Physica C 171 (1990) 223–230
North-Holland

Temperature dependence of the magnetic field penetration depth in $YBa_2Cu_3O_{7-\delta}$ measured on ultra fine powder

V.F. Gantmakher, N.I. Golovko, I.G. Naumenko, A.M. Neminsky and A.V. Petinova
Institute of Solid State Physics, Academy of Sciences of the USSR, Chernogolovka, Moscow District, 142432, USSR

Received 15 August 1990

The temperature dependence of the magnetic field penetration depth $\lambda(T)$ in superconducting $YBa_2Cu_3O_{7-\delta}$ has been determined by measuring the AC susceptibility of fine powder at the frequency 10^5 Hz. The reason for noticeable changes in λ at low temperatures has been found out: strong scattering of carriers by low-frequency excitations. The coupling constant which controls the superconductivity has been estimated. At low temperatures a paramagnetic signal superimposed to the superconductivity one has been observed. It is probably due to alteration in the interaction of the magnetic moments near the surface of the particles.

1. Introduction

In spite of the large number of investigations, the electrodynamics of $YBa_2Cu_3O_{7-\delta}$ in a superconducting state has not been elucidated so far. For instance, the temperature dependence of the magnetic penetration depth $\lambda(T)$ is still to be clarified [1]. Numerous measurements made by different methods (muon spin resonance [2,3], magnetization in weak [4] and strong [5] fields, surface impedance at high frequencies [6–8], etc.) cause some doubts concerning both the presence of a power term in the dependence $\lambda(T)$ at low temperatures and the behaviour of the function $\lambda(T)$ in the vicinity of T_c. In particular, the measurements of AC susceptibility on fine powders [9] gave an extremely great value of $\lambda(O)$ and a large quadratic term at low temperatures.

To get some additional information on the function $\lambda(T)$ we measured the surface impedance (AC susceptibility) of a $YBa_2Cu_3O_{7-\delta}$ fine powder at the frequencies 10^4–10^5 Hz.

2. Experimental

The powder was produced by the spark erosion method [10]. The electrodes to which electric pulses were applied were made of $YBa_2Cu_3O_{7-\delta}$ ceramics and immersed into the liquid oxygen. When pulses power, duration and shape were chosen adequately, the substance evaporating from electrodes was condensed into almost spherical particles of the same chemical composition as that of the electrodes.

The average size of particles in the powder was determined by measuring the specific surface area with the help of the polyatomic adsorption method. The particles had the orthorhombic structure with the parameters $a=3.83$ Å, $b=3.88$ Å and $c=11.16$ Å. No other phases have been observed within the accuracy of the X-ray phase analysis.

The oxygen content was found by the iodometric analysis to be $\delta=0.17$. The parameters of the powder were stable at least during a year if the powder was kept in an argon atmosphere. When the powder was subjected to the pressure of up to 2 kbar, solid samples produced were insulating. This may be the result of the oxygen deficiency in the surface layer of the particles. In this case the measured δ value is a mean one only.

To perform the measurements, the powder was loosely settled in a cylindrical container, the filling factor being about 0.25. To make sure that there were no electric contacts between the particles, in other words, that no superconducting macroloops existed, which would shield the inner part of the container from the penetration of the alternating field, a test mixture of the powder under examination with ZrO_2 powder (with 4:1 weight ratio) was fabricated. A relative volume occupied by a conducting powder in the container with the mixture was $x \approx 12\%$ only, that is, it was below the percolation threshold $x_{cr} \approx 15\%$. The signal from the mixture decreased by the ratio equal to that of the weights of the conducting fraction. This means that even in an undiluted sample contacts between the particles were of no importance [9]. The absolute value of the diamagnetic signal $U(O)/U_{max}$ at low temperatures, determined below (see eq. (3)), served as an extra check.

It should be noted that not all the powder samples could stand up to this set of tests. It seems that sometimes particles adhere into clusters at the stage of powder production. All the results presented below were obtained on the powder of one batch with average particles size $2r = 1920 \pm 40$ Å.

After fabrication the powder was not subjected to any additional thermal treatment. Moreover, an attempt to anneal it in oxygen at $T = 400°C$ for 10 h resulted in emergence of other phases and in broadening of the superconducting transition.

The sample was placed inside one of the two identical couples of induction coils connected towards each other and the disbalance signal was measured. To reduce the change of the background disbalance signal, the coils were disposed in liquid helium over the external surface of a small overturned dewar. The sample together with a thermometer and a heater were placed inside the dewar in gaseous helium. To prevent temperature gradients on the sample, the container for the powder was made of sapphire, its inner diameter being 7 mm, the outer one 9 mm, the height about 10 mm.

Signal U, arising upon introducing the container with the powder into the coil, is caused by the total magnetic moment

$$M = H \cdot \sum \chi \cdot v = H \chi V_p .$$

Here v is the volume of a separate particle, $V_p = P/\eta$ is the volume of the powder determined through its weight P and the "ideal" density $\eta = 6.3$ g/cm^3, H is the external magnetic field and χ is the susceptibility of a particle with its demagnetization factor being taken into account. For a spherical particle when the penetration depth $\lambda = 0$ the value of χ is:

$$\chi = \chi_{max} = -3/8\pi . \qquad (1)$$

For calibration we used the quantity U_{cal} of the signal arising upon introducing the superconducting lead sample with sizes equal to inner sizes of the powder container. This made it possible to take into account the geometric factors resulting from the ratios of the coil-to-sample sizes. Comparing the signal from the powder with that from the sample with the known susceptibility, $-1/4\pi$, we get

$$\frac{\chi}{\chi_{max}} = \frac{U}{U_{max}} = \frac{U}{U_{cal}} \frac{(1/4\pi)}{\chi_{max}} \frac{V}{V_p} = \frac{2}{3} \frac{V}{V_p} \frac{U}{U_{cal}} . \qquad (2)$$

Here V is the volume of a lead sample and U_{max} is the magnitude of the signal, when the flux is completely extruded from the powder ($\lambda = 0$)

$$U_{max} = \frac{3}{2} \frac{V_p}{V} U_{cal} , \qquad (3)$$

and the factor $\frac{2}{3}$ arises on assumption of sphericity of the particles.

Susceptibility χ entering eqs. (1) and (2) describes the magnetic moment produced by the superconducting currents. If the ionic lattice in the surface layer λ has magnetic susceptibility $\chi_{mgn} \neq 0$, then being accessible to the alternating field it provides contribution to the magnetic moment of the sample, which comes both from the susceptibility itself and through its influence on λ. (For instance, under normal skin-effect conditions the thickness of the skin-layer δ is $\delta \sim \mu^{-1/2}$; here $\mu = 1 + 4\pi\chi_{mgn}$.)

The dependence of λ on the susceptibility of the ionic lattice has not been investigated adequately so far. However, such a dependence, if at all, should affect the value of the magnetic flux passing through the sample only when $\lambda \ll r$, $2r$ being a typical size of the sample: $2r \approx v^{1/3}$. As will be shown below, in our case we had $\lambda \geq r$. Under these conditions one may take account only of alteration in the AC field amplitude by a factor $1 + 4\pi(1-n)\chi_{mgn}$ (n is the demagnetization factor). As a result, the magnetic flux $\Phi(T)$, cophased with the external field and normalized over the initial unperturbed flux through the volume occupied by the sample, can be expressed rather precisely in the form (instead of eq. (2)):

$$\Phi = (1 + \tfrac{8}{3}\pi\chi_{mgn}) \cdot (1 - \chi/\chi_{max})$$
$$= (U_{max} - U/U_{max}) . \qquad (4)$$

It can be seen from the next section that the experiment proves the existence of the magnetic contribution to Φ.

The amplitude of the AC field in the main experiments was about 0.08 Oe, the frequency $\omega/2\pi=10^5$ Hz. Some special experiments confirm that there were no nonlinear effects at these amplitudes. Measurements in a magnetically shielded dewar showed that the earth's magnetic field did not matter.

3. Results

Figure 1 depicts an experimental line $\Phi(T)$ characterizing the magnetic flux passing through the assembly of the powder particles. Moving toward lower temperatures one can separate the line up into three domains:

(1) Increase in the vicinity of T_c (see inset).

(2) A descending part where the penetration depth $\lambda(T)$ decreases gradually.

(3) Increase of $\Phi(T)$ in the low temperature range, where $\lambda(T)$ for certain should not vary with temperature.

The investigation of the dependence $\Phi(T)$ in the close vicinity of T_c is by far too incomplete. From the experimental point of view there is an uncertainty arising from the lack of independent measurements of the T_c position. A similar vagueness exists on the ordinate axis: the uncertainty of the position of the point $\Phi=1$ on the axis is comparable to the height of the maximum, that is, it is not clear whether the maximum exceeds the shielding diamagnetic signal induced by the skin-currents in the normal state.

Earlier a similar maximum $\Phi(T)$ was observed upon transition to the superconducting state of fine lead powder with particles of $r\approx 3\mu k$ in radii [11]. As was the case in our experiment, the width of the maximum had been $\Delta T/T_c\approx 0.01$ and its height $\Delta\Phi$ had been also about 0.01. This indicates that the maximum is not related to magnetic moments in the lattice of a superconductor.

The most reliable statement that can be done is as follows. The arising of the maximum is conditioned by small sizes of the particles. No analogous observations on films or massive superconductors are available. The absence of maximum in the measurements of $\chi(T)$ on aluminium powders performed by SQUID magnetometer [12] shows that this phenomenon is related to existence of the AC field. However, no successive calculations of the impedance of small particles have been made for the fluc-

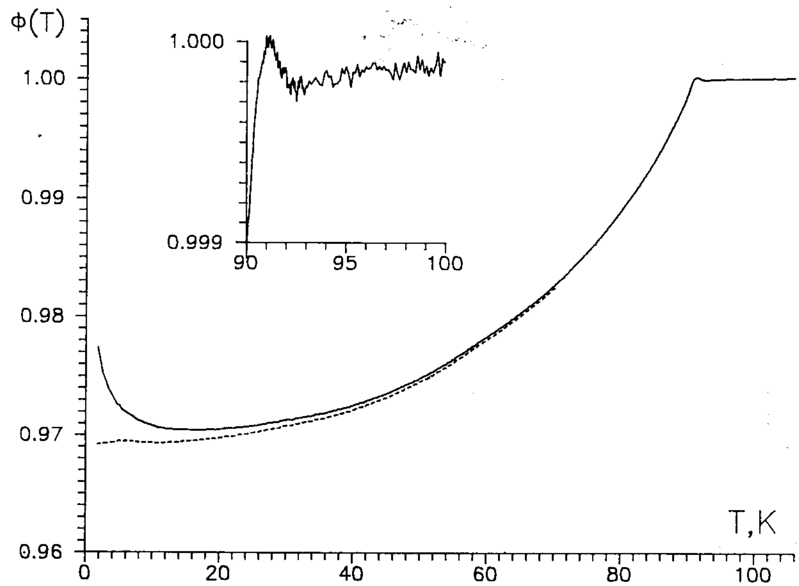

Fig. 1. Temperature dependence of magnetic flux through the powder. Dashed line: the result of dividing by "paramagnetic" factor (see eqs. (4) and (5)).

tuation region. Therefore, this problem is still to be investigated both theoretically and experimentally.

It is easy to separate the contributions to Φ from χ and χ_{mgn}, since they change in different temperature ranges. Plotting Φ as a function of T^{-1}, one may see that at $T \leq 15$ K the Curie law takes place

$$\chi_{mgn} = C/T, \qquad C = 1.1 \times 10^{-3} [\mathrm{K\,sm^{-3}}]. \tag{5}$$

The absolute value of C was obtained by a comparison with signal U_{max}, corresponding to complete extrusion of the flux from the volume V_p. Then dividing Φ by $(1+\frac{8}{3}\pi\chi_{mgn})$, we get function $(1-\chi/\chi_{max})$ at low temperatures. In fig. 1 it is drawn by a dashed line.

The next section will be devoted to the discussion of temperature dependence $\chi(T)$ so obtained. And here we shall discuss in brief the origin of the paramagnetic signal.

As is known, the paramagnetic susceptibility χ_{mgn} is related to the concentration of paramagnetic centers N_{mgn} and to their effective moment $p\mu_B$ by the relation:

$$\chi_{mgn} = N_{mgn}(p\mu_B)^2/3T.$$

Here μ_B is the Bohr's magneton; coefficient p is related to the values of spin s and of g-factor g:

$$p = g[s(s+1)]^{1/2}.$$

From the absolute value of χ_{mgn}, assuming $s=\frac{1}{2}$ and $g=2$, we get that the relative concentration of paramagnetic centers in reference to the total concentration of Cu atoms is:

$$n_{mgn} = N_{mgn}/N_{Cu} \approx 0,1. \tag{6}$$

The value obtained in eq. (6) is too large to attribute the paramagnetic effect to noncontrolled defects or to the admixture of a nonsuperconducting phase. At the same time the value is too low to suggest the existence of free magnetic moments of copper ions. Also, such free moments should have been observed, for instance, while measuring the heat capacity. Therefore, the assumption that the paramagnetic signal is somehow connected to magnetic properties of the surface layers of the particles seems to be the most natural one. A comparison of n_{mgn} with the relative number of copper atoms in the surface layer of h in thickness: $n_{mgn} = 3h/r$ gives $h = 32$ Å, that is, about 3 or 4 elementary cells. Such an effect was earlier observed on vanadium powders [13]: the susceptibility of powder obeyed the Curie law with the coefficient inversely proportional to the mean particle size r.

Detailed experiments are required to draw final conclusions on the origin of the paramagnetic signal.

4. Temperature dependence $\lambda(T)$

A common difficulty in processing experiments with powders is that there is always some uncertainty in respect to the shape and size of particles. However, at a sufficiently small value of $r \leq \lambda$ this uncertainty does not prevent one from measuring the dependence $\lambda(T)$. Indeed, the magnetic moment m of a sphere of radius r in field H and its susceptibility $\chi = m/H_v$ at $r < \lambda$ are described with a good accuracy by the quadratic function [14]:

$$\chi/\chi_{max} = \tfrac{1}{15}u^2, \qquad u = r/\lambda. \tag{7}$$

Using the limiting value of $\chi(0)$ we get from eq. (7) the relation:

$$\frac{\chi(T)}{\chi(0)} = \left[\frac{\lambda(T)}{\lambda(0)}\right]^{-2} \equiv L^{-2}, \qquad L = \frac{\lambda(T)}{\lambda(0)}, \tag{8}$$

which involves neither quantity r, nor the numerical coefficients from eqs. (2) and (7), stipulated by the particle shape. It can be confirmed experimentally, that in our experiments $u<1$ and that eq. (7) is applicable. Indeed, from the value of $1-\Phi(0)$ taken from the graph in fig. 1 one can calculate $u(0) \approx 0.67$. At this u value the value of χ given by eq. (7) differs from the accurate one by 4% [14].

Applying the measured value of $r = 960$ Å, we get $\lambda(0) = 1430$ Å. However, this value, unlike all further results, depends both on r and on the assumption concerning the particle shape.

Figure 2 presents a comparison of the experimental data with three theoretical lines. It is clear that the experiment allows a choice between them. Further discussion will be carried out separately for different regions of variation of $t = T/T_c$.

Region $t<0.4$. As a rule, the variation of $\lambda(T)$ at low temperatures in high-temperature superconductors is discussed with aspiration to choose between an exponential variation of the type

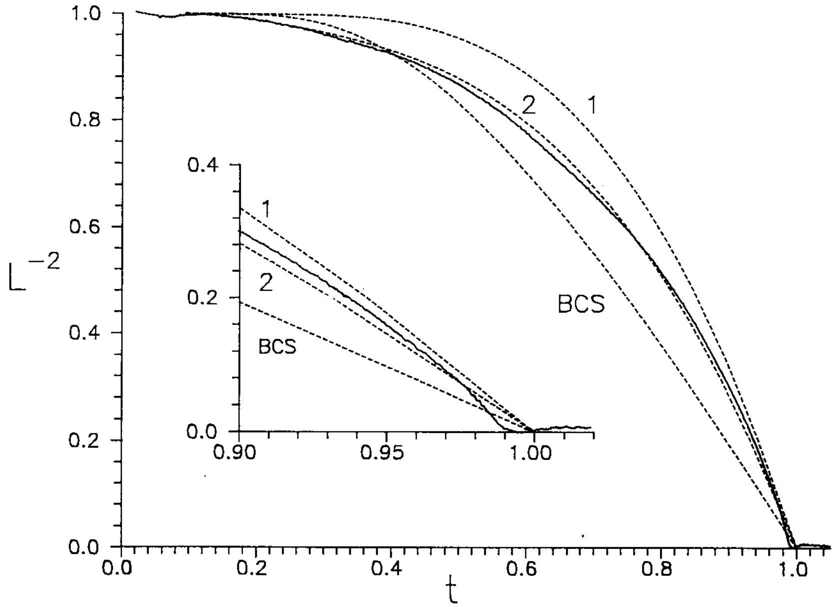

Fig. 2. Temperature dependence of inversed square of normalized penetration depth L^{-2}. Dashed lines are calculated in BCS model (line BCS) and using Eliashberg function (13) with $\Lambda_0=2, \Lambda_1=0$ (line 1) and with $\Lambda_0=2, \Lambda_1=1, \omega_D=\omega_0/5$ (line 2).

$$\Delta\lambda/\lambda=\lambda(T)-\lambda(0)/\lambda(0) \propto (\Delta/T)^{1/2}\exp(-\Delta/T) \quad (9)$$

(T in energy units) and a power law

$$\Delta\lambda/\lambda \propto t^{\nu}, \qquad \nu>0. \quad (10)$$

The latter would have given evidence of turning the gap Δ to zero at some points or lines on the Fermi surface [1]. However, Dolgov, Golubov and Koshelev (DGK) mentioned in [15] another possible line of reasoning in considering the temperature dependence $\lambda(T)$. As it known, the interaction of electrons with virtual phonons defines the transition temperature T_c, and that with thermal phonons leads to the scattering. Provided the coupling constant is large, then with increasing of the temperature the scattering on thermal (acoustic) phonons may gradually transfer the sample from the pure limit $\ell \gg \xi_0$ at $T=0$ to the dirty one $\ell \leq \xi_0$ (ℓ is the mean free path, ξ_0 is the zero-temperature coherence length). In this case λ increases by a factor $(1+\xi_0/\ell)^{1/2}$ [16]. Since length ℓ depends on T, then, apart from unimportant numerical factors,

$$\lambda(T)=\lambda(0)[1+(\Delta/T)^{1/2} \times\exp(-\Delta/T)](1+\xi_0/\ell)^{1/2}, \quad (11)$$

and with $\ell>\xi_0$ and $T<\Delta$ we get

$$\Delta\lambda/\lambda(0)\approx\xi_0/2\ell. \quad (12)$$

Therefore, the power law which, as a rule, describes ℓ dependence on T leads to eq. (10). According to fig. 3, at $t=0.4$ the change in the penetration depth is $\Delta\lambda/\lambda(0)\approx 0.04$. Hence, it follows:

$$\ell(35\,\mathrm{K})\approx\xi_0/0.08\approx 200\,\text{Å}.$$

To show the effect of scattering under strong coupling DGK performed calculation applying the Eliashberg function in the form [15]:

$$\alpha^2F(\omega)=\Lambda_1\left(\frac{\omega}{\omega_D}\right)^2\theta(\omega-\omega_D)+\frac{\Lambda_0\omega_0}{2}\delta(\omega-\omega_0). \quad (13)$$

($\theta(x)=1$ when $x<0$ and $\theta(x)=0$ when $x>0$). Here the first term describes the acoustic part of the spectrum, and the second one the optical part. Line 1 in fig. 2 was drawn with the parameters $\Lambda_0=2$ and $\Lambda_1=0$. It turned out that $T_c=\omega_0/5$. In other words, this is strong coupling with optical phonons without scattering: at $T\leq T_c$ phonons ω_0 are not excited practically and the scattering by them is in-essential. Accordingly, here a very low growth of λ is observed at

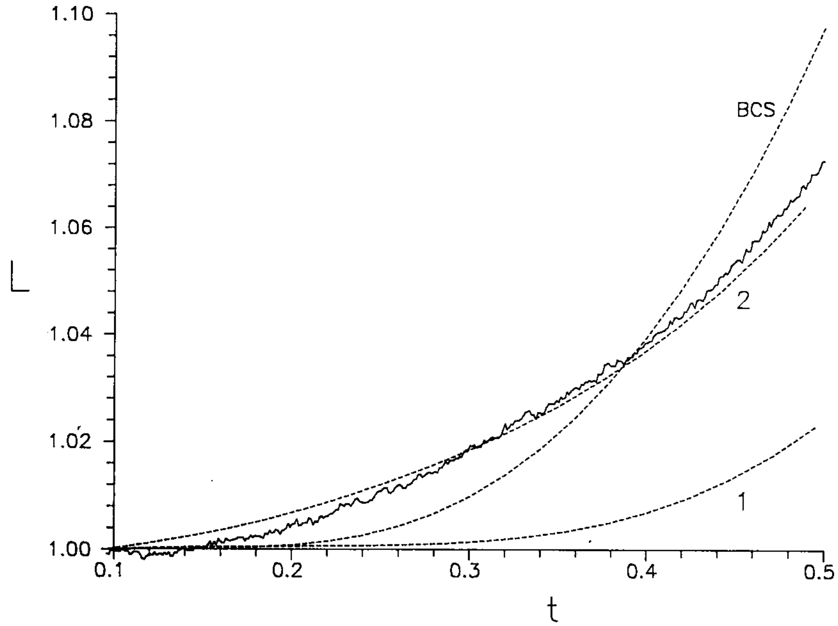

Fig. 3. Low-temperature part of lines in fig. 2 in (L, t)-coordinates.

low temperatures (see fig. 3), as well as in the BCS theory. Line 2 was drawn with $\Lambda_0=2$, $\Lambda_1=1$ and $\omega_D=T_c$. Temperature T_c itself did not undergo any alteration when the interaction with acoustic phonons was switched on and T_c remained equal to $\omega_0/5$ as before. However, the function $\lambda(T)$ altered. It became consistent with the experiment, evidently due to scattering on real phonons with frequencies $0\leq\omega\leq\omega_D\equiv T_c$ at small t.

It should be noted that line 2 does not practically differ from that drawn by Rammer for the case of strong coupling in the "pure" limit [17]. However, since in [17] the coupling constant was large ($\Lambda=6$), and the real phonon spectrum applied involved a low-frequency acoustic part, the phonon scattering evidently had been taken into account automatically.

In many papers ([2,4,5,9], see also [1]) the function $L(t)$ was described in the low-temperature region by a parabola. By comparison, the experimental data and the theoretical line 2 from fig. 3 can both be approximated to a sensible accuracy in the range $0\leq t\leqslant 0.4$ by the function

$$L(T)=1+\alpha t^2\,, \qquad \alpha=0.25\,. \tag{14}$$

A weaker reliable temperature dependence, than the above one, had apparently never been observed. A stronger one seems to be related to the low quality of the sample. For instance, for an annealed powder with other phases admixed we obtained the value of $\alpha=0.6$, as had been obtained in paper [9].

Region $(1-t)\ll 1$. All the theories, not taking account of fluctuations, yield the finite derivative for the function $L^{-2}(t)$ at the point $t=1$ (see inset in fig. 2). This derivative is a very convenient subject for comparison of the theory and the experiment.

To illustrate the possibilities of such comparison, fig. 4 depicts the results of calculating the derivative $D=d(L^{-2})/dt$ as a function of the coupling constant Λ_0 in the model (13) without acoustic scattering, that is, at $\Lambda_1=0$. The most important part of the line is $0\leq\Lambda_0\leq 3$. It demonstrates the growth of the absolute value of D with increasing the coupling constant at an invariable spectrum. However, D depends on the spectrum too. A separate point at $\Lambda_0=2$ demonstrates how D varies, when the low-frequency part of the spectrum (13) with constant $\Lambda_1=1$ is switched on. Therefore, the slope of the line $L^{-2}(t)$ at the point $t=1$ may only be a quantitative characteristic of the coupling constant. However, in the case of $YBa_2Cu_3O_{7-\delta}$ it shows definitely that the coupling cannot be regarded as a weak one.

A slight decrease of D at large Λ_0 in fig. 4. may again

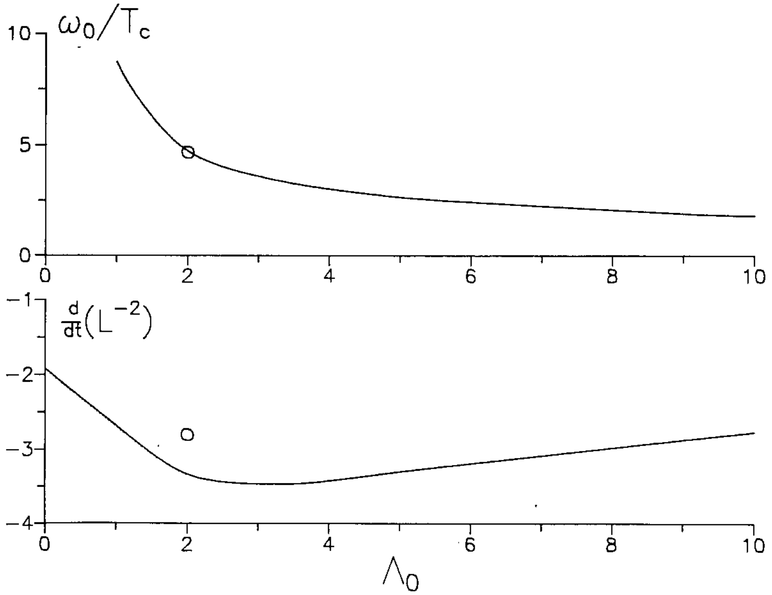

Fig. 4. Dependence of ω_0/T_c and derivative $d(L^{-2})/dt$ on the coupling constant Λ_0 in the model (13) without acoustic scattering, that is, at $\Lambda_1=0$. Point marks the values of ω_0/T_c and $d(L^{-2})/dt$ with acoustic part switched on ($\Lambda_0=2$, $\Lambda_1=1$, $\omega_D=\omega_0/5$).

be explained referring to the scattering, this time, on thermal optical phonons. These phonons exist below the transition temperature due to the decrease in ratio ω_0/T_c accompanying the growth of Λ_0 [18] (the upper curve in fig. 4).

5. Conclusion

A comparison of experimental and theoretical curves $\lambda(T)$ for the low-temperature region $t\leq 0.4$ shows that in $YBa_2Cu_3O_{7-\delta}$ there exists strong coupling with low-frequency excitations, in general, not necessarily with phonons. The scattering of carriers on these excitations is so strong that even at low temperatures $t=0.4$–0.5 it is the main factor limiting the mean free path ℓ. In its turn this results in the power law in the behaviour of the function $\lambda(T)$.

The magnitude of the coupling constant Λ can be obtained by measuring derivative $D=d(L^{-2})/dt$ in the vicinity of the temperature of the superconducting transition $t=1$. Though the calculated value of D depends on a particular spectrum, model (13) can apparently be used for estimates. In any case, $\lambda>1$ for sure.

Acknowledgements

The authors are grateful to A.A. Golubov and A.E. Koshelev for numerous fruitful discussions and for assistance in applying the algorithm of calculating the penetration depth, developed by them, and to G.M. Eliashberg and D.V. Shovkun for valuable comments.

References

[1] J.F. Annett, N.D. Goldenfeld and S.R. Renn, in: Physical Properties of High Temperature Superconductors II, ed. D.M. Ginsberg (World Scientific, New Jersey, 1990) p. 571.
[2] D.R. Harshmann, L.F. Schneemeyer, J.V. Waszczak, et al., Phys. Rev. B39 (1989) 851.
[3] B. Pumpin, H. Keller, W. Kunding, et al., Physica C 162–164 (1989) 151.
[4] L. Krusin-Elbaum, R.L. Greene, F. Holtzberg, et al., Phys. Rev. Lett. 62 (1989) 217.
[5] S. Mitra, J.H. Cho, W.C. Lee, et al., Phys. Rev. B40 (1974) 2674.

[6] J.P. Carini, A.M. Awasthi, W. Beyermann, et al., Phys. Rev. B37 (1988) 9726.
[7] L. Drabeck, J.P. Carini, G. Gruner, et al., Phys. Rev. B39 (1989) 789.
[8] V.F. Gantmakher, V.I. Kulakov, G.I. Leviev, et al., JETF 95 (1989) 1444 (Sov. Phys. -JETP 68 (1989) 833).
[9] J.F. Cooper, C.T. Chu, L.W. Zhou, et al., Phys. Rev. B37 (1988) 638.
[10] G.M. Gryaznov, V.N. Lapovok, I.G. Naumenko, et al., DAN USSR 267 (1982) 619 (Sov. Phys. -Doklady 267 (1982)).
[11] S.G. Gevorgyan, Ph.D. Thesis (Inst. of Research of Armenian Acad. of Sciences, Ashtarak) 1989.
[12] R.A. Buhrman and W.P. Halperin, Phys. Rev. Lett. 30 (1973) 692.
[13] H. Ahoh and A. Tasaki, J. Appl. Phys. 49 (1978) 1410.
[14] D. Shoenberg, Superconductivity, Cambridge, 1952.
[15] O.V. Dolgov, A.A. Golubov and A.E. Koshelev, Solid State Commun. 72 (1989) 81.
[16] M. Tinkham, Introduction to Superconductivity (McGraw-Hill, New York, 1975).
[17] Y. Rammer, Europhys. Lett. 5 (1988) 77.
[18] P.B. Allen and R.C. Dynes, J. Phys. C8 (1975) L158.