Anomalous peak effect in heavy-fermion, intermediate-valence and A15 superconductors: evidence for a Fulde-Ferrell-Larkin-Ovchinnikov state?

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Dedicated to Peter Fulde on the occassion of his 60th birthday

Abstract. An experimental study based upon magnetic and dilatometric measurements is presented for the clean high-κ superconductors UPd_2Al_3, $CeRu_2$ and V_3Si. All three compounds show an enhanced spin susceptibility. Their superconducting state is strongly Pauli limited, and an anomalous peak effect is observed at $T<(0.8–0.9)T_c$, slightly below $H_{c2}(T)$. This phenomenon appears to be qualitatively consistent with a first-order transition between weak and collective pinning, caused by the formation of a staggered order parameter in a generalized Fulde-Ferrell-Larkin-Ovchinnikov phase (M. Tachiki et al., Z. Phys. B, in press).

Keywords: Fulde-Ferrell superconductivity; Clean high-κ superconductors; Vortex pinning.

1 Introduction

In 1964 Fulde and Ferrell (FF) [1] as well as Larkin and Ovchinnikov (LO) [2] predicted a partially polarized superconducting state to form at sufficiently high magnetic fields in clean, Pauli-limited superconductors. The search for the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state has been unsuccessful until recently, when Modler et al. [3] discovered unique anomalies in the sample length, $l(T, H)$, below $H_{c2}(T)$ and $T\leq1.5$ K for the hexagonal, antiferromagnetically ordered (T_N=14.5 K) heavy-fermion (HF) superconductor UPd_2Al_3 ($T_c\leq2$ K) [4]. In a subsequent paper, Gloos et al. [5] suggested that these anomalies mark a first-order transition from the Shubnikov phase to the FFLO state. Their assignment was based on the observations that for UPd_2Al_3 (1) the electronic mean free path l greatly exceeds [4] the coherence length ξ_0 and (2) the Pauli limiting field H_p is much smaller than the orbital field H^*_{c2}, but is (3) almost identical [5] to the upper critical field H_{c2}.

Apart from UPd_2Al_3, the non-magnetic intermediate-valence (IV), cubic Laves-phase compound $CeRu_2$ (T_c=6.1 K [6]) may be considered a promising candidate for

FFLO superconductivity: Huxley et al. [7] and Yagasaki et al. [8] reported striking magnetic anomalies below $H_{c2}(T)$ and for T<5.4 K, similar to those displayed in Fig. 1. The isothermal dc-magnetization curve, M(H), of this compound exhibits a sharp peak (not shown) for fields between the low $H_{c1} \simeq 250$ ϕe (as T → 0) and an also relatively low irreversibility field, H_{irr}. In a wide field range, 15 kϕe<H<30 kϕe, M(H) is almost reversible and, in addition, changes sign. When the field exceeds some critical value H_i, an abrupt change into a regime of strong irreversibility is noticed. At $H_f<H<H_{c2}$, M(H) becomes reversible again. The abrupt increase of diamagnetism at $H>H_i$, the paramagnetic peak observed on reducing the field and the hysteresis of H_i depending on whether H is raised or reduced, indicate a first-order transition between the Shubnikov phase of weak pinning ($H<H_i$) and a state of strong pinning. This resembles the well-known peak effect, often observed near H_{c2} in type-II superconductors with short mean free path [9]. However, like UPd_2Al_3 all the $CeRu_2$ samples referred to in this paper have l values substantially larger than the coherence length ξ_0, and they all exhibit M(H) curves of the kind shown in Fig. 1 [10–12]. Additional support for an abrupt change from weak to strong pinning at $H \geq H_i$ stems from a number of other physical quantities of $CeRu_2$, such as magneto-caloric effect and elastic constants [13, 14], small-angle neutron diffraction [15] and magnetic quantum oscillations [16, 17].

One purpose of the present paper is to demonstrate that, while UPd_2Al_3 shows magnetic anomalies resembling those of $CeRu_2$ [7, 8, 18], the latter compound exhibits dilatometric anomalies which are phenomenologically closely related to the ones reported for UPd_2Al_3 [3, 5]. In addition, we communicate here magnetostriction results on one of the single crystals of the A15 superconductor V_3Si (# 1) for which M(H) results, strikingly similar to those in Fig. 1, have been reported in 1988 by Isino et al. [19]. These experiments have been extended to a very clean V_3Si single crystal (# 5), whose magnetization curves exhibited a peak effect of an only minor size [19].

The paper is organized as follows: In Section 2, we summarize for UPd_2Al_3, $CeRu_2$ and V_3Si some normal-state and superconducting properties emphasizing, apart from a long electronic mean free path, a large value of the normal-state spin

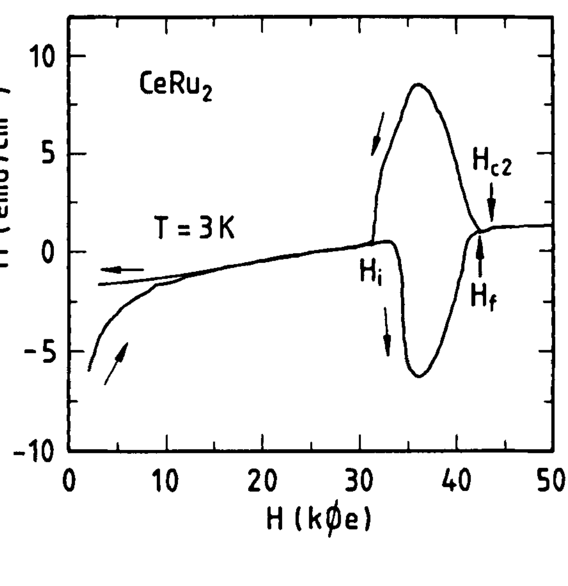

Fig. 1 Isothermal dc magnetization M *vs* H of $CeRu_2$ at T=3 K. Note that a Curie-Weiss contribution due to ≥0.2 at% Ce^{3+} ions has to be subtracted from the data in order to obtain a constant susceptibility $\chi_0=(1.9\pm0.15)\cdot10^{-5}$ emu/cm^3, see text. Arrows indicate measurements done on raising and reducing the field, respectively, as well as onset (H_i), offset (H_f) fields of strong-pinning regime and the upper critical field (H_{c2}). In agreement with the magnetostriction results (Fig. 5), the magnetization is not strictly reversible below $H=H_i$.

susceptibility χ_{spin}. In Section 3 we present for these three superconductors some of the unique anomalies in their magnetic and dilatometric properties that manifest the sudden change from weak to strong pinning mentioned before. In Section 4 we wish to put our results into perspective, i.e. by comparing them with published data on type-II superconductors exhibiting a peak effect of ordinary kind. We shall conclude that our findings for the three title compounds highlight an "anomalous peak effect" which can be qualitatively explained by a recent theory of Tachiki et al. [20] concerning a "generalized FFLO state".

2 Normal-state and thermodynamic superconducting properties

In order to check whether the three compounds of interest meet the strict requirements for the "generalized FFLO state" to form [20], we list in Table 1 relevant information concerning T_c, l, ξ_0, κ, H_{c1} and H_{c2} (taken in the limit $T \to 0$ K). Also included are the values (as $T \to 0$) for both the orbital field $H^*_{c2}=(e^{2+\gamma}/\pi^2)(\Phi_0/2\pi\xi_0^2$ (γ=0.57721 being Euler's constant and ϕ_0 the superconducting flux quantum) [20] and the Pauli-limiting field H_p, as derived from the Clogston criterion: $H_p=0.5\ H_c\ (\pi\chi_{spin})^{-1/2}$ (H_c being the thermodynamical critical field as $T \to 0$). The parameter β, which characterizes the strength of the paramagnetic relative to the orbital pair breaking by the external field, is defined via $\beta=\sqrt{2}\ H^*_{c2}/H_p$ [21]. Finally, estimates are given for the densities of the condensation energy, $\varepsilon_c=H_c^2/8\pi$, as well as of the Zeeman energy, $\varepsilon_z=0.5\ \chi_{spin}H^2_{c2}$. Below, we comment on these numbers for the three materials each in turn.

UPd_2Al_3. Among the heavy-fermion superconductors, the hexagonal compound UPd_2Al_3 is unique in showing microscopic coexistence, below T_c, of superconductivity carried by a "subset of itinerant 5f states" and antiferromagnetic order originating in a "subset of more localized 5f states". From both Knight-shift [22] and specific-heat [23] results the "subset of itinerant 5f states" was characterized by a Pauli spin susceptibility $\chi_p \simeq 3.2\cdot10^{-5}$ emu/cm^3 and by a Sommerfeld coefficient γ_0=125 mJ/K^2 mole. These values for χ_p (=χ_{spin}) and γ_0 have been used to estimate both ε_z and $\varepsilon_c \simeq 0.25\ (\gamma_0/V_{mole})T_c^2$ (V_{mole}=62.94 cm^3/mole).

$CeRu_2$. The $CeRu_2$ single crystal used in this work stems from the same batch as the one recently studied by de Haas-van Alphen experiments [16, 17], the latter yielding an electronic mean free path l=1300 Å. Therefore, we are confident that $l>\xi_0$ holds for our single crystal, too. With V_{mole}=32.23 cm^3/mole and γ_0=29 mJ/K^2 mole [7], we estimate an upper bound of the condensation energy density $\varepsilon_c<84\cdot10^3$ erg/cm^3: Pair breaking by (at least) 0.2at% non-transformed Ce^{3+} "impurities" (with an effective moment of 2.54 μ_B) will reduce $H_c^2/8\pi$. The existence of these paramagnetic pair breakers is inferred from the analysis of our susceptibility results on the same single crystal [24]: The data are reasonably well fitted by $\chi=\chi_0+\chi_i$, where the Curie-Weiss contribution $\chi_i(T)$ with $-\Theta<23$ K is caused by those paramagnetic ions mentioned before, and $\chi_0=(1.9\pm0.15)\cdot10^{-5}$emu/cm^3. Huxley et al. [7] have estimated the normal-state diamagnetic susceptibility due to core and conduction electrons, $\chi_{dia}=-0.3\cdot10^{-5}$emu/cm^3. Thus, the Pauli susceptibility is found to be $\chi_p=\chi_0-\chi_{dia}=2.2\cdot10^{-5}$ emu/cm^3, in reasonable agreement with published data [7, 18]. For the total low-T spin susceptibility we obtain $\chi_{spin}=\chi_p+\chi_i(T\to0)=(2.7\pm0.5)\cdot10^{-5}$ emu/cm^3. The large error bar for χ_{spin} results

Table 1 Normal-state and superconducting properties of UPd_2Al_3, $CeRu_2$ [20] and V_3Si (single crystals # 5 and # 1 from [19]). Values for H_{c1}, H_{c2}, H^*_{c2} (orbital field) and H_p (Pauli-limiting field) are taken for $T \rightarrow 0$. β, ε_c and ε_z are defined in the text.

Compound	T_c (K)	l (Å)	ξ (Å)	κ	H_{c1} (Φe)	H_{c2} (kΦe)	H^*_{c2} (kΦe)	H_p (kΦe)	β	ε_c (10^3 erg/cm^3)	ε_z (10^3erg/cm^3)
UPd_2Al_3	1.85	720	85	50	100	36$\parallel$c 32$\perp$c	61	34	2.5	19.8	20.6$\parallel$c 16.4$\perp$c
$CeRu_2$	6.04	1300	61	16	250	70	118	<79	>2.1	<84	67±12
V_3Si # 5	16.6	300	30	20	700	200	488	220±27	3.1	1080	900±300
# 1	16.7	60									

in a substantial uncertainty of the Pauli-limiting field as well: H_p=(79±8) kϕe. This value will, however, be reduced accordingly by a reduction of the condensation-energy density, i.e. when the action of pair-breaking Ce^{3+} ions is taken into account. Future low-temperature studies of the specific heat and magnetization on the same $CeRu_2$ single crystal are in preparation in order to get more accurate estimates of both ε_c and ε_z. Given the present uncertainty margins, these two quantities are considered to be sufficiently close to each other. Another problem left for future work concerns a striking anomaly in the upper critical field, $H_{c2}(T)$. Compared to the clean limit value of its slope $H'_{c2}=-(dH_{c2}/dT) \simeq H^*_{c2}/0.73\,T_c=26.5$ kϕe/K, we read off Fig. 2b a maximum slope, $H'_{c2,max} \simeq 16.5$ kϕe/K, at $H \simeq 20$ kϕe. The reduction may be explained by the action of the paramagnetic pair breaking effect introduced by a field of this size. Very astonishingly, however, H'_{c2} becomes even smaller at lower fields, an effect that reproduces in the results of different groups [7, 18] and, therefore, appears to be intrinsic.

V_3Si. In Table 1 we have listed the properties of the very clean single crystal # 5 ($l/\xi_0 \simeq 10$) that was thoroughly studied by Isino et al. [19]. This sample undergoes a cubic to tetragonal martensitic transformation at $T_m \simeq 22$ K [19]. The low-T value of the thermodynamic critical field, extrapolated from the data published for T>10 K amounts to $H_c \simeq 5.2$ kϕe. For the "non-transformed" single crystal # 1 (T_m<T_c=16.7 K), the ratio l/ξ_0 is still as large as two [19].

To summarize, the numbers given in Table 1 indicate that all samples discussed in this paper, are fairly clean high-κ superconductors. They are all strongly Pauli limited (i.e. β>1.8 [21]) and exhibit a spin susceptibility large enough to warrant near cancellation of the condensation-energy density ε_c by the Zeeman-energy density ε_z. The latter observation can be expressed by a Pauli-limiting field H_p that is almost the same as the measured upper critical field, H_{c2}.

3 Pinning-related anomalies in magnetic and dilatometric properties

Our investigations on high-quality single crystals of UPd_2Al_3, $CeRu_2$ and V_3Si are based on measurements of the magnetostriction, Δl(H, T=const), thermal expansion, Δl(T, H=const), dc magnetization, M(H, T=const) and M(T, H=const) as well as ac susceptibility, χ_{ac}(H, T=const) and χ_{ac}(T, H=const). For all three of these otherwise very different compounds the above techniques reveal a strikingly similar phenomenology, i.e. an abrupt change from weak to strong pinning giving rise to an "anoma-

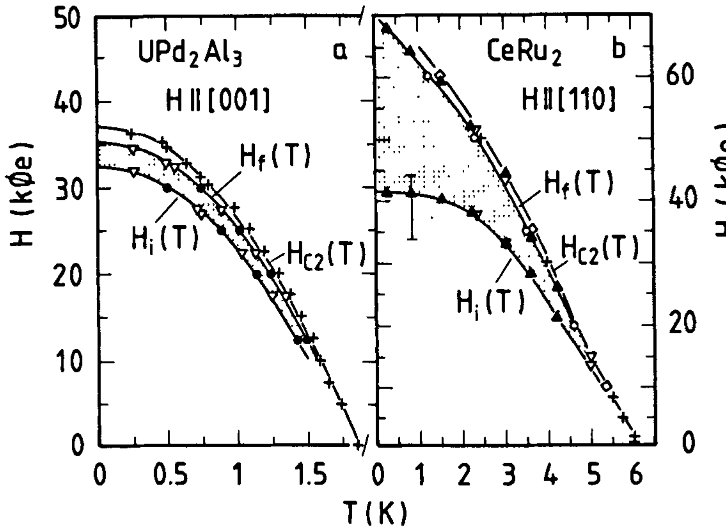

Fig. 2 H-T phase diagrams of UPd_2Al_3 (a) and $CeRu_2$ (b). Anomalies in M(H,T) (▽, Fig. 3), Δl(H) (▲, Fig. 5), Δl(T) (○, Fig. 5) and χ_{ac}(T) (●, Fig. 4) define onset [H_i(T)] and offset [H_f(T)] of irreversibilities, see text. Upper critical field H_{c2}(T) denoted by + [χ_{dc}(H,T)] and [◇ coefficient of thermal expansion $\alpha(T)=1/l\ dl/dT$].

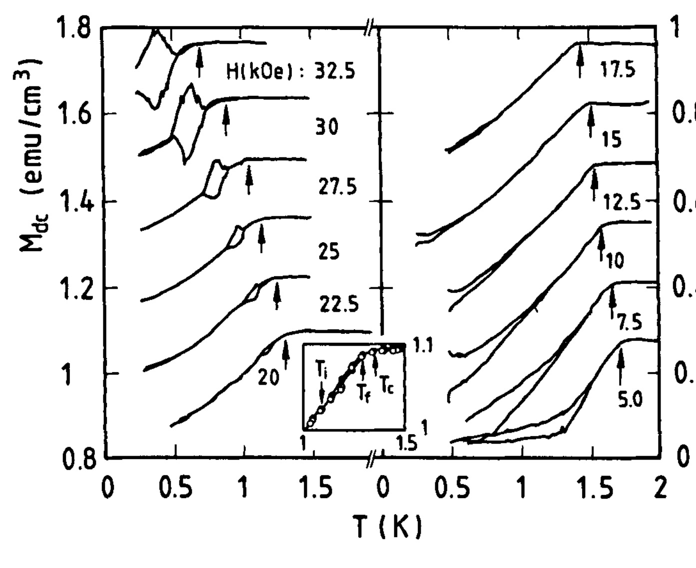

Fig. 3 "Isofield" dc-magnetization M *vs* T for different applied fields for UPd_2Al_3, H‖ [001]. Arrows indicate T_c(H) values. Inset shows 20 kϕe data above T=1 K: ○ and ● denotes values of the hysteretic magnetization taken upon moving the sample up and down within the pick-up coils of the magnetometer, respectively. T_i and T_f mark onset and offset temperatures of irreversibility range.

lous peak effect" somewhat below H_{c2}(T). The onset (H_i, T_i) and offset (H_f, T_f) positions of the strong-pinning regime along with H_{c2}(T) values determined from dc-susceptibility measurements are used to construct the H-T phase diagrams for UPd_2Al_3 and $CeRu_2$ (Fig. 2).

Figure 3 displays a set of dc-magnetization curves on UPd_2Al_3 measured upon cooling in a fixed magnetic field applied along the hexagonal c axis. In the extraction technique used the sample is moved up and down through a set of pick-up coils while the induced voltage is detected with a SQUID system. In the isofield measurements shown in Fig. 3 the temperature was raised in a stepwise manner. Once the

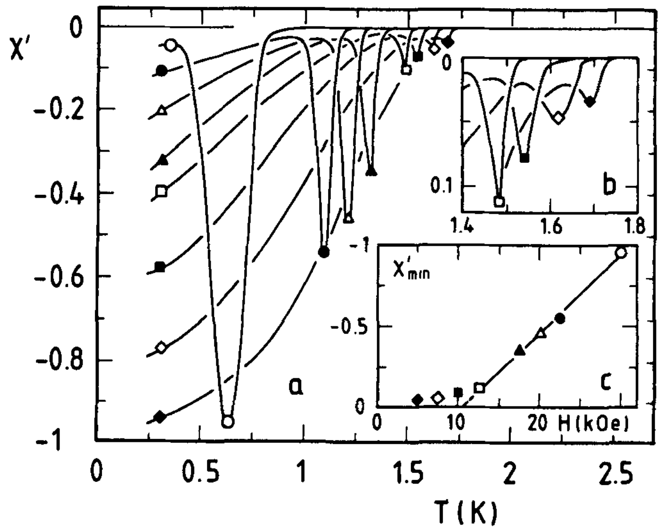

Fig. 4 Real part of χ_{ac}(T, H=const) for UPd_2Al_3 (a, b) at different magnetic fields as indicated by the symbols (c) which mark the magnitude of the minimum *vs* the applied field.

system reached thermal equilibrium, data were collected for both upward and downward movement of the sample. Due to a small field inhomogeneity ($\Delta H/H \leq 4 \cdot 10^{-5}$) the sample experiences a temporal field variation during these extraction processes. The hysteretic behavior observed at the low-T end of the data taken at low fields, H≤17.5 kϕe, indicates a magnetically irreversible state as usually observed below the irreversibility line $H_{irr}(T)$ or $T_{irr}(H)$ of a clean type-II superconductor. Above $T_{irr}(H)$ the experiments reveal an almost reversible magnetization. For magnetic fields H≥20 kϕe, another irreversible range develops at temperatures $T_i \leq T \leq T_f$ somewhat below $T_c(H)$, indicating the onset of strong pinning (cf. insert of Fig. 3). This is consistent with the enhancement of the shielding signal in the same temperature interval as detected in our ac-susceptibility measurements (cf. Fig. 4a). Compared to the dc-magnetization curves, these shielding experiments are able to resolve this phenomenon at even lower fields (Fig. 4b). When plotting the corresponding minimum values of the χ_{ac}(T, H=const) anomaly *vs* field we find a linear decrease upon reducing the field to 12.5 kϕe (Fig. 4c). The deviations of the peaks from this straight line below 10 kϕe are ascribed to demagnetization fields near the edges and corners of our UPd_2Al_3 single crystal.

Clear indications for an abrupt increase of flux pinning can be found also in length measurements performed as a function of both T(H=const) and H(T=const). Figure 5a displays magnetostriction data on the $CeRu_2$ single crystal taken at T=3 K. These data clearly demonstrate that the coupling of the vortex lattice to the crystal lattice mediated via flux pinning increases the stronger the pinning: The abrupt change from weak to strong pinning when raising the field to $H \geq H_i$ causes an enormous increase in the stress, induced by trapped vortices that act on the sample. Like for UPd_2Al_3 [5], for $CeRu_2$ the amplitude of this Δl(H, T=const) anomaly is found to be precipitously reduced upon warming, eventually disappearing completely at $T > T^* \simeq 5.4$ K (corresponding to $H < H^* \simeq 10$ kϕe). A rather abrupt relaxation of the sample length in an almost discontinuous manner can be observed upon warming in a thermal-expansion measurement, when a magnetic field is applied to the sample, following an initial zero-field cooling. This is demonstrated in Fig. 5b for different field histories which correspond to the positions marked by the symbols in Fig. 5a. The rather sharp relaxation which occurs

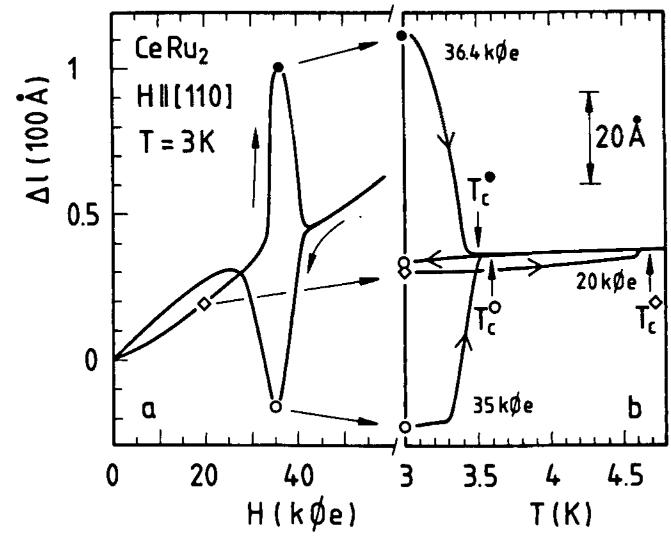

Fig. 5 $CeRu_2$: Isothermal magnetostriction Δl *vs* H (a) and length change Δl *vs* T measured upon warming at different fields (b), starting from different points of the isothermal Δl(H) curve displayed in (a). Δl(T) data are shifted in order to coincide for the normal-state value. Field-cooled curve Δl(T) is shown for H=35 kϕe only.

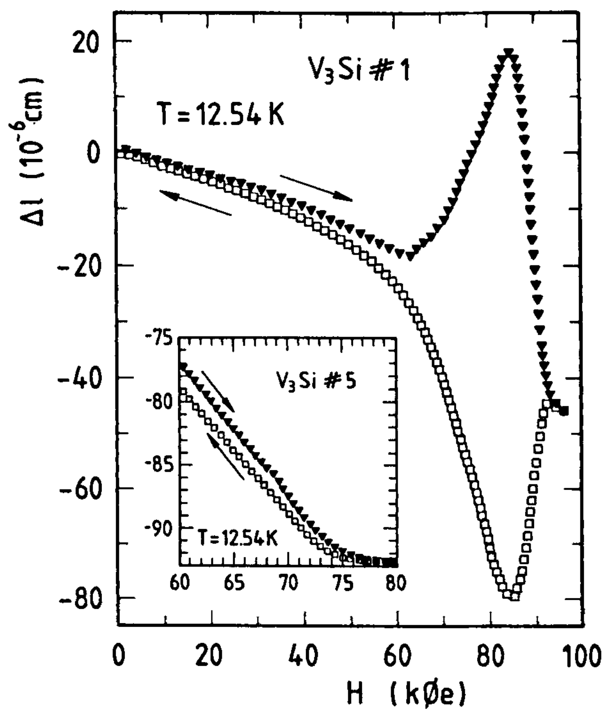

Fig. 6 Isothermal magnetostriction cycle, Δl *vs* H, measured at T=12.54 K for V_3Si single crystal # 1. Inset shows high field data for very clean crystal # 5, showing a weak anomaly at H$\approx$70 kϕe.

slightly below the depinning temperature T_f(H) hints at a substantial weakening of the pinning force.

A strikingly similar phenomenology is found for V_3Si where length measurements have been performed on two single crystals (# 1 and # 5) identical to those studied by Isino et al. [19]. According to these authors crystal # 1 differs from # 5 by a reduced resistance ratio ρ(300 K)/ρ(17 K)=17–12 for # 1 compared to 90 for # 5, indicating a somewhat enhanced defect concentration in the former. The magnetostriction

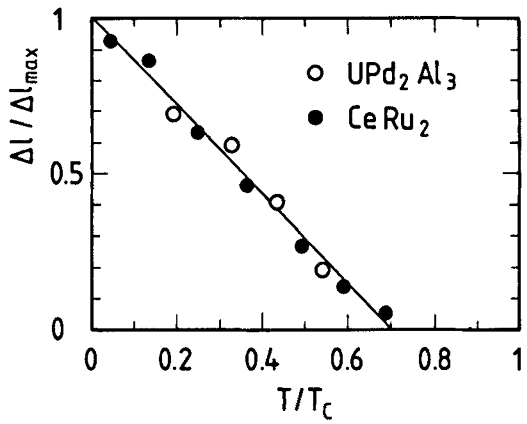

Fig. 7 Amplitude of magnetostriction anomalies *vs* reduced temperature $CeRu_2$ and UPd_2Al_3 single crystals. Data have been normalized to coincide for T/T_c=0.

data of crystal # 1 taken at T=12.54 K are shown in Fig. 6. Similar results on the same crystal were reported already by Isino et al. [19]: Since the latter were obtained at higher temperature (T=15 K) the length change appears to be less pronounced than in Fig. 6. Like for the other two compounds we observe an abrupt change from weak to strong pinning upon increasing the field to H_i followed by a reversible behavior for $H>H_f$. We note that the sudden onset of strong pinning along with the hysteresis of H_i on increasing/decreasing the field indicate that the transition between the two pinning regimes is, in fact, of first order. The analogous magnetostriction experiment performed on # 5 (cf. insert of Fig. 6) demonstrates that a reduced defect concentration in the latter results in a dramatic reduction of the peak height: An only very weak anomaly remains visible near H=70 kϕe. By contrast, a hysteresis of comparable size to that of # 1 is found for fields $H<H_i$. This might indicate that two different sources of flux pinning are responsible for the weak and strong-pinning behavior below and above H_i, respectively. We speculate the former to be dominantly caused by surface pinning due to degradation effects and/or VO_x or SiO_x precipitations. At sufficiently large fields (when the inter-vortex interaction exceeds the pinning force) these pinning centers become ineffective. This might explain the reversible behavior for $H>H_f$.

In Fig. 7 we compare on reduced scales the temperature dependence of the amplitude of the magnetostriction anomalies for UPd_2Al_3 and $CeRu_2$. We refrain from including data for V_3Si, since the field range accessible in our experiment (H≤95 kϕe) does not allow to study the effect down to low enough temperatures, i.e. to below about T/T_c=0.6. Fig. 7 demonstrates a strikingly similar temperature dependence of the Δl(H, T=const) anomalies for both compounds showing a linear reduction with increasing temperature and a complete disappearance at $T/T_c \geq 0.7$.

In Fig. 8 we show length measurements on $CeRu_2$ performed along a closed cycle in the H-T phase diagram. Two observations are worth mentioning: (i) The length balance holds over the full cycle. (ii) A pronounced jump in the sample length occurs along path 2 near $T_f(H)<T_c(H)$, whereas no anomaly can be resolved at lower temperatures, i.e. when warming the sample to $T \geq T_i(H)$. The same observations were made for UPd_2Al_3 [5]. At first glance this is a counter-intuitive result which, however, finds a natural explanation in the fact that in our Δl(T, H=const) measurements there is no driving force acting on the flux lines: (i) Because of the large κ, the high-field magnetization, i.e. the concentration of vortices, does virtually not change upon

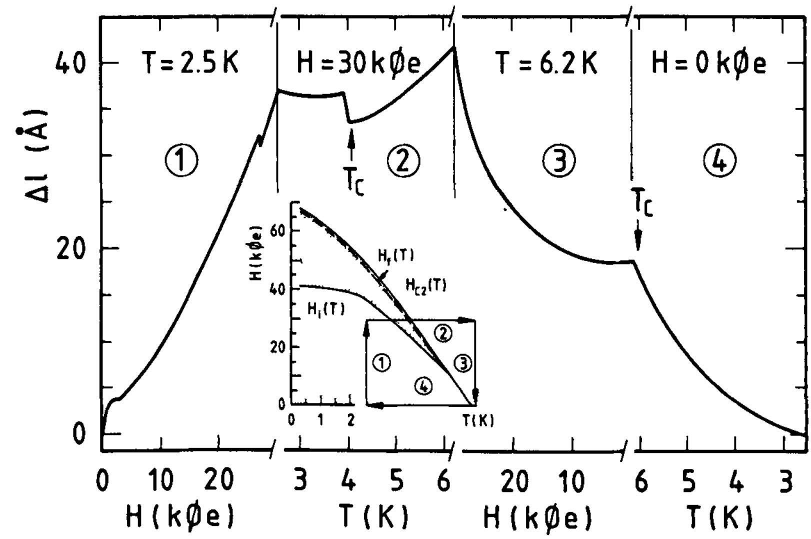

Fig. 8 Dilatometric investigation of a closed cycle in the H-T-plane of $CeRu_2$ (see inset) consisting of four subsequent measurements (H∥[110]): (1) isothermal magnetostriction (T=2.5 K) starting from a superconducting state after zfc, (2) isofield thermal expansion (H=30 kϕe), (3) isothermal magnetostriction (T=6.2 K) and (4) zero-field thermal expansion. Since the length balance is conserved, the small discontinuity in run (1) may be ascribed to a jump of flux bundles within the weak-pinning superconducting state. The characteristic length jump in run (2) occurs at T=(3.90±0.03)K, i.e. below T_c(30 kϕe)=(3.98±0.02)K: both magnitude and sign of the anomaly depend strongly on the prehistory (1), cf. also Fig. 5.

warming. (ii) In the absence of temporal field variations, no Lorentz force is operative which would enable the flux lines to gain energy by taking advantage of the strong pinning at $T \geq T_i$. On the other hand, in the measurements of both dc magnetization (due to the motion of the sample along a small field gradient) and, of course, ac susceptibility the Lorentz force is operative and the abrupt increase in pinning strength can be easily recognized. The above reasoning that Δl(T, H=const) does not react on the prominent first- order transition at $T=T_i$ presumes the existence of a finite pinning potential at $T<T_i$. In fact, hysteresis effects in this part of the H-T phase diagram are clearly resolved in the magnetostriction data (cf. Fig. 5), isothermal dc magnetization [25] as well as in the ac susceptibility (the finite shielding signal for $T<T_i$, cf. Fig. 4). Note that, like for UPd_2Al_3 and V_3Si, no hysteresis is seen in these experiments above $H=H_f$, indicative of a gross reduction of the pinning force upon approaching H_{c2}.

4 Perspective

According to the experimental results presented above three otherwise rather different compounds show very unusual, but similar pinning anomalies: antiferromagnetically ordered HF-UPd_2Al_3, non-magnetic strongly IV-$CeRu_2$ and the A15 compound V_3Si. The development of strong pinning when approaching H_{c2}(T) resembles the peak effect often observed in type-II superconductors. Several mechanisms can cause such a peak effect [9]: (1) Sample inhomogeneities giving rise to normal regions of typical width $d>\xi_0$, (2) "matching" between the array of pins and the vortex lattice and (3) "synchronization" between pins and vortices owing to a softening of the vortex lattice that overcompensates the weakening of the pinning force. None of these mechanisms can, however, explain the results described in Section 3: (1) Sample inhomogeneities are not likely to play an important role in view of the high quality of the samples used in the present investigation. (2) "Matching" should occur at a certain field and, thus, no peak effect should be observable in temperature scans at constant field, in contrast to the results of Figs. 3 and 4. (3) "Synchronization" should result in a gradual rather than in an abrupt increase in pinning. In addition, this mechanism should be operative even close to T_c (close to H=0). As an example, we refer to the critical-current measurements by Wördenweber and Kes [26] on amorphous Nb_3Ge and Mo_3Si films for which synchronization is provided via three- to two-dimensional crossover of the vortex lattice. For these films, the peak effect can be observed even near $T=T_c$ [26], while the magnetostriction anomalies of UPd_2Al_3 and $CeRu_2$ disappear well below T_c (see Fig. 7). We, therefore, conclude that these latter superconductors exhibit a peak effect of novel origin.

An explanation for the anomalous peak effect established for UPd_2Al_3, $CeRu_2$ and, presumably, V_3Si was recently given by Tachiki et al. [20]. Following the theory by Burkhardt and Rainer for a quasi-twodimensional superconductor in a parallel external field [27], they reported the first non-linear theory which addresses the interplay between the Abrikosov vortex lattice and the non-uniform FFLO state. The latter is characterized by a spatially modulated order parameter giving rise to a periodic array of nodal planes (the "LO planes") perpendicular to the vortices. This theory requires the following criteria to be fulfilled: (i) a large electronic mean free path ($l>\xi_0$), (ii) Pauli limiting dominating over the orbital pair-breaking effect by the external field ($\beta>1.8$ [21]), (iii) a Zeeman-energy density that equals the superconducting condensation-energy density and (iv) a short coherence length, or a large GL parameter $\kappa=\lambda/\xi_0$. Table 1 shows that the criteria (i)–(iv) are indeed met for the three superconductors of interest in this paper. Tachiki et al. [20] found a first-order phase transition from the Shubnikov phase to the FFLO state to occur at sufficiently low temperature, or sufficiently high magnetic field. Provided that suitable pinning centers (e.g. point defects) are distributed at random in the superconductor, the afore-mentioned first-order transition ought to be accompanied by an abrupt change from weak to strong pinning, i.e. from a nearly reversible to an irreversible magnetization behavior. This phenomenology can be understood as follows. The weak pinning in the Shubnikov phase is a consequence of the near cancellation of Zeeman- and condensation-energy densities, giving rise both to a very small self energy of the vortex core, $\varepsilon_{core}\simeq\pi\xi_0^2(\varepsilon_c-\varepsilon_z)$, and to a low H_{c1} value, as observed (cf. Table 1). However, once the vortices become truncated by the "LO planes" at sufficiently high field, the vortex segments (with length $\Lambda\simeq$ several 10 ξ_0) can accommodate to the weak random pinning potential more easily than the intact vortices can at lower fields. This results in an enhanced ("collective") pinning of the vortices.

Whereas the formation of the "generalized FFLO state" studied by Tachiki et al. [20] is apt to explain the qualitative behavior of UPd_2Al_3, $CeRu_2$ and V_3Si as described in the preceding section, several specific points need clarification:

1. The criteria (i)–(iv) required for the FFLO state to develop are not sufficient to observe the "anomalous peak effect". For example, a high-quality single crystal of the heavy-fermion superconductor $CeCu_2Si_2$ which fully meets these requirements shows a reversible magnetization near H_{c2} without a peak effect [28, 29]. On the

other hand, for an almost ideal type-II superconductor one would not expect to find the FFLO-induced change from weak to strong pinning. High-quality single crystals of UPd_2Al_3 appear to come close to this limit [30]. Obviously a sufficiently weak random pinning potential is necessary to observe this change. We think that the apparent differences evident in Fig. 6 for the two V_3Si single crystals have to be explained this way: The anomalous peak effect is well pronounced for the crystal # 1 ($l/\xi_0 \simeq 2$), while it is very weak for the high-purity crystal # 5 ($l/\xi_0 \simeq 10$). Furtheron, the relatively strong pinning in $CeRu_2$ (when compared to UPd_2Al_3) highlights the role of pair-breaking Ce^{3+} "impurities", evidenced by a Curie-Weiss contribution to the magnetic susceptibility. This possibility will be explored by future experiments.

2. The onset of the FFLO state should change not only the bulk pinning properties, but should also modify the surface pinning. We plan to study this effect on the V_3Si crystal # 5 in some detail: The hysteretic effects seen for this material below $H=H_i$ both in the magnetization [19] and in the magnetostriction (Fig. 6) point to a degradation of the surface which, in turn, gives rise to strong surface pinning. If one were able to moderately reduce the latter for this pure crystal by careful etching, one might remove the irreversibilities below H_i and then be able to resolve better the onset of the anomalous peak effect as a consequence of the staggered FFLO order parameter influencing both bulk and surface pinning.

3. The unique relaxation of the sample length upon warming in a constant field suggests a precipitous weakening of the pinning force when approaching the depinning temperature, T_f, of the vortex lattice, cf. Fig. 8. Future calculations have to show how temperature-dependent changes of the modulated order parameter are involved in this dramatic jump in the sample length.

4. The absence of hysteresis beyond the peak effect, i.e. for $H_f<H\leq H_{c2}(T)$ and $T_f<T\leq T_c(H)$ suggests a vanishing stiffness of the vortex lattice in the FFLO state upon approaching H_{c2}. Investigations of the elastic properties of the vortex lattice, in particular the shear modulus, have to show whether H_f or T_f mark the transition from a vortex solid to a vortex liquid phase.

5. The existence range for the FFLO state, i.e. $H_i(T)\leq H\leq H_{c2}(T)$ or $T_i(H)\leq T\leq T_c(H)$, is much larger than predicted by the original theories [1, 2]. For example, the theoretical temperature limit is $T^* \simeq 0.56\,T_c$, whereas the unique pinning anomalies described above can be monitored for the three title superconductors up to $T^*=(0.8–0.9)T_c$. While the original theories assume free electrons, realistic electronic structures should be incorporated in a modified version of the theory by Tachiki et al. [20]. In particular, multi-sheeted Fermi surfaces as present at least in the cases of UPd_2Al_3 [31–33] and $CeRu_2$ [16, 17] make antiferromagnetic spin-exchange interactions between carriers probable. This would favor an expanded field range for the FFLO state [27]. For instance, with the normal-state data reported in Section 2 for the IV- compound $CeRu_2$ we estimate a Sommerfeld-Wilson ratio (in SI units) $R=(\chi_p/\mu_0\mu_{eff}^2)/(\gamma_0/\pi^2k_B^2)\simeq 0.8$ ($\mu_{eff}=2.54\ \mu_B$ [34]), yielding a Landau parameter $F_0^a=(1-R)/R\simeq +0.25$. This indicates, in fact, antiferromagnetic electron-electron correlations.

Though the problems mentioned before require intensive future work, it is fair to say that the proposal by Tachiki et al. [20] of the formation of a staggered order parameter in the generalized FFLO state provides a very plausible explanation for the "anomalous peak effect" in UPd_2Al_3, $CeRu_2$ and V_3Si. With respect to the criteria (i)–(iv) that have necessarily to be met, other candidates for FFLO superconductivity can be searched for. Among the heavy-fermion compounds, UBe_{13} [35] and UPt_3 [36] have already been discussed as potential examples. FFLO superconductors may also be found among the new boro-carbide compounds [37]. In view of the fact that all the available evidence for the FFLO high-field phase is indirect, i.e. provided by unique pinning anomalies which are demanding for suitable pinning centers, a direct observation of the proposed staggered order parameter would be highly desirable. For example, scanning-tunneling spectroscopy should be employed to identify the high quasiparticle density of states at the position of those unique planar nodes predicted by Tachiki et al. [20].

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