Flux Dynamics in Iron-Based Superconductors

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Abstract—The newly discovered iron-based superconductors share a common layered structure. After the iron pnictide (1111) family, other iron-based superconductors such as the iron chalcogenides FeTe$_{1-x}$Se$_x$ (11) family have been synthesized. The latter system, characterized by a simple crystal structure, represents an interesting reference system. Indeed, all iron-based superconductors have a stacked structure composed of a layer of iron atoms linked by tetrahedrally coordinated pnictogens (P, As) or a chalcogen (Se, Te) anion: the active layer. The latter is either simply stacked together, as in the FeSe (11), or separated by spacer layers with alkali (e.g., Li), alkaline earth (e.g., Ba), or rare earth oxides/fluorides. In this contribution, we present an ac multiharmonic susceptibility study of the flux dynamic in representative 1111 and 11 iron-based superconductors. Data analysis has been performed in the glass-weak pinning scenario. In particular, the comparison of the third harmonic components vs. temperature and magnetic field returned information on pinning strength and dimensionality. Although in the presence of large thermal fluctuations, because of the high $T_C$, susceptibility data points out a three-dimensional flux dynamic and, unexpectedly, an increase of the pinning amplitude in the Fe-based superconductor systems where the spacer layer is present.

Index Terms—FeSe$_x$, FeSe$_{1-x}$Te$_x$, flux pinning regimes, high magnetic resonant selectors, NdO$_{1-x}$F$_x$FeAs, SmO$_{1-x}$F$_x$FeAs.

I. INTRODUCTION

Among the many types of superconductors (FeSCs) with high critical temperatures triggered many new researches [1]. In fact, these systems are characterized by a quite simple crystalline structure: iron-pnictogen (P, As) [1]–[4] or -chalcogen (S, Se, Te) [5]–[9] superconducting active layers, separated by spacer layers with alkali (Li), alkaline earth (Ba), or rare earth (RE) oxide/fluoride (NdO or SrF), or simply stacked together, as in the FeSe, a material composed with only two elements, certainly the simplest crystalline structure among all iron-based superconductors. However, because superconductivity and magnetism were originally considered as mutually exclusive phenomena, the presence in these systems of a strongly magnetic atom such as $Fe$, makes their study particularly challenging. Indeed, the study of the magnetic response of superconductors is important for practical applications. Actually, the study of the flux dynamic motion is fundamental to determine the pinning characteristics of high temperature superconductors $(HTSC)$, a critical issue that indicates how high can be the critical current carried out by a material using the critical state model [10]. In the 3D magnetic field-current density-temperature $(B – J – T)$ experimental space of a HTSC we may recognize regions where different flux pinning interactions dominate the flux dynamics response of a superconductor [11]. The third harmonic component of the AC susceptibility [12], [13] is the best tool for this kind of analysis not only because it may give information on the non-linear losses, probing directly flux pinning processes [14]–[18] and critical current values, [10] but also because it may recognize the presence of multiple superconducting phases in a sample [12], [19]. In this contribution, from AC multi-harmonic susceptibility data we will discuss flux dynamic information clarifying the pinning types and the dimensionality of the pinning processes occurring in 1111 iron-pnictides and 11 iron-chalcogenides.

II. EXPERIMENTAL SET-UP

The $NdFeAsO_{1−0.14}F_{0.14}$ and $SmFeAsO_{0.85}F_{0.15}$ samples were synthesized in China, the first by high pressure synthesis method from $Nd$ pieces, $As$, Fe, $Fe_2O_3$, $FeF_3$ powders [3], and the second by solid state reaction from stoichiometric amount of $SmAs$, $SmF_3$, $FeAs$ and $Fe_2O_3$ powders and $Sm$ [20]. The $FeSe_{0.88}$, $FeSe_{0.5}Te_{0.5}$ and $FeSe_{0.25}Te_{0.75}$ samples were synthesized in Japan, by solid-state reaction technique from high purity powders of the constituent elements. Further details of the synthesis of these samples are in [21]. The samples were polycrystalline slabs $(10 \times 4 \times 2)$ mm and the $H_{ac} + H_{dc}$ magnetic field was oriented along the 10 mm size.

AC multi-harmonic susceptibility measurements were performed at the LAMPS laboratory of the National Laboratory of Frascati (I.N.F.N.). The susceptibility was measured by a gradiometer based on a bridge of two pick-up coils, connected in series and wounded in the opposite sense, and surrounded by a drive excitation coil. The coils assembly was immersed in a thermally controlled He gas-flow cryostat where a superconducting magnet operated up to 8 T. The AC driving magnetic field frequency may range from 107 to 1070 Hz with variable amplitude up to 10 G. A multi-harmonic lock-in amplifier SIGNAL RECOVERY model 7265 DSP measured the induced signal by the sample. A Pt thermometer probed the temperature together with a carbon resistor placed near the sample and in contact with the sapphire holder.
Fig. 1. Third harmonic $|\chi_3|$ module of the NdFeAsO$_{1-0.14}$F$_{0.14}$ at different frequencies of the ac magnetic field, for $H_{dc} = 0$ T (a) and $H_{dc} = 1$ T (b).

All measurements were performed with the zero-field-cooling (ZFC) set-up, i.e., the sample was slowly cooled down to 4.4 K without magnetic field, then the field was turned on. Analysis have been performed considering the weight of the measured sample volume.

III. RESULT AND DISCUSSION

The frequency dependence of the third harmonic susceptibility $\chi_3$ vs. temperature describes the change of the effective non-linear flux-diffusivity connected with the flux-pinning interaction. As a consequence $\chi_3$ is connected to the critical current $J_c$ [10], in particular, $\chi_3$ is proportional to this value in the temperature range where the penetration depth of the field $B_p$ is less than of the applied magnetic field $H_{app} = H_{ac} + H_{dc}$ and the sample is completely penetrated. In this configuration a large critical current flows in the entire sample volume. The behavior of the $|\chi_3|$ amplitude showed in Fig. 1 indicates for $H_{dc} = 0$ T a weak temperature dependence by the frequency. In fact, the variation of the temperature is small around $\Delta T (frequency) = 0.25$ K, while for $H_{dc} = 1$ T, $\Delta T$ increases only $\sim 1$ K ($\Delta T = 1.25$ K). These regions are outlined as hatched areas in Fig. 1. In the temperature range analyzed, for both values of the DC field, the $|\chi_3|$ amplitudes show higher value at the lowest frequency (107 Hz). The result points out that after the ZFC set-up procedure, the initial superconducting flux dynamic state evolves into a final flux “glass state” with a high enough value of the critical current, emphasizing the occurrence of a strong pinning.

Fig. 2 (H$_{dc} = 0$ T, panel A) shows the same analysis for the FeSe$_{0.88}$ sample. In this sample, still considering that the $|\chi_3|$ amplitude fully penetrate as in the NdFeAsO$_{1-0.14}$F$_{0.14}$ case, we may point out two issues: a) the $|\chi_3|$ values respect to frequency (hatched area) involves a larger temperature range ($\Delta T \approx 0.5$ K); b) the $|\chi_3|$ amplitude shows a small but
opposite frequency trend: as the frequency increases, the amplitude slightly decreases, i.e. the critical current is reduced and shows a weaker pinning during the time evolution. The trend is confirmed by data collected with a small field ($H_{dc} = 0.1$ T) that show only partial $|\chi_3|$ curves limited by the liquid He temperature. Finally, the evolution of the initial critical state in the final glass state is characterized by a clear decrease of the critical current, a behavior compatible with a weaker flux pinning for the $11-FeSe_{0.88}$ if compared to the $1111-NdFeAsO_{1-0.14}F_{0.14}$.

To clarify the time (frequency) behavior showed in Figs. 1 and 2, we underline that each point of the $|\chi_3|$ module vs. temperature is connected to the relationship $\delta = B_{dc}/B_p$ between the applied field and the induced magnetic field penetrated in the sample ($B_p$) in turn proportional to $J_c(B,T)$ [13]. $B_p$ is determined by the metastable equilibrium between the pinning and the Lorentz force at a fixed time (critical state model). The metastable state $B_p$ (or $J_c$) evolves in time (or frequency), i.e., a $J_c(H,T,t)$ dependence occurs. Actually, the value of the $|\chi_3|$ temperature peak (as each $|\chi_3|$ point) defined at a given time (frequency) evolves and the same state will be later recovered at a lower temperature.

In the $B-T-J$ phenomenological diagram (Fig. 3) we qualitatively show the evolution of the two different flux pinning dynamic processes [1111—Panel A and 11—Panel B]. Actually, the current can be considered the time variable through the relation $J(t) \approx [\ln(t/t_0)^{-1/\mu}]$ while $\mu$ is the glass parameter that probes the different pinning regimes [11]. In the next we also discuss some Fe $-$ HTSC compounds of both families.

In Fig. 4, we compared the third harmonic modulus as a function of the reduced temperature $t$ for the $NdFeAsO_{1-0.14}F_{0.14}$, $SmFeAsO_{0.85}F_{0.15}$, $FeSe_{0.88}$, $FeSe_0.5T_{c0.5}$, $FeSe_0.25T_{c0.75}$ and $FeTe_{0.8}S_{0.2}$. In particular, in the comparison between $FeSe_0.5T_{c0.5}$ and $SmFeAsO_{0.85}F_{0.15}$ we considered the small grain signal near $T_{c}$. The $NdFeAsO_{1-0.14}F_{0.14}$ is characterized by a more intense curve and a narrower temperature range respect to other samples, which are broader and weaker. All features enhance after the application of a DC field (Fig. 4 panel B). It is then clear that the strain structural feature due to the “space layer” affects the flux-pinning dynamic of Fe $-$ HTSC superconductors. In fact, if we compare only the grain response Fig. 4(c), the $|\chi_3|$ intra-grain peak of the $SmFeAsO_{0.85}F_{0.15}$ has a sharper peak respect to the $FeSe_{0.5}T_{c0.5}$, where the $|\chi_3|$ curve appears broad and weaker.

The analysis confirms that the pinning in iron-chalcogenides is less efficient for these grains. To gain more insight on the flux pinning characteristics, a dimensional analysis is compulsory. We investigated the Irreversibility Line (IL) of both $NdFeAsO_{1-0.14}F_{0.14}$ and $FeSe_{0.88}$ in a vortex glass-weak pinning regime (Fig. 5). In this framework, $T_R$ is the low-temperature vortex glass phase. The irreversibility temperature ($T_{irr}$) will approach the glass transition temperature $T_g$ in the limit of zero frequency. For each value of the frequency we used for all applied $H_{dc}$ the $T_{onset}$ of the $|\chi_3|$, defined as $T_{irr}$. To obtain $T_g$ we fit the $|\chi_3|$ onset using the method of [23] derived by Wolfs et al. [22] for the 3D case.

Fig. 3. In the B-T-J experimental space [11] of (a) the NdFeAsO1-0.14F0.14 and (b) the FeSe0.88 the current axes is the time variable. After the ZFC temperature procedure (first arrow), HDC has been switch on bringing the sample in a well-defined meta-stable critical state (second arrow). Later it evolves in time in a final stable glass state (third arrow). Other metastable critical states probed at specific time windows defined by the frequency values of the measurements are represented.1

Using the equation below we may estimate the dimensionality ($D$) of these systems:

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

where we have defined: $T_{irr}(H,f)$ as the irreversible temperature; $T_g(H)$ as the glass temperature; $A(H)$ the coefficient depending on the magnetic field applied; $\nu$ and $z$, the static and the dynamic parameters of the glass theory. In Table I, we reported $T_g(H)$, $\nu$ and $z$ values from the fit of Fig. 5 for both samples. These values are consistent with the typical values of the critical exponents of the 3D scaling, while for the NdFeAsO1-0.14F0.14 at 1 T the fit is not consistent [25], [26] and we point out the occurrence a 2D behavior. At $H_{dc} = 0$ T, the $|\chi_3|$ onset of $NdFeAsO_{1-0.14}F_{0.14}$ is independent of the frequency, i.e., the system moves to a final vortex-glass phase with a characteristic creep time rate faster than the explored experimental time window range and shows a three-dimensional (3D) behavior. Also increasing the $H_{dc}$ magnetic field superimposed on a small AC
Fig. 4. Comparison of the modulus of $|\chi_3|$ for NdFeAsO$_{1-0.14}F_{0.14}$, FeSe$_{0.88}$, FeSe$_{0.25}Te_{0.75}$, and FeTe$_{0.8}$ at (a) $H_{dc} = 0$ T and (b) $H_{dc} = 1$ T and for the intragrain signal in the SmFeAsO$_{0.85}F_{0.15}$ and the (c) FeSe$_{0.5}Te_{0.5}$.

Excitation field in the range of 0.1–0.5 T both systems $NdFeAsO_{1-0.14}F_{0.14}$ and $FeSe_{0.88}$ exhibit a 3D flux dynamic. For the $NdFeAsO_{1-0.14}F_{0.14}$ system we also studied the system at $H_{dc} = 1$ T that shows a flux dynamic dimensional transition with a two-dimensional behavior. In other words, if increasing the $DC$ field up to 1 T the FeAs active planes of the 1111 family appear decoupled.

IV. CONCLUSION

In this contribution we compared the ac susceptibility third harmonic modulus of iron-pnictide and iron-chalcogenide superconductors. The analysis point out, under the same condition of frequency and magnetic field, that the first superconductor type have a stronger pinning force, even in the presence of larger thermal fluctuations because of their higher $T_c$. In fact, the $NdFeAsO_{1-0.14}F_{0.14}$ and the $SmFeAsO_{0.85}F_{0.15}$ have the highest $|\chi_3|$ peak intensity. Considering that in iron-pnictides the weak contribution of the $F$ doping flux pinning is comparable to the same mechanism induced by Se vacancies in the $FeSe_{0.88}$ or by $Te$ substitution in $FeSe_{1-x}Te_x$ or...
FeTe\textsubscript{0.8}S\textsubscript{0.2}, the stronger pinning observed in iron-pnictides clearly points out the role of the [Nd(Sm)O] extra layer. We claim that the larger flux pinning may be associated to a coherent strain induced by Nd(Sm)O layers on the superconducting Fe\textsubscript{As} layers. [27], [28].

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REFERENCES


