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Ion beam and thermally-induced interface reaction between high T_c superconductor thin film and metal overlayer

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Abstract. Ion beam and thermally-induced interface reactions between high T_c superconducting thin film of $Y_1Ba_2Cu_3O_{7-x}$ and metal overlayer of Ag are studied with a view to control the interfacial property of contact resistance. The interface reaction is induced by 100 keV Ar^+ ion beam with different ion dose values ranging from 5×10^{13} to 3×10^{14} ions/cm^2. The YBaCuO film-metal interface is characterized by using the small angle XRD to study the structural properties of the interfacial phases. The electrical property of the interface, specifically contact resistance, has been investigated for different dose values and thermal treatments. Three-probe vs four-probe configuration has been adopted to measure the contact resistance.

Keywords. High T_c superconducting YBaCuO film; contact resistance; ion beam mixing.

1. Introduction

The earliest applications of high T_c superconductors are expected to arise in their thin film form. Films capable of carrying current densities above a million Amps/cm^2 at 77 K have been fabricated by different routes. When such films are to be used in applications they will generally have to be connected with external circuits and in this context the properties of superconductor-metal contacts become extremely important (Cava *et al* 1987; Van der Mass *et al* 1987; Tzeng *et al* 1988; Ekin *et al* 1988; Jia and Anderson 1989; Katz *et al* 1989). If a patterned metallization on the superconductor is thermally annealed in vacuum at a sufficiently high temperature for interface reaction to occur then the superconductor is depleted of oxygen. If annealed in oxygen atmosphere, many types of metallizations can degrade. Ekin *et al* (1988) have showed that the contact of In or Al to YBaCuO makes the superconductor surface form non-stoichiometric oxides even at a temperature below 150°C. Thus it is desirable to have low-temperature processing for the improvement in contact properties. Ion beam-induced atomic mixing is one such method which has been well tried in semi-conducting technology (Chiang *et al* 1981). In this work we have shown that ion beam mixing followed by low temperature annealing improves the electrical quality of $Ag/Y_1Ba_2Cu_3O_{7-x}$ interface.

2. Experimental

Thin films of oxide superconductor $Y_1Ba_2Cu_3O_{7-x}$ were deposited by laser beam ablation process using a KrF excimer laser (Lamda Physik EMG200E) producing 1 J energy per pulse of 20 ns duration at $\lambda = 248$ nm. A sintered superconducting pellet with nominal composition of $Y_1Ba_2Cu_3O_{7-x}$, transition temperature of 92 K and the transition width of about 1 K was used as a target. The thin films were deposited on single crystal ZrO_2 (001) substrates. The process parameters such as substrate temperature, laser beam energy density, oxygen partial pressure etc. were optimized as reported elsewhere (Bendre *et al* 1990), so as to obtain good quality thin films.

The silver metal overlayer was deposited in Varian UHV chamber rendering a base pressure of 1×10^{-8} torr. The thin films were appropriately masked by SS strips (2×5 mm dia) to obtain contact pads. To prevent thermal damage to the YBaCuO film during evaporation of the contact metal, a distance of 16 mm from source to substrates was kept during evaporation. The substrate temperature was raised to nearly 100°C, during evaporation, so as to improve the adhesion of the metal overlayer with the YBaCuO film. The evaporation rate was controlled to 2–3 Å/s and the thickness was monitored by a quartz thickness monitor. Typically, metal overlayer of 400 Å thickness was deposited by this method.

The patterned YBaCuO film was irradiated at room temperature by Ar^+ ions using an ion implantation machine indigenously developed in our laboratory. The energy of the ion beam was chosen (100 keV) such that the projected range of the Ar^+ ions reaches the Ag/YBaCuO interface (with straggled range around 300 Å). The samples were loaded for implantation with the SS mask, so that effective beam mixing take place only at the Ag/YBaCuO interface. The ion current density at the target was kept at $0.5\,\mu A/cm^2$ to avoid overheating of the samples. Since the amorphization threshold (White *et al* 1988) in the case of Ar^+ for YBaCuO films is observed at $\sim 10^{15}$ ions/cm^2, the fluence values in this experiment were chosen to be in the range of 10^{13}–10^{14} ions/cm^2 which caused no major damage to T_c or ΔT_c. Of course, the YBaCuO film was masked during implantation and the implantation effects occurred only below the Ag pads.

The processed films were characterized by low angle X-ray diffraction measurements (Rotaflex RU200 B) using CuK_α radiation employing Seeman-Bohlin geometry and the results were obtained for grazing angle values from 0.2° to 2.0° (results shown only for $\alpha = 0.6°$). Lim *et al* (1987) have discussed the X-ray penetration depth in a sample depending on the angle of incidence and critical angle. In our case, the grazing angle of incidence of 0.6° (critical angle for silver = 0.46°) was selected to probe the Ag/YBaCuO interface. The electrical measurements were carried out by employing the three-probe configuration (Khare *et al* 1989) shown in figure 1, so as to obtain the contact resistance. Four-probe measurement was also made to obtain the precise R vs T data above T_c onset.

3. Results and discussion

The resistance (three probe) vs temperature plot of the YBaCuO film with Ag overlayer for various ion beam dose values is shown in figure 2. The variation of three-probe resistance ($R(3P)$) with temperature shows a similar behaviour as in the case of four-probe configuration ($R(4P)$) but with a higher resistance above the onset temperature. Although the three-probe configuration cannot provide an accurate value of the contact resistance R_c, they can qualitatively indicate if the contacts are ohmic. Non-ohmic contacts tend to increase R_c as temperature is decreased (Mizushima *et al* 1988). The contact resistance measured using this configuration is the algebraic addition of metal resistance (for Ag resistivity = 1.59 μohms/cm at 20°C), the Ag/YBaCuO interfacial resistance and the contribution due to the intergranular weak links just below the contact pads. In figure 2, it is observed that the room temperature resistance

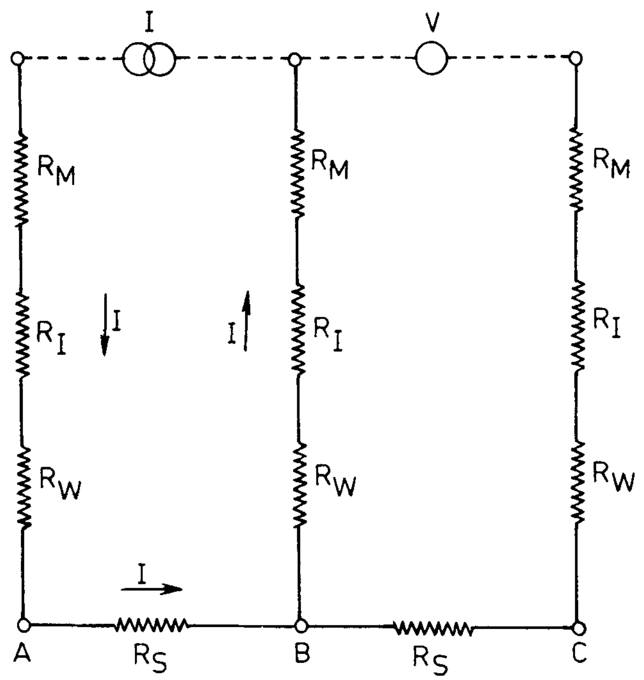

Figure 1. Equivalent circuit configuration of contact resistance measurement. R_M: Contact metal resistance; R_I: YBaCuO and metal interfacial resistance; R_W: Resistance contribution due to intergranular weak links just below the contact pad. $R_c = R_M + R_I + R_W$.

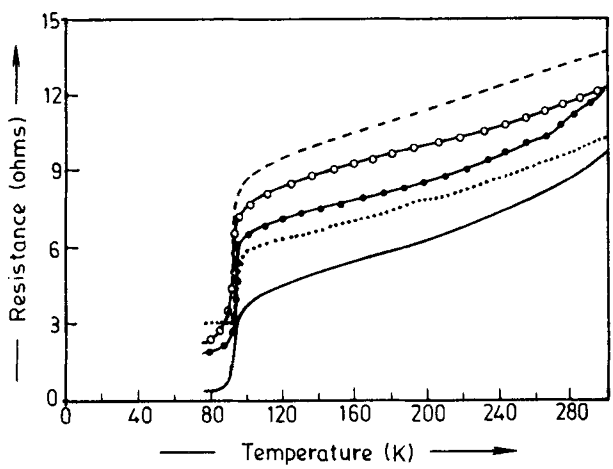

Figure 2. Three-terminal resistances vs temperature plot of YBaCuO film on ZrO_2 (001): (....) As is with Ag overlayer; After implantation of dose values: (–●–●–●–●–) 1.25 × 10^{14} ions/cm^2; (–○–○–○–) 1.75 × 10^{14} ions/cm^2; (------) 3.0 × 10^{14} ions/cm^2; (———) 3.0 × 10^{14} ions/cm^2 and oxygen-annealed at 250°C for 15 min.

increases for increasing dose values. This can be attributed to the implantation-induced damage. The residual resistance at 77 K, essentially the contact resistance, is observed to vary significantly with the ion dose values. This feature is brought out in figure 3. It is clearly seen that the implantation with the initial dose value of 1×10^{14} ions/cm^2 reduces the contact resistance by 42%. The physical interference of the energetic ions at the interface can lead to the mixing of Ag with YBaCuO at the atomic level. In the context of metallization, it has already been shown (Ekin *et al* 1988) that the diffusion of the contact metal in the sample film can contribute in lowering the contact resistance. With the increase in the ion dose value to 1.75–3.00×10^{14} ions/cm^2, the contact resistance is found to increase by 16–18%. To anneal the defects created by implantation process, the films were subsequently subjected to thermal treatment at

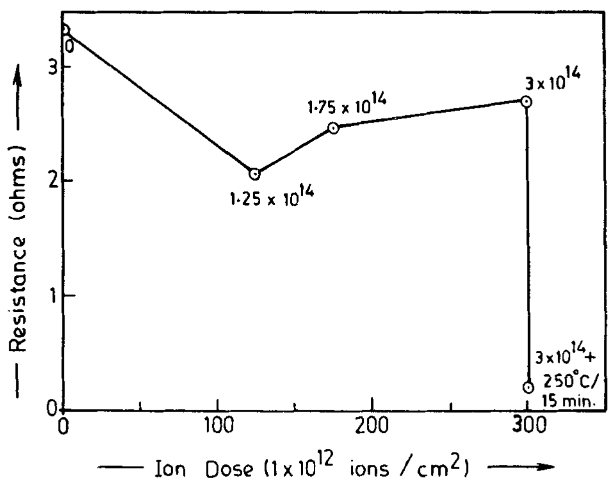

Figure 3. Variation of contact resistance with ion dose values.

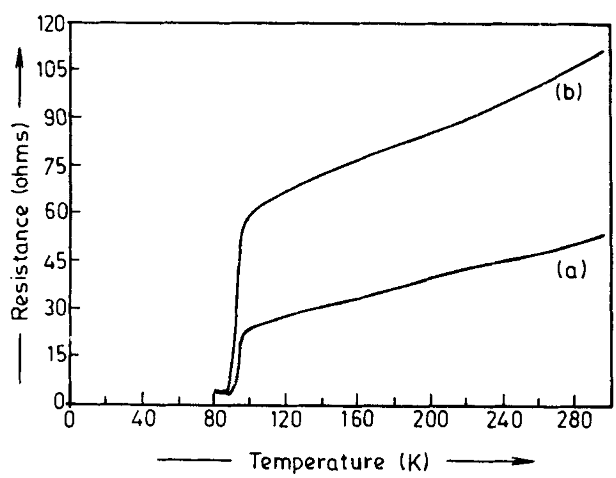

Figure 4. Plot of three-terminal resistance vs temperature of (**a**) As is YBaCuO film with Ag overlayer; (**b**) after thermal annealing in O_2 atmosphere for 250°C for 15 min.

250°C for 15 min. Interestingly, this process reduced the contact resistance by about 92%. In order to confirm the role played by the ion beam mixing in the reduction of R_c, freshly-prepared samples with Ag overlayer were annealed in vacuum (base pressure better than 1×10^{-6} torr) as well as in O_2 atmosphere at 250°C for 15 min. The variation of $R(3P)$ with the temperature for these cases is shown in figures 4 and 5 respectively. In the case of vacuum annealing, the depletion of oxygen even at such low temperature is clearly reflected by the increase in room temperature resistance as well as degrading the transition. On the other hand, in the case of samples annealed in oxygen atmosphere, it is seen that the residual resistance is unaffected while there is a significant increase in the room temperature resistance. This clearly demonstrates the role played by the ion beam mixing at Ag/YBaCuO interface.

In figure 6 are shown the low-angle XRD results for three cases of interest. Figure 6a gives the X-ray data for the superconductor film and it exhibits significant *c*-axis orientation normal to the film surface. Figure 6b shows the data for the as-deposited Ag overlayer on the superconducting film (dashed line) and the sample after ion beam mixing at a dose of 3×10^{14} ions/cm^2. Clearly, after ion beam mixing at this low dose there is no significant change in the basic character of the spectrum, though the Ag film densifies to yield higher line intensities. After annealing, (figure 6c) the character of the spectrum is still the same though the Ag intensities are lowered probably by defect-assisted diffusion across the interface. These results bring out that mixing at this low dose which leads to the observed electrical results is primarily an interface-related effect with minimal bulk mixing.

In conclusion, it has been brought out that the low temperature processing method of ion beam mixing can serve as an important method to improve the quality of metal-superconductor contacts.

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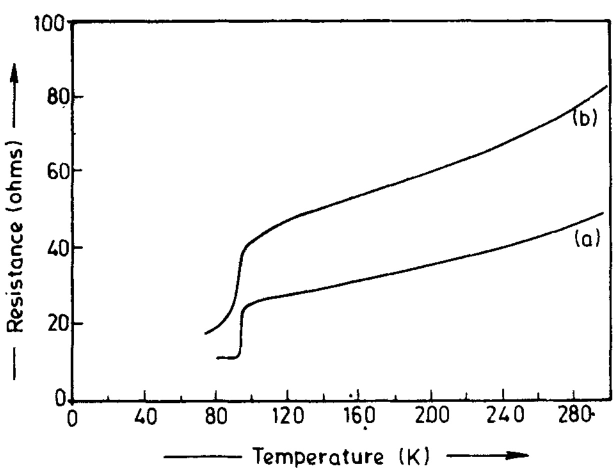

Figure 5. Three-terminal resistance vs temperature graph of (**a**) As is YBaCuO film with Ag overlayer; (**b**) after vacuum annealing at 250°C for 15 min.

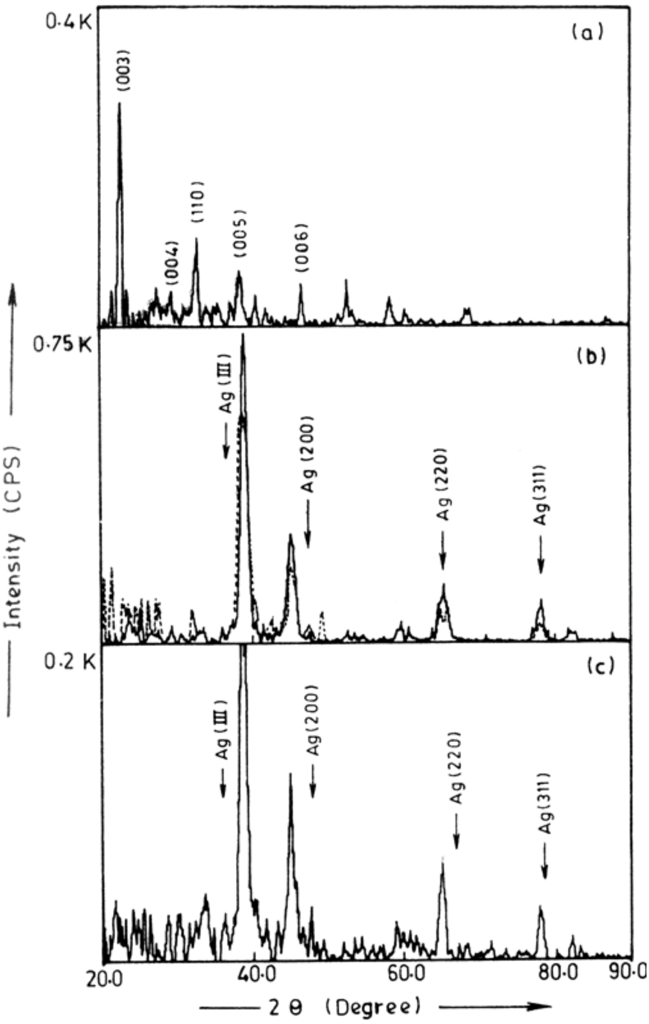

Figure 6. Small angle XRD patterns of (**a**) YBaCuO film on ZrO_2 showing preferential orientation along *c*-axis. (**b**) YBaCuO film with Ag overlayer, as it is (dashed line); YBaCuO film with Ag overlayer and implanting a dose value of 3.0×10^{14} ions/cm^2 (solid line). (**c**) YBaCuO film with Ag overlayer and implanting a dose value of 3.0×10^{14} ions/cm^2 and oxygen annealed at 250° for 15 min.

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