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Corresponding Author: czigany.zsolt@ttk.mta.hu

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Plan-view preparation of TEM specimens from thin films using adhesive tape.

Zsolt Czigány

Research Institute for Technical Physics and Materials Science, Thin Film Physics Laboratory, H-1121 Budapest, Konkoly-Thege M. út 29-33, Hungary

Running Title: Plan-view TEM sample preparation technique

Abstract: A simple plan-view sample preparation technique for transmission electron microscopy (TEM) specimens is proposed for thin films by tearing-off the film with adhesive tape. The demand for very thin samples is highest for nanostructured materials where the structure of 2-5 nm sized features (grains) needs be resolved, therefore, overlapping of nanometer-sized features should be avoided. The method provides thin areas at the fracture edges of plan-view specimens with thickness in the range of the grain size in the film allowing for artefact free high resolution TEM (HRTEM) imaging. Nanostructured materials typically fracture between the grains providing areas with the thickness of the grain size. Besides the swiftness of the method, the samples are free of surface amorphisation artefacts which can occur in ion beam milling up to 1 nm depth even at low energy ion bombardment. The thin film tear-off technique is demonstrated on a CuMn alloy thin film with grain size of 2 nm.

Key words: artefact free, plan-view sample preparation technique, HRTEM, nanostructure, thin films

INTRODUCTION

An obvious difficulty in the high resolution transmission electron microscopy (HRTEM) investigation of nanostructured materials arises from the projective nature of the TEM images. In nanocrystalline materials with grain size of some nanometers the overlapping of the grains may cause Moiré effects which may confuse the true lattice structure. This difficulty is more pronounced in samples such as fullerene-like CN_x which is composed of ~5 nm sized multishell features – so called nano-onions (Czigány et al., 2003). Since the fullerene-like structure does not consist of parallel atomic planes (like single crystals), but curved shells of various radii in leek- or onion-like arrangements the caused projection or Moiré effects in TEM make the interpretation of the images even more complicated. The appearance of the image depends very much both on specimen thickness and defocus (Czigány et al., 2003). Therefore preparation of thin TEM specimens, of thickness compatible with the characteristic feature size, is necessary, where the HR images taken at Scherzer defocus can be interpreted. Besides the specimen thickness, a difficulty in TEM arises from sample preparation induced artefacts, in particular associated with ion beam milling (Barna et al., 1998). The thickness of the amorphised surface layer can be 5-10 nm for conventional ion energies (3-10 keV) and ~1 nm even at 0.25 keV bombarding energy for Si which can be comparable to the nanometer sized features. An alternative method to avoid ion milling in preparation of cross-sectional samples is small angle cleavage technique (SACT) (McCaffrey, 1991) employed for Si, semiconductors and other brittle materials. For plan view investigations the floating-off methods of films from NaCl substrates or alternatively direct deposition of the thin film on carbon coated TEM grids can be employed as an alternative to ion beam milling. However, the thickness of the floated off films can easily exceed the dimensions of the nanofeatures in the film causing the above mentioned projection or Moiré effects. This difficulty could be overcome for CN_x by making the HRTEM investigation at spontaneously formed fracture edges of the film (Czigány et al., 2003), achieving specimen thickness compatible with the characteristic feature size, allowing individual features to be imaged.

In this paper a plan-view sample preparation technique is proposed for HRTEM where the fracture edges of the film formed intentionally to achieve thin specimen areas. The method is quick and avoids ion beam induced artefacts.

EXPERIMENTAL

The proposed sample preparation method was applied to DC magnetron sputtered CuMn alloy thin solid films. The applicability of the method is demonstrated on a CuMn alloy of 66 at% Mn content having the smallest grain size at this composition (Czigány et al., to be publ.). The film was deposited at 100ºC in 2×10^{-3} mbar argon with total deposition rate of 0.25 nm/s at a base pressure of 1.4×10^{-6} mbar. To minimize the necessary sample preparation, the 50 nm thick CuMn alloy film was deposited on a C coated Ni TEM grid to allow for direct TEM investigation.

TEM plan-view sample was produced by placing half of the specimen on the sticky side of an adhesive tape (Kapton® tape by Du Pont or Scotch® tape) with film towards the tape (Fig. 1). A simple stainless steel plate device was constructed to hold the adhesive tape (Fig. 1). When the grid is removed from the adhesive tape, the film in contact with the tape will tear away from the remaining film. The fracture edges of the remaining film will be suitable for HRTEM investigation. The conventional TEM investigation was made in a Philips CM20 electron microscope operated at 200 kV. High resolution TEM investigation was made in a JEOL3010 transmission electron microscope operated at 300 kV with 1.7 Å point resolution.

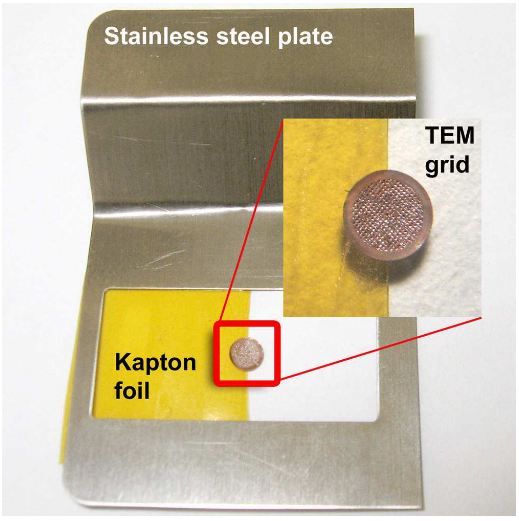

Figure 1: Sample preparation kit for creating thin film areas at fracture edges. The adhesive Kapton tape is fixed on the window of a stainless steel plate. Half of the TEM grid is attached to the Kapton foil with the film side towards the adhesive surface of the tape. After removal, the torn film provides thin areas at the fracture edges.

RESULTS

The as deposited film was suitable for conventional TEM analysis and selected area electron diffraction (SAED). Fig. 2 shows a plan-view bright field (BF) and dark field (DF) image and diffraction pattern of a 50 nm thick CuMn film. The diffraction pattern shows two broad rings due to the nanocrystalline nature of the film. The rings correspond to ~1.25 Å and ~2.13 Å, respectively. Weak rings of cubic MnO at 2.56Å and 1.57Å can also be recognized in the SAED (not marked in Fig 2.). The film shows uniform contrast in the BF image. The

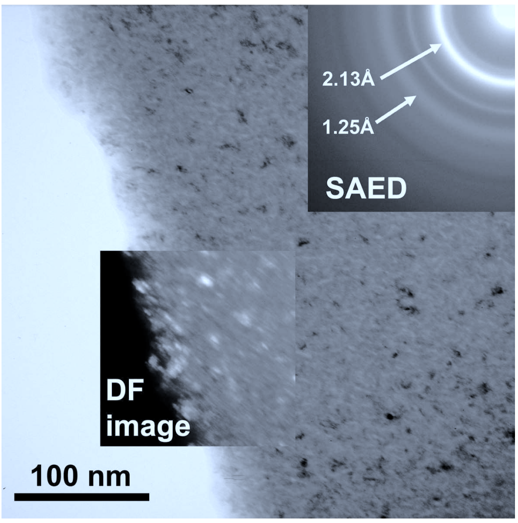

Figure 2: Bright field image of a 50 nm thick CuMn film (66 at% Mn). The SAED pattern (upper inset) consists of broad diffraction rings of the tetragonal CuMn alloy phase due to small grain size which can be concluded from the dark field image as well (lower inset). Weak rings of cubic MnO are also visible (not marked in the SAED).

grain structure can be estimated from the DF image suggesting an average grain size of approximately 5 nm. In addition to the ring broadening in SAED, the small grain size in the DF images indicates that the crystallites are significantly smaller than the film thickness and the large number of overlapping extinguishes the diffraction contrast in the BF image.

The high resolution image (Fig. 3) shows a thin area of the CuMn film at the fracture edge of the sample. At the edge ~2 nm sized equiaxial crystalline grains can be seen with lattice plane spacing of 2.13 Å and 2.56 Å. The 2.13 Å spacing corresponds to the {111} lattice plane spacing of CuMn alloy (inxeded as a tetragonal phase). 2.56 Å spacing corresponds to cubic MnO {111} lattice planes. At a thicker part of the image the lattice planes still can be resolved but the individual grains cannot be clearly distinguished. Thin regions can be found at the fracture edges where the sample thickness is of the order of the grain size and the true structure of the nanomaterial can be imaged. These extremely thin areas can be achieved because the fracture of this nanostructured material apparently takes place between the crystallites, similar to the fracture of CN_x nano-onion films (Czigány et al., 2003), leaving the nano features intact.

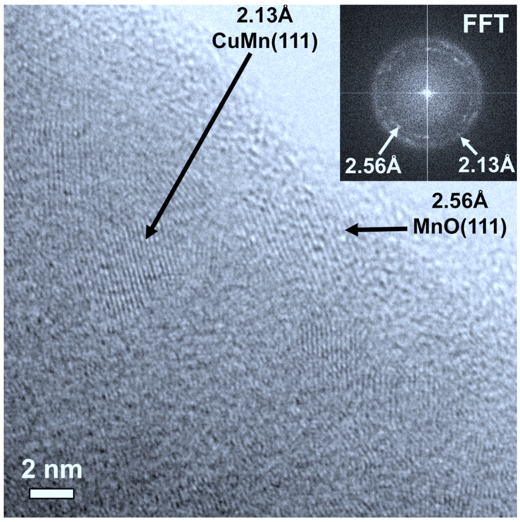

Figure 3: HRTEM plan-view image and its Fourier transform (inset) of a CuMn alloy film shown in Fig 2. Tetragonal CuMn and cubic MnO nanograins can be distinguished based on their lattice spacing.

DISCUSSION

In HRTEM investigations the artefacts from sample preparation can be the limiting factor in the exploration of the structure of materials. According to the damage depth investigation due to ion beam milling, (Barna et al., 1998) reported a damage depth of 5 nm at 3 keV and 1 nm at 0.25 keV bombarding energy for Si and 2.1 nm for GaAs at 1.6keV and non detectable damage at 0.25 keV. In some cases, (e.g. single crystals) the effects of an amorphized surface layer can be decreased by choosing a somewhat thicker part of the sample for investigation (e.g. utilizing a higher acceleration voltage of the TEM). In this way the influence of the amorphized layer can be decreased by improving the thickness-ratio of the original structure to the amorphized surface layer. However, this does not apply for nanostructures with 2 nm sized grains where the amorphised layer of 1-2 nm (on both sides of the specimen) can result that the thickness ratio of the real structure and the artefact can be as low as 30%, even applying low energy ion beam milling. Similar estimate was given in (Czigány et al., 2003) for fullerene-like CN_x in spite of the relatively larger size (~5 nm) of the nano-onions, but in that case, the thick amorphised layer is partially due to high specific sensitivity of CN_x to ion bombardment. The ion beam bombardment can also modify the chemical composition and bonding of compounds as N loss was reported for CN_x films by (Czigány et al., 2003) with XPS at moderate bombarding energies (3-5 keV).

An alternative way for preparation of TEM cross sections to avoid ion beam milling is the small angle cleavage technique (McCaffrey, 1991). SACT was developed for semiconductors and epitaxial single crystal films (McCaffrey, 1991). It provides excellent samples when the film and the substrate share cleavage planes. SACT can also be applied if the film has no preferred fracture directions. Then, the cleavage direction of the film will be determined by the substrate. The presented technique for plan-view sample preparation by tearing-off the film with adhesive tape is a plan-view sample preparation analogue of SACT. Having no substrate, the fracture is determined exclusively by the inherent structure of the film.

CONCLUSIONS

A simple plan-view sample preparation of transmission electron microscopy (TEM) specimens is proposed for thin films by tearing-off the film with adhesive tape. The demand for very thin samples is highest for nanostructured materials where the structure of 2-5 nm sized features (grains) need be resolved; therefore overlapping of nanometer-sized features should be avoided. The method provides thin areas at the fracture edges of plan-view specimens with thickness in the range of the grain size of the film allowing for artifact free high resolution TEM (HRTEM) imaging. Fracture of nanostructured materials apparently takes place between the grains providing areas with the thickness of the grain size. Besides the swiftness of the method, the samples are free of surface amorphisation artefacts which can occur in ion beam milling up to 1 nm even at low energy ion bombardment (Barna et al., 1998). The method is demonstrated on a CuMn alloy thin film (66 at% Mn) with grain size of 2 nm.

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