

## Absence of nonlinear Meissner effect in $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$

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We present measurements of the field and temperature dependence of the penetration depth ( $\lambda$ ) in high purity, untwinned single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  in all three crystallographic directions. The temperature dependence of  $\lambda$  is linear down to low temperatures, showing that our crystals are extremely clean. Both the magnitude and temperature dependence of the field dependent correction to  $\lambda$ , however, are considerably different from that predicted from the theory of the nonlinear Meissner effect for a  $d$ -wave superconductor (Yip-Sauls theory). Our results suggest that the Yip-Sauls effect is either absent or is unobservably small in the Meissner state of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ . [S0163-1829(99)50322-6]

The identification of a strong linear term in the temperature dependence of the magnetic penetration depth ( $\lambda$ ) of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  was a key development in identifying the  $d$ -wave symmetry of the order parameter in high-temperature superconductors.<sup>1</sup> The linear increase of  $\lambda$  with increasing temperature in a  $d$ -wave superconductor reflects a decrease in superfluid density caused by thermal excitations near the nodes in the order parameter, and is insensitive to the direction of the superflow. Yip and Sauls<sup>2,3</sup> proposed that a much more sensitive test of the order parameter symmetry can be made by measuring the field dependence of  $\lambda$  in the Meissner state. They argued that at sufficiently low temperature, a pure  $d$ -wave superconductor should have a weak, linear field dependence, the magnitude of which depends on the relative orientations of the applied field and the nodes in the order parameter.

Although the linear temperature dependence of  $\lambda$  is now well established in many materials, the field dependence has proved much more difficult to isolate. There is to date no *conclusive* evidence as to the presence or absence of this effect in the Meissner state in any material. In this paper we present measurements of both the field and temperature dependence of  $\lambda$  below  $H_{c1}$  in high purity, untwinned single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ , in all three crystallographic directions ( $H\parallel a, b, c$ ). We find that in some crystals there is a relatively strong field dependence to  $\lambda$  which might naively be attributed to the Yip-Sauls effect. However, a closer examination reveals that it is almost certainly of extrinsic origin. Our cleanest crystal shows a field dependence of  $\lambda$  which is up to a factor of 6 lower than that expected from the Yip-Sauls theory and is not suppressed by increasing temperature. This suggests that the Yip-Sauls effect is either absent or unobservably small in the Meissner state of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ .

In the Yip-Sauls theory the field dependence of  $\lambda$  at low temperature is a direct consequence of the presence of the nodes in the order parameter. In the presence of a finite superflow ( $\vec{v}_s$ ) the quasiparticle energy levels are Doppler

shifted by an amount  $\delta\epsilon \propto \vec{v}_s \cdot \vec{v}_f$ , where  $\vec{v}_f$  is the Fermi velocity. In a  $d$ -wave superconductor at low temperature, this effect changes the occupancy of the gapless states near the nodes of the order parameter, resulting in the creation of a jet of quasiparticles moving in the opposite direction to the superflow, thus reducing the effective superfluid density. In an  $s$ -wave superconductor the effect is absent at low temperature because the finite gap prevents the shifted states from being occupied. Yip-Sauls calculate the field dependence in a  $d$ -wave superconductor to be

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

where  $H_0$  is of order the thermodynamic critical field (Xu *et al.*<sup>3</sup> estimated it to be 2.5 T for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ ), and  $\alpha$  depends on the relative orientation of the nodes and the field. For a field directed along an antinode, we calculate from this formula that the slope  $d\lambda_a(H)/dH = 4.5 \times 10^{-2} \text{ \AA/Oe}$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ .<sup>4</sup> The Yip-Sauls effect is very sensitive to temperature and sample purity. At finite temperature  $\Delta\lambda$  is only linear in  $H$  above a crossover field  $H^* \approx k_b T H_0 / 2\Delta_0$  which exceeds  $H_{c1}$  for  $T \gtrsim 3 \text{ K}$  [here  $\Delta_0$  is the maximum energy gap  $\sim 1.9T_c$  (Ref. 3)]. Below  $H^*$ ,  $\Delta\lambda \sim H^2$  and  $d\lambda/dH$  is much reduced. This high sensitivity on temperature can be used to distinguish the Yip-Sauls effect from other possible (nonintrinsic) contributions.

To measure  $\lambda(T, H)$  we utilize a resonant LC circuit driven by a tunnel diode operating at 13 MHz, with frequency stability better than 1 part in  $10^9/(\text{Hz})^{1/2}$ . Well below  $T_c$ , changes in the resonant frequency are directly proportional to changes in the effective screening volume of the sample. For a typical crystal with  $1 \text{ mm}^2$  surface area, we obtain a noise level in  $\lambda$  of  $\sim 0.1 \text{ \AA}$ . This resolution is about 100 times better than previous measurements.<sup>5</sup> A static, dc field (up to 1000 Oe) is provided by a superconducting solenoid which is collinear with the RF probe field. The complete experiment was placed inside a mumetal shielded Dewar; the remnant field was measured, using a

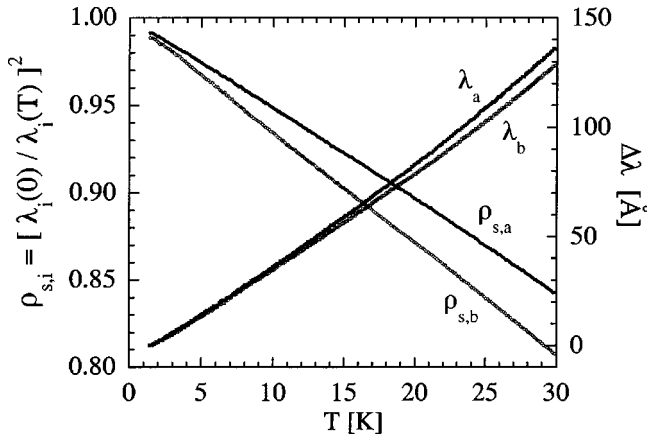


FIG. 1. Temperature dependence of  $\lambda_a$  and  $\lambda_b$  for our untwinned single crystal of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ , in zero dc field. The superfluid density  $[\rho_{s,i} = (\lambda_i(0)/\lambda_i(T))^2]$  is also shown.

fluxgate magnetometer, to be  $\leq 2$  mOe. The RF probe field to which the sample is subjected is estimated to be also  $\leq 2$  mOe. These very low field values help keep to an absolute minimum the number of vortices trapped in the sample as it is cooled through  $T_c$ .

The sample is located on a sapphire rod, the temperature of which is varied independent of the rest of the apparatus. The sample rod may be drawn out of the measurement coil at low temperature so that the background field dependence of the apparatus can be measured *in situ*. We found that this background was highly reproducible at low temperature, but could change markedly upon thermal cycling, making an *in situ* measurement vital. There was a further background originating from the sapphire sample holder, which was temperature dependent, but highly reproducible between runs. This was therefore measured in a separate run with the sample removed. These backgrounds typically give a frequency shift corresponding to  $\sim 3$  Å equivalent change in  $\lambda$  for a  $1 \text{ mm}^2$  single crystal in 100 Oe.

The effective volume of the sense coil was calculated by measuring the shift in frequency when a small sphere of aluminum was inserted. In the  $B\|a,b$  geometry the measured frequency shifts are then easily related to changes in penetration depth simply by measuring the area of the faces of the sample. For the  $B\|c$  geometry a calibration factor is rather more difficult to calculate and so it was estimated by comparing the temperature dependence of the frequency shifts in this orientation to that for  $B\|a,b$ . This factor was then used to relate the field dependent shifts to  $\Delta\lambda(H)$ .

The single crystals measured were grown in yttria stabilized zirconia crucibles as described in Ref. 8. They were annealed so as to give optimal doping. The crystal used for the bulk of the measurements described here had dimensions  $a \times b \times c = 0.80 \times 0.51 \times 0.011 \text{ mm}^3$  and had a  $T_c$  of 91.4 K. The crystal was untwinned (apart from a small region near one corner). X-ray diffraction was used to identify the  $a, b$  axes.

The temperature dependence of  $\lambda$  and the superfluid density  $\{\rho_{s,i} = [\lambda_i(0)/\lambda_i(T)]^2\}$  for the field applied along the  $a$  and  $b$  axes is shown in Fig. 1.<sup>7</sup>  $\lambda(0)$  values were taken from Basov *et al.*<sup>9,10</sup> [ $\lambda_a(0) = 1600$  Å and  $\lambda_b(0) = 1200$  Å]. It can be seen that  $\rho_s$  is essentially linear down to the lowest

temperature measured (1.4 K). A fit to the dirty  $d$ -wave interpolation formula of Hirschfeld and Goldenfeld<sup>11</sup> [ $\lambda(T) = aT^2/(T+T^*)$ ] yields values of  $T^* \leq 1$  K for both directions. The anisotropy in  $d\lambda/dT$  at low temperatures is only  $\sim 3\%$  ( $d\lambda_a/dT = 4.34$  Å/K and  $d\lambda_b/dT = 4.23$  Å/K) which is somewhat less than that reported by Bonn *et al.*<sup>6</sup> This may reflect a lower anisotropy in the zero temperature values of  $\lambda$  in our crystals. We note that our values for  $d\rho_s/dT$  are within 5% of those in Ref. 6. The values of  $T^*$  for this crystal are among the lowest reported to date, showing that our crystal is extremely clean. This property is extremely important as the Yip-Sauls theory predicts that even small amounts of impurities depress the field dependence of  $\lambda$ .

A major problem in measuring the field dependence of  $\lambda$ , by the method described above, is isolating changes in the London penetration depth ( $\lambda_L$ ) from other effects which also increase the effective penetration depth  $\lambda_E$ . The most obvious extrinsic contribution is caused by the motion of vortices. In the mixed state the additional field penetration caused by vortex motion is given by Campbell penetration depth  $\lambda_p = B\phi_0/\mu_0\kappa_p$  ( $\kappa_p$  is the pinning force, and  $\phi_0$  is the flux quantum). The contributions add in quadrature,<sup>12</sup>  $\lambda_E^2(H) = \lambda_L^2(H) + \lambda_p^2$ , and if  $\lambda_p \ll \lambda_L$  then

$$\Delta\lambda_E \approx \Delta\lambda_L + \frac{B\phi_0}{2\mu_0\lambda_L\kappa_p}, \quad \lambda_p \ll \lambda_L. \quad (2)$$

Using values of  $\kappa_p$  taken from Ref. 13 we find that at low temperature the vortex contribution would be  $\sim 25$  Å in 100 Oe (for  $H\|ab$ ) if  $H_{c1}$  was zero and flux was free to enter the sample. For  $H$  less than  $H_{c1}$  the sample is in the Meissner state and should, in principle, be free of vortices. However, in practice vortices may enter well before  $H_{c1}$  at sharp corners and surface imperfections and so in these regions  $\lambda_E > \lambda_L$ . In the measurement configuration  $H\|c$  this problem is likely to be intensified as the field lines are highly distorted due to the flat plate geometry of the sample. Another contribution may come from weak links at the surface. An applied field may dramatically suppress the critical current of any weak link thus causing an increase in  $\lambda_E$ . Finally in the configuration  $H\|ab$  part of the supercurrent flows along the  $c$  axis, and  $\lambda_E \propto \lambda_{ab} + t/l\lambda_c$  (where  $t$  and  $l$  are the dimensions of the sample along the  $c$  and  $a, b$  axes, respectively). Because of the relative weak anisotropy of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  this introduces only a small correction to the measured temperature dependence of  $\lambda$  for thin samples. However, the contribution of  $\lambda_c$  to the field dependence of  $\lambda$  may not be negligible. From the above it is clear that the measured field induced changes in  $\lambda$  can only set an upper limit for the intrinsic field dependence of  $\lambda_L$ , in all field configurations.

The field dependence of  $\lambda$  for our  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  crystal in the  $a$  and  $b$  directions at our lowest temperature (1.4 K) is shown in Fig. 2.  $\Delta\lambda(H)$  is approximately linear in field, with a slope of  $20 \pm 5 \times 10^{-3}$  Å/Oe (i.e.,  $\sim 2$  Å in 100 Oe) in both directions. The data were not hysteretic within the scatter. Although the measured  $\Delta\lambda(100 \text{ Oe})$  is a factor of 2 smaller than the theory prediction for  $T=0$  K, it is comparable with what is predicted at  $T=1$  K. However, the data seem to better follow a straight linear dependence rather than the rounded finite temperature theory curve.

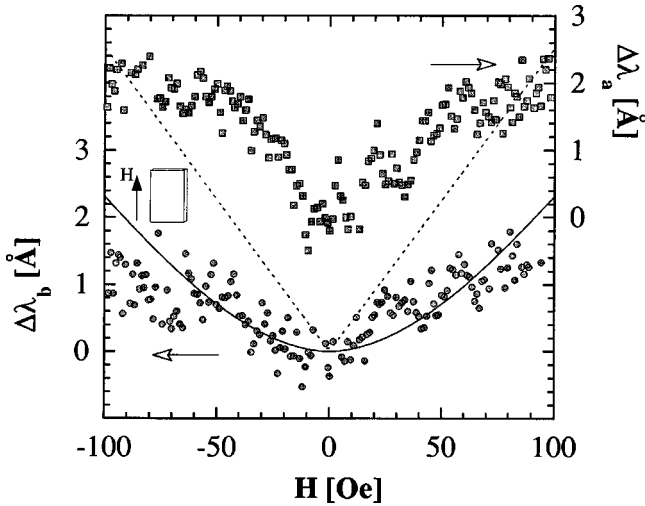


FIG. 2. Field dependence of  $\lambda$  in the  $a$  and  $b$  directions at 1.4 K. The dashed and solid lines show the theory predictions at 0 K and 1.0 K, respectively.

Data for the changes in the average in-plane penetration depth ( $\lambda_{ab}$ ) in the third field configuration ( $H \parallel c$ ) are shown in the upper panel of Fig. 3.  $\Delta\lambda_{ab}(H)$  is linear up to the maximum field measured [a decrease in slope was seen above  $\sim 20$  Oe (not shown)]. Again no hysteresis was observed at these low field values. For this flat sample, in this

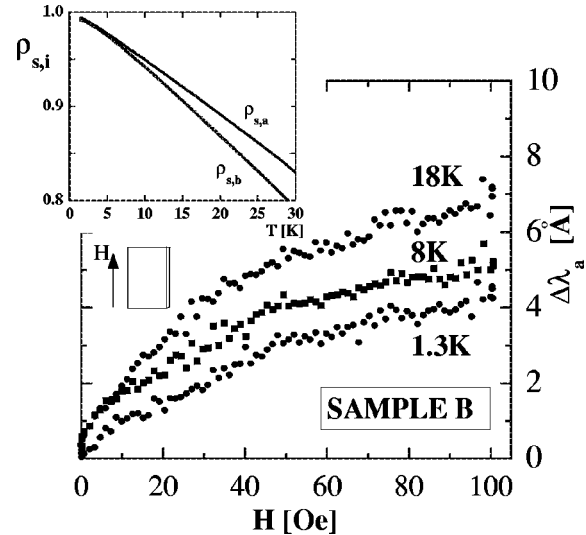


FIG. 4. Field dependence of  $\lambda_a$  for a second sample, at several fixed temperatures (indicated on figure).  $H$  was  $\parallel b$ . The inset shows the temperature dependence of  $\rho_{s,a}$  and  $\rho_{s,b}$  for the same sample.

geometry, the field at the surface of the sample is clearly very different from the applied field. We have estimated the field at the edge of the sample by multiplying the applied field by a demagnetizing factor  $[1/(1-N)]$ , derived from an inscribed ellipsoid approximation. This “demagnetizing” factor was within 20% of that estimated by comparing the known volume of the sample to the measured apparent shielding volume (as measured in a zero field cooled dc magnetization measurement). The actual field on the edge of the sample will not be constant over the whole width, however this “demagnetizing” factor is a rough estimate of the average field. Using  $1/(1-N)=36$ , we estimate the slope  $d\lambda_{ab}/dH=6.3 \times 10^{-3}$  Å/Oe, which is  $\sim 3$  times smaller than for the  $H \parallel a, b$  measurements and more than 6 times smaller than the Yip-Sauls prediction at 300 Oe. We note that with this demagnetizing factor that the change in slope at  $H \sim 20$  Oe corresponds roughly to  $H_{c1}$  in this direction.

The lower left panel of Fig. 3 shows the evolution of the slope,  $d\lambda_{ab}/dH$  as temperature is increased. There is almost no change in  $d\lambda_{ab}/dH$  up to 50 K, in sharp contrast to the predictions of the Yip-Sauls theory. As  $T_c$  is approached  $d\lambda_{ab}/dH$  increases substantially. There was no indication of the predicted change from  $\Delta\lambda(H) \propto H$  to  $\Delta\lambda(H) \propto H^2$  as temperature was increased. We conclude from these observations that the small field dependence that we see does not originate from the Yip-Sauls effect. If we interpret  $\Delta\lambda(H)$  as originating from vortex motion, the above results would imply that at low temperature  $\kappa_p$  is almost constant in our samples.

A close examination of  $\lambda_{ab}(H)$  in Fig. 3 shows that the apparent noise on top of the linear dependence is reproducible with a pattern which is approximately symmetric about  $H=0$ . In fact these modulations in  $\lambda_{ab}$  are quite reproducible and become more pronounced as temperature is increased. In another sample we observed much bigger modulations which resembled closely the type of Fraunhofer pattern typical of the field dependence of the critical current in weak links (see the lower right panel in Fig. 3). We conclude that these effects indicate the presence of weak links

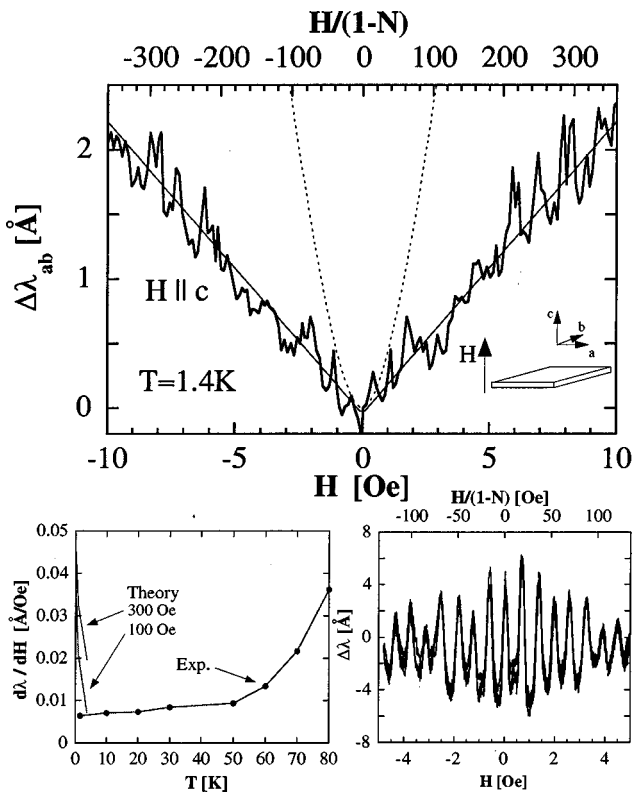


FIG. 3. Top: Field dependence of  $\lambda_{ab}$  with the field applied along  $c$  at 1.4 K. The dashed line is the theory prediction for  $T = 1$  K. Bottom left: The evolution of  $d\lambda_{ab}/dH$  with temperature. Theory curves are the average slope  $\Delta\lambda(H)/H$  with  $H=100$  Oe and 300 Oe from Ref. 3. Bottom right: Fraunhofer-like oscillations in  $\lambda(H)$  in another sample. The data are highly reproducible (five field sweeps are shown).

on the edge of the crystal, the critical current of which is modulated by the static dc field. We note that this structure is only observable because of the extremely low modulation fields used in our experiment.

Finally in Fig. 4 we show data for a second sample of optimally doped, untwinned  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  ( $T_c = 93.6$  K).  $\rho_s(T)$  (inset Fig. 4) for this sample is linear down to only  $\sim 4$  K, showing that it is dirtier than the first sample (cf., Fig. 1).  $\Delta\lambda(H)$  however is larger by around a factor 2 and is linear up to a field of approximately 50 Oe. Again no hysteresis was observed over the field range in the figure ( $H < 100$  Oe). This linear slope increases slightly with increasing temperature in sharp contrast to the predictions of the Yip-Sauls theory. From this fact we conclude that this linear contribution is again of extrinsic origin. The larger magnitude of  $\Delta\lambda(H)$  probably reflects more flux leaking into this sample.

The weak temperature dependence of  $d\lambda_{ab}/dH$  shown here is consistent with the results of Maeda *et al.*<sup>5</sup> for their samples of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ . In their work the lowest temperature at which experiments were performed was 10 K, and little change was found in  $d\lambda/dH$  as temperature was increased. However, these authors report much larger changes in  $\lambda(H)$  than are found here or are indeed predicted by the Yip-Sauls theory. The large size of these effects and the fact that they are not damped out by increasing temperature leads us to conclude that they do not originate from the Yip-Sauls effect, and probably arise from vortex motion.

The large body of evidence collected thus far shows fairly convincingly that  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  is a *d*-wave superconductor and so other possible reasons must be considered for the

apparent absence of the Yip-Sauls effect below  $H_{c1}$ . One possibility could be the effect of nonlocal electrodynamics. Kosztin *et al.*<sup>14</sup> have pointed out that the usual assumption that  $\lambda > \xi$  does not hold at the nodes of a *d*-wave superconductor, and have shown that this has serious consequences for the low *T* excitations. Li *et al.*<sup>15</sup> have recently made calculations of the  $\Delta\lambda(H)$  which include both the nonlocal and nonlinear effects, and they conclude that the nonlocal effects drastically reduce  $\Delta\lambda(H)$  at fields  $\leq H_{c1}$ , rendering the effect essentially unobservable in the Meissner state.

In summary, we have presented measurements of the field dependence of  $\lambda$  on very high-purity single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ , at temperatures down to  $T/T_c = 0.015$ . In our cleanest sample we observe a very weak field dependence in all three crystallographic directions. In the  $H\parallel c$  geometry the slope is around six times smaller than our estimates of what we would expect to see if the Yip-Sauls theory was valid. In the  $H\parallel a, b$  geometry  $\Delta\lambda(H)$  is comparable in magnitude to theory although no indication of finite temperature rounding is seen. A second sample shows a larger effect even though its purity level is lower. In all cases, however, the observed field dependence of  $\lambda$  is not diminished by increasing temperature and so we conclude that it does not arise from the Yip-Sauls effect. The sample dependent  $\Delta\lambda(H)$  we observe most likely originates from vortex motion.

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<sup>1</sup>W. N. Hardy, D. A. Bonn, D. C. Morgan, R. X. Liang, and K. Zhang, *Phys. Rev. Lett.* **70**, 3999 (1993).

<sup>2</sup>S. K. Yip and J. A. Sauls, *Phys. Rev. Lett.* **69**, 2264 (1992).

<sup>3</sup>D. Xu, S. K. Yip, and J. A. Sauls, *Phys. Rev. B* **51**, 16 233 (1995).

<sup>4</sup>Yip and co-workers (Refs. 2 and 3) define  $\lambda = -[d \ln B/dx]^{-1}$ . However, in our experiment the changes in the effective  $\lambda$  are related to  $\int B dV$ , which reduces their  $\Delta\lambda(H)$  values by 2.

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<sup>6</sup>D. A. Bonn *et al.*, *Czech. J. Phys.* **46**, 3195 (1996).

<sup>7</sup>The data were not corrected for the small *c*-axis contribution. For

our crystals the error in  $\Delta\lambda(T=30$  K) is estimated to be less than 1% (Ref. 6).

<sup>8</sup>S. E. Stupp and D. M. Ginsberg, in *Physical Properties of High Temperature Superconductors III*, edited by D. M. Ginsberg (World Scientific, Singapore, 1992).

<sup>9</sup>D. N. Basov *et al.*, *Phys. Rev. Lett.* **74**, 598 (1995).

<sup>10</sup> $\Delta\lambda(T) = \Delta\lambda(T) + \lambda(T=1.45$  K). We estimated  $\lambda(T=1.45$  K) by linearly extrapolating our  $\Delta\lambda(T)$  data to  $T=0$ .

<sup>11</sup>P. J. Hirschfeld and N. Goldenfeld, *Phys. Rev. B* **48**, 4219 (1993).

<sup>12</sup>M. W. Coffey and J. R. Clem, *Phys. Rev. B* **45**, 9872 (1992).

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<sup>14</sup>I. Kosztin and A. J. Leggett, *Phys. Rev. Lett.* **79**, 135 (1997).

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