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Vortex phase transitions in monolayer FeSe film on SrTiO₃

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Abstract

The voltage–current (V – I) characteristics in superconducting monolayer FeSe film on SrTiO₃ (100) under different magnetic fields are investigated. The zero-field V – I result exhibits signatures of a Berezinski–Kosterlitz–Thouless transition, a characteristic of two-dimensional (2D) superconductors. Under an applied magnetic field, with current density lower than the critical current density, the sheet resistance versus current density (R_{sq} – J) dependence changes from ohmic (R_{sq} independent of J) to non-ohmic (a nonlinear dependence of R_{sq} on J) as the temperature decreases, indicative of a vortex phase transition/crossover. We interpret the high-temperature phase as the vortex liquid phase and the low-temperature phase as the vortex slush phase, which has short-ranged vortex lattice correlation, while long-range correlation (i.e. true superconductivity) is absent. No transition into a vortex glass phase is seen, illustrating the importance of thermal fluctuations in a perfect 2D superconductor under a magnetic field.

Introduction

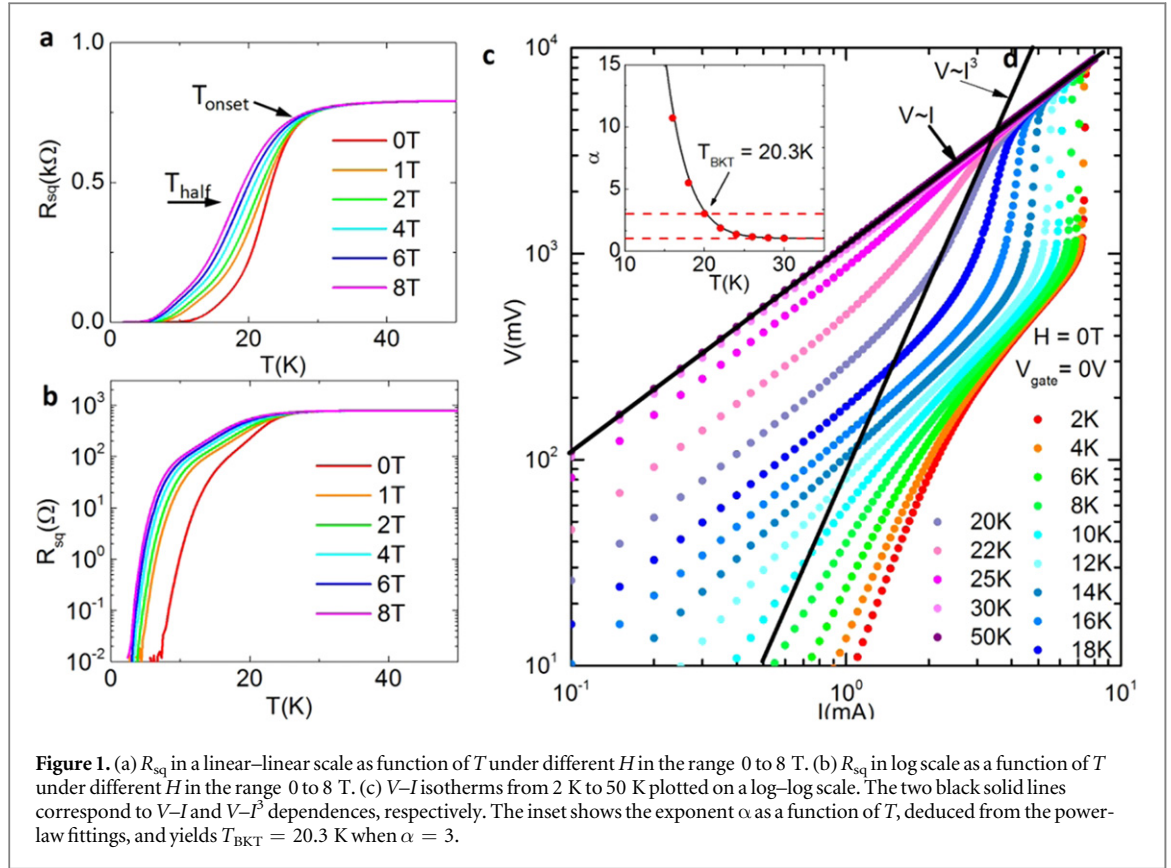
The vortex phase transition is one of the most fascinating research topics in the physics of type-II superconductors under a finite magnetic field. The existence of a vortex liquid–glass transition, which separates a high-temperature vortex liquid (VL) phase from a low-temperature vortex glass (VG) phase [1–3], has been demonstrated in many three-dimensional (3D) superconductors such as cuprates [4–8], MgB₂ [9], Nb [10, 11] and W [12]. In the VG phase, a true superconducting state with zero resistance exists due to pinning of the vortices. On the other hand, a nonzero but exponentially small resistance remains in the VL phase. In the two-dimensional (2D) limit, due to the significantly enhanced thermal fluctuation effects, only the VL phase is present at finite temperatures. The absence of a VL–VG transition at finite temperatures in 2D superconductors has been theoretically predicted and experimentally verified in 2D superconducting films [13, 14].

In this paper, we report experimental evidence of a different vortex phase transition in a recently discovered high-temperature 2D superconductor, a monolayer of FeSe on SrTiO₃ [15–21]. Instead of a VL–VG transition, as the temperature decreases a

transition/crossover from a VL phase to a vortex slush (VS) phase is revealed by examining the voltage–current (V – I) dependence. The VS phase is characterized by a short-range vortex lattice correlation without a long-range correlation. The presence of finite resistance (although exponentially small at low temperatures) makes it distinctively different from the zero-resistance VG phase. While both of the VL–VS and VS–VG transitions have been observed in bulk YBa₂Cu₃O₇ single crystals [22], no VS–VG transition is observed in monolayer FeSe/SrTiO₃ films, potentially due to the strongly enhanced thermal fluctuation effects in 2D superconductors.

Materials and method

The FeSe films studied here were grown using a custom-built ultrahigh vacuum molecular beam epitaxy (MBE) system. The insulating SrTiO₃ (100) substrates with a specific TiO₂-terminated surface were prepared using standard chemical and thermal treatments in a tube furnace. Then, the SrTiO₃ substrates were transferred into the MBE chamber and annealed at 600 °C for 1 h before FeSe film growth. FeSe films were grown at a rate of approximately 0.2



layers/min on the SrTiO₃ substrate by coevaporating Fe(99.995%) from an e-gun source and Se(99.999%) from a Knudsen cell with a flux ratio of 1:10 at 330 °C. A post-grown annealing of the film at 550 °C for 2 h was needed to make FeSe films superconducting. Then, additional 10 unit cell FeTe was deposited on the FeSe film as a capping layer to prevent its atmosphere exposure, similar to a previous method for *ex situ* transport measurement [20]. Transport measurements were performed by using a standard six terminal hall-bar geometry with a width of 0.89 mm and length of 1.25 mm between the two voltage electrodes.

Results and discussion

The sheet resistances (R_{sq}) as a function of temperature (T) were measured with $I = 500$ nA for the 1UC FeSe/SrTiO₃ with 10UC FeTe capping layers. As shown in figures 1(a) and (b), R_{sq} is 790 Ω when $T = 50$ K and starts dropping from ~ 30 K to lower than < 0.04 Ω when T_{zero} is ~ 7 K, below the measurement sensitivity. The superconducting transitions become broader and shift to lower temperatures with increasing H , which are typical characteristics of superconducting transition in thin films. It is natural to attribute the broader transitions of $R_{sq}(T)$ in our 1UC FeSe superconducting layer to the Berezinski–Kosterlitz–Thouless (BKT) phase transition. Strong support for BKT physics is found in the zero-field V – I isotherms

measured over a range of different T . As shown in figure 1(c), the measured V – I curves exhibit I^α power-law dependence, and is precisely what is expected for a system governed by the BKT mechanism [23–26]. In the BKT scenario, the excitation current breaks apart the bound vortex–anti-vortex pairs and give the sample a non-ohmic behavior with increasing current. At higher currents, the rate of increase of V with I is reduced and the I – V curves revert toward ohmic. There are free vortices above the BKT transition temperature, T_{BKT} , and bound vortex–anti-vortex pairs lower energy than the free vortex below T_{BKT} . In the region where V shows the most rapid increase with I , it varies as I^α with a temperature dependent exponent $\alpha(T)$. The exponent α increases gradually from 1 as the temperature decreases and crosses 3 at T_{BKT} , which we assign as the BKT transition temperature ($T_{BKT} = 20.3$ K as shown in the inset of figure 1(c)). Contrary to the theoretical prediction of a sharp jump from 1 to 3 at T_{BKT} , the gradual change of α is likely caused by disorder effects in our monolayer sample, which smears out the sharp transition.

Below, we describe the R_{sq} – J characteristics under perpendicular H , which is numerically computed from the V – I data under H . The R_{sq} – J isotherm at various T from 2 to 50 K at $H = 1$ T is shown in figure 2(a). At the critical current density J_c , R_{sq} shows an abrupt jump to a normal state value, indicating that the superconductivity is completely destroyed above J_c . J_c is found to shift to lower values with increasing T , exhibiting a typical superconducting behavior.

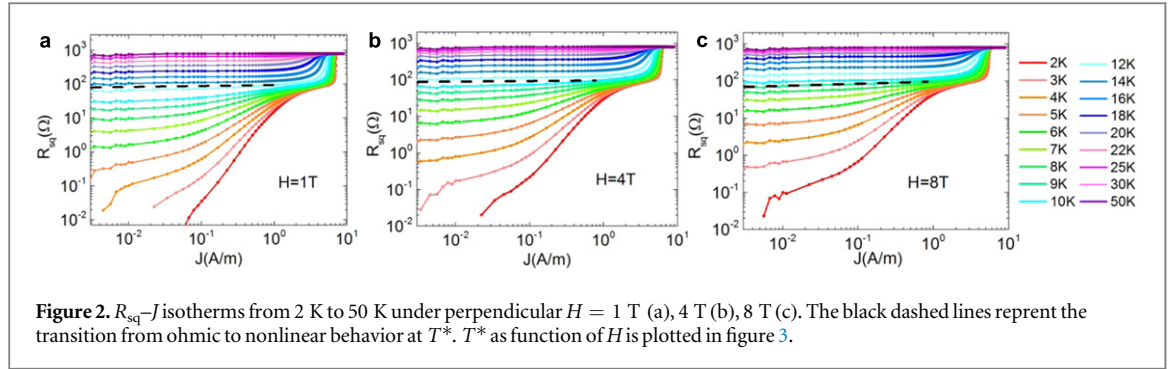


Figure 2. R_{sq} - J isotherms from 2 K to 50 K under perpendicular $H = 1$ T (a), 4 T (b), 8 T (c). The black dashed lines represent the transition from ohmic to nonlinear behavior at T^* . T^* as function of H is plotted in figure 3.

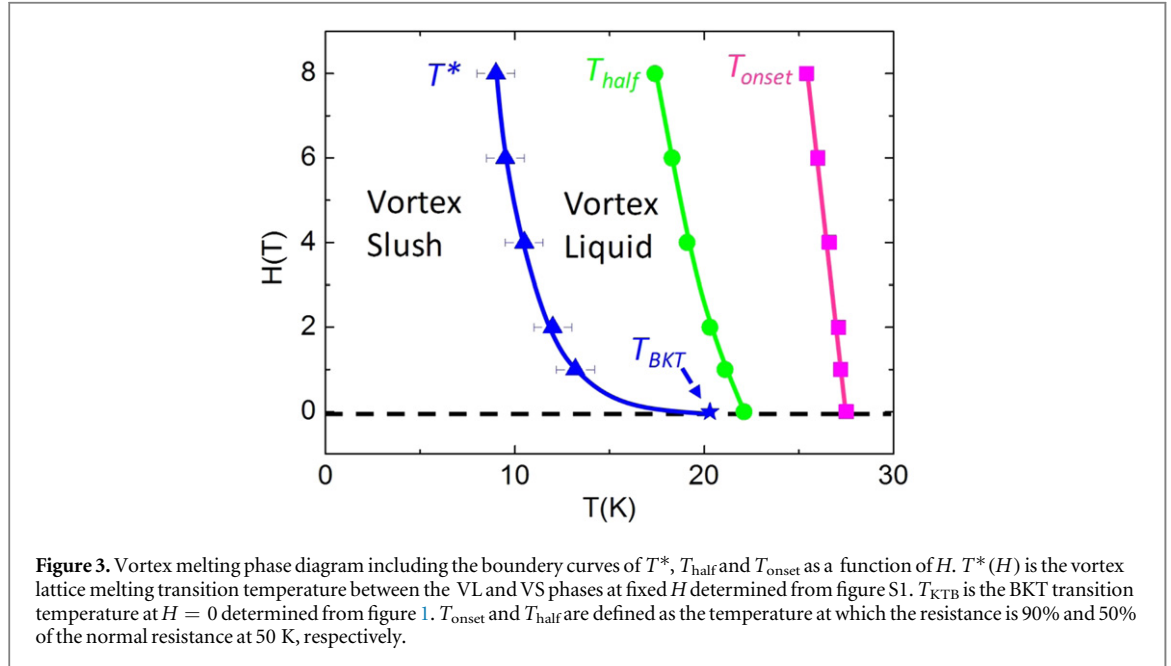
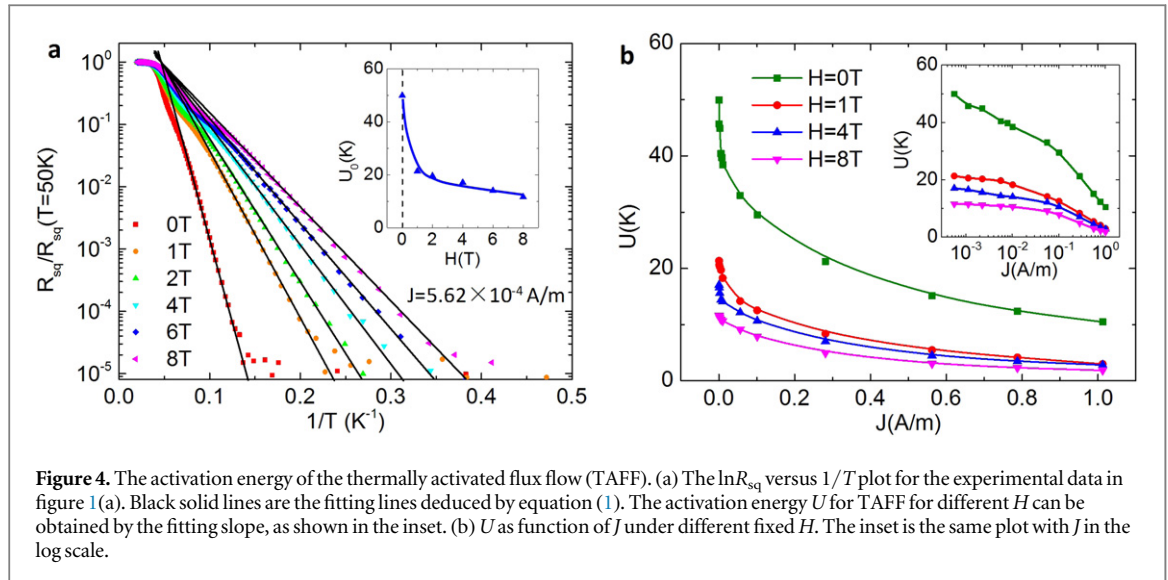


Figure 3. Vortex melting phase diagram including the boundary curves of T^* , T_{half} and T_{onset} as a function of H . $T^*(H)$ is the vortex lattice melting transition temperature between the VL and VS phases at fixed H determined from figure S1. T_{KTB} is the BKT transition temperature at $H = 0$ determined from figure 1. T_{onset} and T_{half} are defined as the temperature at which the resistance is 90% and 50% of the normal resistance at 50 K, respectively.

With J well below J_c , however, R_{sq} - J dependence exhibits two different regimes depending on T , indicative of two phases: (I) when the temperature T is just below the superconducting transition temperature, R_{sq} shows ohmic behaviors (independent of J) until $T = \sim 13$ K at which R_{sq} is nearly an order of magnitude lower than the normal state sheet resistance; (II) when T goes lower than ~ 13 K, R_{sq} starts dropping with nonlinear ‘S sharp’. R_{sq} drops rapidly first when J goes below J_c and then drops slowly when J goes further below ~ 0.1 A m $^{-1}$ and saturates to a very small nonzero value when $J \rightarrow 0$, although the value can be much smaller than those in phase I. We did not take ρ -measurements below 2 K due to the limits of the equipment. Here we define the temperature at which the R_{sq} - J characteristics cross from linear dependence of phase I to nonlinear dependence of phase II as T^* . T^* in the case of $H = 1$ T is ~ 13 K as shown by the black dashed line in figure 2(a). T^* are ~ 10 K and ~ 9 K at $H = 4$ T (figure 2(b)) and $H = 8$ T (figure 2(c)), respectively. According to a previous study [12], the transition temperature for the VG-VL transition can be identified by first plotting the slope (dV/dI) as a function of I for each T . The maximum

value of dV/dI is identified, and T^* is determined at the maximum rate of change of $(dV/dI)^{-1}$ with respect to T . We have used the same method to identify T^* , and have shown procedures in figure S1 in the supplementary materials. We found that the T^* values are 13.2 K, 9.2 K, 8.0 K for $H = 1$ T, 4 T, 8 T, respectively, consistent with the identification in figure 2.

The boundary curve of T^* as function of H is plotted in figure 3, which separates the high-temperature phase I and the low-temperature phase II. Phase I exhibits the typical ohmic characteristics of the VL phase observed in previous experiments. Phase II, which is different from either the VL or VG phase, was not observed in previous experiments for thin films and constitutes the key new result of our work. The R_{sq} - J dependence goes downward nonlinear when J is well below J_c , indicating that the current assisted vortex flow as current increases, and excitations emerge that destroy the short-range correlations. But R_{sq} as $J \rightarrow 0$ is still finite, suggesting long-range correlations are absent, i.e. phase II is not a true VG phase. Consequently, phase II exhibits the typical characteristics of a VS phase observed in defect-enhanced



$\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals by radiation damage [22]. The authors argued that an optimal disorder density is the critical factor to see this VS phase with only short-range vortex lattice correlation. We believe the 2D thermal fluctuation effect of our monolayer FeSe film grown on SrTiO_3 is important for the absence of long-range vortex lattice correlations. We also notice that T^* decreases with increasing H but the slope dT^*/dH becomes less steep with increasing H , as shown in figure 3. This feature is quite similar to the phase diagram observed for the melting of a flux-line solid in anisotropic layered materials such as BSCCO, for which T_{melting} is almost independent of H in a high-field regime [27]. When the external field is high enough, interaction between adjacent pancake vortices in the same layer is stronger than the interaction between those in adjacent layers but on the same vortex line, causing the quasi-2D character for BSCCO.

Now we move on to extracting the energy barrier U , which depends on H and J , for vortex flows. In the region of a thermally activated vortex flow (TAVF), $\ln R_{sq}$ versus $1/T$ can be described by

$$R_{sq}(H, J) = R_0 \exp[-U(H, J)/T] \quad (1)$$

where R_0 is a temperature independent constant and $U(H, J)$ is the activation energy of the flux flow dependent on H and J . As shown in figure 4(a), the experimental data in figure 1(a) with $J = 5.62 \times 10^{-4} \text{ A m}^{-1}$ or $I = 500 \text{ nA}$ exhibit kinks in the $\ln R_{sq}$ versus $1/T$ plots close to $T^*(H)$, suggesting the TAVF activation energy is different below and above T^* . The $\ln R_{sq}$ versus $1/T$ curves at different H are linear in the region below T^* and can be well fitted by equation (1) (black solid lines). The fitting lines obtained from $\ln R_{sq}$ versus $1/T$ at different H can be well extrapolated to meet at the same temperature, $T^{**} = 21 \text{ K}$, which is close to $T_{\text{KT B}}$. In addition, the activation energy $U(H)$ for $J = 5.62 \times 10^{-4} \text{ A m}^{-1}$

can be obtained by the fitting slope. As shown in the inset of figure 4(a), U quickly decreases once H is turned on. We have also compared our data to a logarithmic and a power-law dependence as shown in figure S3 of the supplementary materials, but none of them reproduces our data perfectly. As U not only depends on H but also on J , the J dependence of U under fixed H has also been studied, as shown in figure 4(b). U increases rapidly with decreasing J , but U increases slowly and saturates at a finite value after J is lower than $\sim 0.1 \text{ A m}^{-1}$ (see the inset of figure 4(b)). The J dependence of U can be understood as short-range excitations emerging as J increases, which effectively lower the activation energy for vortex flow. In addition, U saturates to a finite value when $J \rightarrow 0$ indicates the presence of TAVF, consistent with a finite resistance state in the $J \rightarrow 0$ limit shown in figure 2. For a true VG phase, U should diverge when $J \rightarrow 0$. Our observation doesn't match this description due to the presence of the finite U in the $J \rightarrow 0$ limit, further confirming that phase II is not a true VG phase, but a VS phase with short-range correlation.

According to the VG transition theory, the $V-I$ curves at different T near the transition temperature can be scaled into two branches by the scaling equation. It is noted that the 2D VG theory has also been developed, and verified in some cases of extremely thin superconducting films [13, 14]. Interestingly our data cannot achieve a scaling collapse for any combination of the fitting parameters by using either a 3D or 2D scaling equation. The failure is not surprising because the transition/crossover observed in this paper is in fact a transition/crossover between VL and VS instead of a VL-VG transition. It is still unknown whether a true VG phase can occur when T goes to much lower temperatures below 2 K. Future experiments, especially study of $V-I$ characteristics down to mK for this system, are needed.

Conclusion

To conclude, we observed an unexpected vortex slush phase in a monolayer FeSe film on SrTiO₃. The phase is characterized by a short-range vortex lattice correlation while the long-range correlation is absent. The presence of a finite resistance in the $J \rightarrow 0$ limit makes it distinctively different from the zero-resistance vortex glass phase. Thermal fluctuations in a perfect 2D superconductor under a magnetic field is the reason that no transition to the true vortex glass phase is seen in the monolayer FeSe/SrTiO₃ film.

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