

# Magnetic penetration depth in KBaBiO from measurements of the dynamic susceptibility of powder in an alternating magnetic field

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The temperature dependence of the magnetic penetration depth,  $\lambda(T)$ , was determined from measurements of the dynamic susceptibility of superconducting KBaBiO powder. The coupling constant,  $\Lambda \approx 2$ , was estimated from the behavior of this function near  $T_c$ , i.e., this constant is approximately the same as in YBaCuO. In the low-temperature region  $T < T_c/2$  no power-law corrections to the function  $\lambda(T)$  were found.

## 1. INTRODUCTION

The results of many measurements of the temperature dependence of the magnetic penetration depth  $\lambda(T)$  in the high-temperature superconductor YBaCuO comprise two important elements.

The first important result of such measurements is the estimate of the coupling constant  $\Lambda$  in YBaCuO from the  $\lambda(T)$  dependence near  $T_c$ . According to Gantmakher *et al.*<sup>1</sup> the constant  $\Lambda \approx 2$ , i.e., the coupling should be considered strong.

Also, even in the first work,<sup>2–4</sup> where the value of

$$\frac{\Delta\lambda}{\lambda(0)} = \frac{\lambda(T) - \lambda(0)}{\lambda(0)}$$

was measured at low temperatures  $T < T_c/2$ , it was found that it decreases according to a power law. One would expect such a behavior of the function  $\lambda(T)$  if the gap vanishes at some points or lines on the Fermi surface. Doubts about whether the gap in YBaCuO really vanishes somewhere became associated unjustifiably with the experimental observation of the  $\lambda(T)$  dependence. However, the existence of a power law term in  $\lambda(T)$  was confirmed in recent work.<sup>1,5</sup>

In addition, it was shown numerically<sup>1,6</sup> and subsequently also analytically<sup>7,8</sup> that the power term in  $\lambda(T)$  does not necessarily indicate vanishing of the gap even at separate points on the Fermi surface; it can be due to a large coupling constant  $\Lambda$  for interaction with acoustic phonons.

The large value of  $\Lambda$  indicates that we are dealing with a single quantum system “electrons plus lattice” with two excitation branches: electronic with the gap and a gapless phonon branch. The existence of the gapless branch is also the source of the power-law corrections to the system’s thermodynamic parameters at low temperatures. These corrections are greater the larger the coupling constant  $\Lambda$  and the larger the ratio of the superconducting transition temperature  $T_c$  to the Debye temperature  $\Theta$ . We note that the parameters  $\Lambda$ ,  $\Theta$  and  $T_c$  should be considered independent. In two superconductors with identical  $\Lambda$  and  $\Theta$  the transition temperature  $T_c$  can be different owing, for example, to differences in the interaction with the optic branches.

The dependence on the ratio  $T_c/\Theta$  can be explained in the following way. A large  $\Lambda$  indicates strong scattering by thermal phonons. This can convert the specimen gradually from the clean limit  $l \gg \xi_0$  at  $T = 0$  to the dirty limit  $l < \xi_0$  ( $l$  is the mean free path and  $\xi_0$  is the correlation length at  $T = 0$ ). The existence of a finite free path leads to an increase of  $\lambda$  by

$(1 + \xi_0/l)^{1/2}$  times (Ref. 9) and correspondingly to a power law dependence  $\lambda(T)$  because of the  $l(T)$  dependence. Since the number of thermal phonons at a given  $T$  is proportional to  $\Theta^{-3}$ , the effect can be noticeable only for sufficiently large  $T_c$ , in practice for  $T_c \approx \Theta$ . This explains why the question of a power term in  $\lambda(T)$  has never arisen for ordinary superconductors.

What has been said above is the motivation for the experiments described in the present work. Measurements of  $\lambda(T)/\lambda(0)$  were undertaken for another cubic superconductor KBaBiO, using the same method as before:<sup>1</sup> by measuring the dynamic susceptibility of a fine powder. This method goes back to Shoenberg’s work.<sup>10</sup>

KBaBiO belonging to the category of high-temperature superconductors,<sup>11</sup> its possession of a simple cubic structure and a whole array of interesting physical properties<sup>12–14</sup> make this compound extremely attractive for experimental and theoretical investigations. The phase diagram for  $K_x Ba_{1-x} BiO_3$  has been studied in detail and it has been shown that the compound is superconducting for  $x > 0.3$ , while increasing  $x$  from 0.3 to 0.5 decreases the superconducting transition temperature roughly from 30 to 18 K (Refs. 15, 16). The temperature dependence of the gap agrees with the BCS theory while the value of  $2\Delta/kT_c$  is a little larger than the theory demands.<sup>17</sup> The magnitude and temperature dependence of the magnetic penetration depth in this compound has not appeared in the literature, as far as we know.

## 2. THE EXPERIMENT

Powder of the superconducting compound KBaBiO was obtained in the following way.<sup>18</sup>

A mixture of the oxides BaO<sub>2</sub> and Bi<sub>2</sub>O<sub>3</sub>, taken in the stoichiometric ratio 1:0.75 for specimen 1 and 1:1 for specimen 2, was ground thoroughly in an agate mortar, granular KOH was added in molar ratios to the oxides 2.7:1.75 and 5.6:2, and was again homogenized. The mixture was then placed in an alundum crucible and heated in air at 450 °C for an hour. After this the crucible was cooled to room temperature with the oven switched off. Without removing it from the oven, the mixture was heated additionally in an oxygen stream (1 atm) at 450 °C for 0.5 h and also cooled slowly. The product of the synthesis was a solidified KOH melt ( $T_m = 350$  °C) with a finely divided black powder uniformly distributed in it. The excess KOH was removed by a single

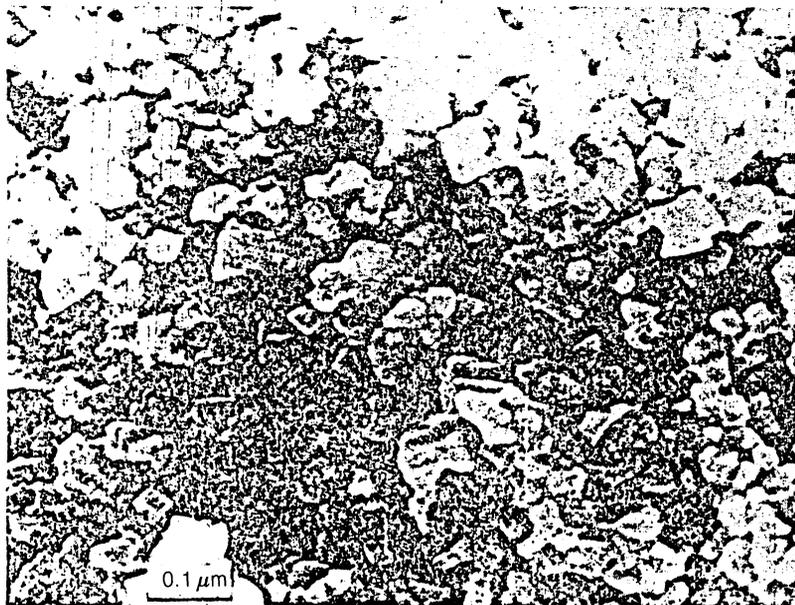


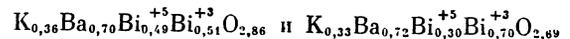
FIG. 1. Appearance of the powder particles in the electron microscope for specimen 1.

treatment with ethyl alcohol. The product, cleaned of KOH, was dried in a dessicator (100 °C) in air. The final product was a finely divided, friable, black powder. X-ray analysis confirmed the existence of a single-phase cubic structure with lattice parameter  $a = 4.293$  (specimen 1) and  $4.283 \pm 0.005$  Å (specimen 2). The absence of bismuth-rich phases in the x-ray powder picture confirmed their removal by the alcohol treatment.

The composition of a specimen was determined from results of local x-ray spectral analysis (LXSA) on a Camebax apparatus with a Link comparison system (probe area 3 μm, accuracy in analysis  $\pm 5$  rel. %); not less than ten points on the specimen surface were analyzed. Taking the concentration of Bi as unity, the mean values obtained for potassium and barium in specimen 1 correspond to the formula  $K_{0.36}Ba_{0.70}BiO_y$  (with a spread in K concentration from 0.31 to 0.42) and  $K_{0.33}Ba_{0.72}BiO_y$  (with a larger spread of K from 0.25 to 0.45) in specimen 2. The fact that the sum of the coefficients for K and Ba differ from unity is probably due not to inaccuracy of the measurements but to the presence of excess K in the interstices of the lattice or to

the presence of vacancies. It is very likely that it is just this difference between the sum of the coefficients and unity that has a strong influence on  $T_c$ .

The oxygen concentration was determined from the  $Bi^{5+}$  ion content in the specimen<sup>19</sup> yielding composition a formula in the form



for specimens 1 and 2 respectively.

The size distribution of particles in the powder (Fig. 2) for specimen 1 was constructed from an electron microscope picture (Fig. 1).

Either pressed powder or powder loosely poured into a cylindrical container was used in the measurements (the powder was pressed in a tube under a pressure of 200 kg/cm<sup>2</sup> to obtain compact specimens). The measurements were carried out in the standard way. The specimen was inserted inside one of two induction coils connected in opposition and the out-of-balance signal was measured.

The signal appearing when the specimen was inserted into the coil is caused by the total magnetic moment

$$M = H \sum \chi_p v_p = H \chi V, \quad (1)$$

where  $v_p$  is the volume of a single particle,  $H$  is the magnetic field in which it is located,  $\chi_p$  is the susceptibility of a particle with its demagnetizing factor taken into account,  $\chi$  is the mean susceptibility of powder particles,  $V = P/\eta$  is the volume of powder determined from its weight  $P$  and its "ideal" density  $\eta = 7.45$  g/cm<sup>3</sup>. For ideal diamagnetic ( $\lambda = 0$ ) spherical particles

$$\chi = \chi_p = \chi_{max} = -3/8\pi.$$

For calibration, the magnitude of  $U_{cal}$  was measured, the signal appearing on inserting a lead cylinder of the same dimensions as the specimen being measured and at the subsequent superconducting transition. This allowed geometrical factors due to the ratio of coil and specimen dimensions to be accounted for automatically. Relating the measured signal

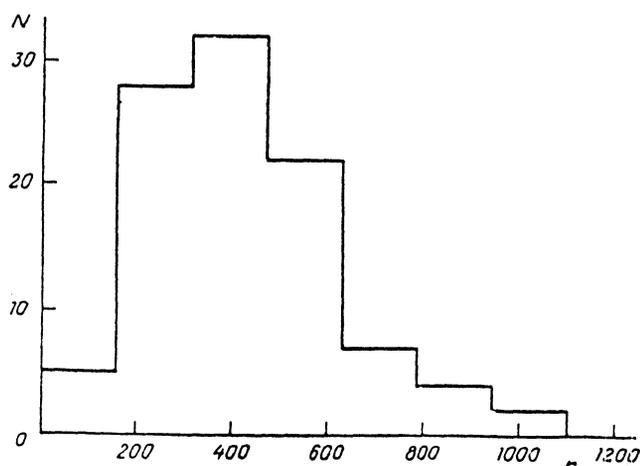


FIG. 2. Size distribution of particles for specimen 1.

to the signal from a known susceptibility  $V_0/4\pi$  ( $V_0$  is the volume of the lead cylinder), we obtain

$$\frac{\chi}{\chi_{\max}} = \frac{U}{U_{\max}} = \frac{U}{U_{\text{cal}}} \frac{1/4\pi V_0}{\chi_{\max}} = \frac{2}{3} \frac{V_0}{V} \frac{U}{U_{\text{cal}}} \quad (2)$$

Here  $U_{\max}$  is the magnitude of the signal on complete expulsion of the flux from the material ( $\lambda = 0$ )

$$U_{\max} = \frac{3}{2} \frac{V}{V_0} U_{\text{cal}},$$

where the factor  $2/3$  is the result of assuming that the particles are spherical. The smallness of the ratio (2) (see below) obtained in the experiment, together with the small demagnetizing factor due to the external specimen shape, allows us to assume that  $H$  entering Eq. (1) is the external magnetic field and that the mutual influence of particles on one another can be neglected (cf. the accurate formula in Ref. 20).

The results of measurements for pressed and for powders freely poured into a tube were identical, which confirmed that even in a pressed specimen there were no conducting contacts between particles. A 100-fold increase in the amplitude of the alternating field did not affect the temperature dependence  $\lambda(T)$ . In the terminology of Gantmakher *et al.*<sup>20</sup> we were always concerned with the "lower limiting curve," when contact between particles does not prevent penetration of the field.

### 3. RESULTS

The magnetic moment of sufficiently small particles with characteristic dimension  $r < \lambda$  is described with high accuracy by the quadratic function<sup>10</sup>

$$\chi/\chi_{\max} \approx k(r/\lambda)^2, \quad (3)$$

where the coefficient  $k$  depends on the shape (for example, if  $r$  is the radius of a sphere, then  $k = 1/15$ ). It follows from this that the measured ratio

$$\frac{\chi(T)}{\chi(0)} = \left[ \frac{\lambda(T)}{\lambda(0)} \right]^{-2} = L^{-2}, \quad L(T) = \frac{\lambda(T)}{\lambda(0)} \quad (4)$$

is independent of both the magnitude of  $r$  and of the coefficient  $k$  which depends on the shape of the particles. That the condition  $r < \lambda$  is really satisfied is confirmed by the fact that  $\chi^{(0)}/\chi_{\max} \approx 0.016$  for all specimens. Assuming that the particles are spherical and  $k = 1/15$ , we obtain for all temperatures

$$r/\lambda < r/\lambda(0) \approx 0,49 < 1. \quad (5)$$

According to Eq. (1) the contribution of an individual particle is proportional to its volume  $v \sim r_p^3$  and the susceptibility  $\chi \sim (r_p/\lambda)^2$ , so that  $r_p^5$  must be averaged to obtain the characteristic dimension  $r_{\text{eff}}$  of the particles. Using the size distribution (Fig. 2) we obtain  $r_{\text{eff}} \approx 650 \text{ \AA}$  and correspondingly  $\lambda(0) \approx 1300 \text{ \AA}$ . This is, of course, only a rough estimate, in particular because there is no certainty that there are not among the powder particles some which just do not become superconducting. However, there is no indication of such a specimen defect even in the measured temperature dependence  $\lambda(T)$ , i.e., in the function (4): all that is required is absence of large particles for which the inequality (5) is violated. We now pass to consideration of this dependence.

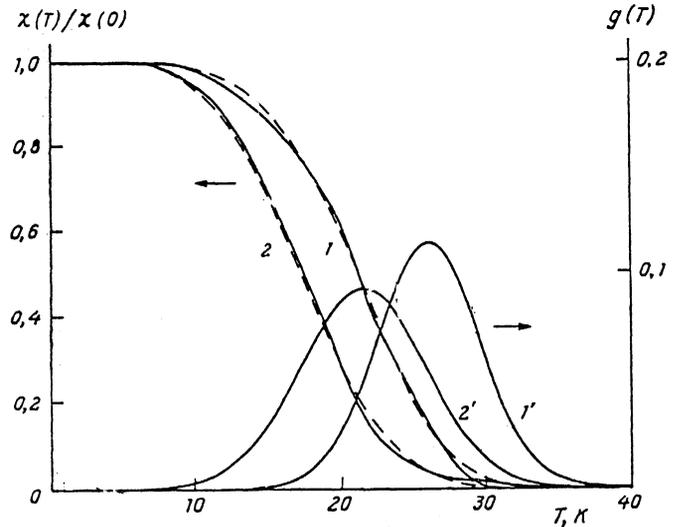


FIG. 3. Temperature dependence of the normalized susceptibility  $\chi(T)/\chi(0)$  for specimens 1 and 2 (continuous lines); dashed lines indicate the theoretical dependences for the optimal values of  $\Lambda$ ,  $\tau$  and  $T_0$ ;  $1'$  and  $2'$  are the calculation transition temperature distribution functions of the particles.

#### 3.1. The neighborhood of the transition temperature

According to theory,<sup>21</sup> the function  $L^{-2}$  has in any case a finite derivative at  $T = T_c$  if fluctuations are not taken into account and the possibility of a zero dimensionality  $\xi_0 \gg r$  is ruled out<sup>22</sup> ( $\xi_0$  is the correlation length at  $T = 0$ ). In some interval  $\Delta T$  this derivative is constant [i.e.,  $\chi(T) \sim L^{-2}(T) = \beta(T_c - T)$ ] and on lowering  $T$  further the modulus of the derivative  $d(L^{-2})/dT$  must decrease. The magnitude of the derivative depends on the phonon spectrum and the coupling constant. For the Éliashberg function in the form

$$\alpha^2 F(\omega) = 1/2 \Lambda \omega_n \delta(\omega - \omega_n) \quad (6)$$

the dependence of  $d[L^{-2}(T)]/dT$  on  $\Lambda$  is given in Fig. 4 of Ref. 1, where Fig. 2 shows also a plot of  $L^{-2}(T/T_c)$  for  $\Lambda = 2$ .

The experimental curves of  $\chi(T)$  for KBaBiO in Fig. 3 appear somewhat different near  $T_c$ . This is doubtless associated with the fact that there is a scatter in the values of  $T_c$  for different grains. This scatter is due to fluctuations in the concentration of K and Ba.<sup>15,16</sup>

On introducing the distribution function for the transition temperatures  $g(T)$  we obtain instead of Eq. (4)

$$\frac{\chi(T)}{\chi(0)} = \int_T^\infty g(t) L^{-2}(T/t) dt. \quad (7)$$

If the effective width of the scatter  $\tau < \Delta T \approx 0.15 T_c$ , then  $\beta(t - T)$  can be substituted in Eq. (7) instead of  $L^{-2}$  and then

$$g(T) = \frac{1}{\beta} \frac{d^2}{dT^2} \left[ \frac{\chi(T)}{\chi(0)} \right],$$

and the maximum of the modulus of the derivative  $D = d[\chi(T)/\chi(0)]/dT$  is determined by the magnitude of the coupling constant  $\Lambda$ .<sup>1,21</sup> In our case  $\tau \sim \Delta T$ . Therefore  $D$  only gives a lower estimate for  $\Lambda$ . We find from Fig. 3 that  $\Lambda > 1$ .

For a more exact comparison we took  $g(T)$  in the form of a normal Gaussian distribution

$$g(T) = (2\pi)^{-1/2} \tau^{-1} \exp[-(T-T_0)^2/2\tau^2] \quad (8)$$

and found for the parameters  $\Lambda$ ,  $\tau$ , and  $T_0$  values which lead to agreement with the experimental results (see Fig. 3). It turned out that for all specimens  $\Lambda = 2$ , as in the case of YBaCuO (Ref. 1).

### 3.2. Low-temperature region

Unlike the region near  $T_c$ , the accuracy in measurements of  $\lambda(T)/\lambda(0)$  at low temperature depends much less on the existence of a scatter in the transition temperatures of the particles. In fact, the magnitude of  $\lambda(T)/\lambda(0)$  depends weakly on  $T_c$  for  $T < 0.5T_c$ , so that if the characteristic width of the distribution function is small,  $\tau \ll T_0$ , then at low temperatures the measured value of  $\lambda(T)/\lambda(0)$  behaves just as though all the particles had a transition temperature  $T_0$ . In other words, far from  $T_0$  we cannot feel the presence of a scatter of the transition temperatures of the particles. The section of the  $\lambda(T)/\lambda(0)$  curve at low temperatures is thus the most accurate result of the experiment.

In order to underline the difference between YBaCuO and KBaBiO, the experimental dependences  $L(T/T_c)$  are shown together for these compounds in Fig. 4 (for KBaBiO we put  $T_c = T_0$ ). One comment should, however, be made here. In the expression for  $\lambda$  discussed by Gantmakher *et al.*,<sup>1</sup>

$$\lambda(T) = \lambda(0) [1 + (\Delta/T)^{1/2} \exp(-\Delta/T)] (1 + \xi_0/l)^{1/2} \quad (9)$$

the last factor has a scaling temperature which is not  $T_c$  but  $\Theta$ . The Debye temperature for both compounds should be about the same. It would therefore seem that to compare  $\lambda(T)$  in these two compounds the abscissa should not be  $T/T_c$  but  $T/\Theta$ , i.e., just  $T$ . The two curves would then almost coincide.

We approached this question from another angle.

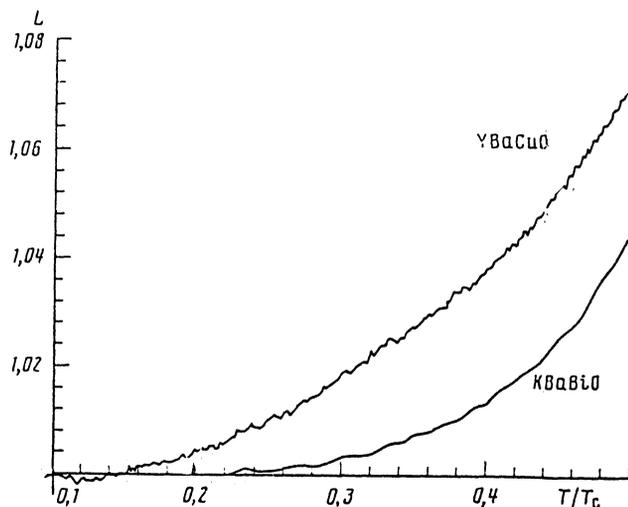


FIG. 4. The low-temperature part of the dependence of the normalized penetration depth on the reduced temperature for YBaCuO (from Gantmakher *et al.*<sup>1</sup>) and KBaBiO. (The temperature  $T_0$  of the maximum of the function  $g(T)$  was taken as  $T_c$ .)

To describe the experimental curve for YBaCuO it was necessary<sup>1</sup> to introduce the acoustic branch with  $\Theta = T_c$  into the Eliashberg function. It was possible to confine oneself to the model in Eq. (6) for KBaBiO, i.e., only to the optic branch; this can be seen from Fig. 3. In other words, the experimental dependence  $\lambda(T)$  could be described without taking account of the last factor in Eq. (9). The ratio  $T/\Theta$  enters in the power-law increment  $\Delta\lambda$  to a high degree, since according to Eliashberg *et al.*<sup>8</sup> the value of the factor  $(1 + \xi_0/l)^{1/2}$ , is the transport scattering length. Even a value  $T_c/\Theta \sim 0.3$  thus makes it impossible to separate out the power-law correction against the background of the exponential term, at least at our experimental accuracy.

### 4. CONCLUSIONS

1. A method is described for low-temperature synthesis of KBaBiO powders with small particle dimensions and fairly narrow superconducting transition temperature distribution for the particles.

2. The magnitude of the coupling constant  $\Lambda$  in KBaBiO cannot be considered small as in the BCS theory, i.e.,  $\lambda(T)$  is well described by strong coupling equations, as in the case of YBaCuO.

3. At low temperatures,  $T \ll T_c$ , the value of  $\Delta\lambda/\lambda(0)$  decreases exponentially with temperature as in usual superconductors. If it is assumed that the nature of the superconductivity in all high-temperature superconductors is the same, then this indicates that the mechanism of superconductivity is the usual one. In any case, judging from our experiments there is no basis for talking of the gap vanishing anywhere on the Fermi surface in KBaBiO. At the same time, this result confirms indirectly the explanation given by Gantmakher *et al.*<sup>1</sup> for the low-temperature dependence  $\lambda(T)$  in YBaCuO which has been mentioned above.

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