beam aligned parallel to the c axis. The samples were irradiated to the fluence of 5×10^{10} and 1×10^{11} ions/cm^2, corresponding to a dose-equivalent flux density of $B_\Phi = 1$ and 2 T, respectively. The resulting damage consisted of a random array of amorphous columns, about 60–70 Å in diameter and continuous throughout the thickness of the crystal, which we confirmed by the transmission electron microscope image. The CD act as strong pinning centers of the vortices; at $B \lesssim B_\Phi$, a substantial portion of the vortices is trapped inside the CD. Particularly below $\sim 0.2 B_\phi$, almost all vortices are expected to be trapped [18]. The CD do not, however, act as strong scattering centers of the electrons, unlike magnetic impurities or point defects. In fact, after the irradiation, $M(H)$ showed a broad peak at $B \sim B_\Phi$ due to the trapping of the vortices by the CD (inset of Fig. 1), but the increase in the resistivity was very small (less than 10%) and the T_c did not change.

Figure 1 plots C_p/T as a function of T^2 for the pristine YNi_2B_2C. We will first remark on the T dependence of C_p. Obviously, the normal state in Fig. 1 (C_p/T vs T^2) that occurs as the superconductivity is suppressed by magnetic fields deviates from a linear dependence. Although this deviation may be attributed to the low energy optical phonons, it is not clear where they originate [19]. Therefore, the quantitative analysis of the T dependence of the electronic contribution obtained by subtracting the phonon terms from the total C_p is precarious.

Figure 2(a) depicts the H dependence of the heat capacity for the pristine YNi_2B_2C at a low temperature, where the phonon contribution is negligible. In order to discuss the heat capacity induced by a magnetic field, we plotted $\Delta C_p(H)/T = [C_p(H) - C_p(0)]/T$ in Fig. 2(a). A significant deviation from a linear H dependence is clearly observed. The inset of Fig. 2(a) plots $\Delta C_p(H)/T$ as a function of $\sqrt{H}$. At a low field, $\Delta C_p(H)/T$ increases with an upward curvature with respect to $\sqrt{H}$. However, at $\sqrt{H} \gtrsim 0.25$ T$^{1/2}$ ($H \gtrsim 70$ mT), $\Delta C_p/T$ obviously increases linearly to $\sqrt{H}$. This $\sqrt{H}$ dependence persists close to H_{c2}. Since the lower critical field H_{c1} determined by the M-H curve is of the order of 30 mT, the $\sqrt{H}$-dependent $\Delta C_p/T$ is observed in almost the whole regime of the

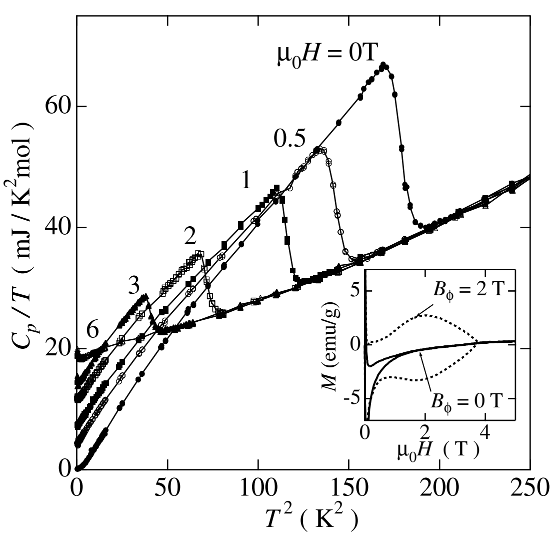

FIG. 1. C_p/T versus T^2 for the pristine YNi_2B_2C. All measurements have been done in the field cooling conditions. Inset: M-H curves for pristine and irradiated ($B_\Phi = 2$ T) crystals at 5 K. While in the pristine crystal $M(H)$ is reversible in a wide H-range, showing very weak pinning centers; $M(H)$ shows a broad peak at $B = B_\Phi$ in the irradiated crystal.

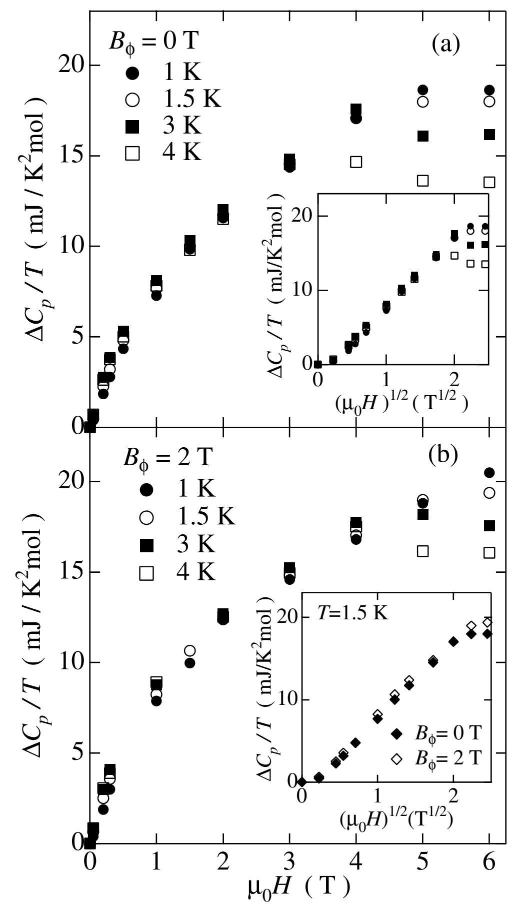

FIG. 2. (a) H dependence of $\Delta C_p(H)/T = \{C_p(H) - C_p(0)\}/T$ for the pristine YNi_2B_2C at low temperatures. Inset: $\Delta C_p/T$ as a function of $\sqrt{H}$. (b) The same plot for the irradiated crystal with $B_\Phi = 2$ T [31]. Inset: the H dependence of $\Delta C_p/T$ before and after the irradiation.

vortex state. Figure 2(b) shows the same plot for the irradiated YNi_2B_2C with $B_\Phi = 2$ T. A significant deviation from the linear H dependence persists even after the irradiation. Surprisingly, $\Delta C_p/T$ is little affected by the introduction of the CD [see the inset of Fig. 2(b)]. We obtained qualitatively similar results for YNi_2B_2C with $B_\Phi = 1$ T. Before discussing these results, we will discuss Z_s in the pristine YNi_2B_2C.

The inset of Fig. 3 shows the T dependence of Z_s in the Meissner phase. Both R_s and X_s decrease rapidly with decreasing T below T_c. Figure 3 shows Z_s as a function of $\sqrt{H}$ at 1.5 K. Let us quickly recall the behavior of Z_s in type-II superconductors. In the Meissner phase, the microwave response is purely reactive and $R_s \simeq 0$ and $X_s = \mu_0\omega\lambda_L$, where μ_0 is the vacuum permeability, ω is the microwave frequency, and λ_L is the London penetration length. On the other hand, the response is dissipative in the normal state and $R_s = X_s = \mu_0\omega\delta/2$, where $\delta = \sqrt{2\rho_n/\mu_0\omega}$ is the skin depth and ρ_n is the normal state resistivity. We determined the absolute values of R_s and X_s from comparison with the dc resistivity and assuming that $R_s = X_s$ in the normal state. In the vortex state, Z_s is determined by the vortex dynamics. Since the present microwave frequency is 3 orders of magnitude higher than the vortex pinning frequency of YNi_2B_2C [20], a completely free flux flow state is realized. In the flux flow state, two characteristic length scales, namely, λ_L and the flux flow skin depth $\delta_f \sim \sqrt{2\rho_f/\mu_0\omega}$, appear in accordance with the microwave field penetration. At a low field, λ_L greatly exceeds δ_f ($\lambda_L \gg \delta_f$). In this regime, R_s and X_s are given as $R_s \sim \rho_f/\lambda_L$ and $X_s \sim \mu_0\omega\lambda_L$. On the other hand, at high fields where δ_f greatly exceeds λ_L ($\delta_f \gg \lambda_L$), the viscous loss becomes dominant and the response is similar to the normal state ($R_s \simeq X_s$), except that δ is replaced by δ_f. In YNi_2B_2C this crossover occurs at a very low field ($\mu_0 H \sim 20$ mT). According to Coffey and Clem [21], Z_s in the flux flow state is expressed as

$$Z_s = i\mu_0\omega\lambda_L\left[\frac{1 - (i/2)\delta_f^2/\lambda_L^2}{1 + 2i\lambda_L^2/\delta_{qp}^2}\right]^{1/2}, \tag{1}$$

where $\delta_{qp} = \sqrt{2\rho_{qp}/\mu_0\omega}$ with the QP resistivity ρ_{qp} is the normal-fluid skin depth. We analyzed the data by means of Eq. (1). We considered two different H dependences of $N(H)$, namely, $N(H) \propto H$ (case I) and $N(H) \propto \sqrt{H}$ (case II). Case I corresponds to the conventional Bardeen-Stephen relation, $\rho_f = (H/H_{c2})\rho_n$ and $\lambda_L^2(H) = \lambda_L^2(0)/(1 - H/H_{c2})$, while case II corresponds to the vortex core shrinking, $\rho_f \sim \sqrt{H/H_{c2}}\,\rho_n$ and $\lambda_L^2(H) = \lambda_L^2(0)/(1 - \sqrt{H/H_{c2}})$. In both cases we used $\lambda_L(0) = 500$ Å [22]. The solid and dashed lines in Fig. 3 represent the theoretical calculations of R_s and X_s by Eq. (1), respectively. Comparing the two cases, case I obviously describes the data much better than case II. Small deviations of the fits from the data in case I may be due to the fact that ρ_f is not strictly linear in H [23].

The results of Z_s and ΔC_p for the pristine and irradiated YNi_2B_2C offer important clues for understanding the QP structure. Figure 3 shows that the linear H-dependent Bardeen-Stephen relation yields a more consistent fit to the data than another model does. Since the QPs localized in the core mainly contribute to the flux flow dissipation, this fact shows that the number of the QPs trapped within each core is independent of H. Therefore, the scenario of the core shrinking with H as an origin of nonlinear H-dependent C_p proposed by several groups [4,9–11] is completely excluded. In addition, and more importantly, the fact that the existence of the CD with a comparable radius with ξ little affects the ΔC_p implies that the QPs within the core radius ξ are not important for the total heat capacity in the pristine YNi_2B_2C. On the basis of these results, we are led to conclude that in YNi_2B_2C *the extended QP states around the vortex core play an important role in determining the superconducting properties, similar to d-wave superconductors*.

Moreover, the effect of the CD on the ΔC_p provides another important piece of information about the microscopic origin of $\sqrt{H}$ dependence. It has been pointed out that in the presence of line nodes there are two sources for the $\sqrt{H}$-dependent $N(H)$, namely, the contributions from localized and delocalized fermions [16], but which of the two dominates has thus far been left unquestioned. Here we shall attempt to address this issue. One arises from the localized QPs in the node directions. Since the extended QP wave function in the node directions is cut off by its adjacent vortices, the area per a single vortex is proportional to the intervortex distance $R \propto 1/\sqrt{H}$. Then $N(H)$ is given as $N(H) \propto N_F\xi RH \propto \sqrt{H}$. The other arises from the delocalized QPs. In the presence of the supercurrent flow with velocity $\boldsymbol{v}_s$, the energy spectrum of the delocalized QPs is shifted by the Doppler effect as $E(\boldsymbol{p}) \rightarrow E(\boldsymbol{p}) + \boldsymbol{v}_s \cdot \boldsymbol{p}$. In superconductors with line

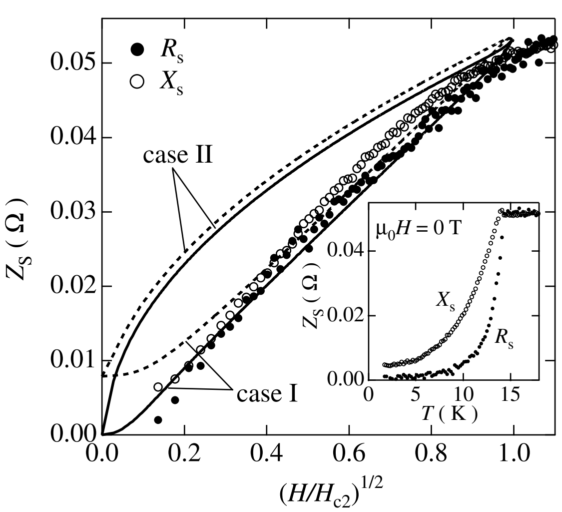

FIG. 3. Inset: T dependence of $Z_s = R_s + iX_s$ in the Meissner phase. Main panel: R_s and X_s as a function of $(H/H_{c2})^{1/2}$ at 1.5 K. The solid and dashed lines represent the result of the theoretical calculations of R_s and X_s by Eq. (1), respectively, assuming $N(H) \propto H$ (case I) and $N(H) \propto \sqrt{H}$ (case II).

nodes such as d-wave symmetry where DOS has a linear energy dependence in the bulk $[N(E) \propto E]$, the Doppler-shifted QPs give rise to the finite DOS at the Fermi level. Then $N(H)$ is obtained by integrating the vortex lattice cell $N(H) \propto R^{-2} \int_{\xi}^{R} \boldsymbol{v}_s(\boldsymbol{r}) \cdot \boldsymbol{p} r\, dr \propto \sqrt{H}$, where $|\boldsymbol{v}_s(\boldsymbol{r})| = \hbar/2mr$ is the velocity of the circulating supercurrents. The very weak influence of the CD on ΔC_p elucidates the fact that *the Doppler shift of the delocalized QPs is mainly responsible for the* $\sqrt{H}$*-dependent* C_p, because the contribution from the localized QPs in the node directions should diminish when the vortex lattice structure is destroyed.

At low fields ($H \lesssim 70$ mT $\approx H_{cr}$), the $\Delta C_p/T$ vs $\sqrt{H}$ curve shows an upward curvature. There are two origins for the deviation from the $\sqrt{H}$ behavior. The first one is the lower critical field $H_{c1} \sim 30$ mT, which is in the same order of H_{cr}. The second one is the crossover from high field to low field scaling. Generally, the heat capacity of superconductors with line nodes is proportional to $\sqrt{H}$ only at *high fields,* where the number of Doppler-shifted QPs exceeds that of thermally excited QPs. The heat capacity at low fields should be described by the zero field scaling $C_p/(\gamma_n T) \sim k_B T/\Delta$, where γ_n and Δ are the Sommerfeld coefficient in the normal state and superconducting energy gap, respectively [24]. The crossover field from high field scaling to low field scaling roughly estimated from the relation $\sqrt{H/H_{c2}} \sim k_B T/\Delta$ with $\mu_0 H_{c2} = 5.5$ T, $T_c = 13.4$ K, and $T = 1.5$ K is $\sim$24 mT, which is also in the same order of H_{cr}. It should be noted that this behavior is also reported in $YBa_2Cu_3O_{7-\delta}$ with d-wave symmetry [25].

The present experiments strongly suggest that the influence of the extended QPs, which has been ignored by many authors, should be taken into account when discussing the vortex lattice structure, H_{c2}, and $M(H)$. In YNi_2B_2C, de Haas–van Alphen oscillations are observed as low as $H_{c2}/5$ [26]. This unusual phenomena may be related to the extended QPs. It is tempting to relate the observed relevance of the delocalized QPs to a three-dimensional d-wave superconductivity [13]. However, recent doping studies on YNi_2B_2C show that the superconductivity survives even in the dirty limit [4,27]. This robustness of the superconductivity against impurities makes a d-wave state unlikely [28]. Therefore, we believe that a strongly anisotropic s-wave state, in which $N(E) \propto E$ is similar to a d-wave state, is most likely for YNi_2B_2C.

We finally comment on the tunneling measurement by Ekino *et al.* which reports a full Bardeen-Cooper-Schrieffer (BCS) gap in YNi_2B_2C [29]. We believe that their break junction method selects a specific q direction in the gap function.

In summary, we have studied the heat capacity and surface impedance of pristine and irradiated YNi_2B_2C at low temperatures. We provided strong evidence that the Doppler shift of the delocalized QP spectrum around the vortex core plays an important role in determining the superconducting properties of YNi_2B_2C.

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