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"Anomalous Peak Effect" — Is it Indicative of a Generalized Fulde-Ferrell-Larkin-Ovchinnikov State?

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Dilatometric and magnetic studies on single crystals of the high-\(\kappa\) superconductors UPd\(_2\)Al\(_3\) and CeRu\(_2\) reveal a spontaneous increase of the volume-pinning force at high magnetic fields of more than one order of magnitude. This “anomalous peak effect” was claimed to be a signature of a new staggered high-field superconducting order parameter. We will review the recent developments and discuss alternative explanations.

1. INTRODUCTION

In the past three years, an exotic superconducting state has attracted much attention in connection with clean type-II superconductors that possess a strongly Pauli-limited upper critical field \(H_{c2}(T)\). The interest was initiated by first results of thermal-expansion and magnetostriction measurements on the heavy-fermion superconductor UPd\(_2\)Al\(_3\) [1]: Within the superconducting state near the upper critical field \(H_{c2}\), distinct anomalies in the length change of a single crystalline sample were found that were interpreted as an indication of a new high-field superconducting Fulde-Ferrell-Larkin-Ovchinnikov state [2].

The first theoretical prediction of such a new high-field superconducting state goes back to 1964, when P. Fulde and R.A. Ferrell, as well as A.I. Larkin and Yu.N. Ovchinnikov (FFLO) [3, 4] studied an alternative, field-induced superconducting pair state: When the Zeeman-energy between singlet-pairing electrons is sufficiently high, a modification of the singlet state is expected to be energetically favorable, which extends the stability of the superconducting phase to higher magnetic fields. The Cooper pair then transports a net moment \(q \neq 0\) which causes a partially depaired superconducting state [3, 4]. The energy gain of the new state with respect to a BCS \((q=0)\) state is of purely magnetic origin: the depaired fraction of electrons gain back their full Zeeman energy [5].

Despite the serious theoretical arguments, up to now, no experimental investigation has been successful to prove the existence of the FFLO superconducting state. However, the aforementioned claim of a possible realization of the FFLO state in UPd\(_2\)Al\(_3\) has provoked controversy impacting both the development of theory [6, 5, 7] and experiment [8, 9]. Additional systems like CeRu\(_2\) [10], V\(_3\)Si [9, 11], Nb/Ti [12], NbSe\(_2\) [13, 14] and Yb\(_3\)Rh\(_4\)Sn\(_3\) [15] have been found to resemble the phenomenology of UPd\(_2\)Al\(_3\) which clearly indicates that the observed phenomenon is of a more general type, being not only restricted to strongly Pauli-limited systems. The diverse type-II superconductors of interest all show similar, pinning-related anomalies near their upper critical field that could be best characterized as an “anomalous peak effect” (APE) i.e. a distinct maximum in the volume pinning force below \(H_{c2}\) that is accompanied by a peak in the critical current density [16]. An interpretation in terms of a superconducting FFLO state [7, 8, 9, 17], however, faces severe problems which will be discussed here. The origin of the “anomalous peak effect” in these systems, thus, remains unclear.

The purpose of this paper is to review recent experimental and theoretical developments in this field. The outline is as follows. Because basic conclusions are drawn from dilatometric experiments, we start with a brief description of how pinning forces affect dilatometric experiments. We will then summarize the experimental status by focusing on exemplary superconductors CeRu\(_2\) and UPd\(_2\)Al\(_3\). Several phenomenological mechanisms will be considered that are expected to provide a “conventional” explanation for the observed peak effect and that are based on the dynamics of the flux-line lattice (FLL). A critical discussion of the generalized FFLO state follows, which also had been considered to give rise to a change in the pinning capability of fluxoids. We will draw some final conclusions and close with an
Fig. 1. Isothermal magnetization $M \equiv H$ of single-crystalline CeRu$_2$ for $H \parallel [110]$. At $H_1$, a hysteresis loop opens abruptly, resembling a first-order phase transition. The hysteresis closes at $H_2$, distinctively below $H_{c2}$ [8].

outlook on future work.

2. EXPERIMENTAL STATUS

2.1. Length changes as a measure of pinning forces

Usually thermodynamic properties of a clean, single-crystalline type-II superconductor dominate the length change $\Delta l$ as a function of magnetic field $H$ or temperature $T$, and the sample deformation originating from pinning forces can be neglected [18]. For yet unknown reasons, however, pinning near $H_{c2}$ generally plays a dominant role in the superconductors described below. First, we focus on the recently developed experimental technique of determining pinning forces by measuring the deformation of the sample [1, 19]. Magnetostriction measurements on a superconducting sample in the presence of strong pinning forces can directly reflect the sum of pinning forces acting on the sample. Applying a field gradient perpendicular to a pinned vortex creates a force that acts on the pinning center in the direction of decreasing field. For an idealized sample (a cylindrical crystal with a random distribution of pinning centers), the external field causes a symmetric deformation of the crystal (see also [19, 12]). Deviations from cylindrical symmetry or an inhomogeneity of pinning centers furthermore create a resulting force on the sample. Since every pinned vortex contributes to the deformation/force on the sample, the dilatometric data give direct access to the volume-pinning force $F_p$: Assuming a linear response of the crystal lattice as well as of the experiment to the acting forces, for a first approach one could presume: $F_p \propto \Delta l$. The transverse deformation [19] also causes via elastic coupling a longitudinal length change which can be measured with a high-resolution dilatometer [1].

2.2. The peak effect in CeRu$_2$

CeRu$_2$ is a non-magnetic intermediate-valent cubic Laves-phase superconductor ($T_c \approx 6.2$K, $\kappa \approx 16$, $\xi_0 \approx 61\AA$) [21, 10]. Recent research activities on this compound were initiated by reports of Huxley et al. [10] and Yagasaki et al. [22] who found anomalies within the superconducting state, similar to those observed in UPd$_2$Al$_3$ (which we will describe in section 2.3).

Fig. 1 shows an isothermal magnetization measurement at $T=3$K on a single crystalline CeRu$_2$ sample. One observes a fairly reversible range of the magnetization $M$ for fields $0 \leq H < H_1$ and a spontaneous opening of a hysteresis loop for $H_1 \leq H \leq H_f$, where $H_f$ is located distinctively below $H_{c2}$. The huge hysteresis loop between $H_1$ and $H_f$ has to be attributed to immense pinning forces. Since the sharp starting point of the anomaly at $H_1$
Fig. 3 Isothermal, longitudinal magnetostriction data $\Delta l$ vs $H$ for different temperatures on the same CeRu$_2$ single crystal for $H\| [1\bar{1}0]$ as used in the measurement of Fig. 2(a). Here the sample is not exactly placed in the center of the solenoid which gives slightly different forces on the sample and thus a different shape of the $\Delta l(H)$ curves. At $T = 0.25K$ : $\Delta l$ for increasing field; $\circ$ : for decreasing field [20].

Fig. 4 H-T phase diagram of CeRu$_2$. The superconducting second-order phase transition $H_{c2}(T)$ is determined by dc-susceptibility (+) and by the thermal-expansion coefficient (\(\phi\)). The irreversible range (dark-shaded region) is taken from magnetostriction ($H_1$; $\Delta l$; $H_1$; $\triangledown$) and dc-magnetization data (\(\bigcirc\)) as well as from measurements of the relaxation of the sample length at $T_1(\square)$. For several values $(T_0, H_0)$ the magnitude of length changes $\delta l$ of small hysteresis loops $H = H_0 \pm 1 k\Omega e$ (indicated by vertical bars 1), are displayed. These provide another relative measure of the pinning forces acting on the sample at different parts of the H-T diagram. The hysteresis of $5A$ at $(T_0=5K, H_0=30k\Omega e)$ in the normal state of the sample gives an estimate of the underlying experimental error for this kind of investigation [8, 20].

itself shows hysteretic behavior with respect to increasing and decreasing field, a first-order transition between weak and strong pinning has been postulated [8]. From the longitudinal magnetostriction data shown in Fig. 2(a), we realize that in the field range below $H_1$, $\Delta l$ is not completely reversible, obviously due to weak pinning forces. However, between $H_1$ and $H_f$, the hysteresis exceeds the previous value by more than one order of magnitude. While increasing field between $H_1$ and $H_f$, the sample is squeezed by the pinned vortices and therefore the longitudinal length increases. When decreasing the field, the sample is stretched transversely, resulting in a longitudinal contraction. Fig. 2(b) shows, that by starting a length measurement in an applied magnetic field as a function of temperature from the different field values assigned in Fig. 2(a), the "frozen-in" strain of the rigid FLL acting on the crystal remains until just below $T_c$. The relaxation of the sample length at $T_1(H) < T_2(H)$ could be due to both depinning or melting of the vortex-lattice and is reminiscent of a first-order transition in the FLL [1, 23]. Similar longitudinal magnetostriction data as in Fig. 2(a) are shown in Fig. 3. The difference between the results of Figs. 2(a) and 3 stems, in the latter case, from a slight off-centric position of the (same) sample (same direction) in the solenoid, that supposedly leads to the more complicated shape of the magnetostriction curves. Nevertheless, the magnitudes of the hysteresis loops still give a relative measure of the resulting volume-pinning force $F_p$, which is plotted vs reduced temperature in Fig. 8. We notice that the anomalies are drastically increasing in size when going to lower temperature, approaching a factor of 100 in comparison to the nearly reversible range below $H_1$. On the other hand, a linear decrease of the volume-pinning force with increasing temperature is observed, the main effect vanishing for $t = T/T_c \rightarrow 0.70$ (Fig. 8). These results are further compiled in the phase diagram, Fig. 4 where the irreversible range is indicated by the dark-shaded area. When comparing this phase diagram with results of other groups, we note a remarkable resemblance of all these phase diagrams obtained on different single- and polycrystalline samples [10, 22, 24, 25, 26, 27, 28]. The peak-effect in CeRu$_2$ seems, therefore, not to be restricted to samples with high defect concentration: For example, a very pure single crystal of CeRu$_2$ with...
Fig. 5(a) H-T phase diagram of UPd$_2$Al$_3$ as measured by ac-susceptibility ($\alpha$: onset/end of irreversibility), dc-magnetization (+: $T_c$, $\bullet$: onset/end) [8, 9] and the isothermal magnetostriction (b) [\$: onset, \$: end]. (b) isothermal magnetostriction data in the strongly irreversible range; a background has been subtracted [2].

an estimated electronic mean free path $l \approx 1340\,\text{Å}$ ($\approx 20 \cdot \xi_p$) shows considerable quantum oscillations in a deHaas-vanAlphen experiment as well as an irreversible range below $H_{c2}$ [29].

2.3. The peak effect in UPd$_2$Al$_3$

UPd$_2$Al$_3$ is an antiferromagnetic heavy-fermion superconductor ($T_N \approx 14.2\,\text{K}$, $T_\alpha \approx 2\,\text{K}$, $\kappa \approx 50$, $\xi_p \approx 85\,\text{Å}$) [32]. The magnetic and the superconducting properties are determined by different subsets of 5f electrons [33]. Although the magnetic order is clearly affected by an external magnetic field, a correlation between the underlying magnetic order and the pinning properties has not yet been found [1]. Figure 5(b) displays isothermal magnetostriction data taken around the irreversible range $H_i < H < H_{c2}$ on a single crystalline UPd$_2$Al$_3$ sample ($T_{c0} = 1.85, H \parallel c$) [1, 2]. Strongly irreversible features in $\Delta l(H)$ on increasing and decreasing the magnetic field are observed [1, 2]. The magnitude of these anomalies again vanish linearly with $t \rightarrow 0.70$ (Fig. 8). In results of the ac-susceptibility on the same sample (Fig. 6), the pinning shows up as a negative peak. Interestingly, this feature in $\chi_{ac}$ can be observed even in very low magnetic fields, which reflects a higher sensitivity of $\chi_{ac}$ compared to the dilatometric data. However, a qualitative change in the field dependence of the magnitude can also be seen at $H \approx 10\,\text{kOe}$ [Fig. 6(c)]. Figure 7 compares the dilatometric to the magnetic data obtained on the same single crystal. From inset (a) follows that a softening of the FLL occurs near $T_f$, where the APE already vanishes. A comprehensive H-T phase diagram from these results is displayed in Fig. 5(a) [8, 9]. The strongly irreversible range is shaded.

3. DISCUSSION

3.1. Phenomenological comparison

In the previous sections, we have summarized experimental facts on two very different superconductors that both show a distinct peak effect. The pinning seems not just only to be related to the density of pinning centers, since the effect could be observed in any bulk sample of the here considered systems with a supposedly minimum requirement of a very diluted distribution of pinning centers. The ranges of strong irreversibility in the $H$-$T$ diagram vary little between single- and polycrystals, whereas the latter are subject to much larger defect concentrations that should severely modify the pinning properties. The magnitude of the anomalies in both compounds scale in the same way suggesting that the same underlying mechanism for the peak effect is active. In comparison to the peak effect found in a superconducting amorphous Nb$_3$Ge thin film (Fig. 8, [30]) it seems to originate from a different mechanism since in this case $F_p$ extrapolates linearly to $t=1$. It has to be stated, however, that in certain Nb$_3$Ge thin films, $F_p$ was reported to vanish also at a $T/T_c < 1$ [31]. The magnitude of $F_p$ in the Nb$_3$Ge amorphous thin film changes only by a factor of about two at low temperatures, whereas, the here described single crystalline samples show a gigantic increase of nearly two orders of magnitude. Inspite of a systemic search for the
peak effect in related compounds, like e.g. in the homologue heavy-fermion superconductor to UPd$_2$Al$_3$, UNi$_2$Al$_3$, and in the heavy-fermion superconductors CeCu$_2$Si$_2$ and UBe$_1$_3, an APE was not observed in any of the investigated samples [34].

We will now turn to a discussion of “classical” phenomenological mechanisms that could engender a peak effect.

3.2. Origin of the peak effect

For type-II superconductors with high defect concentrations, an increase of the volume-pinning force near $H_{c2}$ is a well known phenomenon [16]. Several studies in the past have investigated the influence of defect density on the pinning properties [16, 35, 36]. The occurrence of a peak effect was attributed to the interplay between a more or less rigid vortex lattice and the particular distribution of pinning centers. In the following, we will list the main mechanisms that lead to a peak effect [16] and compare it to the experimental results with respect to the APE.

(i) At a certain magnetic field, the vortex-lattice constant could match a periodic defect structure (“matching”). This mechanism can be evaluated if the maximum of the peak in $F_P$ is at a constant magnetic field for different temperatures. In the cases considered here, we can safely exclude this mechanism.

(ii) The elasticity of the FLL is of importance [16, 37]: (a) The FLL could adjust to the pinning centers as soon as its shear modulus $C_{66}$ softens near $H_{c2}$ (“synchronization”). This is the major mechanism in thin superconducting films, since $C_{66}$ is the only elastic constant that remains relevant in a 2-dimensional case (see e.g. [35, 30]). Though intensively searched for, the APE has not been found in thin films of both UPd$_2$Al$_3$ [38] and CeRu$_2$ [39], but so far appears in all bulk samples and thus, has to be of a different origin. Furthermore, the sharp transition observed at $H_{t1}$, as shown in Fig. 1, should not be caused by a smoothly vanishing shear stiffness of the FLL. (b) Further, the peak effect might be due to the dispersive character of the tilt modulus $C_{44}$ of the FLL [37]. This could, in fact, be an important mechanism and may explain why the ac-susceptibility (frequency $f \cong 10^2$Hz) is so sensitive at very low fields, whereas the main effect in the quasi-static magnetostriction vanishes already at $t=0.7$. However, the ac-susceptibility also shows a qualitative change in the magnitude of the peak, that might suggest different pinning mechanisms for the temperature ranges above and below $H=10k\Omega e$.

(iii) A further interpretation is based on the collective pinning mechanism [40] which occurs when the FLL softens or melts near the super- to normalconducting transition line. Such a mechanism should
be unlikely in the compounds considered here, since we do not observe a relaxation of the strained sample as a function of temperature at $T_1$ [see Fig. 2(b), 7(a)]. It has to be stressed, that the dilatation measurement technique used for these zero-field cooled temperature runs is, in contrast to other techniques, a real static field measurement.

In the following we want to critically review the recent microscopic interpretation of the peak effect that was suggested in Refs. [2, 7, 17].

3.3. Fulde-Ferrell-Larkin-Ovchinnikov superconducting state

An FFLO superconducting state could stabilize superconductivity in a high magnetic field by allowing a small fraction of electrons to become normal conducting and thus recover their Zeeman energy. For this state, the effect of the magnetic field on the spins of the charge carriers has to be dominant, i.e. the superconductor should possess a high spin susceptibility and the upper critical field should be set by Pauli-limiting. Therefore, stringent requirements for the formation of this state are imposed. A lower limit for the parameter $\beta = \sqrt{2} H_{c2,orb}/H_P > 1.8$ was introduced, which is a minimum requirement for the Pauli-limiting field $H_P$ relative to the orbital pair-breaking field $H_{c2,orb}$ [41].

H. Burkhardt and D. Rainer were the first to do a complete calculation of the FFLO state that exceeds the previous approximations of a Landau-type theory [5]. Their calculations were for quasi 2-dimensional systems, with the magnetic field direction parallel to the superconducting sheets, thus neglecting orbital effects. They found a spatially oscillating order parameter with a wavelength $\Lambda = 15...50 \cdot \xi_0$, depending on the external magnetic field. A typical phase diagram is shown in Fig. 9. In fact, the theoretical diagram resembles the experimental phase diagrams [Figs. 4 and 5(a)], however, the temperature range of stability of the FFLO state in these calculations, as well as in most other theoretical approaches, does not exceed a critical value of $t=0.56$ which is also only obtained for a large parameter $\beta$ (see also [6]).

The venture towards a 3-dimensional calculation of the FFLO state was done by M. Tachiki and S. Takahashi who extended the model of Burkhardt and Rainer to include orbital currents [7]. This model should be described here in more detail by first considering the basic energetic conditions of a single flux line. The core energy of a cylindrical vortex with diameter $\xi$, the superconducting coherence length, is given by:

$$E_{\text{core}} = \pi \xi^2 \left[ \frac{H^2}{8\pi} - \frac{1}{2} \chi_{\text{spin}} h^2 \right],$$

where $h$ is a mean magnetic field inside the flux line. In the normal core of the vortex, the Zeeman energy of the charge carriers is gained. A large spin susceptibility thus reduces the core energy. Through this, also the pinning force of the vortex is determined: If $E_{\text{core}}$ is small, the energy gain at a pinning center will be low and thus, the pinning force will be small. Tachiki et al. showed, that if the Zeeman energy $\varepsilon_Z = 0.5 \cdot \chi_{\text{spin}} h^2$ equals the condensation energy density $\varepsilon_c = H_{c2}^2/8\pi$ a further, major requirement for the occurrence of the FFLO state is fulfilled [7]. In this case the pinning forces have consequently to be considered as being relatively small.
The 3d gap function of the generalized FFLO (GF-FLO) state then results in a superposition of the 2d Abrikosov lattice $\tilde{\Psi}(x, y)$ and a spatially oscillating term, $\tilde{\eta}$, pointing in direction of the magnetic field $(z)$:

$$\Delta(\tilde{r}) = \tilde{\Psi}(x, y) \cdot \tilde{\eta}(z),$$

with $\tilde{\eta}(z) = \tilde{\eta}(z + \Lambda)$ and $\Lambda$ the wavelength of the oscillation, which again is of the order $15...35\xi$.

In the GFFLO state, the Abrikosov vortex lattice is thus "cut" into layers of thickness $\sim 10\xi$ which change the background (though beforehand very weak) pinning properties drastically, as shown in Fig. 10. Within the new state, the vortices gain flexibility caused by the nodal planes in the order parameter and consequently can adjust more easily to a random pinning potential. Thus, a transition from very weak pinning to collective pinning should occur, which was calculated to be of first order.

Since this picture is also based on the softening of the FLL at $H_1$, it seems not to be consistent with our experimental observations. Furthermore, stringent conditions, like the temperature-stability range ($t < 0.56$) as well as the parameter $\beta$ (especially for Yb$_2$Rh$_4$Sn$_{13}$ [15]) are thoroughly violated. Thus, a GFFLO state within the present theoretical framework cannot account for the experimentally observed APE. However, a GFFLO state – if it exists – should provide an increase of the volume-pinning force, if one considers an appropriate superconductor that meets the stringent theoretical requirements of a large parameter $\beta$, a high Ginzburg-Landau parameter $\kappa$, a high spin-susceptibility ($\epsilon_z = \epsilon_2$) and a large electronic mean free path. In contrast to the latter requirement, it was shown that the peak-effect in CeRu$_2$ is fairly robust against implantation of inhomogeneities [24], which would clearly violate the clean-limit constrained of an FFLO state. Experimental observations on the, so far, highest purity CeRu$_2$-sample reveal a decrease of $H_{c2}(0)$, while $H_1(0)$ seems to remain at 4T [43]. The extension of the upper critical field, thus, seems to be introduced by a very small amount of defects. Up to now, there is no explanation why the superconducting state in UPd$_2$Al$_3$ and CeRu$_2$ is so much more sensitive to defects compared to other systems. However, in the compounds considered here, the vortex-core energy, as discussed above, should still be an important issue when trying to understand the odd pinning properties in these systems. Interestingly, in CeRu$_2$ as well as in Yb$_2$Rh$_4$Sn$_{13}$ [15], the onset of the APE at $t = 0.7$ coincides with an inflection point of the $H_{c2}$ curve. One could speculate, that the range of the APE is an extension of the superconductivity up to higher fields with respect to a clean-limit ($H_{c2} \rightarrow H_1$) case.

### 4. PERSPECTIVE

In special type-II superconductors like UPd$_2$Al$_3$ and CeRu$_2$, an anomalous peak effect is found: At distinct fields $H_1(T)$ and $H_T(T)$ one finds first-order like transitions that mark a hysteretic strong-pinning range in the $H$-$T$ phase diagrams. In the dilatometric experiments we observe a softening/melting of the FLL at the temperature $T_l \lesssim T_c$. The observed peak effect, thus occurs prior to the melting of the FLL which is similar to the situation in the cuprate superconductor YBa$_2$Cu$_3$O$_{7-\delta}$ [44], illustrating the far-ranging occurrence of the phenomenon. An interpretation in terms of a GFFLO state is not favorable, although, it would provide reason for an APE of different kind. However, it might be realized in systems that meet the stringent theoretical requirements. Most promising candidates for an experimental investigation are e.g. the quasi-2 dimensional cuprates where one can discard orbital effects by applying the magnetic field parallel to the superconducting layers [5].

The objectives for future research should, thus, be twofold: In lack of an explanation for the APE, further experimental investigations have to be considered to unravel the dynamics of the FLL in the region of enhanced pinning activity. E.g. in the system NbSe$_2$ it was pointed out, that the superconducting state is superimposed to a charge density wave, which also could produce unusual pinning properties [14].

On the other hand, the recent theoretical results have given serious constraints for an experimental investigation of a possible FFLO superconducting state in appropriate type-II superconducting systems, so that a systematic search seems to be a promising future task.

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