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Application of MeV Ions to the Analysis and Modification of High Temperature Superconducting Materials

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November 2, 1990

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Subject: Final Report of activities under subcontract B108466 (under DOE Contract W-7405-ENG-48), "Application of MeV Ions to the Analysis and Modification of High Temperature Superconducting Materials"

I. Modification

In analyzing the composition of high T_c superconducting materials using heavy-ion RBS we discovered that the electronic excitation of the target caused mixing to occur. To study this effect we made superlattice structures of alternating thin layers of $Ba_2YCu_3O_{6+x}$ and BaF_2 - as shown by the RBS spectrum in Figure 1. At this point in the process some mixing has occurred, but the individual layers are still visible. However, as the bombardment continued the layers were mixed together - as shown by the RBS spectrum in Figure 2. It may well be that one can use this phenomenon to produce high T_c materials with a wide range of compositions.

We have, however, tried to determine if the ion-beam-induced damage could also be used to modify the properties of the materials in a controlled way. Thus, we have used fission fragment irradiation of Bi-Sr-Ca-Cu-O crystals to determine whether we could modify the transition temperature, resistivity vs temperature, and flux pinning behavior. For these studies we obtained crystalline material from a collaborator at Caltech (R. M. Housley). We made a careful set of measurements, then wrapped the sample in a ^{235}U foil and exposed it to thermal neutrons generated with an Am-Be source. At the fluence used, approximately 10^7 fission fragments/cm² (~ 100 MeV at 100 amu), there were no changes

FIGURE 1

Figure 1: Unannealed $\text{Ba}_2\text{YCu}_3\text{O}_{6+x}$, deposited in layers, prior to ion beam mixing and analysis with the surface barrier detector and 25 MeV Cl ions. The sample, produced by electron beam deposition, was 9 repetitions of Y(200 Å)/BaF₂(750 Å)/Cu(220 Å) on a Si substrate. The dose was $Q = 1.5 \times 10^{12}$ ions. Mixing and analysis were proceeding simultaneously.

FIGURE 2

Figure 2: Unannealed $\text{Ba}_2\text{YCu}_3\text{O}_{6+x}$, deposited in layers, after ion beam mixing. Layers are no longer visible. Previous to this run the total dose was $Q = 2.7 \times 10^{15}$ ions. The run itself put 2.25×10^{15} ions into the sample, in a beam spot $\sim 1 \text{ mm}^2$.

in the superconducting properties. We then attempted to chemically etch the damage tracks left by the irradiation; this showed that the material had so many grain boundaries that its behavior was dominated by this - not the irradiation. Subsequently, we obtained a small ($\sim 1 \text{ mm}^2$) single-crystal sample from IBM. In this case we only looked for the latent track damage by making thin sections for TEM analysis. In the irradiated areas there were indeed the expected damaged regions. We are now waiting for larger samples so that we can make contacts for electrical measurements, followed by another attempt at track etching.

These attempts indicate that high-quality, single-crystal samples are needed to make unambiguous determinations of flux pinning to damage tracks from MeV ions. We hope to have such material eventually, but it is not available to us now.

II. Analysis

In our preliminary measurements of the detailed composition of high T_c samples we employed RBS with MeV heavy ion beams (usually ^{16}O). This initial work used surface-barrier detectors; although the kinematics of the scattering gave good separation of the elements, the detector resolution was not good enough for studying any variation in composition with depth. This is important for these cases because we had found other components (e.g. CuO crystals) to be present at the sample surface or when we were studying the evolution of the material under ion irradiation. Typical cases are shown in Figures 3 and 4.

To improve the depth resolution of the technique we built a time-of-flight (TOF) detector for such RBS studies. In this device the scattered ions pass through two thin carbon foils that are a known distance apart. The secondary electrons generated by the ions in the foils are accelerated and detected in

FIGURE 3(a)

Figure 3: (a) $\text{Ba}_2\text{YCu}_3\text{O}_{6+x}$, sample #111488A, produced by Ar beam sputtering and an O beam on target with the substrate held at 600°C . The annealing was for 11 hours at 450°C . A 16 MeV beam of O was used with the surface barrier detector because of the good resolution of that beam with that detector. The layer is $1760\text{ \AA}$ of $\text{Ba}_{2.07}\text{Y}_{.949}\text{Cu}_3\text{O}_{6.5}$. The substrate is MgO. An .05% impurity of either tantalum or tungsten is visible to the right of barium feature, apparently inadvertently deposited during deposition. This region of the spectrum is shown in detail in Figure 3(b).

FIGURE 3(b)

FIGURE 4(a)

Figure 4: (a) Annealed $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, sputter deposited, sample #031088A. A 16 MeV beam of O^{2+} was used with the surface barrier detector. The sample was a 900 Å layer of $\text{Bi}_2\text{Sr}_{1.68}\text{Ca}_{1.32}\text{Cu}_2\text{O}_8$ on Mg). Included in the analysis was 2% of Ca_2CuO_3 chunks observed on the surface. The diffusion of the Cu into the substrate is modeled as a 600 Å 3.5% layer. An expanded view of the Ca, Cu, and Sr peaks are shown in Figure 4(b).

FIGURE 4(b)

channel plates. By measuring the time difference between the arrival of the ion at each foil we can determine its velocity - and hence its energy (see Figure 5). We have shown that we could get a 0.75% energy resolution for the scattered oxygen ions, which is already a vast improvement over surface barrier detectors (see Figure 6). However, for the work to be done under this subcontract we wanted still better resolution - equivalent to $10\text{-}15\text{ }^{\circ}\text{A}$ in the near-surface region of the sample (see Figure 7).

By studying carefully the behavior of the TOF system at different scattered ion energies we determined that the largest contribution to the resolution was from variations in ion path between the detectors ($\sim 3.5\text{ mm}$ out of 83 cm) (see Figure 8). Correcting this would give an immediate factor-of-two improvement in energy resolution. Under this subcontract we have redesigned the first detector in the system and it has now been constructed. We should be able to test it this month (November 1990).

The original detector configuration was virtually the entire source of path variation. It has now been redesigned so that the two carbon foils are parallel; secondary electrons are bent through 180° from the ion's path through a very uniform magnetic field (isochronous transport) to the channel plate (see Figure 9). We found a very simple and inexpensive way to construct the magnet using a collection of small cylindrical magnets from a toy company (see Figure 10).

III. Future Plans

It is our intention to submit to you a proposal for the continuation of this work. This would consist of the use of the TOF-RBS system to study other materials that were relevant to your program - especially in the form of thin

FIGURE 5

Figure 5: the TOF system shown schematically here has been used to measure RBS spectra with ^{16}O and ^{35}Cl beams of up to 25 MeV primary energy from the EN Tandem Van de Graaff at Caltech. The primary beam crosses a large (60 cm) vacuum chamber and hits the target in a smaller (30 cm) scattering chamber. Particles backscattered at 175° pass through a first secondary electron time pick-off system placed 10 cm away from the target and are stopped in a second detection system back in the large chamber. The flight path can be varied from 40 to 83 cm, which changes the detector geometrical solid angle from 1.3 to 0.36msr. The pulses from the secondary electron devices (SED) are shaped in timing amplifiers (TA), pass through a constant fraction discriminator and trigger a time-to-amplitude converter (TAC). In order to avoid TAC starts without a stop the physical start signal is delayed and "start" and "stop" of the TAC are reversed.

FIGURE 6

Figure 6: The measured energy resolutions for scattered ^{35}Cl (two flight path distances) and ^{16}O ions of different energies detected by our TOF system.

FIGURE 7

Figure 7: 25 MeV ^{35}Cl beam backscattered from a ten layer $\text{In}_{0.18}\text{Ga}_{0.82}\text{As}$ (80 Å)/GaAs (200 Å) superlattice on GaAs(Si). The overlaid Gaussian at the low end of the spectrum represents the energy straggling predicted by the Bohr formula. The Gaussian at the high energy and shows the detector resolution (the flight path was 83 cm). The inset shows the Indium part of the same spectrum taken with a silicon surface barrier detector.

FIGURE 8

Figure 8: Calculated energy resolution versus energy for ^{35}Cl ions including contributions from time resolution (200 ps), flight path differences ($\Delta L/L = 0.3\%$ at $L = 830$ mm), and energy straggling and nonuniformity of the carbon foil of SED1. Points are measured values.

Figure 9 Anode, Faraday Cage, and Magnet Box

Figure 10: B_z in Gauss (7 cm faceplates with 20 magnets) measured for the magnet structure shown in Figure 9.

superlattice structures. The factor of two improvement in energy/depth resolution will allow a 10-15 Å depth resolution in the near surface region of the target material.

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