LOWER CRITICAL MAGNETIC FIELD OF SUPERCONDUCTING Rb_3C_{60}

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The lower critical magnetic field H_{c1} of superconducting Rb_3C_{60} (T_c = 28.5 K) is estimated by different methods. The zero temperature value is found to be given by $H_{c1} = 16.2 \pm 1.0$ mT, and the penetration depth is $\lambda_L = 215 \pm 10$ nm. The Bean model analysis leads to threshold fields of 5.3 mT for T = 5 K, and 4.0 mT for T = 17 K. The big influence of intergranular connections on H_{c1} is demonstrated. Good agreement between the low- and the high-field analysis is found.

The discovery of superconductivity in alkali-doped fullerenes[1–3] resulted in wide interest in this new non-oxide high-T_c superconductors. Presently intensive investigations of the structure (see e.g., Refs. 4 and 5) and physical properties (see e.g., Refs. 6–8) of this materials take place. Quite naturally the lower and upper critical magnetic fields, penetration depth and coherence length are among the most interesting quantities characterizing this new superconductor.

The value of the upper critical magnetic field H_{c2} for Rb_3C_{60} obtained in Ref. 8 and Refs. 7 and 9 and the coherence length evaluated using these results have been in good agreement. However, the determination of the lower critical magnetic field H_{c1} is still controversial. Several groups[7,8] obtained the value H_{c1} = 12–16 mT for Rb_3C_{60} using standard low-field analysis, in which H_{c1} is determined from the point of deviation of the $M(H)$ curve from linear behaviour. Although these results are close to each other, we note that powder samples have been studied. In addition, there is barely enough of a linear part on a $M(H)$ curve but it almost can be considered a parabola starting from zero applied magnetic field. Moreover, ambiguities remain in determining the point of deviation from linear behaviour. Hence we believe it is important to use also different methods to determine the lower critical magnetic field, and compare the results, which is done in the following.

In this paper, we report our investigation of the magnetization curve of the Rb_3C_{60} compound (below T_c) from which we estimate the penetration depth λ_L, and hence the lower critical magnetic field. The Rb_3C_{60} sample was prepared by direct reaction of C_{60} powders with high-purity liquid Rb. For magnetic measurements 27 mg of fine Rb_3C_{60} powder specimen was sealed in a perspex cylindrical container. The electron microscopy study showed that the distribution of size particles ranges from 0.1 to 3 μm, the average grain size being close to 1 μm. More details of the sample preparation and characterization may be found in Ref. 7.

For the measurements, a high sensitivity SQUID magnetometer (Quantum Design) was used. The experiments were performed at magnetic fields -5 T $\leq H \leq$ 5 T within the temperature region 2 K $\leq T \leq$ 300 K. The instrumental sensitivity was verified using high-purity spheres of Bi and Nb, along with a Pt metal standard.

It is known that quantized magnetic field lines (vortices) penetrate into a type-II superconductor when it is immersed in a magnetic field H_a which is larger than the lower critical field H_{c1}. The lower critical field, and hence the penetration depth λ_L (provided the coherence length φ is known from independent measurements of, say, the upper critical field H_{c2}), can be determined from an experimental magnetization curve. Such a curve can be analyzed, and compared with theoretical results, in the low ($H_a \gtrsim H_{c1}$), intermediate ($H_{c1} \ll H_a \ll H_{c2}$), and high ($H_a \lesssim H_{c2}$) field regimes.

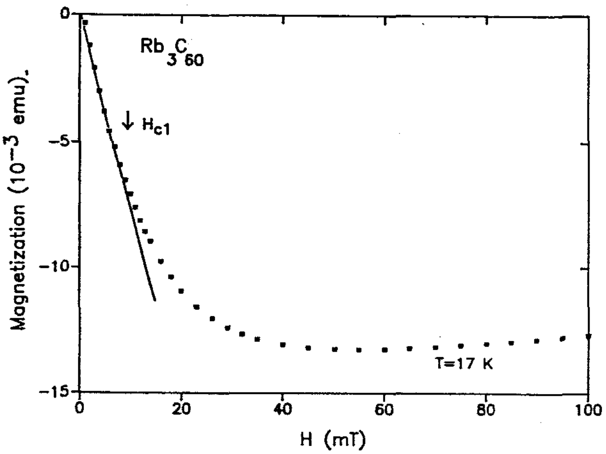

Fig. 1. Magnetization curve for Rb_3C_{60} at $T = 17$ K.

As an example, we show in Fig. 1 the magnetic field dependence of a magnetization at $T = 7$ K, obtained by cooling the specimen from the normal state down to the indicated temperature under zero external magnetic field, followed by a measurement of the magnetization at increasing field. Then we determine H_{c1} as the field where a deviation from a linear $M(H)$ dependence sets in; as mentioned above, this is a quite problematic and very subjective procedure, but we nevertheless proceed along these lines for comparison.

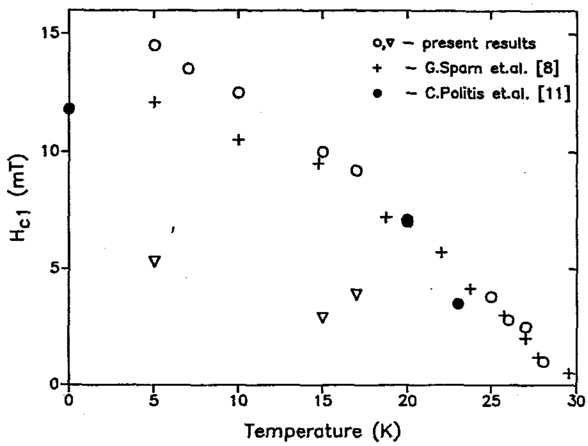

Fig. 2. Comparison of results for the lower critical field vs. temperature. ○: As determined from the deviation from the linear dependence (compare Fig. 1). ▽: Results from the analysis based on Eq. (2), as discussed in the text.

In Fig. 2 we show the temperature dependence of H_{c1} thus determined. Extrapolation of this data to zero temperature gives $H_{c1} = 16.2 \pm 1.0$ mT. Using the standard expression

$$H_{c1}(0) = (\phi_0/4\pi\lambda_L^2)\ln(\lambda_L/\varphi) \tag{1}$$

and the value $\varphi(0) = 2.7$ nm,[9] we find the penetration depth at zero temperature to be given by $\lambda_L(0) = 215 \pm 10$ nm.

In order to make this procedure more quantitative, it is tempting to apply the Bean critical state model[10] for the entry of vortices into superconductors containing pinning centers. According to this theory, for fields above H_{c1}, the magnetization is related to the critical current density j_c (assumed to be field independent for simplicity) by:

$$(4\pi M + H_a) \sim (H_a^2 - H_{c1}^2)/(j_c \cdot D) \tag{2}$$

where D is a characteristic length for the superconducting sample studied. (In our case, for a powder sample, we expect D to be of the order of the grain size, $\sim 1\ \mu$m.) The above relation (2) holds in the range $H_{c1} < H < H^*$, where $H^* \sim j_c D$ is the field at which the field enters completely into the sample. Thus a plot of δM, where $\delta M = M + H_a/4\pi$ is the deviation of the observed magnetization from perfect diamagnetic behaviour, and in particular the threshold field of this plot, should give the lower critical field. Figure 3 shows typical examples of such a plot for our sample for two different temperatures. Indeed, the curves are linear for a reasonable range; we emphasize, however, that the value of H_{c1} thus determinated is about three times smaller than as determinated above (see Fig. 2). In order to illustrate the ambiguities of such an analysis, we remark that we have also plotted $(\delta M)^{1/2}$

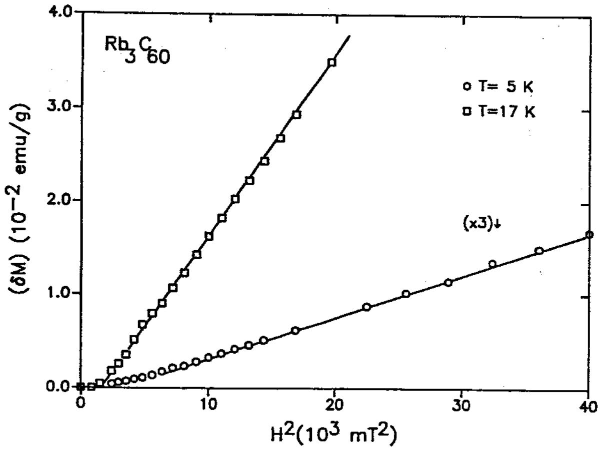

Fig. 3. The deviation of the magnetization $\delta M = M + H_a/4\pi$ vs. applied magnetic field H_a^2 at $T = 5$ K and $T = 17$ K.

vs. H_a; again, this plot is linear over a reasonable range, but the threshold thus determined is about a factor two smaller than the just mentioned result derived from δM vs. H_a^2.

Thus we conclude that the analysis based on the application of the Bean model does not give too reliable results, and we believe this is related to the fact that a powder sample was studied. Such a sample consists of weakly (Josephson) coupled grains, and quite naturally the breaking of the Josephson intergranular coupling is an important effect at low external field. A more detailed theoretical description of this phenomenon is required before a more quantitative analysis of the low-field data can be attempted.

More reliable estimates can, in fact, be obtained from the intermediate and high field range, as described recently.[11] This analysis has been based on the following expressions for the magnetization, valid for intermediate fields ($H_{c1} \ll H_a \ll H_{c2}$):

$$-4\pi M = \frac{\alpha\phi_0}{8\pi\lambda_L^2} \cdot \ln\left(\frac{\beta H_{c2}}{H_a}\right) \tag{3}$$

with suitably chosen constants α, β,[12] and the linear high field ($H_a \lesssim H_{c2}$) or

$$-4\pi M = \frac{H_{c2} - H_a}{1.16(2K^2 - 1)} \,. \tag{4}$$

Applying the above theoretical expressions (3) and (4) to an analysis of data obtained at $T = 20$ K and $T = 23$ K, respectively, and assuming the two-fluid-model temperature dependence to be valid, it was found that $H_{c1}(0) = 8.8 - 11.4$ mT, slightly lower than the present estimate. Essentially, however, these results are consistent with values for K_3C_{60}, namely $H_{c1}(0) = 13.2$ mT and $\lambda_L(0) = 240$ nm, and the results obtained in Ref. 8, $H_{c1}(0) = 12$ mT and $\lambda_L(0) = 247$ nm, which is also apparent from the explicit comparison of data given in Fig. 2. For completeness and easy reference, a comparison of superconductivity parameters is also given in Table 1. (The value K, in contrast to Ref. 11 is computed here as λ_{2fl}/φ_0 since $\lambda_{2fl}(0)$ is believed to be a better approximation to the zero temperature penetration depth than $\lambda_{GL}(T)$ extrapolated to $T = 0$.)

Table 1.

Superconducting parameter	Low-field analysis		Intermediate and high-field analysis[11]
	Ref. 9 and this results	Ref. 8	
T_c(K)	28	29.6	
H_{c1}(mT)	16	12	9–11.4
H_{c2}(T)	46.5	78	
H_c(T)	4.1	4.4	3–3.4
$j_c(10^6$ A/cm$^2)$	4($H = 1$ T, $T = 7$ K)	1.5 ($H = 1$ T, $T = 5$ K)	
φ_0 (nm)	2.7	2.0	
λ_L (nm)	215	247	240–280
$k = \lambda_L/\varphi_0$	80.5	123.5	104–122(λ_{2fl}/φ_0)

In summary, different experiments and procedures to analyse magnetization curves have been studied, from which a consistent picture of critical field results has emerged. The results show unambiguously that A_xC_{60} is an extreme type-II superconductor (with $K \sim 100$). However, as discussed above, open questions of the interpretation of the low-field data in terms of the Bean model remain, which we argued are related to the fact that powder samples have been investigated.

References

1. A. F. Hebard, M. J. Rosseinsky, R. C. Haddon, D. M. Murphy, S. H. Glarum, T. T. M. Palstra, A. P. Ramirez, and A. R. Kortan, *Nature* **350**, 600 (1991).
2. K. Holczer, O. Klein, S. M. Huang, R. B. Kaner, K. J. Fu, R. L. Whetter, and F. Diederich, *Science* **252**, 1154 (1991).
3. M. J. Rosseinsky, A. P. Ramirez, S. H. Glarum, D. W. Murphy, R. C. Haddon, A. F. Hebbard, T. T. M. Palstra, A. R. Kortan, S. M. Zahurak, and A. V. Makhija, *Phys. Rev. Lett.* **66**, 2830 (1991).
4. P. W. Stephens, L. Myhaly, P. L. Lee, R. L. Whetten, S. M. Huang, R. Kaner, F. Diederich, and K. Holtczer, *Nature* **351**, 632 (1991).
5. D. L. Novikov, V. A. Gubanov, and A. J. Freeman, *Physica* **C191**, 399 (1992).
6. K. Holtczer, O. Klein, G. Grüner, J. D. Thompson, F. Diederich, and R. L. Whetten, *Phys. Rev. Lett.* **67**, 271 (1991).
7. C. Politis, V. Buntar, W. Krauss, and A. Gurevich, *Europhys. Lett.* **17**, 175 (1992).

8. G. Sparn, J. D. Thompson, R. L. Whetten, S. M. Huang, R. B. Kaner, F. Diederich, G. Grüner, and K. Holtczer, *Phys. Rev. Lett.* **68**, 1228 (1992).
9. C. Politis and V. Buntar, to be published.
10. C. P. Bean, *Phys. Rev. Lett.* **8**, 250 (1962).
11. C. Politis, A. Sokolov, and V. Buntar, *Mod. Phys. Lett.* **B6**, 351 (1992).
12. Z. Hao and J. R. Clem, *Phys. Rev. Lett.* **67**, 2371 (1991).