Magnetic Field Dependence of the London Penetration Depth of Bi₂Sr₂CaCu₂O_v

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London penetration depth λ_L was investigated as a function of dc magnetic field *H* in the Meissner state of Bi₂Sr₂CaCu₂O_y. At low temperatures, λ_L changes linearly in *H*, which is in contrast to the H^2 behavior observed in conventional superconductors. With increasing temperature, systematic rounding around zero field was observed. Together with the comparative study for a conventional superconductor V₃Si, it is suggested that the superconducting state of Bi₂Sr₂CaCu₂O_y is unconventional. These findings are the first confirmation of a recent new theoretical prediction for *d*-wave superconductors.

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The symmetry of the order parameter (or the energy gap) gives important information on the mechanism of superconductivity. For high- T_c cuprates, the study of the gap symmetry has been one of the most active fields in these materials [1,2]. Among various experimental methods, measurement of the detailed temperature dependence of the magnetic field penetration depth λ is one of the most popular ones, since it is a direct measure of superfluid density. If there are nodes in the gap, as was theoretically expected for strongly correlated materials [3–6], λ changes as T^n at low temperatures, in contrast to the thermally activated behavior for conventional *s*-wave superconductors. The power *n* depends on the type of the node in the **k** space [7].

Recently, high-resolution $\lambda(T)$ data on single crystals have been available by several groups. The current situation, however, has not converged. Maeda et al. observed T^2 behavior in Bi₂Sr₂CaCu₂O_y (Bi-2212) [8]. A similar data was also presented by Beasley [9]. He also observed the T^2 behavior in YBa₂Cu₃O_y (YBCO). In contrast, Hardy et al. observed T-linear $\lambda(T)$ at low temperatures in YBCO [10], and proposed that it is an expected behavior in pure crystals and consistent with a gap with line nodes. Furthermore, Bonn et al. observed a systematic change from T-linear to T^2 behavior with Zn doping in YBCO [11], which is also consistent with $d_{x^2-y^2}$ wave (line node) behavior, in some respects. On the other hand, thermally activated behaviors were reported in films of YBCO, but with a very small gap ($\Delta = 6 \text{ meV}$) [12], and in $(Nd, Ce)_2CuO_4$ [13]. The origin of the controversy has not been specified yet. Since the low-temperature behavior of λ is very sensitive to various extrinsic effects [14], it may not be easy to resolve the current puzzling situation until the ultrahigh quality single crystals are available for the high- T_c cuprates. Thus, a different approach is necessary for the gap study by the electromagnetic response.

Recently, Yip and Sauls (YS) calculated a small change of λ ($\Delta\lambda$) in the Meissner state of a *d*-wave superconduc-

tor by the application of dc magnetic field H [15]. They found that

$$\frac{\Delta\lambda(H,T)}{\lambda(0,T)} = \alpha(T)\frac{H}{H_0(T)},\tag{1}$$

in sharp contrast to the s-wave result [16],

$$\frac{\Delta\lambda(H,T)}{\lambda(0,T)} = \beta(T) \left[\frac{H}{H_0(T)}\right]^2,$$
(2)

where $\Delta\lambda(H,T) \equiv \lambda(H,T) - \lambda(0,T)$ is the change of λ by the application of H, H_0 is a characteristic field of the order of the thermodynamic critical field H_c . $\alpha(T)$ and $\beta(T)$ are factors which depend on the number of quasiparticles. Since the origin of the above mentioned change is a pair-breaking effect, for the s-wave case, reflecting the perfect gap opening, $\beta(T) \rightarrow 0$ for $T \rightarrow 0$. On the other hand, in the case of d-wave symmetry, $\alpha(T)$ remains finite (\sim 1, depending on the experimental configuration) even in the low-temperature limit. Based on this result, they proposed that the detailed study of $\lambda(H)$ can be a new candidate for the gap-symmetry study. Experimentally, prior to the YS prediction, the conventional H^2 dependence was reported in a single crystal of YBCO [17]. However, it suffered a criticism that at relatively high temperature the intrinsic behavior is smeared out by the thermal effect [15]. Furthermore, $\beta(T)$ in Ref. [17] is independent of temperature down to 10 K ($\sim 0.109T_c$) [18], which is contrary to the prediction for an s-wave superconductor. In addition, in principle, in the experimental configuration of Ref. [17] they measure the mixture of the in-plane penetration depth λ_{ab} and the out-of-plane λ_c . Therefore, a further detailed investigation is still needed. In this paper, we performed a systematic study of $\lambda(H)$ in Bi-2212 and found that the result is consistent with the YS prediction for the *d*-wave case. To our knowledge, this is the first confirmation of this new prediction.

Single crystals of Bi-2212 were prepared by a floating zone method. Typical dimensions are $1 \times 1 \times (0.02 -$

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0.05) mm³. They are carefully characterized by dc resistivity and magnetic-susceptibility measurement. The superconducting transition temperature T_c and the transition width ΔT_c are typically 90 and 1.3 K, respectively.

Penetration depth was measured at 45 MHz by an rf resonator method [19]. The sample was put in the solenoid and fixed by sapphire rods. What we are interested in is the in-plane penetration depth λ_{ab} . For Bi-2212, the λ_{ab} measurement is possible only in the configuration of $H_{\rm rf} \perp CuO_2$ [8]. Even for $H_{\rm rf} \perp CuO_2$, however, there may be a possible mixing of λ_c owing to the shape effect because of the large demagnetization effect $[1/(1 - \nu_{ab}) = 14-36$, depending on the sample dimensions, where ν_{ab} is the demagnetization factor]. In our data, the contribution of λ_c owing to the shape effect was considered to be negligible for the following two reasons: (i) We obtained the same results for several samples with different thickness; (ii) when comparing the data taken in $H_{\rm rf} \parallel {\rm CuO}_2$ and $H_{\rm rf} \perp {\rm CuO}_2$ configurations, both have quite different field dependence.

The change in the penetration depth $\Delta\lambda$ of the sample is related to the change in the resonating frequency of the circuit Δf as $\Delta\lambda = G\Delta f/f$, where f is the resonating frequency, and G is a geometric factor determined by the geometry of the sample and the solenoid. The magnitude of G was obtained by using Pb with the same dimension as that of the samples used in this study. For comparison with theories, zero-temperature value $\lambda_{ab}(0)$ is necessary. We obtained $\lambda_{ab}(0)$ by a microwave surface impedance measurement [20]. $\lambda_{ab}(0)$ was found to be 2600 Å, which is one of the best values of $\lambda_{ab}(0)$ of this material. The small value of $\lambda_{ab}(0)$ indicates that our crystals are free from macroscopic interlayer impurities.

dc magnetic field was applied by a solenoid made by ourselves and a current source. Magnetic field was calibrated by using a Hall probe with a resolution of 0.2 G. To cancel a residual field (~0.4 G), a small external counter field was applied during the cooling process [21]. Although we have not measured the actual residual field after this procedure, the main experimental result presented here did not depend on whether this procedure was made or not. Therefore, the residual flux, *if any*, cannot be a dominant origin of the main result presented below. During each field sweep, fluctuation of temperature was maintained within ± 10 mK, which limits the effective sensitivity of our $\lambda(H)$ measurement ($\delta \lambda \sim 20$ Å).

Figure 1 shows the field dependence of the change in λ_{ab} ; $\Delta \lambda_{ab}(H,T)$ for $H \perp CuO_2$ at 60 K. $\Delta \lambda_{ab}(H,T)$ is reversible for the small sweep amplitude within an experimental error. On the other hand, when a maximum field of the sweep exceeds a characteristic field H^* , field variation clearly shows a hysteresis, and $\Delta \lambda(H)$ shows a structure at H^* . For a higher field H_r , $\Delta \lambda_{ab}(H)$ becomes reversible again. In fact, $H_r(T)$ agrees with the so-called irreversibility line for a crystal from the same batch [22].



FIG. 1. Change in the in-plane penetration depth λ_{ab} of a Bi-2212 single crystal as a function of dc magnetic field at 60 K. The origin of each curve was shifted. The inset shows the experimental configuration of λ_{ab} measurement.

Therefore, we identify H_r as the irreversibility field. On the other hand, H^* is a field which gives another boundary between reversible and irreversible region. Thus, we can regard H^* as the measure of the lower critical field H_{c1} . In fact, H^* increases gradually with decreasing temperature [8], and takes 6 G at 10 K for the sample shown in Fig. 1. Considering the surface barrier effect [22], this value is reasonable [23].

Next, we will concentrate on $\Delta \lambda_{ab}(H)$ below H^* in Fig. 1. At low temperatures, $\Delta \lambda(H, T)$ shows a V-shaped curve, which means $\Delta \lambda(H, T)$ changes linearly in H. To see this in more detail, the data below H^* are expanded. Figure 2(a) shows the normalized change in λ_{ab} ; $\ell \equiv \Delta \lambda_{ab}(H,T)/\lambda_{ab}(0,T)$, as a function of the normalized field $h = H/H_0$ at various temperatures. H_0 was defined as $H_0 \equiv H^*(\sqrt{2\kappa}/\log\kappa) \sim (1 - \nu_{ab})H_c$, where κ is the GL parameter. We assumed $\kappa \sim 100$. The temperature



FIG. 2. (a) $\ell \equiv \Delta \lambda_{ab}(H,T)/\lambda_{ab}(0,T)$ as a function of the normalized field $h \equiv H/H_0$ at various temperatures. Temperatures are also normalized to T_c ; $t \equiv T/T_c$. (b) ℓ as a function of h^2 .

was also normalized to T_c . At low temperatures, ℓ behaves linearly in h, as was already seen in Fig. 1. However, around zero field, a very small rounding was observed. This rounding becomes more pronounced with increasing temperature. We also show the same data as a function of h^2 in Fig. 2(b). In Fig. 2(b), the data at each temperature show the concave downward behavior, which clearly demonstrates that ℓ of Bi-2212 contains lower order terms than two in h. We also preformed the $\ell(h)$ vs logh plot, and confirmed the crossover from h^2 to h with increasing h. Therefore, the best description of $\ell(h)$ is as follows. $\ell(h)$ is linear in h except around zero field. Around zero field, $\ell(h)$ is quadratic in h. With increasing temperature, the quadratic region becomes prominent.

Figure 3 shows the temperature dependence of $d\ell/dh$. The derivative was taken at fields where ℓ behaves linearly in *h*. Therefore, in terms of the YS theory, $d\ell/dh$ is just equal to $\alpha(T)$ in Eq. (1). The main reason for large error bars is the uncertainty in determining H^* . Although $d\ell/dh(T)$ is complicated, $d\ell/dh$ is very weakly temperature dependent below 40 K, and remains ~1.9 at 10 K (0.12 T_c). $d\ell/dh(T)$ of 1.9 even at a low temperature of $0.12T_c$ suggests that a large number of quasiparticles of $0.12T_c$ are present down to this temperature.

The *H*-linear $\Delta\lambda$, the systematic rounding around zero field, and $d\ell/dh$ of the order of unity—all of these observations are consistent with the theoretical prediction for a *d*-wave superconductor by Yip and Sauls [15], except for some points discussed later.

We also performed the same measurement for a single crystal of V₃Si, which is a representative of *s*-wave type-II superconductors. The result was shown in Fig. 4, together with the data in Bi-2212 taken at a similar normalized temperature. In V₃Si, ℓ clearly behaves as h'^2 ($h' \equiv H/H^*$). On the other hand, ℓ of Bi-2212 shows a concave downward behavior, as was already shown in Fig. 2(b). We also investigated the temperature dependence of ℓ in V₃Si. Since our measurement system is operative above 7 K, only the data above 0.43 T_c are available. The H^2 behavior was observed in the whole



FIG. 3. The temperature dependence of $d\ell/dh$. See the text for details.



FIG. 4. $\Delta \lambda_{ab}(H,T)/\lambda_{ab}(0,T)$ of V₃Si, and Bi-2212 as a function of $(H/H^*)^2$. The dotted straight lines are guides for the eye.

temperature region investigated in V₃Si, and β is a weakly decreasing function of temperature down to this temperature. Detailed data on this material will be shown in a separate publication. Thus, also from the comparison with conventional superconductors, the *H*-linear $\lambda_{ab}(H)$ observed in Bi-2212 is indicative of a *d*-wave gap rather than an *s*-wave gap in terms of the YS theory.

However, several problems should be discussed. The first one is on the field range where the H-linear behavior was observed. According to YS, the H-linear behavior should become remarkable only above the field $H \sim cH_0(T/T_c)$, where c is $2\pi k_B T_c/3\Delta_0$, and Δ_0 is a maximum of the gap at T = 0 K. In the weak-coupling case, c is estimated to be 0.6 [15]. In the Fig. 2, however, the H-linear behavior is observed clearly from the much smaller field. For high- T_c cuprates, it is often reported that $2\Delta/k_BT_c$ takes the value which is far from the weak coupling value. In particular, several photoemission [2,24] and tunneling measurements [25] reported $2\Delta/k_BT_c \simeq 7-8$. This makes c smaller by a factor of 2– 3. Another possible origin which can decrease effective magnitude of c is the effect of the surface barrier. In some samples of Bi-2212 crystals synthesized by us, the apparent H_{c1} is twice larger than that without the surface barrier [22]. Considering these, the effective c may become ~ 0.1 or much less. This is just the field range where we observed the H-linear behavior.

It should be also discussed if it is possible to obtain *H*-linear $\Delta\lambda(H)$ in samples where T^2 behavior was observed in $\Delta\lambda(T)$ [8,9]. In terms of *d*-wave theories, the T^2 - $\lambda(T)$ comes from the disorder-induced pair breaking, which can also smear out the singularity in $\lambda(H)$ at H = 0. If the T^2 - $\Delta\lambda(T)$ in Bi-2212 is due to the disorder-induced pair breaking, the scenario based on the resonant scattering [26] is necessary [11]. In this theory, the crossover temperature T^* from T^2 - to *T*-linear $\Delta\lambda(T)$ is given by $T^* = 0.83(\Gamma\Delta_0)^{1/2} = 0.83a(\Gamma/\Delta_0)^{1/2}T_c$, where Γ is the scattering parameter, and $a \equiv \Delta_0/T_c$. If we take a = 4, $T^*/T_c = 3.3(\Gamma/\Delta_0)^{1/2}$. On the other hand, for the crossover field H_{τ} from H^{2-} to H-linear $\Delta\lambda(H)$, we obtain $H_{\tau}/H_0 = 0.21(\Gamma/\Delta_0)^{1/2}$, since the new energy scale for smearing out the singularity is $\gamma \equiv 0.63(\Gamma\Delta_0)^{1/2}$, instead of πT . With $T^*/T_c \sim 0.4$, we obtain $\Gamma/\Delta_0 = 0.015$ and $H_{\tau}/H_0 \sim 2.5 \times 10^{-2}$, which is consistent with the data in Fig. 2. Thus, the linear $\lambda(H)$ is observable even when the $T^2 - \Delta\lambda(T)$ is observed.

The next question is, "Is it consistent that the H^2 behavior in YBCO [17] and *H*-linear behavior in Bi-2212 are observed in the same normalized temperature region?". One possibility is that $\Delta \lambda_{ab}(H)$ is really different between YBCO and Bi-2212. Another possibility is that YBCO may also show the similar behavior to our Bi data, if we compare the data within the same criteria as shown in Fig. 2. As was mentioned above, even the data of Ref. [17] are inconsistent with the *s*-wave gap. Thus, $\Delta \lambda(H)$ in YBCO deserves further investigation.

Another problem is the complicated temperature dependence of $d\ell/dh [\alpha(T)]$. However, since the form of $\alpha(T)$ in Eq. (1) has not been given in [15], detailed comparison between the experimental results and the theory is impossible in the present stage.

We should also discuss the extrinsic effects owing to the short coherence length. The effects of the crystal imperfections and the motion of the vortices related to them on $\lambda_L(H)$ were discussed in detail by Halbritter [14]. According to Ref. [14], there are two characteristic fields; the intergrain-weak-link field H_1 (~1 G) and the intragrain-weak-link field H_2 (~100 G). For $H \le H_1$; $\lambda(H) \propto H^2$. For $H_1 \leq H \leq H_2$, $\Delta\lambda(H)/\lambda(0) = b(H/H_0)$, with a clear hysteresis. b is of the order of 100. Above H_2 , λ behaves linearly in H, again, with a smaller slope. Our data are strongly different from the prediction of Ref. [14] in the following points: First, we have never observed a clear hysteresis in the H-linear region. Second, the magnitude of b is highly different from our data by two orders of magnitude. Thus, we believe that these extrinsic effects are not the origin of $\lambda(H)$ in our experiment.

In conclusion, by the high-resolution $\lambda_{ab}(H,T)$ measurement in single crystals of Bi-2212, it was found that $\Delta \lambda_{ab}(H,T)$ behaves linearly in H. Together with the comparative measurement in a conventional superconductor V_3 Si, we conclude that the *H*-linear behavior is strongly indicative of the unconventional superconducting state in the Bi-2212 system. To our knowledge, this is the first confirmation of the new theoretical prediction for unconventional superconductors. Since the origin of the Hlinear $\Delta \lambda$ is the singularity at H = 0 due to the existence of nodes in the gap in terms of the YS theory, various types of the unconventional states other than the *d*-wave state may also lead to the similar behavior. Thus, we cannot specify what type of unconventional state is realized in the Bi cuprate. To answer this question, further studies are necessary, both experimentally and theoretically.

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