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## Unconventional vortex dynamics in the low-field superconducting phases of UPt<sub>3</sub>

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**Abstract.** – The flux dynamics at low magnetic fields in UPt<sub>3</sub> shows a clear distinction between the relaxation of bulk vortices and those close to the surface. In addition, in the high-temperature A-phase, vortices trapped in the bulk of the specimen after cycling it in a magnetic field creep out as expected, while bulk vortices in the B-phase remain strongly pinned, indicating that an intrinsic, novel pinning mechanism exists in the low-temperature superconducting phase of UPt<sub>3</sub>.

Since the discovery of superconductivity in UPt<sub>3</sub> one decade ago [1], many studies have focused on elucidating the nature of the superconducting state in this heavy-fermion compound. Based on specific heat measurements [2] as well as measurements of various other physical quantities, the unconventional ( $H$ - $T$ - $p$ ) phase diagram of UPt<sub>3</sub> is nowadays firmly established. It exhibits three distinct superconducting phases with two transitions at  $T_c^+$  and  $T_c^-$  at  $H = 0$  and  $p = 0$ . Several phenomenological theories have been proposed in order to explain this phase diagram as well as the various signatures of unconventional superconductivity in UPt<sub>3</sub>. A critical summary of them has been given by Joynt [3].

Recently, two microscopic measurements made it possible for the first time to differentiate between the two low-field superconducting phases of UPt<sub>3</sub>. Using point contact spectroscopy, Goll *et al.* [4] showed that gap-related features are only observed in the low-temperature B-phase of UPt<sub>3</sub>. In muon spin rotation-relaxation measurements at zero applied magnetic field, Luke *et al.* [5] detected an increase in the internal magnetic field that occurs only in the lower superconducting phase and which can be explained if the B-phase of UPt<sub>3</sub> is characterized by broken time-reversal symmetry.

Here we present a macroscopic type of measurement, vortex creep, which clearly distinguishes between different vortex dynamics in the two low-field superconducting phases of UPt<sub>3</sub>. In addition, we observe that vortices close to the surface decay from a metastable configuration in a different way than vortices in the bulk; the decay laws are different as well

as their temperature dependences. As far as we know, this is the first superconductor which displays such a distinct behaviour. Bulk vortices are so strongly pinned in the low-temperature, low-field superconducting phase (B-phase) that their creep rate is practically zero, while in the high-temperature, low-field superconducting phase (A-phase) we observe logarithmic creep rates that increase with temperature rather rapidly on approaching  $T_c^+$ .

The single crystal of  $\text{UPt}_3$  was prepared from arc-cast polycrystalline rods by zone melting in high vacuum. Laue X-ray diffraction analysis was used to orient the single crystal which was then cut into smaller pieces with a diamond wheel saw. The single crystal used in this investigation was annealed in vacuum at  $800^\circ\text{C}$  for 6 days and it is  $1.5 \times 2.9 \times 0.9\text{ mm}^3$  in size. The magnetic field was applied parallel and perpendicularly to the  $c$ -axis which is along the shortest dimension of the crystal. The sample was cooled in a  $^3\text{He}$ - $^4\text{He}$  mixture inside the mixing chamber of a dilution refrigerator. Temperature measurements were done with a cerium magnesium nitrate thermometer in the liquid  $^3\text{He}$ - $^4\text{He}$ , calibrated at higher temperatures with Ge resistors. In our experimental set-up the specimen remains stationary inside a superconducting flux transformer which is inductively coupled to an r.f.-SQUID sensor.

Isothermal relaxation curves of the remanent magnetization were taken after cycling the specimen in an external field. In all the decay measurements of  $M_{\text{rem}}$  reported here, the specimen is zero-field cooled to the desired temperature and then a field  $H$  is raised up to  $H^{\text{max}}$  in about 30 seconds and subsequently removed in about 1 second. The measurements of  $M_{\text{rem}}(t)$  start typically at  $t \simeq 1\text{ s}$  from the time when the applied field  $H$  reaches zero. Relaxation of the sample magnetization is measured typically in the time window  $1\text{ s} < t < 10^5\text{ s}$ . After a decay measurement, the specimen is heated above  $T_c^+$  and the expelled flux is recorded in order to obtain  $M_{\text{rem}}$  as the sum of the decayed flux plus the flux expelled during heating. We also measure  $\chi'$  and  $\chi''$ , the in-phase and out-of-phase components of the magnetic susceptibility with a mutual inductance bridge, using the SQUID as a null sensor. With this configuration we could detect changes in the susceptibility of the  $\text{UPt}_3$  crystal of the order of 1 p.p.m. of  $\Delta\chi'$  at  $T_c^+$ .

Decays of the remanent magnetization for two different cycling fields  $H^{\text{max}}$  are given in fig. 1a). By cycling the specimen in fields as small as  $H^{\text{max}} = 3.4\text{ Oe}$ , we probe only the dynamics of vortices very close to the surface. At bigger fields, the critical state is established and the decays reflect the motion of bulk vortices as well as surface vortices. We observe in fig. 1a) that the logarithmic decay rate  $|\partial \ln M / \partial \ln t|$  increases with time. For very low fields, the decays can be well fitted with a stretched exponential law of the form

$$M(t) - M(\infty) = [M(0) - M(\infty)] \exp[-(t/\tau)^\beta], \quad (1)$$

with typical values of the stretched exponent  $\beta$  of the order of 0.6–0.7 [6].

With increasing cycling fields, the critical state is reached and the decays show a non-zero logarithmic rate at short times which is not observed at very low fields. Since the time  $\tau$  in expression (1) does not lie in our time window ( $\tau > 10^5\text{ s}$ ), the fitting of the decays is not trivial. We have chosen to describe the decays with two parameters,  $S_{\text{initial}} = -\partial \ln M / \partial \ln t$  and  $\Delta M(10^4\text{ s})$ , the deviation from a pure logarithmic decay law at  $t = 10^4\text{ s}$ . We identify  $S_{\text{initial}}$  with the relaxation from bulk vortices and  $\Delta M$  with the decay from surface vortices. In fig. 1b) we give  $\Delta M(10^4\text{ s})/M_{\text{rem}}$  and  $M_{\text{rem}}$  as functions of the cycling field  $H^{\text{max}}$ . For the lowest fields, when only surface vortices are involved,  $\Delta M$  in the first  $10^4\text{ s}$  is as high as 38% of the trapped flux. This value is reduced dramatically to only 1% in the critical regime ( $H^{\text{max}} > 400\text{ Oe}$  for  $T = 450\text{ mK}$ ). Undoubtedly, the closer the flux is to the surface of the specimen, the stronger its decay is in a given time interval. A similar relaxation behaviour is observed at low fields for the penetration of vortices. We conclude that the stretched exponential decay corresponds mainly to the decay of the flux close to the surface.

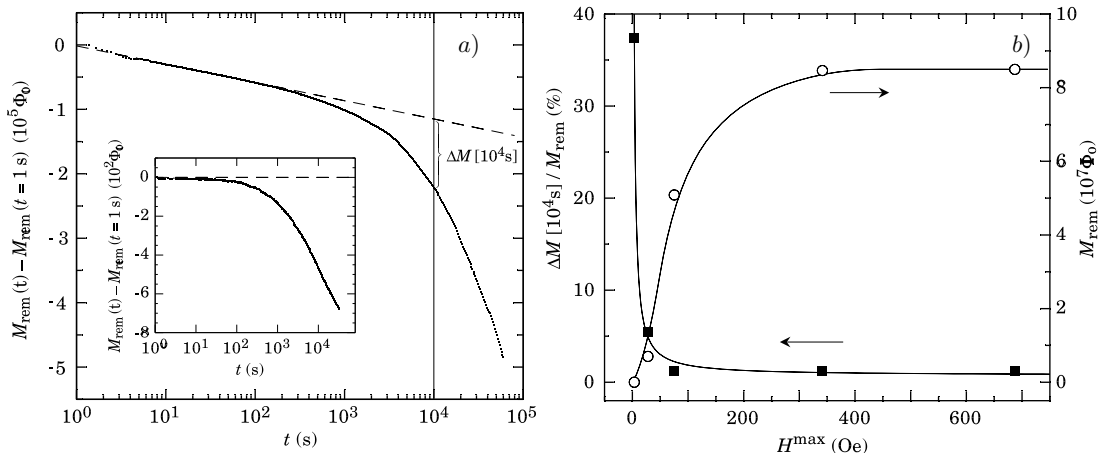


Fig. 1. – *a*) Relaxation curve of  $M_{\text{rem}}$  for the  $\text{UPt}_3$  crystal with  $H \parallel c$  at  $T = 450$  mK after a cycling field  $H^{\text{max}} = 680$  Oe. In the insert, relaxation curve for a smaller cycling field  $H^{\text{max}} = 3.4$  Oe. The remanent magnetization  $M_{\text{rem}}$  is given in the same arbitrary units for both decays (flux quanta  $\Phi_0$  at the sample). *b*) Remanent magnetization  $M_{\text{rem}}$  ( $\circ$ ) and ratio  $\Delta M(10^4 \text{ s})/M_{\text{rem}}$  ( $\blacksquare$ ) for the  $\text{UPt}_3$  crystal with  $H \parallel c$  at  $T = 450$  mK. Here  $\Delta M(10^4 \text{ s})$  represents only the stretched exponential part, *i.e.* the logarithmic part of the decay (dashed lines in fig. 1a)) has been subtracted. The drawn curves in fig. 1b) are only guides to the eye.

We have observed similar strong, stretched exponential decays of surface vortices in at least three different crystals of  $\text{UPt}_3$  in the two low-field superconducting phases with both  $H \parallel c$  and  $H \perp c$  [6] as well as in sintered  $\text{UPt}_3$  powder [7]. In all specimens, the values of  $\Delta M(10^4 \text{ s})/M_{\text{rem}}$  are independent of temperature for  $T/T_c \lesssim 0.5$ – $0.7$ , indicating that this type of diffusive decay results not only from thermal processes but also via a novel form of quantum tunneling.

Quantum tunneling of vortices has been observed at millikelvin temperatures in the high- $T_c$  superconductors [8] as well as in the organic superconductors [9]. Typically, the measured logarithmic creep rates  $|\partial \ln M / \partial \ln T|$  at  $T \rightarrow 0$  are of the order of 1%. These values agree well with the values estimated from the quantum collective creep theory [10]. According to this theory, strong quantum creep rates occur in superconductors with strong anisotropy  $1/\varepsilon$ , high normal-state resistivity  $\rho_n$ , and short coherence length  $\xi$ . A theoretical estimate of the quantum creep rate at  $T \rightarrow 0$  with the corresponding values of  $1/\varepsilon$ ,  $\rho_n$ , and  $\xi$  of  $\text{UPt}_3$  shows that for this superconductor the logarithmic creep rates should be about 1000 times *weaker* than the rates in the high- $T_c$  superconductors. The different relaxation law observed in  $\text{UPt}_3$ , stretched exponential instead of the usual logarithmic or power laws, as well as the strength of the relaxation in a given time interval (38% in  $10^4$  seconds for a cycling field  $H^{\text{max}} = 3.4$  Oe at  $T = 450$  mK) cannot be explained with the existing theories of quantum creep.

It has been suggested by Sigrist, Rice and Ueda [11], [12] that one way to probe an unconventional superconductor is by investigating surface effects. Similar to the A-phase in superfluid  $^3\text{He}$ , where walls have a very significant macroscopic effect [13], one can expect in non-s-wave pairing superconductors a strong influence of boundaries on the order parameter. Our observation of anomalous, giant creep of vortices close to the surface with a stretched exponential decay law may have its origin in the unconventional nature of the order parameter in both superconducting phases.

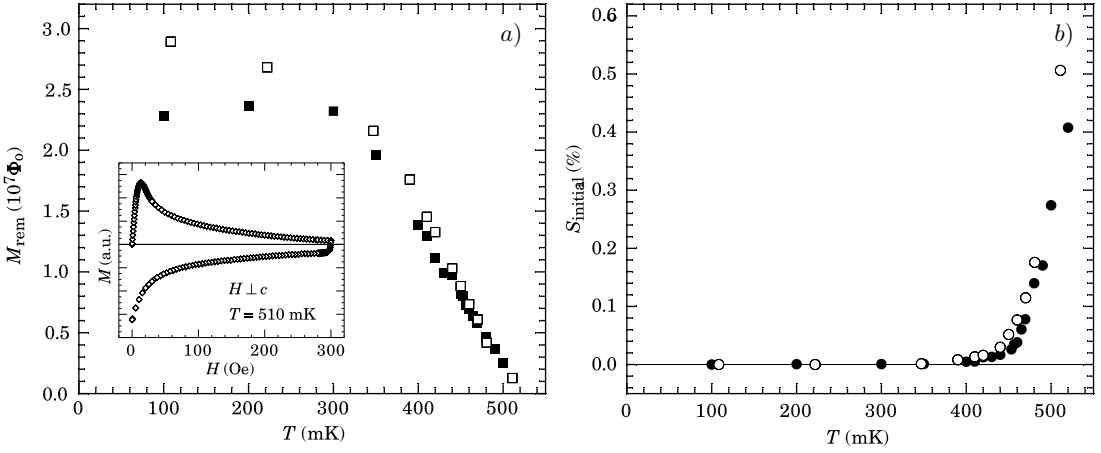


Fig. 2. – a) Saturated remanent magnetization  $M_{\text{rem}}$  vs.  $T$  for  $H \parallel c$  (■) and  $H \perp c$  (□). The insert of this figure shows a typical magnetization curve. b) Initial logarithmic decay rate  $S_{\text{initial}} = |\partial \ln M / \partial \ln t|$  vs.  $T$  for  $H \parallel c$  (●) and  $H \perp c$  (○).

The dynamics of bulk vortices in  $\text{UPt}_3$  also shows special features which are not observed in any other superconductor. In fig. 2a) we show values of  $M_{\text{rem}}$  as function of temperature for  $H \parallel c$  and  $H \perp c$ ; all the points in this figure have been taken with the  $\text{UPt}_3$  crystal cycled to sufficiently high fields, such that the remanent magnetization was independent of the cycling field  $H^{\text{max}}$ . In fig. 2b) we show the initial logarithmic creep rate of  $M_{\text{rem}}$  as a function of temperature. As seen in fig. 2b), for both field directions, the creep rate  $|\partial \ln M / \partial \ln t|$  is practically zero ( $|\partial \ln M / \partial \ln t| < 10^{-4}$ ) up to about 400 mK. Around this temperature it starts increasing slightly and then rapidly reaching a value of  $5 \cdot 10^{-3}$  close to  $T_c^+$ .

The very strong pinning of bulk vortices with an almost zero creep rate in the low-temperature, low-field phase of  $\text{UPt}_3$  cannot be the result of extrinsic quenched disorder. If this were the case, one could not explain the clear change in creep rates that occurs around 400 mK and the increase of the creep rates with temperature in the high-temperature A-phase. The strong reduction of the *bulk* creep in the B-phase is an intrinsic property and probably related to the nature of the order parameter in this phase. In a phase that breaks time reversal symmetry, vortices can be trapped on domain walls between domains of degenerate superconducting phases and decay into fractional vortices which can only exist on domain walls. In this way vortices can be pinned very strongly in a network of domain walls so that ordinary creep is substantially reduced [12], [14].

In fig. 3 we show the a.c. susceptibility for  $H \parallel c$  measured with an amplitude  $H_{\text{ac}} = 6.6$  mOe in the residual d.c. field of the cryostat  $H_{\text{dc}}^{\text{res}} < 2$  mOe. The transition into the superconducting state occurs at  $T_c^+ = 528$  mK and has a width  $\Delta T_c^+ = 11$  mK. Here  $T_c^+$  has been taken as the middle point and  $\Delta T_c^+$  with the 10–90% criteria. At lower temperatures and well separated from the transition at  $T_c^+$ , we have been able to detect an extremely weak peak in  $\chi''$  below  $T = 480$  mK as shown in the insert of fig. 3b). This second dissipation peak is 300 times smaller than the peak in  $\chi''$  at  $T_c^+$ . Between 16 Hz and 160 Hz, this small peak does not depend on frequency. While the position and width of the peak in  $\chi''$  at  $T_c^+$  are practically independent of the amplitude  $H_{\text{ac}}$  of the measuring field between 1.6 mOe and 33 mOe (insert in fig. 3a)), the small peak moves rapidly towards lower temperatures and widens with increasing  $H_{\text{ac}}$  as shown in the inserts of fig. 3a) and b). It is clear that the strong reduction in temperature of

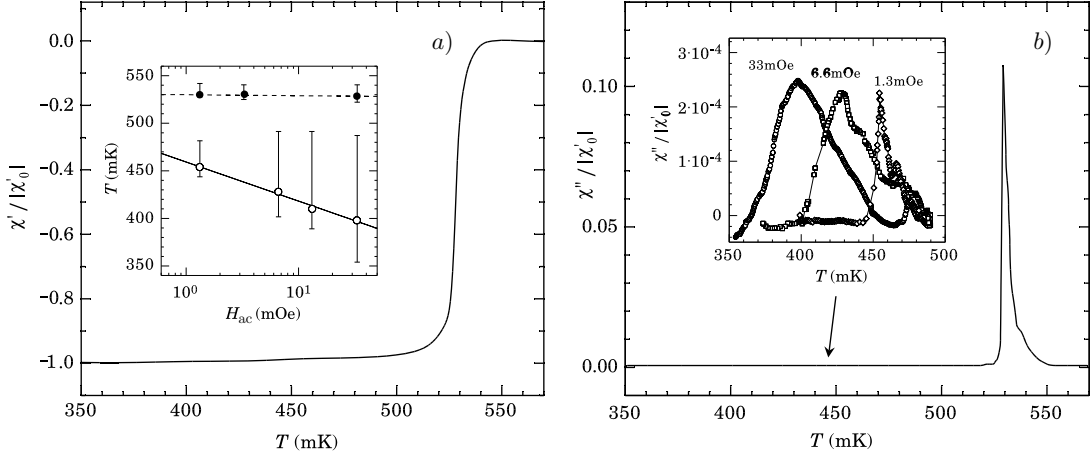


Fig. 3. – A.c. susceptibility of the UPt<sub>3</sub> crystal for  $H \parallel c$ ; in-phase component  $\chi'$  (a)) and out-of-phase component  $\chi''$  (b)). Here  $\chi'_0$  denotes the value of  $\chi'$  at  $T \rightarrow 0$ . The insert of b) is a close-up of the data around  $T = 450$  mK taken at different a.c.-field amplitudes. The insert of a) shows the position of the peaks at  $T_c^+$  (●) and around  $T \simeq 450$  mK (○) with their corresponding widths.

the peak's maximum with field ( $\sim 50$  mK/30 mOe) does not follow the field dependence of the phase boundary between the A and B phases. The onset of dissipation, on the other hand, seems to be independent of  $H_{ac}$  and it occurs at  $T \simeq 480$  mK. Based on the established phase diagram of UPt<sub>3</sub>, we propose to identify the onset of dissipation with  $T_c^-$ .

The second peak around  $T_c^-$  at temperatures where the specimen is fully superconducting has no trivial interpretation. Probably it is related to hysteresis losses which are not present at temperatures above  $T_c^-$ . Dissipative processes can arise due to the build up of domains separated by domain walls in the low-temperature superconducting phase of UPt<sub>3</sub> as well as spontaneous currents and vortices [12], [14]. Domain walls at boundaries of the specimen can act as “channels” for entry or exit of flux. These domain walls can trap fractional vortices that —unless they recombine to give an integer flux quantum— can only move along these walls [12], [14]. Within this tentative picture, dissipation could arise from motion of such vortices and/or domain walls. On further reducing the temperature, the domain walls and fractional vortices are more strongly pinned, so that the maximum in  $\chi''$  is pushed towards lower temperatures for higher  $H_{ac}$  amplitudes.

Summarizing, we present here an investigation of relaxation of the remanent magnetization which shows that the low-field flux dynamics in UPt<sub>3</sub> is different than in any other superconductor. Vortices in the bulk relax from a metastable configuration following a logarithmic decay law while vortices close to the surface relax via a stretched exponential decay law. The bulk relaxation rate is clearly different in the two low-field superconducting phases. In the low-temperature B-phase, the logarithmic creep rate is practically zero while it is finite in the high-temperature A-phase and increases rapidly as the temperature is increased towards  $T_c^+$ . Moreover, a considerable amount of flux penetrates into the specimen at very low cycling fields in both superconducting phases. This flux relaxes very rapidly, for example, at  $T = 450$  mK up to about 38% of the trapped flux decays in the first  $10^4$  seconds. Its relaxation is complex and similar to “glassy” relaxation. Undoubtedly, the anomalous low-field penetration mode in UPt<sub>3</sub> and its unusual dynamics have an effect on surface measurements of the magnetic penetration depth  $\lambda$  and on its temperature dependence. It might also be responsible for the

anomalous Meissner effect of  $\text{UPt}_3$  often reported in the literature. Certainly more work is needed in order to completely understand the vortex phases and their dynamics in  $\text{UPt}_3$ .

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