ac susceptibility of Rb_3C_{60} fine powder

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The temperature dependence of ac susceptibility $\chi(T)$ has been measured at different values of dc magnetic field for Rb_3C_{60} fine powder. The diamagnetic moment of the superconducting powder arises due to both screening currents in separate grains (intragrain contribution) and screening currents in clusters formed by several grains with Josephson junctions between them (intergrain contribution). The intergrain contribution of clusters is proportional to the critical current of the Josephson junctions, j_c, and decreases rapidly in a dc magnetic field H, while the intragrain contribution remains unchanged up to much higher values of H. The $\chi(T)$ measurements at different dc magnetic fields allow us to distinguish between these two contributions and to obtain precisely the temperature dependence of the London penetration depth $\lambda(T)$. We have found that the experimentally obtained $\lambda(T)$ is perfectly described by the weak coupling BCS dependence in the dirty limit.

I. INTRODUCTION

Despite the very recent discovery of superconductivity in the alkali-metal-doped fullerenes there have already been published quite a lot of experimental investigations of their superconducting properties. Recent experimental results obtained with NMR,[1,2] photoemission spectroscopy,[3] muon spin relaxation,[4] and optical measurements[5,6] agree with the expectations of weak coupling Bardeen-Cooper-Schrieffer (BCS) superconductivity. The energy gap Δ_0 obtained in these experiments is close to the weak coupling BCS value $\Delta_0/k_BT_c=1.76$. On the other hand, much larger ratios Δ_0/k_BT_c were obtained by tunneling experiments [2.6 (Ref. 7) and 2.7 (Ref. 8)] by Raman spectroscopy [3.8 (Ref. 9)], and by NMR [2.2 (Ref. 10)]. It is also difficult to explain the high value of T_c by weak coupling BCS theory. There are many theoretical calculations, which invoke strong coupling to attain $T_c\approx30$ K (see Ref. 11 and references therein). In order to understand whether the BCS theory is sufficient to describe the superconductivity in these compounds we need more precise measurements of basic superconducting parameters.

One of the most fundamental characteristics is the London penetration depth λ. The temperature dependence of this value noticeably differs for different theories and is quite sensitive to various parameters. From the experimentally measured $\lambda(T)$ dependence one may check the applicability of BCS theory, understand whether a superconductor is in the clean or dirty limit, and estimate the coupling constant and $\Delta(0)/k_BT_c$ value.

We have already briefly reported on $\lambda(T)$ measurements in Rb_3C_{60} ultrafine powder by ac susceptibility.[12] The ac susceptibility measurements of small [in comparison with $\lambda(0)$] grains is one of the most precise methods to obtain $\lambda(T)$.[13] However, as we have already mentioned in Ref. 12, there is a quite essential difficulty in obtaining λ by such a method. The diamagnetic signal of a powder may arise not only due to screening currents in individual grains, but also due to currents in clusters formed by several grains with Josephson junctions between them. The contribution of individual grains is given only by the value and temperature dependence of λ, while the contribution of clusters depends on the value and temperature dependence of the critical current j_c of the contacts formed by neighboring grains. This cluster contribution may lead to a noticeable error in the extracted $\lambda(T)$ dependence. In Ref. 12 we supposed the contribution of clusters to be small in comparison with the contribution of individual grains. However, for precise determination of $\lambda(T)$ it is nessasary to distinguish experimentally between these two contributions.

In this paper we present the detailed analysis of ac susceptibility measurements of ultrafine Rb_3C_{60} powder at different ac and dc magnetic fields. The value of j_c and hence the intergrain contribution of clusters to susceptibility decrease rapidly with applied magnetic field strength, while the intragrain susceptibility of individual grains remains unchanged up to much higher values of H. This fact allowed us to distinguish between the contribution of individual grains and clusters and to obtain $\lambda(T)$ with a very high precision.

II. SAMPLE PREPARATION

The Rb_3C_{60} powder was produced by solid phase reaction of C_{60} powder with pure (99.99%) Rb. A stoichiometric quantity of Rb was added, in vacuum, to the C_{60} powder in an apparatus made of Pyrex glass. A hermetically sealed ampoule with pure Rb and an ampoule with a known amount of C_{60} powder were connected by a glass capillary with known diameter. The construction was heated to 100 °C and was evacuated for several hours. Then it was sealed up. With the help of a metal cylinder we broke the ampoule with Rb. By gradual heating we filled the capillary with Rb. Then the capillary was sealed at such a point that the Rb remaining below corresponded to the necessary amount. The ampoule with the stoichiometric mixture of C_{60} and Rb was annealed for two days at 200 °C and for 6 h at 250 °C in accordance with the procedure described in Ref. 14. Further annealings did not change the sample's properties.

In order to have good thermal contact we kept the ampoule in an atmosphere of He for several weeks at room

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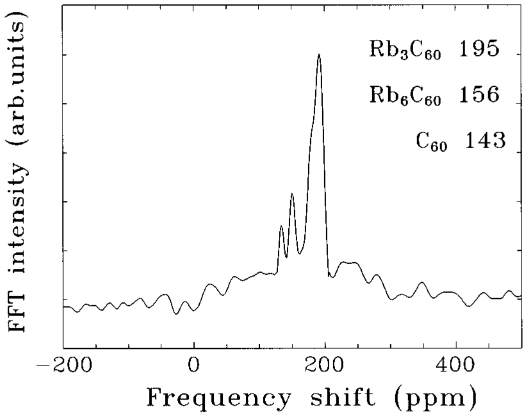

FIG. 1. The static room temperature NMR spectrum of ^{13}C (x axis: frequency shift, ppm; y axis: signal intensity, arb. units).

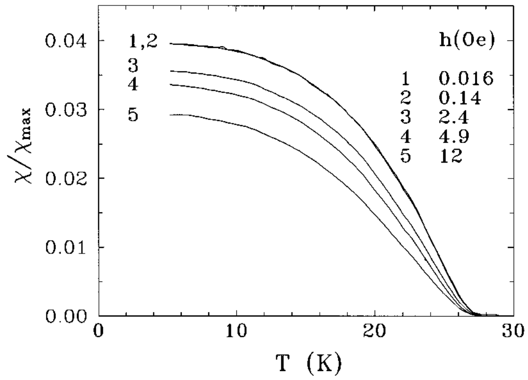

FIG. 2. Set of temperature dependences of ac susceptibility at different amplitudes of alternating magnetic field. 1, 0.016 Oe; 2, 0.14 Oe; 3, 2.4 Oe; 4, 4.9 Oe; 5, 12 Oe.

temperature. This was sufficient for the helium to penetrate inside the ampoule.

The coefficient of filling of the powder was about 25%. The mass of the sample was about 5 mg. The exact volume of Rb_3C_{60} was determined from NMR measurements.

III. SAMPLE CHARACTERIZATION

The phase composition of our sample was determined from the static NMR spectrum of natural-abundant ^{13}C. The fast Fourier transform (FFT) product of the spin-echo signal was obtained with a Bruker MSL-300 pulse spectrometer by averaging over about 10^4 scans. The measurements were carried out at room temperature in a magnetic field of 70.5 kOe. The results are presented in Fig. 1. According to Ref. 15, the resonance peaks at 135, 150, and 192 ppm are attributed to C_{60}, Rb_6C_{60}, and Rb_3C_{60}, respectively. Assuming Gaussian line shapes we have fitted the C_{60} and Rb_6C_{60} peaks and found the ratio of the C_{60}, Rb_6C_{60}, and Rb_3C_{60} peak integrals to be 4:6:90. Since the area of the resonance peak is proportional to the number of nuclei, this means that 90% of the C_{60} powder formed the Rb_3C_{60} phase. Thus from the known mass of C_{60} powder we obtained the Rb_3C_{60} mass m and volume

$$V=\frac{m}{\rho}, \tag{1}$$

where $\rho=2.6$ g/cm^3 is the density of Rb_3C_{60} from x-ray measurements.

IV. EXPERIMENTAL RESULTS

The sample was placed inside one of two identical induction coils connected in opposition to each other. The disbalance signal arising in an alternating magnetic field of amplitude h_z and frequency $\omega=10^5$ Hz was measured. The disbalance signal is proportional to the z component of the magnetic moment of the sample, M_z, varying with the frequency ω of the alternating field. By the z component we mean the projection on the coil axis:

$$M_z=\chi V h_z. \tag{2}$$

The temperature dependence of the magnetic moment $M_z(T)$ was measured with decreasing temperature. We would like to note that when the powder was in vacuum there was a small difference between the curves measured with temperature decreasing and temperature increasing. After filling the ampoule with He, as described above, the results of the measurements with increasing and decreasing temperature became identical.

The ac susceptibility χ is, in general, complex. In our experiments the imaginary part of χ, proportional to the losses, was negligible compared to 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.[16]

The temperature dependence of the normalized ac susceptibility at $H=0$ is shown in Fig. 2. One may see that, at low h_z, there is a linear regime, where χ is independent of h_z. Susceptibility starts to depend on h_z at $h_z>1$ Oe, which is much less than the first critical field of the grains, $H_{c1}=120$ Oe. This shows the presence of weak links.

The dc magnetic field H perpendicular to the ac field h_z was applied at $T>T_c$. The ac susceptibility $\chi(T)$ was measured with temperature decreasing at $h_z<0.1$ Oe, where χ did not depend on h_z. Thus we have always measured $\chi(T)$ with field cooling and in a regime linear with respect to h_z.

Figure 3 shows the experimental $\chi(T)$ dependences at different dc magnetic fields H. From these data we have obtained the $\chi(H)$ dependences at T = const, which are shown in Fig. 4. One may see that χ decreases rapidly at low H, remains nearly constant for 0.5 kOe $<H<$2.5 kOe, and then again decreases noticeably with increasing H.

V. DISCUSSION

The diamagnetic moment of the superconducting powder arises due to screening currents, which may flow inside the grains or in clusters. The intragrain screening may be divided into two parts with respect to the value of the dc magnetic field H. At low field, i.e., $H<H_{c1}$, the grains are in the Meissner state and obviously the dc magnetic field does not change the ac susceptibility of separate grains. Magnetic

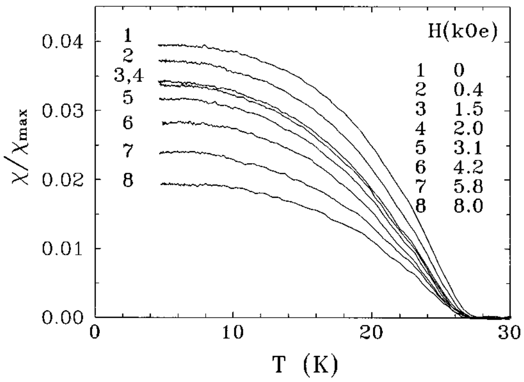

FIG. 3. Set of temperature dependences of ac susceptibility in the linear regime at different dc magnetic fields. 1, 0 kOe; 2, 0.4 kOe; 3, 1.5 kOe; 4, 2 kOe; 5, 3.1 kOe; 6, 4.2 kOe; 7, 5.8 kOe; 8, 8 kOe.

fields higher than H_{c1} create vortices, the vortex density inside the grains being proportional to H. The vortices may move with the frequency of the ac field, so χ may depend on H. On the other hand, there may be Josephson junctions between neighboring grains. Hence the superconducting screening currents may also circulate in clusters formed by several grains.

The total shielding of the powder does not exceed 4%. Such a small value is caused by the small size of the grains in comparison with the penetration depth. In the case of small shielding, the field applied to every grain coincides with the external field. Thus, the magnetic moments of every grain and every cluster may be considered independently. The total magnetic moment is the sum of the magnetic moments of all grains and all clusters:

$$M=\sum_n M_n+\sum_c M_c. \tag{3}$$

Here M_c, the magnetic moment of a cluster, is the magnetic moment due to currents flowing between the grains. We should note that this value does not contain the magnetic moments of the individual grains which form the cluster.

In order to understand the experimental results, let us consider in detail the different possible contributions to the diamagnetic moment of the powder.

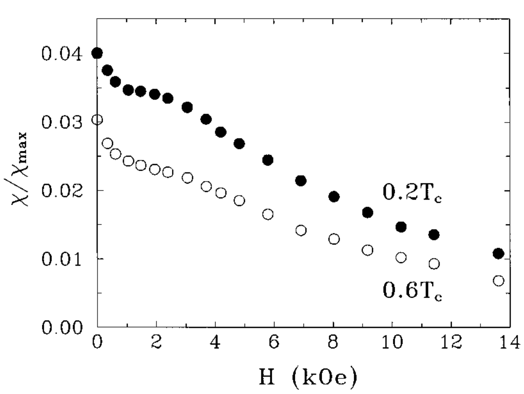

FIG. 4. Field dependence of ac susceptibility in the linear regime at $T=0.2T_c$ and $T=0.6T_c$.

A. Contribution of individual grains in the Meissner state

The magnetic moment M_n of a grain with volume v_n may be expressed through the formula

$$M_n=h_z\chi_{\max,n}v_n f(r_n/\lambda). \tag{4}$$

The function $f(r/\lambda)$ and $\chi_{\max}$ generally depend on the size and shape of a grain. For example, for spherical grain with radius r,[13]

$$f(r/\lambda)=1-3(\lambda/r)\coth(r/\lambda)+3(\lambda/r)^2. \tag{5}$$

However, for grains which are small compared to the penetration depth ($r<\lambda$), the function $f(r/\lambda)$ is described with good accuracy by the quadratic function

$$f(r/\lambda)=k(r/\lambda)^2, \tag{6}$$

where only the coefficient k depends on the grain's shape (e.g., for a sphere $k=1/15$). Thus we may write for the magnetic moment of a small grain

$$M_n=\lambda^{-2}h_z\chi_{\max,n}v_n k_n r_n^2. \tag{7}$$

The total intragrain magnetic moment is determined by summing over all grains:

$$M_z=\sum_n M_n=\lambda^{-2}h_z\sum_n \chi_{\max,n}v_n k_n r_n^2. \tag{8}$$

Consequently, in the case of the separate grains contribution only, the experimentally measured ratio

$$\frac{M_z(T)}{M_z(0)}\equiv\frac{\chi(T)}{\chi(0)}=\left[\frac{\lambda(T)}{\lambda(0)}\right]^{-2}\equiv L^{-2}(T) \tag{9}$$

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

The conclusion that the size of the particles is less than λ for our powders follows directly from the low value of $\chi(0)/\chi_{\max}=0.04$. Taking into account the cubic symmetry of Rb_3C_{60}, we may suppose that in our case all grains have a spherical shape, so $\chi_{\max}=-3/8\pi$ and $k=1/15$. If one independently measures the size of the grains it is possible to determine the value of $\lambda(0)$. However, we failed to determine the grain size by electron microscopy due to the sticking of grains. Therefore we estimated the size of our grains from the known value of $\lambda(0)=4600$ Å.[1] We found the radius of our grains to be $r\approx 4000$ Å.

Equation (9) describes the susceptibility of small grains in the Meissner state. Hence χ should not depend on h_z, for h_z less than the first critical field $H_{c1}\approx 120$ Oe. However, experimentally we found that in a zero dc magnetic field χ starts to depend on h_z at much lower values of alternating field $h_z>1$ Oe (see Fig. 2). Therefore at $H=0$ the diamagnetic signal of the powder is not only due to the intragrain contribution. Usually, the nonlinear dependence of χ at such low values of h_z is characteristic for the screening currents circulating through Josephson junctions.

B. Contribution of clusters

Usually the critical state model for a granular superconductor is employed to calculate the temperature as well as ac and dc magnetic field dependence of the complex ac susceptibility (see, for instance, Refs. 17 and 18). One of the main results of this model is the explanation of the two-stage superconducting transition in $\chi(T)$ which is often observed. Note that such a two-stage transition was observed in the ac susceptibility of Rb_3C_{60} powder.[19] The first stage of the transition (at higher temperature) is due to the grain contribution, while the second one is connected with the intergranular Josephson matrix (cluster of grains) and can be explained in the framework of the critical-state model. According to this model, the second transition occurs when the magnetic field just reaches the center of the sample (see Refs. 17 and 18). Thus, the second transition can be observed only if the cluster extends through the whole sample. In our samples this second transition was not observed (see Fig. 1). Taking into account that the shielding in our sample does not exceed 4%, the critical state does not exist in an intergranular matrix.

We propose another approach which is based on the analysis of the contribution to the ac susceptibility of small clusters, which are practically transparent for the magnetic field.

Let us suppose that N grains form a ring with radius $R=Nr/\pi$. We also suppose that there are Josephson junctions between neighboring grains. Therefore in an ac field h_z there arise not only the screening currents in separate grains but also the screening current j circulating in a ring. In our case the screening is small (see Fig. 2); hence the field everywhere is close to h_z and the total magnetic moment is the sum of the magnetic moments of separate grains and the magnetic moment $\mathbf{M}_c$ of the ring:

$$\mathbf{M}_c=\frac{1}{2c}\int \mathbf{j}\times\mathbf{R}dV. \tag{10}$$

Small shielding also means that in order to calculate the current j, arising in a field h_z, one may neglect the screening of the grains.

The problem of calculating the current j in a ring with many Josephson junctions is very similar to the well-known problem of a dc superconducting quantum interference device (SQUID).[20] The superconducting current density j arising in a magnetic field may be expressed through the formula

$$\mathbf{j}=\frac{c}{4\pi\lambda^2}\left(\frac{\Phi_0}{2\pi}\nabla\Theta-\mathbf{A}\right). \tag{11}$$

Here $\mathbf{A}$ is the vector potential, Θ is the phase of the wave function of the superconducting electrons, and $\Phi_0=\pi\hbar c/e$ is the flux quantum. Let us integrate this expression along the ring, omitting the direct region of contacts. Taking into account that the width of the contacts, d, is negligibly small in comparison with the grain size, $d\ll r$, we obtain the absolute value of j:

$$2\pi Rj=\frac{c}{4\pi\lambda^2}\left(\frac{-\Phi_0}{2\pi}\sum_n \varphi_n+\pi R^2 h_z\right). \tag{12}$$

The phase difference on the nth contact φ_n may be found from the Josephson relation $j=j_{c,n}\sin\varphi_n$, where $j_{c,n}$ is the critical current of the nth contact. If the field h_z is quite small, then $j\ll j_c$ and

$$\varphi_n\approx\frac{j}{j_{c,n}}. \tag{13}$$

Finally we have

$$j\left(1+\frac{1}{Nr\lambda}\sum_n \lambda_{J,n}^2\right)=\frac{c}{8\pi^2\lambda^2}Nrh_z, \tag{14}$$

where $\lambda_{J,n}^2=\Phi_0 c/(16\pi^2\lambda j_{c,n})$ is the Josephson penetration depth.

Certainly the real contacts have different values of j_c; however, we may limit our consideration to two simple cases.

If we suppose that all contacts have the same critical current $j_{c,n}=j_c$ then

$$\frac{\Sigma_n\lambda_{J,n}^2}{Nr\lambda}>\frac{\lambda_J^2}{\lambda^2}\gg 1. \tag{15}$$

Here we took into account that $r<\lambda$ and that for weak contacts $\lambda_J\gg\lambda$. Finally, from Eq. (14) we obtain the current j:

$$j=j_c\frac{2Nr^2}{\Phi_0}h_z. \tag{16}$$

On the other hand, we may suppose that the number of grains forming the ring is not large and there is a significant scatter of j_c. In this case the sum in the left part of Eq. (14) is determined by the critical current of the weakest contact, $j_c=j_{c,\min}$,

$$j=j_c\frac{2N^2r^2}{\Phi_0}h_z. \tag{17}$$

In both cases we obtain that, at sufficiently low h_z, the current j and consequently the magnetic moment of the cluster, M_c, are proportional to the critical current of the contacts and linearly depend on h_z.

C. How to estimate the size of the clusters

We obtained a linear dependence of j on h_z [Eqs. (16) and (17)] at sufficiently small amplitudes, when the condition $j\ll j_c$ is valid. With increasing h_z the screening current j increases and when $j\sim j_c$ Eq. (13) is not valid anymore and obviously the $j(h_z)$ dependence becomes nonlinear. Experimentally (see Fig. 2), we obtained a nonlinear $M_z(h_z)$ dependence for $h_z>1$ Oe. Using this value, the condition $j=j_c$, and $r\approx 4000$ Å we may estimate the size of the cluster N. In the case of identical contacts [Eq. (16)] we find $N<100$. In the case of a dominating weakest contact [Eq. (17)] we obtain $N<10$. This is a rather rough estimation, but the result seems to be reasonable—most of the clusters contain just several grains.

D. Contribution of clusters in dc magnetic field

We have found that in the linear regime the magnetic moment of clusters is proportional to j_c. It is well known that j_c quickly decreases with magnetic field H, so that j_c and hence M_c become negligibly small in a field $H_J\sim\Phi_0/(2\lambda+d)l$, where d is the width and l the length of the contact.[20] In our case $d\ll\lambda$ and $l\sim\lambda$ so that

$H_J \sim \Phi_0/\lambda^2$. At the same time the contributions of individual grains remains the same up to the field of penetration of vortices. At a first glance, the fields H_J (total suppression of the Josephson junctions) and H_{c1} of the grains are of the same order of magnitude $\sim\Phi_0/\lambda^2$. However, if vortex pinning is rigid, the individual grains behave as if they were in the Meissner state up to fields H much higher than H_{c1} (see below). This circumstance is essential for distinguishing between the contributions of separate grains and clusters.

Now we may state that the rapid decrease of $\chi(H)$ at low fields (see Fig. 4) is due to the suppression of the cluster contribution. One may see that this contribution does not exceed 15% in our case. We note that in general the magnetic moment of currents flowing between grains may be not small. It follows from Eqs. (16) and (17) that if j_c is high, or if the characteristic size of the clusters is large, their contribution to susceptibility may be much more than the intragrain contribution. It seems that this situation was observed for Rb_xC_{60} samples with a little excess of Rb ($x \geqslant 3$), where quenching from 250 °C led to a giant increase of screening currents flowing in clusters.[21]

Finally, we obtain that at $H > 0.5$ kOe, where $\chi(H)$ reaches a plateau, the diamagnetic moment is only due to the screening currents flowing in individual grains.

E. Contribution of individual grains in dc magnetic field

At $H < H_{c1}$ the measured ac susceptibility is equal to the Meissner susceptibility [see Eq. (8)]. The Meissner susceptibility does not depend on magnetic field, since the corrections to λ are of the order H/H_{c2} and therefore negligibly small.

At $H > H_{c1}$ apart from the Meissner screening currents one also has to take into account the movement of vortices, which increases the ac penetration depth. The ac field h_z perpendicular to the dc field H tries to turn vortices in the direction of the total field $(\mathbf{H}+\mathbf{h}_z)$. Pinning may prevent vortices from turning along the field, so the magnetic moment $\mathbf{M}$ of the sample may not be parallel to $(\mathbf{H}+\mathbf{h}_z)$. In order to determine M_z and χ in this case one has to know the pinning force. The linear ac penetration depth for an isotropic superconductor containing a vortex lattice is $\lambda_{ac}^2 = \lambda^2 + \lambda_C^2$.[24] Here $\lambda_C^2 = B\Phi_0/(4\pi\alpha_L)$ is the Campbell penetration depth, α_L being the Labusch parameter. B is the magnetic induction; in our case $B=H$ because our particles are practically transparent to the field.

Let us consider pinning to be so rigid ($\lambda^2 \gg \lambda_C^2$) that vortices remain fixed in an ac field. In this case the presence of vortices has no influence on the behavior of the sample in an ac field and both M_z and χ coincide with their values in the Meissner state [Eqs. (8) and (9)]. Experimentally we should obtain a plateau on the $\chi(H)$ dependence up to a field much higher than H_{c1}. Therefore Eq. (9) is valid everywhere on the plateau and one may obtain $\lambda(T)$ dependence with a very high precision.

The observed decrease of χ at $H > 3$ kOe (see Fig. 4) evidently is connected with the movement of vortices in the ac field. With increasing H the Campbell penetration depth becomes comparable to λ and the susceptibility $\chi \propto M_z$ decreases as

$$M_z(H) = \chi_{max} k(r/\lambda_{ac})^2. \tag{18}$$

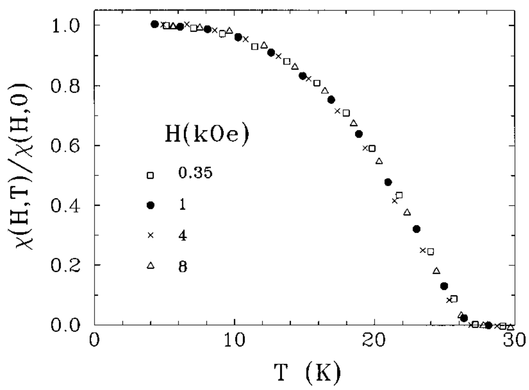

FIG. 5. Temperature dependence of normalized ac susceptibility at different dc magnetic fields.

This expression means that at $H \geqslant 3$ kOe the temperature dependence of χ is determined by the temperature dependence of the Labusch parameter α_L. However, Eq. (18) does not take into account the equilibrium magnetic moment. If the elastic pinning force decreases with increasing H (i.e., α_L increases with H) then we may reach the limit of very weak pinning when M_z is determined by the equilibrium diamagnetic moment.

On cooling in a dc magnetic field H, the sample acquires the equilibrium diamagnetic moment $M(H)$, which for $2H_{c1} < H \ll H_{c2}$ may be expressed as

$$M(H) = \frac{1}{\lambda^2} f(H,\lambda,\xi). \tag{19}$$

Here the function $f(H,\lambda,\xi) \propto \ln(\beta H_{c2}/H)$ (β is constant) scarcely depends on temperature.[22,23] A weak alternating field ($h_z \ll H$) hardly changes the absolute value of magnetic moment $M(H+h_z) \approx M(H)$. However, at each moment h_z tries to turn $\mathbf{M}(\mathbf{H}+\mathbf{h}_z)$ in the direction of the total field $(\mathbf{H}+\mathbf{h}_z)$. If pinning is very weak then $-\mathbf{M}$ is always parallel to the total field $(\mathbf{H}+\mathbf{h}_z)$. In this case $M_z = M(\mathbf{H}+\mathbf{h}_z)\sin\alpha$, where α is the angle between $\mathbf{H}$ and $(\mathbf{H}+\mathbf{h}_z)$. For $h_z \ll H$ we may write

$$M_z(\mathbf{H}+\mathbf{h}_z) \approx M(H)\frac{h_z}{H}. \tag{20}$$

We should note that this expression, derived for very weak pinning and $h_z \ll H$, is valid both at $H > H_{c1}$ and in the Meissner state, since in the Meissner state $\mathbf{M}$ and $(\mathbf{H}+\mathbf{h}_z)$ obviously coincide. It follows from Eq. (20) that χ decreases with increasing field H. If we neglect the logarithmically slow dependence of $M(H)$ then $\chi(H)$ should decrease as $1/H$, which is in good agreement with experiment at $H > 3$ kOe. Equation (20) also predicts the same temperature dependence of normalized susceptibility as in the Meissner state [see Eq. (9)].

Figure (5) shows the experimental temperature dependence of normalized susceptibility, obtained at different dc magnetic fields. We compare the curves obtained on the plateau 0.5 kOe $< H <$ 2 kOe, where pinning is rigid, and at higher H. Really all the curves coincide with a very good accuracy in agreement with Eq. (9). Hence we may conclude that our experimental results at $H < 2$ kOe are in good agreement with the case of rigid pinning, and at $H \geqslant 4$ kOe with the case of very weak pinning.

The transition from rigid to weak pinning takes place in a field $H_p \approx 3$ kOe when $\lambda \approx \lambda_C$. Then we can make a rough estimation of the Labusch parameter $\alpha_L \approx H_p \Phi_0/(4\pi\lambda^2)$. Substituting $\lambda \approx 4600$ Å and $H_p \approx 3$ kOe we obtain $\alpha_L \approx 2\times 10^4$ dyn/cm^2. However, we will show below that this value of α_L may be significantly overestimated.

In the above considerations we used an expression for λ_C which does not take into account the interaction of vortices with the surfaces parallel to **H** and perpendicular to $\mathbf{h}_z$. It may be considered as the interaction of vortices with their images and with the Meissner current, which leads to the well-known Bean-Livingston surface barrier. This interaction exerts on a tilted vortex a force which acts in the same direction as the elastic pinning force.[25] In other words the surface barrier prevents vortices from turning along the field. For small particles this effect may be much more important than the elastic pinning force. In this case λ_C is determined by the surface barrier and increases with H since the surface barrier decreases with increasing H. The surface barrier vanishes in a field $H_c \simeq \Phi_0/(4\pi\lambda\xi)$, which should correspond to the transition from rigid to weak pinning. Substituting the coherence length $\xi \simeq 30$ Å we obtain $H_c \simeq 1$ kOe, which is close to the experimental value $H_p \approx 3$ kOe.

Finally we note that the correct explanation of the field and temperature dependence of χ at $H > 3$ kOe requires exact calculations, which take into account both the elastic pinning force and the surface barrier. However, it does not change the central result that everywhere on the plateau χ is equal to the Meissner susceptibility of individual grains.

VI. TEMPERATURE DEPENDENCE OF LONDON PENETRATION DEPTH

In the preceding sections we established that at $H > 0.5$ kOe the diamagnetic moment of Rb_3C_{60} powder is determined by the intragrain contribution only. The normalized susceptibility exhibits a unique temperature dependence, which is the temperature dependence of the inverse square of the normalized penetration depth $L^{-2}(T)$ in accordance with Eq. (9).

Without taking into account fluctuations,[20] the $L^{-2}(T)$ function has a finite derivative at $T = T_c$. As the temperature decreases this derivative is constant in the temperature interval $\approx 0.15 T_c$ near T_c, i.e.,

$$\chi(T) \propto L^{-2}(T) = \beta(1 - T/T_c). \tag{21}$$

However, the experimental curve deviates from the linear dependence near T_c (Fig. 6). The scale of the fluctuation contribution is given by the Ginsburg number G_i, which is quite small, $G_i \approx 10^{-4}$ in Rb_3C_{60}.[26] Moreover, both Gaussian[27] and critical[28] fluctuations in the three-dimensional (3D) case lead to the increase of the absolute value of the derivative dL^{-2}/dT with increasing T, which is opposite to our experimental results. Thus the deviation of experimental $\chi(T)$ from linear dependence (21) is not 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. (9)

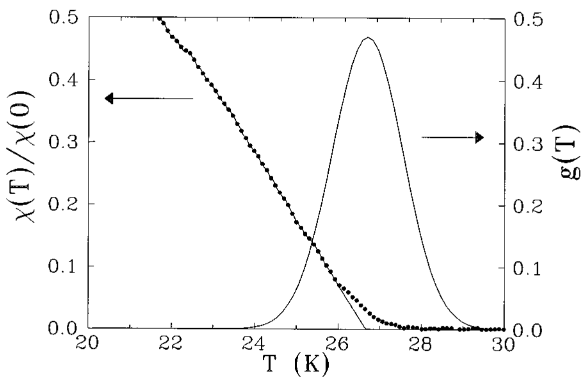

FIG. 6. $\chi(T)$ (solid line), $L^{-2}(T)$ (dotted line), and T_c distribution function $g(T)$ near the superconducting transition. $\chi(T)$ and $L^{-2}(T)$ coincide below 26 K.

$$\frac{\chi(T)}{\chi(0)} = \int_T^\infty g(t) L^{-2}(T/t)\, dt. \tag{22}$$

If the distribution function width is less than $0.15T_c$, we can substitute $\beta(1 - T/t)$ for L^{-2} in Eq. (22), which leads to

$$g(T) = \frac{T}{\beta}\frac{d^2}{dT^2}\left[\frac{\chi(T)}{\chi(0)}\right]. \tag{23}$$

The experimental curve deviates from the linear dependence in a narrow temperature range $\Delta T < 0.07T_c$, so the characteristic width of the distribution function is noticeably smaller than $0.15T_c$. In this case β is equal to the slope of the straight line approximating the experimental $\chi(T)/\chi(0)$ dependence in the range $0.85T_c < T < 0.93T_c$. We find $\beta = 2.6$ and from Eq. (23) we obtain $g(T)$. To estimate the width of the distribution function we fit the $g(T)$ derived from Eq. (23) by the Gaussian distribution

$$g(T) = \frac{1}{\sqrt{2\pi}\epsilon}\exp\left(-\frac{(T - T_0)^2}{2\epsilon^2}\right) \tag{24}$$

which leads to the values of $T_0 = 26.7$ K and $\epsilon = 0.85$ K (Fig. 6). We should note that the real distribution over T_c may differ significantly from the Gaussian one. For example, if $g(T)$ is due to 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, this does not change the slope β because of the small width of the distribution function. Now we discuss the obtained $\lambda(T)$ dependence (see Fig. 7).

The BCS temperature dependence of λ substantially depends on the relation among London penetration depth $\lambda_L = (m^* c^2/4\pi n e^2)^{1/2}$ (here n and m^* are the carrier density and effective mass, respectively, and e is the electron charge), coherence length $\xi_0 = \hbar v_F/\Delta_0$ (v_F is the Fermi velocity), and mean free path l. In particular, the local (London) limit is valid when $\lambda_L \gg \xi$; 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}: $\xi_{GL}(0) \simeq 30$ Å.[14,29–31] In the clean ($\xi_0 \ll l$) limit ξ_0 is approximately equal to $\xi_{GL}(0)$, and $\xi_{GL}(0) \approx (\xi_0 l)^{1/2}$ in the dirty limit ($\xi_0 \gg l$). Hence the upper estimation of ξ is

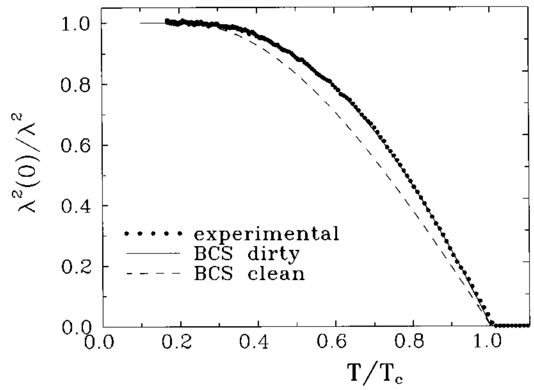

FIG. 7. Comparison of the experimental dependence $L^{-2}(T/T_c)=\lambda^2(0)/\lambda^2(T/T_c)$ with the BCS dependences in clean and dirty limits.

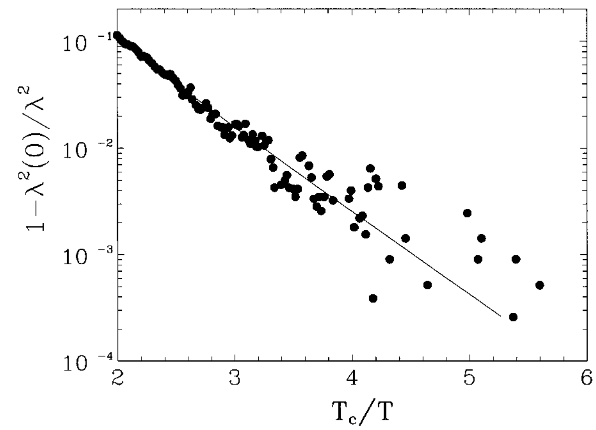

FIG. 8. Experimental $1-L^{-2}(T_c/T)$ dependence at $T<0.5T_c$.

≈ 30 Å. Comparison with the value of $\lambda(0)\approx 4600$ Å shows that in Rb_3C_{60} the local limit is realized for any relationship between ξ_0 and l.

The question about the clean and the dirty limits is still open. Uemura *et al.*[32] estimated the mean free path $l\simeq 70$ Å at $T=T_c$ and comparing it to $\xi\simeq 30$ Å consider that Rb_3C_{60} is a clean superconductor. On the other hand, Palstra *et al.*[33] estimated the coherence length to be $\xi_0\simeq 140$ Å and made the conclusion that Rb_3C_{60} is in the dirty limit. The estimate of $l\simeq 12$ Å at $T=0$ from the upper critical field data[34,35] is also consistent with the dirty limit.

Figure 7 shows the comparison of the measured $L^{-2}(T/T_c)$ with the BCS dependences [calculated using Eq. (5.33) from Ref. 36] in the clean and dirty limits. We note that in the dirty limit

$$\frac{1}{\lambda^2(T)}=\frac{1}{\lambda_L^2}\left(\frac{l}{\xi_0}\right)\left(\frac{\Delta(T)}{\Delta_0}\right)\tanh\left(\frac{\Delta(T)}{2k_BT}\right). \tag{25}$$

There is a significant difference between BCS clean and BCS dirty $L(T/T_c)$ temperature dependences at $T>0.4T_c$. One may see that the experimental $L(T/T_c)$ dependence is perfectly accounted for by the BCS dirty limit. The experimentally obtained slope $\beta=2.6$ is very close to the BCS dirty limit value, $\beta=2.62$, and noticeably differs from the clean limit value, $\beta=2$.

Figure 8 shows the experimental $1-L^{-2}(T_c/T)$ dependence at $T/T_c<0.5$ in a semilogarithmic scale. The results are in good agreement with the low temperature BCS dirty limit expression $L^{-2}=1-2\exp(-\Delta_0/k_BT)$ with $\Delta_0/k_BT_c=1.76$.

It is believed that the low temperature dependence of the penetration depth is a probe of the pairing state. For example, in the case of line nodes in the gap function $L^{-2}(T)$ should exhibit a power law temperature dependence. However, a power law dependence may also arise due to phase fluctuations[37] or due to inelastic scattering.[16,38] The latter two reasons crucially depend on T_c and in Rb_3C_{60} with $T_c\approx 30$ K their contribution should be negligibly small. Thus in Rb_3C_{60} the temperature dependence of the London penetration depth at $T\ll T_c$ is a valid probe of the symmetry of the pairing state. The experimentally obtained exponential behavior of $\lambda(T)$ at low temperatures proves s-wave pairing in Rb_3C_{60}.

Finally, we briefly discuss the relation between $\lambda(0)$ and T_c. The zero temperature penetration depth in the dirty limit may be written as

$$\lambda^{-2}(0)=\frac{4\pi ne^2}{m^*c^2}\left(\frac{l}{\xi_0}\right). \tag{26}$$

Substituting $\xi_0=\hbar v_F/\Delta_0$ with $\Delta_0=1.76k_BT_c$ and introducing the relaxation time $\tau=l/v_F$ we obtain the "conductivity" at $T=0$:

$$\sigma(0)\equiv\frac{ne^2\tau}{m^*}=\frac{\hbar c^2}{4\pi\lambda^2(0)1.76k_BT_c}. \tag{27}$$

We used quotes because $\sigma(0)$ is not a directly measurable conductivity, since Rb_3C_{60} is superconducting below 27 K. Using $\lambda_0\approx 4600$ Å and $T_c\approx 27$ K we find $\sigma(0)\approx 6\times 10^3\ \Omega^{-1}\ \mathrm{cm}^{-1}$. It follows from Eq. (27) that in the case of the dirty limit the remarkable variation $\lambda^{-2}(0)\propto T_c$ found for Rb_3C_{60}, K_3C_{60}, and Na_2CsC_{60} (Ref. 32) means that these superconductors have approximately the same residual conductivity $\sigma(0)$.

In conclusion, we have measured the temperature dependence of the penetration depth $\lambda(T)$ for Rb_3C_{60}. The experimental $\lambda(T)$ is perfectly described by the BCS dependence in the dirty limit with a weak coupling gap $\Delta_0=1.76k_BT_c$.

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