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Magnetization Study of the Superconducting State of UPt_3

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High-resolution *static* magnetization measurements have been performed on high-quality single crystals of UPt_3 at temperatures down to 50 mK and in fields up to 40 kOe, with an attention being paid on the paramagnetic response of the superconducting mixed state. At 50 mK, a small but distinct anisotropy develops in the equilibrium magnetization M_{eq} below the upper critical field H_{c2} which cannot be explained by the effective mass anisotropy nor by the ordinary paramagnetic effect of a spin-singlet pairing. In particular, the discontinuity in dM_{eq}/dH at H_{c2} strongly depends on field direction at low temperature, being vanishingly small for H parallel to the hexagonal basal plane where the normal state spin susceptibility is largest. The results strongly suggest an odd-parity pairing with an appreciable anisotropy in the pair-spin correlation. The features of the magnetization across the B-C phase transition, as well as the irreversibility peak in the magnetization near H_{c2} ("peak-effect"), are also discussed.

KEYWORDS: UPt_3, superconductivity, magnetization measurement, paramagnetic effect, peak effect, triplet pairing

§1. Introduction

Superconductivity in UPt_3 has been attracting much interest because a realization of unconventional order parameters is evident from its complex field-temperature (H-T) phase diagram with three different vortex states (A, B and C phases).[1-7] The phase transitions between these phases have been studied by various methods such as the specific heat,[2,7] magnetocaloric effect,[6] magnetostriction,[5] and acoustic measurements.[3,4] It is now recognized that all these phase boundaries meet at a tetracritical point, irrespective of the direction of an applied field.

An important issue concerning the order parameter of UPt_3 is a possibility of an odd-parity (pseudo-spin triplet) pairing, which has been inferred from a few experimental results. The most crucial one is the NMR Knight shift measurements.[8,9] Recent experiment on high-quality single crystals revealed no change in the ^{195}Pt Knight shift below T_c for any field direction,[9] in striking contrast to the case of a spin-singlet pairing. Another evidence can be seen in the temperature variation of H_{c2}.[10] The absence of the paramagnetic limit for H perpendicular to the hexagonal c-axis ($H \perp c$) is inexplicable by a singlet pairing scenario.[11,12] Several theoretical models have been proposed so far to explain the unprecedented superconductivity of UPt_3,[11-15] though no consensus has been reached yet on the pairing symmetry.

In further elucidating the pairing in UPt_3, it may be of interest to examine the equilibrium magnetization M_{eq} in the superconducting mixed state. In general, there is a contribution to M_{eq} from a paramagnetic polarization of the system, in addition to the diamagnetic orbital currents around the vortices. As is well known, the normal state paramagnetic susceptibility χ_n of UPt_3 is large and anisotropic,[16] with the easy direction being $H \perp c$. Since a substantial part of χ_n is considered to come from the pseudo-spin Pauli paramagnetism,[9,17] field variation of M_{eq} near H_{c2} may strongly depend on the pairing symmetry. In case of an even-parity pairing, for example, the spin polarization should be suppressed below H_{c2} for *all* directions.[18,19] This would lead to a sizable discontinuity in dM_{eq}/dH at H_{c2} especially for $H \perp c$.[19] If this anomaly, the paramagnetic effect, is absent, it would then be a strong implication of the odd-parity states.[20]

This approach is indeed complementary to the NMR Knight shift experiment. It should be noticed, however, that the above NMR results might not be fully compatible with the interpretation of the cross-over[11] in the anisotropy ratio $H_{c2}^{\perp}/H_{c2}^{//}$; the latter assumes $H_{c2}^{//}$ (parallel to the c-axis) to be *paramagnetically limited* at low T. While the NMR experiment strongly suggests that the pair-spin is free to rotate towards the field direction (equal-spin pairing with weak spin-orbit coupling),[9] the anisotropy cross-over in H_{c2} seems to point to a presence of the strong spin-orbit coupling with the pair-spin confined in the basal plane.[12] Whether pair-spin anisotropy is weak or strong deeply concerns the identification of the internal degrees of freedom of the pairing function. In order to clarify this point, it would thus be highly worthwhile to inspect the bulk magnetization of UPt_3 near H_{c2}.

In this paper, we have examined $M_{eq}(H)$ of UPt_3 by means of high-resolution DC magnetization [$M(H)$] measurements on high-quality single crystals, focusing mainly on the paramagnetic response of the high field state (C phase). The results in fact provide a compelling evidence of an odd-parity state, with a small anisotropy in the spin response. It was also our objective to explore the B-C transition by the magnetization measurements. In the course of the experiment, we observed that the magnetization hysteresis exhibits a non-monotonous field variation, taking a pronounced peak just below H_{c2}. This kind of phenomenon, the "peak effect", has recently been drawing attention in reference to a formation of the new vortex state theoretically predicted for spin-singlet superconductors in high fields.[21-28] The implication of the phenomenon in UPt_3 is also discussed.

§2. Experimental Procedures

Two single crystals of UPt_3, sample 3-S and sample 4, were grown by the Czochralski pulling method in a tetra-arc furnace.[29] The starting materials were 99.95% pure U and 99.99% pure Pt. A typical ingot was 3-4mm in diameter and 60mm in length. The ingots were heated by a DC current flow with a density of $1000 A/cm^2$ through the crystal rod and kept at 1200°C for 6 days, in high vacuum of 8×10^{-10} Torr. The crystals were then gradually cooled down to room temperature, taking over 10 days. A clear double superconducting transition was observed in a specific heat measurement. The upper critical temperature T_c was ~520mK. The resistivity ratio $\rho(300K)/\rho(T)$ extrapolated to T=0 from above T_c was in excess of 500 for both samples, indicating excellent quality of the crystals. The crystals were shaped to $2.5\times2.5\times3 mm^3$ (sample 3-S) and $1\times1\times3 mm^3$ (sample 4), with the long axes oriented to the c axis.

Magnetization curves at temperatures ranging from 50mK to above T_c were obtained by a Faraday force magnetometer installed in a dilution refrigerator,[30] in a field gradient of 500~800 Oe/cm produced by independent gradient coils. The magnetic force acting on the sample was detected by a force-sensing parallel plate capacitor. One of its electrodes, on which the sample is mounted, is suspended by thin phosphor-bronze wires, and can move flexibly in the way that the gap varies in proportion to the applied force. The capacitance change is read by a three-terminal autobalance capacitance bridge whose resolution is ~5×10^{-7} pF. Load resolution of better than 10^{-7} N was obtained. By use of the high-sensitivity force-sensing device, the overall displacement of the sample was limited to less than 1μm; there was virtually *no* fluctuation in the magnetic field experienced by the sample. This point is very important in measuring true hysteretic magnetization of the vortex states. Each measurement was carried out after zero-field cooling the sample from above T_c to the desired temperatures. Considering the flux line relaxation, the sweep rate of H was fixed constant (5 Oe/sec) throughout the measurements.

§3. Results and Discussion

3.1 *Magnetization curves and the H_{c2} phase diagrams*

Figure 1 shows the $M(H)$ curves of sample 3-S for both field directions, measured at 50mK. As will be estimated later, the lower critical field H_{c1} of UPt_3 is of the order of ~10 Oe. Because of the small value of H_{c1}, the Meissner region was not well resolved in the magnetization process. The strong irreversibility appearing at low field is due to the ordinary flux pinning effect in the vortex state. The hysteresis rapidly decreases as H increases. The linear magnetization above H_{c2} is due to the normal state paramagnetism, from which we obtain $\chi_n^{//}=5.8\times10^{-6}$ emu/g and $\chi_n^{\perp}=1.1\times10^{-5}$ emu/g at 50mK. It is well known that χ_n of UPt_3 is strongly temperature dependent.[16] For instance, $\chi_n^{\perp}$ first increases on warming in proportion to ~T^2 and takes a broad maximum at around 20K, followed by a Curie-Weiss law at higher temperatures. Within the temperature range of our interest ($T\leq T_c$), however, the thermal variation is small and χ_n does not differ much from the values given above.

The $M(H)$ curves of sample 4 obtained at 50mK are dis-

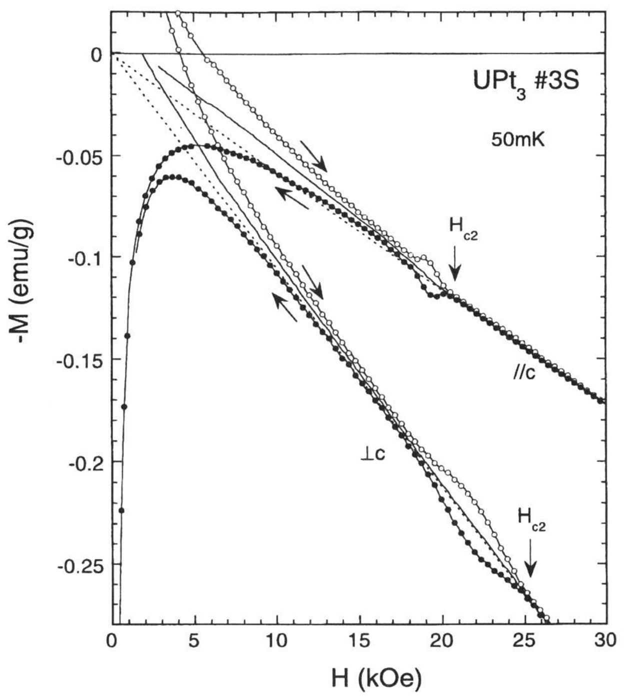

Fig. 1. Magnetization curves of UPt_3 sample 3-S for H parallel and perpendicular to the c-axis. The arrows indicate directions of the field scan. Thin solid lines denote the equilibrium magnetization in the vortex state determined as the average of the field-increasing and decreasing data, while the dotted lines are the extrapolated normal state magnetization. The upper critical field H_{c2} is defined as the field where the magnetization hysteresis disappears. The pronounced peak in the magnetization hysteresis appearing just below H_{c2} is the peak effect, whose onset field is defined as H^*.

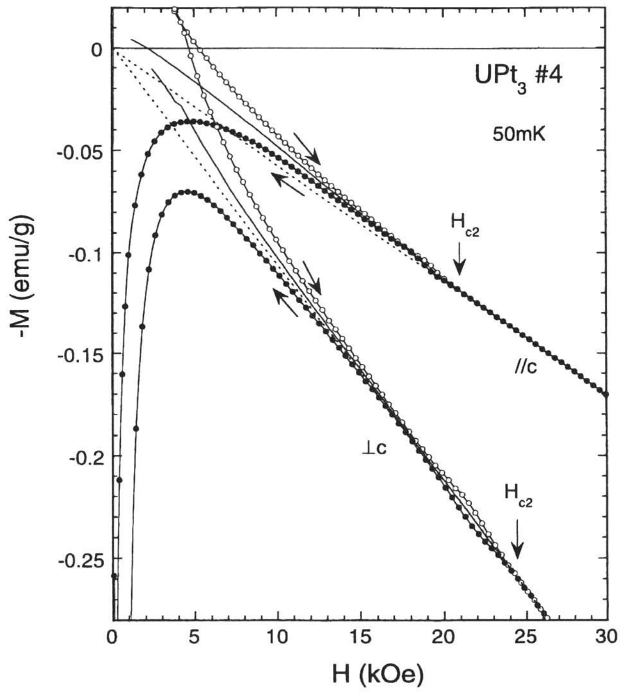

Fig. 2. Magnetization curves of UPt_3 sample 4 for H parallel and perpendicular to the c-axis.

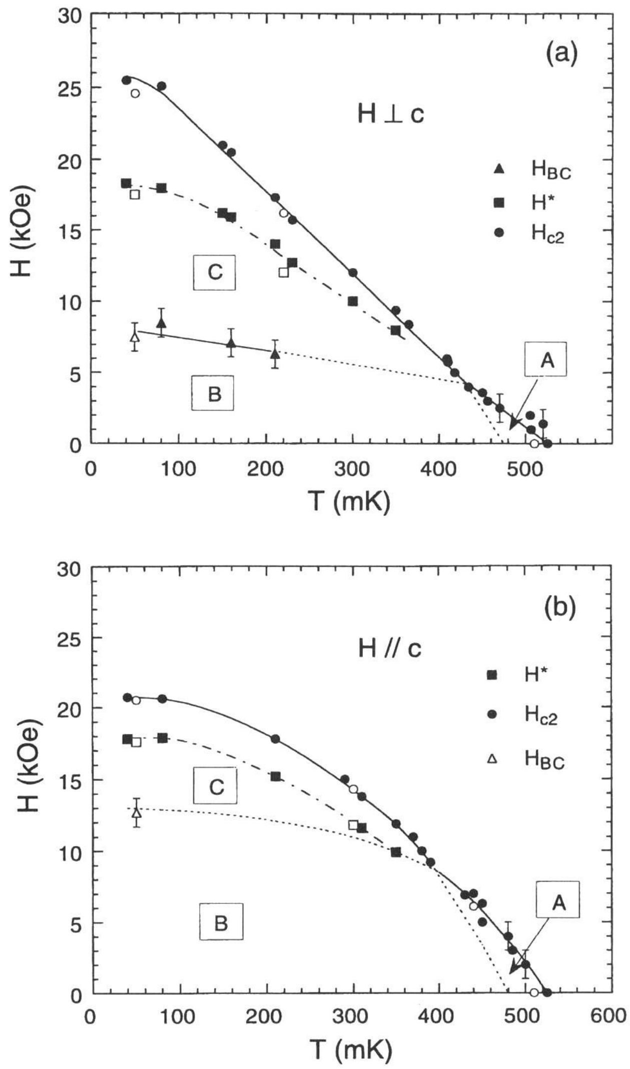

Fig. 3. *H-T* phase diagrams of UPt_3 for $H \perp c$ (a) and $H // c$ (b), obtained from the magnetization measurements. The onset field of the peak effect H^* is also shown. The open and closed symbols indicate the results for sample 3-S and 4, respectively. The dotted lines are the phase boundaries which are not observed in the present experiment.

played in Fig.2. The results are quite similar to those of sample 3-S, except for the magnitude of the hysteresis which linearly depends on the sample dimension and is therefore smaller in sample 4. In fact, the *M(H)* curves of sample 4 become almost reversible at around $H \sim 17$kOe, well below H_{c2}.

In both samples and for both field directions, we observed that the irreversibility in *M(H)* increases again in a narrow region just below H_{c2}.[21] This is a so-called "peak effect", which will be discussed in §3.2. Hereafter, we define H_{c2} as the field where the irreversibility peak vanishes. We show the resulting H_{c2} vs. *T* plots in Figs. 3(a) and 3(b), which are in good agreement with those previously determined by other experimental methods.

When the magnetization hysteresis is small, the equilibrium magnetization M_{eq} of the vortex state can be well approximated by the average of the increasing- and decreasing-field data.[31] The results for M_{eq} are shown in Figs. 1 and 2 by thin solid lines. Similar results for M_{eq} are obtained for both samples. Also shown by the dotted lines are the extrapolated normal state magnetization $M_n^{(//,\perp)}=\chi_n^{(//,\perp)}H$. Evidently, the paramagnetic contribution to M_{eq} is quite large for both field directions. It is very important to notice that the difference between $M_{eq}^{\perp}$ and $M_n^{\perp}$ is vanishingly small near H_{c2}, whereas in $M_{eq}^{//}$ there is a small but abrupt deviation from $M_n^{//}$ below H_{c2} (Fig.2).

3.2 Peak effect

As shown in Figs.1 and 2, the irreversibility in *M(H)* increases again in a narrow region just below H_{c2}. This implies that the flux pinning is enhanced in this region. This phenomenon is known as the "peak effect", which is occasionally observed in type-II superconductors.[31,32] The origin of the peak effect is, however, not always well resolved even in the conventional superconductors, although a certain sort of cross-over in the flux pinning mechanisms is considered to be relevant.[32]

Recently, similar phenomena have been observed in some heavy electron superconductors,[21-24] and discussed in reference to the Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) phase[25-28] which was theoretically predicted to occur for the *spin-singlet* superconductors having a large normal state spin susceptibility. In this phase, the pairing takes place so as to gain the spin Zeeman energy, at the cost of the condensation energy by partially destroying the singlet pairs. As a result, the gap function begins to possess nodes in the real space.[26] Recent theoretical treatment[28] shows that in such case the flux pinning force could be enhanced, since the flux lines become somewhat flexible and thereby have more chances to match to the randomly distributed pinning centers. The predicted FFLO phase should be observable only at low temperatures and high fields, where the pair breaking by the Zeeman energy becomes important. It is separated from the ordinary vortex state by a first order phase boundary, which meets the H_{c2} line at a certain critical temperature expected to locate at $\sim 0.6T_c$ for the s-wave superconductors in the strong paramagnetic limit.[27,28] Across the transition, there should be a jump in the equilibrium magnetization.[27] These points distinguish the FFLO phase from the ordinary peak effect whose occurrence is not always reserved to the high field regions.[32]

In order to show the temperature dependence of the peak effect in UPt_3, we plot the difference M_n-M_{up} in Figs. 4(a) ($H \perp c$) and 4(b) ($H // c$), where M_{up} denotes the magnetization data in the field-increasing scans. There can be clearly seen an irreversibility peak at low temperatures, which becomes weak with increasing temperature and vanishes near 400mK for both directions. The onset field H^* of the peak effect in UPt_3 is also shown in Figs.3(a) and 3(b), whose behavior is somewhat similar to what is predicted for the FFLO phase.[27,28]

Nevertheless, the observed peak effect in UPt_3 cannot be simply related to the FFLO state because of the following reasons. First, there is no evidence of the first-order transition at H^*; neither a discontinuity in M_{eq} nor a hysteresis in H^* has been observed. Second, although the peak effect is clearly seen for $H \perp c$, the paramagnetic effect is apparently absent in $M_{eq}^{\perp}$ as will be discussed in §3.4. In fact, we can show that H^* in the basal plane well exceeds the paramagnetic limiting field H_P that is defined for a singlet pairing state as $\chi_n H_P^2/2 = H_c^2/8\pi$. It seems that the Zeeman energy is not pair breaking in the vortex state for this direction. This would contradict the basic assumption of the FFLO state. These facts might indicate that the peak effect in UPt_3 is of "conventional type".[32] However, there is much to be done before identifying the origin of the peak effect in UPt_3. For instance, we need to answer the question why the peak effect is observable only in the C-phase. These are left to be clarified

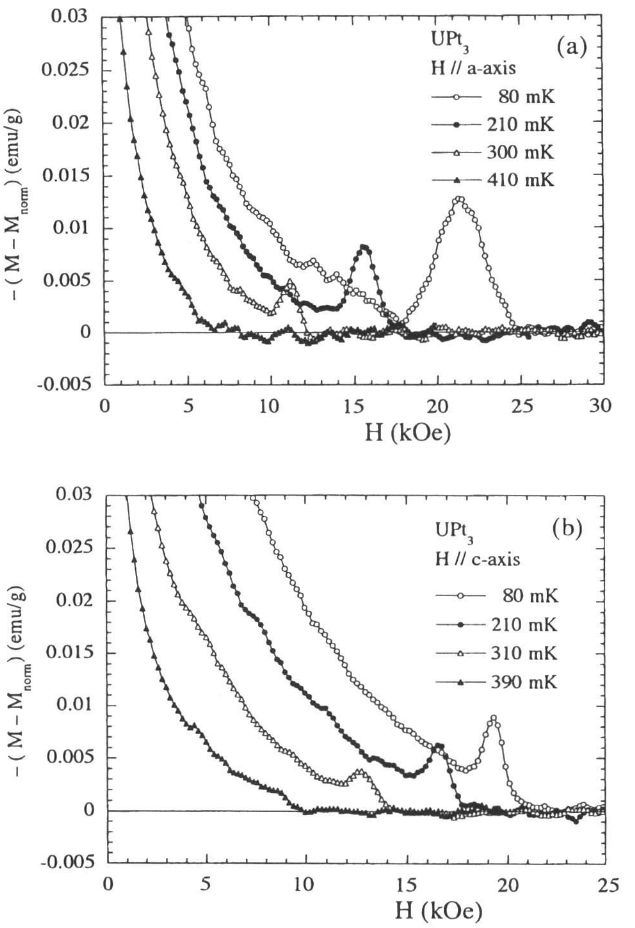

Fig. 4 Magnetization difference M_{up}-M_n of UPt_3 for $H\perp c$ (a) and $H//c$ (b) at various temperatures, where M_{up} denotes the magnetization data in the field-increasing scan. The irreversibility peak can be clearly seen near H_{c2}, whose onset field H^* is indicated by the arrows.

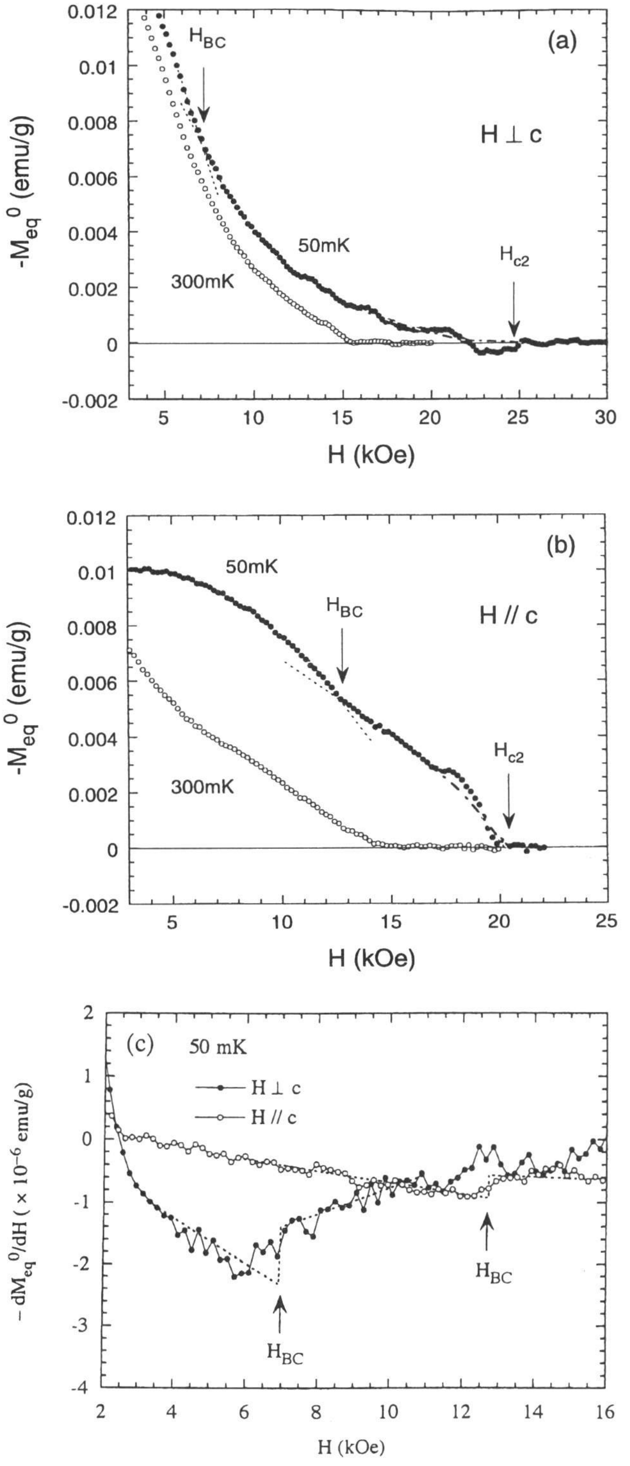

Fig. 5. Magnetization difference M_{eq}^0=M_{eq}-M_n of UPt_3 for $H\perp c$ (a) and $H//c$ (b). The dot-dash lines indicate the expected variation of M_{eq}^0 near H_{c2}. The B-C transition field is shown by the arrows, where a slight change of the slope in M_{eq}^0 is observed. (c): differential susceptibility $M_{eq}^0/\partial H$ at 50mK for both directions. A small jump is observed across the B-C transition, as indicated by the dotted lines.

in the future.

3.3 *Magnetization anomaly across the B-C transition*

From the calorimetric measurements[6,7] it is established that the B-C transition is of second order. We may then expect a change in the slope of M_{eq} across the transition. In Figs. 1 and 2, the B-C phase boundary is expected to locate at $H_{BC}\approx$7kOe ($\perp c$) and $\approx$13kOe ($//c$). Within the resolution of these plots, however, no anomaly is visible in M_{eq} or M at these fields. The change in the magnetization, if any, should be very small.

The behavior of M_{eq} becomes more clear by plotting the magnetization difference M_{eq}^0=M_{eq}-M_n in an enlarged scale in Figs. 5(a) and 5(b), where the results at 300mK are also shown for comparison. The small humps or dips seen in the curves for 50mK near H_{c2} are probably due to the peak effect; hysteresis might not be cancelled out well there, probably due to a non-linear distribution of the vortex lines.[31] It is rather natural to assume a smooth variation of M_{eq}^0, as shown by the dot-dash lines. At 50mK, we can recognize a slight change in the slope of M_{eq}^0 at H_{BC}, as indicated in the figures.

The discontinuity in the differential susceptibility $\Delta(\partial M_{eq}^0/\partial H)$ across the B-C transition can be evaluated from the thermodynamic relations (Ehrenfest relations) that hold for any second order phase transition:

$$\Delta\left(\frac{\partial S}{\partial T}\right)+\Delta\left(\frac{\partial S}{\partial H}\right)\frac{dH_{BC}}{dT}=0, \tag{1a}$$

$$\Delta\left(\frac{\partial M_{eq}^0}{\partial T}\right)+\Delta\left(\frac{\partial M_{eq}^0}{\partial H}\right)\frac{dH_{BC}}{dT}=0. \tag{1b}$$

Here, S is the entropy and ΔX denotes the discontinuity in the quantity X across the transition. Making use of the Maxwell relation $\partial M_{eq}^0/\partial T=\partial S/\partial H$, we obtain

$$\Delta\left(\frac{\partial M_{eq}^0}{\partial H}\right)=\left(\frac{dH_{BC}}{dT}\right)^{-2}\Delta\left(\frac{C}{T}\right). \qquad (2)$$

As $\partial S/\partial H$ always tends to vanish in the limit $T\rightarrow 0$, so does $\Delta(C/T)$ as can be seen from eq.(1a). Nevertheless, $\Delta(\partial M_{eq}^0/\partial H)$ could remain finite. Thus the B-C transition might be better studied by the magnetization measurements. From the specific heat measurement, a small discontinuity of order $\Delta(C/T)=$ 1~2×10^4erg/mole·K^2 has been reported across the B-C transition at T=150mK.[7] Noting $dH_{BC}/dT\sim 4\times10^3$Oe/K around this temperature,[4] we obtain $\Delta(\partial M_{eq}^0/\partial H)\sim 2\times10^{-7}$ emu/g.

Figure 5(c) shows the differential susceptibility $\partial M_{eq}^0/\partial H$ for both directions at 50mK, where we could ascertain a discontinuity of this order as indicated by the dotted lines. The critical field H_{BC} thus obtained is also plotted in Figs. 3(a) ($H\perp c$) and 3(b) ($H//c$). Unfortunately, however, the anomaly in M_{eq}^0 at H_{BC} was smeared with increasing temperature, and could not be traced up to the tetracritical point. We also note that no appreciable change was observed in M_{eq}^0 across the A-B transition.

3.4 *Paramagnetic response in the vortex state*

Now we turn to the paramagnetic effect in the magnetization process. A salient feature of M_{eq}^0 in Figs. 5(a) and 5(b) at 50mK is the marked anisotropy near H_{c2}. There is an order of magnitude difference in the slope of $M_{eq}^0{}_{(//,\perp)}$ at H_{c2}. Moreover, the curvatures of $|M_{eq}^0{}_{(//,\perp)}|$ are different; downward (upward) for $H//c$ ($H\perp c$). In general, M_{eq}^0 near H_{c2} can be expressed in terms of a Ginzburg-Landau (G-L) parameter κ_2 as[18,33)]

$$M_{eq}^0=(H-H_{c2})/[4\pi\beta_A(2\kappa_2^2-1)]\ , \qquad (3)$$

where β_A is a number of order unity that depends on the vortex lattice configuration. From the results in Figs. 5(a) and 5(b), we obtain highly anisotropic values of κ_2 at T=50mK, $\kappa_2^\perp\sim140$ and $\kappa_2^{//}\sim40$; i.e., $\kappa_2^\perp/\kappa_2^{//}\approx3.5$. Here, the κ_2 values were determined from the average slope of M_{eq}^0 between H^* and H_{c2}. Due to the upward curvature of $M_{eq}^0{}_\perp$, the actual value of $\kappa_2^\perp$ and hence the anisotropy ratio could be even larger.

In a conventional superconductor, κ_2 could be direction dependent reflecting the effective mass anisotropy. Although a theoretical treatment of the mass anisotropy in κ_2 seems to be lacking, we may assume that the anisotropy ratio can be scaled by that of the G-L parameter κ. For the uniaxial symmetry, the anisotropy ratio of κ can be approximated as $\kappa^\perp/\kappa^{//}\approx(H_{c2}^\perp/H_{c2}^{//})_{T\rightarrow Tc}$.[34] From the H_{c2} data for UPt$_3$[10], we obtain $\kappa^\perp/\kappa^{//}\sim0.6$; the anisotropy is opposite to what is observed. Evidently, the mass anisotropy does not explain the anisotropy in κ_2 at low T. The low temperature anisotropy in M_{eq}^0 is not likely to be caused by the anisotropic orbital currents; it is rather of spin origin.

It should be noticed that the anisotropy in M_{eq}^0 reverses at low field as can be seen for the curves at 50mK in Figs .5(a) and 5(b); $M_{eq}^0{}_{,\perp}$ becomes larger than $M_{eq}^0{}_{//}$ below H~6kOe. Although the physical implication of this cross-over is not apparent, it is consistent with the thermodynamic constraint

$$H_c^2/8\pi=-\int_0^{H_{c2}}M^0_{eq,(//,\perp)}dH, \qquad (4)$$

where H_c is the *direction independent* thermodynamic critical field. Extrapolating $M_{eq}^0{}_{(//,\perp)}$ to lower fields, we could obtain $H_c(T\ll T_c)\approx2600$Oe for both curves, in agreement with the value that can be estimated from the specific heat data.[2] These facts confirm that the observed anisotropy in M_{eq}^0 is indeed correlated with that in H_{c2}. It is very important to point out, however, that the paramagnetic energy $\chi_n H_{c2}^2/2$ significantly exceeds the condensation energy $H_c^2/8\pi$ by a factor of ~10 ($//c$) and ~25 ($\perp c$). The paramagnetic polarization in the vortex state is essentially large for both directions.

The upward curvature in $|M_{eq}^0{}_\perp|$ with vanishingly small change of the slope at H_{c2} is specific to a clean superconductor *in the absence of* the paramagnetic effect.[35] This implies that the orbital current contribution is predominant in $M_{eq}^0{}_\perp$. By contrast, the downward curvature of $|M_{eq}^0{}_{//}|$ with a sharp change of slope at H_{c2} is the typical behavior of ordinary paramagnetic effect[18,36,37]; a reduction in the spin polarization by pairing enhances $|M_{eq}^0|$ below H_{c2}. This strongly suggests that the spin susceptibility for $H//c$ is somewhat reduced in the C phase of UPt$_3$. Note that these features in M_{eq}^0 become weaker with increasing temperature, as can be seen in Figs. 5(a) and 5(b).

Anisotropic paramagnetic effect itself is not unusual. For instance, in ErRh$_4$B$_4$ where ferromagnetic interactions between Er ions compete with superconductivity of conduction electrons, a strong paramagnetic effect is observed in the magnetization for the spin-easy axis ($H//a$) at low temperatures, while the effect is weak for the hard axis ($H//c$).[37] The crucial point in UPt$_3$ is that the paramagnetic effect in M_{eq}^0 is apparently absent for the direction ($H\perp c$) where the normal state spin susceptibility is *largest*. This is the opposite of what is expected for a clean spin-singlet superconductor. In other words, the spin polarization in the C phase for $H\perp c$ is as large as that in the normal state. It is important to note that the spin-orbit scattering mechanism is irrelevant in this case. In the presence of strong spin-orbit scattering, spin paramagnetism might be recovered in the singlet pairing states.[38] This mechanism, however, would be essentially isotropic and therefore fails to explain the slight but appreciable paramagnetic effect for $H//c$. It should also be noticed that the present samples are in the clean limit; the mean free path of the carriers can be estimated from the residual resistivity ($\rho_{0//}\approx0.2\mu\Omega$cm) to be over 2000Å, much longer than the coherence length of $\xi_0\sim120$Å estimated from the H_{c2} values.

Temperature variation of κ_2 evaluated from M_{eq}^0 is summarized in Fig. 6, which further confirms the above arguments. The anisotropy ratio of κ_2 decreases to $\kappa_2^\perp/\kappa_2^{//}\approx0.8$ at ~450 mK, approaching the value ~0.6 expected from the mass anisotropy. In general, κ_2 coincides with κ as $T\rightarrow T_c$; we thus have $\kappa^\perp\sim50$ and $\kappa^{//}\sim60$. From these κ values and $H_c\sim2600$Oe, we obtain a very small value of the lower critical field $H_{c1}\approx\sqrt{2}H_c\ln\kappa/(2\kappa)$ ~10Oe. Remarkably, $\kappa_2^\perp(T)$ continues to *increase* on cooling without an indication of saturation. This is actually the behavior predicted for a clean superconductor *in the absence of* the paramagnetic effect,[39,40] where $\kappa_2\propto\sqrt{\ln(T_c/T)}$ as $T\rightarrow0$ as shown by the dotted line. By contrast, $\kappa_2^{//}$ continues to *decrease* on cooling; the typical behavior *in the presence of* the paramagnetic effect[18] where the pair breaking by the Zeeman energy becomes important at low temperatures and high fields. As a result, there is a cross-over in the anisotropy ratio $\kappa_2^\perp/\kappa_2^{//}$ at around $T\sim0.7T_c$. Except for this cross-over, the sign reversal in $d\kappa_2/dT$ between the two directions is in agreement with the results inferred from the recent specific heat measurements.[7]

The unusual magnetic response in the vortex state of UPt$_3$ apparently reflects the anisotropic nature of the pairing.

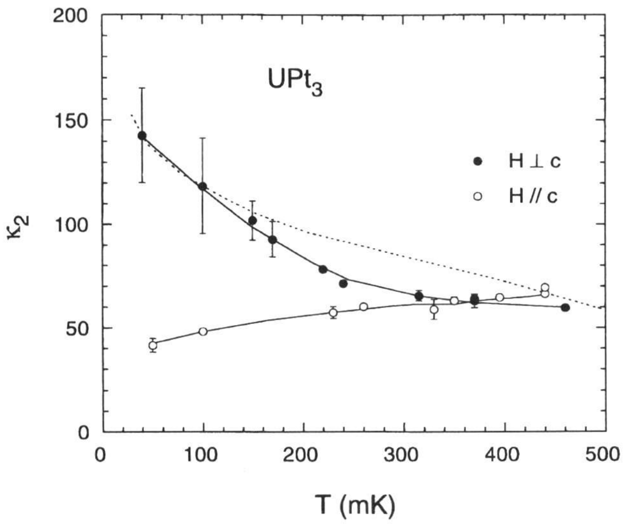

Fig. 6. Temperature variation of the G-L parameter κ_2 for $H \perp c$ (closed circles) and $H // c$ (open circles), obtained from the magnetization measurements. The dotted line indicates the variation expected for a clean superconductor without paramagnetic effect.

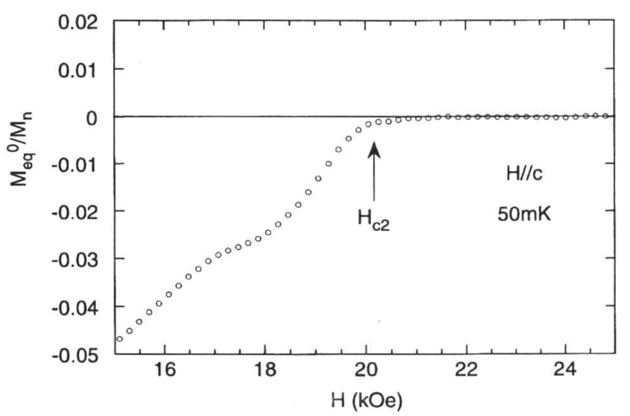

Fig. 7 M_{eq}^{0} divided by the normal-state magnetization for $H//c$ at 50mK. The plot approximately shows a change in the spin susceptibility in the C-phase, which is at most a few percent of the normal state value.

Clearly, the results for $H \perp c$ are incompatible with a spin-singlet pairing, unless we make an unrealistic postulation that the large paramagnetic susceptibility for $H \perp c$ is purely orbital effect. It is rather natural to interpret the results by an odd-parity state, in accordance with the recent NMR experiment on single crystals which reports no change in the Knight shift for *both* principal directions.[9] The results for $H//c$, on the other hand, imply that the pair-spin orientation is somewhat confined in the c-plane. This is in fact the point assumed in the interpretation of H_{c2} anisotropy, and might be an indication of a non-negligible spin-orbit coupling in the pairing channel.[11,12]

We should emphasize, however, that the pair-spin anisotropy is not very strong. To show this somewhat quantitatively, we note that the orbital current contribution to $M_{eq}^{0}{}_{//}$ would be small for $\sim 0.8H_{c2}^{//} \leq H \leq H_{c2}^{//}$, as expected from the behavior of $M_{eq}^{0}{}_{\perp}$. Then $M_{eq}^{0}{}_{//}$ near H_{c2} would mostly consist of the spin susceptibility change $\Delta\chi_s^{//}$ due to pairing, which can be estimated as $\Delta\chi_s^{//}/\chi_n^{//} \sim M_{eq}^{0}{}_{//}/\chi_n^{//}H$. The result is plotted in Fig. 7, which indicates $\Delta\chi_s^{//}$ in the C phase to be at most a few percent of $\chi_n^{//}$. This amount of change might not be detected in the Knight shift measurements[9] within the resolution of the experiment. The present magnetization experiments also reveal the strongly paramagnetic nature of the vortex state of UPt_3, which would be a clue in understanding the unconventional superconductivity in this system.

§4. Summary

We have performed high-resolution DC magnetization measurements on high-quality single crystals of UPt_3 at low temperatures down to 50mK. The results are summarized as follows:

(i) A small change of slope of the equilibrium magnetization $M_{eq}(H)$ is observed across the B-C transition at low temperatures, in agreement with the recently reported specific heat jump at the same field.

(ii) Near H_{c2}, a "peak effect" is observed in the magnetization process at low temperatures below 400mK ($\sim 0.8T_c$). This would not be an indication of a phase transition to a new vortex state such as the FFLO phase, since no anomaly is observed in $M_{eq}(H)$.

(iii) No paramagnetic effect is observed in M_{eq} for the paramagnetically easy direction $H \perp c$, implying that the large normal-state spin-susceptibility for this direction is preserved in the vortex phase. For $H//c$, on the other hand, a significant change of the slope of M_{eq} is observed at H_{c2}, which could be interpreted as a slight suppression of the spin polarization in this direction. These results are in accord with the anisotropy cross-over of H_{c2}, and strongly indicate a triplet pairing with a weak but appreciable anisotropy in the pair-spin orientation. The anisotropy is, however, not very strong, and we consider our results are compatible with the recent NMR Knight shift experiment which reports no change in the Knight shift below T_c for both directions.

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