Quasiparticle Density of States of Clean and Dirty $\delta$-Wave Superconductors in the Vortex State

M. Nohara$^{1,2}$, M. Isshiki$^1$, F. Sakai$^1$, and H. Takagi$^{1,2}$

$^1$Institute for Solid State Physics, University of Tokyo, 7-22-1, Roppongi, Minato-ku, Tokyo 106-8666, Japan
$^2$CREST, Japan Science and Technology Corporation, Japan

(Received January 14, 1999)

The quasiparticle density of states (DOS) in the vortex state has been probed by specific heat measurements under magnetic fields ($H$) for clean and dirty $\delta$-wave superconductors, Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C and Nb$_{1-x}$Ta$_x$Se$_2$. We find that the quasiparticle DOS per vortex is appreciably $H$-dependent even in the clean-limit superconductors, while it is $H$-independent in the dirty superconductors as expected from a conventional rigid normal electron core picture. We discuss possible origins for our observations in terms of the shrinking of the vortex core radius with increasing $H$.

PACS numbers: 74.25.Bt, 74.60.Ec, 74.70.Ad

Determination of the electronic structure of magnetic vortex lines in type-II superconductors has been an issue of long standing interest. Recent attention has focused on its relation with the symmetry of the Cooper pairs: For $s$-wave superconductors with an isotropic gap, there exist low-lying bound quasiparticles around the center of the vortex lines, where the superconducting gap vanishes, and the vortex line has been simply viewed as a “rigid” normal electron cylinder with a radius of the coherence length $\xi$. On the other hand, for $d$-wave superconductors, quasiparticles are delocalized and spread outside the vortex cores because of the presence of the gapless region on the Fermi surface. This difference manifests itself in, for example, a magnetic field ($H$) dependence of the quasiparticle density of states (DOS) at the Fermi level $N_0(H)$: $N_0(H) \propto H$ for $s$-wave superconductors, while $N_0(H) \propto \sqrt{H}$ for $d$-wave superconductors with line nodes. However, even in $s$-wave superconductors, the following unsolved issues may potentially alter the above simple picture of the vortex lines: Shrinking of the core radius with decreasing temperature ($T$), due to the thermal population of the quantized levels for the bound excitations, has been predicted theoretically by Pesch and Kramer. Shrinking of the core radius with increasing $H$ has been proposed based on scanning tunneling spectroscopy (STS) and muon spin rotation (µSR) measurements, though the physics behind this has not been fully explored yet. The de Haas - van Alphen (dHvA) effect has been observed in the mixed state where the cyclotron radius is much larger than $\xi$. This suggests the existence of delocalized quasiparticles even in the $s$-wave superconductors under high magnetic fields.

In this letter, we report the low-temperature specific heat of pure and alloyed Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C and Nb$_{1-x}$Ta$_x$Se$_2$ single crystals under magnetic fields, focusing on the $H$ dependence of the quasiparticle DOS. We will show that the $H$ dependence of the quasiparticle DOS at the Fermi level $N_0(H)$ is significantly influenced by the presence of disorder: While the DOS shows the $N_0(H) \propto H$ behavior in the alloyed samples, $N_0(H)$ shows significant deviation from the $N_0(H) \propto H$ behavior in the pure samples. We will discuss possible mechanisms of the deviations from the $H$-linear behavior and their relation with quasiparticle scattering.

Single crystals of Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C were grown by a floating zone method, while iodine-assisted chemical transport was employed for the growth of Nb$_{1-x}$Ta$_x$Se$_2$. Table I lists the superconducting and normal state parameters for the samples used in this study. As evident from the table, the pure samples, YNi$_2$B$_2$C and NbSe$_2$, are in the clean limit ($\xi \ll l$, where $l$ is the mean free path), while the alloyed samples, Y(Ni$_{0.8}$Pt$_{0.2}$)$_2$B$_2$C and Nb$_{0.8}$Ta$_{0.2}$Se$_2$, are in the intermediate range near the dirty limit ($\xi \approx l$). To determine $N_0(H)$ in magnetic fields, we have measured low-temperature specific heat ($C$) from 1.4 K to 20 K under magnetic fields up to 12 T using a thermal relaxation calorimeter. The relative resolution of the measurements was better than 0.5 %, and the absolute accuracy determined from the measurement of a Cu standard was better than 1 %.

Specific heat data for Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C under various magnetic fields parallel to the $c$ axis are shown in Fig. 1, plotted as $C/T$ versus $T^2$ for two samples (a) $x = 0.0$ and (b) $x = 0.2$. The measurements were performed with increasing temperature after field cooling from a temperature well above the superconducting transition temperature $T_c$. For the pure sample, a clear jump is seen in the zero-field data at $T_c = 15.4$ K. The upper critical field $H_{c2}(T)$ was determined from the specific heat data for a fixed $H$. The value at $T = 0$, $H_{c2}(0)$, was estimated by fitting the low-temperature data below 5 K to $H_{c2}(T) = H_{c2}(0)[1 - h(T/T_c)^2]$, where $h$ is a constant, giving an estimate of $H_{c2}(0) = 8.0$ T. This is consistent with a reported value of $H_{c2}(0) = 8.1$ T.

The least squares fit of the 9 T data, which represent the normal state specific heat, to $C = \gamma_N T + \beta T^3$ between 1.4 K and 5 K provides a Sommerfeld constant $\gamma_N = 20.6$ mJ/K$^2$mol. These values agree reasonably with those reported previously. For the $x = 0.2$ sample, we obtain...
$T_c = 12.1$ K, $\gamma_N = 14.7$ mJ/K²mol, and $H_{c2}(0) = 4.3$ T.

Under magnetic fields, a finite amount of $T$-linear term, $\gamma(H)T$, appears in the specific heat well below $T_c$ as shown in Fig. 1. The $\gamma(H)$ values at each field were estimated by extrapolating the $C/T$ versus $T^2$ data between 1.4 and 2.5 K linearly to $T = 0$. The zero-field value $\gamma(0)$ for the pure sample is negligibly small, indicative of the high quality of the sample. A small residual $\gamma(0)$ of about 0.4 mJ/K²mol is seen for the $x = 0.2$ sample, which very likely originates from a tiny amount of normal-state secondary phase. In order to see the magnetic field dependence, $\gamma(H)/\gamma_N$ is plotted in Fig. 3(a) as a function of $H/H_{c2}$. A substantial difference between the two samples is evident. For the pure sample, $\gamma(H)$ first shows a marked increase at low fields, then gradually approaches the normal state value. Notably, $\gamma(H)$ reaches a value almost half that of $\gamma_N$ at 1 T ($H/H_{c2} \approx 0.13$). We observed essentially the same $H$-dependence of $\gamma(H)$ in the field direction $H \perp c$. Analogous nonlinear dependence of $\gamma(H)$ was also observed for LuNi$_2$B$_2$C. [14] In contrast, for the $x = 0.2$ sample, $\gamma(H)$ increases linearly with $H$ as $\gamma(H) = \gamma(0) + \gamma_N H/H_{c2}$ over a wide field range below $H_{c2}$. $\gamma(H)$ does not show any field dependence above $H_{c2}$, indicating that this region is indeed in the normal state.

Qualitatively similar behavior for $\gamma(H)$ was observed in Nb$_{1-x}$Ta$_x$Se$_2$. Shown in Fig. 2 are the specific heat data for Nb$_{1-x}$Ta$_x$Se$_2$ under magnetic fields applied parallel to the $c$ axis. For the pure sample, $x = 0.0$, we obtained $T_c = 7.3$ K, $\gamma_N = 19.3$ mJ/K²mol, and $H_{c2}(0) = 4.4$ T. For the alloyed sample, $x = 0.2$, we obtained $T_c = 5.1$ K, $\gamma_N = 15.1$ mJ/K²mol, and $H_{c2}(0) = 3.2$ T. The magnetic field dependence of $\gamma(H)$ was estimated in the same manner as mentioned above, and is displayed in Fig. 3(b). A nonlinear behavior of $\gamma(H)$ is noticeable for the pure NbSe$_2$ sample, though not as significant as that observed in YNi$_2$B$_2$C. Similar nonlinear behavior of $\gamma(H)$ in the pure sample was reported by Sanchez et al. [3,14]

In contrast, for the alloyed sample, Nb$_{0.8}$Ta$_{0.2}$Se$_2$, $\gamma(H)$ exhibits a linear dependence on $H$ below $H_{c2}$ as observed in Y(Ni$_{0.8}$Pt$_{0.2}$)$_2$B$_2$C.

Since the number of vortices is scaled by $H$, the nonlinear behavior of $\gamma(H)$ in the pure superconductors implies that the DOS per single vortex depends on $H$ when the sample is clean. This contradicts the conventional picture that the vortex line should be viewed as a “rigid” normal electron cylinder, where the bound quasiparticles give rise to a $H$-independent DOS proportional to $N_0 \pi \xi^2$ per vortex line, where $N_0$ is the DOS in the normal state. However, once the electron mean free path $l$ is substantially reduced due to strong impurity scattering and $l \leq \xi < l \pi \xi^2$ intervortex distance $R$, the conventional picture appears to hold true. We point out that the unusual $\gamma(H)$ behavior and the strong influence of disorder are universally observed for $s$-wave superconductors, although the detailed $\gamma(H)$ data are available only for a limited number of compounds: A nonlinear dependence of $\gamma(H)$ was reported on a clean $s$-wave superconductor CeRu$_2$, where the deviation is compatible with that observed in NbSe$_2$. By contrast, a linear behavior was reported on a dirty superconductor Nb$_{77}$Zr$_{23}$. [6,17] Both $H$-linear and nonlinear $\gamma(H)$ were observed on V$_3$S$_5$. [6,17] which is very likely due to different degrees of disorder in the samples.

We suggest that the apparent breakdown of the “rigid” normal electron picture originates from the $H$-dependent core radius. In the conventional picture, the vortex core is assumed to be rigid. However, if the core radius is $H$ dependent, the quasiparticle DOS per vortex should also be $H$ dependent, resulting in a nonlinear $\gamma(H)$. Indeed, direct measurement of the local DOS in NbSe$_2$ by STS gives evidence for the shrinking of the core radius. [6] Using the experimentally observed $\gamma_N$ and $\gamma(H)$, we estimate the effective core radius $\rho_V$ by defining $2\pi \rho_V (H)^2 \gamma_N = \gamma(H)/(H/H_{c2})$, where $H_{c2}$ is the flux quantum. Note that $\rho_V (H_{c2}) = \xi(0)$. The variation of $\rho_V (H)$ estimated in this way is shown in the inset to Fig. 3, and compared with those determined by STS for NbSe$_2$. The $H$ dependence of $\rho_V$ from these two independent probes appears to agree reasonably with each other. The difference in the magnitudes should not be taken as significant since there is an arbitrariness of a factor in the definition of $\rho_V$.

The shrinking of the core radius may be supported by an anomalous temperature dependence of $H_{c2}(T)$ near $T_c$. In Fig. 4(a), $H_{c2}(T)$ curves deduced from the specific heat data are shown for the pure YNi$_2$B$_2$C where the deviation of $\gamma(H)$ from the linear behavior is most significant. A pronounced positive curvature is clearly seen near $T_c$. This may be understood in terms of the shrinking of $\rho_V$ (and hence $\xi$), which would lead to an enhancement of $H_{c2}(T)$ with increasing $H$ through the relation $H_{c2} = \Phi_0/2\pi \xi^2$. Indeed, from the suppressed initial slope at $T_c$, $dH_{c2}/dT|_{T_c} = -0.30$ T/K, we can obtain an estimate for $\xi(0) \approx 115$ Å using $\xi(0) = 0.54[\Phi_0/T_c(dH_{c2}/dT)|_{T_c}]^{1/2}$, which is substantially larger than $\xi(0) = 65$ Å, but close to the estimate of the enhanced $\rho_V \approx 130$ Å from $\gamma(H)$ at low magnetic fields limit (Fig. 3). Furthermore, as clearly seen in Fig. 4(a), the alloyed sample Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C does not show any noticeable positive curvature, consistent with the $H$-independent $\rho_V$. [8] A small positive curvature of $H_{c2}(T)$ near $T_c$ can be seen in the pure NbSe$_2$ as shown in Fig. 5(b) (See also ref. [8] and CeRu$_2$, [8]) which is consistent with the observation of a definite but less pronounced deviation from the $H$-linear $\gamma(H)$ in these materials.

By introducing a certain (repulsive) vortex-vortex interaction in the vortex lattice states, it was shown theoretically that the core radius indeed shrinks with increasing $H$. [4,5,19] The physical origin of the vortex interac-
tion is not clear yet. We speculate that this interaction is mediated by a coherent transfer of quasiparticles between the cores. The observation of dHvA oscillations in the mixed state of clean YNi$_2$B$_2$C and NbSe$_2$, where the cyclotron radius is by far larger than the core radius, clearly indicates the presence of delocalized and highly coherent quasiparticles outside the vortex cores. These delocalized quasiparticles may give rise to a certain vortex-vortex interaction and result in a shrinking of the core radius with increasing $H$. Then the critical sensitivity of the $\gamma(H)$ behavior can be naturally understood, since the coherent motion of these quasiparticles will be substantially suppressed by the impurity scatterings, when $l \sim \xi < R$. One may argue that the DOS associated with the delocalized quasiparticle itself may be the origin of nonlinear $\gamma(H)$ behavior. Hedø et al. recently proposed that the nonlinear behavior of $\gamma(H)$ in CeRu$_2$ originates from the contribution of delocalized quasiparticles with momentum $k$ perpendicular to $H$. The discussion is based on the theoretical study by Brandt et al., who showed that near $H_{c2}$, while excitations with $k$ parallel to $H$ have a gap, those in planes perpendicular to $H$ are gapless due to the formation of the vortex line lattice. However, the theory by Brandt can be applicable only near $H_{c2}$, and another theoretical approach with emphasis on the low field regime indicates that the local DOS outside the core is almost zero when $H/H_{c2} < 0.4$. [21]

Finally, we would like to comment on the pronounced nonlinear $\gamma(H)$ behavior in pure YNi$_2$B$_2$C compared with that of NbSe$_2$. We suspect that the presence of substantial gap anisotropy plays a role in YNi$_2$B$_2$C. The presence of a reduced gap region is suggested by the dHvA measurement. As seen in Fig. 1(a), the zero-field specific heat of pure YNi$_2$B$_2$C shows $T^3$ behavior rather than a thermally activated behavior down to low temperatures. This also supports the presence of a substantially small gap region on the Fermi surface, although $d$- or $p$-wave pairing is unlikely in YNi$_2$B$_2$C because of the insensitivity of the superconductivity to nonmagnetic Pt substitution. The reduced gap region on the Fermi surface may enhance quasiparticle transfer between the vortices and may result in the pronounced nonlinear $\gamma(H)$ in terms of the above speculative picture. In NbSe$_2$, zero-field specific heat exhibits thermally activated behavior at low temperatures, indicative of the presence of a relatively isotropic gap. This is consistent with the smaller deviation from the linear behavior of $\gamma(H)$ and positive curvature of $H_{c2}(T)$ in this compound.

In summary, we have measured the specific heat of Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C and Nb$_{1-x}$Ta$_x$Se$_2$ in the vortex state. Our results show that, in clean superconductors, the coefficient of the $T$-linear term in the specific heat, $\gamma(H)$, shows nonlinear behavior with $H$, suggesting that the DOS per vortex is $H$ dependent, while in dirty superconductors, $\gamma(H)$ is linear, suggesting a constant DOS per vortex. We suggest that these results may be understood in terms of the shrinking of the vortex core radius with increasing magnetic field. To strengthen this scenario, it is critically important to examine the $H$ dependence of $\rho_v$ in dirty superconductors by direct probes such as STS and $\mu$SR. Such attempts are now in progress.

We would like to thank K. Machida and M. Ichioka for valuable discussions. We acknowledge the critical reading of the manuscript by N. E. Hussey. This work was supported by a grant-in-aid of the Ministry of Education, Science, Sports, and Culture of Japan.

[1] C. Caroli, P. G. de Gennes and J. Matricon: Phys. Lett. 9 (1964) 307.
[2] G. E. Volovik: JETP Lett. 58 (1993) 469.
[3] W. Pesch and L. Kramer: J. Low Temp. Phys. 15 (1974) 367.
[4] A. A. Golubov and U. Hartmann: Phys. Rev. Lett. 72 (1994) 3602.
[5] J. E. Sonier et al.: Phys. Rev. Lett. 79 (1997) 1742.
[6] J. E. Graebner and M. Robbins: Phys. Rev. Lett. 36 (1976) 422.
[7] T. Terashima et al.: Phys. Rev. B 56 (1997) 5120.
[8] H. Takeya, T. Hirano and K. Kadowaki: Physica (Amsterdam) 256C (1996) 220.
[9] C. S. Oglesby et al.: J. Crystal Growth 137 (1994) 289.
[10] M. Nohara et al.: J. Phys. Soc. Jpn. 66 (1997) 1888.
[11] H. Takagi, M. Nohara and R. J. Cava: Physica (Amsterdam) 237-238B (1997) 292.
[12] H. Michor et al.: Phys. Rev. B 52 (1995) 16165.
[13] D. Sanchez et al.: Physica (Amsterdam) 204B (1995) 167.
[14] A. Junod: in Studies of High Temperature Superconductors, ed. A. Narlikar (Nova Science Publishers, New York, 1996) Vol. 19, p. 1.
[15] M. Hedo et al.: J. Phys. Soc. Jpn. 67 (1998) 272.
[16] A. P. Ramirez: Phys. Lett. A 211 (1996) 59.
[17] G. R. Stewart and B. L. Brandt: Phys. Rev. B 29 (1984) 3908.
[18] Recently, Shulga et al. discussed that the positive curvature of $H_{c2}(T)$ in YNi$_2$B$_2$C can be understood in terms of two bands. However, it is not clear whether their scenario can consistently explain the observed systematic correlation between $\gamma(H)$ and $H_{c2}(T)$. See, S. V. Shulga et al.: Phys. Rev. Lett. 80 (1998) 1730.
[19] M. Ichioka, A. Hasegawa and K. Machida: preprint.
[20] U. Brandt, W. Pesch and L. Tewordt: Z. Phys. 201 (1967) 209.
[21] M. Ichioka, N. Hayashi and K. Machida: Phys. Rev. B 55 (1997) 6565.
**FIG. 1.** Specific heat divided by temperature $C/T$ for (a) YNi$_2$B$_2$C and (b) Y(Ni$_{0.8}$Pt$_{0.2}$)$_2$B$_2$C, as a function of $T^2$ under various magnetic fields. The solid lines are guides to the eye.

**FIG. 2.** Specific heat divided by temperature $C/T$ for (a) NbSe$_2$ and (b) Nb$_{0.8}$Ta$_{0.2}$Se$_2$, as a function of $T^2$ under various magnetic fields. The solid lines are guides to the eye.

**FIG. 3.** The coefficient of $T$-linear specific heat at low temperatures normalized by the Sommerfeld constant, $\gamma(H)/\gamma_N$, as a function of reduced magnetic fields, $H/H_{c2}$, for (a) Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C and (b) Nb$_{1-x}$Ta$_x$Se$_2$. The solid lines are guides to the eye. The insets show the effective vortex core radius, $\rho_V$, determined by assuming $2\pi\rho_V \Delta^2 = \gamma(H)$, as a function of $H/H_{c2}$. $\rho_V$ of NbSe$_2$ determined by STS at $T/T_c = 0.33$ are shown (open squares) for comparison.

**FIG. 4.** Temperature dependence of the upper critical field $H_{c2}(T)$ for Y(Ni$_{1-x}$Pt$_x$)$_2$B$_2$C and Nb$_{1-x}$Ta$_x$Se$_2$. The solid lines are guides to the eye. The broken lines indicates the fit of the initial slope of $H_{c2}(T)$ at $T_c$. 

\[ H(0)/H_{c2} \]
I. TABLE

TABLE I. Superconducting and normal state parameters for the Y(Ni\(_{1-x}\)Pt\(_x\))\(_2\)B\(_2\)C and Nb\(_{1-x}\)Ta\(_x\)Se\(_2\) samples studied. The mean free path \(l\) was calculated from the resistivity \(\rho_0\) just above \(T_c\) by using \(l = \hbar/(3\pi^2)\^{1/3}/e^2n^{2/3}\rho_0\). We used an electron density of \(3 \times 10^{22} \, e/\text{cm}^3\) for Y(Ni\(_{1-x}\)Pt\(_x\))\(_2\)B\(_2\)C and \(1.6 \times 10^{22} \, e/\text{cm}^3\) for Nb\(_{1-x}\)Ta\(_x\)Se\(_2\). The Ginzburg Landau coherence length parallel to the ab plane, \(\xi(0)\), was determined from \(H_{c2}(0)\).

| \(x\) | \(T_c\) (K) | \(\rho_0\) (\(\mu\Omega\text{cm}\)) | RRR | \(\xi(0)\) (\(\text{Å}\)) | \(l\) (\(\text{Å}\)) |
|------|-----------|-----------------|-----|------|---------|
| Y(Ni\(_{1-x}\)Pt\(_x\))\(_2\)B\(_2\)C | 0.0 | 15.4 | 0.87 | 37.4 | 65 | 1500 |
| | 0.2 | 12.1 | 35.5 | 2.6 | 90 | 38 |
| Nb\(_{1-x}\)Ta\(_x\)Se\(_2\) | 0.0 | 7.3 | 4.7 | 20.0 | 85 | 450 |
| | 0.2 | 5.1 | 25.4 | 4.2 | 100 | 80 |