

Linear temperature dependence of lower critical field in MgB₂

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The lower critical field H_{c1} has been carefully measured on a well-shaped cylindrical dense sample of the new superconductor MgB₂ fabricated by high-pressure synthesis. The penetration depth λ is calculated from the H_{c1} data. It is found that a linear relation of $H_{c1}(T)$ appears in the whole temperature region below T_c . Furthermore, a finite slope of dH_{c1}/dT and $d\lambda(T)/dT$ remains down to the lowest temperature (2 K). These are inconsistent with the expectation for an isotropic s -wave superconductivity in MgB₂.

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I. INTRODUCTION

Recently discovered superconductor MgB₂ has generated a new round of interest in the field of superconductivity.¹ Many important properties have already been derived, such as the upper critical field $H_{c2} = 15\text{--}20.4$ T,²⁻⁵ the Ginzburg-Landau parameter $\kappa \approx 26$,⁵ and the flux dynamics and vortex phase diagram.⁵⁻¹¹ Recently, a Hall-effect measurement has been done showing that the charge carriers are holes with very high density $n_s > 1.5 \times 10^{23}$ cm⁻³.¹² Elegant transport measurement¹³ on the MgB₂ bulk sample shows also an anomalous Hall effect, which is similar to that of the high-temperature superconductors (HTS). Measurement on the Seebeck coefficient¹⁴ signals also positive charges in MgB₂. Theoretically there are two major diverse pictures accounting for the superconductivity.^{15,16} Based on band calculations, Kortus *et al.*¹⁵ proposed that the superconductivity results from the strong electron-phonon interaction and the high phonon frequency associated with the light B element. This picture is supported by the recent isotope measurement on ¹⁰B and ¹¹B yielding an isotope exponent $\alpha \approx 0.26$.¹⁷ In the second picture, Hirsch¹⁶ suggested an alternative explanation, which conjectures the same mechanism of superconductivity in MgB₂ as in high-temperature cuprate superconductors. Recently high-pressure measurement¹⁴ shows that the superconducting transition temperature decreases linearly at a rate of $dT_c/dP = -1.6$ K/GPa, in contrast to that predicted by Hirsch's theory. In order to know the superconducting mechanism, probing the nature of the superconducting gap and symmetry seems to be very important. Recent data from specific heat,¹⁸ tunneling spectroscopy,¹⁹⁻²¹ and NMR study²² commonly suggest a phonon-mediated BCS picture with the isotropic s -wave superconducting energy gap Δ ranging from 2 to 8 meV corresponding to the coupling strength from weak to strong. It is important to point out that all these are based on some kind of fitting with parameters to the experiment data. For an isotropic s -wave superconductor, it needs a minimum energy to excite the charges from the superconducting condensate to the excited states. Thus one can expect an exponentially activated temperature dependence in the low-temperature region for many thermodynamic parameters, such as the lower critical field H_{c1} and penetration depth λ , etc. Therefore one should observe vanishing slopes $dH_{c1}(T)/dT = 0$ and $d\lambda(T)/dT = 0$ in the low-

temperature region. In this paper, we present the determination of the lower critical field H_{c1} in the whole temperature region *without any fitting parameters*. Our data show a linear temperature dependence of the lower critical field, which is clearly in contrast to an isotropic s -wave superconductivity in MgB₂.

II. EXPERIMENT

Samples of MgB₂ investigated here were fabricated by high-pressure synthesis ($P = 6$ GPa at 950 °C for 0.5 h) described in detail elsewhere.²³ The x-ray-diffraction analysis shows that the major diffraction peaks are from the MgB₂ phase. Our samples are very dense with a metallic shiny surface after polishing. The calculated density is about 2.2 g/cm³, which is very close to the ideal density 2.4 g/cm³. The dc magnetic measurements were carried out by a vibrating sample magnetometer (VSM), Oxford 3001, and a superconducting quantum interference device (SQUID), Quantum Design MPMS 5.5 T. During the magnetic measurements, the sample axis was always aligned along the field direction.

III. RESULTS

Figure 1 shows the resistive transition of one plate sample cut from the same high-pressure synthesized bulk as our cylindrical sample. The resistive transition shows that the onset

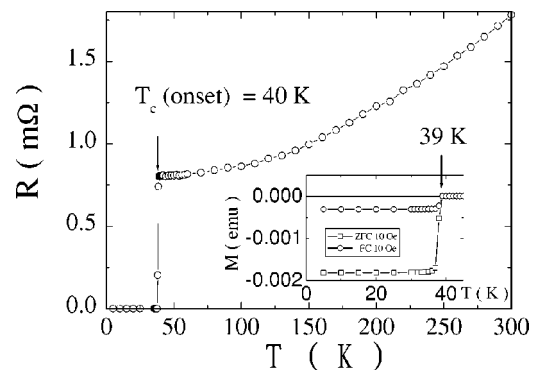


FIG. 1. The resistance measurement of MgB₂. The onset T_c is 40 K, and the transition width is less than 1 K. The inset gives the ZFC and FC diamagnetization at 5 Oe.

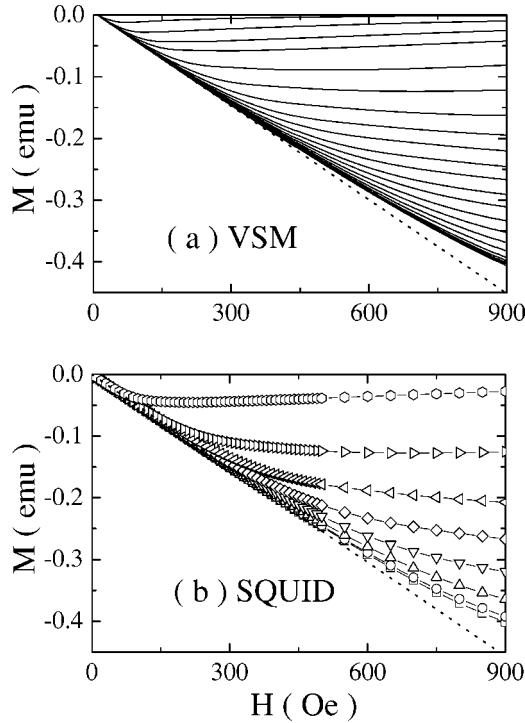


FIG. 2. The magnetization curve $M(H)$ measured by using (a) VSM and (b) SQUID. The dotted line is the “Meissner line” defined in the text. The temperature is selected as (a) 2–6 K, step 1 K; 8–32 K, step 2 K; 34–38 K, step 1 K; and (b) 2 K, 5–35 K, step 5 K. It is very clear that the initial slope of all the curves is the same.

T_c^{onset} is 40 K and the transition width is less than 1 K. The inset of Fig. 1 shows the diamagnetic transition of the cylinder investigated in this work. The $M(T)$ curves measured in both the field-cooled (FC) and zero-field-cooled (ZFC) processes reveal a sharp transition. To minimize the demagnetization factor, the sample was carefully cut and ground to a cylinder with a diameter of 1.58 mm and 3.50 mm in length. The demagnetization factor was about 0.15 when the field was applied along the axis of the cylinder. After zero-field cooled from 50 K to a desired temperature, the magnetic field was applied slowly up to 900 Oe, which is much higher than H_{c1} . Figures 2(a) and 2(b) show the dc magnetization curves obtained by VSM and SQUID, respectively. All curves show clearly the common linear dependence of the magnetization on field caused by the Meissner effect at low fields.

IV. DISCUSSION

The value of H_{c1} is determined by examining the point of departure from linearity on the initial slope of the magnetization curve. To do this, a linear fit with points between 10 Oe and 100 Oe at 2 K is made. This line passes also through the data points measured at other temperatures in the low-field region. The fitted linear line is called the “Meissner line” (ML), shown as the dotted line in Fig. 2, which represents the magnetization curve of Meissner state. The results of subtracting this ML from magnetization curves of SQUID measurements are plotted in Fig. 3. The data from the VSM

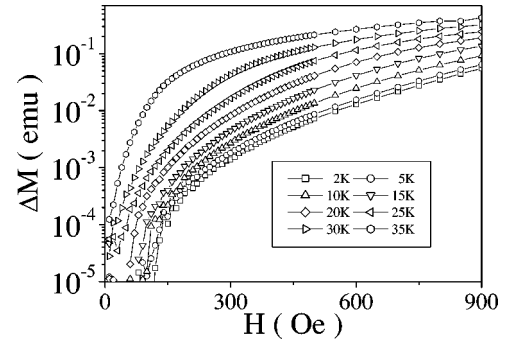


FIG. 3. The difference between the linear line and the $M(H)$ curves measured on the SQUID. ΔM is shown in logarithmic scale. All curves show a fast drop when the real H_{c1} is approached. By using a proper criterion, e.g., 10^{-3} emu, H_{c1} is easily obtained.

bear close resemblance to that from the SQUID, thus they are not shown here. The value of H_{c1} is easily obtained by choosing a proper criterion of ΔM . In Fig. 4(a), we plot the H_{c1} acquired by using criteria of $\Delta M = 1 \times 10^{-3}$ and 10^{-4} emu, respectively. Because of the low precision of the VSM, there is only the SQUID result when choosing the 10^{-4} emu criterion. If not specially mentioned, the data based on the criterion of $\Delta M = 1 \times 10^{-3}$ emu is chosen hereinafter. The H_{c1} measured by VSM and SQUID coincide

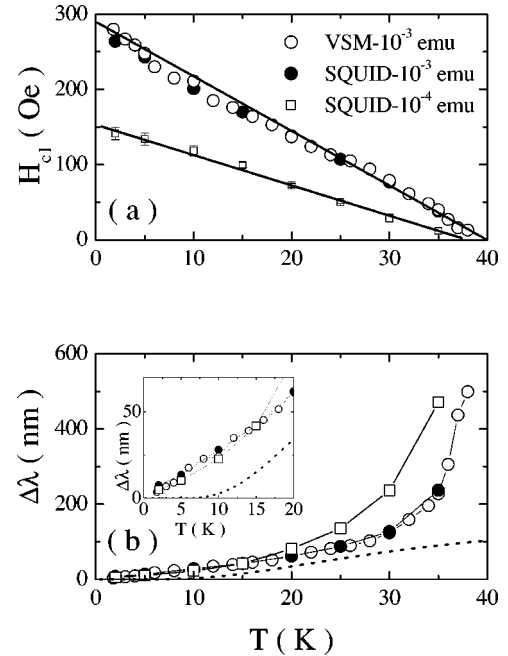


FIG. 4. The temperature dependence of the (a) nominal H_{c1} and (b) $\Delta\lambda$. The circles and square symbols are obtained by using criteria of $\Delta M = 1 \times 10^{-3}$ emu and $\Delta M = 10^{-4}$ emu, respectively. For the criterion of $\Delta M = 10^{-4}$ emu, error bars are given for determining the nominal H_{c1} . The low-temperature part is enlarged in the inset of (b), which shows a clear linear T dependence of $\Delta\lambda$. The dotted line in (b) and its inset is a theoretical calculation using Eq. (2). Because there is no consistent value of the energy gap Δ yet, we choose an intermediate value, $\Delta = 5$ meV. It should be noted here that the value of Δ only affects the magnitude of $\Delta\lambda$ and does not change the low-temperature behavior of $\Delta\lambda$.

in the same line, showing that our results are independent of measuring device and technique. Principally by extrapolating the nominal $H_{c1}(T)$ curve to zero temperature, we can get $H_{c1}(0)$. Since the value of $H_{c1}(T)$ determined here is criterion dependent, it is difficult to have a comparison with the values derived by other authors. However, from our data we can conclude that the effective first flux penetration occurs at an applied field H_a of about 150 Oe to 300 Oe depending on the criterion. At an even lower criterion, since ΔM drops sharply with decreasing magnetic field, therefore the $H_{c1}(T)$ determined will not be far from that determined with the criterion of 10^{-4} emu. Taking the demagnetization factor (0.15) into account, we find that $H_{c1}(0) = 150/(1-0.15) \approx 176$ Oe at the criterion of 10^{-4} emu. A striking feature here is that the nominal $H_{c1}(T)$ (without correcting to the demagnetization factor) shows an approximate linear behavior in the whole temperature region as shown by the straight line in Fig. 4(a). This approximate linear behavior holds also for the nominal $H_{c1}(T)$ determined with other criteria, such as $\Delta M = 10^{-4}$ emu.

In order to investigate the temperature dependence of the penetration depth λ in the low-temperature region, we use the expression

$$H_{c1} = \frac{\Phi_0}{4\pi\lambda^2} \ln \kappa, \quad (1)$$

where $\Phi_0 = hc/2e \approx 2 \times 10^{-7}$ G cm² is the flux quantum, and $\kappa \approx 26$ (Ref. 5) is the Ginzburg-Landau parameter. Using $H_{c1}(0) = 176$ Oe obtained above, we find $\lambda(0) \approx 1700$ Å. The deviation $\Delta\lambda(T)$ of the penetration depth from its zero-temperature value $\Delta\lambda = \lambda(T) - \lambda(0)$ is calculated from the data of H_{c1} and shown in Fig. 4(b). Here we take $\ln \kappa$ as a constant since κ is a weak temperature-dependent function. It is remarkable that the dependence of $\Delta\lambda$ on temperature below 20 K ($T_c/2$) is close to a linear relation, i.e., $\Delta\lambda \propto T$, which is more explicit in the inset.

One may argue that the nominal H_{c1} relation obtained in our experiment may not reflect the true H_{c1} but the flux entry field because of the Bean-Livingston surface barrier.²⁴ But it is clear that the influence of surface barrier is not important in our cylindrical sample because the magnetic hysteresis loops are very symmetric even up to 35 K. Another strong point against this argument is that an extremely small and unreasonable H_{c1} will be obtained when following the scenario of the Bean-Livingston surface barrier. As presented in Ref. 24, the entry field $H_s = \Phi_0/4\pi\lambda\xi = H_{c1}\kappa/\ln \kappa$, which is about eight times larger than H_{c1} assuming $\kappa \sim 26$. Therefore, the true $H_{c1}(0)$ would be about 22 Oe and $\lambda(0) \approx 5400$ Å at 0 K if the surface barrier is very important, which contradicts most experiments.^{4,5} The reason for not seeing the importance of the Bean-Livingston barrier in our present sample may be that the surface of our sample, although polished, is not smooth at all leading to a very weak image attractive force.

Another question would concern whether the H_{c1} determined here is the lower critical field of grains because of the polycrystalline nature of our sample. As we mentioned previously our present sample is a high-pressure synthesized

sample with very high density. Therefore, H_{c1} measured here is not the granular but the bulk property. Recently, muon spin rotation (μ SR) and ac susceptibility measurements²⁵ as well as microwave surface resistance measurements²⁶ also gave results consistent with ours.

In order to know whether this linear behavior is induced by the uncertainty in using the criterion for ΔM , we have used different criteria, such as 10^{-4} emu, for the determination of H_{c1} , but the linear dependence is not changed. This linear dependence contradicts the prediction for an isotropic s -wave superconductivity supported by some experiments mentioned in the first paragraph of this paper. For an isotropic s -wave superconductor, the finite-energy gap manifests itself with an exponentially activated temperature dependence of many thermodynamic properties. In the isotropic s -wave BCS theory, $\Delta\lambda$ is given by²⁷

$$\frac{\Delta\lambda(T)}{\lambda(0)} \sim 3.3 \left(\frac{T_c}{T}\right)^{1/2} \exp\left(-\frac{\Delta}{k_B T_c} \frac{T_c}{T}\right), \quad (2)$$

where Δ is the energy gap. The exponential term causes the very weak temperature dependence of $\Delta\lambda(T)$, i.e., $d\lambda(T)/dT \approx 0$ in the low-temperature region as shown by the dotted line in the inset of Fig. 4(b). It is easy to see that the linear dependence of $\Delta\lambda(T)$ versus T shown in Fig. 4(b) and its inset is clearly different from the prediction by the isotropic s -wave BCS theory. The finite slope of $H_{c1}(T)$ and $\lambda(T)$ in the low-temperature region found here may be understood in the frame of an anisotropic energy gap.^{13,28,29} There was an argument contributing this linearity to the superconducting phase fluctuation.³⁰ This possibility can be however ruled out by the observation of a very weak critical fluctuation effect in MgB₂ characterized by the nonrounded onset superconducting transitions measured at different fields.

This linear behavior is similar to that in the YNi₂B₂C system³¹ and in high-temperature superconductors (HTS). It may also strongly relate to the approximate linear dependence of upper critical field $H_{c2}(T)$ in the wide temperature region for MgB₂ as recently observed by experiment² and calculated by Haas and Maki³² by assuming an anisotropic s -wave gap symmetry. In cuprate superconductors, the finite slope of $d\lambda(T)/dT$ at 0 K has been ascribed to the d -wave symmetry^{33,34} of the energy gap based on the consideration as follows. In a pure d -wave superconductor, the energy gap along the node directions ($k_x = \pm k_y$) vanishes and the spectrum $N_s(E)$ of quasiparticle excitations in the superconducting phase is gapless, which results in the linear dependence of $N_s(E)$ on E at low excitation energies. A finite temperature will excite certain quasiparticles leading to a linear dropping of the superfluid density ρ_s with temperature. As to other p -wave superconductors, $\Delta\lambda$ also shows nonexponential dependence on T because of the nodes of the order parameter.³⁵ The linear $H_{c1}(T)$ measured in MgB₂ may not imply d -wave or p -wave superconductivity, but calls certainly for a new understanding. For example, according to the theory by Hirsch,¹⁶ it appears to be the same ‘‘hole superconductivity’’ in HTS and MgB₂. Theorists may get an answer to the question of whether the finite slope of $H_{c1}(T)$

and $\Delta\lambda(T)$ at low temperatures for HTS, $\text{YNi}_2\text{B}_2\text{C}$, and MgB_2 is due to the same cause.³⁶

V. CONCLUSIONS

In conclusion, we have measured the M - H curve of a cylindrical MgB_2 sample and obtained its lower critical field $H_{c1}(T)$. A striking linear curve $H_{c1}(T)$ has been observed in the whole temperature region. Further calculation of the penetration depth shows also a finite slope in the low-temperature limit. All these are at odds with the expectations

for an isotropic s -wave superconductivity. The reason is unknown but warrants certainly further investigation in order to get a final consensus of the order-parameter symmetry of MgB_2 .

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- ¹J. Nagamatsu, N. Nakagawa, T. Maranaka, Y. Zenitani, and J. Akimitsu, *Nature (London)* **410**, 63 (2001).
- ²S.L. Bud'ko, C. Petrovic, G. Lapertot, C.E. Cunningham, P.C. Canfield, M.H. Jung, and A.H. Lacerda, *Phys. Rev. Lett.* **63**, 220503(R) (2001).
- ³P.C. Canfield, D.K. Finnemore, S.L. Bud'ko, J.E. Ostenson, G. Lapertot, C.E. Cunningham, and C. Petrovic, *Phys. Rev. Lett.* **86**, 2423 (2001).
- ⁴Y. Takano, H. Takeya, H. Fujii, H. Kumakura, T. Hatano, K. Togano, H. Kito, and H. Ihara, *cond-mat/0102167* (unpublished).
- ⁵D.K. Finnemore, J.E. Ostenson, S.L. Bud'ko, G. Lapertot, and P.C. Canfield, *Phys. Rev. Lett.* **86**, 2420 (2001).
- ⁶H.H. Wen, S.L. Li, Z.W. Zhao, Y.M. Ni, Z.A. Ren, G.C. Che, H.P. Yang, Z.Y. Liu, and Z.X. Zhao, *Chin. Phys. Lett.* **18**, 816 (2001).
- ⁷H.H. Wen, S.L. Li, Z.W. Zhao, Y.M. Ni, Z.A. Ren, G.C. Che, H.P. Yang, Z.Y. Liu, and Z.X. Zhao, *cond-mat/0103521* (unpublished).
- ⁸J.R. Thompson, M. Paranthaman, D.K. Christen, K.D. Sorge, H.J. Kim, and J.G. Ossandon, *cond-mat/0103514* (unpublished).
- ⁹M.J. Qin, X.L. Wang, H.K. Liu, and S.X. Dou, *cond-mat/0104112* (unpublished).
- ¹⁰Marc Dhalle, Pierre Toulemonde, Concetta Beneduce, Nicolas Musolino, Michel Decroux, and Rene Flukiger, *cond-mat/0104395* (unpublished).
- ¹¹S. Patnaik, L.D. Cooley, A. Gurevich, A.A. Polyanskii, J. Jiang, X.Y. Cai, A.A. Squitieri, M.T. Naus, M.K. Lee, J.H. Choi, L. Belenky, S.D. Bu, J. Letteri, X. Song, D.G. Schlom, S.E. Babcock, C.B. Eom, E.E. Hellstrom, and D.C. Larbalestier, *cond-mat/0104562* (unpublished).
- ¹²W.N. Kang, C.U. Jung, Kijoon H.P. Kim, Min-Seok Park, S.Y. Lee, Hyeong-Jin Kim, Eun-Mi Choi, Kyung Hee Kim, Mun-Seog Kim, and Sung-Ik Lee, *cond-mat/0102313* (unpublished).
- ¹³R. Jin, M. Paranthaman, H.Y. Zhai, H.M. Christen, D.K. Christen, and D. Mandrus, *cond-mat/0104411* (unpublished).
- ¹⁴B. Lorenz, R.L. Meng, and C.W. Chu, *Phys. Rev. B* **64**, 012507 (2001).
- ¹⁵J. Kortus, I.I. Mazin, K.D. Belashchenko, V.P. Antropov, and L.L. Boyer, *Phys. Rev. Lett.* **86**, 4656 (2001).
- ¹⁶J.E. Hirsch, *cond-mat/0102115* (unpublished).
- ¹⁷S.L. Bud'ko, G. Lapertot, C. Petrovic, C.E. Cunningham, N. Anderson, and P.C. Canfield, *Phys. Rev. Lett.* **86**, 1877 (2001).
- ¹⁸R.K. Kremer, B.J. Gibson, and K. Ahn, *cond-mat/0102432* (unpublished).
- ¹⁹G. Rubio-Bollinger, H. Suderow, and S. Vieira, *Phys. Rev. Lett.* **86**, 5582 (2001).
- ²⁰G. Karapetrov, M. Iavarone, W.K. Kwok, G.W. Crabtree, and D.G. Hinks, *Phys. Rev. Lett.* **86**, 4374 (2001).
- ²¹A. Sharoni, I. Felner, and O. Millo, *Phys. Rev. B* **63**, 220508 (2001).
- ²²H. Kotegawa, K. Ishida, Y. Kitaoka, T. Muranaka, and J. Akimitsu, *cond-mat/0102334* (unpublished).
- ²³Z.A. Ren, G.C. Che, Z.X. Zhao, H. Chen, C. Dong, Y.M. Ni, S.L. Jia, and H.H. Wen, *Chin. Phys. Lett.* **18**, 589 (2001).
- ²⁴P. G. Gennes, *Superconductivity of Metals and Alloys* (Addison-Wesley, New York, 1989), p. 79.
- ²⁵C. Panagopoulos, B.D. Rainford, T. Xiang, C.A. Scott, M. Kambara, and I.H. Inoue, *cond-mat/0103060* (unpublished).
- ²⁶A.A. Zhukov, K. Yates, G.K. Perkins, Y. Bugoslavsky, M. Polichetti, A. Berenov, J. Driscoll, A.D. Caplin, L.F. Cohen, Ling Hao, and J. Gallop, *cond-mat/0103587* (unpublished).
- ²⁷B. Muhlshlegel, *Z. Phys.* **155**, 313 (1959).
- ²⁸C.T. Chen, P. Seneor, N.C. Yeh, R.P. Vasquez, C.U. Jung, Min-Seok Park, Heon-Jung Kim, W.N. Kang, and Sung-Ik Lee, *cond-mat/0104285* (unpublished).
- ²⁹F. Simon, A. Jánossy, T. Fehér, F. Murányi, S. Garaj, L. Forró, C. Petrovic, S.L. Bud'ko, G. Lapertot, V.G. Kogan, and P.C. Canfield, *Phys. Rev. Lett.* **87**, 047002 (2001).
- ³⁰E. Roddick and D. Stroud, *Phys. Rev. Lett.* **74**, 1430 (1995).
- ³¹S. Oxx, D.P. Choudhury, B.A. Willemsen, H. Srikanth, S. Sridhar, B.K. Cho, and P.C. Canfield, *Physica C* **264**, 103 (1996).
- ³²S. Haas and K. Maki, *cond-mat/0104207* (unpublished).
- ³³W.N. Hardy, D.A. Bonn, D.C. Morgan, R.X. Liang, and K. Zhang, *Phys. Rev. Lett.* **70**, 3999 (1993).
- ³⁴D.A. Bonn, S. Kamal, K. Zhang, R.X. Liang, D.J. Baar, E. Klein, and W.N. Hardy, *Phys. Rev. B* **50**, 4051 (1994).
- ³⁵J.F. Annett, *Physica C* **317-318**, 1 (1999).
- ³⁶For example, Lee *et al.* have given a possible explanation to this linear temperature dependence of penetration depth in all superconductors based on the incomplete condensation of Cooper pairs. Jae-Weon Lee *et al.*, *cond-mat/0101011* (unpublished).