Interplay of magnetism and superconductivity in $CeCu_2Si_2$ under hydrostatic pressure

1998 J. Phys.: Condens. Matter 10 L749

(http://iopscience.iop.org/0953-8984/10/47/003)

J. Phys.: Condens. Matter **10** (1998) L749–L755. Printed in the UK PII: S0953-8984(98)98112-7

LETTER TO THE EDITOR

Interplay of magnetism and superconductivity in $CeCu_2Si_2$ under hydrostatic pressure

I Sheikin†‡, D Braithwaite†, J-P Brison§, A I Buzdin‖ and W Assmus¶

† Département de Recherche Fondamentale sur la Matière Condensée, SPSMS/LCP, CEA/Grenoble, 17 Rue des Martyrs, 38054 Grenoble Cedex 9, France
‡ Institute for High Pressure Physics, 142092, Troitsk, Moscow District, Russia
§ Centre de Recherches sur les Très Basses Températures, CNRS, BP166, 38042 Grenoble Cedex 9, France
‖ Centre de Physique Théorique et de Modélisation de Bordeaux, Laboratoire de Physique Theorique, Rue du Solarium, 33174 Gradignan Cedex, France
¶ Physikalisches Institut, Universität Frankfurt, D-60054 Frankfurt, Germany

Received 30 September 1998

Abstract. We present transport measurements under hydrostatic pressure, low temperature and magnetic field on a single crystal of the heavy fermion superconductor $CeCu_2Si_2$. The presumably magnetic A-phase is shown to collapse rapidly with pressure, and to almost disappear at 3.2 kbar. We also present measurements of the upper critical field $H_{c2}(T)$. We find evidence for a relation between the A-phase and the presence of a maximum on the temperature dependence of the H_{c2}. The magnetic susceptibility deduced from the analysis of the variation of $H_{c2}(T)$, taking into account the exchange interaction, shows a sharp variation at the A-phase boundary obtained from resistivity measurements in the normal state. Our analysis shows that the sign of the exchange integral should be negative, thus superconductivity is enhanced by the paramagnetic susceptibility as in the Jaccarino–Peter effect.

The interplay of magnetism and superconductivity is at present one of the fascinating topics in the physics of heavy fermion systems. In addition to several uranium-based superconductors (UPt_3, URu_2Si_2, UPd_2Al_3) which exhibit well defined antiferromagnetic order, $CeCu_2Si_2$, which lies close to a magnetic instability, is another good candidate for the investigation of this phenomenon. Since the discovery of heavy fermion superconductivity in $CeCu_2Si_2$ [1], this compound has been the object of many experimental and theoretical studies. It is now known that in the low-temperature B–T phase diagram, the superconducting (SC) phase is embedded in another phase (labelled A-phase) as shown in figure 1. In zero magnetic field, the transition into the A-phase occurs below the temperature $T_A \sim 0.7$ K, which is only slightly higher than the superconducting critical temperature T_c. When a magnetic field is applied, the behaviours of the two phases are quite different: the upper critical field, H_{c2}, of the superconducting phase is about 2 T, whereas the A-phase exists up to much higher fields, exceeding 7 T when the temperature approaches zero for $H \parallel a$ [2]. However, the low-temperature phase diagram of $CeCu_2Si_2$ is extremely sensitive to small differences in stoichiometry [3]. Samples can be of the S-type (SC with no signature of the A-phase), A/S-type (coexistence of superconducting and A-phases) or A-type (only A-phase). The A-phase was first detected as an anomaly in magnetoresistance measurements [4] and was shown to be of magnetic origin by NMR [5] and muon spin relaxation (μSR) [6] measurements. However, the type of magnetic state remains a mystery, all attempts at direct observation of the magnetic structure by neutron diffraction having failed so far. For

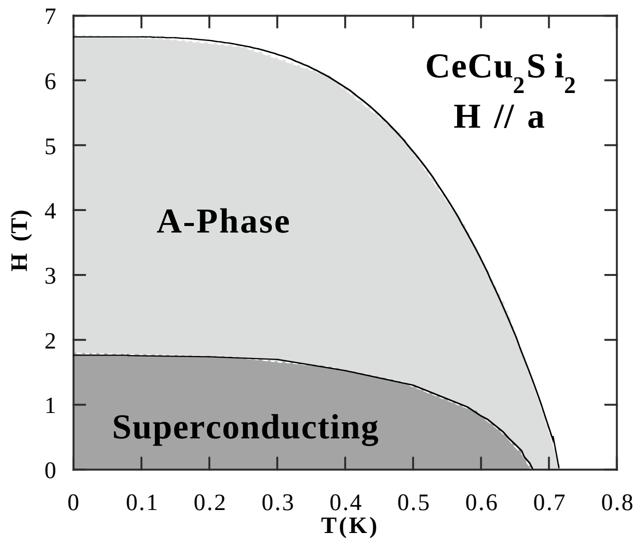

Figure 1. Generic diagram of the superconducting and A-phases in $CeCu_2Si_2$ with the field applied in the (a, b) plane.

example, a recent neutron elastic scattering experiment on a single crystal failed to detect any long-range order in the $(h, 0, l)$ plane [7]. Recent μSR results [8, 9] suggest that spin density wave or spin glass type ordering are most likely. But even the microscopic signatures of the appearance of the A-phase are obscure: new NQR experiments on cerium-depleted samples show that the A-phase may exist above the temperature where a clear broadening occurs in the Cu NQR or NMR signal [10]. Several high-pressure studies have been performed on $CeCu_2Si_2$, revealing that the pressure dependence of T_c exhibits a sharp increase from about 0.7 K to over 2 K above 16 kbar [11, 12], but remains almost constant up to this pressure. Hence relatively little attention was paid to the low-pressure range and in particular no observations of the A-phase under pressure were reported until a recent study showed that it is completely suppressed at 7 kbar [13]. This study still provided little information about the evolution of the B–T phase diagram of the A-phase under pressure. It is also known that pressure can induce superconductivity in A-type samples which are non-superconducting at ambient pressure [14]. In the present study, transport measurements have been performed in magnetic field, in order to determine precisely how the A-phase is suppressed with hydrostatic pressure. We have also measured the upper critical field to look for evidence of interplay between the magnetic and SC phases. We will not solve the problem of whether the A-phase is a real magnetic phase transition here, but we will stress that it has a clear magnetic origin with feedback effects on the temperature variation of the upper critical field. By the application of pressure, T_A decreases and collapses near 4 kbar. As for antiferromagnetic superconductors, the interplay between superconductivity and magnetism appears clearly when the magnetic ordering temperature is lower than the superconducting critical temperature.

We have carried out transport measurements on a single crystal of $CeCu_2Si_2$ in a piston–cylinder type pressure cell with a liquid pressure transmitting medium [15]. The sample was a single crystal of the A/S-type which was found to display a strong anomaly in the resistivity corresponding to the A-phase boundary at ambient pressure. The cell was placed in a dilution cryostat with a superconducting magnet to apply fields up to 8 T parallel to

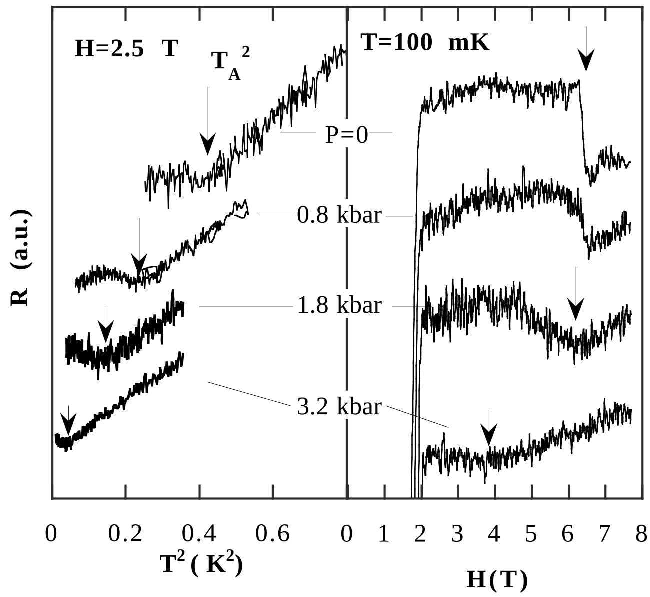

Figure 2. Examples of field sweeps (right) where the A-phase boundary appears as a sharp drop in resistance at low pressure or as a minimum in resistance at higher pressure, and temperature sweeps (left) where the A-phase boundary is determined by the point of deviation from a T^2 law. In both plots curves correspond to 0, 0.8, 1.8 and 3.2 kbar and are successively shifted downwards.

the cell axis, which corresponds to the *a* axis of the sample. The sample resistance was measured by a standard AC technique with lock-in detection. The measuring current of 30 μA at a frequency of 17 Hz was applied along the *c* axis of the sample. The pressure was calibrated at low temperature by measuring the superconducting critical temperature of tin. At ambient pressure the sample showed a superconducting transition with T_c of 0.68 K and transition width of 25 mK (10–90%), the residual resistivity was 7.8 $\mu\Omega$cm. These results are among the best reported in the literature and confirm the high quality of the sample. To determine the upper critical field and the A-phase diagram, we performed temperature and field sweeps as shown in figure 2. In the field sweeps at low pressure, the A-phase boundary appears as a sharp drop of about 15% of the magnetoresistance [4]. At higher pressures the feature becomes broader, changing finally to a rather smooth minimum, and transition points were determined by the minimum of the magnetoresistance. On the temperature sweeps the A-phase boundary is marked by a change of regime: below T_A, a deviation from the T^2 law occurs [16]. At intermediate temperatures and fields both types of measurement were performed and the criteria used produced consistent results. H_{c2} was determined by the superconducting transition midpoint. Figure 3 shows the evolution of the A-phase under pressure. We have found that the A-phase disappears very rapidly with pressure. It appears that the initial effect is mainly to depress the temperature of the A-phase boundary at low fields, while the critical field of the A-phase at low temperature is hardly affected up to 1.7 kbar. At a pressure of 3.2 kbar we observed only tiny signs of the A-phase. Contrary to the lower pressure results, the A-phase boundary obtained at 3.2 kbar lies at a low field even at low temperature. This suggests that the A-phase should completely disappear at a pressure only slightly higher than 3.2 kbar. The high-field part of

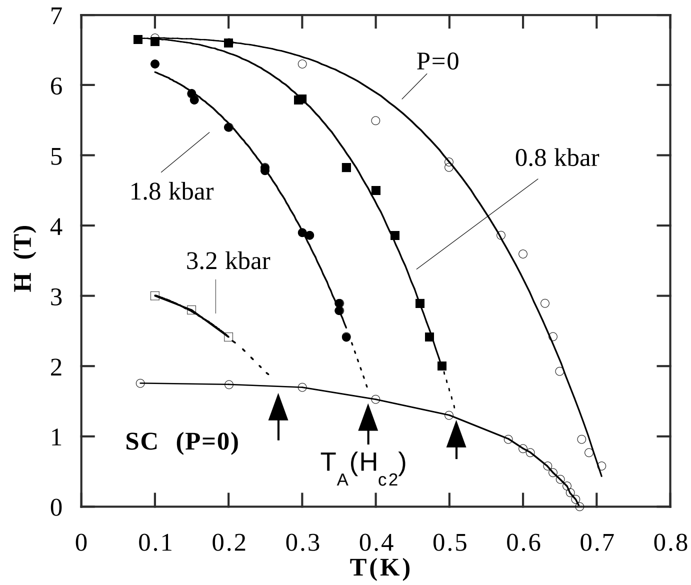

Figure 3. Evolution of the A-phase diagram under pressure shown with the superconducting phase at zero pressure. Dashed lines show the extrapolation of a fit of the form $(1 - T/T_c)^{\alpha}$, in order to estimate the temperature of the intersection between the superconducting and A-phase boundaries, $T_A(H_{c2})$, indicated by arrows.

the temperature dependence of the upper critical field H_{c2} renormalized by T_c for different pressures is shown in figure 4. The low field points, i.e., close to T_c (not shown) are identical for all pressures up to 3.2 kbar within the experimental resolution. However, the low-temperature parts of the $H_{c2}(T)$ curves are found to vary significantly with pressure. The most striking result is the appearance of a rather broad maximum on the temperature dependences of H_{c2} when the A-phase boundary is shifted to a lower temperature. This maximum becomes more pronounced at higher pressure, but, like the A-phase, disappears at pressures above 3.2 kbar. This anomaly can be also seen in the temperature sweeps at a magnetic field slightly lower than the maximum value of H_{c2} (figure 4 inset) where re-entrant behaviour is found. Such a maximum on H_{c2} in this compound has already been observed in some samples at ambient pressure [17], although less pronounced than here. Several possible explanations were proposed, but without a final conclusion. At that time the existence of the A-phase was unknown, however, our new results suggest that the origin of this maximum of H_{c2} is due to the interplay between the magnetic A-phase and superconductivity. Several points support this idea: we have not found this behaviour either at higher pressure, when the A-phase does not exist, or at zero pressure, when the temperature of the transition to A-phase at zero field is slightly higher than T_c. Moreover, when the anomaly occurs, it is in the temperature region of $T_A(H_{c2})$, the temperature where the A-phase boundary crosses the upper critical field (see the arrows in figure 3). The influence of the appearance of a magnetic phase on H_{c2} should come from a change in the (paramagnetic) Pauli limitation. For a quantitative estimate of this change through a realistic model of H_{c2}, a proper account of the mean free path [18] and strong coupling parameters [19, 20] would be necessary. Because the microscopic nature of the A-phase is still unknown, this is, at least at present, out of reach. However, in order to test qualitatively how the anomalous behaviour of H_{c2} can arise from a modification of the Pauli limitation

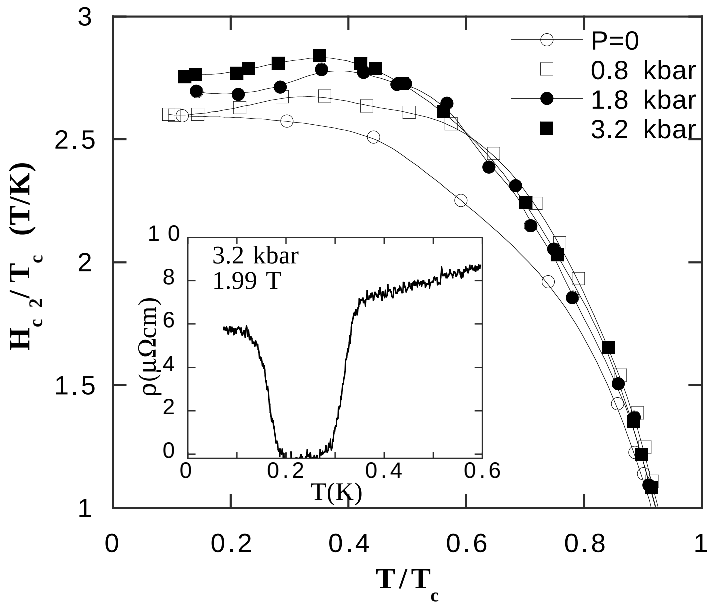

Figure 4. Temperature dependence of the upper critical field H_{c2} renormalized by T_c for different pressures. The low field points, i.e. close to T_c (not shown) are identical for all pressures within the experimental resolution. The inset shows a temperature sweep at a field slightly below the maximum value of H_{c2} exhibiting re-entrant behaviour.

by the A-phase, a simple model of H_{c2} should be sufficient. To this end we have analysed the temperature dependences of H_{c2} assuming weak coupling and clean limit. In this case, the Pauli limitation is entirely controlled by the value of the gyromagnetic ratio, g, of the conduction electrons ($g = 2$ for free electrons). With the same hypothesis, the orbital limitation is determined by a mean Fermi velocity which can be deduced from the value of the slope of H_{c2} at T_c, $(\mathrm{d}H_{c2}/\mathrm{d}T_c)_{T=T_c}$. We mentioned above that, within the experimental error bars, this slope is constant at about 25 T/K in the pressure range considered here. So g is the only parameter which can change with pressure, and inside the A-phase. Let us note that, in the clean limit scenario, the calculation of the paramagnetic effect on H_{c2} must include the appearance of the Fulde–Ferrel–Larkin–Ovchinnikov state [21], which gives realistic though slightly overestimated values of H_{c2} at low temperature, owing to the fact that $CeCu_2Si_2$ is probably in an intermediate regime between the clean and dirty limits [18]. In a simple description of the magnetism of $CeCu_2Si_2$ arising from (paramagnetic) conduction electrons and magnetic ions, the Pauli limitation of H_{c2} is controlled by the pair-breaking effect of the external field, $g\mu_B B$, and by an additional term due to the internal exchange field, $\chi(T)JB$, where J is the exchange integral between the conduction electrons and the magnetic moments, and $\chi(T)$ is the uniform susceptibility of the magnetic ions. We can therefore write the effect of an external field as an effective temperature dependent gyromagnetic factor $g_{eff}(T)\mu_B B$ where

$$g_{eff}(T) = g + \chi(T)J/\mu_B. \qquad (1)$$

We then fit the $H_{c2}(T)$ curve allowing g_{eff} to vary with temperature. As neither J nor g should vary with temperature, $g_{eff}(T)$ reflects the temperature dependence of the susceptibility. This variation of $g_{eff}(T)$ is shown in figure 5. g_{eff} is practically constant at

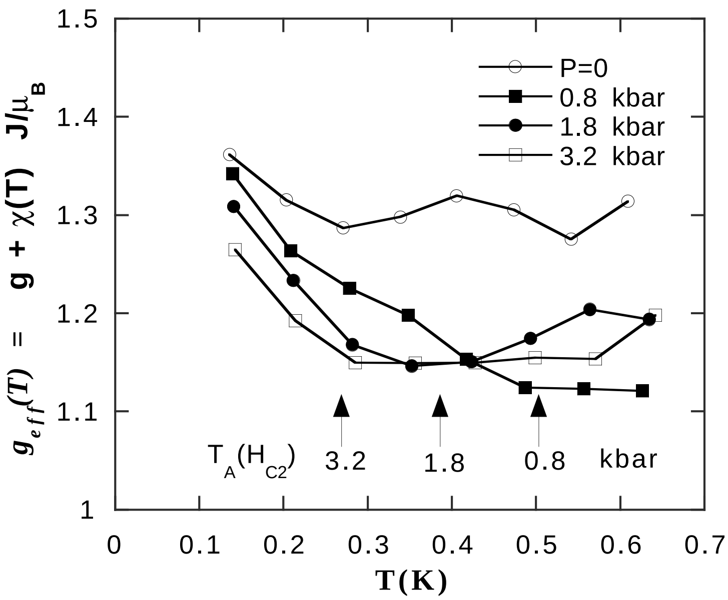

Figure 5. Temperature dependence of the effective g factor obtained from the fit of the $H_{c2}(T)$ curves. Note that for each pressure the upturn of $g(T)$ occurs at a temperature close to the intersection of the A-phase and superconducting phase boundaries indicated by the arrows.

ambient pressure, but at higher pressures shows a clear upturn. The most significant result is that the temperature where this upturn occurs coincides very well with the temperature of the intersection of the A-phase boundary with H_{c2} (indicated by arrows). A second, weaker upturn can be seen at lower temperatures on the ambient pressure curve, but this probably arises from the shape of the theoretical curve of H_{c2} which tends to overestimate H_{c2} at low temperatures. In a more realistic model g_{eff} is expected to remain constant at ambient pressure and the increase would probably saturate when T decreases towards zero at the higher pressures.

The fact that this upturn of g_{eff}, found from the shape of H_{c2}, occurs at almost exactly the same temperature as the transition to the A-phase deduced from magnetoresistance in the normal phase, is in itself strong evidence towards our suggestion that the maximum of H_{c2} is due to the interaction with magnetism. However, if, as is most likely, the A-phase corresponds to the onset of an order with antiferromagnetic correlations, one would expect the susceptibility to decrease, or at least remain constant below the ordering temperature, whereas g_{eff} is found to increase on cooling. This is not necessarily contradictory as the relation between χ and g_{eff} depends on the sign of J. If J is negative, the two terms in equation (1) compensate each other, and the paramagnetic susceptibility enhances H_{c2} in the presence of an external field, according to the Jaccarino–Peter effect [22]. Then if $\chi(T)$ has a maximum below T_c, $H_{c2}(T)$ will also exhibit a maximum as found here.

Several studies have shown that the superconducting and A-phases tend to repel each other [2, 8, 9]. Here we have assumed their coexistence at least in a region close to the H_{c2} boundary. We stress that this is not contradictory with an expulsion of the A-phase when moving deeper into the superconducting phase. However, our results support the idea that at the onset of superconductivity the two phases coexist homogeneously on a microscopic level, and not, as sometimes suggested, in separate macroscopic regions. The critical field of the A-phase, H_A, increases on cooling and decreases with pressure. This supports the idea of a real magnetic long-range ordered state for the A-phase.

This kind of measurement may provide a unique way of obtaining information on very weak variations of the magnetic susceptibility, as its effect on the conduction electrons can be greatly amplified by the exchange integral. Our results show that interplay of magnetism and superconductivity must probably be taken into account to understand the upper critical field of $CuCu_2Si_2$ at low temperature and ambient pressure.

We thank J Flouquet for his constant encouragement and helpful discussions. One of us (IS) is grateful to Centre International des Etudiants et Stagiaires for financial support.

References

[1] Steglich F, Aarts J, Bredl C D, Lieke W, Meschede D, Franz W and Schäfer H 1979 *Phys. Rev. Lett.* **43** 1892
[2] Bruls G, Wolf B, Finsterbusch D, Thalmeier P, Kouroudis I, Sun W, Assmus W, Lüthi B, Lang M, Gloos K, Steglich F and Modler R 1994 *Phys. Rev. Lett.* **72** 1754
[3] Steglich F, Gegenwart P, Geibel C, Helfrich R, Hellmann P, Lang M, Link A, Modler R, Sparn G, Büttgen N and Loidl A 1996 *Physica* B **223&224** 1
[4] Rauchschwalbe U, Steglich F, de Visser A and Asayama K 1987 *J. Magn. Magn. Mater.* **63&64** 347
[5] Nakamura H, Kitaoka Y, Yamada H and Asayama K 1988 *J. Magn. Magn. Mater.* **76&77** 517
[6] Uemura Y J, Kossler W J, Yu X H, Schone H E, Kempton J R, Stronach C E, Barth S, Gygax F N, Hitti B, Schenck A, Baines C, Lankford W F, Onuki Y and Komatsubara T 1989 *Phys. Rev.* B **39** 4726
[7] Fåk B 1998 private communication
[8] Feyerherm R, Amato A, Geibel C, Gygax F N, Hellman P, Heffner R H, MacLaughlin D E, Müller-Reisener R, Nieuwenhuys G J, Schenck A and Steglich F 1995 *Physica* B **206&207** 596
[9] Luke G M, Keren A, Kojima K, Le L P, Sternieb B J, Wu W D, Uemura Y J, Onuki Y and Komatsubara T 1994 *Phys. Rev. Lett.* **73** 1853
[10] Ishida K, Tabuchi K, Kashima K, Kawasaki Y, Kitaoka Y, Asayama K, Geibel C and Steglich F 1998 *Proc. SCES 98 (Paris, 1998) Physica* B at press
[11] Bellarbi B, Benoit A, Jaccard D, Mignot J-M and Braun H F 1984 *Phys. Rev.* B **30** 1182
[12] Thomas F, Ayache C, Fomine I A, Thomasson J and Geibel C 1996 *J. Phys.: Condens. Matter* **8** 51
[13] Steglich F, Buschinger B, Gegenwart P, Lohmann M, Helfrich R, Langhammer C, Hellmann P, Donnevert L, Thomas S, Link A, Geibel C, Lang M, Sparn G and Assmus W 1996 *J. Phys.: Condens. Matter* **8** 9909
[14] Link A, Laube F, Donnevert L, Sparn G, Geibel C and Steglich F 1998 *J. Magn. Magn. Mater.* **177–181** 401
[15] Itskevich E S, Voronovskii A N, Gavrilov A F and Sukhaparov V A 1967 *Cryogenics* 359
[16] Gegenwart P, Lohmann M, Lang M, Helfrich R, Langhammer C, Köppen M, Geibel C, Steglich F and Assmus W 1997 *Physica* B **230–232** 572
[17] Assmus W, Herrmann M, Rauchschwalbe U, Riegel S, Lieke W, Spille H, Horn S, Weber G, Steglich F and Cordier G 1984 *Phys. Rev. Lett* **52** 469
[18] Rauchschwalbe U, Ahlheim U, Bredl C D, Mayer H M and Steglich F 1987 *J. Magn. Magn. Mater.* **63&64** 447
[19] Vargoz E, Jaccard D, Genoud J Y and Brison J-P 1998 *Solid State Commun.* **106** 631
[20] Thomasson J, Okayama Y, Sheikin I, Brison J-P and Braithwaite D 1998 *Solid State Commun.* **106** 637
[21] Saint James D, Sarma D and Thomas E J 1969 *Type II Superconductivity* (New York: Pergamon)
[22] Jaccarino V and Peter M 1962 *Phys. Rev. Lett.* **9** 290