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Vortex pinning and irreversibility fields in $\text{FeS}_{1-x}\text{Se}_x$ ($x = 0, 0.06$)

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We report strong vortex pinning and large irreversibility fields in single crystals of tetragonal $\text{FeS}_{1-x}\text{Se}_x$ ($x = 0, 0.06$). Vortex dynamics is characterized by crossover in field dependence of the depinning energy U_0 , indicative of single flux surface pinning to the region of collective flux pinning on point-like defects. The close proximity of the irreversibility lines to the upper critical field (H_{c2}) is consistent with strong pinning in FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$, pointing that new materials with building-blocks of FeS_4 tetrahedra are likely to host high critical currents.

Since the discovery of superconductivity in $\text{LaOFeAs}_{1-x}\text{F}_x$ many new iron-based superconductors have been discovered.¹ FeSe features rather simple crystal structure characterized by FeSe_4 tetrahedra and superconductivity below about 8 K. FeSe_4 building blocks can be incorporated in various space groups and materials such as $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$, $\text{K}_x(\text{NH}_3)_y\text{Fe}_2\text{Se}_2$, and $(\text{Li}_{0.8}\text{Fe}_{0.2})\text{OHFeSe}$ where superconducting T_c rises up to 44 K.²⁻⁴ FeSe_4 tetrahedra also constitute potential basis of high-temperature superconductivity since FeSe films on SrTiO_3 have shown T_c as high as 100 K.⁵

Superconductivity at 5 K was also discovered in FeS.⁶ FeS has the same crystal structure and similar electronic band structure to FeSe.⁷ Large magnetoresistance, non-linear Hall effects and multiband electronic conduction are observed in FeS.⁸ Low temperature heat transport and specific heat study of FeS crystal suggest nodal superconducting gap whereas theoretical studies point to possible $d_{x^2-y^2}$ pairing symmetry.^{7,9,10}

The critical current densities in FeS are comparable to iron based superconductors with much higher superconducting transition temperature, such as FeSe and $\text{K}_x\text{Fe}_{2-y}\text{Se}_2$, but lower than $\text{FeSe}_{0.5}\text{Te}_{0.5}$ and iron pnictide superconductors.¹¹⁻¹⁵ Though FeS itself has T_c too low to be used in practical application, it is of interest to study irreversibility fields, vortex dynamics and pinning properties, which will be helpful for understanding new FeS-based superconductors. The irreversibility line determines the region in the magnetic field vs temperature (H, T) phase diagram where the dissipationless property of the thermodynamical superconducting phase is preserved with partial penetration of the magnetic field inside the superconductor. This is the boundary between the magnetically irreversible region with nonzero critical current (J_c) and reversible region with zero J_c in the (H, T) and depends on the flux pinning strength, dimensionality, and the grain size.¹⁶

The ac susceptibility is a powerful tool to investigate the flux dynamics and irreversibility via low-frequency characteristics of the vortex motion. Here we report the ac susceptibility study on FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$. We provide insight into the dynamical properties of vortices in the crossover between the liquid flux phase (LFP) and liquid glass phase (LGP) via dynamic features of the mag-

netic flux penetration¹⁷ and we also present irreversibility lines, pinning mechanism, and depinning energy barriers within the thermally activated flux motion framework. The close proximity of the irreversibility line to the H_{c2} in the (H, T) phase diagram suggests strong vortex pinning in FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$. This indicates that materials with FeS_4 building blocks and higher T_c 's are highly desirable for applications.

Single crystals of FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$ were synthesized by hydrothermal reaction method.¹¹ Ac susceptibility measurements were performed in Quantum Design MPMS-XL5 in magnetic field $H_{ac} = 1$ Oe and frequency ν_m ranged from 1 to 1000 Hz superimposed on a constant dc field.

Figure 1 shows the temperature dependence of the in-phase χ' and the out-of-phase χ'' ac susceptibility for FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$ in different dc magnetic fields. Both samples exhibit quite sharp superconducting transitions. The peak in χ'' arises due to resonant absorption of energy in the thermally activated flux-creep model.¹⁷ Only one peak is observed in FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$, indicating the absence of grain effects.¹⁸ With the dc field increasing, the transitions of $\chi'(T)$ and $\chi''(T)$ are shifted to low temperature. The peak height of $\chi''(T)$ initially increases with field, and is constant with further dc field increase. This is similar to other high- T_c Fe-based and Cu-based superconductors.¹⁹ The χ' represents the magnetic flux shielding, and χ'' is related to the dissipation. As shown in Fig.1(d), we define T_c as the onset of $\chi'(T)$ decrease, and denote the peak position of $\chi''(T)$ as T_p . The position of T_p usually appears a few Kelvin below the onset of the diamagnetic signal in $\chi'(T)$, and coincides with the peak of $d\chi'(T)/dT$ in the Debye-like relaxation framework.¹⁷ The peak of χ'' can be attributed to the resonant absorption of energy when the frequency of the excitation matches the inverse characteristic relaxation time of the vortices in the pinning potential dip. The magnetic field dependence of T_p can be used to determine the irreversibility line; this is often used χ''_{ac} criterion which we use in the following analysis.^{17,19}

Figure 2 (a) shows the typical curves of the temperature dependence of the out-of-phase ac susceptibility around T_p at $H = 1000$ Oe for different frequencies. T_p increases with the frequency increase, since the gain in

FIG. 1. (Color online). Temperature dependence of the in-phase χ' and the out-of-phase χ'' ac susceptibility for FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$ at various dc magnetic fields with the ac amplitude $H_{ac} = 1$ Oe and frequency $f = 1$ Hz. (a) $H//c$ and (b) $H//ab$ for FeS respectively; (c) $H//c$ and (d) $H//ab$ for $\text{FeS}_{0.94}\text{Se}_{0.06}$ respectively.

the unrelaxed value of current density must compensate for its loss due to relaxation. The dc field H_p vs T_p phase diagram for frequencies is shown in Fig. 2(b). All data can be fitted by the formula

$$H_p(T) = H_0(1 - t_p)^\beta \quad (1)$$

where $t_p = T_p/T_c$ is the reduced temperature, β is an index of the pinning strength for the vortex system. As shown in the inset of Fig. 2(b), with the frequency increasing from 1 to 1000 Hz, β decreases from 1.31 to 1.26. We note that $\beta = 4/3 - 3/2$ is commonly found in the fitting of the irreversibility line in cuprate superconductors.²⁰

In the high- T and high- H regions above irreversibility line of the vortex phase diagram, flux lines are in the thermally assisted flux flow (TAFF) regime where the thermally activated vortex jumps between the metastable states of the vortex lattice. In the low-temperature and low-field region below the irreversibility line, vortex is frozen in a glassy-like state and flux motion has to overcome a large pinning potential well. This is thermally activated flux creep (TAFC) regime. The movement of

FIG. 2. (Color online). (a) Temperature dependence of the out-of-phase ac susceptibility under various frequencies with $H = 1000$ Oe and $H//c$ for FeS. (b) Temperature dependence of out-of-phase peak (T_p) at different frequencies. inset of (b) shows the index of the pinning strength β as a function of frequencies. $1/T_p$ as a function of frequency of (c) FeS, and (d) $\text{FeS}_{0.94}\text{Se}_{0.06}$.

the flux lines is governed by the depinning energy barrier U_0 . In the thermally activated framework where the mechanism of driving the fluxoid dynamic relaxation goes via thermally activated processes, U_0 can be obtained by fitting the dependence of $1/T_p$ on frequency f using: $\frac{f}{f_0} = \exp(-\frac{U_0(H)}{k_B T_p})$, where f_0 is a constant, and k_B is Boltzmann constant. As shown in Fig. 2(c) and 2(d), $1/T_p$ as a function of frequency of FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$ can be reasonably well explained, and the extracted depinning energy U_0 on various dc field is presented in Fig. 3.

Even though the critical currents in $\text{FeS}_{0.94}\text{Se}_{0.06}$ are three times larger than that in FeS, both FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$ have similar depinning energy of the order of 10^3 K, also similar to FeSe grown by the vapor transport method.¹³ This is smaller than in $\text{K}_{0.79}\text{Fe}_{1.86}\text{Mn}_{0.01}\text{Se}_2$,¹⁹ probably reflecting much smaller lattice imperfections in FeS and $\text{FeS}_{0.94}\text{Se}_{0.06}$. The observation of quantum oscillation in FeS is in agreement with our interpretation.²¹

As shown in Fig. 3, there is a crossover in the field

FIG. 3. (Color online). Magnetic field dependence of the effective depinning energy, The solid lines are the lines fitted with the formula $U_0 = H^{-n}$.

dependence of depinning energy U_0 . Both FeS and FeS_{0.94}Se_{0.06} have the same crossover field $H_{cr} = 600$ Oe. The data on either side of H_{cr} can be fitted with $U_0 = H^{-n}$, $n = 0.38, 0.65$ for FeS and FeS_{0.94}Se_{0.06} at $H < H_{cr}$, while $n = 1.21, 1.28$ at $H > H_{cr}$. The crossover in the field dependence of depinning energy has been observed in high- T_c superconductors,^{17,22} and is usually interpreted by the transition from a single flux pinning to the collective flux pinning region.²³ In the low field region the number of vortices is lower than the number of pinning centers. Then, single vortex pinning effect gives rise to weak field dependence of U_0 . In the higher-field region the existence of the short range order of the flux-line lattice results in $\sim H^{-1}$ field dependence. Strong plastic pinning is expected to produce a $\sim H^{-0.5}$ dependence on the magnetic field.²⁴ However, the pinning can also occur from the local strong pinning centers in the low field region and will result in strong intrinsic pinning. It is reported that the planar defects are responsible for $U \sim H^{-1/2}$ and the point defects are responsible for $U \sim H^{-1}$.^{25,26} Considering the weak interlayer coupling of FeS due to deintercalation synthesized method, the crossover could denote the transition of dominant pinning mechanism from surface-like pinning to point-like pinning with field increase. This is consistent with the vortex pinning mechanism derived from the critical current density measurement.¹¹

The χ'' criterion was used to draw the irreversibility line because the weak dependence of T_p on frequencies f was observed in all applied DC fields. Therefore, we choose the lowest frequency $f = 1$ Hz to determine T_p . There is usually a second peak region in most iron-based superconductors, which divide the vortex phase diagram into vortex glass region and plastic motion region.^{15,27} We did not observe the second peak effect from the dc magnetization above 2 K.¹¹ Be-

FIG. 4. (Color online). Out-of-plane (a),(c) and in-plane (b), (d) vortex phase diagram for the superconducting state for FeS and FeS_{0.94}Se_{0.06}, respectively.

cause the second peak effect depends on temperature, such as in YBCO,²⁸ we cannot rule out the possibility of the second peak existence at much lower temperatures. So the region below the irreversibility line is likely plastic motion region. We establish the vortex phase diagram based on T_p and T_c obtained from Fig.1, as shown in Fig.4. The irreversibility line divides the phase diagram into vortex liquid and plastic motion region. We use the Werthamer-Helfand-Hohenberg (WHH) formula $H_{c2}(0) = 0.693[-(dH_{c2}/dT)]_{T_c} T_c$ to determine the upper critical field in the low-temperature region.²⁹ From our data, the upper critical fields are estimated to be $\mu_0 H_{c2}^c(0) = 0.37$ T, $\mu_0 H_{c2}^{ab}(0) = 2.14$ T for FeS, and $\mu_0 H_{c2}^c(0) = 0.40$ T, $\mu_0 H_{c2}^{ab}(0) = 2.30$ T for FeS_{0.94}Se_{0.06} whereas the anisotropy parameter $\gamma_H = \mu_0 H_{c2}^{ab}(0)/\mu_0 H_{c2}^c(0) \approx 5.8$ for both FeS and FeS_{0.94}Se_{0.06}. Small Se doping suppresses T_c and increases the upper critical field somewhat, but the results are in agreement with previous report from electrical transport measurements.⁸ All irreversibility lines can be fitted by Eq. (1), which gives $\beta_c = 1.28$ $\beta_{ab} = 1.32$ for FeS and $\beta_c = 1.17$, $\beta_{ab} = 1.04$, smaller than typical values of $\beta = 4/3 - 3/2$ in cuprate superconductors. However, it is evident that the irreversibility lines are much closer to H_{c2} when compared to 1111 iron based superconductors.¹⁷ This indicates strong pinning in both FeS and FeS_{0.94}Se_{0.06} and relatively stronger pinning in FeS_{0.94}Se_{0.06} when compared to FeS. This is consistent with the results of critical current measurement.¹¹

In summary, we report irreversibility fields and vortex dynamics in tetragonal FeS and FeS_{0.94}Se_{0.06} single crystals. We observed the crossover from a single flux surface pinning to the region of collective flux pinning on point-like defects, probably due to layered crystal structure and weak interlayer coupling in FeS-based superconductors. Our results indicate that new superconducting

materials with FeS_4 tetrahedra are likely to be good candidates for low-temperature and high-field applications if critical temperature could be raised. Given the recent discovery³⁰ of superconductivity below 14 K in BaFe_2S_3 with ladders of FeS_4 ,¹¹ search for new FeS-based superconductors could be of high interest.

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