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Flux Pinning in Bi-2212 Single Crystals with Various Oxygen Concentrations[†]

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By systematic variation of the oxygen concentration it is demonstrated that the oxygen vacancies in the CuO_2 planes are indeed the predominant pinning centers in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212) single crystals. Both the critical current density and the pinning energy increase proportional to the square root of the vacancy concentration. We also show that a change of the superconducting parameters upon annealing, predominantly of $\lambda_L(0)$, gives rise to a considerable decrease of the critical current.

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PACS. 74.60.Ge— Flux pinning; flux creep, and flux-line lattice dynamics

Although there are good reasons to believe that flux pinning in as-grown Bi-2212 single crystals is caused by oxygen vacancies in the CuO_2 layers [1,2], there is as yet no hard experimental evidence for this belief. Such evidence can only be provided by studying the effect of a systematic change of the defect concentration on the critical current and pinning energy. Earlier work along these lines [3] gave incomplete support for the case of oxygen vacancies. We followed a slightly different scenario and started with a careful investigation of oxygen diffusion under different annealing conditions [4]. According to this work annealing in air between 500 °C and 800 °C reversibly removes interstitial oxygen from the BiO layers, whereas annealing in nitrogen at 500 °C and 600 °C irreversibly removes oxygen from the CuO_2 sheets. We therefore cut from the same single crystal four equally large ($1.5 \times 1.5 \times 0.02 \text{ mm}^2$) pieces and prepared 4 samples (Bi1 to Bi4) by giving them a different heat treatment, see Table I. The difference between Bi1 and Bi2 is that oxygen was removed from the BiO layers as evidenced by a correlated increase in both T_c , the c-axis lattice parameter and the London penetration depth. The latter parameter was obtained from magnetization measurements by plotting dM/dT vs $\ln H$ [5]. The increase of $\lambda_L(0)$ upon oxygen loss shows that Bi1 is in the overdoped regime and Bi2 optimally doped. Further removal of oxygen, i.e. for Bi3 and Bi4, does not change these parameters anymore, giving strong support to the assumption that for these samples the oxygen also

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comes out of the CuO_2 sheets creating more vacancies in these layers. If pinning is caused by oxygen vacancies we thus expect that the magnetization curves become more irreversible going from Bi2 to Bi3 to Bi4. This effect is indeed observed, as can be seen in Fig. 1. We also see that this increase in pinning is preceded by a strong decrease going from Bi1 to Bi2. As will be shown below this decrease can be related to the change of $\lambda_L(0)$.

For a confrontation of pinning experiments with theory the data in Fig. 1 are not suitable as they are influenced by flux creep. The basic pinning properties at low temperature can be deduced from magnetic relaxation measurements and a Maley analysis [6]. The results of such an analysis for our samples at a field of 2 T are shown in Fig. 2 where the energy barrier for flux creep U is plotted versus the current density j . The logarithmic behavior $U(j) = U_c \ln(j_c/j)$ is typical for creep in the single-vortex pinning regime [7,8]. The pinning energy U_c and the critical current density j_c can be determined from the linear extrapolation to $U = 0$. Results for our samples are given in Table I.

As shown in Ref. 2, the relations for j_c and U_c at $T = 0$, are

$$j_c = A_1 (B_c^2 / \mu_0 \phi_0 s) \sigma_{tr} \xi_0 n_{\square}^{1/2} \quad (1)$$

$$U_c = j_c \phi_0 s r_f = A_2 j_c \phi_0 s \xi. \quad (2)$$

Here n_{\square} is the areal density of oxygen vacancies in the CuO_2 double layers, σ_{tr} is the electron scattering crosssection of a vacancy; s the crystal lattice periodicity, r_f the range of the pinning potential and A_1 and A_2 are constants of order 10 and 1, respectively. For Bi-2212 we use the following parameter values: $B_c(0) = 0.5$ T, $\xi(0) = 2.1$ nm, $\xi_0 = 2.8$ nm, $s = 1.54$ nm and $\sigma_{tr} = \pi D^2/4$ with $D = 2.9$ nm.

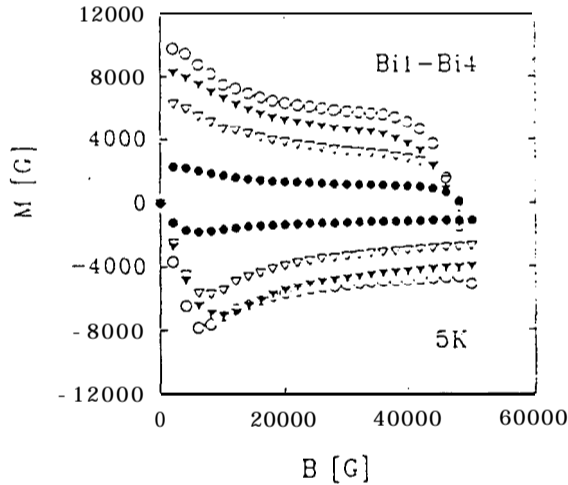


FIG. 1. Magnetization curves at 5K of Bi-2212 single crystals with different oxygen content: Bi1(\circ), Bi2(\bullet), Bi3(∇), Bi4(\blacktriangledown). The decrease of the irreversibility from Bi1 to Bi2 is related to a change of superconducting parameters, whereas the increase in going from Bi2 to Bi4 is due to an increase of vacancies in the CuO_2 layers.

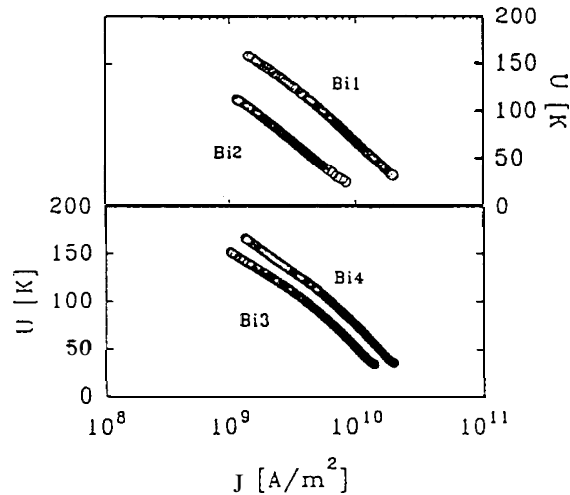


FIG. 2. Creep energy vs current density determined from a Maley analysis [6,7] of magnetic relaxation experiments at 2 T on samples Bi1-Bi4.

TABLE I. Characteristic data of Bi-2212 samples after different heat treatments.

	T_{ann} (°C)	Atm.	T_c (K)	λ_L (nm)	Δx	J_c (10^{10} Am^{-2})	U_c (K)	n_{\square} (10^7 m^{-2})	n_{\square} (%)
Bi1	500	air	83.5	180	0	3.5	55	0.7	0.3
Bi2	800	air	89.5	260	0.023	1.3	53	0.7	0.3
Bi3	500	N ₂	89.0	260	0.054	2.3	56	2.2	0.8
Bi4	600	N ₂	89.0	260	0.089	3.3	61	4.8	1.7

In order to compare our data with these predictions we plot in Fig. 3 j_c^2 and U_c^2 versus the change in oxygen concentration Δx . The latter is related to n_{\square} via $n_{\square} = 6.9 \times 10^{18} \Delta x \text{ m}^{-2}$. Within experimental accuracy the data fall on straight lines. From the slopes we determine $A_2 = 2.0$. We thus find that $\tau_f \simeq 2\xi$, which is quite reasonable. It shows that the changes in j_c and U_c are consistently described by the pinning theory for single pancake vortices. The experimental value of $dj_c^2/d\Delta x$ is $1.6 \times 10^{22} (\text{A/m}^2)^2$. Comparison with Eq. (1) yields $A_1 \simeq 4.1$, which is also a quite reasonable value. We therefore conclude to have convincingly shown that pinning in Bi-2212 is predominantly caused by oxygen vacancies in the CuO_2 layers.

Given the uncertainties in both the parameter values for Bi-2212 and the value of A_1 (in Ref. 2 we assumed $A_1=17$), it is only possible to deduce a rough estimate for n_{\square} . The

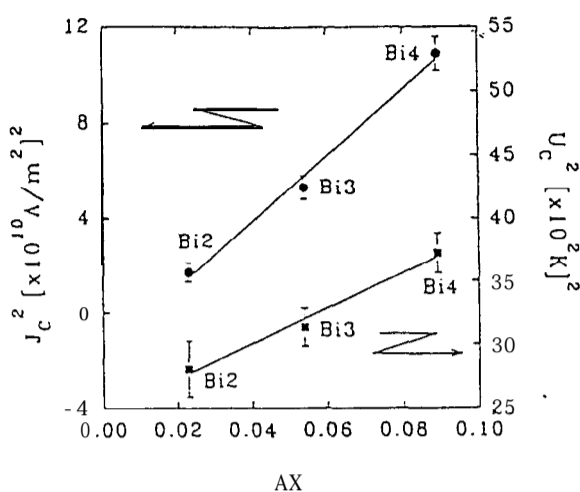


FIG. 3. j_c^2 and U_c^2 vs change in oxygen concentration for Bi2, Bi3 and Bi4 determined from data in Fig. 2.

following consideration may help. The data in Fig. 3 actually show an upward curvature. This would be expected if the vacancy concentration for Bi2 is too low to justify use of Eq. (1). This relation requires that there should at least be one vacancy per vortex core, i.e. $n_{\square} > (\pi\xi^2)^{-1} \approx 7 \times 10^{16} \text{ m}^{-2}$ corresponding to a vacancy concentration of 0.26 percent in the double layer. Assuming these values to be reasonable estimates for Bi2, we find for Bi3 and Bi4 the values listed in Table I. Concentrations of up to 2 percent in the CuO_2 layers have been reported for a related compound, i.e. $\text{La}_{1.8}\text{Sr}_x\text{Ca}_{1.2-x}\text{Cu}_2\text{O}_{6-\delta}$ [9].

Next we discuss the difference between Bi1 and Bi2. Although the data in Fig. 3 indicate that there may be an additional pinning mechanisms active in Bi2, we ignore this and try to explain the difference by investigating the effect of a change in the superconducting parameters between Bi1 and Bi2 assuming Eq. (1) describes the pinning and n_{\square} does not change. It follows from Eq. (1) that $j_c \propto B_c^2 \xi_0$ which with $B_c = \Phi_0 / \xi_0 \lambda_L$ can be written as $j_c \propto \Phi_0^2 / \lambda_L^2(0) \xi_0$. The increase of $\lambda_L^2(0)$ by a factor of 2.1 (see Table I) would largely account for the drop in j_c between Bi1 and Bi2. However, it seems likely that the reduction in charge carriers will give rise to a slight decrease of ξ_0 . This effect may partially compensate the increase of λ_L^2 . For a 2D BCS model we would obtain $j_c \propto 1/\lambda_L(0)$ which only qualitatively explains the difference in j_c between Bi1 and Bi2.

In summary, a systematic variation of the oxygen concentration by annealing Bi-2212 in air or nitrogen, decreases the critical current density by a factor of two due to an increase of the penetration depth and subsequently enhances j_c back to the original value by an increase of the oxygen vacancy concentration in the CuO_2 layers from about 0.3 percent to about 1.7 percent.

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