Temperature Dependence of the Magnetic Field Penetration Depth in Rb_3C_{60} Measured by ac Susceptibility

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The temperature dependence of the magnetic field penetration depth $\lambda(T)$ is determined down to $0.2T_c$ by the ac-susceptibility measurements on the Rb_3C_{60} fine powder with small, about 1 K, variation in the grains' superconducting transition temperatures. The $\lambda(T)$ temperature dependence is exponential at low temperatures, which allows us to determine the energy gap $\Delta_0 = 1.7k_BT_c$. Deviations of the experimental dependence $\lambda(T)$ from BCS are small and can be explained by strong-coupling theory.

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The discovery of superconductivity with moderately high T_c in the alkali-doped fullerenes, A_3C_{60} (A =K,Rb,Cs), has created considerable interest in the mechanism of supercoductivity in these compounds. It is still unclear whether the BCS theory can explain superconductivity in the A_3C_{60} or if another approach is necessary. The energy gap Δ_0 close to the BCS value $\Delta_0/k_BT_c = 1.76$ obtained from optic [1], NMR [2], and muon spin relaxation (μSR) [3] experiments and the existence of the Hebel-Slichter peak in μSR [3] are in favor of BCS. On the other hand, the large value of $\Delta_0/k_BT_c = 2.6$, measured in the tunnel experiment [4], and the absence of the Hebel-Slichter peak in NMR [2] contradict the BCS. The authors of Ref. [5] have concluded on the basis of these divergences from BCS that the strong-coupling theory is necessary for the explanation of the superconductivity in A_3C_{60}.

It is well known that strong-coupling effects substantially alter the temperature dependence of the magnetic field penetration depth expected from BCS. In the present Letter we report $\lambda(T)$ dependence determined by ac-susceptibility measurements in fine Rb_3C_{60} powders with mean grain size less than the penetration depth. We find that this dependence is close to the BCS one in local dirty limit. The gap Δ_0 obtained from this dependence is also close to the BCS one. The small deviation of $\lambda(T)$ dependence from BCS can be explained by the strong-coupling theory with coupling strength $\Lambda \approx 0.8$. We know only one other work [6] in which the dependence of $\lambda(T)$ for K_3C_{60} was measured by μSR.

The Rb_3C_{60} powder was produced by solid phase reaction of the C_{60} powder with the pure Rb. A stoichiometric quantity of Rb was added, in vacuum, to the C_{60} powder in the apparatus made of Pyrex glass. Then the ampoule with the mixture of C_{60} and Rb was sealed and annealed for 2 d at 200 °C and for 6 h at 250 °C in accordance with the procedure described in Ref. [7]. Further annealings did not change the sample properties.

Thus three samples were produced with the coefficient of filling of the powder of about 25%. The results obtained for all the samples were the same. There was also exact reproducibility from one measurement to another. It was possible to move the powder from one end of the ampoule to another. We also tried to shake the ampoules in order to change the properties of the contacts between grains. However, such a procedure did not influence the experimental results at all.

The sample was placed inside one of two identical induction coils connected with each other and the disbalance signal, which is proportional to the magnetic moment M of the sample, was measured in an alternating magnetic field of 10^5 Hz frequency: $M = H\chi V$ (see, for instance, Ref. [8]). Here H is the amplitude of the alternating magnetic field, χ is the ac susceptibility, $V = m/\rho$ is the sample volume, $\rho = 2.6$ g/cm^3—density of the Rb_3C_{60} powder from the x-ray measurements. To determine the Rb_3C_{60} mass we supposed that C_{60} with known mass completely reacted with Rb. The mass of each sample was about 5 mg.

The ac susceptibility χ is generally complex. In our experiments the imaginary part of χ, proportional to the losses, was negligible, compared with the real part of χ, proportional to the shielding. We have normalized the measured ac susceptibility by the susceptibility of an ideally diamagnetic ($\lambda = 0$) sphere $\chi_{\max} = -3/8\pi$. For this purpose the setup was specially calibrated [8]. The temperature dependence of the normalized ac susceptibility is shown in Fig. 1. The shielding did not exceed 4%. Such a small value is caused by the small size a of powder grains in comparison with the penetration depth λ. In the case of small shielding, a field applied to every grain coincides with the external field, hence the total magnetic moment is determined by summing over all grains [9]:

$$M = H \sum_n \chi_{\max,n} v_n f(a_n/\lambda) \ . \quad (1)$$

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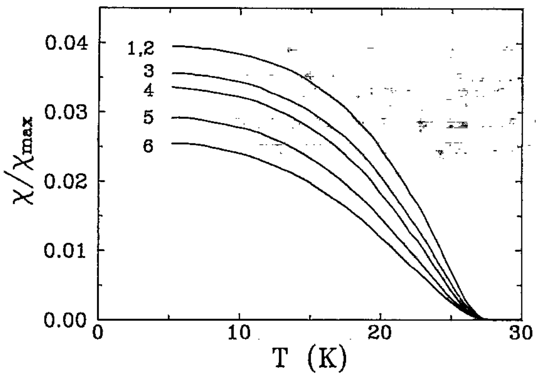

FIG. 1. Set of temperature dependences of ac susceptibility at the different amplitudes of alternating magnetic field curves. 1: 0.016 Oe, 2: 0.14 Oe, 3: 2.4 Oe, 4: 4.9 Oe, 5: 12 Oe, 6: 24 Oe.

Here $\chi_{\max}$ and the function $f(a/\lambda)$ generally depend on the size and shape of a grain. For example, for spherical grains with diameter a [9],

$$f(a/\lambda) = 1 - 6(\lambda/a)\coth(a/2\lambda) + 12(\lambda/a)^2 \ . \qquad (2)$$

However, for small grains, compared with the penetration depth $a < 2\lambda$, the function $f(a/\lambda) = k(a/\lambda)^2$ (for a sphere $k = 1/60$) and

$$M = \chi H \sum_n v_n = \lambda^{-2} \sum_n \chi_{\max,n} v_n k_n a_n^2 \ . \qquad (3)$$

Consequently, the experimentally measured ratio

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

defines the temperature dependence $\lambda(T)$.

The conclusion that the size of the particles is less than λ for our powders follows directly from the low value of $\chi(0)/\chi_{\max} = 0.04$. If one independently measures the size of the grains it is possible to determine the value of $\lambda(0)$.

We failed to determine the grain size by electron microphotography due to the sticking of grains. Therefore we estimate the size of the grains using the known value of $\lambda(0)$. If $\lambda(0) = 4600$ Å Ref. [2] for spherical grains we estimate the diameter from Eq. (2) to be $a \approx 7400$ Å. Unfortunately there is a significant dispersion of $\lambda(0)$ obtained by different groups. Thus Politis *et al.* [10] investigated the magnetic moment of Rb_3C_{60} powder in a strong magnetic field and obtained the estimation $\lambda(0) = 2500$ Å. However, it was shown later in Ref. [11] that it is necessary to reanalyze the data of Ref. [10] taking into account the small size of the grains, which leads to the estimation $\lambda(0) = 3500$ Å, which is not very different from the value of Ref. [2]. [In Ref. [11] one can find the value of $\lambda_{\rm GL} = \lambda(0)/\beta^{1/2}$, β is the coefficient in Eq. (5), according to our measurements $\beta = 2.85$.]

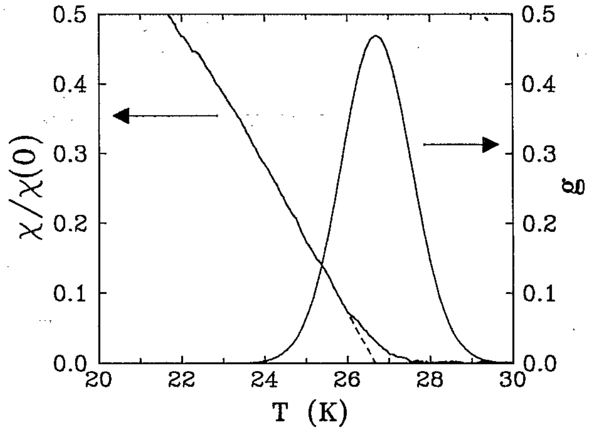

FIG. 2. $\chi(T)$ (solid line), $L^{-2}(T)$ (dashed line), and T_c distribution function $g(T)$ near superconducting transition. $\chi(T)$ and $L^{-2}(T)$ are coincident below 26 K.

Without taking into account fluctuations [12], the $L^{-2}(T)$ function has a finite derivative at $T = T_c$. As temperature decreases this derivative is constant in the temperature interval $\approx 0.15T_c$ near T_c, i.e.,

$$\chi(T) \propto L^{-2}(T) = \beta(1 - T/T_c) \ . \qquad (5)$$

At lower temperatures its absolute value decreases. However, the experimental curve deviates from the linear dependence near T_c (Fig. 2). This deviation is substantially greater than that caused by fluctuations and should be associated with the grains' T_c variation. Introducing the transition temperature distribution function $g(T)$ we get instead of Eq. (4):

$$\frac{\chi(T)}{\chi(0)} = \int_T^\infty g(t) L^{-2}(T/t)dt \ . \qquad (6)$$

If the distribution function width is less than 0.15 T_c, we can substitute $\beta(1 - T/t)$ for L^{-2} in Eq. (6), then

$$g(T) = \frac{T}{\beta}\frac{d^2}{dT^2}\left[\frac{\chi(T)}{\chi(0)}\right] \ . \qquad (7)$$

To estimate the width of the distribution function we fit the $g(T)$ derived from Eq. (7) by the Gaussian distribution

$$g(T) = \frac{1}{\sqrt{2\pi}\,\sigma}\exp\left(-\frac{(T-T_0)^2}{2\sigma^2}\right) \ , \qquad (8)$$

which leads to the values of $T_0 = 26.7$ K, $\sigma = 0.85$ K, and $\beta = 2.85$ (Fig. 2). We should note that the real distribution over T_c may differ significantly from the Gaussian one. For example, if the $g(T)$ is due to the local fluctuations in the Rb stoichiometry, the distribution function is asymmetric and is skewed to lower T_c's since the dependence of T_c on the Rb concentration has a maximum. However, it does not change the slope β because of the small width of the distribution function, so β coincides with the slope of the straight line approximating the experimental $\chi(T)/\chi(0)$ dependence close to T_c.

Solving Eq. (6) with $g(T)$ given above we obtain the dependence of $L^{-2}(T)$.

The serious problem in evaluating the $\lambda(T)$ dependence from our data is the dependence of susceptibility on the amplitude of the alternating magnetic field H starting from an amplitude of 1 Oe which is less than $H_{c1}(0) = 120$ Oe in Rb_3C_{60} [13]. This shows the presence of weak links in the grains of powder. More exactly several grains are supposed to form a cluster with Josephson junctions. In this case it is possible to show that Eq. (3) is still valid in the linear regime (in the case of low amplitudes) if one uses, instead of grain size a, the effective size of the cluster $a_{\rm eff}$. With the field amplitude increasing the screening current in the cluster exceeds the critical current of the junctions. In this case the current flows mainly in the separate grains, so the effective size of the cluster decreases. Herewith, the susceptibility decreases, being proportional to $(a_{\rm eff}/\lambda)^2$. We intend to discuss in detail the field dependence in another paper. Here we just note that $a_{\rm eff}$ does not greatly exceed the size of grains, because at sufficiently large amplitude $H = 24$ Oe when most of the contacts are already destroyed, the susceptibility decreases to not more than half of its initial value.

Thus the $\chi(T)$ temperature dependence in the linear case (upper line in Fig. 1) gives the $\lambda(T)$ dependence in accordance with Eq. (4) and Eq. (6). Now we discuss this dependence.

The BCS temperature dependence of λ substantially depends on the relation between London penetration depth $\lambda_L = (m^*c/4\pi e^2)^{1/2}$ (here m^* is the carrier effective mass, e is the electron charge), coherence length $\xi_0 = \hbar v_F/\pi\Delta_0$ (v_F is the Fermi velocity), and mean free path l. In particular, the local (London) limit is valid when $\lambda_L \geq \xi$, here ξ is the Pippard coherence length ($1/\xi = 1/\xi_0 + 1/l$). The Ginzburg-Landau coherence length is known from the measurements of H_{c2}: $\xi_{\rm GL}(0) \simeq 30$ Å [7,13–15]. In the clean ($\xi_0 \ll l$) limit ξ_0 is approximately equal to $\xi_{\rm GL}(0)$, and $\xi_{\rm GL}(0) \approx (\xi_0 l)^{1/2}$ in the dirty limit ($\xi_0 \gg l$). Hence, the upper estimation of ξ is ≈ 30 Å. Comparison with the value of $\lambda(0) \approx 4600$ Å shows that in Rb_3C_{60} the local limit is realized at any relationship between ξ_0 and l.

The relation between ξ_0 and l for K_3C_{60} was discussed in [14,16]. The value of l is determined by v_F which is not known exactly. In Ref. [16] $v_F = 5 \times 10^6$ cm/s was supposed and the values of $l = 20$ Å and $\xi_0 = 26$ Å were obtained, which corresponds to the interface of clean and dirty limits. If $v_F = 1.8 \times 10^7$ cm/s is assumed then $l = 11$ Å and $\xi_0 = 140$ Å [14], which corresponds to the dirty limit.

The comparison of the measured $L^{-2}(T/T_c)$ with the BCS dependences [calculated using Eq. (5.33) from Ref. [17]] in dirty and clean limits is shown in Fig. 3. We note that in the dirty limit

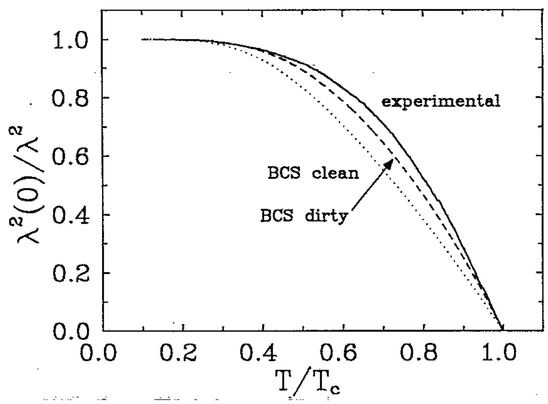

FIG. 3. Comparison of the experimental dependence $L^{-2}(T) = \lambda^2(0)/\lambda^2(T)$ with the BCS dependences in clean and dirty limits.

$$\frac{1}{\lambda^2(T)} = \frac{1}{\lambda_L^2}\left(\frac{l}{\xi_0}\right)\left(\frac{\Delta(T)}{\Delta_0}\right)\tanh\left(\frac{\Delta(T)}{2k_BT}\right). \quad (9)$$

One can see that BCS dependence in the dirty limit coincides with the experiment better.

The dependence $L^{-2}(T/T_c)$ at $T/T_c < 0.4$ is described in BCS by the expression $L^{-2}(T/T_c) = 1 - \gamma\exp(-\Delta_0/k_BT)$. In the clean limit $\gamma = (2\pi\Delta_0/k_BT)^{1/2}$, in the dirty limit $\gamma = 2$. The fitting of the experimental curve by this expression gives $\Delta_0/k_BT_c = 1.7 \pm 0.2$ and $\gamma = 2 \pm 0.5$, which coincides well with the BCS dirty limit ($\Delta_0/k_BT_c = 1.76$, $\gamma = 2$). The exponential behavior of $\lambda(T)$ at the low temperatures proves s-wave pairing in Rb_3C_{60}.

We would like to note that in oxide superconductors the experimental $\lambda(T)$ dependences are still controversial. Thus even the latest $\lambda(T)$ measurements in $YBa_2Cu_3O_{7-x}$ show at $T < 0.4T_c$ the significantly different power law temperature variations or a weak exponential dependence [18].

The small discrepancy between our experimental results and BCS dependence $L^{-2}(T/T_c)$ can be caused by the effects of strong coupling. It is convenient to use the value of the slope $\beta = -dL^{-2}(t)/dt$ near T_c [see Eq. (5)], ($t = T/T_c$) as the parameter of the coupling strength. The experimental value $\beta = 2.85$ is greater than the dirty limit value $\beta_d = 2.62$ (the BCS clean limit value $\beta_c = 2$). The increase of the coupling strength $\Lambda = 2\int \frac{\alpha^2F(\omega)}{\omega}d\omega$, [$\alpha^2F(\omega)$ is the Eliashberg function] leads to an increase of the slope β as compared with the BCS value. We note that $\beta = 4$ in the two-fluid Gorter-Casimir model. The β values in the strong coupling theory are situated between the BCS and two-fluid values. Generally the dependence $L^{-2}(T/T_c)$ in the strong coupling theory is determined not only by the coupling strength constant Λ, but also by the overall $\alpha^2F(\omega)$ function. For the estimation of Λ we have used the simplest Eliashberg function

$$\alpha^2F(\omega) = \tfrac{1}{2}\Lambda\omega_0\delta(\omega - \omega_0), \quad (10)$$

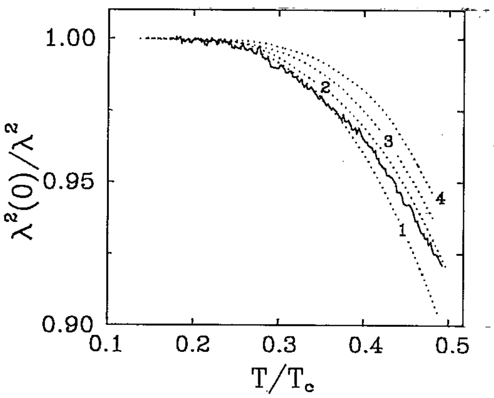

FIG. 4. Comparison at low temperatures $L^{-2}(T) = \lambda^2(0)/\lambda^2(T)$ with theory. Solid line: experiment; dotted line 1: BCS, dirty limit; 2: one-peak model, $\Lambda = 0.8$, dirty limit; 3: one-peak model, $\Lambda = 1.3$, clean limit; 4: two-peak model, dirty limit.

which describes the coupling with only one phonon mode (one-peak model). In this model in the dirty limit we have the best agreement with the experimental value of $\beta = 2.85$ at $\Lambda = 0.8$. (The penetration depth was calculated by the numerical solution of the Eliashberg equations [19] by using expressions from Ref. [17], the pseudopotential μ^* was assumed to be zero.) In this model the value of β coincides with the BCS value at $\Lambda \leq 0.5$. In the one-peak model we can obtain the same value of $\beta = 2.85$ in the clean limit. In this case $\Lambda = 1.3$, which is substantially greater than Λ in the dirty limit.

However, the value $\Lambda = 0.8$ fits the low temperature region, where $L^{-2}(T/T_c)$ is proportional to $\exp(-\Delta_0/k_BT)$, much better (see Fig. 4). This is due to the increase of the energy gap Δ_0/k_BT_c with increasing Λ. The value of Δ_0/k_BT_c in the one-peak model with $\Lambda = 0.8$ still does not differ significantly from the BCS one.

Of course, the very simple one-peak model can serve only as an estimate. In Ref. [5] the two-peak-model Eliashberg function was proposed in which, besides the high-frequency mode ($\omega_2 = 1000$ cm^{-1}) with the coupling strength $\Lambda_2 = 0.5$, there is the low-frequency mode ($\omega_1 = 40$ cm^{-1}) with coupling strength $\Lambda_1 = 2.7$ (the summary coupling strength is 3.2). This model in the dirty limit gives our experimental value of the slope $\beta = 2.85$, however, the obtained value of $\Delta_0/k_BT_c = 2.6$ is greater (as in the tunneling experiment [4]). Hence, in this model the low temperature part of the dependence $L^{-2}(T/T_c)$ strongly differs from the measured one (Fig. 4).

In conclusion, we have measured the temperature dependence of the penetration depth and determined the gap Δ_0 for Rb_3C_{60}. This dependence and the value of the gap are close to those known from the BCS theory. We have also estimated the electron-phonon coupling strength.

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