# Effect of columnar defects on the irreversibility line in pristine and iodine-intercalated Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub> single crystals

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#### Abstract

We have investigated the influence of columnar defects (CD) on the vortex dynamics in pristine and iodine-intercalated  $Bi_2Sr_2CaCu_2O_{8+\delta}$  single crystals from dc SQUID magnetization measurements. Especially, the temperature dependence of the irreversibility fields,  $H_{irr}(T)$ , were studied. Anisotropy ratio  $\gamma$ , estimated from the fitting to the 2-dimensional melting model (A. Schilling *et al.*, Phys. Rev. Lett. 71 1899 (1993)) in higher fields than the matching field  $B_{\phi}$  at low temperature region, turns out to be decreased by the iodine-intercalation and additionally by the heavy-ion irradiation.

Keywords: Iodine-intercalation, Columnar defect, Irreversibility line, Anisotropy

## I. Introduction

A mount of information on the vortex dynamics in high- $T_c$  superconductors have been obtained from the magnetic phase diagram based on the irreversibility line  $H_{irr}(T)$ , which has been usually determined from the dc magnetization measurements at the temperature where the zero field cooling (ZFC) and field cooling (FC) magnetization merges and/or from the onset of a measurable resistivity [1], and/or from the onset of the third harmonic transmittivity in ac susceptibility measurements [2, 3]. But the meaning irreversibility line in the heavy ion irradiated system is still controversial in the sense that it was assisted to be the melting line from vortex solid to liquid [4,6] or to be the bose-glass to vortex liquid [7]. It may be due to the fact that the jump in reversible magnetization at a characteristic field [4],  $B_{FOT}$ , which was interpreted as an symptom of the first order melting of vortex solid, was not observed in irradiated samples unlike in pristine samples. Recently, van der Beek et al. [3]

have assisted that it is due to the enhancement of the irreversibility field over to the characteristic field  $B_{FOT}$  of the first order transition, i.e.,  $B_{FOT}(T) << H_{irr}(T)$ .

Kuroda *et al.*, [8] have reported that the Bose-glass behavior is observed at fields below the matching field B<sub>↓</sub> in irradiated Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub> (Bi2212), but not in irradiated La-substituted Bi<sub>2</sub>Sr<sub>2</sub>CuO<sub>6+δ</sub> (Bi2201). They have assisted that it can be due to the extremely weak coupling between pancakes vortices along the c-axis in Bi2201 even under the presence of columnar defects.

Halogen (for example, I<sub>2</sub>) intercalated [9] and following heavy ion irradiated Bi2212 single crystals can be used to test the idea of Kuroda's. Since it was well-known that iodine can be intercalated between the double *Bi-O* bilayer of Bi2212 and therefore tune the interlayer coupling strength. [9, 10] Structural studies using X-ray diffraction and TEM [9,10] of Bi2212 single crystals reveal that the iodine intercalation expands the c-axis unit cell dimension,

while it has little effect on the in-plane parameters. In terms of transport [10], the effect of iodine intercalation is to depress the superconducting transition temperature  $T_c$  by about 10 K. Moreover, the temperature dependence of the resistivity along the c axis,  $\rho_c$  changes from a semiconducting behavior to a metallic one after iodine-intercalation to Bi2212 single crystals [10]. From the extensive studies of Xray photoemission [11, 12], Hall effect and pressure dependence [13], systematic doping control [14, 15], and IR [16], the metallization of c-axis resistivity have been thought to be due to the increase of hole carrier concentration on CuO, plane. But it was also tried to explain the metallization by the enhanced c-axis coupling [9, 17] against the increased c-axis constant by 3.5 Å. On the other hand, it assisted by Winkel [18] that an extrinsic, defect mediated mechanism along c-axis can cause a strong decrease of  $\rho_c$ , which was argued from the fact that the position of magnetic irreversibility line in the H-T plane remains unchanged upon iodine intercalation.

Even though it is not fully adopted on what causes the decrease in  $T_c$  through the intercalation, it is thought that the decrease in Tc is the plus effect of the increasing hole concentration in  $CuO_2$  plane and the reduced coupling between the superconducting  $CuO_2$  layers. Nowadays the former is thought to be predominant over the latter.

It is the purpose of this paper to investigate the influence of the iodine-intercalation on the interlayer coupling (anisotropy) and the impact on the phase diagram of heavy-ion irradiated IBi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub> (Irr-IBi2212). From the analysis of irreversibility line  $H_{irr}(T)$  presented below (Sec. III), it turns out that the anisotropy ratio  $\gamma$  is decreased by the iodineintercalation, which is assumed to give rise to the metallization in  $\rho_r$ . The anisotropy was also decreased by introducing columnar defects, which can be the cause that make a similarity in  $H_{irr}(T)$  between the moderately anisotropic high-Tc superconductor (for examples,  $YBa_2Cu_3O_{7-\delta}$ ,  $\delta=0.4$ , 0.62 (Y-123), La<sub>1.86</sub>Sr<sub>0.14</sub>CuO<sub>4</sub> (La-214), Y<sub>1-x</sub>Pr<sub>x</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>6.97</sub> (Y(Pr)-123) of Ref. [19,20]) and highly anisotropic but heavy-ion irradiated Bi2212 single crystals (Irr-Bi2212).

#### II. Experiments

Bi2212 crystal boules have been grown in an infrared radiation furnace by the traveling solvent

floating zone method [21]. In order to control various oxygen content  $\delta$ , single crystals were annealed in a quartz tube furnace under the various oxygen partial pressures at 700 °C for 10 h, then slowly cooled to 300-400 °C (~0.4 °C/min) while keeping the equilibrium oxygen pressures, which was carried out by Watanabe et al., [22] to reach more homogeneous phase against the oxygen diffusion during the annealing. Finally, they were rapidly cooled to room temperatures at a rate of 50 °C/min. For the actual control of the oxygen partial pressures from 3.8x10<sup>-4</sup> Torr to 30 mTorr, the sample tube was evacuated using a rotary pump with a flow of the oxygen. To obtain a highly underdoped samples, the quartz tube was evacuated down to 3.8x10<sup>-7</sup> Torr without the oxygen flow using a turbomolecular pump at 700 °C for 10 h. Magnetization studies were carried out using a dc SQUID magnetometer (Quantum Design, MPMSXL. The onset superconducting transition temperature,  $T_c$ , measured at low field (10 Oe) dc magnetization, was changed from 85 K (as-grown) to 89 K (30 mTorr) to 85 K (3.8x10<sup>-4</sup> Torr) to 60 K (without the oxygen flow). The highly underdoped samples were obtained as many smaller pieces than original one, the annealing condition (700 °C, 3.8x10 -7 Torr) turns out to be near the decomposition temperature and pressure of Bi2212 crystals.

Iodine intercalation was performed by heating each sample with an amount of iodine (mass corresponding to about 2 atm at 190 °C) in a vacuum-sealed pyrex tubes at 180-200 °C for 120 h.

The columnar defects were formed along the c-axis of Bi2212 single crystals by 1.3 GeV uranium-ion irradiation at Argonne national laboratory in USA. The total planar density of the defects was  $5 \times 10^{10}$  cm<sup>-2</sup>, which corresponds to the matching field  $B_{\phi} \sim 1$  T.

The mass of the first sample (G2I-2) was changed from  $1.2\pm0.1$  mg (for the pristine Bi2212), to  $1.4\pm0.1$  mg (after iodine-intercalation), finally to  $1.6\pm0.1$  mg (after heavy-ion irradiation). The first increase in mass is due to the iodine-intercalation, but the second increase after irradiation may be due to the uncertainty in accuracy or due to remained photoresist, which is used to attach the sample to copper block for heavy-ion irradiation. The mass of the second sample(G5-2) is  $0.7\pm0.1$  mg after heavy-ion irradiation.

For the complete penetration, the thickness of samples less than 30 m were chosen. Taking the unit cell of the stage-1 intercalated material as IBi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+8</sub> (IBi2212) the theoretical density is

6.13 g/cm³, compared to pristine Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub>, with a theoretical density of 6.53 g/cm³. [23]

To obtain information on the oxygen contents  $\delta$ , we used the empirical relation between  $T_c$  and the hole concentration p in  $CuO_2$  plane  $T_c/T^{max}_c = 1-82.6(p-0.16)^2$  which is satisfied for a number of High-Tc superconductors [24, 35] and we use  $T^{max}_c = 90$  K. The value of  $\delta$  was estimated from the relation: [22]  $|\delta - 0.24| \propto |p - 0.16|$ . From above analysis, the samples were regarded to be changed from overdoped (85 K, as-grown) to optimal doped (89 K) to underdoped (85K) to highly underdoped (60 K) phases.

#### III. Results and Discussion

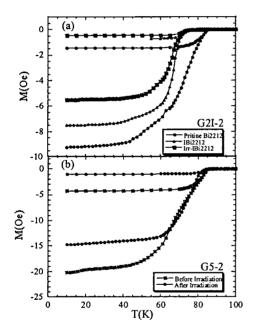


Fig. 1. Temperature dependence of dc magnetization of  $Bi_2Sr_2CaCu_2O_{8+\delta}$ .

Fig. 1 shows the changes in the temperature dependence of dc magnetization at low field (10 Oe) of pristine Bi2212 single crystals through iodine-intercalation (IBi2212) and heavy-ion irradiation (Irr-IBi2212). The onset  $T_c$  is decreased from 89 K to 78 K after iodine-intercalation, as reported by other groups. [9, 10, 23] The homogeneity of the iodine intercalated sample is enhanced as compared to that of pristine

Bi2212 crystals. The superconducting volume fraction estimated from the susceptibility turns out to be suppressed by the iodine-intercalation and additionally by the heavy-ion irradiation.

From the X-ray diffraction analysis shown in Fig. 2, the c-axis lattice parameter of iodine intercalated crystal was estimated to be 18.9 Å, which is indicative of stage-1 intercalation. [[9, 10, 26]

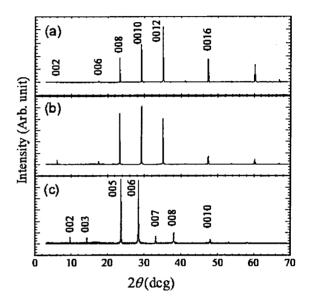


Fig. 2. X-ray diffraction of (a) Pristine Bi2212 (G5-2), (b) after heavy-ion irradiation (G5-2), and (c) iodine-intercalted and heavy-ion irradiated Bi2212 (G2I-2).

Fig. 3 shows the various characteristics in irreversibility lines of Bi2212 single crystals measured in magnetic fields H parallel to the c-axis of the crystal, which depend on their type of disorder and the amount of hole in  $CuO_2$  plane.

First we will discuss the behavior at  $T/T_c > 0.6$ . Unirradiated samples, shown with square symbols above about 45 K, show the power law behavior [5] in temperature dependence of irreversibility fields  $H_{irr}(T) = B_0 (1 - T/T_c)^n$  with  $B_0 \cong {}_0{}^5 c_L{}^4/16\pi^3$   ${}_{ab}{}^4(0)\gamma^2(k_BT_c)^2$  and  $n \sim 2$ , which is well-known as a thermal-fluctuation-induced 3 dimensional flux-line solid to liquid melting near  $T_c$ . [27] Below 45 K,  $H_{irr}(T)$  have changed their curvature, which is a typical symptom in the dimensional crossover from 3D to 2D vortex line. From the best

fitting, the characteristic crossover field [28]  $B_{cr} \cong {}_{0}/(s\gamma)^{2}$  is an order of 1 kG, where s is the interdistance between  $CuO_{2}$  layers.

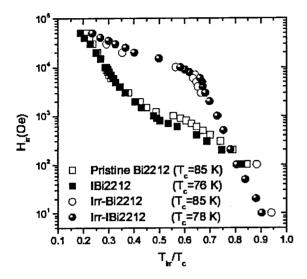


Fig. 3. The temperature dependence of the irreversibility line of pristine Bi2212 (empty square, from Ref. 5), IBi2212 (solid square, from Ref. 5), Irr-Bi2212 (empty circle, G5-2), and Irr-IBi2212 (sphere, G2I-2). 0.1 G criterion for irreversibility test was used.

After heavy-ion intercalation (samples with circle symbols), the irreversibility temperature is increased, which is well-known due to the pinning of the twodimensional pancake vortices in the superconducting layers by the columnar defects. [29, 30] Except the rapidly increasing region of 3500 < H < 5500 Oe (approximately corresponding to the region  $1/3 B_{\phi} < H$ < 1/2  $B_{\phi}$  Hirr (T) appears to follow the form of  $H_{irr}(T) \propto \exp(-T/T_0)$ . This behavior was also observed in heavy-ion irradiated Bi2212 single crystals [3, 31] for  $B_{\phi} > 30$  mT and in moderately anisotropic high-Tcsuperconductors. [19, 20, 27] The difference is that at unirradiated anisotropic and moderately superconductors (Y-123, La-214, Y(Pr)-123 ),  $H_{irr}(T) \propto \exp(-T/T_0)$ . at a temperature region below  $T/T_c < 0.8$ -0.9 appears to be followed by power law behavior. The presence of this irreversibility line has been interpreted by the existence of a breakdown field in which a proximity effect induced superconductivity of charge reservoir layer (for example, SrO-BiO-BiO-SrO in Bi2212 system) is destroyed. [32] But for my best knowledge, there is no further evidences for the proximity effect in the charge reservoir block and/or Andreev reflection behavior between CuO<sub>2</sub> layers. It should be further investigated about it.

Recently, van der Beek [33] tried to explain it by assuming that the irreversibility line can coincide with the bose-glass transition line and that the bose-glass transition in a layered supercondctor can be identified by the unbinding of the bound pancake vacancy-interstitial pairs within the same layer, *i.e.*, the "quartets" suggested by Feigel'man. [34] According to van der Beek [33] the bose-glass transition temperature  $T_{BG}$  was estimated from the equality between the thermal energy and the energy of the quartets, *i.e.* 

$$k_B T_{BG} = (R_I) \quad _0 s(R_I/)^2$$
 (1)

where  $R_I$  is the distance between a bound vacancy and interstitial,  $_0 = _0{}^2/4\pi\mu_0$   $_{ab}{}^2$  the typical vortex energy per unit length, and  $=(_a{}_b{}^{-1}+(\gamma s)^I)^{-1}$  the generalized penetration depth taking account both magnetic and Josephson coupling. From Eq. (1), the bose-glass transition line was given as follows

$$B_{BG} = B_{\Lambda} \left( \frac{\varepsilon_0 s}{k_B T} \right) \exp \left( \frac{\varepsilon_0 s}{k_B T} \right), (B_{\Lambda} \ll B \ll B_{\phi})$$
 (2)

with  $B = _{0}/^{2}$ ,  $_{0}$  and to be evaluated at  $T_{BG}$ . We have carried out a least square fit of our data to Eq. (2) shown in Fig. 3. From the best fit, the values of  $_{0}$  (0)  $s/k_{B}$  of Irr-Bi2212 and Irr-IBi2212 were estimated to be 701 K and 1198 K, respectively. The values of  $_{ab}$  (0) were estimated to be 211 nm and 158 nm, respectively, which are typical values reported by other group [3, 23]. But the value of B a re an order of  $10^{-3}$  and  $10^{-7}$  Oe, respectively, which are unphysically small.

Using their results of best fitting, van der Beek [33] reported that the exponential decrease in  $H_{irr}$  at high T is steeper for the overdoped crystal, therefore which has the smaller  $_{ab}(0)$  and  $\gamma$  than those of the optimal doped Bi2212 single crystal., According to the study on the relation between the penetration depth and the doping level, [35], this argument is valid only in a restricted region. This point can be tested in our experiments with advantage of iodine-intercalation to Bi2212 single crystals, which was known to be able to increase the hole doping level of copper-oxide

superconducting layer as well as the to increase the *c*-axis lattice parameter. Through following analysis, we found that the penetration depth in overdoped samples (intercalated ones) can be larger than that in optimally doped samples.

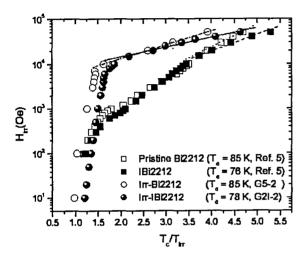


Fig. 4. The temperature dependence of the irreversibility line of pristine Bi2212 (empty square, from Ref. 5), IBi2212 (solid square, from Ref. 5), Irr-Bi2212 (empty circle, G5-2), and Irr-IBi2212 (sphere, G2I-2).

We have fitted the data in high field at low temperature region to the thermal fluctuation-induced 2D melting model [27] as shown in Fig. 4.

$$B_m \approx \frac{\phi_0}{(s\gamma)^2} \exp\left(\frac{\phi_0^2 c_L^2 s}{8\pi \lambda_{ab}^2(0) k_B T}\right) \quad (3)$$

From the best fit, the values of  $_{o}'$  ( $_{s}\gamma$ )<sup>2</sup> were estimated to be 108, 846, 3300, and 5600 G for pristine Bi2212 ( $_{s}$  = 15.0 Å), IBi2212 ( $_{s}$  = 18.9 Å), Irr-Bi2212, and Irr-IBi2212, respectively. As a result, the anisotropy ratios  $_{g}$  have values of 292, 83, 53, and 32, respectively. The values of  $_{o}^{2}c_{L}^{2}s$  /8  $_{a}$   $_{b}^{2}$  (0) $_{b}$ 8 turn out to be 112, 59, 54, and 41 K, respectively, which is consistent to the intuition that the penetration depth will increase if we introduce the intercalation and further increase with columnar defects. This interpretation is based on the fact that  $_{c}^{2}$  was decreased with iodine-intercalation and further decreased by heavy ion-irradiation and that  $_{c}^{2}$  is

proportional to  $ab^{-2}$  at zero temperature limit. [35] Using general value of  $c_L$ , 0.15, the values of ab (0) were estimated to be 134, 207, 193, and 249 nm, respectively, which is typical values reported by others. [5,23] But it should be noticed that the value of  $c_L$  will be decreased by iodine-intercalation, but will be increased by the columnar defects in the range of 0.1  $c_L$  0.4[36] In our experiments, we could not determine the dependence of  $c_L$  on the iodine-intercalation and on the columnar defects.

Although there is an uncertainty in determining the value of  $H_{irr}(T)$  using a fixed criterion, but we assist that the value from extracted from the slope will reflect the nature.

Table 1 Thermodynamic parameters of pristine, IBi2212, Irr-Bi2212, and Irr-IBi2212. From the best fit to Eq. (3).  $_{ab}(0)$  was estimated using the value of  $c_L = 0.15$ .

Parameters	Pristine	IBi2212	Irr- Bi2212	Irr- IBi2212
$o/(s\gamma)^2$ (G)	108	846	3300	5600
$\frac{\phi_0^2 c_L^2 s}{8\pi\lambda_{ab}^2(0)k_B} $ (K)	112	59	. 54	41
γ	292	83	53	32
$_{ab}(0)$ (nm)	134	207	193	249

### IV. Conclusions

We have investigated the influence of columnar defects on the vortex dynamics in pristine and iodine-intercalated  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals from dc SQUID magnetization measurements. Especially, the temperature dependence of the irreversibility fields,  $H_{irr}(T)$ , were studied. Anisotropy ratio  $\gamma$  in higher fields than the matching field  $B_{\phi}$  at low temperature region, turns out to be decreased by the iodine-intercalation and additionally by the heavy-ion irradiation.

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