First-order transition between weak and strong pinning in clean superconductors with enhanced spin susceptibility

R. Modler, Philipp Gegenwart, M. Lang, M. Deppe, M. Weiden, T. Lühmann, C. Geibel, F. Steglich, C. Paulsen, J. L. Tholence, N. Sato, T. Komatsubara, Y. Ōnuki, M. Tachiki, S. Takahashi

Angaben zur Veröffentlichung / Publication details:

Modler, R., Philipp Gegenwart, M. Lang, M. Deppe, M. Weiden, T. Lühmann, C. Geibel, et al. 1996. "First-order transition between weak and strong pinning in clean superconductors with enhanced spin susceptibility." *Physical Review Letters* 76 (8): 1292–95. https://doi.org/10.1103/physrevlett.76.1292.

First-Order Transition between Weak and Strong Pinning in Clean Superconductors with Enhanced Spin Susceptibility

R. Modler, P. Gegenwart, M. Lang, M. Deppe, M. Weiden, T. Lühmann, C. Geibel, and F. Steglich
Institut für Festkörperphysik, TH Darmstadt, D-64289 Darmstadt, Germany

C. Paulsen and J. L. Tholence
CRTB, CNRS, F-38042 Grenoble Cedex 9, France

N. Sato and T. Komatsubara
Physics Department, Tohoku University, Sendai 980, Japan

Y. Ōnuki
Faculty of Science, Osaka University, Toyonaka, Osaka 560, Japan

M. Tachiki and S. Takahashi
Institute for Materials Research, Tohoku University, Sendai 980, Japan
(Received 12 September 1995)

Dilatometric and magnetic experiments on high-quality single crystals of the superconducting compounds UPd_2Al_3 and $CeRu_2$, which have enhanced spin susceptibilities, demonstrate a first-order transition between weak and strong pinning at $T < 0.9T_c$ ($H > 10$ kOe). This is compatible with a staggered order parameter due to the formation of a "Fulde-Ferrell-Larkin-Ovchinnikov" state.

PACS numbers: 65.70.+y, 74.60.Ge, 74.70.Tx

Staggered superconducting order parameters have attracted much interest in connection with the internal multilayer structure of high-T_c cuprates [1] and, more recently, with "odd-frequency-pairing" superconductors (SC) [2]. More than three decades ago Fulde and Ferrell (FF) [3] as well as Larkin and Ovchinnikov (LO) [4] proposed that, in a very clean and strongly Pauli-limited singlet SC, a partially polarized superconducting state should form at high magnetic fields. This spatially modulated "FFLO" state should raise the upper critical field $H_{c2}(T)$ at which the superconducting-normal transition takes place. While the search for the FFLO state among classical SC was unsuccessful, there have been recent claims for its existence in certain heavy-fermion (HF) SC. For example, the record value $H_{c2}(0) \simeq 140$ kOe established for a high-quality single crystal of cubic UBe_{13} ($T_c = 0.94$ K) was ascribed to the formation of the FFLO state [5]. Furthermore, Modler *et al.* [6] discovered unique anomalies in the sample length, $l(T,H)$, below $H_{c2}(T)$ and $T \lesssim 1.5$ K for the hexagonal compound UPd_2Al_3 which exhibits [7] a coexistence of HF superconductivity ($T_c \lesssim 2$ K) and strong antiferromagnetic order ($T_N = 14.5$ K). Emphasizing that UPd_2Al_3 fully meets the following strict requirements, i.e., is (1) a very clean SC (electronic mean free path $\ell_0 \simeq 700$ Å $\gg$ superconducting coherence length $\xi_0 \simeq 85$ Å [7]) and (2) subject to strong Pauli limiting ($H_{c2}(0) \simeq H_P$[in 10 kOe] $= 1.84T_c$[in K]), Gloos *et al.* [8] speculated that these $l(T,H)$ anomalies mark a first-order transition from the mixed state to the modulated FFLO state. New research activities were initiated by the reports of Huxley *et al.* [9] and Yagasaki *et al.* [10] that the nonmagnetic ("strongly intermediate-valent") cubic Laves-phase SC $CeRu_2$ [11] ($T_c = 6.1$ K) exhibits striking magnetic anomalies below $H_{c2}(T)$ and $T < 5.5$ K [12], leading to an H-T phase diagram similar to that of Fig. 1(b). Subsequently Goshima *et al.* [13] showed that in $CeRu_2$, along with these magnetic features, anomalies show up in both the magnetocaloric effect and the elastic constants which appear to be related to the dilatometric ones found in UPd_2Al_3 [6,8]. Since the $CeRu_2$ samples studied in Refs. [9,10,13] were clean

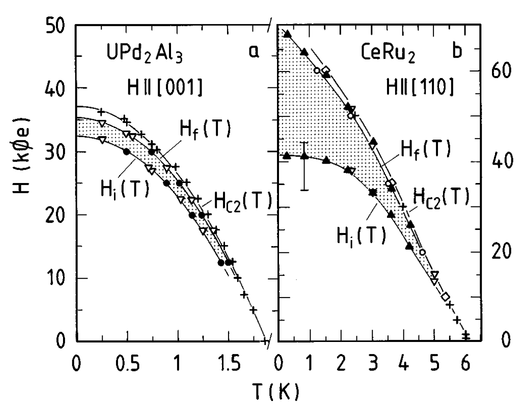

FIG. 1. *H-T* phase diagrams of (a) UPd_2Al_3 and (b) $CeRu_2$. Anomalies in *M(H,T)* (▽, Fig. 2), $\Delta l(H)$ (▲, Fig. 3), $\Delta l(T)$ [○, Fig. 4(a)] and $\chi_{ac}(T)$ (●, Fig. 5) define onset [$H_i(T)$] and offset [$H_f(T)$] of irreversibilities, see text. Upper critical field $H_{c2}(T)$ defined by +[$\chi_{dc}(H,T)$] and ◇ [$\alpha(T)$, Fig. 4(b)].

 0031-9007/96/76(8)/1292(4)$06.00

SC ($\ell_0 \gg \xi_0 = 61$ Å [9]), a naive interpretation in terms of the ordinary, i.e., disorder-induced, "peak effect" in the critical current density often observed for dirty type-II SC [14] can be safely discarded.

In this Letter we report a comparative study on high-quality single crystals of UPd_2Al_3 and $CeRu_2$, based on measurements of the magnetostriction [$\Delta l(H, T = \text{const})$], thermal dilatation [$\Delta l(H, T = \text{const})$], dc magnetization [$M(H, T = \text{const}), M(T, H = \text{const})$], and ac susceptibility [$\chi_{ac}(H, T = \text{const}), \chi_{ac}(T, H = \text{const})$]. Our salient results are as follows: (i) As far as the anomalies mentioned in the introductory paragraph are concerned, these otherwise very different compounds appear phenomenologically related to each other; cf. their H-T phase diagrams in Figs. 1(a) and 1(b). (ii) We are able to infer the main criteria for these anomalies to occur, i.e., large values not only of the electronic mean free path, but also of the spin susceptibility. (iii) We can explain quite naturally the first-order line $H_i(T)$ in Figs. 1(a) and 1(b) that separates weak ($H < H_i$) from strong ($H > H_i$) pinning. This marks a transition from the mixed state, the usual Abrikosov vortex lattice, into a "generalized FFLO" state, the latter exhibiting nodal planes perpendicular to the vortices [15].

For our present investigations we used the same UPd_2Al_3 single crystal ($T_c = 1.85$ K) studied in [7,8]. As before, we obtained similar results for the H field applied along and perpendicular to the hexagonal c axis. A $CeRu_{1.7}$ ingot was prepared from the starting materials (4N Ce, 3N Ru) by arc melting. The ingot was subsequently zone melted in a cold crucible placed under vacuum in an induction furnace. Parts of the resulting $CeRu_2$ single crystal were used for de Haas–van Alphen measurements which yielded an electronic mean free path $\ell_0 \simeq 1300$ Å [16]. The thermal-dilatation and magnetostriction measurements were carried out using a parallel-plate capacitance technique, with a dilatation cell manufactured from 5N Ag. The magnetization measurements on $CeRu_2$ were done at THD with the aid of a SQUID system of small field inhomogeneity ($\Delta H/H \leq 2 \times 10^{-4}$), those on UPd_2Al_3 were done using the low-T, high-H SQUID facilities at CRTBT ($\Delta H/H \lesssim 4 \times 10^{-5}$). Both dc-magnetization experiments were quasistatic extraction measurements. The latter setup also rendered the possibility to measure the ac susceptibility, i.e., by using a high-resolution SQUID sensor (longitudinal ac field $\simeq$1 Oe and frequency $\simeq 10^2$ Hz).

The isothermal magnetization curves of UPd_2Al_3 ($T = 0.25$ K) and $CeRu_2$ ($T = 3$ K) display in both cases a sharp peak at H_{c1}, and, in addition [cf. Figs. 2(a) and 2(b)], an almost reversible $M(H)$ dependence for $15 \lesssim H \lesssim 30$ kOe, a hysteresis "loop" between H_i and $H_f < H_{c2}$, and reversible behavior at higher fields. The $CeRu_2$ data [Fig. 2(b)] clearly indicate a first-order transition from weak to strong pinning. This is corroborated

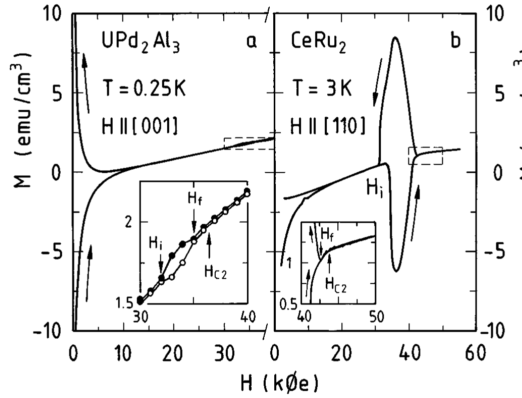

FIG. 2. Isothermal dc magnetization M vs field H of (a) UPd_2Al_3 and (b) $CeRu_2$. Insets: H_i and H_f indicate the onset and offset fields of the hysteresis loop. H_{c2}, the upper critical field, is defined by the change in slope of the high-field data. ○ and ● in the inset of (a) denote values of the hysteretic magnetization taken upon moving the sample up or down within the pickup coils of the magnetometer.

by our magnetostriction results [Fig. 3(a)] which demonstrate that at $H \geq H_i$ there is an enormous increase in the stress induced by the trapped vortices that act on the sample. As for UPd_2Al_3 [7,8], for $CeRu_2$ (i) the amplitude of the $\Delta l(H, T = \text{const})$ anomaly is found to be precipitously reduced upon warming, and (ii) the sample length $\Delta l(T, H = \text{const})$ relaxes slightly below the depinning temperature $T_f(H)$, hinting at a pronounced weakening of the pinning force [17]. For UPd_2Al_3, different signs for these quasidiscontinuous length changes were observed in different runs on the same sample; cf. Refs. [7,8]. The $CeRu_2$ data of Fig. 3(b) reveal that both the size and sign of this length jump depend on the field history, i.e., the way by which the magnetic field had been applied to the sample [Fig. 3(a)], following an initial zero-field cooling.

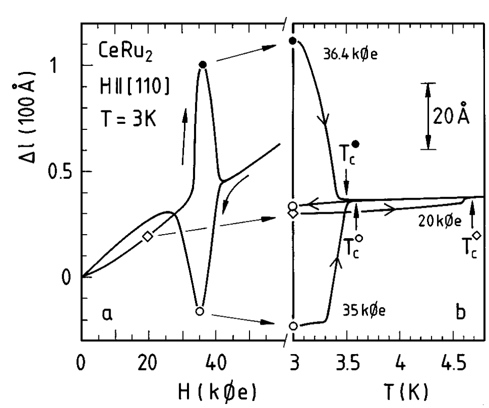

FIG. 3. $CeRu_2$: (a) Isothermal magnetostriction Δl vs H and (b) length change Δl vs T measured at different fields, starting from different points of the isothermal $\Delta l(H)$ curve displayed in (a). $\Delta l(T)$ data are shifted in order to coincide for the normal-state values. Field-cooled curve $\Delta l(T)$ is shown for $H = 35$ kOe only.

A compilation of the results from different techniques for the UPd_2Al_3 sample is shown in Fig. 4. These measurements were carried out at $H = 25$ kOe as a function of temperature. The change in $\Delta l(T)$ [Fig. 4(a)] occurs well below the superconducting transition temperature T_c as determined from the mean-field-type anomaly in the thermal-expansion coefficient $\alpha(T) = l^{-1}\partial l/\partial T$, measured by slowly cooling in constant field [Fig. 4(b)]. Figures 4(c) and 4(d) demonstrate that for $T_i < T < T_f$ the magnetization is irreversible, consistent with a pronounced negative peak in $\chi_{ac}(T)$ and indicating strong pinning. Comparison with Fig. 4(a) shows that the relaxation of the sample length takes place slightly above the temperature of this peak and slightly below the depinning temperature T_f. The pinning-related anomalies in $\Delta l(T,H)$ and $M(T,H)$ become negligibly small below $H \simeq 10$ kOe for both compounds, i.e., above $T \simeq 1.5$ K for UPd_2Al_3 and above $T \simeq 5.5$ K for $CeRu_2$. As seen in Fig. 5, the depths of the minima in the $\chi_{ac}(T, H = \text{const})$ curves [18] decrease linearly upon reducing the field down to 12.5 kOe. We ascribe the peaks below 10 kOe [Fig. 5(b)], which deviate from the straight line in Fig. 5(c), to demagnetizing fields near the edges and corners of our UPd_2Al_3 single crystal.

Turning now to an interpretation of our experimental results, we first wish to stress that both UPd_2Al_3 and $CeRu_2$ exhibit large low-T spin susceptibilities χ_S [3.2×10^{-5} and $(2.7 \pm 0.5) \times 10^{-5}$ emu/cm^3] [17]. Therefore, the quasiparticles inside the vortex cores gain a large Zeeman energy density which, close to H_{c2}, becomes almost identical to the superconducting condensation energy density $H_c^2/8\pi \simeq 0.25(\gamma_0/V_{\text{mole}})T_c^2$; here H_c is the thermodynamic critical field, γ_0 (125 and 29 mJ/K^2 mole for UPd_2Al_3 [17] and $CeRu_2$ [9]) the Sommerfeld coefficient of the normal state specific heat and V_{mole} (62.94 and 32.23 cm^3 mole^{-1}) the molar volume. Consequently, one estimates a very small self-energy of the vortex core, $E_{\text{core}} \simeq \pi\xi_0^2(H_c^2/8\pi - 0.5\chi_S h^2)$, h being the magnetic field induced at the vortex core by the shielding currents surrounding it. This explains [15] why H_{c1} (as $T \rightarrow 0$) is as low as 100 and 250 Oe for UPd_2Al_3 and $CeRu_2$, respectively, and why the pinning is that weak in a wide field range below H_i; see Figs. 2(a) and 2(b).

Including the large Zeeman term in the BCS Hamiltonian for a singlet SC with sufficiently weak spin-orbit scattering off impurities ($\ell_{so} > \ell_0 > \xi_0$, as realized in our samples), Tachiki *et al.* [15] obtained for the gap equation a solution that varies periodically along the direction of the H field. The wavelength of the oscillation Λ varies between $\simeq 40\xi_0$ at H_i and $\simeq 15\xi_0$ at H_{c2}, depending on the strength of the (dominating) paramagnetic relative to the orbital pair-breaking effect of the external field. In the new state, the order parameter exhibits a periodic array of nodal planes perpendicular to the vortices, at intervals Λ [19]. Apart from a much larger layer separation, this "generalized FFLO state" resembles the vortex state of layered cuprate SC in magnetic fields perpendicular to the CuO_2 planes.

As argued above, the pinning force on rigid vortices in such a clean SC with enhanced spin susceptibility is very weak. In the generalized FFLO state, however, we expect a significant change in the pinning capability of the SC. Here the occurrence of planar nodes of the order parameter at H_i leads to a segmentation of the vortices into pieces with a length of several times the coherence length. As a result, these vortex segments become flexible in a qualitatively similar way as the quasi-two-dimensional vortex disks ("pancakes") in cuprate SC. If the vortices are subject to weak disorder, e.g., as in the presence of point defects, we expect the individual vortex segments to accommodate more easily to the random

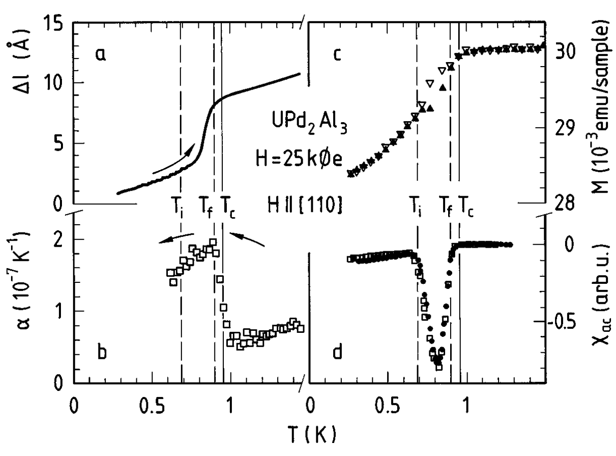

FIG. 4. Isofield results on UPd_2Al_3 indicate that the sample length (a) relaxes at $T \lesssim T_f < T_c$. (b) T_c is determined from the midpoint of the jump in the thermal-expansion coefficient $\alpha(T)$. The temperatures T_i and T_f mark the onset and the offset of the strong-pinning regime as observed in (c) *M(T)* and (d) $\chi_{ac}(T)$, respectively; full (open) symbols (c) have the same meaning as in Fig. 2(a), (d) denote measurements upon heating (cooling).

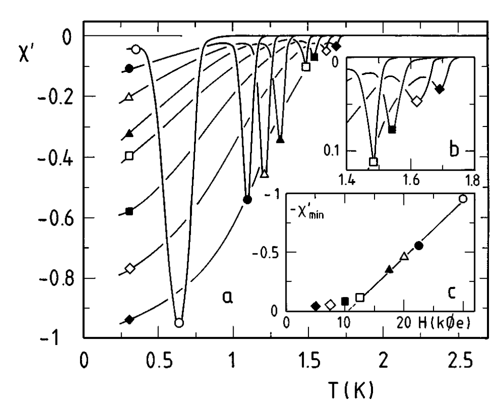

FIG. 5. (a),(b) real part of $\chi_{ac}(T, H = \text{const})$ for UPd_2Al_3 at different magnetic fields as indicated by (c) the symbols which mark the magnitude of the minimum vs the applied field.

pinning potential, thereby becoming efficiently pinned by the collective action of weak pinning centers [20].

Several points need clarification by future investigations: (1) Compared to the results of the original theory dealing with BCS superconductors [3,4], which restricts the FFLO state to $T < 0.56T_c$, we find a greatly enhanced existence range, $T < 0.9T_c$, for both UPd_2Al_3 and $CeRu_2$ [21]. (2) The existence range of the FFLO state on the field axis is, especially for $CeRu_2$, much wider than expected from Refs. [3,4] where spherical Fermi surfaces are assumed. However, both UPd_2Al_3 [22] and $CeRu_2$ [16] exhibit complex Fermi surfaces with disjunct portions. Therefore, an antiferromagnetic exchange interaction among electronic quasiparticles is highly probable [23], which should favor an expanded field range for the FFLO state, cf. Ref. [19].

In conclusion, for two SC that behave differently in many respects we have observed surprisingly similar anomalies in the magnetic and dilatometric properties, indicating highly unusual pinning properties: the antiferromagnetically ordered HF compound UPd_2Al_3 and the nonmagnetic, strongly intermediate-valent compound $CeRu_2$. We have presented arguments that, apart from a large spin-orbit mean free path and sufficiently weak orbital pair breaking, an enhanced spin susceptibility is a prerequisite for the formation of the generalized FFLO state. Owing to its generic nodal structure, the latter can explain naturally the observed first-order transition from weak to strong pinning. In order to observe the "anomalous peak effect" described in this Letter, a suitable weak random pinning potential appears to be required; cf. Ref. [17]. Future experiments, e.g., of scanning-vacuum-tunneling spectroscopy, should be addressed to directly observe the unique nodal structure of the staggered FFLO order parameter.

We thank P. Fulde, K. Gloos, M. B. Maple, and D. Rainer for stimulating conversations. This work was supported by the SFB 252 Darmstadt-Frankfurt-Mainz. R. M. gratefully acknowledges a fellowship by the Forschungszentrum Karlsruhe-*Technik und Umwelt.*

[1] M. Tachiki *et al.,* Z. Phys. B **80**, 161 (1990).
[2] P. Coleman, Phys. Rev. B **49**, 8955 (1994); Phys. Rev. Lett. **74**, 1653 (1995).
[3] P. Fulde and R. A. Ferrell, Phys. Rev. **135**, 550 (1964).
[4] A. I. Larkin and Yu. N. Ovchinnikov, Zh. Eksp. Teor. Fiz. **47**, 1136 (1964) [Sov. Phys. JETP **20**, 762 (1965)].
[5] F. Thomas *et al.,* J. Low Temp. Phys. (to be published).
[6] R. Modler *et al.,* Int. J. Mod. Phys. B **7**, 42 (1993).
[7] C. Geibel *et al.,* Z. Phys. B **84**, 1 (1991).
[8] K. Gloos *et al.,* Phys. Rev. Lett. **70**, 501 (1993).
[9] A. D. Huxley *et al.,* J. Phys. Condens. Matter **5**, 7709 (1993).
[10] K. Yagasaki *et al.,* J. Phys. Soc. Jpn. **62**, 3825 (1993).
[11] B. T. Matthias *et al.,* Phys. Rev. Lett. **1**, 449 (1958).
[12] See also S. B. Roy and B. R. Coles, J. Phys. Condens. Matter **6**, L663 (1994); H. Sugawara, *et al.,* Physica (Amsterdam) **206B-207B**, 196 (1995); M. B. Maple (private communication).
[13] H. Goshima *et al.,* Physica (Amsterdam) **206B-207B**, 193 (1995).
[14] A. M. Campbell and J. E. Evetts, Adv. Phys. **21**, 327 (1972).
[15] M. Tachiki *et al.,* Z. Phys. B (to be published).
[16] M. Hedo *et al.,* J. Phys. Soc. Jpn. (to be published).
[17] F. Steglich *et al.,* Physica (Amsterdam) C (to be published).
[18] See, also T. Lühmann *et al.,* Physica (Amsterdam) **235C-240C**, 2433 (1994); A. Ishiguro *et al.,* J. Phys. Soc. Jpn. **64**, 378 (1995).
[19] This state was already obtained by H. Burkhardt and D. Rainer [Ann. Phys. **3**, 181 (1994)] for quasi-two-dimensional SC, where the H field is applied *parallel* to the superconducting layers and, thus, orbital effects can be neglected.
[20] Note that a peak effect precursive to the vortex-lattice melting has been observed in YBCO single crystals [W. K. Kwok *et al.,* Phys. Rev. Lett. **73**, 2614 (1994); G. D'Anna *et al.,* Europhys. Lett. **25**, 225 (1994)].
[21] M. R. Norman, Phys. Rev. Lett. **71**, 3391 (1993).
[22] Experiment: Y. Inada *et al.,* Physica (Amsterdam) **206C-207C**, 33 (1995); Theory: L. M. Sandratskii *et al.,* Phys. Rev. B **50**, 15 834 (1994).
[23] For intermediate-valent $CeRu_2$, exhibiting an intrinsic Pauli susceptibility [17] $\chi_P = 2.7 \times 10^{-4}$ (in SI units), the Sommerfeld-Wilson ratio $R = (\chi_P/\mu_0\mu_{\rm eff}^2)/(\gamma_0/\pi^2 k_B^2) \simeq 0.8$ (with $\mu_{\rm eff} = 2.54\mu_B$ [24]). Thus, the Landau parameter $F_0^a = (1 - R)/R \simeq +0.25$, in fact, indicating antiferromagnetic interactions.
[24] D. M. Newns and A. C. Hewson, in *Valence Fluctuations in Solids,* edited by L. M. Falicov, W. Hanke, and M. B. Maple (North-Holland, Amsterdam, New York, Oxford, 1981), p. 27.