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VSM measurements of critical currents for $^2H^+$ and $^{16}O^+$ low energy ion implanted $YBa_2Cu_3O_{7-x}$ thin films

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Abstract

A Vibrating Sample Magnetometer (VSM) was used to study the magnetic properties of $YBa_2Cu_3O_{7-x}$ (YBCO) thin films sputtered onto $LaAlO_3$, $SrTiO_3$ and MgO substrates, before and after ion implantation with deuterium and $^{16}O^+$. By applying the Bean model, the irreversible magnetisation M, was converted into a critical current density J_c, using a length scale Λ determined from a non-destructive technique, at each temperature. The implantation led to significant changes in the superconducting properties of the films. Most notably, for the deuterium implantation in lower T_c films (84K typically), T_c was enhanced by several degrees and M/Λ was enhanced by 1.1 - 1.25 times. In higher T_c films (92K), T_c and J_c were unaltered at the present fluencies used. A similar observation was made for films with a range of T_c values implanted with $^{16}O^+$; when the T_c was unaltered after implantation, M/Λ was also unchanged, but where T_c was depressed M/Λ was enhanced by a factor of 2.3. Possible mechanisms are discussed.

1. Introduction

Fundamentally a change in the T_c of a HTS sample is important because it implies that the mechanism creating the superconductivity is being altered in some way. Previous reports of T_c enhancement in bulk YBCO samples after implantation of protons have been made by Yokota [1] and Pang [2], in beams of 500 and 200 keV and fluencies of 1 x 10^{17} and 1 x 10^{15}/cm^2, respectively. Yokota explains the change as either the result of an improvement in the electronic band structure, a rearrangement of oxygen vacancy sites or a fortuitous elastic distortion of the unit cell caused by the implanted H^+ ions. Alternatively, Kato [3] has reported drops in T_c in bulk samples when ion implanted with 400 keV protons at fluencies between 7.6 x 10^{15} and 2.3 x 10^{17}/cm^2. This was attributed to deoxygenation of the sample by H^+ ions.

Several other groups have reported depression of T_c and then recovery of original value by annealing in oxygen [4,5]. In particular Matsui et al [5], using x rays, measured an increase in the c axis lattice parameter when the T_c of his thin YBCO films dropped after irradiating with 200 keV neon ions at doses between 1 x 10^{14} and 1 x 10^{17}/cm^2. This implies that the oxygen content was lowered by the implantation process in this case.

From a technological aspect, it is crucial to investigate any mechanisms which can enhance the flux line pinning and therefore the current carrying capacity of the HTS films. Several groups have used magnetisation experiments to determine J_c from the Bean model. However unless the length scale of the screening current is determined the Bean formula can not be applied accurately [6]. Very recently a report of J_c enhancement in 0.9 T by a factor of two was made by H+ or Xe+ irradiation using good quality thin films [7]. It has become popular to quote J_c(B) enhancement [7,8] but this is somewhat arbitrary depending on the particular sample and ion implantation. As we show later a much more rigorous approach is to measure the irreversibility line

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before and after ion implantation and quote J_c at reduced field $b = B_{applied} / B_{irr}$.

We have initiated a programme of ion implantation in films of various thicknesses, smoothnesses and substrate materials. Comparisons are made between J_c and T_c for each film, pre- and post-implantation and the effect of rapid thermal annealing in oxygen and nitrogen.

2. Experimental Method

Thin YBCO films were implanted with deuterium at an energy of 50 keV and at fluencies of 1 x 10^{16} (high dose) and 1 x 10^{12} (low dose)/cm^2. Four films were implanted: NPL1b and NPL2 on $LaAlO_3$ (initial T_{ci} = 83K, 92K respectively), CAM3 on MgO (T_{ci}= 78K) and CAM4 on $SrTiO_3$ (T_{ci}= 81K). Each film was broken into two pieces for the implantation of low and high doses. The distribution of the deuterium through the thickness of the sample and substrate was measured using Secondary Ion Mass Spectrometry (SIMS) for the high dose case. The distribution maximum was found to lie in the region of the substrate. This process was repeated for $^{16}O^+$ ion at an energy of 200 keV and fluence of 1 x 10^{12} (low dose)/cm^2, implanted on three different films: NPL1a (initial T_{ci}= 82K) grown on $LaAlO_3$, CAM1 (T_{ci}= 78K) on MgO and CAM2 (T_{ci} = 69K) on $SrTiO_3$.

T_c was measured inductively and J_c was calculated from M - H loops using a VSM (Oxford Instrument 3001). In order to use the Bean formula correctly [9], a non destructive technique was employed which gives independent estimates of the length scale on which the screening current flows, and the sheet critical current density k_c=$J_c t$ (6), where t is the sample thickness. The length scale technique is based on a susceptibility argument. For samples with large aspect ratios (resulting in large demagnetisation factors when H is applied parallel to the c-axis), the initial slope during field reversal is equivalent to a differential susceptibility χ', where χ' α Λ/t. J_c was determined at three temperatures: $0.9T_c$, $0.3T_c$ and 10K. Unless otherwise stated J_c was measured from the remnant magnetisation from a 0.1T loop (which is well above the critical state penetration field). Also x - ray diffraction two theta measurements were taken to look at the crystal structure.

Where possible the samples were again broken in two for rapid thermal annealing (RTA) in oxygen and nitrogen (at 800 °C for twenty seconds). The samples were then characterised as before.

3. Results

For the low dose deuterium implantation T_c increased by 1 - 3K in the lower T_c films. An example of the T_c enhancement is shown in fig. 1 for the NPL1b film. The length scales decreased by between 1.4 - 5 times and the calculated $J_c(0)$ proportional to M/Λ, increased by between 1.1 - 1.7 times. Since other groups use the sample dimension D, to calculate J_c from the Bean formulae, fig.2 shows M/D and fig.3 M/Λ, for sample CAM3 (MgO substrate) at 10K, (a) for the unimplanted piece (T_{ci} = 78K) and (b) for the implanted piece (T_c = 79K). In this case although the piece broken off for implantation was twice as long as the unimplanted piece, its effective sample size at 0.1T and 10K from the length scale analysis was only half the unimplanted value.

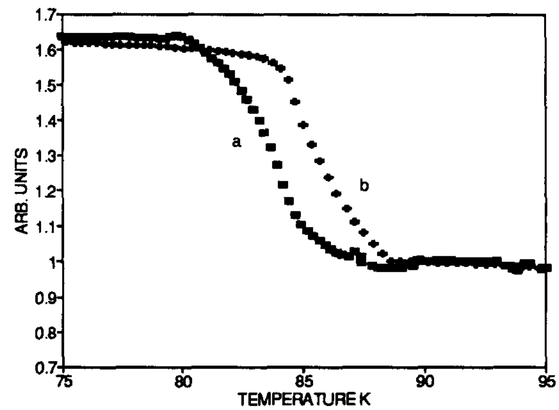

Fig. 1 Inductive transition of NPL1b film for (a) unimplanted and (b) $^2H^+$ low dose implanted.

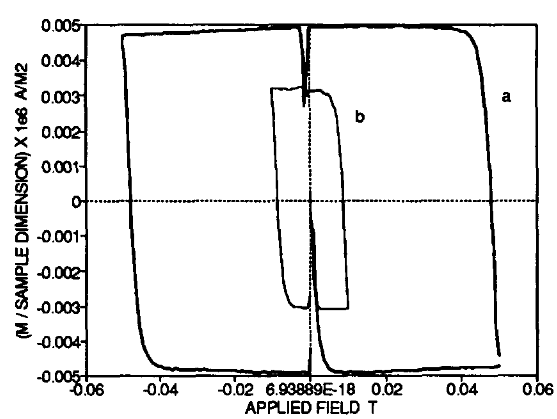

Fig. 2 Variations of the magnetisation/sample dimension, with applied field B(T) at 10K for (a) unimplanted (b) $^2H^+$ low dose implanted CAM3.

The T_c of the higher T_{ci} film (T_{ci} = 92K), was not changed by implantation of deuterium at the lower dose and M/Λ remained the same at 10K, but at 30K there is evidence for a deterioration of superconducting properties. ie a relative decrease in $J_c(0)$. Although this was a better film, as reflected in its narrow inductive transition width of 0.5K at 92K, fig. 6 shows that this film has been degraded after implantation, under the present conditions.

After the rapid thermal anneal in oxygen, the original T_{ci} value was not usually recovered. The value was either less than the initial T_{ci} or the superconducting transition disappeared completely. When annealing in nitrogen, the superconductivity was destroyed in all cases. The high dose deuterium implantation also removed the superconducting transition but it was recovered after annealing in oxygen with a lower value than T_{ci}.

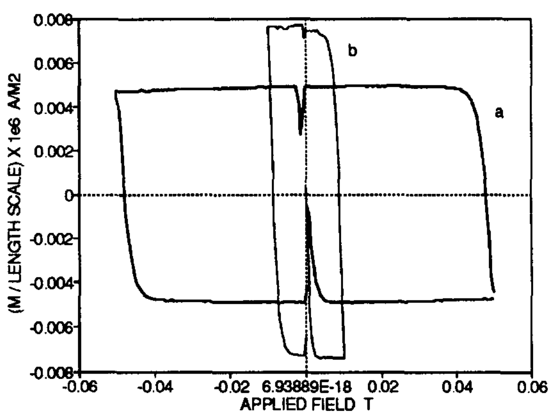

Fig. 3 Variation of the (magnetisation/length scale of screening current) with applied field B(T) at 10K for (a) unimplanted (b) $^2H^+$ low dose implanted CAM3 film.

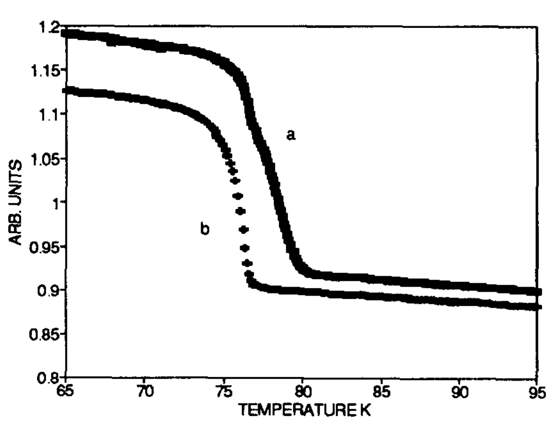

Fig. 4 Inductive transition of CAM1 for (a) unimplanted (b) $^{16}O^+$ implanted films.

Summarising our results for the $^{16}O^+$ implantation. After the low dose implantation, T_c either remained constant or decreased (see fig. 4). The length scale dropped by a factor between 0.3 - 0.8 and $J_c(0)$ increased by a factor between 1.5 - 2.3. That is to say that M/Λ increased significantly in some cases. Figure 5 shows M/Λ (a) before (T_{ci} = 78K) and (b) after (T_c= 76K) ion implantation on CAM1 (MgO substrate) at 20K.

Figure 6 shows the irreversibility line for NPL2 (T_c = 92K) (a) before and (b) after implantation. Although at 10K M/Λ was not altered, the irreversibility line was depressed after implantation of $^2H^+$ ions (low dose).

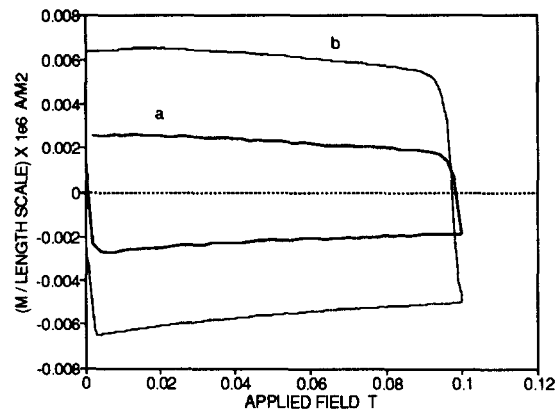

Fig. 5 Variation of (magnetisation/length scale of screening current) with applied field B(T) for CAM1 at 20K (a) unimplanted (b) $^{16}O^+$ implanted piece.

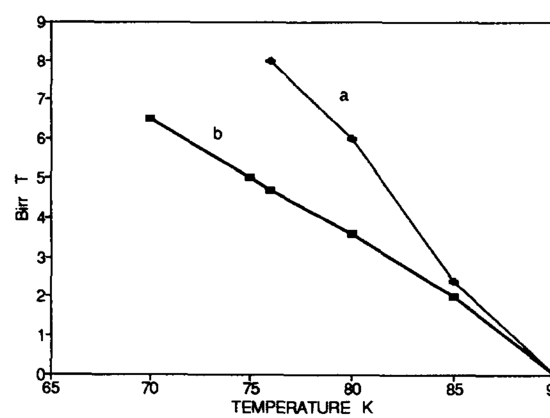

Fig. 6 Irreversibility line for NPL2 film (a) unimplanted (b) implanted.

4. Discussion

In all cases the length scale decreased after implantation. In some cases this is related to the fact that the implanted piece of film was smaller than the unimplanted one, but more usually the implantation creates some granularity. The beam energies employed in this study would probably only leave the highest quality films, which do not have any extended defects, undamaged. The broadness of the inductive transition indicates that our sample do not fall into this category (fig. 1).

Although the magnetisation critical current is enhanced, the transport current would in all probability be degraded because the weak links have been created in the film. Under these conditions, the irreversibility line would be a good measure of the enhanced intragrain pinning. For a film showing J_c enhancement (depression) where there is a significant increase (decrease) in the B_{irr} line (fig. 6), to quote J_c enhancement at an arbitrary field is misleading. Obviously the J_c enhancement will be particularly large if the field and temperature are chosen close to the irreversibility line for the unimplanted film. A much more consistent system is to quote the enhancement of J_c at a normalised field $b = B_{applied}/B_{irr}$

The most striking result is that of the increase in T_c. This only occurs for the deuterium implantation. There appear to be two possible explanations. Enough hydrogen may stay in the film to have an effect on the electronic band structure as reported by Yokota. But from initial calculations from the SIMS data, the density of deuterium in the film is very small, compared with the volume of a unit cell of YBCO. In future implantations, the number of ions coming to a rest in the film will be varied by increasing the beam energy, or by making the 'channelling' of the particles less significant [10].

The other possibility is that the deuterium may disturb the oxygen atoms as they pass through the film, knocking them into different positions. This argument is strengthened by examining the critical current density which is not increased significantly (10 - 25%), in the films where T_c is greatly enhanced. It has been suggested that specific oxygen vacancy sites could act as pinning centres. If these are filled in the process of the deuterium ions passing through the film, it might be expected that a decrease in J_c would result. However the ions are also creating some local damage enhancing the pinning and increasing J_c. To observe a small overall J_c in association with T_c enhancement, might be anticipated as a result of the competition between these two processes. The recovery of T_c in some cases by oxygen annealing suggests that the high doses of deuterium are disturbing the oxygen atoms in the film somehow.

The low dose of $^{16}O^+$ at the same fluency as the deuterium leaves T_c unaltered or suppressed. The suprěssed T_c case yielded the largest J_c enhancement. There could be several reasons why this behaviour is different to the deuterium implantation. For example the size of the ion and the fact that ^{16}O naturally resides in the crystal structure.

5. Conclusion

From our preliminary study we have illustrated the importance of using the length scale technique to determine J_c from magnetisation experiments. We have also shown that using a reduced field when quoting the enhancement of J_c at field is not only more rigorous but would facilitate comparison of results between different studies. Taking the length scale into account the best $J_c(0)$ enhancement (of a factor of 2.3 at 20K), was achieved with 200keV $^{16}O^+$ ions at a fluency of 1 x $10^{12}/cm^2$. We draw attention to the trend that the T_c enhancement may be related to a process whereby key oxygen sites which contribute to the pinning, have been removed, competing with the process of increased pinning through local ion damage and resulting in only small J_c enhancement. In films where T_c is suppressed, a larger J_c enhancement is achieved.

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10 Channelling is a phenomenon whereby the deuterium may ricochet through the crystal lattice without having the effect one might statistically expect, as the target cross-section is reduced. A tilt of about seven degrees away from the perpendicular is used in RBS to take this into account.