Ion beam thinning and polishing of $YBa_2Cu_3O_7$ films

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Successive ion beam milling at grazing angles of a 2400-Å-thick, *c*-axis-oriented $YBa_2Cu_3O_7$ film is shown to give smooth films which superconduct at thicknesses on the order of tens of angstroms. The thinning and polishing process is characterized at successive milling stages using resistance transitions, x-ray analysis, scanning electron microscopy, and Rutherford backscattering and channeling analysis of composition and surface quality. As thinning proceeds, scanning electron microscopy and diffraction features associated with *a*-axis texture are removed and crystalline quality, as measured by x rays and channeling, markedly improves.

Ion beams have found a wide variety of uses in the post-deposition processing of high T_c thin films. For example, ion beam etching and focused ion beam lithography techniques[1,2] have been used to obtain micron size features. In addition, tailoring of materials properties, such as the transition temperature T_c and the critical current density, can be achieved by introducing controlled ion damage or implantation into selected areas, usually weak links.[3,4] To our knowledge, aside from a report of the high milling rate of bulk ceramic $YBa_2Cu_3O_7$,[5] there have not been any detailed reports on thinning and/or polishing of high T_c films with ion beams. However, with respect to nonsuperconducting materials, this area of application has received much attention and is reviewed in a number of places.[6,7]

Although metallic films have been successfully thinned,[8] there are critical material-dependent factors which must be taken into consideration to minimize ion-induced texturing of the surface. Usually, the ion beam is incident at a low grazing angle to facilitate the removal of high spots. Surface damage, either by implantation of bombarding ions or sputtering of target atoms, can be reduced by lowering the energy and angle of incidence of the bombarding ions. Other factors affecting the extent of surface damage include temperature, chemical composition, crystal orientation, surface topography, type of bombarding ion, and the heat of sublimation of the bombarded material.[6-8]

In this letter, we present results showing that ion (Xe^+) beams can be successfully used to thin *and* polish $YBa_2Cu_3O_7$ films. Although we do not make any effort to optimize the milling parameters to minimize surface damage, we do show that a 2400 Å *c*-axis-oriented $YBa_2Cu_3O_7$ film, grown on a (100) $SrTiO_3$ substrate, can be milled to an "electrical" thickness of <40 Å without losing its superconducting porperties. Characterization after each successive milling stage by x rays, scanning electron microscopy (SEM), and Rutherford backscattering spectroscopy (RBS) and channeling confirms that polishing with a concomitant removal of a textured *a*-axis component is occurring.

The results reported here are on a 2400-Å-thick *c*-axis-oriented $YBa_2Cu_3O_7$ film made in a three-source evaporator. Film robustness with respect to degradation by water vapor and atmospheric contaminants was obtained by using BaF_2 in one of the sources.[9] Details of the film preparation and post-deposition annealing are reported elsewhere.[10] The initial film thickness was measured with a surface profilometer. Two films from the same substrate, one for electrical characterization and the other for x ray, SEM, and RBS characterization, were milled for selected times and then remeasured after each milling. This process was repeated until the films were milled to a thickness approaching tens of angstroms where the conductivity was beginning to rapidly approach zero. During milling, the films were mounted on a substrate platform which was rotated at ~ 2 Hz and oriented at a 10° glancing angle of incidence with respect to the impinging beam. The xenon ions with an energy of 1000 eV and a current density of 150 μA/cm^2 were extracted from a Kaufman ion source[11] operating at a pressure of 1.5×10^{-4} Torr.

Electrical measurements were taken using the Van der Pauw technique with four equally spaced indium contacts soldered at the boundary of the film. The contacts remained in place for the complete set of measurements. Figure 1 is a

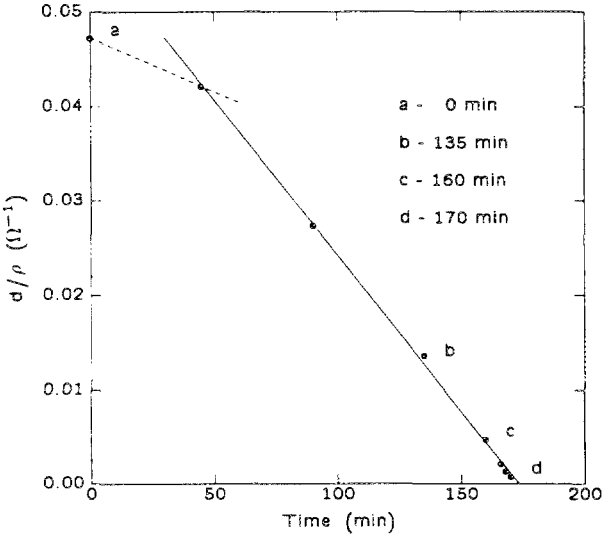

FIG. 1. Plot of the room-temperature sheet conductance vs milling time. The labeled points, with milling times identified in the legend, correspond to the resistive transitions in Fig. 2. The dashed line corresponds to the slow removal of *a*-axis-oriented material initially at the surface and the solid line indicates a uniform removal of *c*-axis material.

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plot of the room-temperature sheet conductance, equal to the film thickness d divided by the film resistivity ρ, versus milling time. After 45 min of milling time, the trend in the data is linear (solid line), hence indicating a uniform milling rate. The use of the sheet conductance as a direct measure of d presupposes that ρ is independent of thickness. We do not expect size effect in the resistivity to be important because of the short scattering length along the c axis. The final thickness after 170 min of milling time [(d) in Fig. 1] is calculated to be roughly equal to the 2400 Å starting thickness multiplied by the ratio of the ordinate values for d/ρ at (d) and (a). This thickness of 40 Å, achieved after 170 min of milling, represents an *electrical* thickness and does not include a possible insulating dead layer which would not contribute to the conductivity.

Selected resistive transitions corresponding to the points (a)–(d) in Fig. 1 are shown in Fig. 2. Initially, the film had a zero-resistance transition at 90 K and a critical current density at 77 K of 1.8×10^6 A/cm^2. A second post-deposition anneal at 300 °C inadvertently resulted in a degradation of the film to the broader and lower T_c transition shown in (a) of Fig. 2. Separate films having initial resistive transitions <1 K wide (10–90% normal-state criterion) at 90 K show a significantly reduced degradation of T_c with thinning. Nevertheless, the salient feature of these data presented here is that the metallic character of the initial 2400-Å-thick film has been preserved at all stages of milling, and a zero resistance T_c of 40 K for an electrical thickness of 40 Å has been obtained.

The SEM micrograph of the surface of the unmilled film shown in Fig. 3(a), which corresponds to label (a) of Figs. (1) and (2), reveals appreciable surface texture. The trellis-like lattice of rods is attributed to an a-axis-oriented component[12] which, as shown in Fig. 3(b), is mostly removed after 45 min of milling. The removal of this more open highly textured material at the surface of the film has less effect on the electrical properties and proceeds at a slower rate (dashed line of Fig. 1) than does the removal of the more dense subsurface material. The sputtering yield for a-axis-oriented material may also be different than for c-axis-oriented material. For longer milling times, additional SEM micrographs (not shown) reveal an increasing smoothness and paucity of surface features, observations consistent with improvements in planarization proportional to milling time.

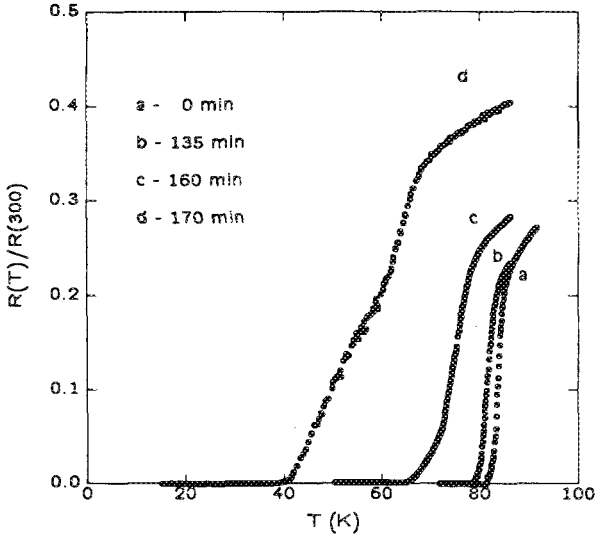

FIG. 2. Plot of the resistive transitions, normalized to the respective room-temperature resistances, for four different stages of milling identified in the legend and in Fig. 1. The temperature coefficient of resistance is positive up to room temperature.

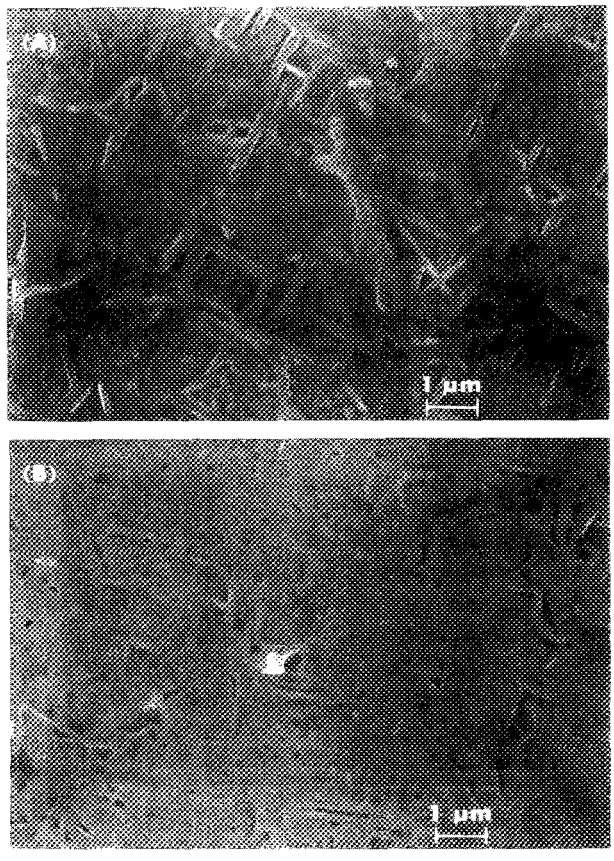

FIG. 3. SEM micrographs of (a) the unmilled film and (b) the same film after milling for 45 min.

The c-axis orientation of the films was established by x-ray scans along [00l] (θ-2θ scans normal to the substrate) on films annealed for 0–135 min. Figure 4 is a portion of two scans showing data for the as-grown film (closed circles) and after milling for 45 min (open circles). Nearly 100 % of the film is oriented with the c axis normal to the substrate as indicated by the near absence of a peak at the (200) scattering angle. [Other films prepared under different conditions have shown substantial (200) peaks that can even be larger than (00l) peaks.] The disappearance of the (200) shoulder after 45 min of milling is consistent with the disappearance of the trellis-like structure shown in Fig. 3 and suggests that the a-axis-oriented material is confined to the film surface.

The peaks marked "x" in Fig. 4 are unidentified impurity peaks of $SrTiO_3$ that, within a factor of 3, occur with the same intensity in scans of the bare substrate. The changes in the intensity of the impurity peaks probably result from a spatial variation of the impurity phase concentration since the x-ray beam scatters from a slightly different volume in each scan. The x-ray absorption of the film is negligible as

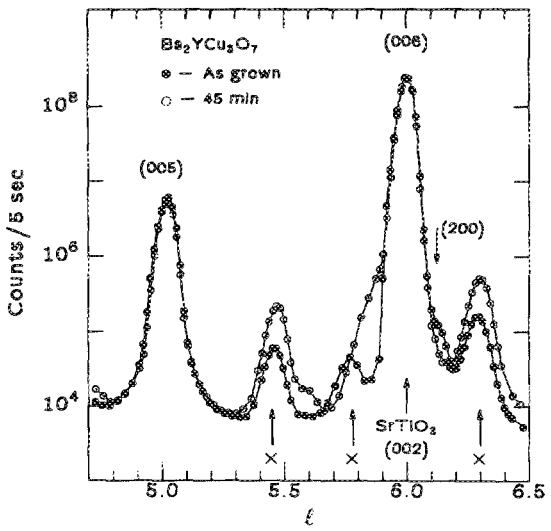

FIG. 4. Scan along [00*l*] for the as-grown film (●) and after 45 min of milling (○). The peaks marked "×" are $SrTiO_3$ impurities also seen in the bare substrate. Note that the weak (200) feature, identified as an *a*-axis component of the film, disappears after the first stage of milling.

indicated by the lack of intensity change in the $SrTiO_3$ peaks as the film is thinned. As expected, the intensities of all (00*l*) peaks scale with the same milling time dependence as the resistance data of Fig. 1.

RBS data using 2 MeV He^+ ions scattered at 170° were also recorded after each milling step. There was no apparent variation of composition with milling time. The RBS thicknesses calculated using the theoretical density for $YBa_2Cu_3O_7$ (7.57×10^{22} atoms/cm^2) scale roughly linearly with time and thus agree with the electrical thickness dependence shown by the solid line in Fig. 1. More important, χ_{min}, the ratio of the channeled yield to the random yield taken in the Ba region of the spectrum, decreases by a factor of 3 from 34% for the unmilled film [(a) of Figs. 1 and 2] to 11% for the film milled for 135 min [(b) of Figs. 1 and 2], indicating that the degree of film alignment has increased from 66 to 89%. This result is in accord with the x ray and SEM results and confirms our conclusion that, as the films are milled and polished, the disordered and textured *a*-axis component is removed leaving an ordered 100% *c*-axis phase with low χ_{min}.

From the results reported here we conclude that ion beams can be successfully used as a thinning and polishing agent for high T_c *c*-axis-oriented $YBa_2Cu_3O_7$ films. We presume that the success of this technique for this case derives primarily from anisotropic sputtering rates correlated with the intrinsic anisotropies of these atomically layered materials. Clearly, ion-induced surface damage caused by preferential sputtering and associated nucleation and diffusion of defects has a deleterious effect on the T_c of ultrathin films. It is difficult to ascertain how much of this T_c reduction occurs because of interdiffusion of impurities across the substrate-film interface, a process enhanced by the high temperatures (~800 °C) used during post-deposition annealing, or because of ion beam induced damage during milling. Nevertheless, the observation of $T_c = 40$ K in a film milled to a thickness of 40 Å is an encouraging result for the use of this technique in the fabrication of ultrathin films for both device applications and fundamental physics studies.

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