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## Vortex dynamics and irreversibility line in $\text{FeSe}_{0.25}\text{Te}_{0.75}$

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### Abstract

AC multi-harmonic susceptibility measurements were performed in the  $\text{FeSe}_{0.25}\text{Te}_{0.75}$  superconductor. A large frequency-magnetic field-temperature ( $f$ - $H$ - $T$ ) region of the superconducting phase diagram was mapped under a wide range of the DC magnetic field. In particular the irreversibility line was accurately drawn up to 20 T as a function of both  $T$  and  $f$ . The frequency dependence of the irreversibility line was analyzed using the 3D vortex glass model, well describing the experimental behavior up to high magnetic field. The data evidence the evolution in time of the metastable critical state in a final stable vortex glass phase by decreasing the frequency. Moreover, from the Werthamer, Helfand and Hohenberg theory we estimated the upper critical field  $\mu_0 H_{c2}(0 K) \sim 62$  T. The observed 3D flux-pinning response points out that this system is suitable for carrying quite high critical currents.

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**Keywords:**  $\text{FeSe}_{0.25}\text{Te}_{0.75}$ ; flux pinning; AC multi-harmonic susceptibility; iron based superconductors; irreversibility line; upper critical field.

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## 1. Introduction

The discovery of  $\text{FeSe}_x\text{Te}_{1-x}$  compounds [1-3] belonging to the (11) family of iron chalcogenides attracted the attention of both theoreticians and experimentalists because of their simple crystal structure yet rich phase diagram where magnetic and superconducting phases may coexist [4,5] forming two networks at the nanoscale. Thanks to the high values of the upper critical field [6] and to the high critical current density [7]  $\text{FeSe}_x\text{Te}_{1-x}$  compounds are ideal candidates for many technological applications. However, the lack of understanding regarding the correlation dynamics between the flux lines in the mixed state represents a real bottleneck. A major obstacle for reliable technological applications is also the reduction of the dissipative phenomena due to the magnetic vortex motion inside the material. Therefore, in order to improve both the fabrication process and the application perspectives of these materials, a crucial issue is to investigate their flux-pinning and vortex dynamics properties. In this respect the accurate determination of the Irreversibility Line (IL) is important. This line divides the magnetic field-temperature phase diagram between a true superconducting vortex glass region where the flux lines are pinned and the current is carried without dissipation, and a vortex liquid region where the flux lines move in response to the applied current and the material shows an Ohmic behavior. In this contribution, using AC multi-harmonic susceptibility, we investigated the vortex dynamics in the  $\text{FeSe}_{0.25}\text{Te}_{0.75}$  superconductor. The AC susceptibility is indeed a powerful technique to explore the superconducting state, because it directly probes the dynamic magnetization of the material [8,9]. In particular the high harmonic coefficients ( $\chi_n$ ) probe the irreversible flux-pinning response of the superconducting phases occurring in the sample [10].

The work is mainly focused in the reconstruction of the temperature-magnetic field-frequency phase diagram in the superconducting state. In particular the irreversibility line was drawn in detail up to high magnetic fields ( $< 20$  T) as a function of temperature and frequency. Moreover, we extrapolated the irreversibility line in the limit of zero frequency, in the framework of the 3D vortex glass theory. By decreasing the frequency, the  $\chi_3$  temperature onset data show the time evolution of the critical state into a final stable vortex glass phase. The observed 3D flux-pinning behavior of the system in a wide range of the magnetic field points out the possibility to carry high critical currents in the material, even under high magnetic field. Finally, from the Werthamer, Helfand and Hohenberg (WHH) theory the upper critical field  $\mu_0 H_{c2}(0 K)$  was estimated  $\sim 62$  T [11]. These results help to clarify the complex behavior of the flux lines in the mixed state of this superconductor.

## 2. Experimental

The  $\text{FeSe}_{0.25}\text{Te}_{0.75}$  sample was synthesized in Tsukuba, Japan, by a solid state reaction technique from high purity powders of the constituent elements. Further details of the synthesis are given elsewhere [12]. AC susceptibility multi-harmonic measurements were performed at the High Field Magnet Laboratory (HFML) in Nijmegen, using a probe based on a set of two counter-wound pick-up coils, surrounded by a drive excitation coil, especially developed to work in the high field magnet ( $H_{dc}$  up to 33 T) [13]. The sample is mounted in a plastic holder at the center of one of the two coils while the other remains empty. With such a setup the background noise is predominantly compensated. The coil assembly is immersed in a thermally controlled He gas-flow cryostat which is mounted in the main magnet. The length of the probe is adjusted to place the center of the two pick-up coils in the uniform field region of the magnet, in order to reduce the background signal from the DC field.

All measurements were performed in the zero field cooling (ZFC) setup, i.e., the sample was slowly cooled without a magnetic field, then the field was switched on. The probed frequency range was 107 - 1070 Hz, in the AC magnetic field amplitude  $\mu_0 H_{ac} \sim 9.8$  G and DC field amplitude range 0 T – 20 T.

The critical temperature  $T_c(H)$  was obtained from the diamagnetic temperature onset of the first harmonic modulus  $\chi_1$  vs.  $T$  curves at each value of the DC magnetic field, as shown schematically in Fig. 1a (black arrow), where the measurements of the most representative fields at 507 Hz are reported. This was the frequency with the best signal to noise ratio. To increase the sensitivity, first derivative data were always analyzed for determining the onset.

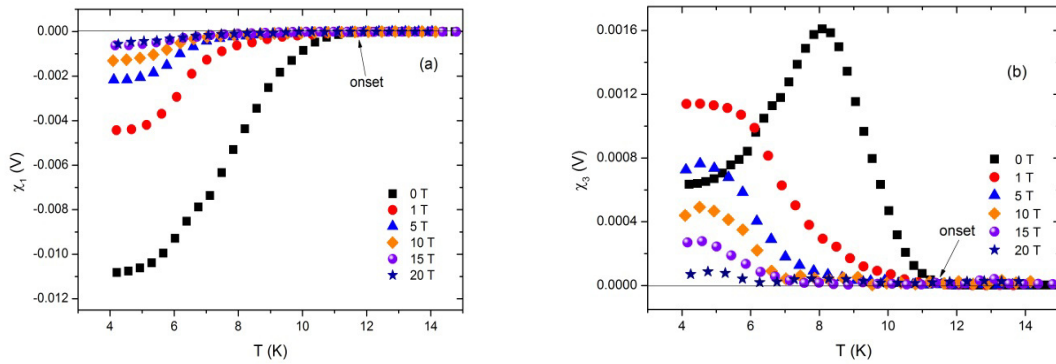


Fig. 1. AC susceptibility measurements vs. temperature and DC magnetic field at 507 Hz. The black arrow indicates the onset. (a) First harmonic modulus of the susceptibility  $\chi_1$  vs.  $T$ ; (b) third harmonic modulus of the susceptibility  $\chi_3$  vs.  $T$ .

For this sample,  $T_c(0 T) \sim 14$  K was found at zero field and 1070 Hz. Moreover, from these data the critical field curve  $\mu_0 H_{c2}(T)$  was deduced up to 20 T.

Following the same procedure, the irreversibility line was obtained from the temperature onset of third harmonic modulus  $\chi_3$  vs.  $T$  curves (Fig. 1b, black arrow). This method allows enhancing the sensitivity on the non-linear flux pinning dynamic processes in the superconducting phase. Indeed the higher harmonic components of the susceptibility,  $\chi_n$ , are the result only of the non-linear response of the material to the excitation of the AC field [14,10] and describe the change of the effective flux-diffusivity connected with the flux-pinning interaction [8]. In particular, in addition to directly discriminating between linear losses and non-linear flux-pinning response, compared with the other harmonics, the third one has the best signal to noise ratio [14].

### 3. Results and discussion

Fig. 2 shows the phase diagram of the  $\text{FeSe}_{0.25}\text{Te}_{0.75}$  superconductor deduced from the onsets of the  $\chi_1$  vs.  $T$  and  $\chi_3$  vs.  $T$  curves at 507 Hz (Fig. 1 a, b), as described in the previous section. The full squares mark the irreversibility line. Below this line, a non-reversible hysteretic behavior of the magnetization sets in, as a result of the disordered pinning of flux lines in the material forming a *vortex glass phase*.

This state is of particular interest for technological applications, because even in a magnetic field, it is a true superconducting phase where high current densities can be supported without dissipation. The region above the irreversibility line is the *vortex liquid phase*, where the thermal activation allows unpinning of flux lines which now move in response to the applied current, leading to Ohmic behavior and dissipation.

The full circles track the upper critical field  $\mu_0 H_{c2}(T)$ , obtained from the diamagnetic onset of the  $\chi_1$  vs.  $T$  curves. Data in the low field region near  $T_c$  have an almost linear dependence with a slope  $dH_{c2}/dT = -6.4 \pm 0.2$  T/K (see the linear fit in Fig. 2). Then, from the Werthamer, Helfand and Hohenberg theory [11]:

$$H_{c2}(0 K) = 0.693 T_c \left| \frac{dH_{c2}}{dT} \right|_{T \sim T_c}, \quad (1)$$

we evaluated  $\mu_0 H_{c2}(0 K) \sim 62$  T, in the limit of vanishing temperature and in agreement with the literature for a similar sample [15].

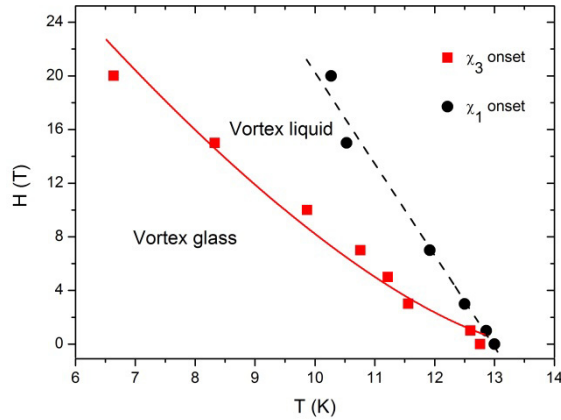


Fig. 2. Phase diagram of the FeSe<sub>0.25</sub>Te<sub>0.75</sub> at 507 Hz. The full squares mark the irreversibility line (the continuous line is the fit with the Eq. 2 in the text). The full circles track  $H_{c2}(T)$  (the dashed line is a linear fit).

The irreversibility line can be fitted with a power-law function (see the continuous line in Fig. 2):

$$H \propto (1 - T/T_c)^\beta, \tag{2}$$

with the exponent  $\beta = 1.4 - 1.5$ . This form, already found for cuprates [16, 17] and pnictides [18,19], describes the behaviour of a spin-glass system, i.e., the de Almeida-Thouless line [20]. Also the frequency behaviour of the irreversibility line was explored, plotting the temperature onset of  $\chi_3$  as a function of the frequency, at each value of the DC magnetic field. The resulting curves are reported in Fig. 3 again for the most representative fields.

In this case, a slight increase of the onsets with the frequency is observed, connected with the time evolution of the metastable superconducting phase described by the critical state towards a final stable vortex glass phase. Indeed this frequency dependence may be understood in the framework of the vortex glass theory. According to this model, near the vortex glass phase-transition temperature  $T_g$ , a power-law divergence of the correlation length  $\xi \propto |T-T_g|^{-\nu}$

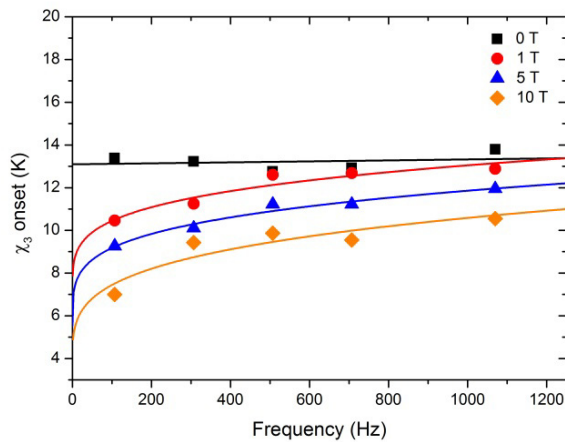


Fig. 3. Temperature onsets of  $\chi_3$  vs. frequency for the most representative DC magnetic fields. Continuous lines are fits with the Eq. 3 in the text. The straight line is the linear fit of data at 0 T.

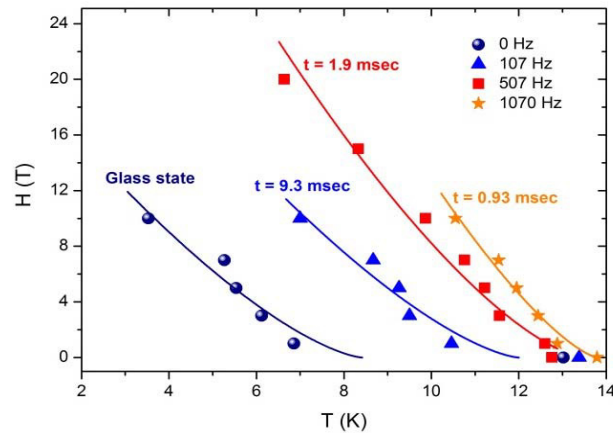


Fig. 4. Irreversibility line at the most representative frequencies. The full spheres track the irreversibility line in the limit of zero frequency, extrapolated from the fit of the  $\chi_3$  vs.  $f$  (Fig. 3) with the Eq. 3 in the text. Continuous lines are the best fits with the Eq. 2 in the text.

characterized by a critical exponent  $\nu$  is observed. Moreover, a critical slowdown of the dynamics near  $T_g$ , with a power-law divergence of the relaxation time  $\tau \propto |T - T_g|^{-\nu z}$ , where  $z$  is the dynamical critical exponent, is also expected. By inverting the latter relationship and taking care of the system dimensionality  $D$ , we obtained the frequency dependence of the irreversible temperature [21, 22]:

$$T_g(H, f) = T_g(H) + A(H) f^{1/\nu(z+2-D)}. \quad (3)$$

In the plot at  $\mu_0 H_{dc} = 0$  T, the  $\chi_3$  onsets are almost independent of the frequency as shown by the straight line in Fig. 3. A possible explanation is that the flux lines arrange in the vortex-glass phase with a characteristic creep time rate, faster than the explored experimental time window range. Using equation (3) we fitted the experimental data under the assumption of a 3D flux-pinning interaction and extrapolated the irreversible temperature  $T_g(H, 0)$  in the limit of the zero frequency at each DC field. The resulting irreversibility line is shown in Fig. 4 (full spheres), where we show also the IL of the metastable superconducting states at the most representative frequencies. The measurements at different frequencies are windows on the time evolution of the superconducting system in the H-T plane. In Fig. 4 the flux-pinning state time evolution is clearly pointed out. In the framework of the spin-glass theory the continuous lines are the best fits with the equation (2) and  $\beta = 1.4 - 1.5$  [20]. Deviations from the fit are observed at small fields near  $T_c$ , especially at low frequencies and may be explained with the occurrence in the sample of weak links among superconducting grains, really poorly connected near the critical temperature [23].

#### 4. Conclusion

A wide region of the superconducting  $\text{FeSe}_{0.25}\text{Te}_{0.75}$  phase diagram was mapped through multi-harmonic AC susceptibility measurements, as a function of temperature (4 – 25 K), DC magnetic field (< 20 T) and frequency (107 – 1070 Hz). The flux-pinning dynamics in this system were analyzed at high magnetic field and the irreversibility line was accurately drawn up to 20 T, as a function of both temperature and frequency. The IL experimental behavior can be reasonably well described in the framework of a 3D vortex-glass model up to high magnetic field applied, although deviations at small field and low frequency are observed near  $T_c$ . The 3D flux-pinning response of the  $\text{FeSe}_{0.25}\text{Te}_{0.75}$  system in a wide range of applied magnetic field points out the possibility of a significant increase of the critical current in this system. Moreover, data clearly show the time evolution of the metastable superconducting state in the final stable vortex glass phase. Finally, from the WHH theory an upper critical field  $\mu_0 H_{c2}(0 \text{ K}) \sim 62 \text{ T}$  was estimated. These results help to clarify the complex behavior of the flux lines in

the superconducting phase of this superconductor with a simple crystal structure but where magnetic and superconducting phases may coexist at the nanoscale.

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## References

- [1] Hsu, F.-C., Luo, J.-Y., Yeh, K.-W., Chen, T.-K., Huang, T.-W., Wu, P. M., Lee, Y.-C., Huang, Y.-L., Chu, Y.-Y., Yan, D.-C. and Wu, M.-K., 2008. Superconductivity in the PbO-type structure  $\alpha$ -FeSe. *Proceedings of the National Academy of Sciences of the United States of America* 105, 14262–14264.
- [2] Yeh, K.-W., Huang, T.-W., Huang, Y.-L., Chen, T.-K., Hsu, F.-C., Wu, P. M., Lee, Y.-C., Chu, Y.-Y., Chen, C.-L., Luo, J.-Y., Yan, D.-C. and Wu, M.-K., 2008. Tellurium substitution effect on superconductivity of the  $\alpha$ -phase iron selenide. *Europhysics Letters* 84, 37002.
- [3] Sales, B. C., Sefat, A. S., McGuire, M. A., Jin, R. Y., Mandrus, D. and Mozharivskiy, Y., 2009. Bulk superconductivity at 14 K in single crystals of  $\text{Fe}_{1+y}\text{Te}_x\text{Se}_{1-x}$ . *Physical Review B* 79, 094521.
- [4] Fang, M. H., Pham, H. M., Qian, B., Liu, T. J., Vehstedt, E. K., Liu, Y., Spinu, L. and Mao, Z. Q., 2008. Superconductivity close to magnetic instability in  $\text{Fe}(\text{Se}_{1-x}\text{Te}_x)_{0.82}$ . *Physical Review B* 78, 224503.
- [5] Khasanov, R., Bendele, M., Amato, A., Babkevich, P., Boothroyd, A. T., Cervellino, A., Conder, K., Gvasaliya, S. N., Keller, H., Klauss, H.-H., Luetkens, H., Pomjakushin, V., Pomjakushina, E. and Roessli, B., 2009. Coexistence of incommensurate magnetism and superconductivity in  $\text{Fe}_{1+y}\text{Se}_x\text{Te}_{1-x}$ . *Physical Review B* 80, 140511(R).
- [6] Klein, T., Braithwaite, D., Demuer, A., Knafo, W., Lapertot, G., Marcat, C., Rodière, P., Sheikin, I., Strobel, P., Sulpice, A. and Toulemonde, P., 2010. Thermodynamic phase diagram of  $\text{Fe}(\text{Se}_{0.5}\text{Te}_{0.5})$  single crystals in fields up to 28 tesla. *Physical Review B* 82, 184506.
- [7] Tsurkan, V., Deisenhofer, J., Günther, A., Kant, Ch., Klemm, M., Krug von Nidda, H.-A., Schrettle, F., Loidl, A., 2011. Physical properties of  $\text{FeSe}_{0.5}\text{Te}_{0.5}$  single crystals grown under different conditions. *The European Physical Journal B* 79, 289–299.
- [8] Di Gioacchino, D., Marcelli, A., Puri, A., Bianconi, A., 2010. The a.c. susceptibility third harmonic component of  $\text{NdO}_{1-0.14}\text{F}_{0.14}\text{FeAs}$ : A flux dynamic magnetic analysis. *Journal of Physics and Chemistry of Solids* 71, 1046–1052.
- [9] Poccia, N., Ricci, A., Campi, G., Fratini, M., Puri, A., Di Gioacchino, D., Marcelli, A., Reynolds, M., Burghammer, M., Saini, N. L., Aeppli, G. and Bianconi, A., 2012. Optimum inhomogeneity of local lattice distortions in  $\text{La}_2\text{CuO}_{4+y}$ . *Proceedings of the National Academy of Sciences of the United States of America* 109, 15685–15690.
- [10] Di Gioacchino, D., Celani, F., Tripodi, P., Testa, A. M. and Pace, S., 1999. Nonuniversal temperature dependencies of the low-frequency AC magnetic susceptibility in high- $T_c$  superconductors. *Physical Review B* 59, 11539.
- [11] Werthamer, N. R., Helfand, E. and Hohenberg, P. C., 1966. Temperature and Purity Dependence of the Superconducting Critical Field,  $H_{c2}$ . III. Electron Spin and Spin-Orbit Effects. *Physical Review* 147, 295.
- [12] Mizuguchi, Y. and Takano, Y., 2010. Review of Fe Chalcogenides as the Simplest Fe-Based Superconductor. *Journal of the Physical Society of Japan* 79, 102001.
- [13] Wiegers, S. A. J., Christianen, P. C. M., Engelkamp, H., den Ouden, A., Perenboom, J. A. A. J., Zeitler, U., Maan, J. C., 2010. The High Field Magnet Laboratory at Radboud University Nijmegen. *Journal of Low Temperature Physics* 159, 389–393.
- [14] Shaulov, A. and Dorman, D., 1988. Investigation of harmonic generation in the alternating magnetic response of superconducting Y-Ba-Cu-O. *Applied Physics Letters* 53, 2680.
- [15] Kida, T., Matsunaga, T., Hagiwara, M., Mizuguchi, Y., Takano, Y. and Kindo, K., 2009. Upper Critical Fields of the 11-System Iron-Chalcogenide Superconductor  $\text{FeSe}_{0.25}\text{Te}_{0.75}$ . *Journal of the Physical Society of Japan* 78, 113701.
- [16] Malozemoff, A. P., Worthington, T. K., Yeshurun, Y., Holtzberg, F. and Kes, P. H., 1988. Frequency dependence of the AC susceptibility in a Y-Ba-Cu-O crystal: A reinterpretation of  $H_{c2}$ . *Physical Review B* 38, 7203(R).
- [17] Yeshurun, Y. and Malozemoff, A. P., 1988. Giant Flux Creep and Irreversibility in an Y-Ba-Cu-O Crystal: An Alternative to the Superconducting-Glass Model. *Physical Review Letters* 60, 2202.
- [18] Prando, G., Carretta, P., De Renzi, R., Sanna, S., Palenzona, A., Putti, M. and Tropeano, M., 2011. Vortex dynamics and irreversibility line in optimally doped  $\text{SmFeAsO}_{0.8}\text{F}_{0.2}$  from AC susceptibility and magnetization measurements. *Physical Review B* 83, 174514.
- [19] Prando, G., Carretta, P., De Renzi, R., Sanna, S., Graf, H.-J., Wurmehl, S. and Büchner, B., 2012. AC susceptibility investigation of vortex dynamics in nearly optimally doped  $\text{RFeAsO}_{1-x}\text{F}_x$  superconductors (R = La, Ce, Sm). *Physical Review B* 85, 144522.
- [20] de Almeida, J. R. L. and Thouless, D. J., 1978. Stability of the Sherrington-Kirkpatrick solution of a spin glass model. *Journal of Physics A: Mathematical and General* 11, 983.
- [21] Fisher, D. S., Fisher, M. P. A. and Huse, D. A., 1991. Thermal fluctuations, quenched disorder, phase transitions, and transport in type-II superconductors. *Physical Review B* 43, 130.
- [22] Wolfus, Y., Abulafia, Y., Klein, L., Larkin, V. A., Shaulov, A. and Yeshurun, Y., 1994. Diverging Time Scales for Onset of Irreversibility in High-Temperature Superconductors. *Physica C* 224, 213–220.
- [23] Yeshurun, Y., Felner, I. and Sompolinsky, H., 1987. Magnetic properties of a high- $T_c$  superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_7$ : Reentry-like features, paramagnetism, and glassy behavior. *Physical Review B* 36, 840(R).