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Bulk superconductivity in the heavy-fermion superconductor UPt_3

T. T. M. Palstra, P. H. Kes, and J. A. Mydosh
Kamerlingh Onnes Laboratorium der Rijks-Universiteit Leiden,
2300 RA Leiden, The Netherlands

A. de Visser, J. J. M. Franse, and A. Menovsky
Natuurkundig Laboratorium der Universiteit van Amsterdam,
1018 XE Amsterdam, The Netherlands

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We have investigated the superconducting properties of UPt_3 by determining the Meissner effect via ac and dc susceptibility and magnetization measurements. We found an upper and lower limit of the superconducting volume fraction of 80% and 30% with magnetization and flux expulsion experiments, respectively. Thus, the superconductivity of UPt_3 is a bulk property.

Recently a new class of superconductors, the heavy-fermion superconductor, has attracted a great deal of interest. These superconductors are characterized by a high effective mass of the pairing electrons, which can reach up to 200 times the free-electron mass. This enhancement is explained by a strong interaction of the conduction electrons with the $4f$ or $5f$ electrons, that are located in a narrow band at the Fermi level. This interaction results in a high correlation of the electron system and causes the high effective mass. These systems have a high value of the specific-heat coefficient γ, which can be of order 1 $J/mol\,K^2$, and they also have very large values for the initial slope of the critical field $B_{c2}'(T_c) = -(dB_{c2}/dT)|_{T=T_c} \approx 5$–25 T/K. At present a number of compounds have been shown to belong to this class of superconductor, e.g., $CeCu_2Si_2$ (Ref. 1) and UBe_{13} (Ref. 2).

The compound UPt_3, discussed in this article, was also claimed to be a heavy-fermion superconductor.[3] However, in distinction to the above materials, UPt_3 exhibits strong spin fluctuations and even local moments at higher temperatures.[3-6] The superconductivity was deduced from the behavior of the resistivity, ac susceptibility, and specific heat. Nevertheless, the results of the first two methods can be explained by the superconductivity of filamentary exclusions of nonstochiometric UPt_3. Furthermore, the discontinuity in the specific heat at T_c was only 30% of the value predicted by the BCS theory and the temperatures attained in this experiment were not low enough to cover the complete superconducting transition.

In this Rapid Communication we demonstrate bulk superconductivity by measuring the Meissner effect by means of magnetization and dc susceptibility experiments. We have studied a sample with a superconducting transition temperature $T_c = 490$ mK and found that the superconducting volume fraction in the Meissner state lies in-between 30% and 80% at 350 mK. We have also measured the temperature dependence of the critical fields B_{c1} and B_{c2} and obtained values for some of the parameters characterizing the superconducting state of this heavy-fermion superconductor.[5]

A polycrystalline UPt_3 sample was prepared by arc melting the appropriate amounts of pure U and Pt in a titanium gettered atmosphere. After casting of the melt into a water-cooled crucible, a cylindrical sample was obtained, with a diameter of 4.8 mm and a length of 4.5 mm. Then it was annealed for 48 h at 900 °C. Also a powdered sample was made, which was annealed for 48 h at 800 °C. Ultrapure Cd ($T_c = 0.52$ K) and Sn ($T_c = 3.72$ K) cylinders of the same dimensions served as reference samples.

All measurements were performed in a continuous cycling ^3He cryostat spanning temperatures from 0.3 to 10 K. The coil system consisted of superconducting primary coils, each having two secondary pick-up coils of copper wire. The temperature was measured with a calibrated Ge resistor and controlled within 1 mK by a PID temperature controller. A magnetic field up to 1 T could be applied by means of a superconducting magnet. The magnetoresistance of the Ge resistor resulted in temperature deviations of at most 2 mK.

The ac susceptibility χ_{ac} was measured by means of a standard mutual inductance technique, using a frequency of 10.9 Hz and a driving field of 0.05 mT. We measured the dc susceptibility χ_{dc} by recording the induced voltage V_{ind} of the pick-up coils while ramping the magnetic field at a typical rate of 0.2 mT/s. The signals are calibrated by comparing the results with those of the Cd and Sn reference samples. In order to avoid flux pinning effects, the sample was heated above T_c and cooled in zero field before ramping the magnetic field. The magnetization can be obtained by numerical or analog integration. Additionally the magnetization was measured directly by means of a flux transformer method, similar to that described by Andres and Wernick.[7] Here the primary coil of 30 turns of superconducting NbTi wire was wound directly on the UPt_3 cylinder.

In Fig. 1 we show a typical trace of the dc susceptibility of the UPt_3 cylinder versus the magnetic field. This figure clearly demonstrates a nearly total Meissner effect and also shows that the virgin curve differs considerably from the flux pinned state. Another method put forward to measure the superconducting volume fraction[8] is to cool the sample in a constant dc magnetic field through T_c and measure the flux expulsion $\Delta\Phi$. The results are presented in Fig. 2 together with the virgin magnetization curve. Here, the initial slopes at $H = 0$ yield the superconducting volume fractions. Figure 3 shows these fractions as obtained by both methods plotted as a function of temperature. The two methods give an upper and lower limit to the superconducting volume fraction as will be discussed below.

In Fig. 4 we plot the magnetization of UPt_3 at 352 mK. This curve was obtained by the flux transformer method. The curve is rounded at B_{c1} probably because of demagne-

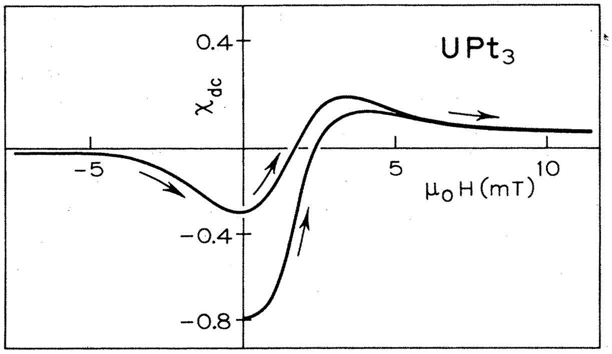

FIG. 1. dc susceptibility (normalized with respect to Cd and Sn) vs magnetic field of UPt$_3$ at 353 mK ($t = T/T_c = 0.73$). The lower curve is the virgin curve. For decreasing field the curve is a mirror image with respect to the ordinate.

tizing effects. The anomaly at B_{c2} cannot be seen in this plot because the Ginzburg-Landau parameter κ is extremely high and thus B_{c2} is much larger than B_{c1}. The dc susceptibility and magnetization ascertained that $B_{c1} = 2.2$ mT at 353 mK and from the ac susceptibility we found that $B_{c2} = 0.6$ T at this temperature.[5] From $B_{c2}/B_{c1} \approx 2\kappa^2/\ln\kappa$ one calculates that $\kappa \approx 20$.

Figure 5 shows the temperature dependence of B_{c1} and B_{c2}. We have taken B_{c1} as the field where $\chi_{dc} = 0$, thus, where $M = M(H)$ has a maximum. We derived B_{c2} from ac susceptibility measurements, where $T_c(B_{c2})$ was defined as the 50% point of the superconducting transition. We failed to observe the anomaly at B_{c2} in $\chi_{dc}(H)$ and $M(H)$. This anomaly is in the order of $(2\kappa^2)^{-1}$ and because $\kappa \approx 20$, the anomaly is smaller than our experimental accuracy.

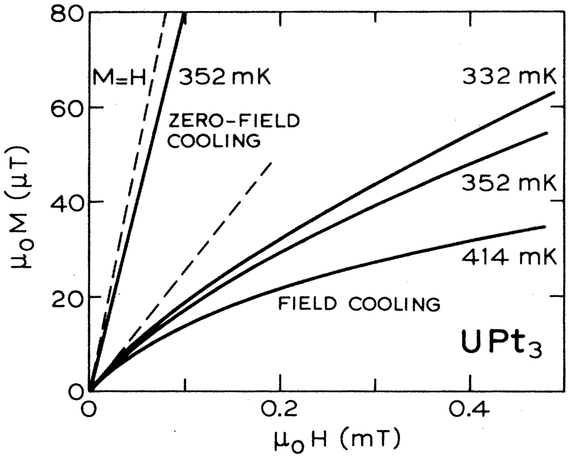

FIG. 2. Magnetization vs magnetic field of UPt$_3$ obtained by the two methods. Zero-field cooling denotes the virgin magnetization curve. The curves denoted by "field cooling" were drawn through the data points (not shown) obtained by measuring the flux expulsion at constant magnetic field as a function of temperature. The dashed lines represent the full Meissner effect ($M = H$) and the initial slope of the 332-mK curve.

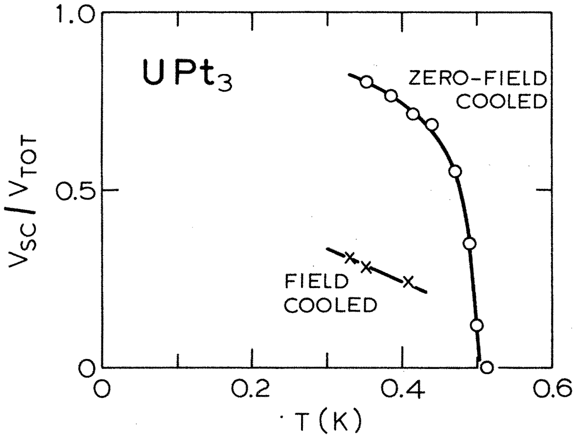

FIG. 3. Superconducting volume fraction of UPt$_3$ vs temperature as obtained by the field cooling (×) and the zero-field cooling (○) methods.

The superconducting transition of UPt$_3$ at 0.5 K in the presence of spin fluctuations is evidenced from dc resistivity, ac susceptibility, and specific-heat experiments.[3-6] The observed high value of $B_{c2}' = 4.4$ T/K, the enormous specific-heat coefficient $\gamma = 422$ mJ/mol K^2, and a residual resistivity of $\rho_0 \approx 3 \times 10^{-8}$ Ω m (Ref. 6) yield an effective mass for the pairing electrons of $m^* = 180 m_0$, thereby classifying this material as a heavy-fermion superconductor. This analysis,[5] similar to that for CeCu$_2$Si$_2$,[9] assumes a spherical Fermi surface and that UPt$_3$ is not a strong-coupling superconductor. Using the above values, a coherence length $\xi_0 = 2.0 \times 10^{-8}$ m, a mean free path $l = 3.6 \times 10^{-8}$ m, and the London penetration depth of $\lambda = 3.6 \times 10^{-7}$ m have been calculated. The Ginzburg-Landau parameter is found to be $\kappa = 23$ in agreement with the value obtained from the critical fields. The remaining question is whether this behavior represents a bulk property or if it arises from filamentary and/or surface effects. So far, specific-heat measurements could not exclude the latter possibility, because the jump in the specific heat was only 30% of the value expected from BCS theory.

The method used to measure the Meissner effect of CeCu$_2$Si$_2$ by cooling in field[8] gives only a lower limit for the superconducting volume fraction, because no correction can be made for flux trapping processes, which seem to be of

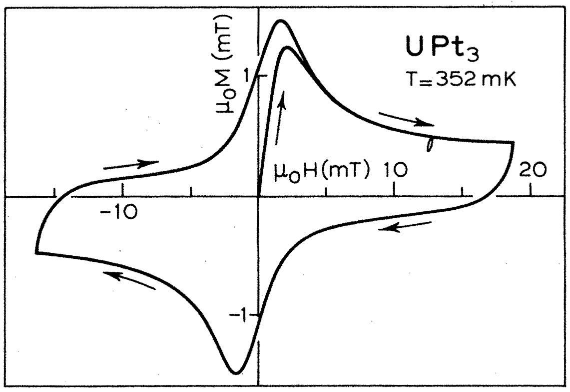

FIG. 4. Virgin magnetization curve and magnetization loop of UPt$_3$ at 352 mK. A minor hysteresis loop determined by χ_{ac} at 13 mT is also included.

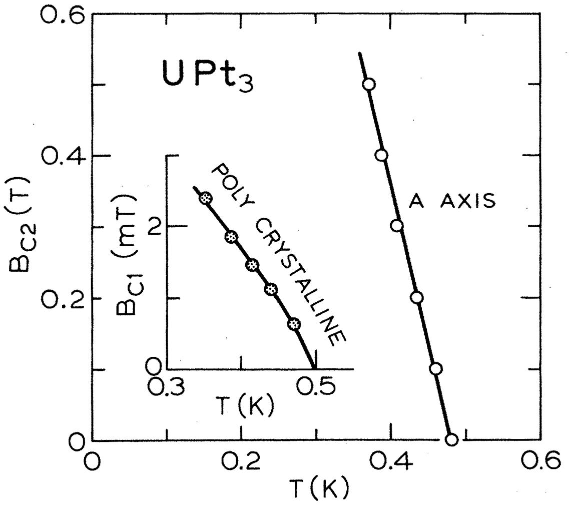

FIG. 5. Temperature dependence of the critical fields B_{c1} (inset) for a polycrystalline sample, and B_{c2} for a single crystal along the *a* axis. See also Ref. 5.

particular importance. It is well known that the superconducting transition propagates more quickly along the surface than into the bulk.[10] Furthermore, one can calculate that any superconducting surface in a small magnetic field forms an energy barrier impeding the passage of flux lines.[11] Thus, when cooling the sample below T_c, initially the surface becomes superconducting, forming this energy barrier. When the superconducting transition propagates farther into the bulk, the flux lines should be expelled from the inner part of the sample. However, if the pinning forces are large, the flux lines will be trapped at the surface barrier. Although the sample may be a good superconductor, it is well possible that very little flux is expelled. These pinning forces are especially large when the surface is oriented parallel to the magnetic field,[12] as was the case in the experiments on the CeCu$_2$Si$_2$ bars. The pinning forces can decrease by more than two orders of magnitude when the surface is not parallel to the magnetic field and this explains why the powdered samples will always show a more complete expulsion of flux than cylinders oriented parallel to the magnetic field.[8]

Besides the surface pinning, bulk pinning will also occur. Here, the flux lines are pinned at defects within the crystal lattice. The hysteresis of the magnetization loop (see Fig. 4) shows the results of these enormous pinning effects. Nevertheless, the expelled flux will still set a lower limit of the superconducting volume fraction.

In order to avoid such difficulties with flux pinning, we have cooled our samples below T_c in zero field and subsequently measured χ_{dc} or M, while sweeping the magnetic field. The initial slope of M vs H determines the superconducting volume fraction. However, in this experiment it is impossible to discern normal regions embedded in superconducting regions. Thus, this method will set the upper limit of the superconducting volume fraction. We think that for UPt$_3$ it gives a more reliable estimate because of the enormous pinning effects. ac susceptibility, which is also limited by flux pinning, will only allow a poor estimate of the superconducting volume fraction.

In addition, we have measured a UPt$_3$ powdered sample which gave similar results as the cylinder. The superconducting volume fraction at 357 mK, obtained from dc susceptibility, was 80% for the cylinder and 50% for the powder. This difference can be ascribed to a different transition temperature, $T_c(\text{bulk}) = 0.50$ K, $T_c(\text{powder}) = 0.45$ K, and a different transition width, $\Delta T_c(\text{bulk}) = 0.10$ K and $\Delta T_c(\text{powder}) > 0.10$ K. These quantities, T_c and ΔT_c, seem to be very dependent on the annealing or cold working procedures.[5]

We conclude from our measurements that the superconductivity of UPt$_3$ is a bulk property. The superconducting behavior of UPt$_3$ seems to be dominated by the 5*f* electrons of the U atoms. This can be inferred from the high effective mass of the pairing electrons. It has been argued[3] that the superconducting electron pairs would have a parallel alignment (*p* wave) because the superconducting transition occurs in the presence of spin fluctuations which favors a parallel alignment. Further experimental investigations, such as tunneling and Knight-shift measurements, and the effect of nonmagnetic impurities, are needed to verify this hypothesis.[13]

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