

Interface broadening due to Ar⁺ ion bombardment measured on Co/Cu multilayer at grazing angle of incidence

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A Co/Cu multilayer system, with double layer thickness of about 60 nm was Auger electron spectroscopy depth profiled applying 1 keV Ar⁺ bombardment, varying the angle of incidence in the range of 78°–86°. It was found that (a) the relative sputter rate changes with the angle of incidence; (b) the interface broadening of the Co/Cu transition is larger than that of the Cu/Co transition and depends on the depth; (c) the broadening of the Co/Cu transition increases with increasing relative sputter rate; and (d) the broadening of the Cu/Co transition does not depend on the relative sputter rate. These features will be explained as the consequences of the strong preferential sputtering of Cu (with respect to Co) in this angular range. Because of the preferential sputtering ion bombardment induced roughening and smoothening (which was directly observed by atomic force microscopy measurements) occurs on the Co/Cu and Cu/Co interface, respectively. D-TRIM simulation has been also performed which suggested that ion mixing also causes larger broadening on the Co/Cu than on the Cu/Co interface. © 2003 American Vacuum Society. [DOI: 10.1116/1.1559921]

I. INTRODUCTION

Specimen preparation (ion milling) for cross-sectional transmission electron microscopy (XTEM) studies frequently applies low energy ion bombardment at grazing angle of incidence. Destructive depth profiling techniques [Auger electron spectroscopy (AES), x-ray photoelectron spectroscopy depth profiling, and secondary ion mass spectrometry] also apply ion bombardment of similar sputtering conditions to remove the material in a sectioning way. During ion bombardment of a solid only part (generally smaller) of the energy is used for removing atoms, and the rest is dissipated in the material resulting in various defects; the lower the energy the lower the damage. Since the goal in these applications is to measure damage-free surface the ion energy is reduced as much as possible. Part of the dissipated energy causes surface roughening. The roughening weakly depends on ion energy, but strongly depends on the angle of incidence.^{1–3} Many papers have been published on surface roughening. The theoretically well based approaches, using various master equations, seem inadequate to predict the morphology development at a wide range of the sputtering parameters and material structures.^{4,5} However, phenomenological approaches based on quite different assumptions^{3,6,7} seem to be applicable for the description of the morphology development for sputtering parameters used in ion milling and AES depth profiling. These theories predict smoothing if grazing angle of incidence is used for ion sputtering, and the specimen is rotated during sputtering. These predictions are in

excellent agreement with the vast number of ion milling experiences¹ and AES depth profiling studies.^{8–10}

In this article we will show, however, that there are deviations from the above general rule. Co/Cu multilayer structure with a double layer thickness of roughly 60 nm was AES depth profiled using 1 keV Ar⁺. The angle of incidence varied in the range of 78°–86°. We found that the sputter rate of Cu is larger than that of Co and the ratio strongly changes in this range of the angle of incidence. The interface broadening turned to be different for the two, Co/Cu and Cu/Co, interfaces; it is larger for the Co/Cu interface than for the Cu/Co interface. The broadening of the Co/Cu interface increases with increasing relative sputter rate, which will be explained by ion bombardment induced interface roughening and ion mixing.

II. EXPERIMENT

The copper/cobalt multilayer structure was sputter deposited on polished single-crystal (111) silicon substrates in a plasma beam sputter deposition system (Sputron Balzers). The thickness of the individual layers was controlled using a quartz crystal microbalance during sputter deposition. The nominal thickness of all layers was about 30 nm.

The thickness of the individual layers was determined by Rutherford backscattering spectroscopy (RBS), which applied ⁴He⁺ bombardment. The detection angle with respect to the beam was 165°. The sample was tilted to 70° to improve the depth resolution.

Cross-sectional transmission electron microscopy (XTEM) was used to estimate the intrinsic interface roughness. XTEM studies were carried out using the usual recipe;

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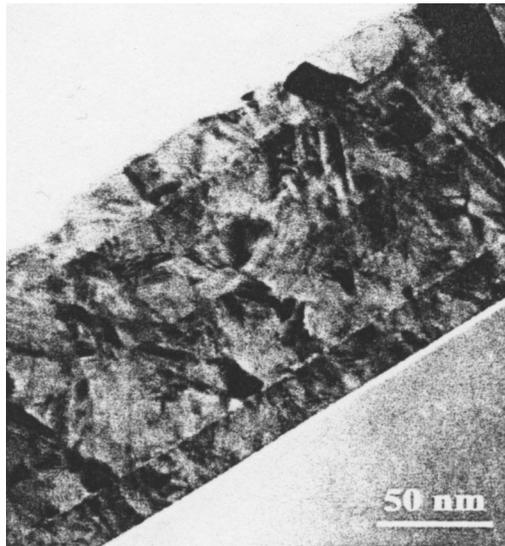


FIG. 1. XTEM image of the specimen.

two parts of the specimen were glued together face-to-face, and this unit was ion milled using the usual scheme.¹¹ 10 keV ion energy was applied for removing the major part of the material, approaching the perforation, ion energy was decreased to 0.25 keV to get rid of the damaged layer. The specimen was rotated during ion sputtering and the angle of incidence was 88°.

Auger depth profiling was carried out using a dedicated depth profiling device.¹² The projectile was Ar⁺, with energy of 1 keV. The depth profiles were recorded at angles of incidences of 78°, 82°, 83°, 84°, 85°, 85.5°, and 86° (with respect to the surface normal). The specimen was rotated during sputtering. The target current proportional to the bombarding current was measured during sputtering, and was used to keep the sputtering current constant.

The Cu 920 eV and Co 656 eV (to avoid the overlapping with Cu) Auger peaks were recorded in direct current mode by a preretarded cylindrical mirror analyzer (STAIB DESA 100). The measured curves were numerically differentiated and the peak-to-peak heights were used to characterize the surface concentration.

III. RESULTS

The actual structure was determined by RBS and was found to be Cu 26.3 nm/Co 23.3 nm/Cu 25.8 nm/Co 22.2 nm/Cu 26.0 nm/Co 22.5/Si substrate (instead of the nominal 30 nmCu/30 nm Co).

Figure 1 shows a typical XTEM image; the interfaces are flat and smooth and do not change with depth. The intrinsic roughness of the interface can be estimated to be less than 2 nm (rms).

Figure 2 shows two as recorded depth profiles [Auger peak-to-peak heights, Co (656 eV) and Cu (920 eV), as a function of the sputtering time]. For both measurements the ion energy (Ar⁺) is 1 keV, while the angles of incidence are 85° and 78°, respectively. The amount of surface contamination varied from specimen to specimen and thus the begin-

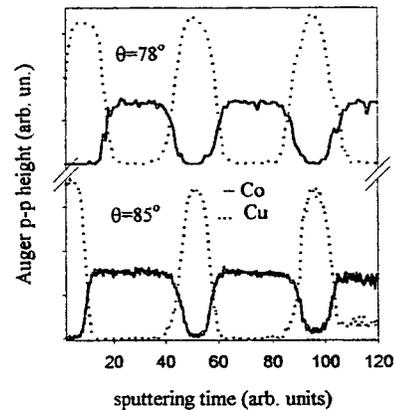


FIG. 2. As measured AES depth profiles (shifted to each other) of the Cu/Co multilayer system. Sputtering conditions: Ar⁺, 1 keV, angles of incidence 78° and 85°, and rotated specimen.

ning part of the profiles are not shown. The apparent widths of the Co and Cu layers are different; the Co layers seem to be thicker than the Cu ones just contrary to actual thicknesses of the layers. It is also evident that apparent thickness of the layers depends on the angle of incidence of the sputtering. The transition from the Co layer to the Cu is thicker (broader) than the transition from the Cu to Co layer.

For the detailed evaluation of the depth profile we should transform the sputtering time scale to depth and the Auger peak-to-peak heights to concentration. The latter can be easily done¹³ if we know the relative sensitivity factor, the backscattering factors, and the inelastic mean free paths (IMFP) for both elements. The relative sensitivity factor is the ratio of the Auger peak heights measured in pure Cu and Co. The backscattering factors are calculated from the expression of Shimizu,¹⁴ while IMFP values [1.14 and 1.54 nm for Co (656 eV) and Cu (920 eV)] are taken from the NIST database.¹⁵

To transform the sputtering time to depth we suppose that the sputter rate is proportional to the local concentration.¹⁶ In this case the erosion rate can be given by two parameters; e.g., the sputtering rate of Co, Y_{Co} , and the relative sputter rate $Y = Y_{Cu}/Y_{Co}$. The ratio of the thickness of the layers depends only on Y , while their absolute values on Y_{Co} and Y . Four sputtering times (to remove a given layer) could be determined from each experiment (the results for the first and last layers were ignored because of experimental problems). Knowing the actual layer thickness from the RBS measurement Y and Y_{Co} were determined for all sputtering conditions.¹⁷ Y values are shown in Fig. 3.

Figure 4 shows part of the AES depth profile (the region around the second Cu layer) recorded at 85° angle of incidence (lower curve in Fig. 2) after the coordinate transformation was carried out. To characterize the transitions the depth resolution, which is the distance of the points belonging to 84% and 16% concentrations,¹⁸ was calculated. The depth resolution has physical meaning only at that case if the interface broadening is a Gaussian type. To check the validity of the assumption we fitted two error functions to the measured curve shown in Fig. 3. In the case of the Co/Cu transition the agreement is quite good while in the case of the

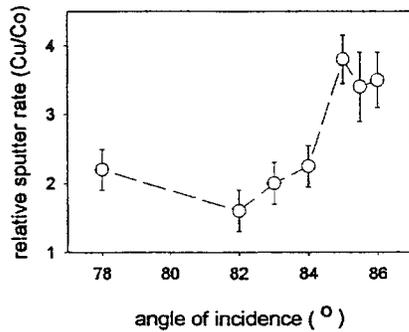


FIG. 3. Relative sputter rate of Cu/Co, Y , as a function of the angle of incidence. Sputtering conditions: Ar⁺, 1 keV, and rotated specimen. The points are connected to lead the eyes.

Cu/Co transition the fitting is poor. This means that the transition between the layers cannot be described by a simple Gaussian broadening. To cope with this problem we fitted a linear function to the transition to characterize it as well. Obviously both quantities depend on the applied algorithm of the transformation of the axes. As an example Table I shows the depth resolution values and the corresponding slopes measured on the depth profile of ion energy 1 keV and angle of incidence 84°.

IV. DISCUSSION

Considering Table I we make some observations. First the slope and depth resolution values show similar trends. Both show that the transition from Cu to Co is steeper than from Co to Cu. Second, in the case of Cu/Co transitions, both values (slope and depth resolution) convincingly show that the interface broadening increases (depth resolution increases, slope decreases) with depth. In the case of the Co/Cu transition, however, the interface broadening only slightly increases with depth.

The depth resolution and slope values were determined for all angles of incidences (78°, 82°, 83°, 84°, 85°, 85.5°, and 86°). The trends were similar to those demonstrated with the help of Table I, but the actual values depended in some

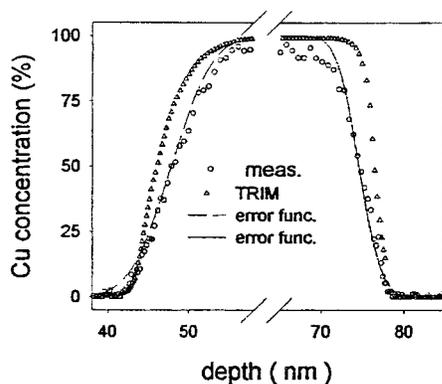


FIG. 4. AES depth profile after transformation of axes (circles) of the second Cu layer and the corresponding simulated profile (triangles). Angle of incidence in both cases is 85°. Full and dotted lines show the error functions, fitted to the measured data.

TABLE I. Depth resolution and slope values determined after transformation of the axes in the case of sputtering conditions of projectile Ar⁺, energy 1 keV, and angle of incidence 82°.

| | Depth resolution (nm) | Slopes (%/nm) |
|--------|-----------------------|---------------|
| 1Cu-Co | 2.75 | -16.98 |
| 2Cu-Co | 3.9 | -14.65 |
| 3Cu-Co | | -13.29 |
| 1Co-Cu | 6.1 | 8.53 |
| 2Co-Cu | 6.4 | 8.42 |

not understood way on the angle of incidence. Additional difficulty is that the broadening of the interface depends on the depth. Avoiding the problems connected to this dependence in the following we will compare data belonging to the same transitions. The transitions will be signed according to the number of layer (counted from the surface) they belong to. For example, 2Cu/Co stands for the transition from the 2 Cu layer to Co. If we compare the same transitions as a function of angle of incidence the picture is not much clearer. On the other hand a clear correlation appears if we show the results as a function of the relative sputtering rate. This is demonstrated in Fig. 5; it shows the slopes determined in transitions 1Co/Cu and 2Cu/Co. Two different behaviors can be observed. In the case of the Co/Cu transition the broadening increases (the slope decreases) with the increase of the relative sputter rate. On the other hand for the Cu/Co transition, weak, if any, dependence on the relative sputter rate is found. Though the actual values of the slopes depend on the transformation of the axes (the accuracy of which strongly depends on the accuracy of the RBS measurement that provides the thickness of the layer) the ratios are independent of the transformation.

The strongly asymmetric behavior observed correlates with the relative sputtering rate, thus any explanation should be based on the difference of the sputter rates of Co and Cu in this angular range. The two most important processes causing interface broadening are ion induced roughening and mixing. We will check how these processes are affected by the large difference of the sputtering rates.

First we deal with the effect of interface roughening. Let us suppose that the instantaneous surface proceeds from the

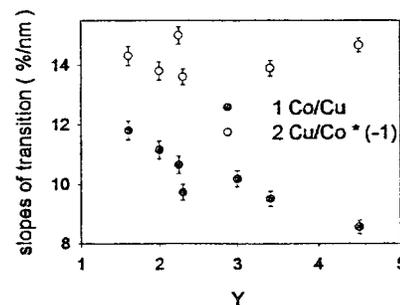


FIG. 5. Slopes determined on the 1 Co/Cu and 2 Cu/Co transitions as a function of the relative sputter rate. The slopes measured on the Cu/Co interfaces are negative; for the sake of better demonstration they were multiplied by -1 .

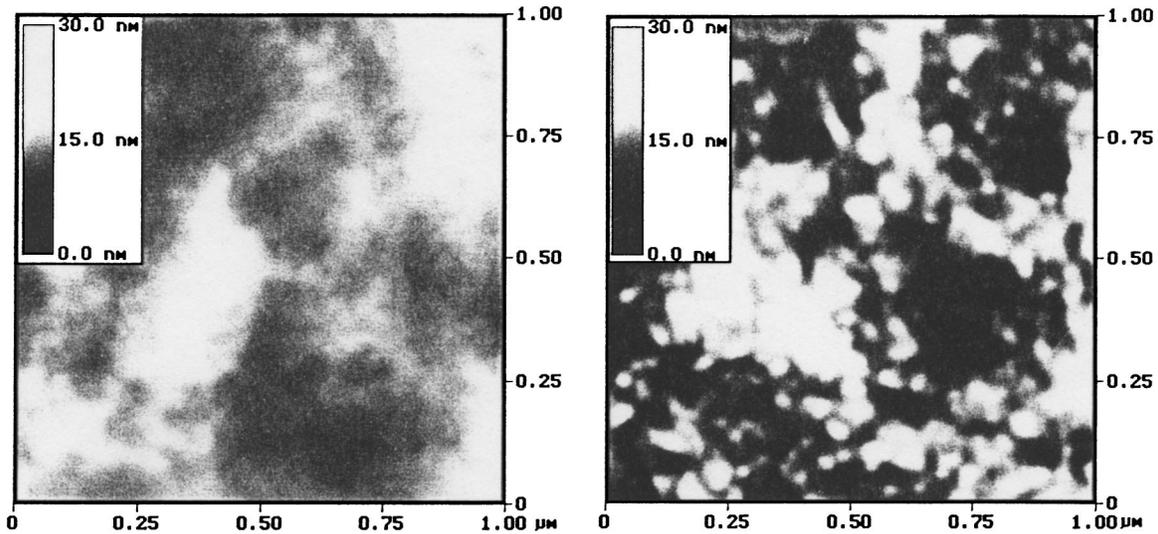


FIG. 6. AFM images of surface at 50% composition of (a) 1Co/Cu and (b) 2Cu/Co transitions. The scanned areas are $1 \times 1 \mu\text{m}^2$.

first Co layer to the second Cu one. We might suppose that due to several reasons (polycrystalline layer, rough initial surface, presence of impurities on the surface, etc.) the instantaneous surface is somewhat rough. Approaching the Cu layer first only on some patches of the instantaneous surface appears Cu. The removal rate of the Cu regions is higher than that of regions still covered by Co. Consequently the surface roughness increases. The higher the relative sputter rate, the higher the surface roughening. Though sample rotation and grazing angle of incidence cause smoothing by means of step movements,^{3,11} they cannot compensate this process completely. If all the cobalt is removed, that is, when the pure copper region is reached due to the grazing angle of incidence bombardment and rotated specimen, the step movements are active and the surface gets smoother, but not necessarily completely flat. Thus the instantaneous surface is smoother when it reaches the Cu/Co interface than that when it reached the pure copper region. Additional smoothing occurs entering the next cobalt layer. The sputtering rate of Co is lower than that of Cu, thus the “fell back” parts can “catch up.” Thus in the Cu/Co transition the smoothing continues resulting in a steeper transition than that in the Co/Cu one. In the pure Co layer further smoothing occurs. Since the extent of the smoothing depends on the removed layer thickness, complete healing can only be expected if the layer thicknesses are rather large.³ If this condition is not met (present case) the process is self-accelerating resulting in degradation of the depth profile, which was also observed as depth-dependent depth resolution.

The validity of the above speculation can be easily checked by measuring the roughness of the instantaneous surface in the transition layers. For this measurement we prepared two surfaces by terminating the depth profiling (angle of incidence 85°) when the concentration was 50% in the 1Co/Cu and 2Cu/Co transitions, respectively. The surfaces were measured *ex situ* by an atomic force microscope (AFM; DI Nanoscope E) in contact mode using a Si tip with

radius of curvature of 10 nm. The images were recorded with a resolution of 512 points/line at 1 to 2 Hz scan rate. The roughness was given by root mean square (rms) of the average of height deviations taken from the mean data plane. Figures 6(a) and 6(b) show the AFM images of surfaces of Co/Cu and Cu/Co transitions, respectively. It is clear without any additional evaluation that the surface belonging to the Co/Cu transition is rougher than that belonging to the Cu/Co transition. Several rms values were calculated for the surfaces by placing $500 \times 500 \text{ nm}^2$ areas on various spots. The average of these measurements yielded 4.1, and 2.1 nm rms for the Co/Cu and Cu/Co transitions, respectively.

We should also consider the effect of ion mixing, since the altered layer composition and thus its thickness depends on the relative sputtering rate. To study the ion mixing we apply our standard method.¹⁰ We started with the structure determined by RBS and simulated the sputtering by means of a dynamic TRIM code.¹⁹ This calculation provided us with in-depth distribution along the sputtering from which the simulated depth profile can be derived. The main problem with the applied code is that by using its default parameters it cannot reproduce the relative sputter rate observed in the experiment. In Ref. 17 we showed that by reducing the surface binding energy of copper to 1.52 eV (which is unrealistic from a physical point of view) the sputtering rate versus angle of incidence curve was similar to the measured one. Thus we simulated the depth profile by using 1.52 eV for the copper surface binding energy while all other input parameters were the default ones. Figure 4 shows the result (triangles). It is clear that the TRIM also predicts an unexpected large and asymmetric broadening of the Cu/Co and Co/Cu interfaces. The depth resolutions are 6.1 and 2.3 nm for the Co/Cu and Cu/Co transitions, respectively.

It is obvious that even the unexpectedly high interface broadening is less than the experimentally found values. We must, however, consider the combined effect of the ion mixing and roughening. Assuming that the two processes are

independent we can utilize that the summation rule of the independent probabilities.¹⁶ According to this rule the square of the individual broadening should be added and the square root of the sum is the resulting broadening. For the present case we get 7.4 and 3.3 nm for the 1Co/Cu and 2Cu/Co transitions, respectively. These values agree reasonably well with the ones, 8.5 and 5.5 nm for the 1Co/Cu and 2Cu/Co transitions, experimentally found.

The sputtering conditions applied in this study were introduced to eliminate the sputtering induced roughening. In many cases this expectation is fulfilled and one gets depth independent depth resolution even after removing hundreds of nanometers of material, which drastically improved the capability of AES depth profiling.^{8–10} In the present case, however, applying grazing angle of incidence sputtering induced roughening and enhanced ion mixing occurs because of the high relative sputter rate, resulting in extremely poor depth resolution. Thus for the Co/Cu system depth profiling applying grazing angle of incidence, e.g., 85°, is not recommended. Rather one should use an angle of incidence where the relative sputter rate is minimal, e.g. 78°.

The most important conclusion of this work is that one should consider, in planning a depth profiling study, the relative sputtering rates of the components involved. Unfortunately data of sputter rates at grazing angle of incidence are rare.

This study also emphasizes that the interface broadening is a complex process depending on, besides the sputtering condition (projectile, energy, and angle of incidence), the relative sputter rate as well. It is also evident that the broadening of the interface might be different for the A/B and B/A interface. It follows that the term depth resolution used in sputter depth profiling techniques has a limited applicability as it has been pointed out earlier.¹⁰

V. CONCLUSION

A Co–Cu multilayer system was depth profiled by 1 keV Ar⁺ ions varying the angle of incidence in the range of 78°–86°. The specimen was rotated during ion sputtering. Using these sputtering conditions we have found:

- (1) the relative sputter rate of Cu to Co strongly depends on the angle of incidence;
- (2) the broadening of the Co/Cu interface is much stronger than that of the Cu/Co interface; and

- (3) the interface broadening of the Co/Cu interface increases with the increase of the relative sputter rate.

These phenomena can be explained by ion bombardment induced interface roughening and mixing. Both processes cause different broadening on the Co/Cu and Cu/Co interfaces. The difference of the ion induced roughening on the two interfaces was verified by direct AFM measurement.

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