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THE DETERMINATION OF STEP HEIGHTS ON THE NON-UHV HEAT-TREATED MgO (100) SURFACE BY CBED

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ABSTRACT

Conditions are established whereby step heights may be measured in the acid-washed and air-annealed MgO single-crystals currently employed as substrates for pulsed-laser-deposition. Preliminary data indicates that the steps are typically a few nanometers in height.

INTRODUCTION

The annealing of (100)-oriented MgO substrates, which has been shown to promote the formation of relatively large and well-defined surface steps [1], is known to improve the quality of thin films, such as YBCO [2], deposited onto their surfaces. YBCO, in particular, has been shown to exhibit grapho-epitaxy, in which steps on the surface promote the nucleation of islands and influence the subsequent growth into an interconnected film [3]. It has been suggested, however, that the formation of this interconnected film may be influenced by the height of the steps on the substrate (see reviewer's comments to reference [3]). It is conceivable that the height of the steps may also play a role in the island nucleation event itself. Before these phenomena may be analyzed further, however, an investigation of the step heights in the acid-washed and annealed substrates employed for the above studies is of paramount importance. This paper, therefore, establishes initial conditions for the determination of step heights by convergent-beam electron-diffraction (CBED) and present some preliminary results.

EXPERIMENTAL

The method of preparation of the TEM foils, based on reference [4], is given in reference [5]. The TEM was performed in a Philips CM30 EM operated at 200kV and CBED patterns were obtained in micro-probe mode using a nominal spot-size of 5.6 nm. and a diffraction vector $\vec{g}=\langle 220 \rangle$. In nanoprobe mode, beam damage resulting from the focused probe, and which for low contaminant levels is generally not a problem in normal imaging, was found to be unacceptably rapid.

Magnesium oxide is an insulator and, as such, is prone to charging. Additionally, it has been noted that the build-up of, mainly carbonaceous, contamination over time (a few weeks), from ambient sources, correlated strongly with the build-up of a large steady charge on the specimen during TEM. This surface contamination also leads to dramatically accelerated rates of beam damage, presumably resulting from primary surface-carbon-atom displacements. The steady charge is most evident when traversing from the center to the periphery of a large ($>100\mu$) hole in the specimen; in one case, resulting in a displacement equivalent to $\sim 4^\circ$ ($4g_{220}$) in the back-focal plane of the transmitted spot towards the edge of the specimen. As the specimen is further translated, towards thicker areas, the beam gradually retraces its path until the intensity is lost.

The difficulties presented by specimen charging are generally surmountable in normal imaging. However, when it is desired to employ 'spot-mode' techniques, such as CBED, on well-defined areas, actions such as small specimen translations, or changing objective apertures (and thereby altering the effective capacitance of the specimen-objective aperture plate array) as well as changes in the illumination conditions can displace the beam from the desired area and/or change the effective beam-tilt. Whilst this effect is notably reduced, and often barely detectable for samples without heavy surface contamination, the nature of these experiments, which rely on the validity of the 2-beam dynamical approximation, demands the adoption of the following procedures in addition to those customarily adopted [6,7]:

1) The objective aperture is not removed or changed during the experiment. A 70μ aperture was found to be ideal in that, suitably positioned, it allowed through both the transmitted and the

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that most steps could be resolved in the image. The beam tilt resulting from displacing the aperture to center it around the transmitted spot was not detectable.

2) The area from which the CBED pattern is taken is located using two double-exposures before and after each pattern is photographed. Initially the condensor lens is defocused sufficiently to give a recognizable image. After the desired terrace is centered this image is photographed. The probe is then focused onto the center of this terrace and a double-exposure taken of the focused probe. Typical exposure times are 30s and 0.044s, respectively. This procedure is repeated, in reverse, after the CBED pattern has been photographed. These images later may be compared with another taken under conditions of better step visibility (e.g., zone-axis BF, or weak-beam micrograph) and the precise position determined from which the CBED pattern was taken. Only those patterns for which the corresponding images show no drift are analyzed.

Certain other experimental constraints are also imposed: First, since it is desired to extract information from neighboring terraces, and, furthermore, to be sure that the probe be contained (and remain) wholly within a terrace, a region of the specimen must be located which is comparatively flat, i.e. which has sufficiently large, relatively widely-spaced terraces. This region should also contain, or be bordered by unmistakable, highly visible features, such as pores or the edge of the foil (see above). Second, the step traces, or at least the majority of them, must be visible under the 2-beam dynamical conditions, and with the objective aperture, employed: For the bright-field case, in which the contrast variations between neighboring terraces are extremely small, this places a practical upper limit on the specimen thickness beyond which absorptive losses render the step traces invisible. Third, sufficient minima (i.e., two) must be visible in the CBED pattern. This imposes a lower limit on the specimen thickness, the value of which is dependent on the imaging characteristics and microscope parameters employed. In practise, it was found that this combination of parameters restricts these experiments to regions between $\sim 850\text{\AA}$ and $\sim 1300\text{\AA}$ thick.

Finally, the CBED patterns are analyzed according to the technique of Kelly et al [4], as modified by Allen [5]. Measurements of the fringe separations were made optically, using a ruler marked with 1mm divisions on large prints. A typical distance between the transmitted and diffracted beam on these prints was 150mm.. Errors given on data points, thicknesses and individual extinction distances are mean errors derived via error propagation from the measurement errors (± 0.5 mm) in estimating the positions of extrema.

RESULTS

An area used for CBED analysis is shown in figure 1. This is a near-weak-beam image with the precise positions from which CBED patterns were taken indicated by filled circles. The specimen shown in figure 1 had been prepared only a few hours before this CBED experiment was conducted; a surface film was not in evidence, beam damage was apparently very slow and the

charging effect was minimal. The results of the analyses of the CBED patterns from these points is plotted in figure 2. Notice that the graphs of figure 2 are all very nearly parallel.

An area of a second specimen, which had been annealed for 10 minutes at 1350°C , is shown in figure 3. At the time of this experiment, this specimen was already several weeks old, and had been in the microscope many times. Heavy contamination was evident, resulting in severe beam damage, and particularly noticeable charging effects as outlined above. The analyses of the CBED data are shown in figure 4. These graphs are not always parallel to one another. The thicknesses and extinction distances determined from these analyses are listed in table I.

Figure 1: MgO (100) foil annealed in air for 15.5 mins at 1350°C

Figure 3: BF image with the locations indicated from which CBED data is taken.

Table I - Thicknesses and Extinction Distances derived from CBED patterns

Figure 2			Figure 4	
	Thickness (Å)	ξ_g 220 (Å)	Thickness (Å)	ξ_g 220 (Å)
1	901±12	871±37	954±10	865±27
2	921±10	869±30	1031±16	902±37
3	932±10	871±29	1069±15	887±52
4	967±11	868±27	1088±11	904±44
5	1011±11	857±23	1114±12	865±37
6	1031±11	867±23	1185±13	896±38
7	1096±15	863±46		
Av.		866.6±5.0		887±18

DISCUSSION

Inspection of figures 1 and 3, coupled with the thickness data of table 1, enables determinations of the step heights to be made. For example, looking first at figure 3, the contrast level only appears to undergo one discrete change between points 3 and 4, and 4 and 5, suggesting individual step heights of 17Å and 26Å, respectively. Step 2-3 of figure 4 (31Å) apparently forks into two at the bottom of the figure, which might suggest individual step heights ~16Å. Step 5-6 (71Å) is plainly composed of two separate steps, whilst the precise character of step 1-2 (74Å) is uncertain, but appears to contain at least 2 components. Again employing error propagation, the errors on these step heights are ~15Å, however, the data does appear to suggest a higher accuracy. A possible explanation for this discrepancy is that the individual measurements were more accurate than the commonly used error of one half of the smallest measuring division ($\pm 0.5\text{mm}$), since tenths of a mm. were estimated. Efforts are underway to improve the potential accuracy of these determinations. The interpretation of figure 1 is more complex, but the series 1-2-3-4-6-5-7 (height differentials of 20, 11, 35, 64, 20, 85Å, respectively) may be representative of the morphology of the material. Step 4-5 (44Å) plainly is composed of two steps, whilst discontinuities 5-7 (85Å) and 6-7 (65Å) appear to be large single steps. Lastly, a solitary very large step (1-7), at 195Å has been measured. The narrow-band contrast arising from these larger steps is thought to arise from inclined step faces [5].

The observation that laser-ablated YBCO thin film, although interconnected, is still not entirely continuous at thicknesses of 120Å [3] and 240Å [4], coupled with the initial findings of this study that steps on these annealed substrates are typically (with one exception) considerably lower than these heights would appear to support the view that YBCO on (100) MgO grows by a Volmer-Weber growth mechanism irrespective of step height. With an alternative view, that YBCO is merely constrained to grow as islands by the steps, one might expect a continuous film to form soon after a thickness ~ the step height had been reached. However, further determinations of step heights are necessary before a more definite conclusion can be reached.

The discrepancy in values of the apparent extinction distance between the two experiments outlined in this paper, as given in table 1 (the error quoted is one standard deviation), and particularly the lack or presence of significant experimental spread, may be ascribable to the presence of the thick carbonaceous layer. Further details of this will be given at a later date.

These results form part of a larger study aimed at a determination of the step heights as a function of annealing time and temperature. Whilst certain trends have already become apparent, these results also will be presented at a later date.

CONCLUSIONS

An experimental method, employing the well-known CBED technique, has been developed and successfully applied to the measurement of step heights in annealed MgO substrates. Initial determinations have found steps ranging in height from ~10Å to ~190Å, with a typical value for a single step being ~20-30Å in these specimens.

REFERENCES

- [1] M.G.Norton, S.R.Summerfelt, C.B.Carter, Appl. Phys. Lett., **56**, 2246-2248 (1990)
- [2] B.H.Moeckly, S.E.Russek, D.K.Lathrop, R.A.Buhrman, J.Li, J.W.Mayer; Appl. Phys. Lett., **57**, 1687-1689 (1990)
- [3] M.G.Norton, C.B.Carter, Scanning Microsc., **6**, 385-398 (1992)
- [4] M.G.Norton, C.B.Carter, J. Cryst. Growth, **110**, 641-651 (1991)
- [5] S.L.King, C.B.Carter - these proceedings.
- [6] P.M.Kelly, A.Jostsons, R.G.Blake, J.G.Napier, phys. stat. sol. (a), **31**, 771-780 (1975)
- [7] S.M.Allen, Phil. Mag. A, **43**, 325-335 (1981)
- [8] The authors acknowledge the support of the Department of Energy under grant number DE-FG02-89ER45381.

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