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IRRADIATED and OXYGEN DEFICIENT MATERIALS

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# **MAGNETIZATION and FLUX PINNING in HIGH- $T_c$ CUPRATES: IRRADIATED and OXYGEN DEFICIENT MATERIALS**

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## **ABSTRACT**

This work surveys recent studies of the intragrain current density  $J$  and vortex pinning in high  $T_c$  superconductors. Materials include  $Y_1Ba_2Cu_3O_{7-\delta}$  and  $Bi_2Sr_2Ca_1Cu_2O_8$  single crystals and aligned polycrystals. To probe the flux pinning, we modified the strength, number, and morphology of defects. Varying the oxygen content ( $7-\delta$ ) in  $Y_1Ba_2Cu_3O_{7-\delta}$  or irradiating the materials with ions, having either light or heavy masses, gives systematic changes in the character of the all-important defects.

## **INTRODUCTION and EXPERIMENTAL FEATURES**

After discovery of high temperature superconductors, it soon became apparent that these materials have both great promise and formidable difficulties. To conduct large, dissipation-free electric currents in high magnetic fields, the material must contain microscopic imperfections that change the superconducting properties over small regions of space. The purpose of these "defective" regions is to immobilize the vortices that penetrate the superconductor, since energy dissipation accompanies vortex motion. Given the potentially high operating temperatures of HTSC's, thermal energy  $k_B T$  easily activates vortex motion. We have studied several factors that influence the "pinning" of vortices: modification of the superconductive matrix in  $Y_1Ba_2Cu_3O_{7-\delta}$  by varying the oxygen content ( $7-\delta$ )  $\geq 6.8$ , where progressive removal of oxygen weakens most superconductive properties; irradiation with light ions (3 MeV protons) to create controlled densities of point-like defects; and irradiation with energetic heavy ions (580 MeV Sn ions) to form parallel columns of highly defected material, aligned along the ion beam

direction. Consequently, we can study vortex pinning sites with different strengths and morphologies, in order to understand the easy vortex movement and to establish the maximum realizable current density  $J$  in these materials.

We have investigated the "static" (dc) magnetization in several cuprate superconductors, using SQUID-based and vibrating sample magnetometers. Studies of the mixed state magnetization at temperature  $T$ , magnetic field  $H$  ( $H \parallel c$ -axis), and time  $t$  yield the in-plane current density  $J$  and the effective pinning energy  $U_{\text{eff}}$  for vortices. The circulating supercurrent density  $J$  was determined using the Bean model<sup>1</sup> with appropriate transverse crystal dimensions. Decay of the circulating current density was studied in the SQUID magnetometer, in flux creep measurements of  $M$  vs. time  $t$ . The quantity  $U_{\text{eff}}$  depends on the instantaneous current density  $J(t)$  and increases strongly as  $J$  decreases, due to collective pinning effects.

To assess the impact of chain-site oxygen vacancies on current density and pinning energy, a magnetically aligned composite of small  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  crystallites was formed.<sup>2</sup> The small grain size of  $\sim 7 \mu\text{m}$  radius allowed easy and reversible variation of oxygen content ( $7-\delta$ ). Oxygen deficiencies  $\delta$  in the range 0-0.2 were established *in situ*, using a thermogravimetric apparatus (TGA).

The irradiation studies used thin single crystals of  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$  and  $\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_8$ . Point-like defects (vacancies, interstitials, site-antisite defects, etc.) were formed by irradiating YBCO crystals with 3 MeV protons to fluences near  $10^{16}$  ions/cm<sup>2</sup>. Alternatively, highly energetic heavy ions (580 MeV  $\text{Sn}^{30+}$  ions) produced highly disordered, semicontinuous columns. These linear defects, with diameter of  $\sim 50 \text{ \AA}$ , form by inelastic electronic excitations, if the energy deposition rate is large enough. In our case, the rate was (2.6-2.0) keV/ $\text{\AA}$  to a depth of  $20 \mu\text{m}$  in the crystals, for both YBCO and BSCCO. The ion beam traveled at a small angle ( $\sim 2^\circ$ ) from the  $c$ -axis, to avoid channeling effects. Sn-ion fluences of  $(0.5 - 4) \times 10^{11}$  ions/cm<sup>2</sup> were used; multiplying the fluence by the flux quantum  $\phi_0 = 2.07 \times 10^{-11}$  Tesla-cm<sup>2</sup> reexpresses the fluence as an equivalent vortex density, i.e., a matching magnetic field  $B_\phi$  of (1-8) T.

### PERSISTENT CURRENT DENSITY and IRREVERSIBILITY LINE

Studies of the equilibrium magnetization of oxygen deficient  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  materials have shown that the superconductive properties degrade continuously with removal of oxygen.<sup>2</sup> For example, the superconductive condensation energy  $F_c = H_c^2/8\pi$  decreases sharply with

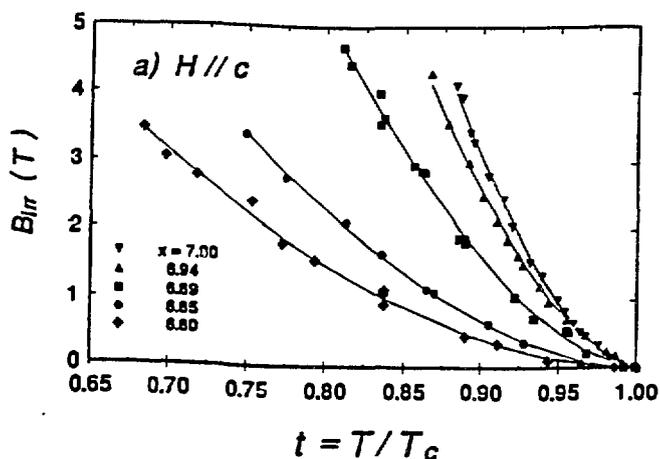


Fig. 1. Irreversibility field  $B^*$  for  $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  with oxygen deficiency  $\delta$  versus reduced temperature.

deficiency  $\delta$ . In Fig. 1 is a second example, the irreversibility line  $B_{irr}(T)$  obtained from an ac magnetic response study with  $H \parallel c$ -axes. This line in the  $H$ - $T$  plane separates a low temperature region with finite  $J_c > 0$  from a higher temperature region with  $J_c = 0$ . As oxygen is removed, the irreversible region with  $J_c > 0$  contracts rapidly. The solid lines in Fig. 1 are fitted to the function  $B_{irr}(T) = B_o^* [1 - (T/T_c)^n]$ , with  $n \approx 1.7$ . A third example is the magnitude of the intragrain critical current density. With decreasing oxygen content ( $7-\delta$ ),  $J_c$  decreased monotonically, for almost all conditions of field and temperature  $T$ . Note that in this study, the single sample contained a *constant* number of strong, preexisting defects such as site-antisite exchanges. All of these quantities are interrelated by a simple, single site model for vortex pinning.<sup>2</sup> The model provides that  $J_c \propto F_c \xi_{ab}$ , where  $\xi_{ab}$  is the coherence length within the Cu-O planes. This correlation is shown in Fig. 2, where  $B_o^*$  and  $J_c$  (at 4.2 K and 6.5 tesla) are plotted vs. the pinning parameter  $F_c \xi_{ab}$ . The linear relations show that these quantities are indeed controlled by fundamental superconductive properties, such as the condensation energy. Similar proportionalities arise in more sophisticated (and more realistic) theory for collective vortex pinning.

Ion irradiation provides a facile tool to create controlled numbers of defects with transverse size of  $\sim 1$ -10 nm. For HTSC materials, this dimension is comparable to the coherence length  $\xi \approx$  (radius of the vortex core). Consequently, these defects can effectively pin flux and increase  $J_c$ . An example is shown in Fig. 3, a plot of current density vs. temperature. The data are for a  $Bi_2Sr_2Ca_1Cu_2O_8$  crystal, both unirradiated and irradiated with Sn-ions to a fluence  $B_\phi = 5$  T, for several applied fields. Addition of columnar defects dramatically increases the current density in this material.<sup>3,4</sup> In addition, heavy ion irradiation displaced the irreversibility line upward in temperature by 10-25 K or more.

The effects of ion irradiation on single crystals of  $Y_1Ba_2Cu_3O_7$  are similar, although some important differences are apparent. Both point-like<sup>5</sup> and line-like<sup>6</sup> defects from light and heavy ion irradiation, respectively, increase  $J_c$  substantially. At low to moderate temperatures, the two defect morphologies give similar, very high values for  $J_c$ . Near  $T_c$ , however, columnar defects are far more effective than point-like defects and give much higher current densities. Linear defects also

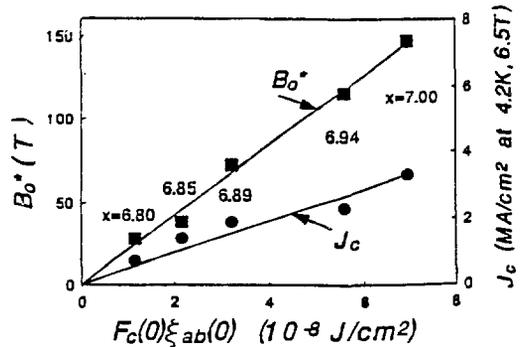


Fig. 2. Correlation between the model pinning parameter  $F_c \xi_{ab}$  and (1) the irreversibility field  $B_o^*$  extrapolated to  $T=0$  and (2) persistent current density at 4.2 K.

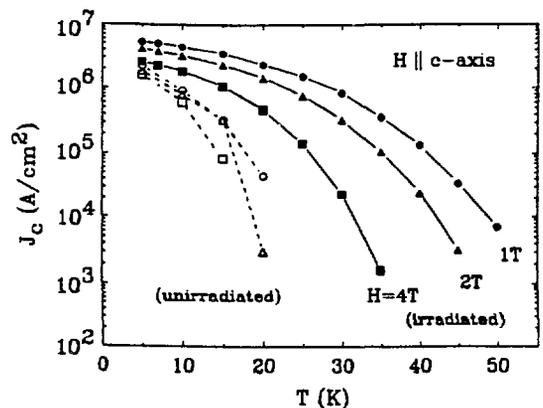


Fig. 3. Persistent current density vs. temperature for a  $Bi_2Sr_2Ca_1Cu_2O_8$  crystal before and after heavy ion irradiation to form columnar defects. Values of field  $H \parallel c$  are shown.

displace the irreversibility line upward in field and temperature, unlike point defects.

### EFFECTIVE VORTEX PINNING ENERGY $U_{\text{eff}}(J, T)$

Associated with vortex motion is a decay of persistent currents with time. Since the irreversible magnetization is proportional to  $J(t)$ , the time dependence of  $M(t)$  depends intimately on the relationship between  $J$  and pinning energy  $U_{\text{eff}}(J)$ . This follows from thermal activation of flux from a pinning well of depth  $U_{\text{eff}}$ . The probability of excitation is proportional to an Arrhenius factor,  $\exp[-U_{\text{eff}}(J, T)/k_B T]$ . From the resulting master rate equation, Maley et al.<sup>7</sup> developed a procedure to obtain the effective pinning energy from experimental quantities:

$$U_{\text{eff}}(J, T) = -T[\ln(dM/dt) - C] \quad (1)$$

Here  $C$  is a temperature independent constant whose value is fixed by requiring that, at low temperature,  $U$  is a continuous function of  $J$ . We set  $k_B = 1$ .

An example of this analysis is given in Fig. 4, a plot of  $U_{\text{eff}}(J, T)$  versus  $J$  on logarithmic axes. These results (lower curve) were obtained from  $M(t)$  data at the temperatures shown, with  $H \parallel c\text{-axis} = 1$  Tesla. The material was a single crystal of  $Y_1\text{Ba}_2\text{Cu}_3\text{O}_7$  containing a nearly optimum density of columnar defects,  $B_\phi = 3$  T. With increasing temperature, the  $U(J, T)$  segments "sag" or fall below any reasonable extrapolation of the lower temperature data. According to Nelson and Vinokur,<sup>8</sup> entropy considerations cause just such an effect, as vortex lines wander at higher  $T$ . For columnar pins, they predict that  $U_{\text{eff}} = U_\phi f(T/T^*)$  with  $f(0) = 1$  and  $f(x) \approx x^2 \exp(-x^2)$  for  $x \gg 1$ . To test this phenomenon and obtain experimental values for  $f$  at temperatures  $T$ , we force  $U_{\text{eff}} \cdot (1/f)$  to be a continuous function of  $J$  at higher

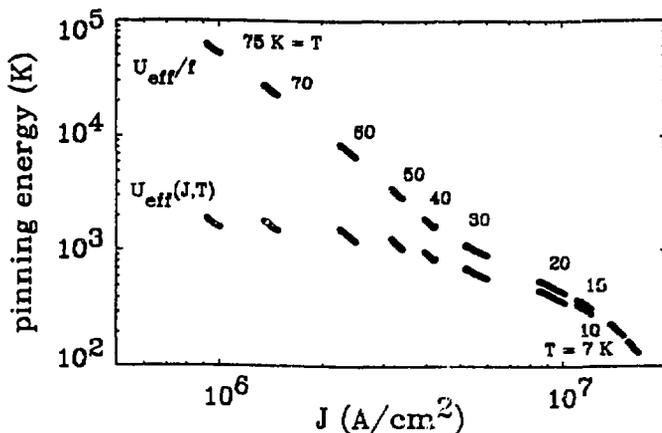


Fig. 4. Lower set of segments show effective vortex pinning energy  $U_{\text{eff}}(J, T)$  vs.  $J$ , for temperatures shown. Upper semicontinuous curve shows  $U_{\text{eff}}/f$ , where factor "f" accounts for thermal smearing of the pinning potential.

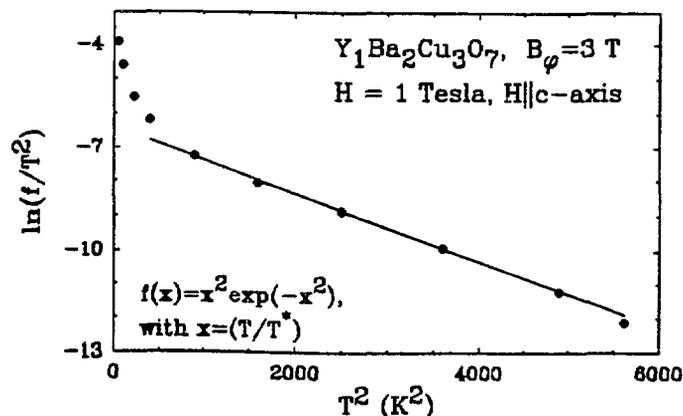


Fig. 5. "Thermal smearing" analysis. A plot of  $\ln(f/T^2)$  vs.  $T^2$ , with pinning reduction factor "f" obtained from Fig. 4. Theory predicts a linear relation for  $T > T^*$ . Here  $T^* = 32\text{K}$ .

temperature. The resulting function  $U_{\text{eff}}/f$  is shown in Fig. 4 also (upper curve). Figure 5 plots the quantities  $\ln(f/T^2)$  vs.  $T^2$ . According to the theory, this plot should be linear at high temperature, as it is. The slope corresponds to  $T^* = 32$  K, which is about half of that estimated for YBCO.<sup>8</sup> As far as we know, this is the first experimental determination of the boundary temperature  $T^*$  of the Bose glass regime. This theoretical configuration of linearly pinned vortices occurs at low temperatures  $T \leq T^*$ , for fields  $B < B_\phi$  as is the case here. Further details will be presented elsewhere.

In summary, defects of varying strength and morphology lead to a rich variety of interesting and practical phenomena in high  $T_c$  superconductors. While many questions remain, it is clear that maximizing  $J_c$  requires a maximum oxygen content in  $Y_1\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$  materials that contains preexisting pins. Over a wide range of conditions, columnar defects pin vortices extremely effectively, perhaps as well as can be achieved.

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