Flux Dynamic Changes by Neutron Irradiation in BISCCO: High Harmonics AC Susceptibility Analysis

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***Abstract*—High harmonics ac susceptibility measurements joint with neutron irradiations on quasi-bi-dimensional high temperature superconductor (quasi-2D-HTSC) are good tools to study the flux dynamics and its interaction with pinning processes in these superconductors. Flux neutron intensity of $5 * 10^{17}$ n $* \mathrm{cm}^{-2}$ shows a deep change in the flux pinning dynamics in Bi-Sr-Ca-Cu-O (BSCCO) system. Third harmonic susceptibility signal increases in amplitude after the neutron irradiation, followed by a rise of the pinning and critical current. Moreover, after the irradiation, the measurements underline the demise of the anomalous peak effect (PE) associated with a three-dimensional/two-dimensional (3D/2D) flux lattice transition.**

***Index Terms*—BiSCCO, flux pinning regimes, high harmonic magnetic ac susceptibility measurements, neutron irradiation.**

I. Introduction

THREE-DIMENSIONAL phenomenological phase diagram magnetic field-temperature-current density (H-T-J), for high temperature superconductors (HTSC), is divided in several separate regions where different flux pinning interactions dominate flux dynamics response of the superconductors via different critical current ($J_c(A/cm^2)$), dependences on temperature (T(K)) and magnetic field (H(T)) [1]. In the quasi-bi-dimensional HTSC (quasi-2D-HTSC) like Bi-Sr-Ca-Cu-O (BSCCO) compounds, as well known, the disorder gives random pinning potential energy for the fluxons correlated with the same superconducting layer and uncorrelated in different layers [1]. The pinning in this case, is less efficient because of the large thermal fluctuation influence, with a consequent limitation of the critical current. Regarding these bi-dimensional superconductors, what is needed is to increase the pinning force in the H-T-J regions operative in the experimental applications.

One of the most efficient ways to improve the flux pinning capability consists of exposing these materials to a suitable irradiation environment. Large pinning enhancement is achieved by irradiation with protons and other ions, but these three-dimensional columnar defects are not efficient for the decoupled 2D pancake vortices [2] dominant in the 2D-HTSC flux dynamics. The enhancement of flux pinning after fast neutron irradiation is obtained in different HTSC tapes [3]–[10]. In 2D-HTSC materials the neutron irradiation gives more efficient pinning due to the cascade defects of 50–100 Å in diameter and point defects produced in the individual copper oxides (CuO_2) planes [2], [9]. It is useful to understand these effects on the flux dynamics in composite superconductors that are of great importance for technological applications, such as silver sheathed $(Bi,Pb)_2Sr_2Ca_2Cu_3O_{10}$ (Bi-2223) tapes [11]. In this paper $Bi_{1.8}Pb_{0.4}Sr_{1.8}Ba_{0.2}Ca_2Cu_3O_x$ bulk systems irradiated by neutrons are examined. Some experimental data on flux dynamics is analyzed using ac multi-harmonic susceptibility measurements. The results are compared with the un-irradiated half portion of the samples.

II. Experimental Set-Up

Samples with the chemical composition $Bi_{1.8}Pb_{0.4}Sr_{1.8}Ba_{0.2}Ca_2Cu_3O_x$ were prepared by using the standard solid-state reaction method. According to X ray diffraction (XRD) measurements the sample is approximately 97% Bi-2223 phase. Samples were irradiated using fast neutron channel of 'Training Research Isotopes General Atomics' (TRIGA) reactor in standard aluminum blocks suspended in the center of the channel 36/6 inside the hot chamber of the nuclear reactor in "Horia Hulubei" National Institute for Nuclear Physics and Engineering in Bucharest, (IFIN-HH). The neutron flux density is $2.13 \times 10^{13}\ \mathrm{cm}^{-2} * \mathrm{s}^{-1}$ and the final fluency is $5 \times 10^{17}\ \mathrm{cm}^{-2}$.

During the irradiation, the sample temperature was not measured, but the channel temperature was 40°C. After irradiation, the samples remained in the hot chamber for 7 days in order to remove the residual activity. The non-irradiated sample is named "NIR" while that irradiated is defined "IR".

The ac-susceptibility, including high harmonic components, was measured with a first derivative gradiometer based on double pick-up coil surrounded by a driven coil [12]. The sample was mounted on a sapphire holder inserted in the pick up coils. Temperature was measured with a platinum thermometer (PT100) in a good thermal contact with the samples. The whole assembly was cooled in zero magnetic field (ZFC), in a thermally controlled He gas flow cryostat provided with an 8 T superconducting magnet. Measurements were done sweeping the temperature with a rate of 0.3 K/min up to a temperature greater than the zero field critical temperature of the samples (between 10 and 120 K). ac driving magnetic field

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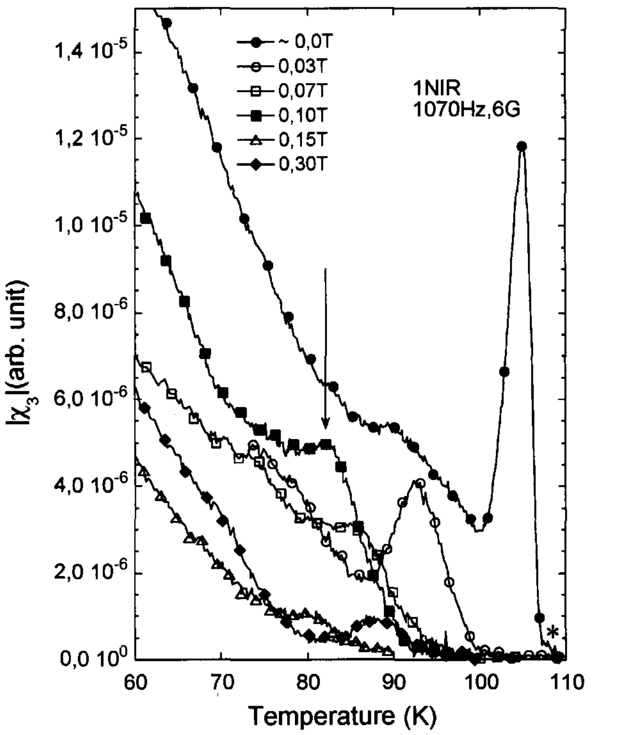

Fig. 1. The $|\chi_3|$ modules versus temperature in different magnetic dc field for non-irradiated sample (1NIR). The arrow indicates the anomalous peak, the $*$ point to the de-couple grain signal.

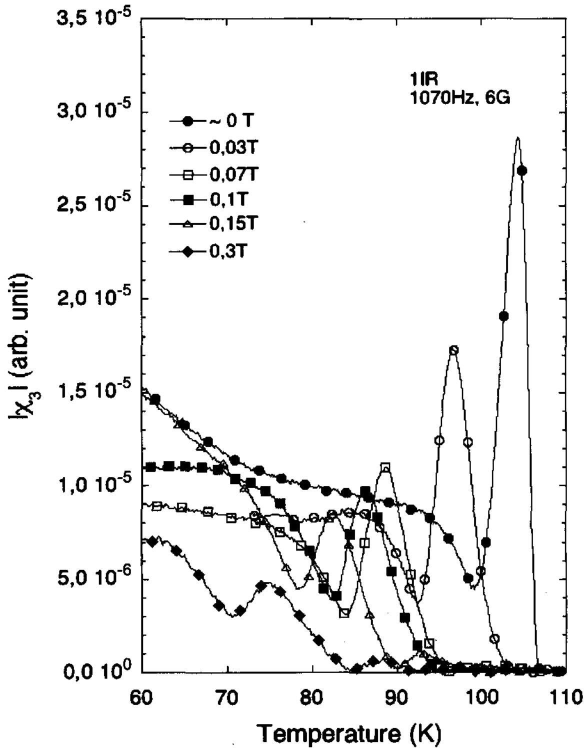

Fig. 2. The $|\chi_3|$ module for irradiated sample (1IR) in different magnetic dc field. Same experimental conditions of the previous non-irradiated sample have been adopted. The anomalous peak effect (PE) is not present.

had an amplitude of 6 Gauss at a frequency f $=$ 1070 Hz. dc magnetic fields were swept from 0 to 1.5 T. Induced signal was measured with a multi-harmonic EG&G lock-in amplifier. Both ac and dc fields were applied in parallel to the longest size of the sample.

III. Results and Discussion

The ac high harmonic components give information on the non-linear losses directly connected with the flux pinning processes [13], critical current values [14] and are able to detect in the samples multiple superconducting phases [15].

Figs. 1 and 2, show the module of third harmonic $|\chi_3|$, for sample 1NIR and 1IR in function of the temperature for different dc magnetic field amplitudes (H_{dc}) in the range [$0\ T \leq H_{dc} \leq 0.3\ T$].

The $|\chi_3|$ behaviors indicate two distinct phases connected with inter-granular properties, one exhibits an evident peak, and the other shows shoulders at lower temperatures. 1NIR sample also exhibits a very small peak amplitude due to the superconducting response of the de-coupled crystals at $H_{dc} = 0$ T very close to T_c ($*$ in Fig. 1).

It is well known, that increasing the applied field, the peak shifts to lower temperature and the peak amplitude decreases. In our case the behavior and flux dynamics structure appears more complex. In the 1NIR sample (Fig. 1), an anomalous peak effect (PE) takes place (indicated with an arrow). In the PE range the pinning response increases. Normally this superconducting properties improvement is explained as a crossover transition from three-dimensional (3D) to quasi-two dimensional (quasi-2D) vortex dynamics [16]–[21]. The 2D-flux pancakes tune better to the random pinning present on the CuO_2 planes. The value of the crossover magnetic field in this case is $H_{dc} = H_{cr} = 0.1$ T. For values of magnetic fields around H_{cr} the peak amplitude rate decreases faster. This fact confirms the transition between the two flux transports, where "2D pancake/pinning" couplings are weaker than the "3D fluxon/pinning" interactions.

The $|\chi_3|$ amplitudes of the 1IR sample (Fig. 2) exhibit an increase in respect to the NIR values in the whole of the magnetic range explored, furthermore, the PE effect vanished. This is better evidenced in Fig. 3 where the $|\chi_3|$ peak versus H_{dc} is shown for the two samples.

Flux dynamics analysis can be done using the 'brick-wall' model [22]. This model, considers the microstructure of the sample. In schematic representation, the inter-granular superconducting transport can be affected by two (or more) different weakly linked Josephson Junctions (JJ), one with a stronger superconducting connection in respect to the other. The 1IR sample at low magnetic field (Fig. 3) shows that all JJ types between the grains are connected and the inter-granular pinning features display the power law [22]. When the crossover magnetic field (H_{cr}) is achieved, weaker JJ are broken and only strong ones remain. If the induced current in the sample, is lower in respect to the strong JJ critical current, J_c^{sjj}, the transport is determined by the intra-grain pinning behavior described by the exponential law [22]. In this law the parameter strongly depends on T(K), type and concentration of defects. If the induced current is greater than J_c^{sjj} only the thermal fluctuations are important so the pinning regime can be ignored, but the presence of new defects can reduce the thermal fluctuation of pancakes.

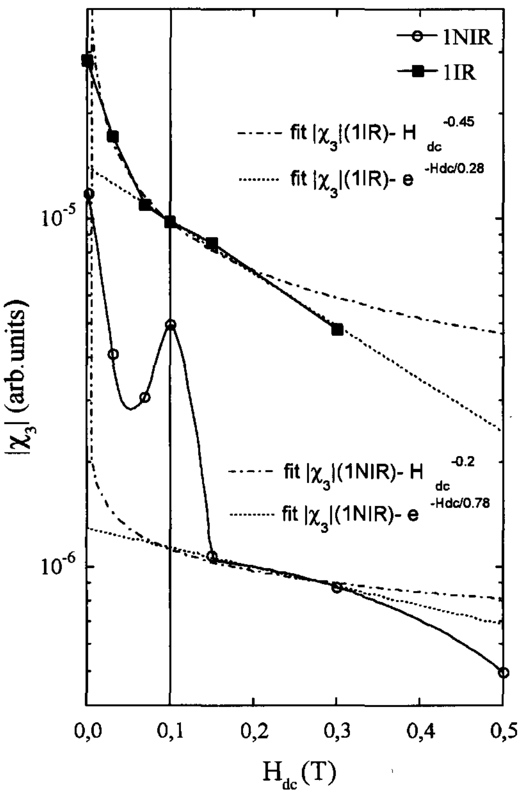

Fig. 3. The $|\chi_3|$ peak in function of dc magnetic field for not irradiated and irradiated samples (1NIR, 1IR).

Also in this case the exponential law describes the transport behavior [22]. This controversial point remains unclear, but considering the magnetic and temperature ranges explored in this paper, the density current is not greater than the inter-granular strong J_c^{sjj}. For this reasons, the $|\chi_3|$ exponential behavior (Fig. 3) describes the intra-grain pinning processes and not the thermal fluctuations.

The mathematical expressions of the critical current in two flux dynamic regimes are [22]:

A) $J_c \propto B^{-\nu}$, (low magnetic field)
B) $J_c \propto \exp(-B/B_0)$, (high/medium magnetic field)

In the A) case 'power law', the parameter exponent ν depends on the pinning strength disorder between the grains.

While in the B) case the exponential magnetic behavior can be described by intra-granular pinning mechanism.

In irradiated sample (Fig. 3), the non-linear (pinning) response increase is mainly due to the pinning centers enhancement, which is a normal consequence of neutron irradiation. In an irradiated sample with nuclear reactor neutron flux, some nuclear reactions take place. In our case, "n + ^{63}Cu" is considered relevant. The reaction channels are principally two apart from elastic scattering:

Reaction Products	Q-value/ Threshold (MeV)	Cross section barn
$^{60}Co+\alpha$	1.71448/0.0	$1.8\ 10^{-3}$
$^{63}Ni+p$	0.71541/0.0	$7.9\ 10^{-2}$
$^{63}Cu+n$	0.0/0.0	2.03

Three months after the irradiation, the gamma counter (slow measurement for 24 hours) measured the presence of a great amount of ^{60}Co. The measurement has confirmed the first nuclear reaction occurrence shown in table. This reaction produces a double effect on pinning:

a) substitution of ^{63}Cu (paramagnetic atom) with ^{60}Co (magnetic atom) due to different 3d orbitals filling. This brings about a change in the electronic structure of the $Cu - O_2$ planes;
b) production of helium atoms (α) of 1.71 MeV, leading to a cascade of defects, spatially uncorrelated, grater than point defects.

Second nuclear reaction of the table produces similar pinning effects:

a) substitution of ^{63}Cu (paramagnetic atom) with ^{63}Ni (magnetic atom) with substantial change in the electronic structure in the $Cu - O_2$ planes;
b) production of protons, generating uncorrelated point defects [2].

Slightly different superconducting material described as $Bi_{1.8}Pb_{0.4}Sr_{1.8}Ba_{0.2}Ca_2Cu_{3\delta}Co_\alpha Ni_\beta O_x$ where $\delta = \alpha + \beta$ has been produced. From the neutron fluency and cross section of the reaction products, we expect to have 48×10^{13} of ^{60}Co and 19×10^{15} of ^{63}Ni atoms in this new superconducting material.

These new defects are efficient in individual CuO_2 planes causing an enhanced pinning in the 2D-flux pancakes as well as in the 3D-flux line. The irradiation changes the flux dynamic in the temperature-magnetic ranges studied. The sharp decoupling flux pinning transition, known as PE, present in the non-irradiated sample at $H_{dc} = 0.1$ T vanishes (Fig. 3).

It is remarkable (Fig. 3) that the field crossover value ($H_{cr} = 0.1$ T) between the two inter-granular flux regimes for irradiated and non-irradiated samples does not change. The decoupling of the 'weak JJ' between grains remains unalterable by neutron irradiation, in other words the tilt between the grain borders does not change. 1IR flux dynamics behavior is in contrast with pinning behavior that similar HTSC [23] exhibit, i.e. $(B_{0.65}C_{0.35})Ba_{1.4}Sr_{0.6}Ca_2Cu_3O_x$ sample, irradiated with same neutron fluency by same TRIGA nuclear reactor. This difference can be attributed to the Boron, used in the [23]. Boron has a high cross section for the neutron capture but it migrates and is accumulated in the grain edges due to the Boron high diffusion coefficient and thermal sintering processes. By neutron irradiation high quantities of Lithium in the grain edges are produced and a worsening of the inter-granular superconductive properties is expected.

The comparison (Fig. 3), between 1IR and 1NIR samples shows other flux dynamics changes. In 1IR sample, the inter-granular power law at low magnetic field gives an increase of the parameter exponent ν, $|\nu|(1IR) = 0.45$; in respect to 1NIR sample, where $|\nu|(1NIR) = 0.2$. The weaker $|\chi_3|$ slope of the 1IR sample in applied magnetic field, is a normal behavior due to the pinning increase. While for magnetic field range $H > H_{cr}$, the exponential behavior of the 1IR gives a parameter value $B_0(1IR) = 0.28$ T less than $B_0(1NIR) = 0.78$ T. The exponentially decreasing slope after the irradiation is steeper and weakening of the superconducting properties occurs. However, this non-clear behavior can be illusory, since the B0 value calculated in the 1NIR sample is overestimated by the presence of the anomalous peak.

Moreover, in Fig. 3 the 1NIR $\chi|3|$ behavior shows a coincidence between the PE magnetic field value of $H = 0.1$ T (normally explained as "a transition occurs from weakly pinned 3D line vortices to decoupled 2D pancake vortices in the grain"[24]) and the magnetic field crossover H_{cr} where the weak JJ are broken between the grains. Seemingly, two disconnected intragranular and inter-granular processes take place at $H = 0.1$ T. Possible explanation of the PE in the inter-granular dynamic phenomena context is considering the competition between two, weak and strong, 'JJ grain boundary' flux dynamics [25]. This is a prototype of the collective transport in disordered systems with two different pinning regimes [25]. At the crossover between dynamics, (tuned by applied magnetic field) a PE can be expected due to a better interaction among the particular 'JJ strong' grain boundary configuration and the flux lattice.

Above-mentioned 'casual' flux dynamics condition vanishes after the neutron irradiation; in fact, a new pinning arrangement is now present in the sample (Fig. 3).

IV. Conclusion

The introduction of cascade defects, point defects and new elements by neutron irradiation in quasi-bi-dimensional HTSC, like our $Bi_{1.8}Pb_{0.4}Sr_{1.8}Ba_{0.2}Ca_{.2}Cu_3O_x$ sample, modifies the pinning nature and increases critical current in the analysed magnetic field range. Slightly different material that can be described as $Bi_{1.8}Pb_{0.4}Sr_{1.8}Ba_{0.2}Ca_{.2}Cu_{3-\delta}Co_{\alpha}Ni_{\beta}O_x$ is produced. An increase of the pinning response via rising of the pinning strength occurs. The neutron irradiation clearly diminishes the effect of the magnetic field in low amplitude, where all Josephson Junction (JJ) dominate the flux dynamics between superconducting grains and 'casual' anomalous peak (PE) disappears. Moreover, the H_{cr}, value remains unvaried, where a partial limitation of the electric transport, due to the rupture of 'weak JJ', begins. Neutron irradiation does not vary grain boundary tilts. For $H > H_{cr}$, in the H-T range studied, the current induced in the sample is lower than 'inter-granular strong JJ' critical current. Electric transport is described by exponential magnetic field dependence of the intra-grain J_c.

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