Vortex phase diagram of the layered superconductor \( \text{Cu}_{0.03}\text{TaS}_2 \) for \( H \parallel c \)

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Abstract
The magnetization and anisotropic electrical transport properties have been measured in high quality \( \text{Cu}_{0.03}\text{TaS}_2 \) 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 \( \text{Cu}_{0.03}\text{TaS}_2 \) 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 YBa$_2$Cu$_3$O$_7$ 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 YBa$_2$Cu$_3$O$_7$. 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 YBa$_2$Cu$_3$O$_7$ ($t > 0.84$) [37]. Hence, the $\rho$–$H$ relation for lower $t$ remains unexplored. Interestingly, a similar transition in 2H–NbSe$_2$ single crystal was observed from the magnetoresistance results for $H \parallel c$ (measured at $T = 4.2$ K) [38], though it was not discussed.

In our previous work, high quality Cu$_{0.03}$TaS$_2$ single crystals with $T_{onset}^C = 4.2$ K were grown successfully [39]; these crystals are isostructural with 2H–NbSe$_2$. The Ginzburg number ($G_\lambda$) is a measure of the importance of thermal fluctuations according to the traditional collective pinning theory [29] and is given by the formula $G_\lambda = \frac{\kappa_B T_c / H_{C2}^2(0) \xi(0) \xi_0^2}{\eta_0^2}$. The estimated $G_\lambda$ for Cu$_{0.03}$TaS$_2$ is of the order of $10^{-5}$ [39], comparable with that for NbSe$_2$ ($ \approx 3 \times 10^{-4}$) [7]. Thus, Cu$_{0.03}$TaS$_2$ should be a good candidate for PE.

When the Lorentz–Magnus force ($F_L = J \times B$, where $B$ is the magnetic flux density) exerted by the current density ($J$) overcomes the pinning forces, the vortices will move viscously with a mean velocity $V_L$ generating an electric field $E = B \times V_L$ [40]. The viscous friction depends on the energy dissipation process of quasi-particles in and around the normal core of the vortices. Therefore, the dc resistivity $\rho = E/J$ measured in the vortex liquid phase is related to the magnetic field and the vortex motion (or flux flow). The flux flow resistivity ($\rho_F$) is defined as the slope of the $E$–$J$ curve ($\partial E/\partial J$), which is independent of the pinning force [40]. The Bardeen–Stephen (BS) law predicts that if other forces such as vortex-defect interaction (pinning) are negligible, the flux will flow freely and the free flux flow (FFF) resistivity will be proportional to the magnetic field $H$, $\rho_F = \rho_{ab} H/\sqrt{H_{C2}}$, where $\rho_{ab}$ is the normal state resistivity [41]. The BS law has been confirmed in most dirty and clean s-wave systems. In an extremely clean NbSe$_2$ single crystal, the confirmation of the BS law has been observed from the magnetic field dependence of resistivity and the Hall effect [42]. However, in some layered superconductors with gap nodes or with multiple bands gap, such as Bi$_2$Sr$_2$CuO$_{6+\delta}$ [43], Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ [44] and MgB$_2$, [45], the BS law breaks down in microwave impedance experiments. The origin of these phenomena has been attributed to the different energy dissipation influenced by the abnormal gap structures [43].

In this study, we report the discovery of the PE and the transition from activated to diffusive behavior of the vortex liquid phase in Cu$_{0.03}$TaS$_2$ for $H \parallel c$. Finally, a simplified vortex phase diagram of Cu$_{0.03}$TaS$_2$ is proposed.

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) mm$^3$ 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 $J_C = 20(M^+ - M^-)/w(1 - \frac{\rho_{ab}}{\rho_F})$ according to the Bean critical state model [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. $H_{int}$ 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 $H_p^{onset}$, $H_p$, $H_{int}$ 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 from $T = 2.0$ to 3.6 K. All the curves are shifted for clarity except for $T = 2$ K. The normalized
Figure 1. The angular dependence of the in-plane resistivity \( \rho_{ab} \) at \( T = 3.7 \text{ K} \) and \( H = 0.8 \text{ T} \). The arrows mark the degrees corresponding to \( H \parallel ab \) and \( H \parallel c \). The insets show the sketches of the sample arrangement with respect to the direction to the field.

Figure 2. A superconducting \( M-H \) loop for Cu_{0.03}TaS_2 measured at \( T = 2.0 \text{ K} \) with \( H \parallel c \). 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. \( H_{\text{onset}} \), \( H_p \), \( H_{\text{irr}} \) are marked by arrows.

Figure 3. The magnified plots of \( M-H \) loops for Cu_{0.03}TaS_2 measured at different temperatures. All curves are shifted for clarity and the dashed lines represents \( M = 0 \) at each temperature. The up down arrow represents 1 emu cm\(^{-3}\).

Figure 4. The log–log plot of the normalized critical current density \( j_c \) \( (J_c / J_{c,0}) \) as a function of normalized field \( (h = H / H_{irr}) \) of Cu_{0.03}TaS_2. The dashed line serves as a guide for the eye. The arrow shows the direction of increasing temperature.

The data of \( j_c (J_c / J_{c,0}) \), where \( J_{c,0} \) represents the critical current density under zero field) as a function of normalized field \( (h = H / H_{irr}) \) are extracted from the \( J_c (T, H) \) data.

Figure 4 shows 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 SmMo_6S_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 (\( H_{\text{onset}} / H_{irr} \)) 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_xTaS_2 [50] or Ni_xTaS_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 \( H_{ct} \). This can also explain why only 200 ppm of Fe doping causes a significant effect of PE broadening and weakening in \( \text{NbSe}_2 \) [52]. Thus, our data support the view that OD transition is the origin of the PE in \( \text{Cu}_{0.03}\text{TaS}_2 \).

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 \( H_{ct} \). 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 \( H_{ct} \), where \( \rho_{ab} \) remains zero; in the second one, \( \rho_{ab} \) starts to increase abruptly from \( H_{ct} = 1.3 \) T to a kink at \( H \sim 1.9 \) T; in the third one, \( \rho_{ab} \) increases slowly following a power law up to \( H_{ct} = 6.95 \) 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 \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 \) [53] and \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) [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 \( \text{Cu}_{0.03}\text{TaS}_2 \). Thus, the transition from the vortex liquid to another vortex liquid phase observed in \( \text{Cu}_{0.03}\text{TaS}_2 \) 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 \( \text{Cu}_{0.03}\text{TaS}_2 \) has a typical value of 300–900 K, which is far from the experimental temperature. The plastic barriers \( U_p \) and \( T_m \) (the first order thermal melting temperature) can be related via the Lindemann number [2, 58]: \( T_m \approx 2.7c_{\perp}^2U_p \). Assuming \( U_p \) is associated...
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 $\rho_{ab}/\rho_{c}$ ($\rho_{ab}$ represents the $\rho_{ab}$ at $T = 5$ K) with $H \parallel c$. As shown in figure 8, in the activated regime, $\rho_{ab}/\rho_{c} \sim h$ 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 $\rho_{ab}/\rho_{c}$ for $H \parallel c$. Obviously, $\rho_{ab}/\rho_c \sim h^0$ in the diffusive regime when $T \leq 3.2$ K. The $\rho_{ab}/\rho_c \sim h^0$ 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 $\rho_{ab}/\rho_c$ 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 $\rho_{ab} \sim \rho_n(H/H_{c2})^{0.03}$ 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 $V_1 \sim H^{-2/3}$ relation can be deduced from $\rho_{ab} = E/J = B \times V_1/J$. Thus, with increasing $H$, $V_1$ increases in the activated regime from $H_{irr}$, then reaches a peak at $H$ and further decreases with a relation of $V_1 \sim H^{-2/3}$.

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 $H_{c1}^{\text{ons}}$, $H_p$, $H_{irr}$ and $H_{c2}$ [4]. 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 phase of Cu$_{0.03}$TaS$_2$ 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 phase...
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Figure 9. The $H–T$ vortex phase diagram for Cu0.03TaS2, depicting the temperature dependence of $H_p$, $H_a$, and $H_T$. For details of the different vortex states, see the text.

phase of 2H–NbSe2 may also be complicated, as in Cu0.03TaS2 and HTCS.

4. Conclusion

In summary, the PE was observed from the superconducting magnetic hysteresis loops of Cu0.03TaS2. 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 Cu0.03TaS2 for $H \parallel c$ is given. Our results indicate that high density disorder in TaS2 intercalated superconductors will lead to the disappearance of the PE and the transition from activated to diffusive behavior of the vortex liquid phase.
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