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# Analytical electron microscopy of multilayered thin films using microcleavage

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**Abstract.** Microcleavage transmission electron microscopy (MTEM) has been applied to the study of many properties of multilayered samples. We illustrate the unique capabilities of this technique for obtaining a detailed structural picture of the multilayer in order to study long-range perpendicular thickness drifts, lateral variations, roughness, substrate quality, adherence, thermal stability, composition, and crystallinity.

*Subject terms: x-ray multilayered optics; multilayered thin films; microcleavage; electron microscopy.*

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## 1. INTRODUCTION

Interest in soft x-ray optics has been enhanced by recent developments in synchrotron x-ray beam lines and by progress in thin-film deposition techniques. For some time, multilayers have been used as powerful tools for studying interfaces and diffusion,[1] opening up the possibility of producing high quality soft x-ray mirrors.[2-4] More recently, superlattices have received renewed attention because of their unique structural,[5] elastic,[6] and electronic properties.[7]

We present here the unique capabilities offered by electron microscopy, combined with a microcleavage technique developed earlier,[8] for studying the structure, crystallinity, adherence, and roughness of multilayers. We present several representative examples of which electron microscopy has been used to study some of the problems outlined above.

The properties of multilayer samples are heavily dependent on their structural properties. The main nondestructive characterization tool used to date has been x-ray diffraction, especially for samples in which the layer thickness is less than ~100 Å. As is well known, in x-ray diffraction measurements the phase information is lost, and therefore these studies rely heavily on proper structural modeling. In addition, x-ray diffraction generally arises from large volumes of the sample, therefore providing only an average, nonlocal picture of the structure.

Electron microscopy, on the other hand, provides information that is more local (from a volume ~$10^6$ to $10^9$ smaller than in x-ray diffraction). The phase information is not lost until the image is produced. In addition to the standard diffraction and imaging techniques, a variety of filtering techniques such as dark field imaging (DFI),[9] electron energy loss spectroscopy (EELS), and high resolution transmission electron microscopy (HREM) have been used to obtain a detailed structural picture of the multilayer. We also have used a hot stage grid holder to study the temperature stability.[10] To obtain an image that is easy to interpret, the sample size has to be reduced to microscopic size (less than 1000 Å), which is usually accomplished by performing extensive chemical and mechanical treatment and ion milling. Clearly, all of these sample prepara-

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**TABLE I. Systems for which the microcleavage transmission electron microscopy technique has been applied. Period thicknesses range from 15 to 300 Å, and the total number of bilayers ranges from 1 to 100. WRe/C, WRe/B, Pb/Ge, and Cu/Si were prepared using molecular beam epitaxy; the others were prepared using sputtering.**

| Multilayer | | Substrate | | |
|---|---|---|---|---|
| High Z material | Low Z material | Glass | Silicon | Other |
| W, Mo, Ta Au, Pt | C or Si | x | x | |
| Co, Cr, W WRe alloy | C | | x | |
| WRe alloy | B | x | | |
| W | $SiO_2$ | | | $Al_2O_3$ |
| Ta | $SiO_2$ | x | | |
| MoN | AlN | | x | |
| NbN | AlN | | x | |
| Pb | Ge | | x | |
| Cu | Si | | x | |

N.B.: The technique has also worked for bilayers with two low Z materials, such as (Si, $SiO_2$) and ($Si_3N_4$, $SiO_2$).

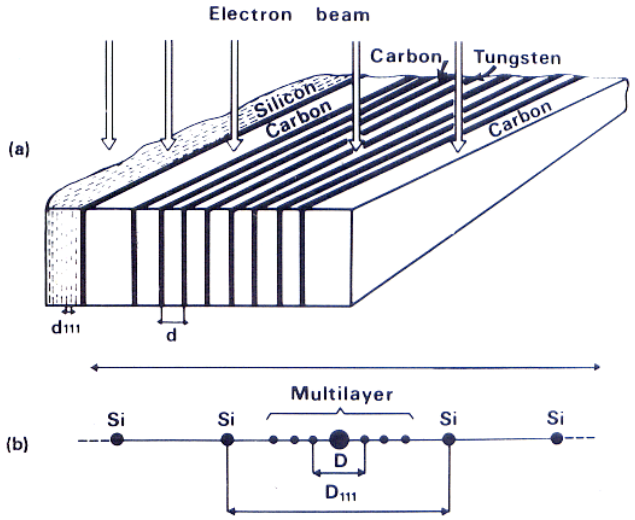


**Fig. 1. (a) Microfragment showing Si substrate, W/C multilayer, and direction of the electron beam. (b) Idealized diffraction image. The internal calibration[18] comes from $d \times D = d_{111} \times D_{111}$.**

tion techniques might produce some artifacts that are not easily controllable.

## 2. TECHNIQUE

We have successfully used the microcleavage transmission electron microscopy (MTEM) technique[11] to study the structural properties of a large number of different multilayer systems (Table I). In this technique, the substrate is scratched and the multilayer-substrate combination is cleaved along the scratch. Several microfragments of uncontrolled size and shape are collected on a microscope grid by rubbing the edge of the break. Generally, the fragments are wedge shaped, as shown in Fig. 1(a). To obtain a clean image, the planes of the layers are aligned parallel to the electron beam, as shown in the figure. In addition to the direct transmission image, a diffraction pattern can also be obtained, as shown in Fig. 1(b). Notice that by proper alignment, diffraction spots from both the substrate and the multilayer can be obtained so that the substrate acts as an internal calibration. An important advantage of the technique is its simplicity, allowing a quick characterization of the samples, which can be used as feedback information into the sample-making process.

### 2.1. Depth variation

Figure 2(a) shows a direct transmission image (obtained for a W/C multilayer ($2d \cong 48$ Å) in combination with the diffraction pattern [Fig. 2(b)] from the same sample. The spots close to the direct beam arise from the multilayer, and the further spot arises from the substrate. In this way, the diffraction from the substrate provides an internal calibration for the superlattice diffraction spots.[8] We should point out that the image for thin layers can be used to determine only the bilayer, not the individual layer, thickness. This is because the relative contrast is dependent on the focusing conditions of the microscope.

Figure 2(a) shows the bilayer thickness decreasing gradually toward the left of the figure due to a drift in preparation condition. This is much harder to deduce from a model-dependent reconstruction of the structure using x-ray or electron diffraction techniques, as is clearly seen from the electron diffraction picture [Fig. 2(b)], where the superlattice peaks are all merged into a continuous image that makes detailed modeling of the intensities mandatory. In contrast to diffraction, MTEM immediately allows the study of long-range drifts in preparation conditions, as shown for diode (Fig. 3(a)] and triode [Fig. 3(b)] sputtering.

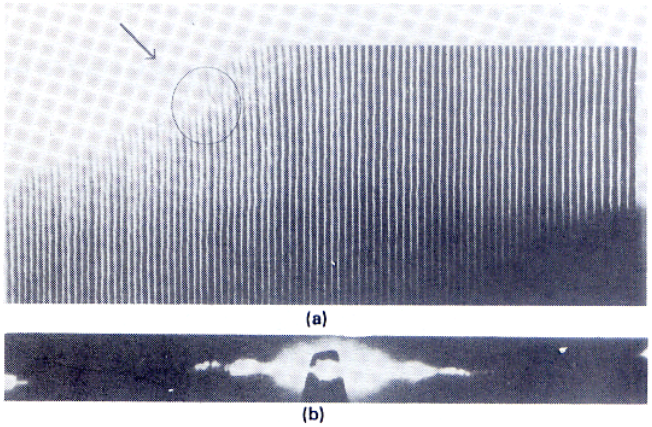


**Fig. 2. (a) Cross section of a W/C multilayer ($2d \cong 48$ Å). Notice the gradual decrease in the period toward the left. (b) Electron diffraction from the sample in (a), showing the effect of this decrease on the multilayer diffraction spots. The arrow in (a) shows a bifurcation structure[12] that we believe to be only an artifact of the electron microscopy technique.**

### 2.2. Lateral variation

Because of the finite sizes of the evaporation or sputtering sources, a lateral variation of the layer thickness may appear on the sample, as shown by Figs. 4(b) and 4(c), which are MTEM images of the top and bottom portions of the sample, respectively. Comparison of these two images shows the thickness differences between the two portions of the sample. This variation is also observable in a photographic image of

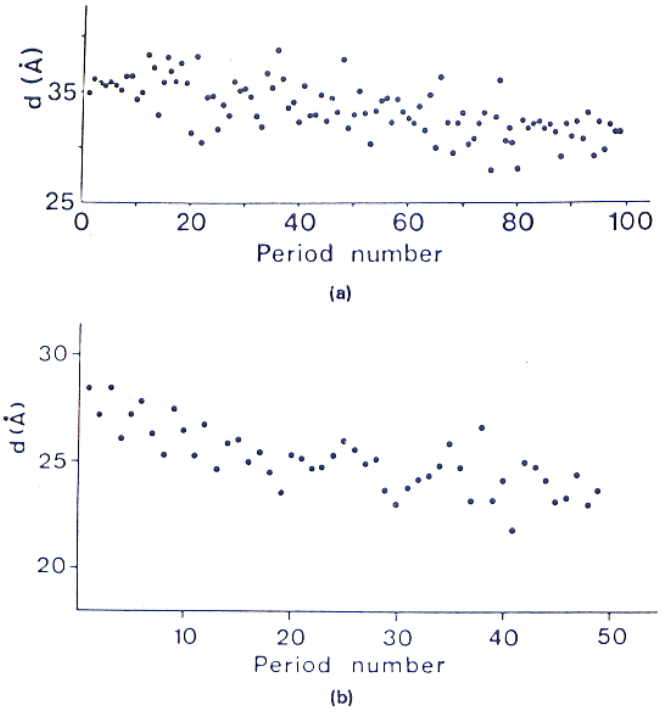


Fig. 3. Drifts in period obtained from figures similar to Fig. 2 for (a) diode and (b) triode sputtering.

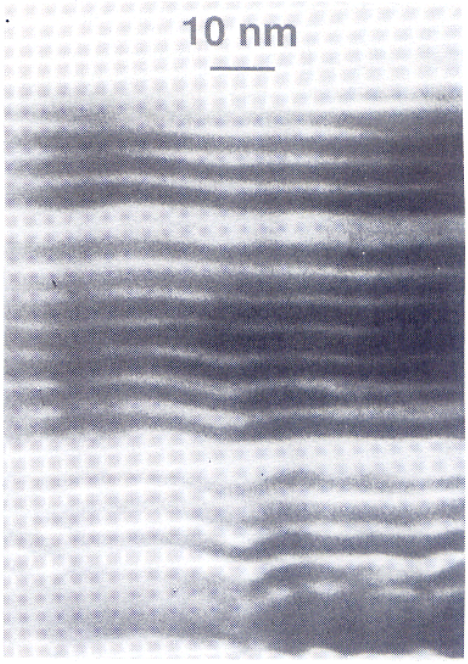


Fig. 5. Effect of roughness artificially induced by the evaporation of small (~10 to 30 Å) Pt beads on the substrate. In this case the roughness heals toward the top of the sample. Periodicity is 54 Å.

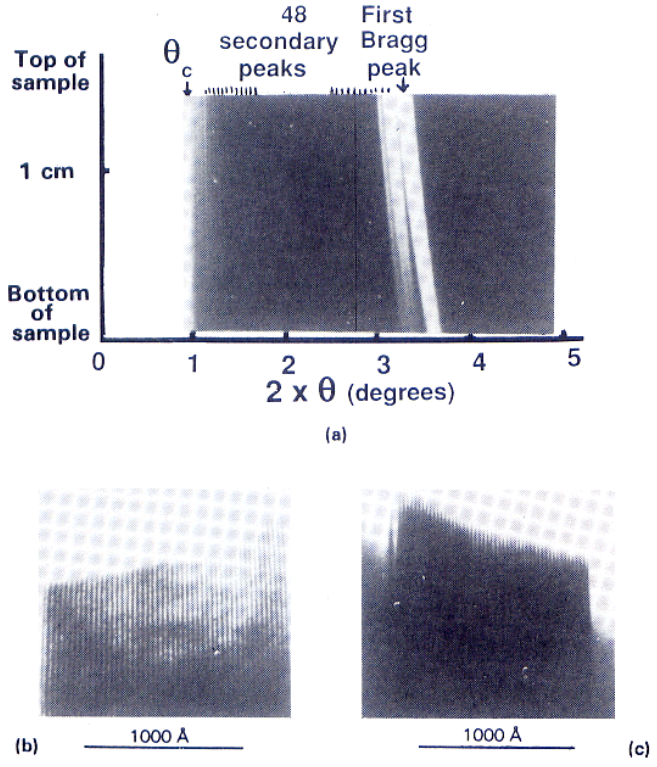


Fig. 4. (a) Lateral variation of layer thickness is observed in a photographic image of small-angle x-ray diffraction ($\theta_c$ = critical angle). Notice the 48 secondary peaks between $\theta_c$ and the first Bragg peak. MTEM images of (b) top and (c) bottom of the sample shown in (a).

the low-angle x-ray (CuK$\alpha$) diffraction, Fig. 4(a). For this the x-ray detector is replaced by a photographic plate that captures the variation of the x-ray intensity *along* the sample. Note that the angle at which the multilayer peaks appear varies continuously along the sample.

### 2.3. Roughness

The substrate or interfacial roughness also contributes to the deterioration of the multilayer x-ray optics performance. To illustrate the effect of substrate roughness on W/C multilayer deposition, we deposited small (~10 to 30 Å) Pt balls on the substrate. It is easy to see (Fig. 5) that the roughness tends to heal toward the top of the sample, although this effect varies from system to system. For this particular case it seems reasonable to conclude that the amorphous carbon is responsible for the smoothing effect. We have observed a similar behavior when carbon was used as a buffer between a rough substrate and a multilayer.

The effect of defects on the substrate ("substrate quality") and sample surface roughness can be observed in an image obtained by *transmission* in the direction perpendicular to the plane of the substrate and multilayer (Fig. 6). A detailed study of the surface roughness using transmission electron microscopy has not been performed, and it is possible that better height resolution can be obtained using scanning electron microscopy.[13]

The lower limit for the individual layer thicknesses is limited to about 10 Å in the case of W/C. This is because the individual layers are no longer continuous, as illustrated in Fig. 7, although on the average a modulation is still present.

Adherence to the substrate and an interruption of the multilayer preparation process can also be observed as additional breaks in the microcleavage.

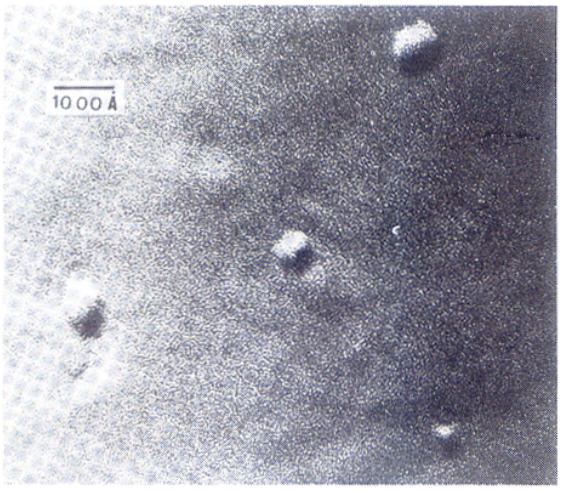


Fig. 6. Transmission image perpendicular to the layers, showing substrate defects and surface roughness.

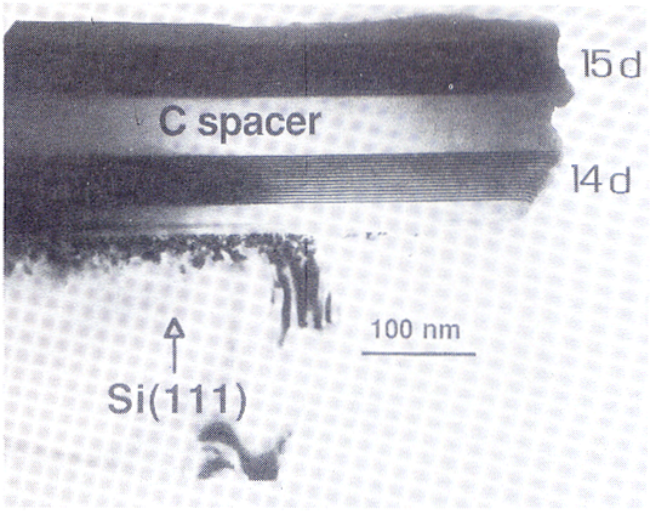


Fig. 8. Cross section of a Fabry-Perot structure showing well-formed continuous layers. The period is 32.5 Å, and the C spacer is 480 Å.

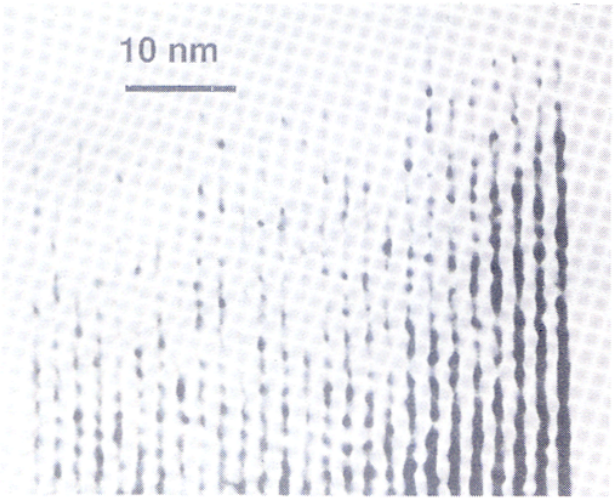


Fig. 7. Island structure of thin (<10 Å) W layers. Periodicity is 21 Å.

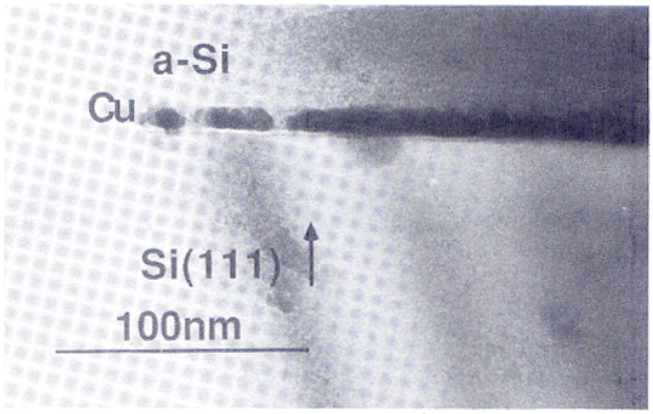


Fig. 9. Cross section of amorphous Si (420 Å) film and single-crystal Si substrate with a thin Cu layer (40 Å) sandwiched between.

### 2.4. Structures observable

Cross section images can be used to observe clearly and directly a large number of structures that are prepared using thin-film techniques. For instance, Fig. 8 shows a W/C Fabry-Perot structure,[14] and Fig. 9 shows a copper layer sandwiched between a single-crystal Si substrate and a sputtered amorphous Si film. Table I lists the large variety of structures for which the technique has been applied.

### 2.5. High resolution

A complete characterization of thin-film multilayered structures requires knowledge of the position of each atom in the multilayer. This might be possible with crystalline superlattices. Figure 10 shows the use of high resolution electron microscopy (HREM) with a Philips 420 microscope on a C/W sample where the individual layers are amorphous. The (111) planes of the silicon substrate give a direct and accurate scale for the transmission image; however, since the layers are amorphous, atomic planes are not observed in the multilayer. This type of high resolution microscopy can be done routinely with the latest generation of 400 kV microscopes. The main advantage of the HREM of the substrate-multilayer combination is that the substrate atomic planes provide an internal calibration scale, allowing accurate measurement of the layer thickness.

### 2.6. Chemical mapping by electron energy loss spectroscopy

EELS has been used to perform chemical mapping of the constituents.[15] Figure 11 shows the chemical profile of the carbon (top) and the tungsten (bottom) in a W/C multilayer. The annular dark field (ADF) gives the tungsten profile by selecting electrons scattered at high angles. In such a preliminary experiment, the spatial resolution is of the order of 10 Å.

## 3. CONCLUSION

It is clear that the richness of variability in multilayer structures makes electron microscopy ideally suited for their study. This is especially true when the microcleavage combined with

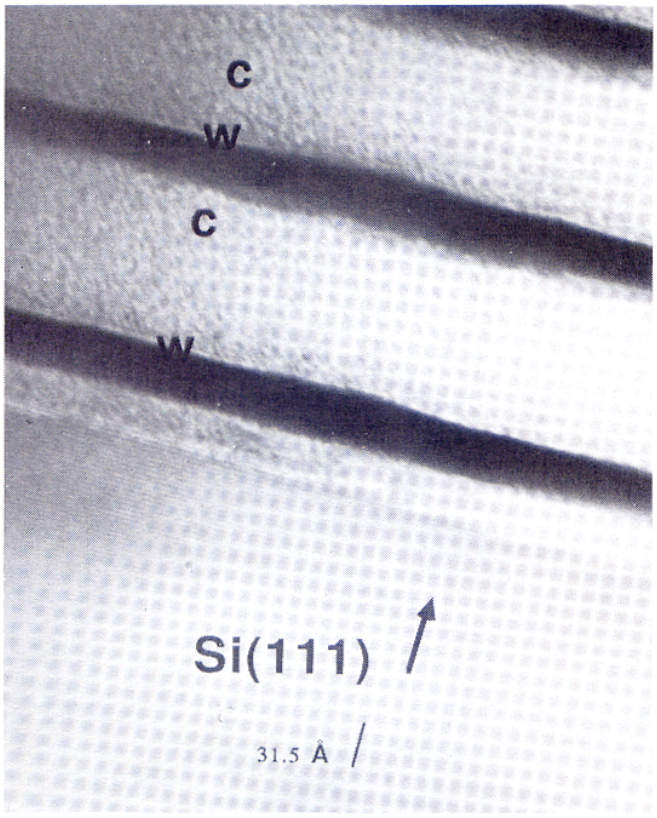


**Fig. 10. Cross section using high resolution electron microscopy with a Philips 420 microscope. The substrate provides an accurate scale: $10d_{111} = 31.5$ Å.**

an analytical microscopy technique is compared with ordinary diffraction techniques. The technique presented here is limited to samples for which (a) a proper cross section can be prepared, (b) the microcleavage does not affect the property of the samples, and (c) the contrast is sufficient to allow the formation of an image.

## 4. ACKNOWLEDGMENTS

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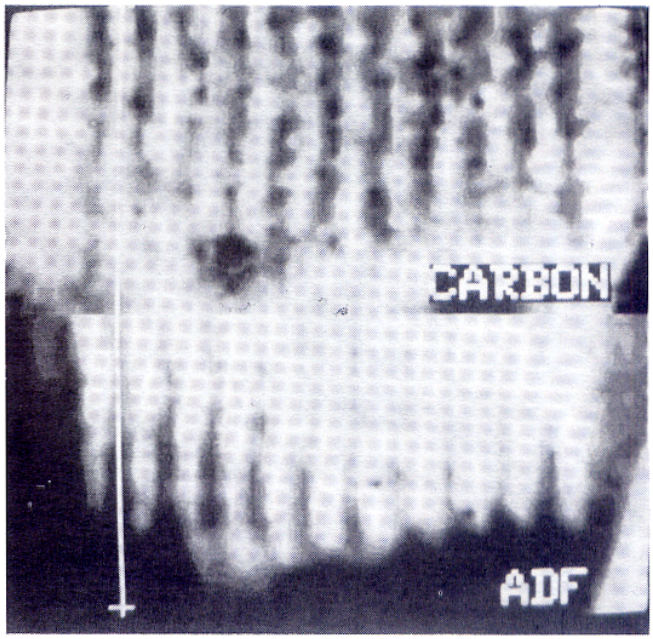


**Fig. 11. Chemical mapping using EELS (courtesy C. Colliex).**

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