Microcleavage transmission electron microscopy applied to the interfacial structure of multilayers and microstructure of small particles on a substrate

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We have developed a new technique useful in imaging microstructures such as small particles, multilayers, and superlattices on a substrate. By producing images at various angles with respect to the substrate we have been able to obtain new information on interfacial structure not yet available with other techniques.

The preparation, characterization, and study of small structures are problems of major current activity.[1,2] Many of the physical properties of microstructures such as small particles, multilayers, or superlattices are strongly connected to their morphology. Structural characterization is mostly accomplished with the use of diffraction (x ray, neutron or electron) and imaging [transmission (TEM) and scanning (SEM) electron microscopy] techniques. In the past, TEM images of epitaxial structures have been obtained from cross-sectional specimens observed at different foil orientations.[3] In those cases the final thinning of the sample was carried out by ion bombardment. We present here a novel microcleavage technique applied to the imaging of the structure of small particles and superlattices. We believe this is the first time that (a) transmission electron microscopy has been used to image small particles *on a substrate* and (b) an interfacial image of *a multilayer* has been obtained.

Microcleavage transmission electron microscopy[4] (MTEM) is a technique which was developed for the fast imaging of microstructures, allowing immediate feedback to the preparation technique and avoiding long and cumbersome ion milling techniques. In MTEM the overlayer (superlattice, small particle, etc.)/substrate (usually Si) combination is cleaved. A standard microscope grid (300–400 mesh, copper) is rubbed against the fresh cleavage and thus randomly shaped fragments of the substrate and overlayer are collected on the grid. Because the fragments are of microscopic size they adhere quite firmly to the grid possibly due to electrostatic charging. We should stress that because the fragments are of microscopic size they cannot be manipulated and they have to be collected by a random process. In most cases, a short search on the grid, easily accomplished with a goniometric head, reveals a fragment as shown in Fig. 1 for a multilayer. The electron beam can thus be oriented at various angles [Fig. 1(a) parallel and Fig. 1(b) at an angle θ] with respect to the multilayer planes and images of the overlayer and substrate are produced in this fashion.

A variety of small particles (Ag, Au, Cu, Cr, Ni, Al) was produced using electron beam gun evaporation (at 10^{-7} Torr) on a room-temperature single crystal of Si(111). The rate (typically 2–10 Å/s) was controlled in a feedback loop using quartz crystal oscillators. MTEM of a *nominally* 26-Å-thick Ag film at various angles with respect to the substrate is shown in Fig. 2. The existence of a distribution of sizes is clear from the pictures. A statistical study of the sizes [for Ag on Si(111)] shows them to have a Gaussian distribution centered at about four times the nominal thickness ($\sim$100 Å in this case) and a width of about 20 Å. This should be compared to the log normal distribution ordinarily found in small particles.[2] The exact distribution and detailed shape of the particles depend on the nature of the overlayer. The most striking feature of this work is that as the sample is tilted closer to the parallel direction (0°) the averaging along the direction of the electron beam produces an image which looks like an almost continuous layer on the substrate.

We also have prepared a W/C multilayer on a Si(111) substrate, with the W varying from 5 to 20 Å, using sputter deposition. A MTEM picture at two different angles with respect to the substrate is shown in Fig. 3. Note that if the sample is perfectly aligned (0°) with respect to the electron beam [Fig. 3(a)] the image shows clearly well-formed layers with a minimal apparent roughness. However, an image of the multilayer obtained at a tilt angle of $\sim$15° reveals additional structure inside the W component of the multi-

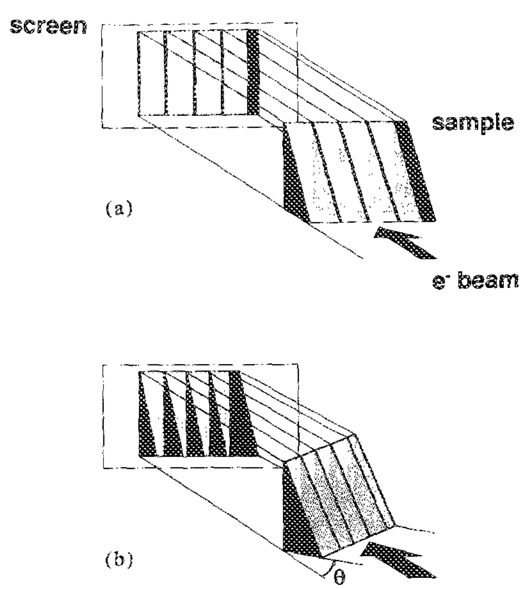

FIG. 1. Schematic of the microcleavage transmission electron microscopy (MTEM) wedge and two different geometrical arrangements with respect to the electron beam and screen. (a) In this geometry the beam is oriented parallel to the layers so the image consists of parallel lines which are a result of the averaging in the direction parallel to the electron beam. (b) In this case the sample is tilted with respect to the electron beam and the screen. In this fashion the electron beam penetrates through the interface. Note that each layer appears as a triangular shaped object.

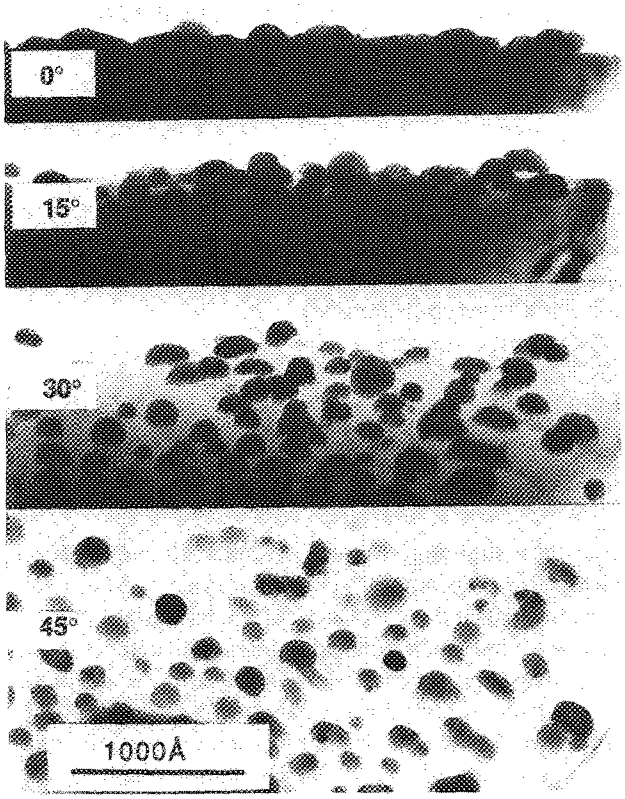

FIG. 2. MTEM images of small Ag particles on a silicon substrate at different angles. Note the averaging that occurs if the beam is parallel to the substrate-particle interface (0°) and that in some cases it is even possible to observe one particle behind another.

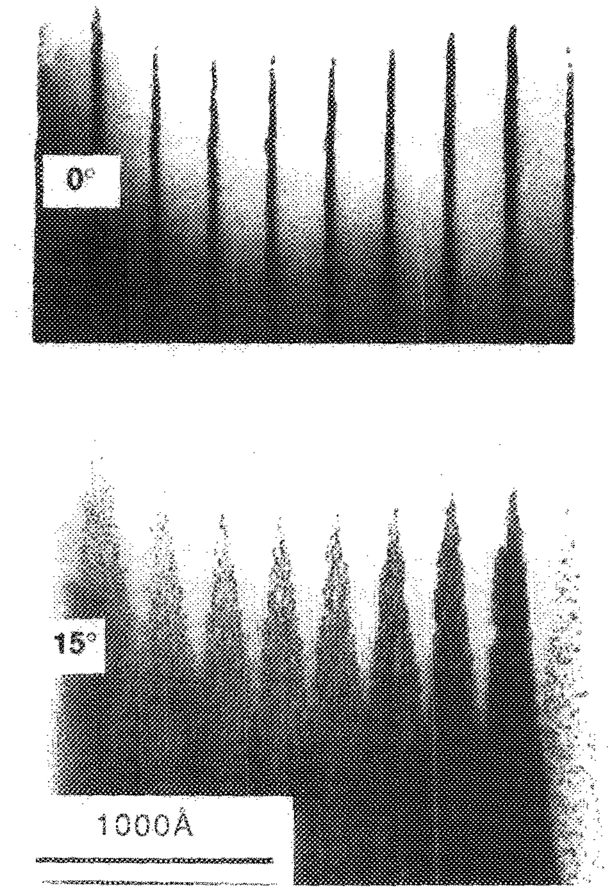

FIG. 3. MTEM images of a W/C multilayer with variable W thickness. The thinnest layer is at the left of the figure and the last layer at the right is pollution on the substrate. Note that the thinner the W layer is the more discontinuous it appears to be in the tilted images.

layer. Note that the "thinner" W layer shows a finer grained aspect than the thicker W layer. The *detailed* interpretation of the structure in the tilted image is not clear at the present time. However, we believe that it probably arises from the interfacial roughness. In the zero degree orientation, the averaging along the substrate parallel direction produces images which arise from *seemingly* continuous layers with minimal roughness. Although the technique has been applied to more than 20 different multilayer combinations, in all cases one of the components was a brittle nonmetal (insulator or semiconductor). At present, it is not clear whether due to lack of contrast metal/metal multilayers cannot be imaged in this fashion or whether the mechanical properties are such that the microfragments do not form with our preparation technique.

Presently, we are not quite sure of the implications of these measurements for other cross-section images of multilayers. It is clear, however, that MTEM combined with tilted images is a powerful technique which gives directly interfacial information at short length scales.

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[1]See, for instance, *Synthetic Modulated Structures*, edited by L. L. Chang and B. C. Giessen (Academic, Orlando, 1985).
[2]For recent work see Surf. Sci. **106** (1981) and **156** (1985).
[3]M. S. Abrahams and C. J. Buiocchi, J. Appl. Phys. **45**, 8 (1974).
[4]Y. Lepêtre, I. K. Schuller, G. Rasigni, R. Rivoira, R. Philip, and P. Dhez, SPIE Proc. **563**, 258 (1985).