Magnetic field studies of superconducting UBe_{13} thin films

P. M. Tedrow and J. H. Quateman*

Francis Bitter National Magnet Laboratory, Cambridge, Massachusetts 02139

We have made superconducting thin films of UBe_{13} by vapor codeposition onto heated sapphire substrates. The critical magnetic field and normal-state magnetoresistance of the films are similar to those of bulk material. The magnetoresistance has been measured in the temperature range 0.4 to 40 K in fields up to 20 T. We find that plots of isotherms of the magnetoresistance $[R(0)-R(H)]/R(0)$ versus magnetic field H show a universal behavior for $T > 2$ K if the scale of the magnetic field is compressed as T increases.

I. INTRODUCTION

The superconductivity of heavy-electron materials has attracted the attention of theoretical and experimental physicists in recent years[1] because of the possibility that these materials may manifest some form of cooperative state more exotic than the ordinary BCS phonon-mediated superconductivity. Unfortunately, there is a dearth of qualitative experiments which can be done on bulk samples and which offer a clear distinction between BCS and other proposed forms of superconductivity. Thus, while there are suggestive results from, for example, NMR (Ref. 2) or ultrasonic attenuation[3] or specific-heat[4] measurements, controversy still exists.

Some hope has been raised that proximity-effect or electron-tunneling measurements might clarify the nature of the electron transport in these heavy-electron materials.[5,6] These measurements are most easily carried out on thin-film samples. The purpose of this article is to report the successful production[7] of a thin-film superconducting heavy-electron material; in this case, UBe_{13}. We have made transition-temperature, critical magnetic field, and normal-state magnetoresistance measurements which demonstrate the similarity of the films to bulk material. In addition, we have made x-ray measurements which verify the existence of the proper phase.

We have chosen to study UBe_{13} because, of the known heavy-electron materials, it appeared to be the easiest to make from a metallurgical point of view and has the highest T_c,[1] about 0.9 K. Other possible candidates require very-well-ordered material or appear to be too complex. The disadvantage of using UBe_{13} is that one must deal with the toxicity of Be.

Bulk samples of the compound UBe_{13} have been extensively studied by many techniques. Superconductivity was first observed by Bucher *et al.*[8] Subsequently, critical magnetic field,[7,9–11] specific-heat,[4,12] magnetic susceptibility,[12] magnetoresistance,[7,11,13] ultrasonic attenuation,[3] and NMR relaxation-time[2] measurements have been made by various groups. Many of these properties are anomalous compared to the predictions for simple BCS superconductors, but there does not yet appear to be conclusive evidence as to the nature of the superconducting state.

II. EXPERIMENTAL RESULTS

The UBe_{13} films were made by vapor codeposition onto heated sapphire substrates. The Be was evaporated from an electron-gun source, while the U, in the form of 0.05-mm spheres, was sprinkled onto a hot tungsten filament using a magnetic vibrator. The amount of each material being deposited was monitored separately using two quartz-crystal monitors. The key to success appears to be the achievement of a controlled U deposition rate. The low vapor pressure and high rate of oxidation of U make this goal difficult to achieve. Film thicknesses were in the range of 100–500 nm. The substrates were held at 700 °C during deposition and were heated to 1100 °C for about 1 min following deposition. Measurements of the film resistance were made using the standard dc four-terminal technique. The measuring current was usually about 10 μA, corresponding to a current density of the order of 1 A/cm^2. The films were cooled using a booster-pumped ^3He cryostat. Magnetic fields up to 20 T were employed for the normal-state magnetoresistance measurements. In addition, x-ray diffractometer scans were used to verify the existence of the UBe_{13} phase. The films were shown to consist of randomly oriented crystallites. The films were very stable and retained their properties for at least
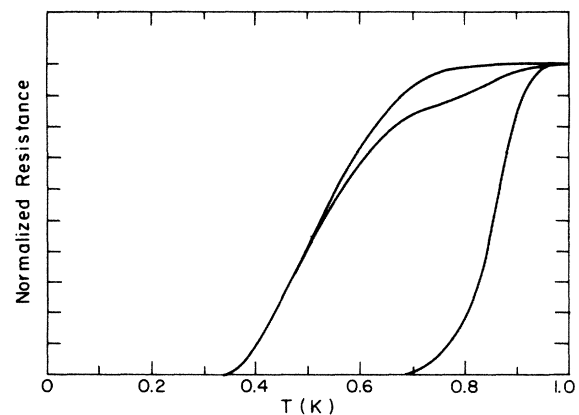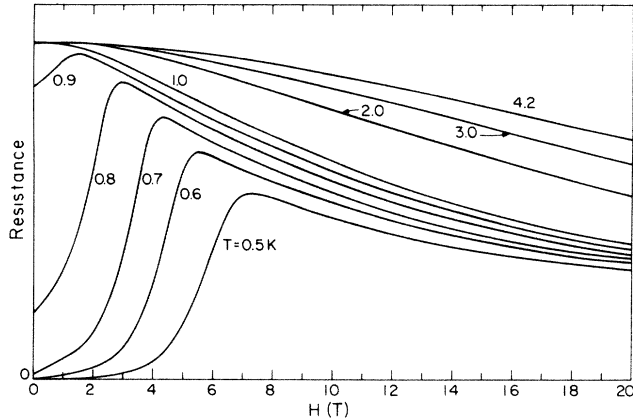

FIG. 1. Resistive transitions of three UBe_{13} films.

FIG. 2. Resistance versus magnetic field at various temperatures for a UBe_{13} film.

up to a year without special storage precautions.

The resistive transition as a function of temperature for several films is shown in Fig. 1. The transition temperature for bulk UBe_{13} is about[8–11] 0.9 K. The best films showed an onset of superconductivity between 0.9 and 0.95 K. The transition was about 0.2 K wide, much broader than those observed in bulk samples. Many films had lower transition temperatures while some showed a double transition. Examples are shown in Fig. 1. We did not observe any transitions at temperatures between those of the films represented in the figure. A number of films which showed an increase in resistance on cooling and a large negative magnetoresistance did not become superconducting.

Figure 2 shows the resistance of a UBe_{13} film versus applied magnetic field for various temperatures above and

below T_{c0}. The strong negative magnetoresistance observed in bulk samples[9–11] is evident in these data as well. This negative magnetoresistance could be seen at temperatures up to about 40 K. At higher temperatures, the magnetoresistance appeared to be small and positive.

Figure 3 shows the critical magnetic field of the same film as a function of temperature. The data of Maple et al.,[9] Rauchschwalbe et al.,[10] and of Remenyi et al.[11] for bulk samples are also indicated. Because of the unusual shape of the resistive transitions (Fig. 2), the critical field was defined as the field at which the resistance reached one-half of the peak value for each temperature. We note that there is a small anisotropy in H_{c2} with $H_{c2||} > H_{c2\perp}$. Also, $H_{c2\perp}$ shows an upward curvature with decreasing temperature. Whether these features are fundamental or artifacts of the method of defining H_{c2} must be examined further; however, the data of Rauchschwalbe et al.[10] and Remenyi et al.[11] show similar curvature.

III. DISCUSSION

A. Magnetoresistance

We have calculated $R_M = [R(0) - R(H)]/R(0)$ for various temperatures from data such as those shown in Fig. 2, and have found that by adjusting the scale of H, we can produce the curves of R_M versus $H/H_0(T)$ in Fig. 4. The curves of R_M versus $H/H_0(T)$ for temperatures from 2 to 40 K coincide to within the width of the line in the figure. As shown by the points for $T = 1$ K in Fig. 2, however, for temperatures below 2 K, R_M rises more rapidly for small H and less rapidly for large H than the universal curve for higher T.

Figure 5 shows $H_0(T)$ versus T, demonstrating that H_0 is linear in T for high T. We arbitrarily choose the scale of $H_0(T)$ to be such that the slope $dH_0/dT = k_B/\mu_B$, Boltzmann's constant divided by the Bohr magneton. This choice of scale is also used in Fig. 4. The linear portion of $H_0(T)$ extrapolates to $19T$ at $T = 0$. Scaling of R_M for bulk samples has been observed previously.[13]
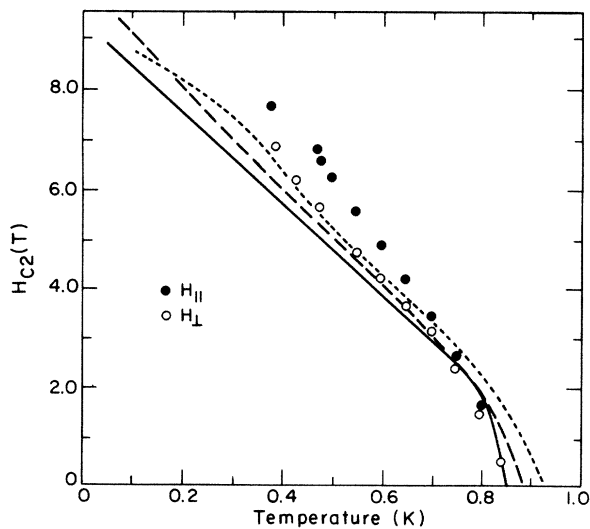

FIG. 3. Critical magnetic field versus temperature for a UBe_{13} film in parallel (●) and perpendicular (○) fields. Data on bulk samples from Maple et al. (Ref. 9) ———, Rauchschwalbe et al. (Ref. 10) — — —, and Remanyi et al. (Ref. 11) — — are also shown.

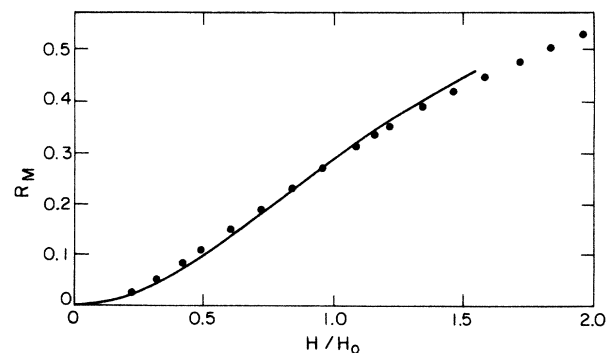

FIG. 4. $R_M \equiv [R(0) - R(H)]/R(0)$ vs H/H_0. The solid line represents data for $2 < T < 40$ K. The points are for $T = 1$ K. See text for choice of H_0.
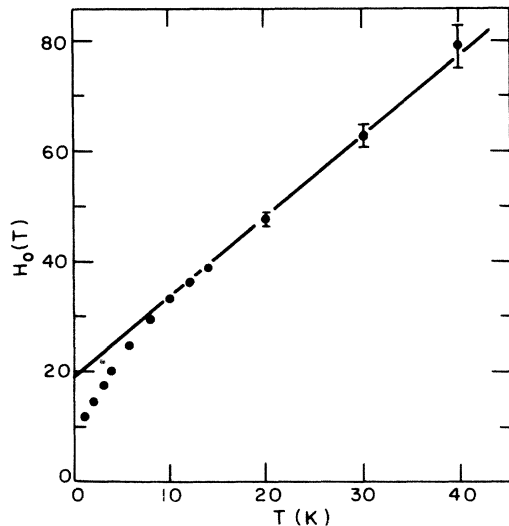
FIG. 5. H_0 vs T, with the scale of H_0 chosen to make the slope of the straight line equal to k_B/μ_B.

B. Critical magnetic field

As shown in Fig. 3, the critical magnetic field of the films is qualitatively similar to measurements on bulk samples. The upturn in H_{c2} with decreasing temperature was seen also by Rauchschwalbe et al.[10] and Remenyi et al.[11] The former authors also showed that the inclusion of temperature- and field-dependent normal-state resistivity, specific heat, and magnetic susceptibility in the theory of H_{c2} could produce qualitatively this temperature dependence. More exact comparisons of experimental data with the theory await measurement of these normal-state properties.

The other qualitative feature of the critical field measurements is the difference between parallel and perpendicular H_{c2}. Since the x-ray analysis of the films indicates randomly oriented crystallites, the anisotropy of H_{c2} cannot be attributed to intrinsic crystalline anisotropy of UBe₁₃. It presumably is a geometrical effect arising from the fact that the film thickness is large compared to the coherence length and small compared to the penetration depth.

C. Critical temperature

All of the films made according to the technique described earlier showed an increase in resistance with decreasing temperature similar to that for bulk UBe₁₃. In addition, these films all showed large negative magnetoresistance. Not all, however, were superconductors, at least above 0.3 K. Of those that were superconductors, most had a T_c of about 0.6 K. Some had the bulk T_c, while a few showed a partial transition at 0.9 K and a broad decrease of resistance at lower temperature. The systematics of these sample variations has thus far not been elucidated, but films thinner than about 100 nm were never superconducting. The cause of the problem of making reproducible films lies in the affinity of U for oxygen and the low vapor pressure of U. These two properties make evaporation of U difficult without a powerful e-gun source (U quickly dissolves any refractory metal boat). Our 3-kW gun could not evaporate the U fast enough to keep the charge from oxidizing. Once a skin of oxide formed, the gun did not have enough power to break through it. Thus we were forced to use the flash evaporation method which is difficult to control and gassy.

IV. SUMMARY

We have produced superconducting films of UBe₁₃ and studied their properties as a function of magnetic field. The transition temperature, critical field, and normal-state magnetoresistance were similar to those of bulk samples. Our results demonstrate the feasibility of making UBe₁₃ films by coevaporation and that, if a well-equipped evaporator could be dedicated to such a project, high quality UBe₁₃ films could be deposited reliably.

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*Present address: Raytheon Co., Research Division, 131 Spring Street, Lexington, MA 02173.

[1]See review articles by G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984); and P. A. Lee, T. M. Rice, J. W. Serene, L. J. Sham, and J. W. Wilkins, Comments Condensed Matter Phys. **12**, 99 (1986).

[2]D. E. MacLaughlin, C. Tien, W. G. Clark, M. D. Lan, Z. Fisk, J. L. Smith, and H. R. Ott, Phys. Rev. Lett. **53**, 1833 (1984); C. Tien, D. E. MacLaughlin, M. D. Lan, W. G. Clark, Z. Fisk, J. L. Smith, and H. R. Ott, Physica **135B**, 14 (1985).

[3]B. Batlogg, D. J. Bishop, E. Bucher, B. Golding, C. M. Varma, Z. Fisk, J. L. Smith, and H. R. Ott, Physica **135B**, 23 (1985); B. Batlogg, D. J. Bishop, B. Golding, C. M. Varma, Z. Fisk, J. L. Smith, and H. R. Ott, Phys. Rev. Lett. **55**, 1319 (1985);

B. Golding, D. J. Bishop, B. Batlogg, W. H. Haemmerle, Z. Fisk, J. L. Smith, and H. R. Ott, ibid. **55**, 2479 (1985).

[4]H. R. Ott, H. Rudigier, T. M. Rice, K. Ueda, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **52**, 1915 (1984); G. R. Stewart, Z. Fisk, J. L. Smith, J. O. Willis, and M. S. Wire, Phys. Rev. B **30**, 1249 (1984); G. R. Stewart, Z. Fisk, J. L. Smith, H. R. Ott, and F. M. Mueller, in Proceedings of the 17th International Conference on Low Temperature Physics (LT17), Karlsruhe, 1984, edited by U. Eckern et al. (North-Holland, Amsterdam, 1984), p. 321; M. H. Mayer, U. Rauchschwalbe, C. D. Bredl, F. Steglich, H. Rietschel, H. Schmidt, H. Wühl, and J. Beuers, Phys. Rev. B **33**, 3168 (1986).

[5]A. J. Millis, Physica **135B**, 69 (1985); E. W. Fenton, ibid. **135B**, 60 (1985); Solid State Commun. **34**, 918 (1985); B.

Ashauer, G. Kieselmann, and D. Rainer, J. Low Temp. Phys. 63, 349 (1986).

[6]E. L. Wolf, R. J. Noer, S. Han, K. W. Ng, T. P. Chen, D. K. Finnemore, and L. Tanner, Physica 135B, 65 (1985); S. Han, K. W. Ng, E. L. Wolf, H. F. Brown, L. Tanner, Z. Fisk, J. L. Smith, and M. R. Beasley, Phys. Rev. B 32, 7567 (1985).

[7]J. H. Quateman and P. M. Tedrow, Physica 135B, 154 (1985).

[8]E. Bucher, J. P. Maita, G. W. Hall, R. C. Fulton, and A. S. Cooper, Phys. Rev. B 11, 440 (1975).

[9]M. B. Maple, J. W. Chen, S. E. Lambert, Z. Fisk, J. L. Smith, H. R. Ott, J. S. Brooks, and M. J. Naughton, Phys. Rev. Lett.

54, 477 (1985).

[10]U. Rauchschwalbe, U. Ahlheim, F. Steglich, D. Rainer, and J. J. M. Franse, Z. Phys. B 60, 379 (1985).

[11]G. Remenyi, D. Jaccard, J. Flouquet, A. Briggs, Z. Fisk, J. L. Smith, and H. R. Ott, J. Phys, (Paris) (to be published).

[12]H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. 50, 1595 (1983).

[13]B. Batlogg, A. P. Ramirez, G. Boebinger, Z. Fisk, J. L. Smith, E. Bucher, and H. R. Ott, Bull. Am. Phys. Soc. 31, 445 (1986).