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X D Zhu$^1$, J C Lu$^2$, Y P Sun$^{1,3}$, L Pi$^2$, Z Qu$^1$, L S Ling$^1$, Z R Yang$^3$ and Y H Zhang$^{1,2}$

$^1$ High Magnetic Field Laboratory, Chinese Academy of Sciences, Hefei 230031, People’s Republic of China
$^2$ Hefei National Laboratory for Physical Sciences at Microscale, University of Science and Technology of China, Hefei 230026, People’s Republic of China
$^3$ Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, People’s Republic of China

E-mail: xdzhu@hmfl.ac.cn and zhangyh@ustc.edu.cn

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Abstract

The magnetization and anisotropic electrical transport properties have been measured in high quality Cu$_{0.03}$TaS$_2$ single crystals. A pronounced peak effect has been observed, indicating that high quality and homogeneity are vital to the peak effect. A kink has been observed in the magnetic field, $H$, dependence of the in-plane resistivity $\rho_{ab}$ for $H \parallel c$, which corresponds to a transition from activated to diffusive behavior of the vortex liquid phase. In the diffusive regime of the vortex liquid phase, the in-plane resistivity $\rho_{ab}$ is proportional to $H^{0.3}$, which does not follow the Bardeen–Stephen law for free flux flow. Finally, a simplified vortex phase diagram of Cu$_{0.03}$TaS$_2$ for $H \parallel c$ is given.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

When a type-II superconductor is placed under a magnetic field $H$ above the lower critical field ($H_{C1}$) and below the upper critical field ($H_{C2}$), the magnetic field penetrates into the superconductor through the vortex arrays, each of which carries one quantum flux surrounded by circulating supercurrent [1]. This state is called the mixed state. Now, it is well known that for many type-II superconductors, the mixed state is actually composed of many complex vortex phases, such as the vortex solid phase, the vortex glass phase and the vortex liquid phase [2–4], instead of the simple vortex lattice predicted by Abrikosov [5]. Although these phases have been studied extensively in the past, the details and the transitions between these phases remain issues of debate.

The peak effect (PE), referring to the anomalous increase and the pronounced maximum of critical current density ($J_C$) or magnetization prior to the irreversible field ($H_{irr}$), has been observed in many type-II superconductors, such as Nb [6, 7], CeRu$_2$ [8, 9], V$_3$Si [10], layered NbSe$_2$ [11–15], ReNi$_2$B$_2$C (Re = Dy, Ho, Er, Tm, Y, Lu) [16, 17], MgB$_2$ [18]. In addition, a similar phenomenon has been observed in high temperature cuprate superconductors (HTCS) [19–25] and the recently discovered iron based superconductors [26, 27] etc that is believed to have different origins and is usually called the fishtail effect. Experimentally, PE is usually observed in weakly pinned and high quality single crystals of layered superconductors for $H$ along the $c$-axis ($H \parallel c$). An incredible number of publications dealing with the underlying physics of the PE have been published during the past few decades, but the interpretation remains a controversial issue [28–35]. So far, it is widely accepted that the PE is related to a vortex phase transition, though the details of the vortex phases remain controversial. Among the proposed mechanisms of PE, phenomenological pictures based on an order–disorder (OD) transition from a quasi-ordered Bragg glass (weakly pinned elastic glass) phase [32] to a disordered phase with proliferation of topological defects [33, 34] (or with these two co-existing phases [35]) explain a broad number of related experimental results.

The vortex liquid phase lies between $H_{irr}$ and $H_{C2}$. Phase transition not only occurs between vortex solid (or vortex glass) and vortex liquid phases, but also between different regimes of vortex liquid phases. An unusual reversible
second order phase transition between two vortex liquid phases has been discovered in YBa2Cu3O7 single crystal by heat capacity and magnetization measurements [36]. In addition, the transition between two different regimes of vortex liquid phase has been observed from the magnetic field dependence of resistivity ($\rho$) behavior and the abnormal negative minimum of Hall resistivity in YBa2Cu3O7. It was explained in a phenomenological picture of transition from ‘activated’ to ‘diffusive’ behavior of vortex motion [36]. However, because of the extremely large $H_{C2}$, the limitation of the applied magnetic field and the fluctuation of superconductivity in HTCS, the normalized superconducting transition temperature $t = (T/T_c$, where $T$ is the temperature and $T_c$ is the superconducting transition temperature) is rather limited in YBa2Cu3O7. As marked by an ellipse in figure 2, a pronounced anomalous PE feature is observed. The high reversibility around the PE transition width (10%–90%) of $\sim 0.1$ K, indicating the high quality of the sample. The measurements discussed in this study were carried out on a sample with a dimension of 3.18 mm ($l$, the longest dimension) $\times$ 1.25 mm ($w$, the width) $\times$ 0.34 mm ($t$, the shortest dimension) mm3 with $t$ along the c-axis. The isothermal dc magnetization hysteresis measurements were performed using a Quantum Design superconducting quantum interference device (SQUID) system (1.8 K $\leq T \leq$ 300 K, 0 T $\leq H \leq$ 7 T). The $H$ dependences of the $J_c$ were extracted from the isothermal magnetization loop results using the formula $J_c = 20(M^+ - M^-)/w(1 - \frac{E}{H})$ [46], where $M^+$ and $M^-$ represent the magnetization (emu cm$^{-3}$) measured during the process of decreasing and increasing field, respectively, and $w$ and $l$ are measured in centimeters.

2. Experimental details

Platelets of single crystal Cu$_{0.03}$TaS$_2$ with $T_c = 4.2$ K used in this study were grown via the iodine vapor transport method as described in our previous report [39]. The onset superconducting transition temperature is 4.2 K with a transition width (10%–90%) of $\sim 0.1$ K, indicating the high quality of the sample. The measurements discussed in this study were carried out on a sample with a dimension of 3.18 mm ($l$, the longest dimension) $\times$ 1.25 mm ($w$, the width) $\times$ 0.34 mm ($t$, the shortest dimension) mm3 with $t$ along the c-axis. The isothermal dc magnetization hysteresis measurements were performed using a Quantum Design superconducting quantum interference device (SQUID) system (1.8 K $\leq T \leq$ 300 K, 0 T $\leq H \leq$ 7 T). The $H$ dependences of the $J_c$ were extracted from the isothermal magnetization loop results using the formula $J_c = 20(M^+ - M^-)/w(1 - \frac{E}{H})$ [46], where $M^+$ and $M^-$ represent the magnetization (emu cm$^{-3}$) measured during the process of decreasing and increasing field, respectively, and $w$ and $l$ are measured in centimeters.

The anisotropic transport property measurements were performed using the standard four probe method in a Quantum Design physical property measurement system (PPMS) (1.8 K $\leq T \leq$ 300 K, 0 T $\leq H \leq$ 16 T). In order to make sure that the direction of $H$ would be exactly parallel to the ab plane ($H \parallel ab$) and the c-axis ($H \parallel c$) of the single crystal, a rotating sample holder was used. We measured the angular dependence of the in-plane resistivity ($\rho_{ab}$) at $T = 3.7$ K and $H = 0.8$ T to determine the direction of the magnetic field. As shown in figure 1, $H \parallel ab$ was determined as the angle corresponding to the minimal resistance ($\theta_{ab}$) and $H \parallel c$ was determined as $\theta_{ab}$ plus 90°. During all the measurements, the excitation current was kept at 5 mA ($J \approx 1.1$ A cm$^{-2}$) and the contact resistance was less than 1 $\Omega$.

3. Results and discussion

3.1. Peak effect

Figure 2 shows the isothermal superconducting magnetization hysteresis ($M-H$) loop for Cu$_{0.03}$TaS$_2$ measured at $T = 2.0$ K. As marked by an ellipse in figure 2, a pronounced anomalous PE feature is observed. The high reversibility around the PE region indicates the high quality of the sample. The right inset panel shows the $M-H$ plot on an expanded scale to emphasize the presence of the PE anomaly around a field of 1.1 T. The fields corresponding to the onset and peak of the PE are marked as $H_{P}^{onset}$ and $H_{P}$, respectively. $H_{irr}$ is estimated from the field where the two branches of the hysteresis loops meet, as shown in the right inset of figure 2. The obtained $H_{P}^{onset}$, $H_{P}$, $H_{irr}$ are $\sim 0.98$ T, 1.1 T, 1.3 T, respectively.

In order to investigate the PE further, more $M-H$ loops were measured at different temperatures. Figure 3 shows the portion of $M-H$ loops (the first and fourth quadrants) for Cu$_{0.03}$TaS$_2$ measured at $T = 2.0$ to 3.6 K. All the curves are shifted for clarity except for $T = 2$ K. The normalized
The angular dependence of the in-plane resistivity ($\rho_{ab}$) at $T = 3.7$ K and $H = 0.8$ T. The arrows mark the degrees corresponding to $H || ab$ and $H || c$. The insets show the sketches of the sample arrangement with respect to the direction to the field.

A superconducting $M$--$H$ loop for Cu$_{0.03}$TaS$_2$ measured at $T = 2.0$ K with $H || c$. The arrows on the curve show the processes of increasing field and decreasing field during the measurement. The left inset shows the dimensional sketch of the sample with respect to the field. The PE region is marked by an ellipse. The right inset shows the magnified plot of the $M$--$H$ curve in the vicinity the PE region.

The log–log plots of the $h$ dependence of $j_C$ at different temperatures for Cu$_{0.03}$TaS$_2$. Obviously, in the range of $0.01 < h < 0.1$, the $j_C$--$h$ relation follows a power law relation $j_C \propto h^{-n}$ (with $n \approx 1$) and overlaps very well except for the PE region for different temperatures. Interestingly, the power law behavior of $j_C$--$h$ has been discovered in NbSe$_2$ [47] and SnMo$_6$S$_8$ [48], in agreement with predictions of the weak collective pinning theory [29] that attributes its origin due to the inter-vortex interactions [49]. Individual pinning should be dominant below the weak collective pinning region, which is separated by a kink on the $j_C$--$H$ curve [48]. In the PE region, the value of the normalized peak position ($H_P/H_{irr}$) of PE is almost unchanged ($h \sim 0.71$) initially, but it decreases gradually with increasing temperature.

The PE has not been observed in directly synthesized Na$_x$TaS$_2$ [50] or Ni$_x$TaS$_2$ grown from NaCl/KCl flux [51], whose superconducting transition widths are larger than that of Cu$_{0.03}$TaS$_2$ [39]. This indicates that the high quality and homogeneity of the sample are necessary for the PE. The inhomogeneity of intercalates leads to the $T_C$ fluctuation in real space and provides additional pinning centers when the $T$ approaches $T_C$. The high density inhomogeneity caused by
intercalates will induce disorder and lead to the disappearance of the OD transition. It will further lead to the disappearance of the PE near $H_{at}$. This can also explain why only 200 ppm of Fe doping causes a significant effect of PE broadening and weakening in NbSe$_2$ [52]. Thus, our data support the view that OD transition is the origin of the PE in Cu$_{0.03}$TaS$_2$.

3.2. The transition from activated to diffusive behavior of the vortex liquid phase

Figure 5(a) depicts the $M$–$H$ curve and the magnetic field dependence of $\rho_{ab}$ measured at $T = 2.0$ K. The inset is the sketch of the sample arrangement with respect to $H$ and its contacts. Figure 5(b) depicts the log–log plot of the $\rho_{ab}$–$H$ curve, whose inset shows the determination of $H_{C2}$. As shown in figure 5(a), with increasing $H$, the $\rho_{ab}$–$H$ curve can be divided into three regimes: the first one is the superconducting regime below $H_{at}$, where $\rho_{ab}$ remains zero; in the second one, $\rho_{ab}$ starts to increase abruptly from $H_{at} = 1.3$ T to a kink at $H \sim 1.9$ T; in the third one, $\rho_{ab}$ increases slowly following a power law up to $H_{C2} = 6.95$ T (as shown in figure 5(b)).

Figures 6(a) and (b) show the measured $\rho_{ab}$–$H$ curves at different temperatures for $H \parallel ab$ and $H \parallel c$, respectively. The insets of figures 6(a) and (b) show the sketches of the direction of the $H$, $J$ and the vortex motion. As shown in figure 6, with increasing $T$, the superconducting transition moves to lower field both for $H \parallel ab$ and $H \parallel c$. However, there is no kink in the flux flow region of the $\rho_{ab}$–$H$ curve for $H \parallel ab$.

Apparently, the kink only occurs in the vortex liquid phase for $H \parallel c$, which should be related to a vortex phase transition. We define the $H$ corresponding to the kink as $H_k$. Interestingly, no anomaly can be observed from the $M$–$H$ curves at $H_k$. In pure and high quality Bi$_2$Sr$_2$CaCu$_2$O$_y$ [53] and YBa$_2$Cu$_3$O$_7$ [54] single crystals, there is a transition in the temperature dependence of resistivity under magnetic field, separating the abrupt increasing region from zero resistivity and the slowly broadening region. The transition has been reported to originate from the first order melting of the vortex lattice, with a characteristic of a discontinuous change of the magnetization [53]. However, as shown in figure 5, no such discontinuous change of the magnetization can be observed from the $M$–$H$ curve for Cu$_{0.03}$TaS$_2$. Thus, the transition from the vortex liquid to another vortex liquid phase observed in Cu$_{0.03}$TaS$_2$ should be attributed to the transition of activated to diffusive behavior in the vortex liquid phase rather than the first order melting of the vortex lattice.

In the activated regime, the vortex line is activated from the strong pinning barrier, which leads to the abrupt increase of $\rho_{ab}$ from zero with increasing $H \parallel c$. The analysis of the activated regime of the $\rho_{ab}$–$H$ curve with the thermal activated plastic motion model did not give any satisfactory fits [55–57]. Therefore, the temperature dependence of $\rho_{ab}$ is measured to obtain the thermal activation energy ($U_{act}$). The activated regime in the vortex liquid phase is more obvious in $\rho_{ab}$–$T$ curves than those in $\rho_{ab}$–$H$ curves. Figure 7 depicts the $H$ dependence of the obtained $U_{act}$, which is found from the $\ln \rho_{ab}/1/T$ curves shown in the inset according to $\rho \approx \rho_0 e^{-U_{act}/T}$ [55]. The $H$ dependence of $U_{act}$ fits well with the power law $U_{act} \sim H^{-1.34}$. The $U_{act}$ of Cu$_{0.03}$TaS$_2$ has a typical value of 300–900 K, which is far from the experimental temperature. The plastic barriers $U_{pl}$ and $T_m$ (the first order thermal melting temperature) can be related via the Lindemann number [2, 58]: $T_m \approx 2.7C_1^2U_{pl}$. Assuming $U_{pl}$ is associated

![Figure 5](image1)

![Figure 6](image2)
with $U_{act}$ and $c_L \sim 0.2$, an unreasonable value of 30–90 K is derived for $T_m$, which further confirms that the transition in the vortex liquid phase does not originate from the first order thermal melting.

In the diffusive regime, the thermal activated plastic motion model is not valid and the $\rho_{ab}$ mainly depends on the energy dissipation of vortex motion for $H \parallel c$, indicating a broad transition. In contrast, for $H \parallel ab$, crossing the van der Waals gap between the superconducting TaS$_2$ layers provides the main energy dissipation because of the strong intrinsic pinning. Thus, for $H \parallel ab$, the transition of activated to diffusive behavior in the vortex liquid phase does not appear in 2H–NbSe$_2$ [38] or Cu$_{0.03}$TaS$_2$.

### 3.3. The breakdown of the BS law

Figure 8 shows the $h$ dependence of the normalized resistivity $\rho_{ab}/\rho_0$ ($\rho_0$ represents the $\rho_{ab}$ at $T = 5$ K) with $H \parallel c$. As shown in figure 8, in the activated regime, $\rho_{ab}/\rho_0 = h$ curves measured at different temperatures overlap with each other; in the diffusive regime, the curves almost overlap with each other when $T \leq 3.2$ K and then turn up rapidly when $T > 3.2$ K. This can be explained by the extra energy dissipation that contributes to the flux flow resistivity when $T$ is near $T_C$. The inset shows the log–log plot of the $h$ dependence of the normalized resistivity $\rho_{ab}/\rho_0$ for $H \parallel c$. Obviously, $\rho_{ab}/\rho_0 \sim h$ curves of the diffusive regime are almost logarithmic when $T \leq 3.2$ K. The $\rho_{ab}/\rho_0 \sim h$ curves can be fitted to $f(h) = h^{0.3}$ very well when $T < 3$ K, which is depicted as a dashed curve in figure 8. Although lower temperature cannot be achieved due to the instrumental limit, our results strongly suggest that the $\rho_{ab}/\rho_0 \sim h$ relation for $T \rightarrow 0$ should be also logarithmic and near $h^{0.3}$. It should be noted that the $\rho_{ab} - H$ curve of YBa$_2$Cu$_3$O$_7$ at $t \sim 0.85$ is very similar to that of Cu$_{0.03}$TaS$_2$ for $t > 0.8$ ($T > 3.2$ K) [37]. For YBa$_2$Cu$_3$O$_7$, the fluctuation will be weaker and the vortex phases will be far from the multi-critical point of the phase diagram at lower $t$ [2]. But the $\rho_{ab} - H$ of YBa$_2$Cu$_3$O$_7$ may have a different relation. Thus, more experiments with higher magnetic field and lower $t$ are needed to study the vortex liquid phase for YBa$_2$Cu$_3$O$_7$ and other HTCS.

As discussed above, in the diffusive regime of the vortex liquid phase, the motion is almost free and the $\rho_{ab} \propto \rho_0(H/H_{C2})^{0.03}$ behavior in Cu$_{0.03}$TaS$_2$ does not follow the BS law for free flux flow. Assuming that in the diffusive regime only flux flow contributes to the $\rho_{ab}$ and $B$ equals $H$, the $V_L \propto H^{-2/3}$ relation can be deduced from $\rho_{ab} = E/J = B \times V_L/J$. Thus, with increasing $H$, $V_L$ increases in the activated regime from $H_{int}$, then reaches a peak at $H_k$ and further decreases with a relation of $V_L \propto H^{-2/3}$.

Interestingly, the Na$_{x}$TaS$_2$ and the Ni$_{x}$TaS$_2$ single crystal do not show any sign of the vortex liquid to liquid transition. Therefore, the inhomogeneity makes vortex-defect interaction the primary energy dissipation channel as opposed to the flux flow and therefore the diffusive regime is not established. Thus, it can be concluded that the high quality and weaker pinning of the sample is vital to the PE and the transition from activated to diffusive behavior of the vortex liquid phase.

### 3.4. The vortex phase diagram

Figure 9 shows the vortex phase diagram for Cu$_{0.03}$TaS$_2$, depicting the temperature dependence of $H_{C1}^{onset}$, $H_{P}$, $H_{int}$ and $H_{C2}$. All critical fields have almost linear $T$ relations in the experimental temperature range. The vortex phases of the PE region are just called the ‘peak effect’. Based upon the discussions above, the vortex phase diagram is composed of the Bragg glass phase, the PE region, the activated and the diffusive regimes of the vortex liquid phase.

2H–NbSe$_2$ shares the same structure with Cu$_{0.03}$TaS$_2$. So far, PE in 2H–NbSe$_2$ has been extensively studied, but not the vortex liquid phase. Our results suggest that the vortex liquid...
Figure 9. The $H$–$T$ vortex phase diagram for Cu$_{0.03}$TaS$_2$, depicting the temperature dependence of $H^\text{mod}_1$, $H_3$, $H_4$, and $H_5$. For details of the different vortex states, see the text.

4. Conclusion

In summary, the PE was observed from the superconducting magnetic hysteresis loops of Cu$_{0.03}$TaS$_2$. A transition from activated to diffusive behavior of the vortex liquid phase was observed from the magnetoresistance experiment for $H \parallel c$. In the diffusive regime of the vortex liquid phase, the in-plane resistivity $\rho_{ab}$ shows a $\rho_{ab} \propto H^3$ relation, which does not follow the BS law for free flux flow. Finally, a simplified vortex phase diagram of Cu$_{0.03}$TaS$_2$ for $H \parallel c$ is given. Our results indicate that high density disorder in TaS$_2$ intercalated liquid phase.

References

[27] Pippard A B 1969 Phil. Mag. 19 217
[34] Higgins M J and Bhattacharya S 1996 Physica C 257 232 and references therein
Bean C P 1964 Rev. Mod. Phys. 36 31