PHYSICAL REVIEW B, VOLUME 63, 180405(R)

Reanalysis of the magnetic field dependence of the penetration depth: Observation of the nonlinear Meissner effect

Klaus Halterman,[1] Oriol T. Valls,[1] and Igor Žutić[2]
[1]*Department of Physics and Minnesota Supercomputer Institute, University of Minnesota, Minneapolis, Minnesota 55455*
[2]*Department of Physics and Center for Superconductivity Research, University of Maryland, College Park, Maryland 20742*
(Received 12 October 2000; published 23 April 2001)

We examine recent high-precision experimental data on the magnetic field, **H**, dependence of the penetration depth $\lambda(H)$ in $YBa_2Cu_3O_{7-\delta}$ for several field directions in the a-b plane. In a theoretical analysis that incorporates the effects of orthorhombic symmetry, we show that the data at sufficiently high magnetic fields and low temperatures are in quantitative agreement with the theoretical predictions of the nonlinear Meissner effect.

DOI: 10.1103/PhysRevB.63.180405 PACS number(s): 74.20.De, 74.25.Nf, 72.20.Ht

The nonlinear Meissner effect (NLME) is potentially a very important tool for the study of the pairing state in high-temperature superconductors (HTSC's), as well as other materials in the ever increasing list of those for which the proposed order parameter (OP) leads to an energy gap with nodes. It is widely accepted[1] that the symmetry of the OP in HTSC's is at least predominantly *d* wave, vanishing at nodal lines approximately 90° apart in a quasi-two-dimensional Fermi surface (FS). Many details of the OP in these materials remain quite unclear, however. Are the nodes exactly at right angles? Are they true nodes or only very deep minima "quasinodes?"[2] Addressing these and similar questions is important for obtaining clues about the nature of the superconductivity. They are particularly difficult to answer for the *bulk* OP, which may well differ[3] from the more easily observed surface state. To probe the bulk OP it is best to use electromagnetic techniques, since electromagnetic fields penetrate the sample over a depth of the order of the London penetration length λ, which is, in these materials, several orders of magnitude larger than the coherence length ξ that characterizes the range of typical surface probes. Indeed, one of the early key results[4] in support of bulk *d*-wave superconductivity was the measurement of the linear temperature dependence of the penetration depth in a high-purity $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystal. However, such data indicated only the existence of nodal lines without the angular resolution needed to identify their position. Consequently, intensive efforts to precisely determine the structure of the bulk OP have continued.

It was first pointed out years ago[5] that nodes in the OP yield distinctive and measurable nonlinear effects in the angular and field dependencies of the penetration depth when the superconductor is in the Meissner state. In subsequent theoretical work[6–9] more emphasis was placed on the existence, due to this NLME, of a component of the diamagnetic moment normal to the applied field and on the torque associated with this transverse component. These phenomena were deemed to be easier to measure than the changes in λ itself. It was shown[9] in this context that the NLME can be used to perform *node spectroscopy*, that is, not just to infer the existence of nodes, but to *locate their positions* on the FS and to determine whether they are true nodes. Yet, the experimental situation is rather confusing. The best experimental effort to measure the transverse diamagnetic moment[10] in YBCO was inconclusive. Subsequently, results[11] for the magnetic-field-dependent penetration depth to a precision of ~ 0.1 Å were reported.[12] The NLME should be observable in such a high-precision experiment, more precise than existing transverse moment measurements. Unfortunately, no theory of the NLME contribution to the penetration depth for orthorhomobic structures such as YBCO was available when Ref. 11 was written. Only very recently[13] have the necessary calculations been performed. This has resulted in contradictory claims as to whether observed results are in agreement with NLME theory. Thus, a certain amount of skepticism has developed as to the observability of the NLME.

In this paper we show that measurements of the field-dependent penetration depth $\lambda(\psi,H)$ as a function of the angle ψ that an applied field **H** in the a-b plane forms with the *a* axis must be analyzed very carefully. The anisotropy of the linear penetration depth tensor has a drastic effect[13] on the NLME for $\lambda(\psi,H)$. This may have been overlooked because the anisotropy effects in the transverse moment are known[9] to be relatively minor. One must also contend with several other factors that may mask the signal at low fields and high temperatures, and which are very difficult to account for theoretically. Thus we reanalyze here the best data available for the penetration depth in YBCO. We find that, although some questions remain, the low-temperature data are in quantitative agreement with theoretical expectations for the NLME in this material.

We focus here on YBCO, the most experimentally studied[10,11,14,15] HTSC in this context. Hence, the relevant material parameters are well known, thus reducing the uncertainty in the fitting procedures. We perform our analysis primarily on the most complete available high-resolution data of Ref. 11, which includes results for four different directions of the applied field in the a-b plane.

The angular- and field-dependent increase in the penetration depth due to the NLME for materials with orthorhombic anisotropy of the YBCO type was first calculated in Ref. 13. The details will not be repeated here. The sample is assumed to have its larger faces parallel to the a-b plane (this is the case for crystals grown by the usual methods) and thickness large compared with the penetration depth. One has for the quantity $\Delta\lambda(\psi,H)\equiv\lambda(\psi,H)-\lambda(\psi,0)$

$$\Delta\lambda(\psi,H)=\frac{1}{6}\frac{H}{H_0}\lambda\mathcal{Y}(\psi). \tag{1}$$

Here λ is the geometric mean of the two in-plane principal values λ_a and λ_b of the zero-field penetration depth tensor, H_0 is a characteristic field of order $\Phi_0/\pi^2\lambda\xi$ (Φ_0 is the flux quantum), and $\mathcal{Y}$ carries the angular dependence. The orthorhombicity, very important in this case, is incorporated into $\mathcal{Y}$ through two parameters:[13] one is the ratio $\Lambda\equiv\lambda_a/\lambda_b$, and the other is the angle α that the Fermi velocity at the node located in the first quadrant forms with the a direction. Because of the orthorhombic distortion of the FS, this angle does not have to exactly equal $\pi/4$ even for a pure $d_{x^2-y^2}$ state, while the quantity Λ, for YBCO, considerably exceeds unity. Here we take the zero-field quantities λ_a and λ_b fixed at their experimental[16] values (1050 and 1575 Å), giving $\Lambda=1.5$. In this case, the full expressions[13,17] for $\mathcal{Y}(\psi)$ simplify somewhat and can be written as

$$\mathcal{Y}(\psi)=\frac{18\Lambda}{2+\Lambda}\cos^2\alpha\sin\alpha\cos\psi\sin^2\psi+\frac{2}{\Lambda^2(1+2\Lambda)}$$
$$\times\sin^3\alpha\cos^3\psi\left[1+2\Lambda+(4\Lambda-1)\left(\frac{\tan\psi}{\tan\psi_1}\right)^{3\Lambda/(\Lambda-1)}\right]$$
$$+\frac{2\Lambda^2(2\Lambda^2-10\Lambda-1)}{(2+\Lambda)(1+2\Lambda)}\cos^3\alpha\sin^3\psi\left(\frac{\tan\psi}{\tan\psi_1}\right)^{3/(\Lambda-1)},$$
$$\psi\in[0,\psi_1], \tag{2a}$$

$$\mathcal{Y}(\psi)=\frac{18}{1+2\Lambda}\sin^2\alpha\cos\alpha\cos^2\psi\sin\psi+2\Lambda^2\cos^3\alpha\sin^3\psi,$$
$$\psi\in\left[\psi_1,\frac{\pi}{2}\right], \tag{2b}$$

where the angle ψ_1 is given by $\psi_1\equiv\arctan(\tan\alpha/\Lambda)$.

Because of the orthorhombicity, the angular dependence of $\Delta\lambda(\psi,H)$ is quite different[13] for $\Lambda=1.5$ than that found for the tetragonal case ($\Lambda=1,\alpha=\pi/4$). This is unlike the situation for the transverse magnetic moment,[9] which from symmetry considerations vanishes when **H** is along the a or b axes. Thus, moderate orthorhombic anisotropy induces only a relatively minor distortion in the curve between $\psi=0$ and $\psi=\pi/2$ since these points are, so to speak, anchored. This is not the case for $\Delta\lambda(\psi,H)$: when the field is applied along $\psi=0$ the currents flow over a region of thickness determined by λ_b, while if $\psi=\pi/2$ the relevant skin depth is λ_a. The effect is nonzero, and different, in either case. This difference is compounded by the nonlinearity and the apparent overall symmetry of $\mathcal{Y}$ is π rather than $\pi/2$ even for moderate orthorhombicity. Failure to take this into account leads to erroneous conclusions concerning the angular dependence of $\Delta\lambda(\psi,H)$.

To analyze data in terms of Eqs. (1) and (2), additional considerations are needed. These expressions, indicating that

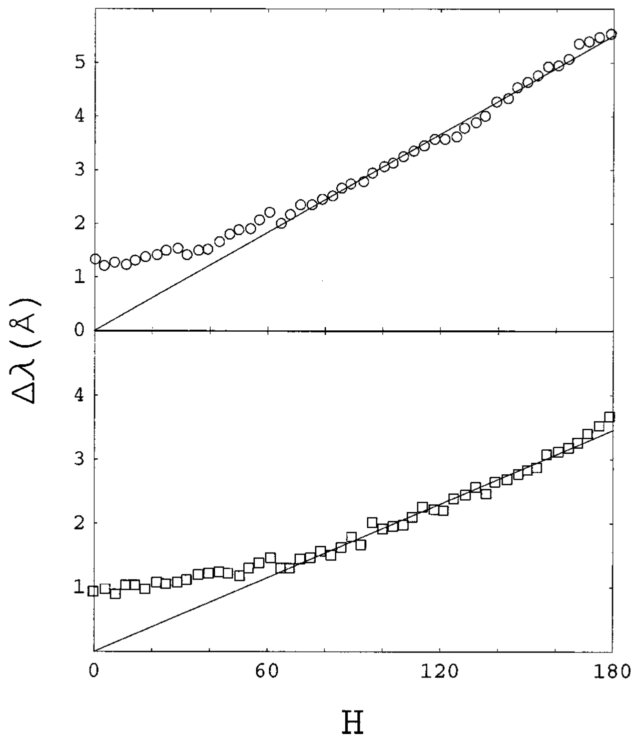

FIG. 1. Magnetic field (H) dependence of $\Delta\lambda$ (see text). The straight lines are fits to the 1.2-K data (circles and squares) of Fig. 3, Ref. 11, for $H>60$ g. Top: **H** applied along the b axis. Bottom: **H** along the a axis.

$\Delta\lambda$ is proportional to H, are valid at low temperature. Here, ‘‘low’’ temperature is a *field-dependent* concept. The characteristic temperature separating the high- and low-T regimes is[6,7] $T^*(H)\approx\Delta_0(H/H_0)$, where Δ_0 is the gap amplitude. At any finite T, the validity of the above equations will break down at sufficiently small H. Further, the effect of impurities is not included. For the clean samples used in experiments[10,11,14] this should affect[6,7] only the small field results. The same is true of possible nonlocal effects.[18] If they are present at all in this geometry,[19] they would affect results at fields below[20] 20 g. Ideally, one would like to take into account all of these effects by modifying the above formulas. However, it is not feasible at present to include all of these factors *simultaneously* in a reliable manner. It is therefore best to perform the analysis in a consistent manner in terms of data in the higher range of fields available, where these additional effects are all weak, and the above expressions are valid.

In Fig. 1 we show best straight-line fits to the 1.2-K data of Ref. 11 for **H** along the a and b directions. All data in the range $H>60$ g are included in the fit. The cutoff of 60 g was chosen as the point below which deviations from a straight line begin and it will be shown below to lead to a consistent interpretation. The straight line does not intercept the origin of the original plot, which has to be shifted downward. This is as expected, since the experimental $\Delta\lambda$ includes the previously mentioned temperature,[21] impurity, and possible nonlocal effects that increase this quantity with respect to the theoretical, clean, zero-temperature local value. The shift is

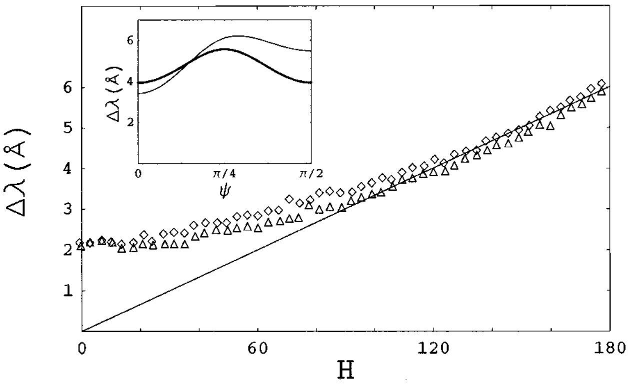

FIG. 2. $\Delta\lambda(H)$ for H along $\psi=|\pi/4|$. The straight line is the theoretical result at higher fields with the parameters extracted from Fig. 1. Diamonds and triangles are the experimental data with **H** applied at $\psi=\pm\pi/4$. Inset: predicted angular dependence (thin curve) of $\Delta\lambda(\psi)$ including anisotropy. Bold curve: result for a tetragonal system. The amplitudes of both curves correspond to $H=180$ g.

small, of order 1 Å, indicating that the sample is clean and any such spurious effects are small. The two slopes of the lines obtained from these fits are the quantities $\mathcal{Y}(0)\lambda/H_0$ and $\mathcal{Y}(\pi/2)\lambda/H_0$, respectively. From Eq. (2), $\mathcal{Y}(0)/\mathcal{Y}(\pi/2)=(1/\Lambda^4)\tan^3\alpha$. We have fixed Λ to its independent experimental value of $\Lambda=1.5$. We then determine the α that fits this ratio and subsequently find the characteristic field H_0 from either one of the slopes. The results are very sensible: we obtain $\alpha=\pi/4+\pi/17$ and $H_0=5660$ g. The value for the angle between the Fermi velocity at the node and the a axis exceeds $\pi/4$ by a small amount, as one would expect for a pure $d_{x^2-y^2}$ pairing state and a tight-binding FS with a slight orthorhombic distortion. The value of the characteristic field is consistent with expectations[8,9] and also with our cutoff choice for the field: in the range of fitting we have $H/H_0>0.01$. This means that in this field range, the characteristic temperature $T^*(H)$ introduced above is of the order of 4–12 K. Hence the 1.2-K data included in the fitting are in the low-temperature regime and the procedure is consistent.

Up to now, we have, however, fit two quantities with two parameters, although the reasonable values obtained for these parameters are encouraging. To go beyond, we now use the obtained values of α and H_0 to plot the predicted slope of the high-field data at $|\psi|=\pi/4$ without any additional parameters. This is done in Fig. 2. Experimental results for fields applied in the $\psi=\pm\pi/4$ directions are included. These results ought to be identical (even with the orthorhombic distortion) and their small discrepancy reflects systematic errors in the experiment. Nevertheless, the fit is excellent in the high-field range. We also plot (inset) with these parameter values the predicted angular dependence of $\Delta\lambda$ for YBCO. One can see that with the orthorhombicity, this angular dependence differs considerably from that obtained for a tetragonal system, also plotted for comparison. The actual curve is not symmetric about $\pi/4$ and its maximum is much less pronounced than that for the tetragonal case, which is characterized[5] by a factor of $\sqrt{2}$ between maxima and minima. Because of these differences, the attempt made by the authors of Ref. 11 to reconcile the angular dependence of their data with the theory of the NLME in a tetragonal system had to fail.

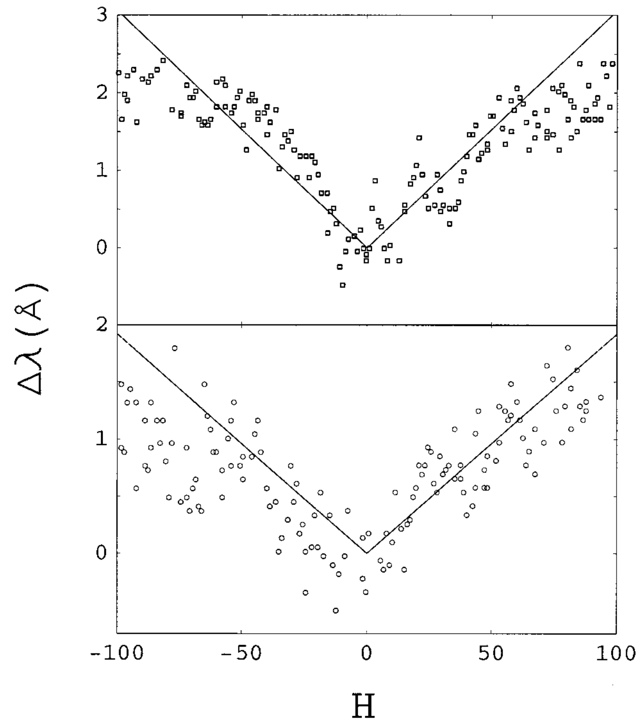

FIG. 3. $\Delta\lambda$ as a function of H. The straight lines are the theoretical results with the same parameters found in Fig. 1. The symbols are experimental data of Ref. 14. Top panel: **H** at $\psi=\pm\pi/2$. Bottom panel: **H** along $\psi=0,\pi$.

In Fig. 3 we compare the theoretical results with other more recent data[14] on YBCO at $T=1.4$ K. The parameters used are exactly the same as previously obtained. No new fits were performed. Results for the two directions available (field applied along the two principal axes) are shown. This data is in a more restricted, lower-field range, and it has considerably more scatter than that of Bidinosti *et al.*[11] All that can be said with certainty is that it is consistent with the NLME theory with the same parameter values.

In summary, the main result of the analysis presented here is that the best low-temperature, high-field data[11] on the nonlinear penetration depth in YBCO is in quantitative agreement, in its magnitude and angular and field dependence, with NLME theoretical expectations. Other data[14] are also consistent with theory. Failure to observe the NLME in the transverse moment[10] seems to be attributable to the actual precision in that experiment being just slightly less than what was in fact required.

Two remarks must be added. First, the crucial influence of the orthorhombic anisotropy in the angular dependence of $\Delta\lambda$, which becomes very different (see Fig. 2) from that found for tetragonal symmetry, must be emphasized. Second, one sees the need to finesse the temperature, impurity, and possibly other problems associated with smaller fields by obtaining and analyzing data at the highest possible fields below that of first flux penetration. Fortunately, this field is in the range 200–400 g (Refs. 10, 11, and 15) for typical YBCO crystals.

The question of the temperature dependence of the results[11,14] is less clear and needs further discussion: results obtained at 7 K for the same sample mainly discussed here are[11] not substantially different from those at 1.7 K. With the characteristic temperature $T^*(H)$ in the range estimated above, it can be that the high-field results are not yet affected by the temperature at 7 K while those at low fields are dominated by largely temperature-independent impurity effects. Indeed, it appears that a straight-line fit to the 7-K data at the highest fields (see Fig. 4 of Ref. 11) has a larger (in absolute value) vertical axis intercept than that for the 1.7-K data, which would be consistent with this scenario. Nevertheless, the weak temperature dependence of the data will remain a puzzle so long as a rigorous calculation including impurities, temperature, and possibly nonlocal and other effects is not feasible. It is possible that these effects combine to yield a temperature dependence weaker than what the naive theory would predict.

Finally, our analysis indicates that there is no significant is admixture to the $d_{x^2-y^2}$ gap, since such an admixture would lead to quasinodes and to[13] a considerable reduction in $\Delta\lambda$. Furthermore, the nearness of α to $\pi/4$ is consistent with the absence of a real s component as well.

It would be desirable to perform measurements of $\Delta\lambda$ in YBCO at additional values of the angle ψ to verify in more detail if the curve in the inset of Fig. 2 is indeed closely followed. The comments made here on the proper way to analyze experimental data should also be taken into account in any attempts to use the NLME to elucidate the pairing states of other suspected unconventional superconducting materials for which it is estimated[13] that the sensitivity of present penetration depth measurements is sufficient to probe the NLME.

We thank C. P. Bidinosti and A. Bhattacharya for very useful information about their respective measurements. This work was supported in part by the Petroleum Research Fund, administered by the ACS (at the University of Minnesota), and by DARPA and ONR (Grant No. N00140010028) at the University of Maryland.

[1] J.F. Annett, N. Goldenfeld, and A.J. Leggett, *Physical Properties of High Temperature Superconductors V*, edited by D.M. Ginsberg (World Scientific, Singapore, 1996); J. Low Temp. Phys. **105**, 473 (1996).

[2] These questions are sometimes phrased in terms of the possible existence of an s (for the first question) or is (for the second) admixture to the predominant d-wave OP.

[3] S.R. Bahcall, Phys. Rev. Lett. **76**, 3634 (1996); M. Covington *et al.*, *ibid.* **79**, 277 (1997); M. Fogelström, D. Rainer, and J.A. Sauls, *ibid.* **79**, 281 (1997).

[4] W.N. Hardy *et al.*, Phys. Rev. Lett. **70**, 3999 (1993).

[5] S.K. Yip and J.A. Sauls, Phys. Rev. Lett. **69**, 2264 (1992).

[6] B.P. Stojković and O.T. Valls, Phys. Rev. B **51**, 6049 (1995).

[7] D. Xu, S.K. Yip, and J.A. Sauls, Phys. Rev. B **51**, 16 233 (1995).

[8] I. Žutić and O.T. Valls, Phys. Rev. B **54**, 15 500 (1996).

[9] I. Žutić and O.T. Valls, Phys. Rev. B **56**, 11 279 (1997).

[10] A. Bhattacharya *et al.*, Phys. Rev. Lett. **82**, 3132 (1999).

[11] C.P. Bidinosti *et al.*, Phys. Rev. Lett. **83**, 3277 (1999).

[12] C.P. Bidinosti and W.N. Hardy, Rev. Sci. Instrum. **71**, 3816 (2000).

[13] K. Halterman, O.T. Valls, and I. Žutić, Phys. Rev. B **63**, 014501 (2000).

[14] A. Carrington *et al.*, Phys. Rev. B **59**, R14 173 (1999).

[15] J. Buan *et al.*, Phys. Rev. B **54**, 7462 (1996).

[16] D.N. Basov *et al.*, Phys. Rev. Lett. **74**, 598 (1995).

[17] The limit $\Lambda=1$ cannot be taken directly in Eq. (2). Recourse to the fuller expressions of Ref. 13 is required.

[18] M.-R. Li, P.J. Hirschfeld, and P. Wölfle, Phys. Rev. Lett. **81**, 5640 (1998).

[19] I. Kostzin and A.J. Leggett, Phys. Rev. Lett. **79**, 135 (1997).

[20] A. Bhattacharya *et al.*, Phys. Rev. Lett. **83**, 887 (1999).

[21] The temperature contribution to this shift is seen in Fig. 14 of Ref. 7.