c-AXIS PENETRATION DEPTH IN Bi₂Sr₂CaCu₂O₈₊δ SINGLE CRYSTALS MEASURED BY ac-SUSCEPTIBILITY AND CAVITY PERTURBATION TECHNIQUE

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The c-axis penetration depth \( \Delta \lambda_c \) in Bi₂Sr₂CaCu₂O₈₊δ (BSCCO) single crystals as a function of temperature has been determined using two techniques, namely, measurements of the ac-susceptibility at a frequency of 100 kHz and the surface impedance at 9.4 GHz. Both techniques yield an almost linear function \( \Delta \lambda_c(T) \propto T \) in the temperature range \( T < 0.5T_c \). Electrodynamic analysis of the impedance anisotropy has allowed us to estimate \( \lambda_c(0) \approx 50 \mu m \) in BSCCO crystals overdoped with oxygen \( (T_c \approx 84 K) \) and \( \lambda_c(0) \approx 150 \mu m \) at the optimal doping level \( (T_c \approx 90 K) \).

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Cuprate high-temperature superconductors (HTS) are layered anisotropic materials. Therefore the electrodynamic problem of the magnetic field penetration depth in HTS in the low-field limit is characterized by two length parameters, namely, \( \lambda_{ab} \) controlled by screening currents running in the CuO₂ planes (in-plane penetration depth) and \( \lambda_c \) due to currents running in the direction perpendicular to these planes (out-of-plane or c-axis penetration depth). The temperature dependence of the penetration depth in HTS is largely determined by the superconductivity mechanism. It is known (see, e.g., Ref. [1] and references therein) that \( \Delta \lambda_{ab}(T) \propto T \) in the range \( T < T_c/3 \) in high-quality HTS samples at the optimal level of doping, and this observation has found the most simple interpretation in the d-wave model of the high-frequency response in HTS [2]. Measurements of \( \lambda_c(T) \) are quoted less frequently than those of \( \lambda_{ab}(T) \). Most of such data published by far were derived from microwave measurements of the surface impedance of HTS crystals [3–11]. There is no consensus in literature about \( \Delta \lambda_c(T) \) at low temperatures. Even in reports on low-temperature properties of high-quality YBCO crystals, which are the most studied objects, one can find both linear, \( \Delta \lambda_c(T) \propto T \) [4, 9], and quadratic dependences [11] in the range \( T < T_c/3 \). In BSCCO materials, the shape of \( \Delta \lambda_c(T) \) depends on the level of oxygen doping: in samples with maximal \( T_c \approx 90 K \) \( \Delta \lambda_c(T) \propto T \) at low temperatures [7, 8]; at higher oxygen contents (overdoped samples) \( T_c \) is lower and the linear function \( \Delta \lambda_c(T) \) transforms to a quadratic one [8]. The common feature of all microwave experiments is that the change in the ratio \( \Delta \lambda_c(T)/\lambda_c(0) \) is smaller than in \( \Delta \lambda_{ab}(T)/\lambda_{ab}(0) \) because in all HTS \( \lambda_c(0) \gg \lambda_{ab}(0) \). The length \( \lambda_c(0) \) is
especially large in BSCCO crystals, \( \lambda_c(0) > 10 \mu m \) and, according to some estimates, it ranges up to \( \sim 500 \mu m \). The large spread of \( \lambda_c(0) \) is caused by two factors, namely, the poor accuracy of the techniques used in determination of \( \lambda_c(0) \) and effects of local and extended defects in tested samples, whose range is of order of 1 mm and comparable to both \( \lambda_c \) and total sample dimensions.

Recently we suggested \cite{12} a new technique for determination of \( \lambda_c(0) \) based on the measurements of the surface barrier field \( H_f(T) \propto 1/\lambda_c(T) \) at which Josephson vortices penetrate into the sample. The field \( H_f \) corresponds to the onset of microwave absorption in the locked state of BSCCO single crystals. This paper suggests an alternative technique based on comparison between microwave measurements of BSCCO crystals aligned differently with respect to ac magnetic field and a numerical solution of the electrodynamic problem of the magnetic field distribution in an anisotropic plate at an arbitrary temperature. Moreover, since \( \lambda_c(0) \) in BSCCO single crystals is relatively large, we managed to determine \( \lambda_c(T) \) from the temperature dependences of ac-susceptibility and compare these measurements to results of microwave experiments.

Single crystals of BSCCO were grown by the floating-zone method \cite{13} and shaped as rectangular platelets. This paper presents measurements of two BSCCO samples with various levels of oxygen doping. The first sample (#1), characterized by a higher critical temperature, \( T_c \approx 90 \) K (optimally doped), has dimensions \( a \times b \times c \simeq 1.5 \times 1.5 \times 0.1 \) mm\(^3\) \((a \approx b)\). The second (#2, \( a \times b \times c \simeq 0.8 \times 1.8 \times 0.03 \) mm\(^3\)) is slightly overdoped \((T_c \approx 84 \) K\).

When measuring the ac-susceptibility \( \chi = \chi' - i\chi'' \), we placed a sample inside one of two identical induction coils. The coils were connected to one another, and the out-of-phase and in-phase components of the imbalance signal were measured at a frequency of \( 10^5 \) Hz. These components are proportional to the real and imaginary parts of the sample magnetic moment \( M \), respectively: \( M = \chi v H_0 \), where \( v \) is the sample volume and \( H_0 \) is the ac magnetic field amplitude, which was within 0.1 Oe in our experiments.

Fig.1. Curves of the ac-susceptibility of sample #1 versus temperature in different orientation with respect to ac magnetic field: \( H_\omega \parallel c \) (full circles); \( H_\omega \perp c \), \( H_\omega \) is parallel to the b-edge of the crystal (up triangles); \( H_\omega \perp c \), \( H_\omega \) is parallel to the a-edge of the crystal (down triangles). Left-hand inset: transverse (T) orientation, \( H_\omega \parallel c \), the arrows on the surfaces show directions of the screening current. Right-hand inset: Longitudinal (L) orientation, \( H_\omega \perp c \).

Fig.1 shows temperature dependences \( \chi'(T)/|\chi'(0)| \) in sample #1 for three different sample alignments with respect to the ac magnetic field: the transverse (T) orientation, \( H_\omega \parallel c \), (the inset on the left of Fig.1), when the screening current flows in the ab-plane.
(full circles); in the longitudinal (L) orientation, \( H_\omega \perp c \), (the inset on the right of Fig.1, \( H_\omega \) is parallel to the \( b \)-edge of the crystal), when currents running in the directions of both CuO\(_2\) planes and the \( c \)-axis are present (up triangles); in the L-orientation, \( H_\omega \perp c \), whose difference from the previous configuration is that the sample is turned around the \( c \)-axis through 90° (down triangles). Fig.1 clearly shows that at \( T < T_c \), \( \chi'_{ab}(T) \) is notably smaller in the T-orientation than \( \chi'_{ab+c}(T) \) in the L-orientation (the subscripts of \( \chi' \) denote the direction of the screening current). The coincidence of \( \chi'_{ab+c}(T) \) curves at \( H_\omega \perp c \) and the small width of the superconducting transition at \( H_\omega \parallel c \) (\( \Delta T_c < 1 \) K) indicate that the quality of the tested sample #1 is fairly high. This is supported by precision measurements of surface impedance \( Z_s(T) = R_s(T) + iX_s(T) \) of sample #1 at frequency \( f = 9.4 \) GHz in the T-orientation, which are plotted in Fig. 2. The measurement technique was described in detail elsewhere [1]. It applies to both surface impedance components \( R_s(T) \) and \( X_s(T) \):

\[
R_s = \Gamma_s \Delta(1/Q), \quad X_s = -2\Gamma_s \delta f/f.
\]

Here \( \Gamma_s = \omega \mu_0 \int_V H_\omega^2 dV/\int_S H_s^2 dS \) is the sample geometrical factor (\( \omega = 2\pi f, \mu_0 = 4\pi \times 10^{-7} \) H/m, \( V \) is the volume of the cavity, \( H_\omega \) is the magnetic field generated in the cavity, \( S \) is the total sample surface area, and \( H_s \) is the tangential component of the microwave magnetic field on the sample surface); \( \Delta(1/Q) \) is the difference between the values 1/\( Q \) of the cavity with the sample inside and empty cavity; \( \delta f \) is the frequency shift relative to that which would be measured for a sample with perfect screening, i.e., no penetration of the microwave fields. In the experiment we measure the difference \( \Delta f(T) \) between resonant frequency shifts with temperature of the loaded and empty cavity, which is equal to \( \Delta f(T) = \delta f(T) + f_0 \), where \( f_0 \) is a constant\(^{1}\). The constant \( f_0 \) includes both the perfect-conductor shift and the uncontrolled contribution caused by opening and closing the cavity. In HTS single crystals, the constant \( f_0 \) can be directly derived from measurements of the surface impedance in the normal state; in particular, in the T-orientation \( f_0 \) can be derived from the condition that the real and imaginary parts of

\(^{1}\) We note that \( \delta f(T) \) includes the frequency shift due to the sample thermal expansion, which is essential for \( T > 0.7 T_c \) in the T-orientation [1].
the impedance should be equal above $T_c$ (normal skin-effect). In Fig. 2 $R_s(T) = X_s(T)$ at $T \geq T_c$, and its temperature dependence is adequately described by the expression 
\[ 2R_s^2(T)/\omega\mu_0 = \rho(T) = \rho_0 + bT \]
with $\rho_0 \approx 13 \mu\Omega \cdot cm$ and $b \approx 0.3 \mu\Omega \cdot cm/K$. Given $R_s(T) = \sqrt{\omega\mu_0\rho(T)/2} \approx 0.12 \Omega$, we obtain the resistivity $\rho(T_c) \approx 40 \mu\Omega \cdot cm$. The insets to Fig. 2 show $R_s(T)$ and $\lambda(T) = X_s(T)/\omega\mu_0$ for $T < 0.7T_c$ plotted on a linear scale. The extrapolation of the low-temperature sections of these curves to $T = 0$ yields estimates of $\lambda_{ab}(0) \approx 2600 \AA$ and the residual surface resistance $R_{res} \approx 0.5 m\Omega$. $R_{res}$ is due to various defects in the surface layer of the superconductor and it is generally accepted that the lower the $R_{res}$, the better the sample quality. The above mentioned parameters of sample #1 indicate that its quality is fairly high. In the T-orientation, linear functions $\Delta R_s(T)$ and $\Delta \lambda_{ab}(T)$ in the low-temperature range were previously observed in optimally doped BSCCO crystals at a frequency of about 10 GHz [7, 8, 14]. In the slightly overdoped sample #2 we also observed $\Delta \lambda_{ab}(T)$, $\Delta R_s(T) \propto T$ at low temperature, moreover, the measurement $R_{res} \approx 120 \mu\Omega$ is, to the best of our knowledge, the lowest value ever obtained in BSCCO single crystals.

In both superconducting and normal states of HTS, the relation between electric field and current density is local: $j = \sigma E$, where the conductivity $\sigma$ is a tensor characterized by components $\sigma_{ab}$ and $\sigma_c$. In the normal state, ac field penetrates in the direction of the $c$-axis through the skin depth $\delta_{ab} = \sqrt{2/\omega\mu_0\sigma_{ab}}$ and in the CuO$_2$ plane through $\delta_c = \sqrt{2/\omega\mu_0\sigma_c}$. In the superconducting state all parameters $\delta_{ab}$, $\delta_c$, $\sigma_{ab} = \sigma_{ab}' - i\sigma_{ab}''$, and $\sigma_c = \sigma_c' - i\sigma_c''$ are complex. In the temperature range $T < T_c$, if $\sigma' \ll \sigma''$, the field penetration depths are given by the formulas $\lambda_{ab} = \sqrt{1/\omega\mu_0\sigma_{ab}''}$, $\lambda_c = \sqrt{1/\omega\mu_0\sigma_c''}$. In the close neighborhood of $T_c$, if $\sigma' \gg \sigma''$, the decay of magnetic field in the superconductor is characterized by the functions $\text{Re}(\delta_{ab})$ and $\text{Re}(\delta_c)$, which turn to $\delta_{ab}$ and $\delta_c$, respectively, at $T \geq T_c$.

In the L-orientation of BSCCO single crystals at $T < 0.9T_c$ the penetration depth is smaller than characteristic sample dimensions. If we neglect the anisotropy in the $ab$-plane and the contribution from $ac$-faces (see the inset to Fig. 1), which is a factor $\sim c/b$ smaller than that of the $ab$-surfaces, the effective impedance $Z_{ab+c}^s$ in the $L$-orientation can be expressed in terms of $Z_{ab}^s$ and $Z_{c}^s$ averaged over the surface area [1, 5] (the superscripts of $Z_s$ denote the direction of the screening current). Thus, given measurements of $\Delta \lambda_{ab}(T) = \Delta X_{ab}^s(T)/\omega\mu_0$ in the T-orientation and of the effective value $\Delta \lambda_{ab+c}(T) = \Delta X_{ab+c}^s(T)/\omega\mu_0$ in the L-orientation, we obtain

\[ \Delta \lambda_c = [(a + c) \Delta \lambda_{ab+c} - a \Delta \lambda_{ab}]/c. \tag{2} \]

This technique for determination of $\Delta \lambda_c(T)$ was used in microwave experiments [3–9] at low temperatures, $T < T_c$. Even so, it cannot be applied to the range of higher temperatures because the size effect plays an important role. Really, at $T > 0.9T_c$ the lengths $\lambda_c$ and $\delta_c$ are comparable to the sample dimensions. In order to analyze our measurements in both superconducting and normal states of BSCCO, we used formulas [15] for field distributions in an anisotropic long strip ($b \gg a, c$) in the L-orientation. These formulas neglect the effect of $ac$-faces of the crystal, but take account of the size effect. In addition, in a sample shaped as a long strip, there is a simple relation between its surface impedance components and ac-susceptibility, which is expressed in terms of parameter $\mu$ introduced in Ref. [15]:

\[ \Delta (1/Q) - 2i \delta f/f = i\gamma\mu\nu/V, \quad \chi = -1 + \mu, \tag{3} \]

\[ \chi = -1 + \mu, \tag{3} \]
where $\gamma = V H_0^2 / [\int_V H_o^2 dV] = 10.6$ is a constant characterizing our cavity [1]. At an arbitrary temperature, the complex parameter $\mu = \mu' - i\mu''$ is controlled by the components $\sigma_{ab}(T)$ and $\sigma_c(T)$ of the conductivity tensor:

$$\mu = \frac{8}{\pi^2} \sum_n \frac{1}{n^2} \left\{ \frac{\tan(\alpha_n)}{\alpha_n} + \frac{\tan(\beta_n)}{\beta_n} \right\},$$

(4)

where the sum is performed over odd integers $n > 0$, and

$$\alpha_n^2 = -\frac{a^2}{c^2} \left( \frac{i}{2} + \frac{\pi^2 \delta_{ab}^2 n^2}{c^2} \right), \quad \beta_n^2 = -\frac{c^2}{a^2} \left( \frac{i}{2} + \frac{\pi^2 \delta_{c}^2 n^2}{a^2} \right).$$

In the superconducting state at $T < 0.9 T_c$ we find that $\lambda_{ab} \ll c$ and $\lambda_c \ll a$. In this case, we derive from Eq. (4) a simple expression for the real part of $\mu$:

$$\mu' = 1 + \chi' = \frac{2\lambda_c}{a} + \frac{2\lambda_{ab}}{c}. \quad (5)$$

One can easily check up that in the range of low temperatures the change in $\Delta \lambda_c(T)$ prescribed by Eq. (5) is identical to Eq. (2). Fig.3 shows measurements of $\Delta \lambda_c(T)$ in sample #1 (circles) and sample #2 (squares) at $T < 0.9 T_c$. The open symbols plot low-frequency measurements obtained in accordance with Eq. (5), the full symbols plot microwave measurements processed by Eq. (2). Agreement between measurements of sample #2 (lower curve) is fairly good, but in fitting together experimental data from sample #1 (upper curve) we had to divide by a factor of 1.8 all $\Delta \lambda_c(T)$ derived from measurements of ac-susceptibility using Eq. (5). The cause of the difference between $\Delta \lambda_c(T)$ measured in sample #1 at different frequencies is not quite clear. We rule out a systematic experimental error that could be caused by misalignment of the sample with respect to the ac magnetic field because the curves of $\Delta \lambda_c(T)$ were accurately reproduced when square sample #1 was turned through an angle of 90° in the L-orientation. It seems more plausible that Eqs. (2) and (5), which neglect the contribution of ac-faces,
Fig.4. Temperature dependences of \( \Delta (1/Q) \) (open squares) and \(-2\Delta f/f\) (open circles) in the L-orientation of sample #1 at \( T > T_c \). Solid line shows the function \(-2\delta f(T)/f\) deriving from Eq. (3). Left-hand inset: \( \Delta (1/Q) \) and \(-2\Delta f/f\) as functions of temperature in the neighborhood of \( T_c \). Right-hand inset: \( \rho_c(T) \) in sample #1 (triangles).

yield inaccurate results concerning sample #1: its ac-faces, which have a notable area (sample #1 is thick), can host a lot of defects (for example, those of the capacitive type), and the latter can affect the character of field penetration as a function of frequency.

The curves of \( \Delta \lambda_c(T) \) at \( T < 0.5 T_c \) plotted in Fig.3 are almost linear: \( \Delta \lambda_c(T) \propto T \). The inset to Fig.3 shows the low-temperature section of the curve of \( \Delta \lambda_c(T) \) in sample #1. Its slope is 0.3 \( \mu m/K \) and equals that from Ref. [8]. Note also that changes in \( \Delta \lambda_c(T) \) are smaller in the oxygen-overdoped sample #2 than in sample #1.

We can estimate \( \lambda_c(0) \) by substituting in Eq. (1) \( \delta f(0) \) obtained by comparing of \( \Delta (1/Q) \) and \( \Delta f = \delta f - f_0 \) measurements taken in the T- and L-orientations to numerical calculations by Eqs. (3) and (4), which take account of the size effect in the high-frequency response of an anisotropic crystal. The procedure of comparison for sample #1 is illustrated by Fig.4. Unlike the case of the T-orientation, the measured temperature dependence of \( \Delta (1/Q) \) in the L-orientation deviates from \(-2\Delta f/f\) owing to the size effect. Using the measurements of \( R_s = \sqrt{\omega \mu_0/2\sigma_{ab}} \) at \( T > T_c \) in the L-orientation (Fig. 2) for determination of \( \sigma_{ab}(T) \), alongside the data on \( \Delta (1/Q) \) in the L-orientation (open squares in Fig. 4), from Eqs. (3) and (4) we obtain the curve of \( \rho_c(T) = 1/\sigma_c(T) \) shown in the right-hand inset to Fig. 4. Further, using the functions \( \sigma_c(T) \) and \( \sigma_{ab}(T) \), we calculate \(-2\delta f/f\) versus temperature for \( T > T_c \), which is plotted by the solid line in Fig. 4. This line is approximately parallel to the experimental curve of \(-2\Delta f/f\) in the L-orientation (open circles in Fig. 4). The difference \(-2(\delta f - \Delta f)/f\) yields the additive constant \( f_0 \). Given \( f_0 \) and \( \Delta f(T) \) measured in the range \( T < T_c \), we also obtain \( \delta f(T) \) in the superconducting state in the L-orientation. As a result, with due account of \( \lambda_{ab}(T) \) (the inset to Fig. 2), we derive from Eqs. (3) and (5) \( \lambda_c(0) \), which equals approximately 150 \( \mu m \) in sample #1. A similar procedure performed with sample #2 yields \( \lambda_c(0) \approx 50 \mu m \), which is in agreement with our measurements of overdoped BSCCO obtained using a different technique [12]. We also estimated \( \lambda_c(0) \) on the base of absolute measurements of the susceptibility \( \chi'_c(0) \) from Eq. (5), and we obtained \( \lambda_c(0) \approx 210 \mu m \) for sample #1 and \( \lambda_c(0) \approx 70 \mu m \) for sample #2. These results are in reasonable agreement with our
microwave measurements if we take into consideration the fact that the accuracy of $\lambda_c$ measurements is rather poor and the error can be up to 30%.

In conclusion, we have used the ac-susceptibility and cavity perturbation techniques in studying anisotropic high frequency properties of BSCCO single crystals. We have observed almost linear dependences $\Delta \lambda_c(T) \propto T$, which are in fair agreement with both experimental [7, 8, 12] and theoretical [16] results by other researchers. We have also investigated a new technique for determination of $\lambda_c(0)$, which is a factor of three higher in the optimally doped BSCCO sample than in the overdoped crystal. The ratio between the slopes of curves of $\Delta \lambda_c(T)$ in the range $T \ll T_c$ is the same. These facts could be put down to dependences of $\lambda_c(0)$ and $\Delta \lambda_c(T)$ on the oxygen content in these samples. At the same time, we cannot rule out influence of defects in the samples on $\lambda_c(0)$, even though their quality in the $ab$-plane is fairly high, according to our experiments. In order to draw ultimate conclusions concerning the nature of the transport properties along the $c$-axis in BSCCO single crystals, studies of more samples with various oxygen contents are needed.

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