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Effects of Y or Gd Addition on the Structure of Al Thin Films

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The addition of Y or Gd to Al remarkably decreased a grain size of Al matrix and suppressed growth of hillocks at high temperatures (350 ℃ - 450 ℃), in association with large number of segregation of metallic compounds mostly at grain boundaries. Their resistivities after annealed at above temperatures showed very low values less than 5 μΩcm.

I. Introduction

Sputter-deposited aluminum thin films are widely used as interconnector lines in very large-scale integration (VLSI) circuits or as gate and source electrode bus-lines in thin- film-transistor-liquid-crystal-display (TFT-LCD) arrays . However, since the feature size of the conductor line width is being greatly reduced in advanced devices, the microstructure of thin-film conductors is of vital importance to their reliability. Y. K. Lee et al. reported that Al-Y alloy prepared by evaporation shows relatively high resistance to growth of hillocks, but that this resistance becomes low after annealing at high temperature [1]. In this report, we study in detail the effects on the structure of Al of adding Y or Gd in order to control its microstructure and thus to inhibit growth of hillocks and to obtain high conductivity.

II. Experimental

Al-Y and Al-Gd alloy films about 400 nm thick were deposited on a 7059 glass substrate by using a DC magnetron sputtering apparatus at an Ar pressure of 0.7 Pa. The composition of the films was varied by using a composite target consisting of 100 mm Al disks (4N) and 5 x 5 x 1 mm^3 Y or Gd chips (3N) . The film composition was determined by Inductively Coupled plasma (ICP) spectroscopy. The film's electrical resistance was measured by using a conventional four-point probe at room temperature. Its reflectivity was measured at room temperature for a wavelength of 780 nm. The film structures were examined by using a X-ray diffractometer and a transmission electron microscope (TEM) operated at 300 kV, respectively.

III. Results and Discussion

X-ray diffraction measurements showed that the growth of the Al (111) plane was largely suppressed by adding only a small amount of Y or Gd (less than 0.5 at%) to pure Al and the diffraction intensity of their main peaks tends to increase again with futher increase of adding elements. These are shown representatively for Al-Gd alloy thin films in Fig.1. Though the solid solubility of Y or Gd in Al is very low (less than about 0.03 at%),

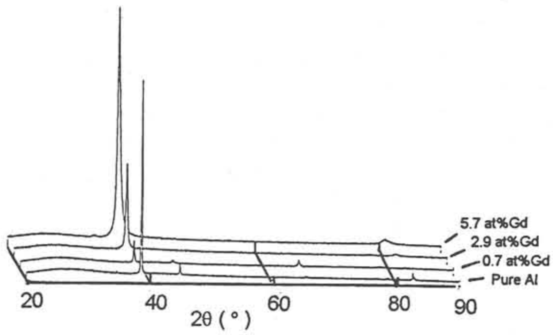

Fig. 1 X-ray diffraction profiles for Al-Gd alloy thin films as a function of Gd content.

these as-made alloy films cotaining about 3 at% of added elements show very fine grains less than 100 nm in size, and their structure is in the mixed state of a highly supersaturated solid solution of the Al phase and very fine metallic compounds , such as Al_2X or Al_3X (where X denotes Y or Gd). The representative TEM observations of as-deposited $Al_{96.5}Y_{3.5}$ and $Al_{97}Gd_3$ alloy thin films are exhibited in Fig.2. The corresponding energy dispersive X-ray

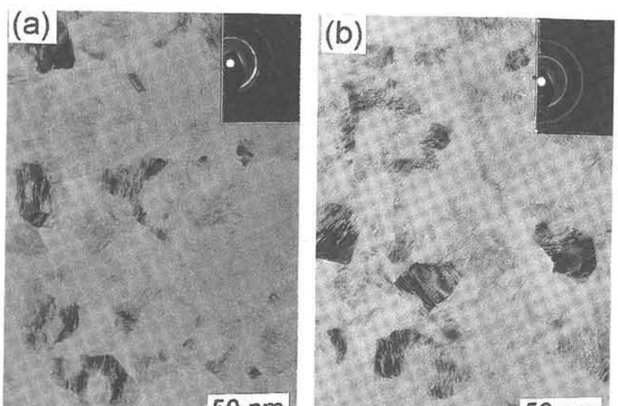

Fig. 2 Bright field images and diffraction patterns of as deposited (a) $Al_{96.5}Y_{3.5}$ and (b) $Al_{97}Gd_3$ alloy thin films.

analysis (EDX) of TEM samples in Fig.1 revealed that adding Y or Gd elements homogeneously distributed in Al matrix. The Al-Y and Al-Gd alloy films (with about 3 at% of added elements) were isochronally annealed in a vacuum (less than 10^{-4} Pa.) at each temperature for 30 min. The changes in their resistivity and reflectivity are shown as functions of the annealing temperature in Fig.3. Note that even though their resistivities as made are very high-more than 20 μΩcm-they significantly decrease above 300℃, and approach the values

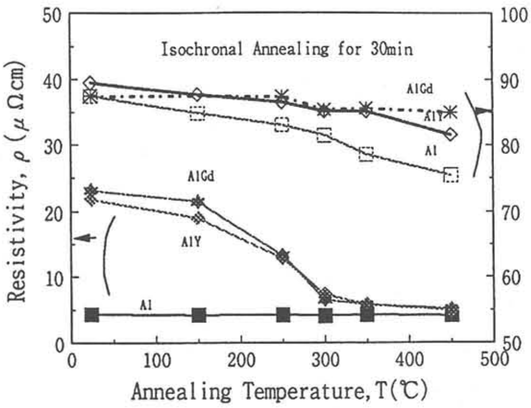

Fig.3 The change of resistivity, ρ and reflectivity, R of Al alloys after annealed isochronally for 30 min.

for pure Al at 450℃. On the other hand, the reflectivity changes much less markedly than the resistivity. Figure 4 shows the change in the resistivity of samples with the same composition as shown in Fig.3 when they are isothermally annealed

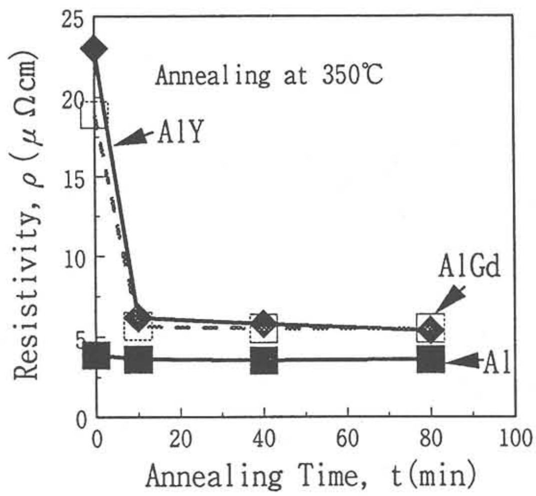

Fig.4 Change in resistivity ρ of Al alloys after isothermal annealing at 350℃.

in a vacuum at 350℃. It should be noticed that their resistivities decrease largely down to low values of about 5 μΩcm within a very short annealing time, and almost saturate on further annealing. Both X-ray diffraction and TEM observation revealed that fine metallic compounds of Al_3Y and Al_3Gd in Al-Y and Al-Gd alloy systems were respectively segregated in an Al matrix (mostly at the grain boundaries of Al), as representatively shown in Figs. 5 and 6 respectively.

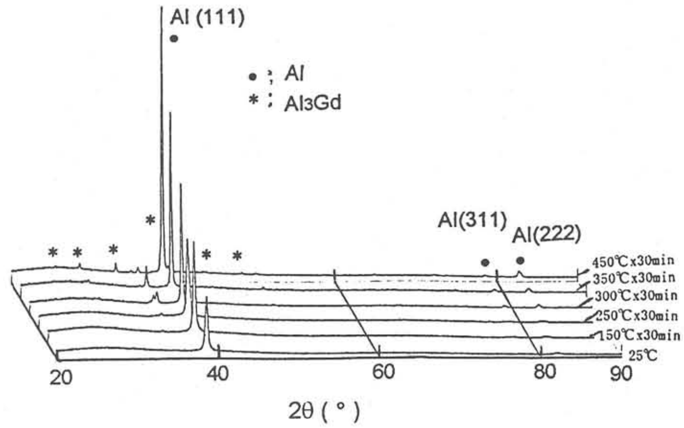

Fig. 5 X-ray diffraction profiles of $Al_{97}Gd_3$ alloy thin films isochronally annealed up to 450 ℃

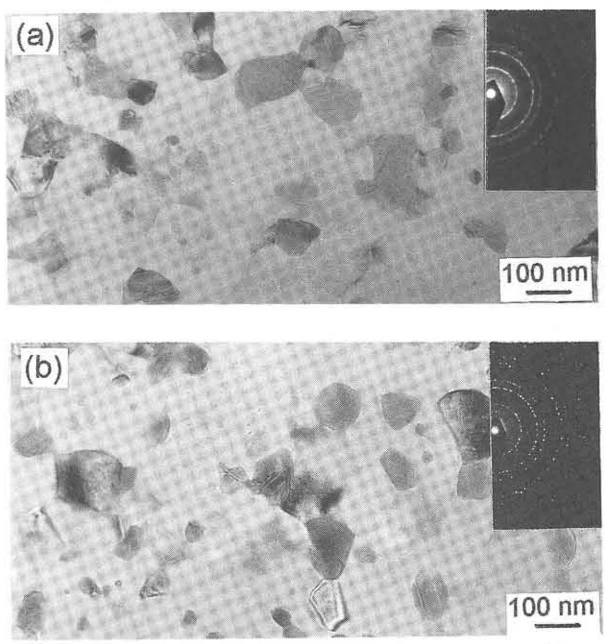

Fig. 6 Bright field images and diffraction patterns of (a) $Al_{96.5}Y_{3.5}$ and (b) $Al_{97}Gd_3$ thin films annealed at 350 ℃ for 80 min.

Both EDX and selective area diffraction analyses verified that relatively dark fine grains in Fig. 6 were Al-Y or Al-Gd metallic compounds. Furthermore, their mean grain sizes were very small-less than 200 nm-in comparison with that of pure Al (more than 500 nm). Surprisingly, no growth of hillocks was observed on the surface of the above AlY and AlGd alloy film samples after annealing at 450℃, nor after annealing at 350℃ for 80 min. These representative results are shown in Fig.7, where those of Al-Zr and Al-Nb alloy thin film samples with the same amounts of added elements are also shown for comparison. In the case of Al-Zr and AlNb samples, defects of hillocks and whiskers (white spots and lines respectively in Fig. 7) started to form on their film surfaces at 250℃.

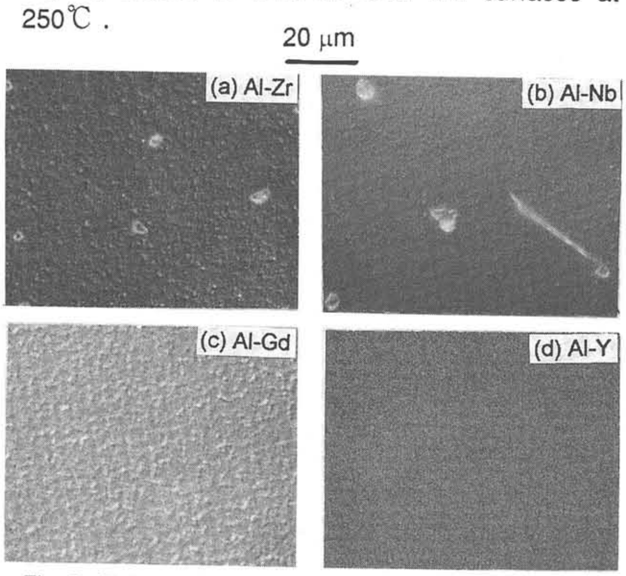

Fig. 7 Polarized optical micrographs of Al alloy thin film surfaces annealed at 450 ℃ for 30 min.

Figure 8 shows the change in both the resistivity and reflectivity of samples with the same alloy composition as those in Figs.3 and 4 as a function of the substrate temperature during sputtering. In contrast to the trend in Fig.3, reflectivity abruptly decreases at a substrate temperature of 350 ℃, in association with the growth of large grains. Note that no growth of hillocks is observed on film surfaces of any samples in Fig. 8.

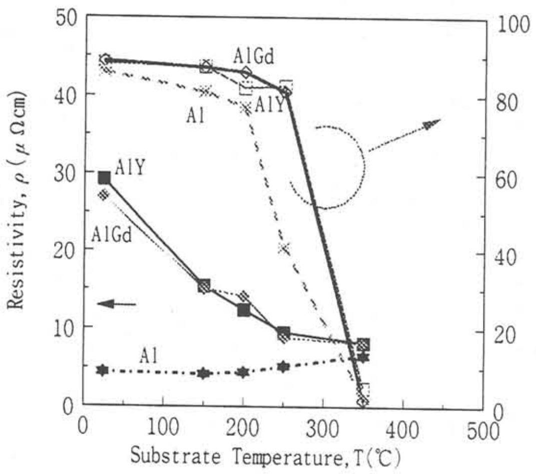

Fig.8 The change of resistivity, ρ of Al-X alloys (X=Y, Gd) with substrate temperatures, Tsub

It is worth mentioning that the above results of low resistivity after annealed at high temperature and high thermal stability without no hillock growth are also generally true for other Al-rare-earth-transition alloy films (Al-RE, RE = La,Pr,Nd,Sm,Tb, Dy, Ho, and Er). The representative isochronal annealing curves of resistivities for these alloy thin films (containing about 3 at% of added elements) are shown in Figs 9 and 10. Note that all high values of resistivities of as-made samples largely decrease above 350 ℃,and approach the values for pure Al.

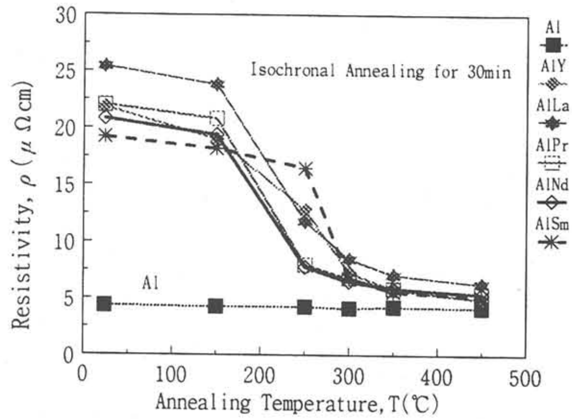

Fig.9 The change of resistivity, ρ of Al alloys after annealed isochronally for 30 min.

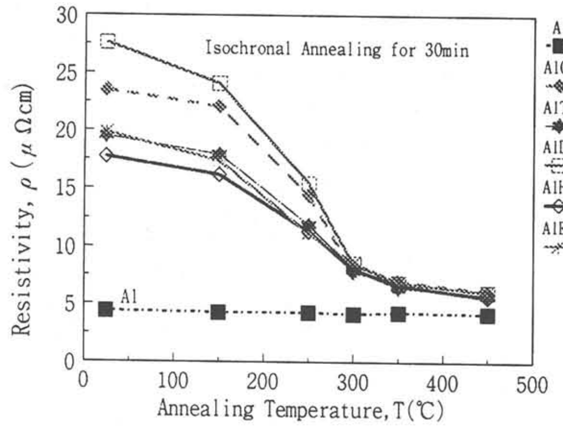

Fig.10 The change of resistivity, ρ of Al alloys after annealed isochronally for 30 min.

IV. Summary

Al-rare-earth-transition alloy films (Al-RE, RE = T, La, Pr, Nd, Sm, Gd, Tb, Dy, Ho, and Er) showed very low resistivity less than 5 μΩcm after annealed at more than 350 ℃, associating with a large number of segregation of metallic compounds mostly at grain boundaries. They also showed high thermal stability without growth of hillocks at high temperatures in comparison with other Al alloys, such as Al-Zr and Al-Nb thin films.

References

(1) Y. K. Lee, N. Fujimura, and T. Ito, J. Vac. Sci. Technol. B9, (1991) 2542.