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*Title:* MICROSTRUCTURE AND PROPERTIES OF in-situ RF SPUTTERED $YBa_3CU_3O_7$ THIN FILMS FOR MICROWAVE APPLICATIONS

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# MICROSTRUCTURE AND PROPERTIES OF IN-SITU R.F. SPUTTERED $YBa_2Cu_3O_{7-x}$ THIN FILMS FOR MICROWAVE APPLICATIONS

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## ABSTRACT

The residual surface resitance of a number of films of $YBa_2Cu_3O_7$, prepared by off-axis sputtering onto MgO substrates, has been measured using a parallel-plate resonator technique. Deposition conditions were kept constant, apart from the substrate temperature. There is no correlation between surface resistance and other important microscopic parameters, such as $T_c$ and c-axis lattice parameter. There is, however, a trend to higher $R_s$ with increasing volume fraction of in-plane misoriented material, although the correlation is not perfect. Furthermore, we have found that most of the misoriented material is localized at the film substrate interface and therefore is probably not responsible for most of the RF losses. The data suggest that at higher deposition temperatures, there is an increasing tendency for $45^o$-misoriented material to appear in the films, and it may be that a significant fraction of this material is present closer to the free film surface. STM qualitatively supports this conclusion.

## INTRODUCTION

The development of microwave technology has been one of the areas of focus forhigh temperature superconductor (HTS) applications. The early stages of research have emphasized passive microwave components such as filters, resonators, and delay lines. The performance of these devices depends on the surface resistance of the material and lower loss results in superior components. Surface resistance has a very complex origin, even in the case of traditional superconductors. At temperatures low compared with $T_c$, microstructural deviations from perfect crystallinity, rather than any intrinsic losses associated with the material, will determine the residual surface resistance. This is clearly the case in HTS cuprates, where second phases, or material that is not crystallographically well aligned, are known to increase the surface resistance. Studies of c-axis oriented sputtered films have suggested [1] that the microwave loss is proportional to the amount of material rotated $45^o$ around the c-axis. The results reported here were obtained on films deposited using the same technique, under similar conditions, and examine this suggestion further.

## EXPERIMENTAL TECHNIQUES

The films were deposited using off axis RF magnetron sputtering [2] from a 2" diameter stoichiometric target of $YBa_2Cu_3O_7$. The substrates (1 $cm^2$ (100) MgO) were used as received from the vendor [3] and were fastened to the heater block using a silver-containing cement. The deposition parameters are given in the table below. Temperature was monitored using an IR pyrometer reading on the substrate heater block.

The microwave surface resistance was measured with a parallel plate resonator technique [4]. The method uses a thin dielectric (0.5-mil-thick teflon) sandwiched between two superconducting films. This parallel plate structure has a very low impedance and, because the high impedance outside of the resonator, the edge of the structure effectively has an open boundary condition. The radiation Q of this structure is high (~$10^5$) and the surface area to volume ratio is large, leading to a sensitivity of a few microohms. The samples must be used in pairs and in this study we used single films paired with a niobium reference. Measurements were made at 4K and approximately 10 GHz. At this temperature all of the measured surface resistances should correspond to residual values. All of the films were completely c-axis perpendicular to the limit of detection of the diffractometer. Lattice parameters were obtained from the (007) reflection. In-plane orientation was obtained by comparing the relative intensities of the off-axis {012} reflection in successive $45^0$ increments in an azimuthal (chi-) scan. Critical temperatures were obtained by measurement of the reactive impedance of a coil inductively coupled to the film, operated at a frequency of 10 kHz.

Table I
Deposition Parameters and Film Properties

| FILM I.D. | Pow er (W) | Deposit ion T (C) | Oxygen (mtorr) | Argon (mtorr) | Thicknes s (nm) | c-axis (nm) | %45 | Tc (K) | Rs mW |
|---|---|---|---|---|---|---|---|---|---|
| 4/5/91-s1 | 50 | 708 | 60 | 40 | 182 | 1.1703 | 3.35 | 85 | 195 |
| 6/10/91-s1 | 100 | 706 | 40 | 110 | 499 | 1.1710 | 1.03 | 80 | 126 |
| 6/18/91-s1 | 100 | 705 | 40 | 110 | 773 | 1.1773 | 0.68 | 72 | 110 |
| 7/10/91-s1 | 100 | 726 | 40 | 110 | 522 | 1.1719 | 4.9 | 85 | 1650 |
| 7/11/91-s1 | 100 | 715 | 40 | 110 | 447 | 1.1712 | 1.9 | 81.5 | 772 |
| 7/12/91-s1 | 100 | 686 | 40 | 110 | 387 | 1.1751 | 0.94 | 79 | 53 |
| 7/19/91-s1 | 100 | 705 | 40 | 110 | 350 | 1.1700 | 2.3 | 84 | 172 |
| 7/25/91-s1 | 100 | 668 | 40 | 110 | 418 | 1.1760 | 0.97 | 77 | 61 |
| 7/26/91-s1 | 100 | 652 | 40 | 110 | 362 | 1.1800 | 1.54 | 70 | 73 |
| 8/6/91-s1 | 100 | 636 | 40 | 110 | 415 | 1.1850 | 1.38 | 56 | 338 |

## RESULTS AND DISCUSSION

Surface resistance is plotted against deposition temperature in Fig. 1. We obtain a broad minimum in $R_s$ at a deposition temperature of about $685^oC$. Deposited under these conditions the films have surface resistances that are slightly higher than the best values reported in the literature. Note, however, that pressure, power, etc., have not been optimized here.

When $T_c$ and c-axis lattice parameter are examined (Fig. 2) it is clear that the films are microscopically far from perfect. The highest $T_c$'s are obtained for films deposited with substrate temperatures above 700°C, whereas the lowest values of $R_s$ occur at significantly lower temperatures. The c-axis lattice parameter is longer than that found in stoichiometric bulk materials (1.168 nm). In agreement with others [1], therefore, we find no obvious correlation between microscopic disorder and surface resistance.

We have attempted to assess mesoscale disorder by x-ray texture analysis and STM studies of the surface topology of a large number of films. The surface resistance is plotted as a function of the volume fraction of 45°-misoriented material in Fig. 3. Although the trend in the plot suggests a connection between these quantities the correlation is quite weak, implying that other factors are also important.

Because the films have different thicknesses, we have measured of the amount of 45°-misoriented material as a function of thickness, both for the films shown in Table 1, and for an additional series of films deposited at a constant temperature, but for different lengths of time. All of these results are shown in Fig. 4. It is clear that there is a very strong dependence of the amount of misoriented material on thickness. In fact there is an excellent reciprocal relationship between these quantities. The simplest interpretation of this result is that most of the misoriented material lies in a very thin layer at the film/substrate interface. This is qualitatively supported by the STM studies. In the films with the lowest $R_s$ we were unable to find any grains of misoriented material at the surface. Such grains are normally apparent because of the polygonization which causes them to exhibit a square pyramidal growth morphology. In films deposited at higher temperatures grains rotated 45° are more common and easily visible.

The relationship shown in Fig. 4 allows us to normalize the volume % of misoriented material with respect to the variable film thickness. In Fig. 5 we therefore plot the product of volume fraction of misoriented material and film thickness vs the deposition temperature. We find that there is a broad minimum in this quantity and above about 700°C there is evidently an increased tendency to produce misoriented material.

The observation that most of the misoriented material lies at the interface with the substrate complicates the interpretation of the surface resistance results. In the thicker films (which are several London penetration depths thick), the misoriented material is in a region of little RF field. It should, therefore, not contribute significantly to the losses. It is possible, however, that films containing a large amount of 45° misoriented material at the film substrate interface also contain larger amounts throughout the depth of the film, leading to the loose correlation evident in Fig. 3. Alternatively, losses may be due to some other mesoscale disorder (e.g. low-angle grain boundaries [5]) which are also present, but not quantified by the present experiments. The situation is further complicated by the STM observation that surface grain size increases considerably as the film thickness increases [6].

## CONCLUSIONS

The principal finding of this study may be summarized as follows.

1. There is no obvious correlation between $R_s$ and microscopic materials properties such as $T_c$ or c-axis lattice parameter.
2. There is a loose correlation between $R_s$ and volume % of $45^o$-misoriented material.
3. X-ray data suggest that most of the misoriented material lies at the film/substrate interface and would therefore be relatively ineffective in leading to microwave losses in the experimental configuration used here.
4. Under the deposition conditions used here, the volume % of misroriented material is not a strong function of deposition temperature but shows some evidence of an increase above $700^oC$, which may be associated with the rapid increase in surface resistance observed in films deposited above this temperature.

In addition we know that the oxygen partial pressure plays a critical role in determining the orientation of grains. It is by no means certain that the dependence of $R_s$ on substrate temperature observed here is necessarily true under other deposition conditions.

## References

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Figure Captions

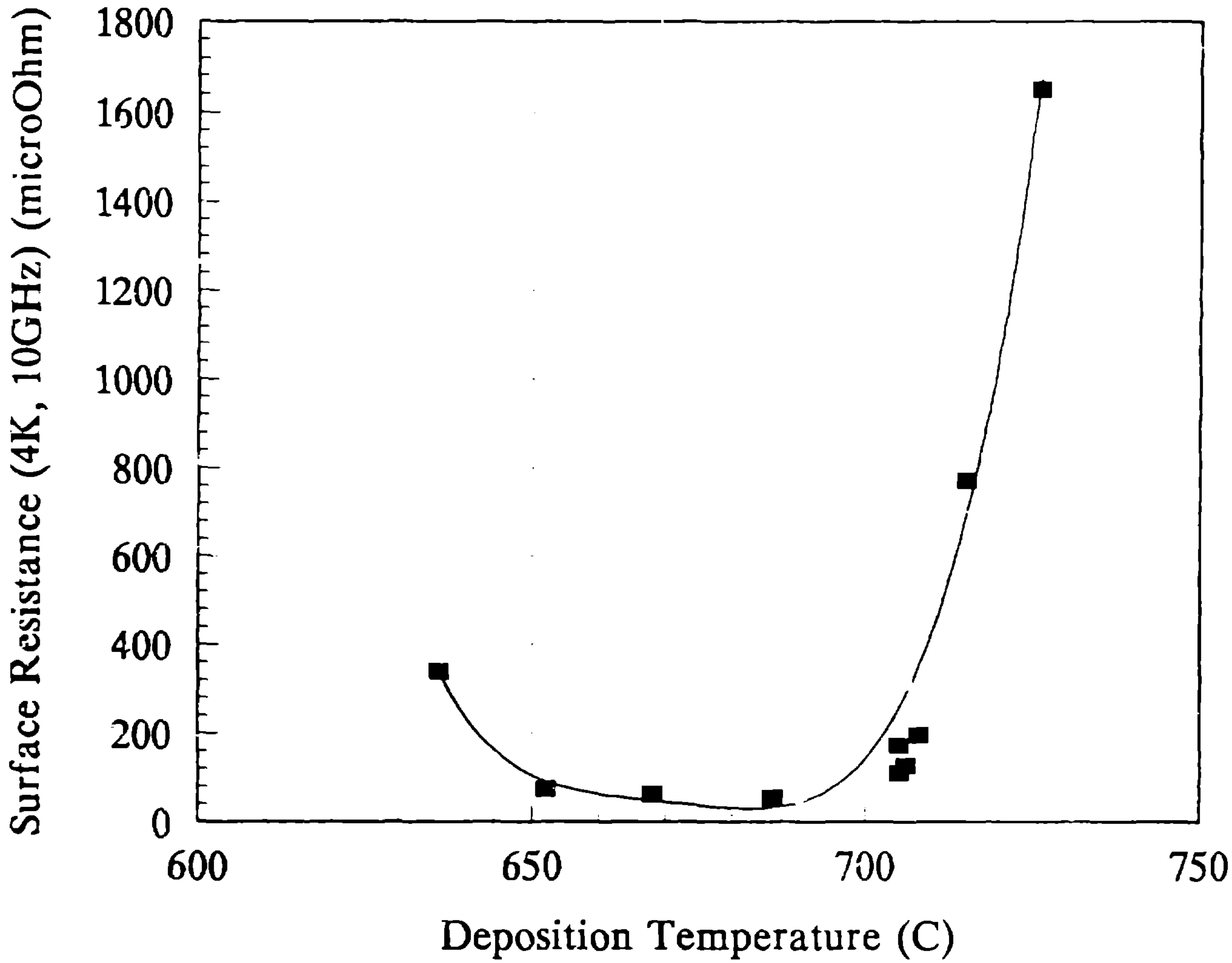
Fig. 1. The surface resistance measured at 10GHz and 4K plotted against deposition temperature.

Fig. 2. Critical temperature and c-axis lattice parameter plotted vs deposition temperature.

Fig. 3. The surface resistance plotted vs total amount of 45$^o$-rotated material.

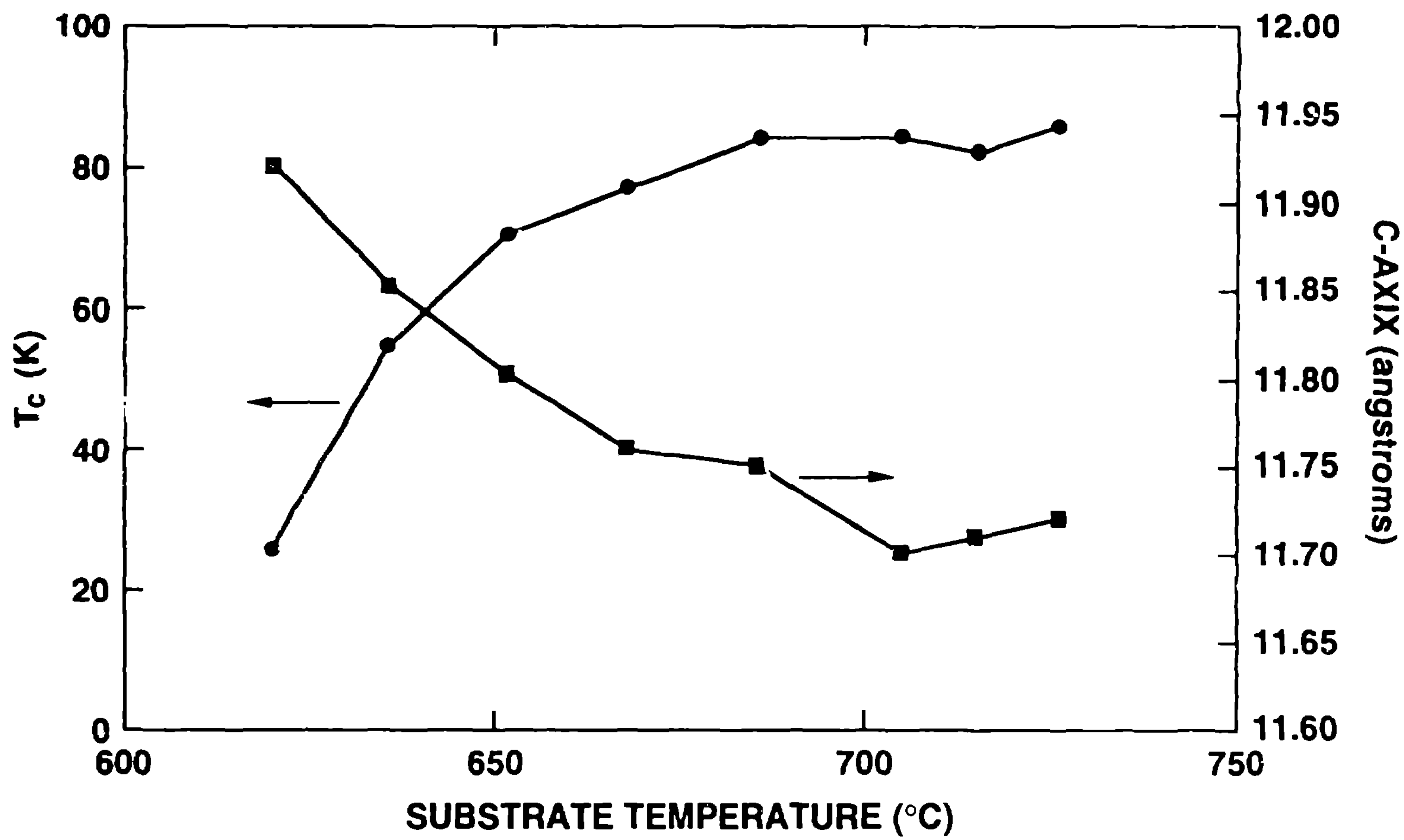
Fig. 4. The percent of 45$^o$-misoriented material vs film thickness.

Fig. 5. Product of volume % misoriented material and film thickness plotted against deposition temperature.



Surface Resistance (4K, 10GHz) (microOhm)
1800
1600
1400
1200
1000
800
600
400
200
0
600
650
700
750
Deposition Temperature (C)


Fig. 2


100
80
60
40
20
0
$T_c$ (K)
12.00
11.95
11.90
11.85
11.80
11.75
11.70
11.65
11.60
C-AXIX (angstroms)
600
650
700
750
SUBSTRATE TEMPERATURE (°C)

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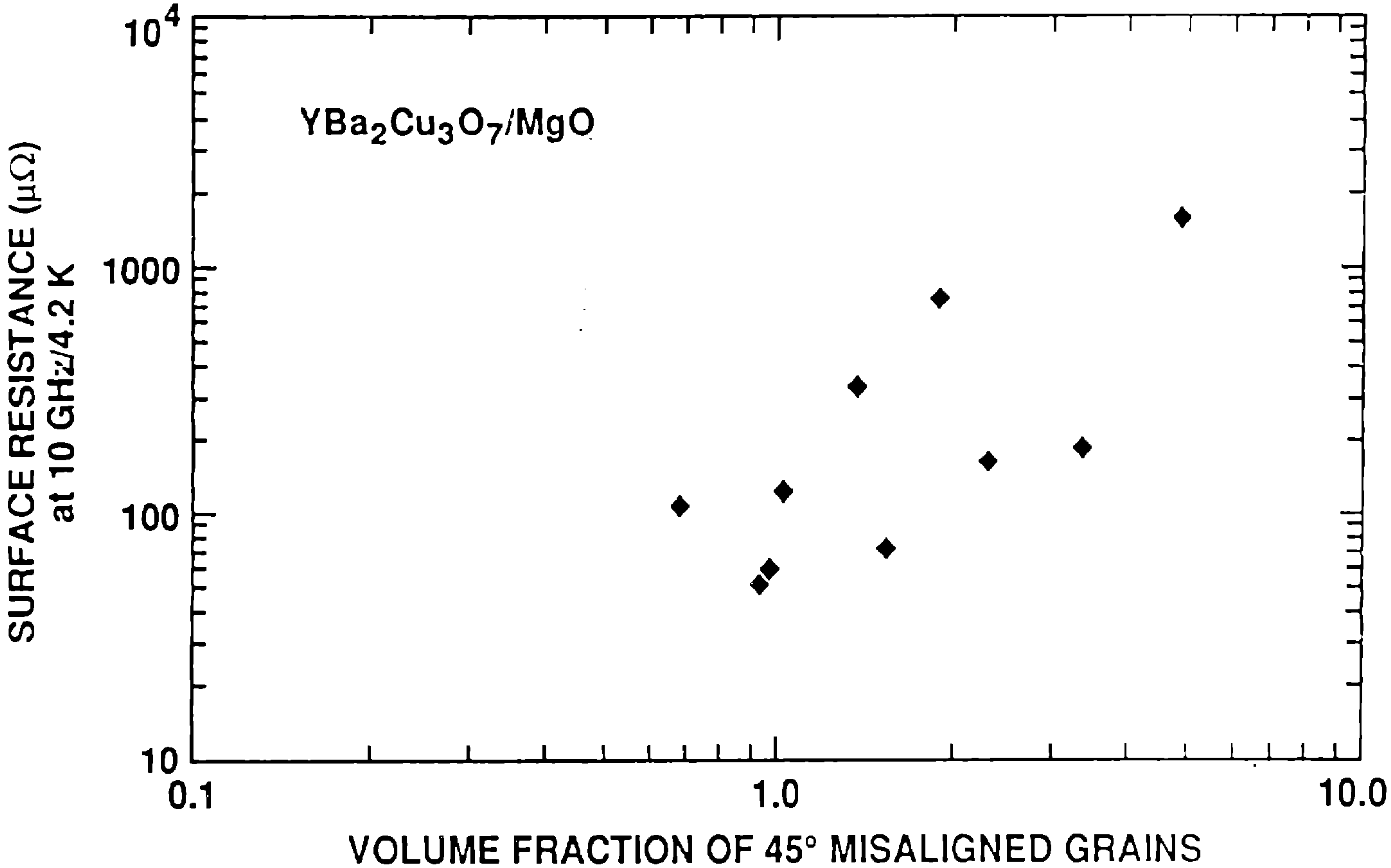
## Correlation Between Surface Resistance and Presence of Misaligned Grains

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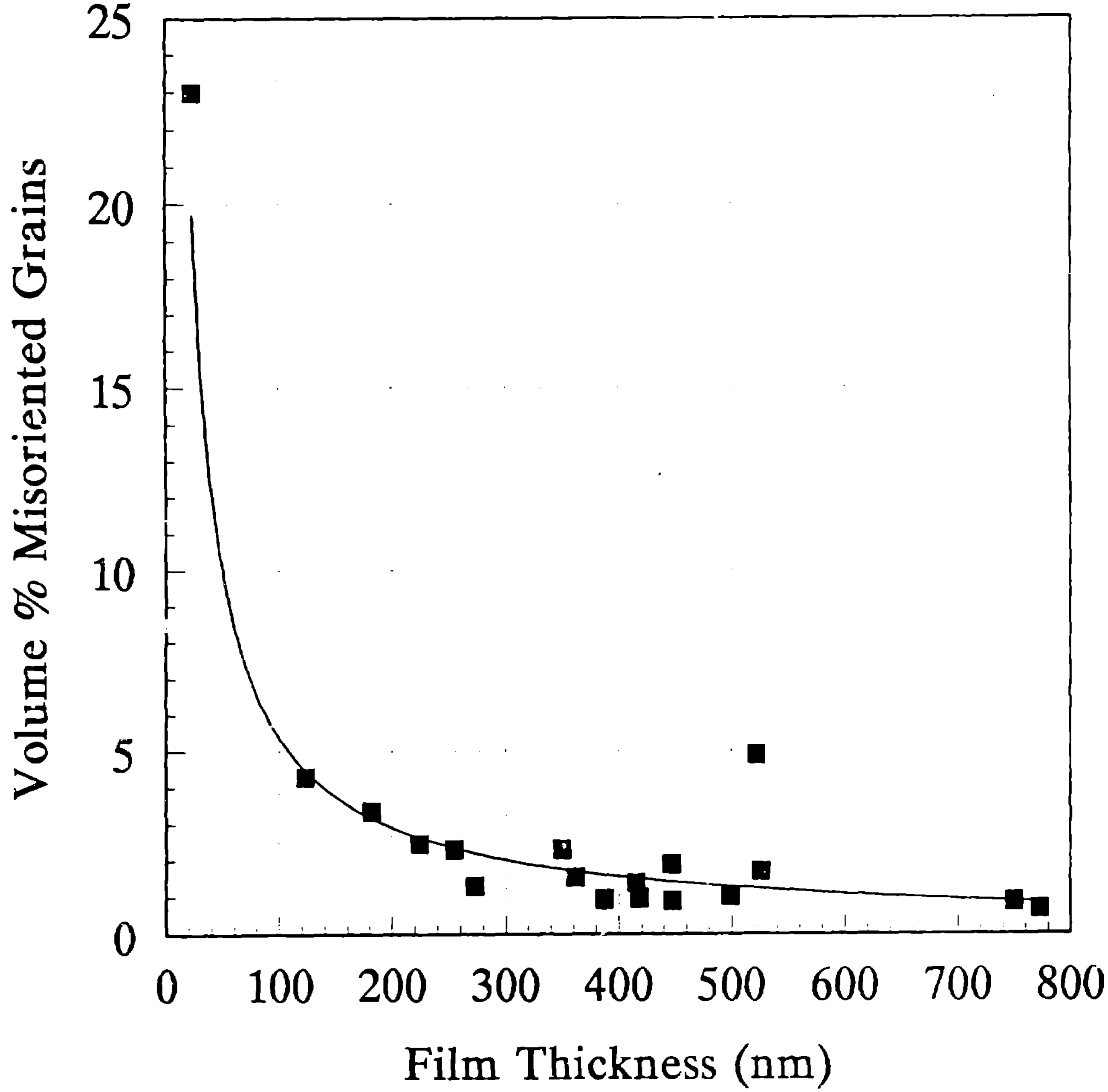
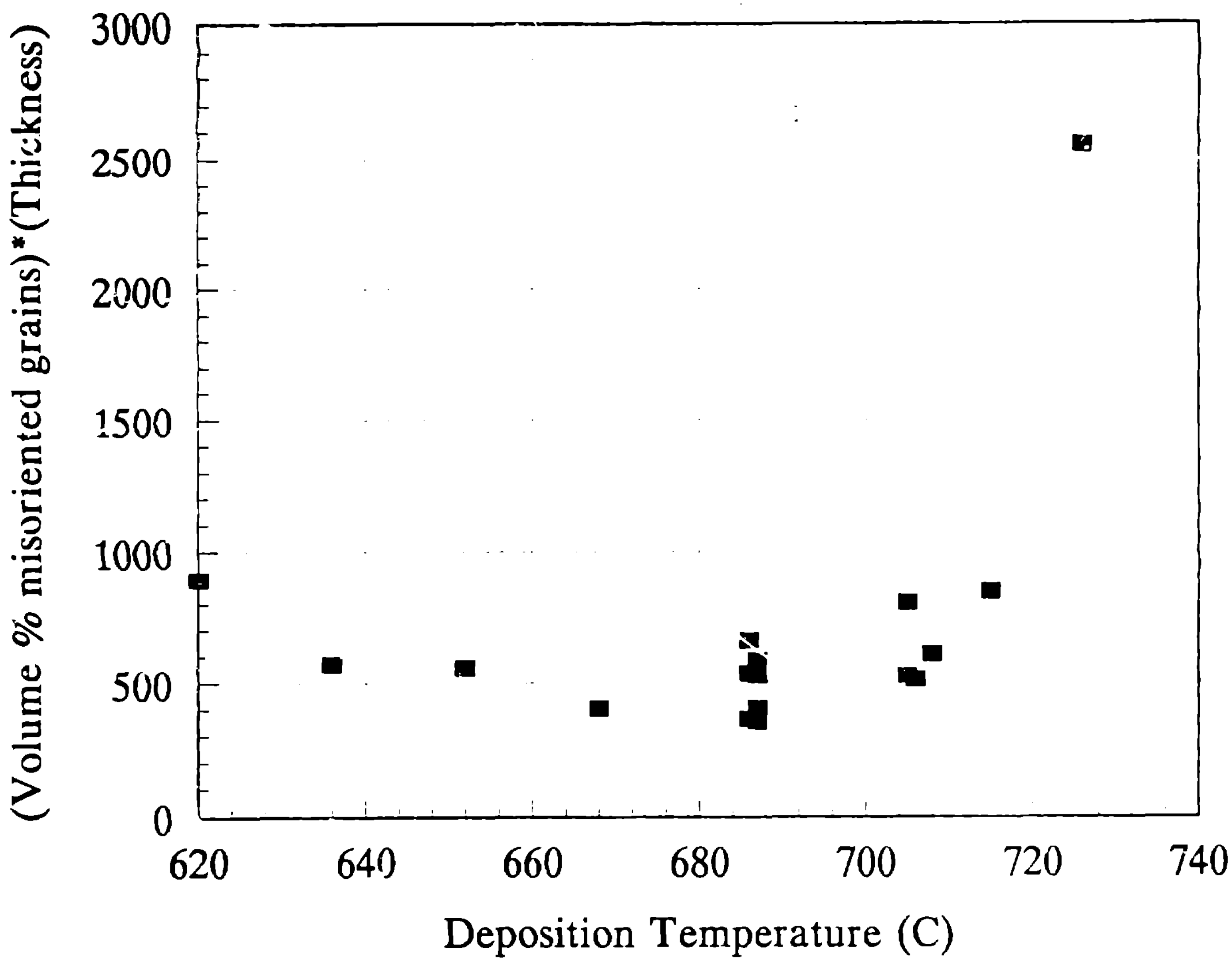


Volume % Misoriented Grains
25
20
15
10
5
0
0
100
200
300
400
500
600
700
800
Film Thickness (nm)



(Volume % misoriented grains)*(Thickness)
3000
2500
2000
1500
1000
500
0
620
640
660
680
700
720
740
Deposition Temperature (C)