

# Anomalous hysteresis studies in Bi-2223/Bi-2212 superconductors by non-resonant microwave absorption (NRMA) with special reference to energy stabilized Josephson (ESJ) fluxons

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**Abstract :** Magnetic hysteresis of granular high temperature superconducting Bi-2223/Bi-2212 pellets has been studied by non-resonant microwave absorption (NRMA) method. It has been found that the nature of hysteresis is anomalous at all temperatures below  $T_c$ . This nature has been analyzed by considering the role of energy stabilized Josephson (ESJ) fluxons, which was not attempted in its totality previously. A comparative study has also been done which is based on data acquired from literature of both NRMA and DC magnetization measurements on granular superconductors. This comparison distinctly shows the difference, i.e. normal hysteresis in DC magnetization studies, whereas, anomalous hysteresis in NRMA studies under similar conditions of low temperatures and low magnetic fields. The discussion brings out the suggestion that, the anomalous hysteresis observed by NRMA method is a manifestation of inevitable generation and detection of ESJ fluxons.

Keywords: : Superconductors Sintering Electron Paramagnetic Resonance Hysteresis

## 1. Introduction

In intrinsically granular unconventional high- $T_c$  superconducting materials like cuprates, the magnetization hysteresis due to trapping of magnetic fluxons at various defects has been studied previously by using (1) direct magnetization methods like: DC magnetic field ( $H_{DC}$ ) variation of (i) flux density ( $B$ ), (ii) microwave surface resistance ( $R_s$ ), (iii) transport critical current density ( $J_c$ ) [1–7] and (2) modulated magnetic field variation of non-resonant microwave absorption ( $P$ ) [8–19].

However with regard to nature of hysteresis, the direct magnetization studies ( $B$  vs.  $H$ ,  $R_s$  vs.  $H$  and  $J_c$  vs.  $H$ ) [1–7] in such superconductors have shown normal (i.e. increasing magnetic field curve lying below decreasing magnetic field curve) as well as anomalous (i.e. increasing magnetic field curve lying above decreasing magnetic field curve) hysteresis in different conditions. Normal hysteresis generally observed at low temperatures (LT) and/or low magnetic fields (LH), whereas, anomalous hysteresis at high temperatures (HT) and/or high fields (HH).

It is striking that, in contrast to the above direct magnetization methods, the magnetic field modulated hysteresis in high- $T_c$  cuprates by non-resonant microwave absorption (NRMA) method

[8–15,18,19] has generally shown anomalous nature even at LT/LH. This behavior has also been observed in other variety of superconducting systems like:  $YPd_5B_2C_{0.35}/YNi_2B_2C$  [20], and  $MgB_2$  [21a,22], granules of  $Nb_3Sn$ ,  $NbGeAl_2$ ,  $V_3Si$ ,  $As$ ,  $Al$ , etc. [23–26].

It is surprising that despite the voluminous literature on the NRMA hysteresis [8–26], only a few groups [11,12,18] have mentioned about its anomalous nature and related it to the most successful model by Ji et al. [6] and its modifications by Mahel et al. [11] and Ramachandran et al. [18]. However, these modifications have their own limitations too. For example, the former one is applicable only for a limited modulation amplitude range. Whereas the latter one suffers from its stringent requirement of ‘threshold’ field value. Therefore, a closer look of Ji et al. model for the anomalous hysteretic nature observed in granular superconductors is necessary.

It is known that Ji et al. [6] modified conventional Bean model [27] to explain HT and/or HH anomalous hysteresis in granular superconductors by including the role of intergranular Josephson (hereafter called as IGBJ) fluxons in addition to intragranular Abrikosov/Pancake fluxons (hereafter called as IG). According to this model, the higher number density and higher mobility of IGBJ fluxons [10] at HT and/or HH in comparison to that at LT and/or LH leads to anomalous hysteresis. However, the general observation of anomalous hysteresis at LT and/or LH by NAMA method cannot be explained even by considering the contribution of IGBJ fluxons.

The aim of this paper is to consider the existing status of experimental observations and analysis to explain the general anomalous hysteretic nature observed in present Bi-2212/Bi-2223 samples

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studied by NRMA method by including the role of a different kind of mobile fluxons which are generated due to tilt motion of IGBJ/IG fluxons [14,28,29]. Since such fluxons are formed in order to stabilize energy of the flux lattice, therefore, named as energy stabilized (ESJ) fluxons hereafter. In the present study BSCCO system was chosen, as it has found that the formation of ESJ fluxons is more likely due to strong anisotropy and layered structure [28,29]. A comparative study based on NRMA and DC magnetization hysteresis data acquired from literature has also been made.

## 2. Experiment

In this work, we have carried out some NRMA studies of pure  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  (Bi-2212) and  $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+x}$  (Bi-2223) sintered pellets. Details of preparation and characterization of these samples have been reported elsewhere [16,17]. These sintered Bi-2212 and Bi-2223 pellets are nearly single phasic as suggested by X-ray diffraction and have  $T_c \sim 96$  K [16] and 117.2 K [17] respectively.

For NRMA studies, Bruker ER 200D X-band (9.47 GHz) EPR (Electron Paramagnetic Resonance) spectrometer equipped with an Oxford ESR 900 continuous helium gas flow temperature variation accessory was used.

The measurements were done at different values of temperature (5–130 K) by cooling the sample in zero field and then sweeping the static magnetic field  $H$ . Fixed moderate modulation amplitude of typically 4 G was used ensuring that the modulation amplitude dependent hysteresis and phase reversal do not complicate the results [8,10,11,13]. Modulation frequency was 100 kHz and microwave power was 20 mW. It was checked and confirmed that the signal shapes are not distorted.

The signals were recorded in the low as well as high field ranges for both forward and reverse scans of the field. For high field scans, the field was varied between  $-50$  (+50) to  $+1000$  ( $-1000$ ) G. For low field scans, the field was varied between  $-50$  (+50) to  $+50$  ( $-50$ ) G. EPR signal of standard reference sample of diphenyl picryl hydrazil (DPPH) under identical conditions was also taken in order to see the phase of the NRMA signal. Other details of NRMA measurements are given in our earlier papers [16,17].

Signals were recorded in the derivative form [i.e. field derivative ( $H$ ) of microwave power absorption ( $P$ ):  $dP/dH$ ] resulting from magnetic field modulation and phase-sensitive detection technique when  $H_{mw}$  is perpendicular to  $H$ . This provides information on extremely small temperature and magnetic field dependent microwave absorption ( $P$ ) changes. These changes are related with thermal forces/microwave modulation magnetic field induced motion of inter/intragranular fluxons in granular superconductors when subjected to external magnetic fields.

## 3. Results and discussion

Here, first we will discuss the (i) present NRMA results obtained in Bi-2223/Bi-2212 sintered pellets followed by a discussion based upon a comparative study of the existing data of: (ii) NRMA magnetic hysteresis in different granular superconducting systems and (iii) a comparison of DC/NRMA magnetic hysteresis in identical superconducting systems.

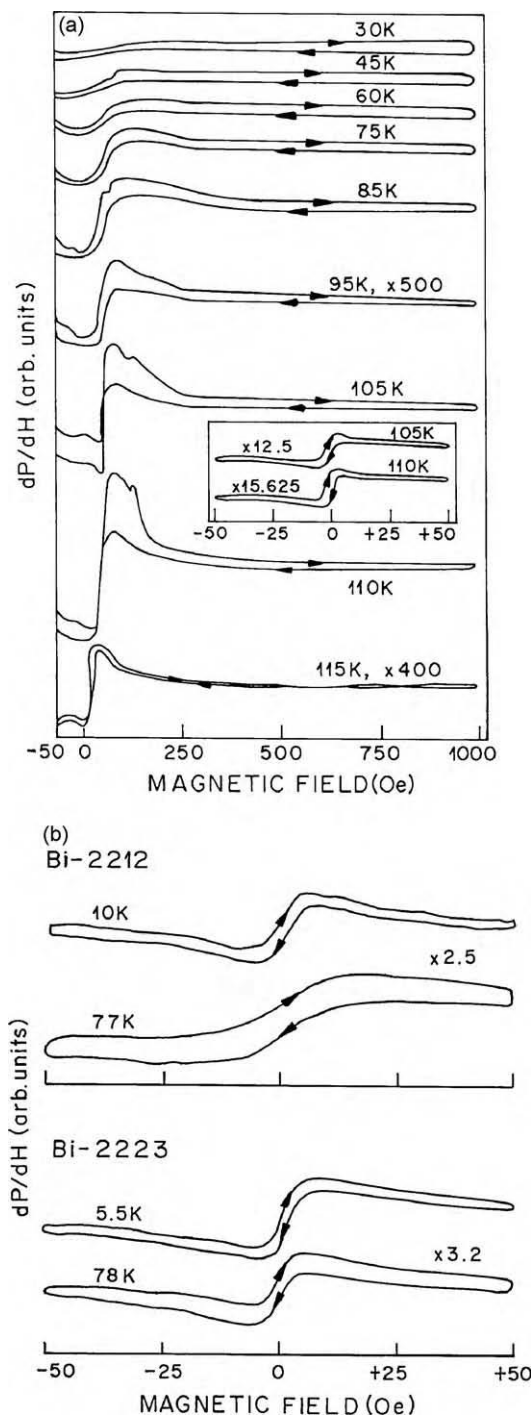
### 3.1. Present results

Fig. 1a shows  $dP/dH$  vs.  $H$  curves of NRMA signals recorded for Bi2223 sintered pellets at various temperatures in the high field scans for both forward ( $-50$  to  $+1000$  G) and reverse ( $+1000$  to  $-50$  G) directions. The arrow shows DC magnetic field sweep direction.

It can be seen from these curves that for most of the temper- Fig. 1. (a)  $dP/dH$  vs.  $H$  hysteresis curves for Bi-2223 sintered samples showing atures studied from 30 K to 115 K, the signal intensity is nearly anomalous nature in the entire field range from  $-50$  (+50) G to  $+1000$  ( $-1000$ ) G (through 0 G) at all temperatures below 115 K, except at 105 K and 110 K for near

independent of magnetic field particularly for large field values. Such quasi linear dependence of dissipation on field has been reported and associated with the motion of IG fluxons

around zero field and a hump like feature  $-50$  G through 0) Bi-2212 and Bi-2223 sintered samples showing anomalous nature around 120 G also appeared at 30 K, which vary in amplitude and not only at HT (77 K, 78 K) but also at LT (5.5 K, 10 K), width with increasing temperature. Similar signals around zero field [8–26,30,31] and broad hump like feature around 20–120 G [21b,30,31] have been reported earlier in high temperature super-IGBJ fluxons and ESJ fluxons. On increasing temperature up to 95 K conductors also. The former signal at zero field has been associated both the signals were found to grow in intensity indicating that the with motion of IGBJ fluxons [8–26]. Whereas the latter broad hump number of IGBJ fluxons and ESJ fluxons increases as the temperature signal been associated with motion of ESJ fluxons [21b,30,31]. Due increases. to the similarity in the present study, the observed zero field signals Along with these observations, forward and reverse field sweeps and the broad small signal have also been respectively assigned to of these signals show hysteresis, which becomes pronounced as the



zero field scans, where it appears no hysteresis. Shown in inset at 105 K and 110 K hysteresis curves for low field scans (from  $-50$  G to  $+50$  G through 0 and back to  $-50$  G) [10,21b]. Other signals

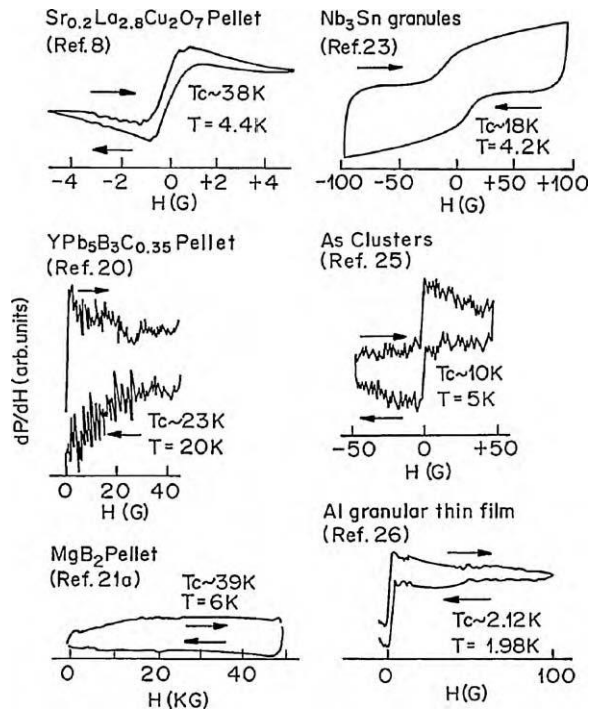


Fig. 2. NRMA hysteresis curves in different granular superconducting systems both conventional as well as unconventional systems showing anomalous nature in the LT/LH regimes.

temperature increases up to 95 K. This behavior can be attributed to the role of IGBJ and ESJ fluxons in hysteresis.

At 105 K and 110 K, in low fields around 0 G in the high field scans hysteresis was not found so clearly. To confirm the low field behavior at these temperatures, low field signals were recorded with field varying from  $-50$  to  $+50$  and from  $+50$  to  $-50$  G. A clear presence of hysteresis is obvious (shown in inset). Beyond 110 K the broad signal diminished and at 115 K, only a zero field single line remained and the hysteresis disappeared, indicating a flux flow regime.

Similar results were obtained in Bi-2212 samples. The low field scans for both the Bi-2223 and Bi-2212 at low (5.5 K, 10 K) and high (77 K, 78 K) temperatures are shown in Fig. 1b.

Interestingly, for entire temperature range of studies, i.e. 5.5 K to 110 K and for different field [high field (Fig. 1a) as well as low field (Fig. 1b)] scans, the decreasing H curve lies below the increasing H curve for both the samples (Bi-2212 and Bi-2223). This indicated that the nature of NRMA magnetic hysteresis is anomalous even at temperatures as low as 5.5 K. Probably this anomaly in magnetic hysteresis is due to the presence of ESJ fluxons in addition to IGBJ fluxons.

It would be appropriate to give a brief account of earlier reports related particularly with nature of NRMA hysteresis in different granular superconducting systems and its comparison with that of DC magnetization method, before any conclusive discussion on the role of ESJ fluxons in anomalous hysteresis observed in present samples by NRMA results.

### 3.2. NRMA magnetic hysteresis in different granular superconducting systems

Fig. 2 exhibits the results of NRMA hysteresis reported earlier in (a) unconventional granular superconductors like La-Sr-Ca-Cu-O [24], YPb<sub>5</sub>B<sub>3</sub>Co<sub>0.35</sub> [20] and MgB<sub>2</sub> [22] and (b) conventional granular superconductors like Nb<sub>3</sub>Sn granules [23], As-clusters in GaAs layers [25], Al granular thin films [26]. It is worth noting that in

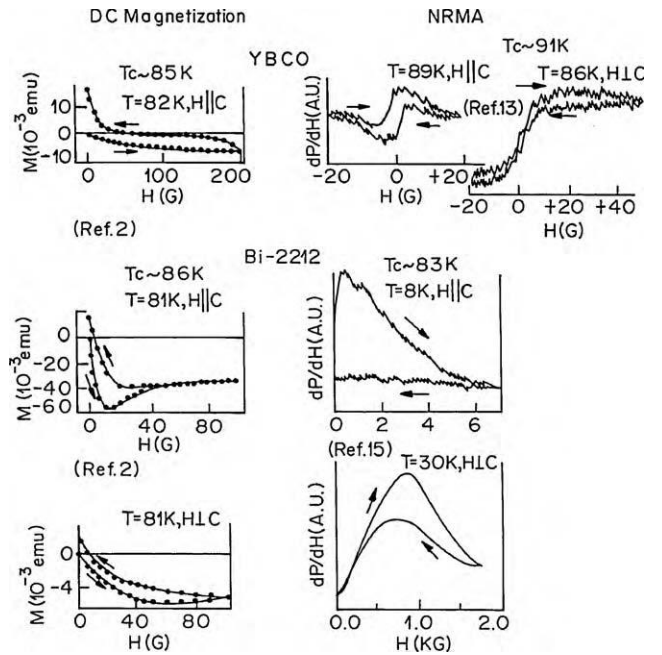


Fig. 3. Comparison of NRMA and DC magnetic hysteresis in YBCO/Bi-2212 single crystals showing the anomalous hysteresis as seen in NRMA in contrast to normal in the DC magnetization under identical H  $\parallel$  c-axis & H  $\perp$  c-axis orientation conditions.

all the NRMA curves, the decreasing H curve lies below the increasing H curve (anomalous hysteresis behavior) at temperatures much below  $T_c$  and in low field regions as observed in the present Bi2212/Bi-2223 samples (Fig. 1). This confirms the anomalous nature of NRMA hysteresis at LT/LH in all these different granular unconventional/conventional superconducting systems also. This makes the scenario very interesting and intriguing. In order to investigate further, in the following, a comparison with the nature of hysteresis as seen for example by DC magnetization method is also made.

### 3.3. A comparative study of DC and NRMA magnetic hysteresis

In order to have a comparison, DC and NRMA magnetic hysteresis data available in single crystals of YBCO system [2,13] and Bi-2212 system [2,15] for both H  $\parallel$  c-axis and H  $\perp$  c-axis orientations are shown in Fig. 3. It further confirms that in contrast to normal hysteresis observed in DC magnetization the nature of hysteresis shown by NRMA method is anomalous under similar conditions of temperature and magnetic field.

Thus all the above data showing anomalous hysteresis in different granular superconducting systems under LT/LH conditions as seen in NRMA method. This data support the present results. This suggests the involvement of some fluxons other than IGBJ fluxons. It is these fluxons, which not only increase the total number of more mobile fluxons at LT and/or LH, etc. but also are detected due to the extra sensitivity of the NRMA method.

As mentioned before that although there exist earlier reports on the presence of thermal/Lorentz forces induced fluxons (named as ESJ fluxons) in Bi-2212 single crystal by rf absorption measurements [30]/NRMA measurements [21b,31], however, their role in hysteresis has not been included so far. The presence of these fluxons and the anomalous hysteresis in the present samples, the role of ESJ fluxons seems a possibility. However, to come to an unambiguous conclusion, it is essential to highlight the experimental features of NRMA method, which makes it unique. For example, its ability: (i) in creating a situation for enhanced ESJ fluxons, (ii) in detecting even minute changes in the fluxon density and (iii) in

distinguishing between different types of fluxons, where all other DC magnetization techniques unable to do so.

In this context it is known that NRMA technique uses EPR (Electron Paramagnetic Resonance) spectrometer, where DC magnetic field ( $H_{DC}$ ) is modulated with a modulation field ( $H_m$ ) superimposed parallel on  $H_{DC}$ . Due to this  $H_m$  of magnetically modulated hysteresis are observed unlike direct field hysteresis measurements. This modulation field in addition to  $H_{DC}$  (used in B vs. H and  $J_c$  vs. H methods) and  $H_{DC} + H_{mw}$  (both used in  $R_s$  vs. H method) gives a few advantages. For example, it provides extra energy (due to a combined effect of three fields:  $H_{DC} + H_{mw} + H_m$ ) required to overcome the strength of the weak links; as a result, IGBJ/IGJ fluxons enter at much lower temperature/lower magnetic fields. Another is a stronger Lorentz force, particularly due to  $H_m$  induced oscillatory currents, which tilt the IGBJ/IGJ fluxons, and probably becomes a volume source of generation of ESJ fluxons.

Further, a combination of the modulation field in conjunction with phase-sensitive detection makes EPR an extremely sensitive and powerful tool to detect any feeble change and to distinguish between different types of fluxons [10,21b,31]. Furthermore, the sensitivity of the NRMA method, which uses EPR, is even five orders of magnitude greater than the sensitivity limit of the EPR spectrometer [24].

All these parameters make EPR technique an extra sensitive and unique in comparison of other techniques. Therefore, any change, however minute and at times seemingly intractable, related with density/distribution/motion and different types of fluxons depending upon temperature/magnetic field/sample form, etc. can be easily detected in NRMA. Furthermore, NRMA is the only method [10,21], which is able to distinguish between above-mentioned different types of fluxons, i.e. IGBJ, IG and ESJ, etc.

#### 4. Summary and conclusion

The present NRMA measurements made on Bi-2212 and Bi-2223 samples showing anomalous magnetic hysteresis at all temperatures below  $T_c$  have been analyzed and explained by including the role of ESJ fluxons. An analytical comparative study based on NRMA data reported by others in a variety of granular superconducting systems and its comparison with DC magnetic hysteresis data has revealed that anomalies in hysteresis probably, are the manifestation of combined effect of additional fields like: oscillatory MW field and modulation field, essentially in the formation of ESJ fluxons.

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