Correlation between J_c and screw dislocation density in sputtered $YBa_2Cu_3O_{7-\delta}$ films

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Electric transport properties of sputtered $YBa_2Cu_3O_{7-\delta}$ films were studied as a function of screw dislocation density, ranging from $5 \cdot 10^7$ cm^{-2} to $1.3 \cdot 10^9$ cm^{-2} as determined at the film surface. A correlation was found between the number of screw dislocations and the critical current density (J_c). Films with higher screw dislocation densities have higher critical current densities and a slower drop of J_c as a function of applied magnetic field H.

Introduction

Critical current densities (J_c) up to $8 \cdot 10^6$ A/cm^2 at 77 K have been observed under self-field conditions in epitaxial $YBa_2Cu_3O_{7-\delta}$ films prepared by sputtering [1, 2] and laser ablation [3]. To account for such high critical current densities, which are about one order of magnitude higher at 4.2 K, strong pinning sites for the magnetic flux quanta are required. Although pinning at twin boundaries [4, 5], at the surface [4], at impurity phase inclusions [6], and due to the layered structure of the high-T_c cuprates [4, 7] has been observed to some extent, strong pinning sites capable of supporting the high current densities observed have yet to be identified. Various other pinning sites have been proposed for high-T_c materials, such as dislocations [8–10], and point defects [8, 11].

Scanning tunneling microscopy (STM) of the as-grown surfaces of sputtered and laser-ablated $YBa_2Cu_3O_{7-\delta}$ films has revealed the presence of high densities ($\approx 10^9$ cm^{-2}) of screw dislocations [1, 12]. Subsequent studies have shown that the screw dislocation densities of $YBa_2Cu_3O_{7-\delta}$ films can be controlled by varying the growth conditions [13]. Utilizing this ability to vary the screw dislocation density, we report in this paper on the observation of a correlation between the screw dislocation density and J_c of $YBa_2Cu_3O_{7-\delta}$ films.

Dislocations, being line defects, may be effective pinning sites for the case where the flux lines are oriented parallel to the line of the dislocations, since pinning can then occur over a substantial fraction of the flux line length. In the present study this is the relevant configuration; the flux lines generated by the self-field of the transport current or by the externally applied field are oriented predominantly[1] parallel to the film's c-axis, (normal to the film surface) and are thus parallel to the dislocation core (Fig. 1).
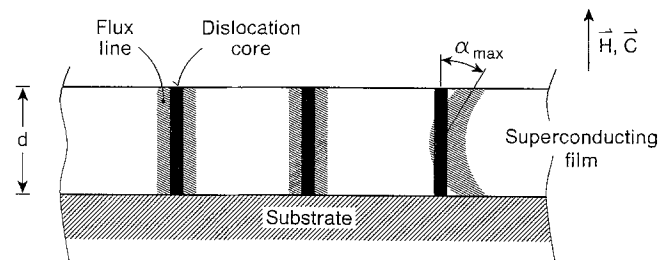
Fig. 1. Sketch of the sample configuration. The flux line at the right-hand side is drawn with a parabolic shape to illustrate the angle α_{max} as described in the text

[1] Note that when the externally applied field is small, some curvature of the flux lines is expected. As pointed out by Brandt [14], the equilibrium configuration of flux lines in a film of thickness d, where d is much greater than the magnetic penetration depth λ, carrying a current density J in the presence of a transverse applied magnetic field H, is one of parabolically shaped flux lines tilted from the film surface normal by an angle α that reaches a maximum value at the film surface given by $\alpha_{max} = \tan^{-1}(Jd/2H)$ (Fig. 1). This relation provides an upper limit for the maximum tilt of the flux lines at the film surface in our studies: For $d = 100$ nm and $J_c = 5 \cdot 10^7$ A/cm^2, $\alpha_{max} \simeq 45°$ for $H \simeq 0.03$ T. The actual tilt is expected to be much smaller for several reasons: the thicknesses of the films investigated are small compared to λ, and the effects of the line tension of the vortices and pinning at line defects were not included in the derivation of α_{max}

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The maximum core pinning force per unit length, f_p^{max}, on a flux line parallel to a dislocation core can be estimated by [11]

$$f_p^{max}(T) = \frac{\Phi_0^2}{16\pi\mu_0 \lambda^2(T)\,\xi(T)} \qquad (1)$$

where Φ_0 is the magnetic flux quantum, $\lambda(T)$ is the magnetic penetration depth, and $\xi(T)$ is the coherence length (MKS units). Note that pinning may also result from electrostatic effects, as proposed by Pan et al. [9]. For c-axis oriented $YBa_2Cu_3O_{7-\delta}$ films, Eq. (1) yields a maximum pinning force per unit length of $f_p^{max} \approx 2\cdot10^{-3}$ N/m at 4.2 K. In the critical state the pinning force f_p equals the Lorentz force f_L per unit length: for $J_c = 5\cdot10^7$ A/cm^2, $f_p = f_L = J_c\Phi_0 = 10^{-3}$ N/m, or $f_p \approx 1/2 f_p^{max}$. This shows that pinning at dislocations may account for the observed critical current densities. As will be discussed below, additional defects are necessary to explain the observed critical current densities in magnetic fields greater than about 0.05 T. This is in accordance with the work of Hylton and Beasley [11], who concluded that extended defects, such as dislocations, could not account for the pinning characteristics of high-T_c films, if those were the *only* pinning sites present. Our results also agree with calculations of the pinning force of screw dislocations based on Thuneberg's formalism [15], as shown in recent work by Kes [16].

Experimental

The c-axis oriented $YBa_2Cu_3O_{7-\delta}$ films, 100–150 nm thick, were grown by DC hollow cathode magnetron sputtering on nominally (100) oriented $SrTiO_3$ substrates. The sputtering parameters used were a pressure of 87 Pa ($Ar:O_2 = 2:1$), a plasma discharge at 150–180 V with a current of 260–500 mA, a heater block temperature of 750–780 °C and growth rates ranging from 0.05 nm/s to 0.18 nm/s [13]. Electrical transport measurements indicated typical T_c ($\rho = 0$) values of 87–89 K, resistivity ratios (ρ_{300}/ρ_{100}) of 2.5–3, and $J_c = 1$-$8\cdot10^6$ A/cm^2 at 77 K and $J_c = 1$-$7\cdot10^7$ A/cm^2 at 4.2 K.

Each sample for J_c measurements, cut from the same polished substrate as the corresponding two samples for STM investigation, was mounted on the substrate heater block between the STM samples for the deposition of the $YBa_2Cu_3O_{7-\delta}$ film. Transport measurements were made using a 4-point geometry, and magnetic fields up to $\mu_0H = 8$ T were applied perpendicular to the film surface ($H \parallel c$). The current at which a 1 μV voltage drop was observed across the 8 μm·100 μm bridge was taken as the critical current. The J_c measurement was repeated on several samples after cycling them to room temperature. The scatter in the resulting data was about ten percent, giving a lower bound on the precision of the J_c values.
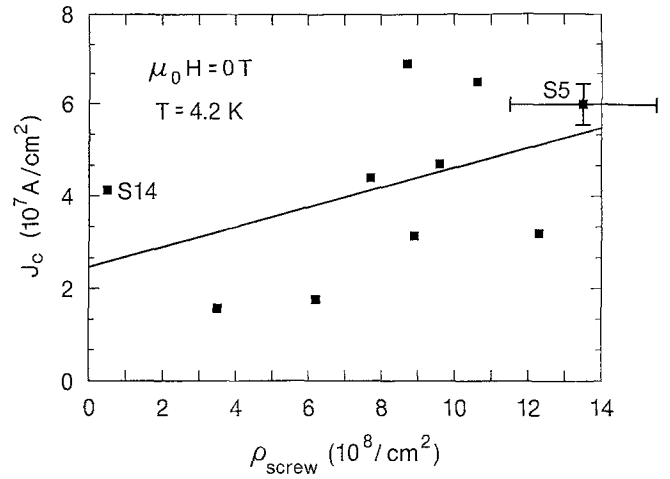
Fig 2. Critical current density (J_c) in the absence of applied magnetic field at 4.2 K as a function of the surface screw dislocation density (ρ_{screw}) measured by STM. The error bars on a typical data point are indicated, as is the line of best fit. (Samples # S5 and S14 refer to Figs. 3–5 and [13])
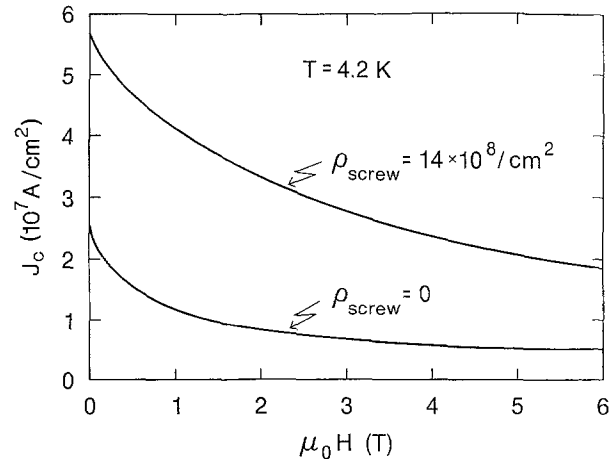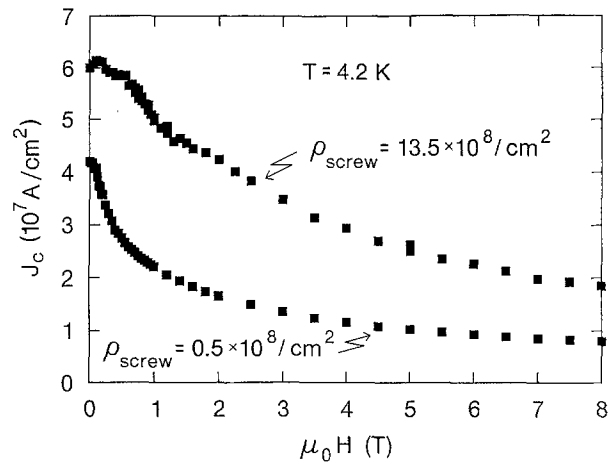
Fig. 3a, b. Critical current density (J_c) as a function of the applied magnetic field ($H \parallel c$) at 4.2 K **a** for two sputtered $YBa_2Cu_3O_{7-\delta}$ films having surface screw dislocation densities of $0.5\cdot10^8$ cm^{-2} and $13.5\cdot10^8$ cm^{-2} (samples S14 and S5 in [13]) and **b** extrapolated $J_c(H)$ curves corresponding to $\rho_{screw} = 0$ and $\rho_{screw} = 14\cdot10^8$ cm^{-2} extrapolated from lines of best fit to $J_c(\rho_{screw})$ (of the same type shown in Fig 2) for μ_0H varied from 0–6 T

Results and discussion

Experimentally, we observe a correlation between J_c and the screw dislocation density ρ_{screw}. This trend is shown in Fig. 2 for the case of zero external magnetic field and a temperature of 4.2 K for all of the samples investigated. The scatter in the data in excess of the measurement uncertainties of J_c and ρ_{screw} (as indicated by the estimated error bars) is attributed to variations in film quality, owing to the large parameter window in which the samples were grown. As the line of least squares fit indicates, J_c in self-field increases with a slope of 20 mA/cm^2 per screw dislocation/cm^2 reaching the film surface. However, this correlation does not prove a causal relationship between screw dislocations and J_c.

The magnetic field dependence of J_c at 4.2 K is shown in Fig. 3a for two films having $\rho_{screw} = (0.5 \pm 0.2) \cdot 10^8$ cm^{-2} and $(13.5 \pm 2) \cdot 10^8$ cm^{-2}. Even for the film with the higher screw dislocation density the flux line density exceeds the screw dislocation density for applied fields $\mu_0 H \gtrsim 0.03$ T. To account for the observed critical current density in fields where the Lorentz force on the flux lines exceeds the maximum volume pinning force of the screw dislocations $f_L \dfrac{B}{\Phi_0} = J_c B \geq f_p^{max} \rho_{screw}$, which occurs for $B \gtrsim 0.05$ T, additional pinning sites must be present in the films.

It might be expected that characteristic features would appear in the $J_c(\mathbf{H})$ curves at the crossover between the low field (in which the screw dislocations could account for J_c) and the high field regime. Indeed, as shown in Fig. 4, $J_c(\mathbf{H})$ decreases in low fields more slowly with increasing ρ_{screw}. This is also demonstrated by the two samples shown in Fig. 3a. To produce $J_c(\mathbf{H})$ curves representative of all samples, linear least squares fits to the $J_c(\mathbf{H})$ versus ρ_{screw} data of all the samples (of the same type shown in Fig. 2) were made for $\mu_0 H = 0$–6 T. The $J_c(\mathbf{H})$ values corresponding to these lines of best fit are shown for $\rho_{screw} = 0$ cm^{-2} and $\rho_{screw} = 14 \cdot 10^8$ cm^{-2} in Fig. 3b. These curves confirm that in
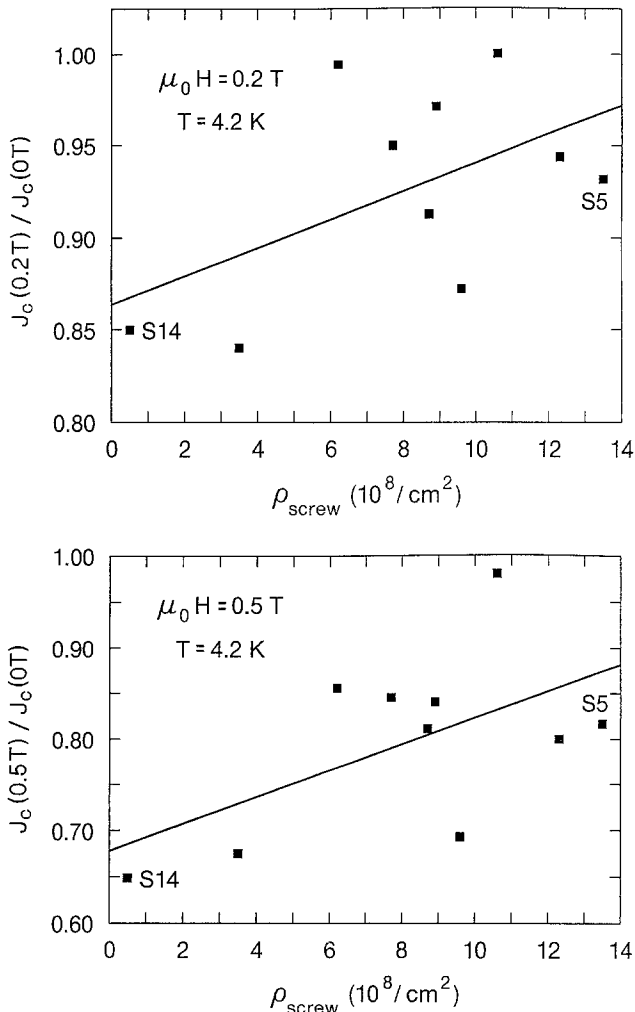
Fig. 4a, b. Critical current density (J_c) at 4.2 K in an applied magnetic field ($\mathbf{H} \| c$) of a $\mu_0 H = 0.2$ T and b $\mu_0 H = 0.5$ T, normalized to that in zero applied field as a function of the observed surface screw dislocation density (ρ_{screw}) for sputtered YBa$_2$Cu$_3$O$_{7-\delta}$ films grown on nominally (100) oriented SrTiO$_3$ substrates
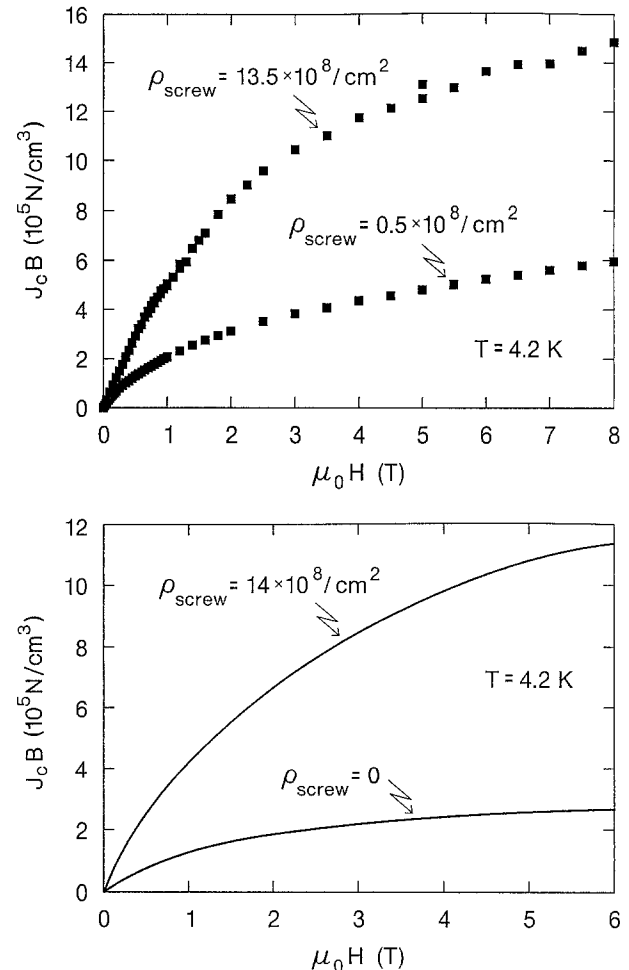
Fig. 5a, b. The volume pinning force ($J_c B$) as a function of applied magnetic field at 4.2 K a for two sputtered YBa$_2$Cu$_3$O$_{7-\delta}$ films having surface screw dislocation densities of $0.5 \cdot 10^8$ cm^{-2} and $13.5 \cdot 10^8$ cm^{-2} (samples S14 and S5 in [13]) and b extrapolated $J_c(\mathbf{H})$ curves corresponding to $\rho_{screw} = 0$ and $\rho_{screw} = 14 \cdot 10^8$ cm^{-2} extrapolated from lines of best fit to $J_c(\rho_{screw})$ (of the same type shown in Fig. 2) for $\mu_0 H$ varied from 0–6 T

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low fields a more gradual decrease in $J_c(\mathbf{H})$ for higher ρ_{screw} is a general trend. The absence of a pronounced kink in these curves, denoting the crossover region, is attributed to the averaging involved in performing the linear extrapolations, the random spatial distribution of the screw dislocations, the variation of their pinning strengths (not all screw dislocations necessarily extend through the entire film thickness), and to other pinning sites in the samples. Additional pinning sites may also be the reason why the crossover of the $J_c(\mathbf{H})$ curve appears at ≈ 0.5 T, one order of magnitude higher than the crossover regime estimated above.

Figure 5 shows the magnetic field dependence of the volume pinning force and least squares fit data for the same two samples presented in Fig. 3a. As demonstrated by this figure, even in fields of 8 T the volume pinning force increases as a function of field strength which provides evidence for a high density of additional pinning sites. Furthermore, Fig. 5b reveals that the additional pinning sites are correlated with the density of threading screw dislocations, and thus have to be defects, which contribute to about 75% of the total pinning strength in the films.

Likely candidates for the vortex pinning sites required to explain the behavior of J_c in high magnetic fields are point defects [11], edge dislocations [9, 10], impurity

Fig. 6. High resolution cross-sectional TEM image of a $YBa_2Cu_3O_{7-\delta}$ film (sample S8 in [13]) showing the presence of a small elongated precipitate (indicated by the arrow). The inset shows a dark field view of the same precipitate at lower magnification

phase inclusions [6], or twin boundaries [4]. In view of the fact that measurements made on the same twinned and detwinned single crystals showed a negligible role of twin boundaries on flux pinning at low temperatures [5], and that films produced with different twin spacings do not exhibit corresponding changes in volume pinning force [17], and that the highest critical currents (up to $1.1 \cdot 10^7$ A/cm^2 at 77 K) for $YBa_2Cu_3O_{7-\delta}$ layers were observed in twin-free $YBa_2Cu_3O_{7-\delta}/(Nd, Ce)_2CuO_x$ multilayers [10], twin boundaries appear unlikely as strong pinning sites. But $YBa_2Cu_3O_{7-\delta}$ films having high critical current densities ($8 \cdot 10^6$ A/cm^2 at 77 K) [1, 3] have been found to contain many defects including stacking faults [3, 18, 19], small precipitates [3, 18, 19], and dislocations [1, 18]. An example of a small precipitate revealed by transmission electron microscopy (TEM) investigations of these films [19] is shown in Fig. 6. This elongated impurity phase inclusion measuring about 5 nm across and nearly extending through the entire 150 nm thick film, has a shape well suited to pinning. The total density of small precipitates (of all shapes) observed in these sputtered $YBa_2Cu_3O_{7-\delta}$ films is of the order of 10^{16} cm^{-3} [19], suggesting that the defects themselves or the stress fields associated with them may contribute significantly to pinning. Edge dislocations may be particularly strong sources of pinning. Very high densities of edge dislocations ($\simeq 10^{11}$ cm^{-2}) have been reported recently for ultrathin $YBa_2Cu_3O_{7-\delta}$ films grown on MgO [20]. In that study the edge dislocations were found in the regions of island coalescence during film growth. This suggests that in our films edge dislocations may be present around the perimeter of the growth spirals in order to accommodate slight misorientations between neighboring growth spirals as they coalesce. Although the location of such defects with respect to the growth spirals could not be identified yet, high resolution electron microscopy along the c-axis provided evidence for a high density of edge dislocations ($\simeq 10^{11}$ cm^2) with Burgers vectors $b = [100]$ and $[010]$ [19]. The pinning energy of such edge dislocations is expected to be particularly high as they are located in the valleys between the growth spirals which are low energy positions for the flux lines. Pinning by edge dislocations is also suggested in studies of incoherent $YBa_2Cu_3O_{7-\delta}/(Nd, Ce)_2CuO_x$ multilayers [10] for which high critical current densities were found for layer thicknesses at which the formation of edge dislocations was expected. Ways in which the growth process of $YBa_2Cu_3O_{7-\delta}$ films may give rise to edge dislocations are described elsewhere [12, 13]. It is also reasonable to assume that point defects such as missing oxygen atoms, contribute to pinning. But if the pinning were only due to point defects, point defects with densities $> 4 \cdot 10^{19}$ cm^{-3} would be required [11], which seems to be a rather high number.
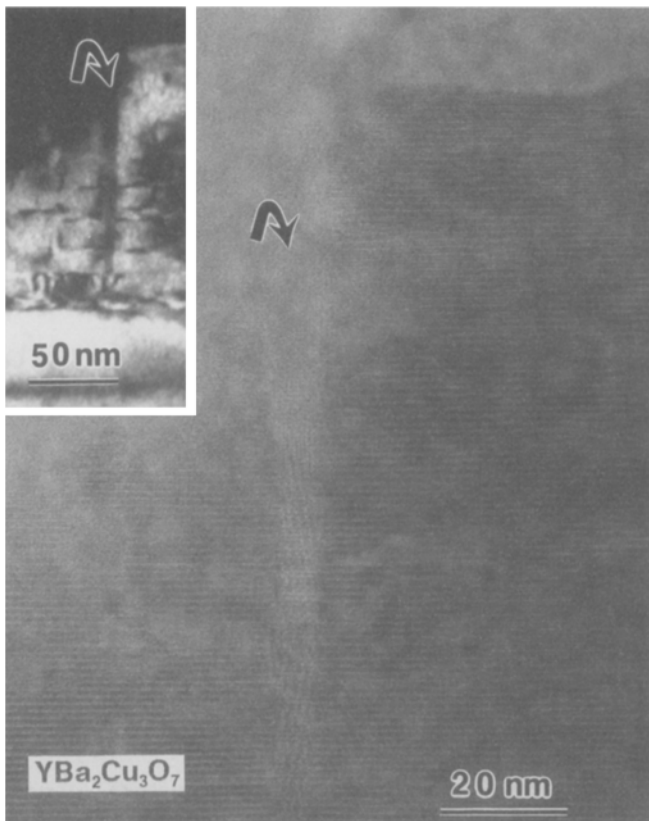
Summary

In summary, a correlation between the density of screw dislocations and J_c has been observed. Films with higher

screw dislocation densities have higher J_c, a slower drop of $J_c(H)$, and higher volume pinning forces. The observed number of screw dislocations is sufficient to account for the observed J_c in small magnetic fields ($\lesssim 0.05$ T). Additional pinning sites, created by defects apparently correlated to the presence and density of the screw dislocations, are required to explain the J_c observed in higher magnetic fields.

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