Has the nonlinear Meissner effect been observed?

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We examine recent high-precision experimental data on the magnetic field, \( \mathbf{H} \), dependence of the penetration depth \( \lambda(H) \) in \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) (YBCO) for several field directions in the \( a-b \) plane. In a new 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.

It is widely accepted [1] that the symmetry of the order parameter (OP) in high temperature superconductors (HTSC’s) is at least predominantly \( d \)-wave, vanishing at nodal lines approximately ninety degrees 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 \( \lambda \) which is, in these materials, several orders of magnitude larger than the coherence length \( \xi \) which 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 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 [5] years ago that nodes in the OP yield distinctive and measurable nonlinear effects in the field and angular dependence of the penetration depth when the superconductor is in the Meissner state. In subsequent theoretical work [6] more emphasis was placed on the existence, due to this nonlinear Meissner effect (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 \( \lambda \) itself. It was shown [7] 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 or not.

Thus, the NLME is potentially a very important tool for the study of the pairing state in HTSC’s, as well as other materials in the ever increasing list of those for which the proposed OP leads to an energy gap with nodes. Yet, the experimental situation is rather confusing. The best experimental effort to measure the transverse diamagnetic moment [8] in YBCO was inconclusive. Subsequently, results [9] for the magnetic field dependent penetration depth to a precision [10] of \( \sim 0.1\text{Å} \) were reported. 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 orthorhombic structures such as YBCO was available when Ref. [11] was written. Only very recently [12] have the necessary calculations been performed. This has resulted in contradictory claims as to whether observed results are in agreement or not 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 \( \psi \) that an applied field \( \mathbf{H} \) 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 [14] to be relatively minor. One must also eliminate several other factors that may mask the signal at low fields 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 quantitatively in agreement with theoretical expectations for the NLME in this material.

We focus here on YBCO, the most experimentally studied \( \text{HTSC} \) 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 $\Delta \lambda(\psi, H) \equiv \lambda(\psi, H) - \lambda(\psi, 0)$:

$$\Delta \lambda(\psi, H) = \frac{1}{\Phi_0} \frac{H}{H_0} \lambda Y(\psi).$$  \hspace{1cm} (1)

Here $\lambda$ is the geometric mean of the two in-plane principal values, $\lambda_a$ and $\lambda_b$, of the zero field penetration depth tensor, $H_0$ is a characteristic field of order $\Phi_0/\pi^2 \lambda \xi$ ($\Phi_0$ is the flux quantum), and $Y$ carries the angular dependence. The orthorhombicity, very important in this case, is incorporated into $Y$ through two parameters [13]: one is the ratio $\Lambda \equiv \lambda_a/\lambda_b$, and the other is the angle $\alpha$ 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 $\Lambda$, for YBCO, considerably exceeds unity. Here we take the zero field quantities $\lambda_a$ and $\lambda_b$ fixed at their experimental [10] values (1050 Å and 1575 Å), giving $\Lambda = 1.5$. In this case, the full expressions [13,17] for $Y(\psi)$ simplify somewhat and can be written as:

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

where the angle $\psi_1$ 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 [3] 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 the $b$ axis. 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 $\lambda_b$ while if $\psi = \pi/2$ the relevant skin depth is $\lambda_a$. The effect is nonzero, and different, in either case. This difference is compounded by the nonlinearity and the apparent overall symmetry of $Y$ is $\pi$ 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 $\Delta \lambda$ 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 $T^*(H) \approx \Delta_0 (H/H_0)$, where $\Delta_0$ 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 [67] only the small field results. The same is true of possible nonlocal effects [13]. If they are present at all in this geometry [19], they would affect results at fields below 20 gauss. 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.

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 $H > 60$ gauss are included in the fit. The cutoff of 60 gauss 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 which increase this quantity with respect to the theoretical, clean, zero temperature, local value. The shift is 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, where from Eq.\,(2), $\mathcal{Y}(0) = (2/\Lambda^2) \sin^3 \alpha$, and $\mathcal{Y}(\pi/2) = 2\Lambda^2 \cos^3 \alpha$. Thus, the ratio of the two slopes depends only on $\alpha$ since we have used the independent experimental value $\Lambda = 1.5$. We then determine the $\alpha$ 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 $H_0 = 5660$ gauss. 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\,[3] 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 order of 4 to 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 $\alpha$ 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.\,3. 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\,[3] 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.

In Fig.\,3 we compare the theoretical results with other more recent data\,[12] 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 the NLME theoretical expectations. Other data\,[14] are also consistent with theory. Failure to observe the NLME in the transverse moment\,[14] 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 gauss\,[10,11,12] 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 7K 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 $\alpha$ 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 $\psi$ 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.

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[1] J.F. Annett, N. Goldenfeld, and A.J. Leggett, *Physical properties of High Temperature Superconductors V*, D.M. Ginsberg (Ed.), 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.*, Phys. Rev. Lett. 79, 277 (1997), M. Fogelström, D. Rainer and J.A. Sauls, Phys. Rev. Lett. 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. Stoiković and O.T. Valls, Phys. Rev. B 51, 6049, (1995).

[7] D. Xu, S.K. Yip and J.A. Sauls, Phys. Rev. B 51, 16233, (1995).

[8] I. Žutić and O.T. Valls, Phys. Rev. B 54, 15500, (1996).

[9] I. Žutić and O.T. Valls, Phys. Rev. B 56, 11279, (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. Inst. 71, 3816 (2000).

[13] K. Halterman, O.T. Valls and I. Žutić, cond-mat/0007510, to appear in Phys. Rev. B.

[14] A. Carrington *et al.*, Phys. Rev. B 59, R14173 (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. Wölle, 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. [2].

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. [13], for $H > 60$ gauss. Top: $H$ applied along the $b$–axis. Bottom: $H$ along the $a$–axis.

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 $H = 180$ gauss.

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 $\psi = 0, \pi$. 

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Fig. 1 Halterman et al

\begin{align*}
\Delta \lambda (\AA) & \quad H \\
\end{align*}
Fig. 2 Halterman et al.

![Graph showing the relationship between \( \Delta \lambda \) (Å) and \( H \).]
Fig. 3 Halterman et al.

Graph showing the relationship between $H$ and $\Delta \lambda$ (Å). The graph contains a scatter plot with data points and two lines indicating a trend. The x-axis represents $H$ ranging from -100 to 100, while the y-axis represents $\Delta \lambda$ (Å) ranging from 0 to 3.