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A transmission electron microscopy study on the crystal structure and atomic arrangement of Al–Nd thin films deposited on glass substrates

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ABSTRACT

The crystal structure and atomic arrangement of Al–2 at.% Nd thin films were investigated by a transmission electron microscopy (TEM) study. As a result, we have revealed that Nd atoms exist in Al matrix as solid solutions in the state of as-deposited and annealed at low temperature, while the intermetallic compounds with the composition of Al_4Nd are precipitated as the annealing temperature increased above 300 °C. The precipitated Al_4Nd has a tetragonal structure with nine cyclic layers, and the atomic arrangement was identified by the fast Fourier transform (FFT) and Fourier filtered images as well as the experimental high-resolution transmission electron microscopy (HRTEM) images. Consequently, Nd atoms, forms of solid solutions or precipitates, effectively suppress the grain growth of Al.

1. Introduction

Active-matrix liquid crystal displays (AMLCDs), especially thin film transistors (TFTs)-LCDs, have been surprisingly developed in the last decade. As the panel size and the resolution of TFT-LCDs have been increased, interconnections which required low resistivity in TFT panels have been widely investigated [1]. Aluminum (Al), as a metal electrode, has merits of low resistivity and low cost, however the formation of hillock which may lead to device failure during thermal processes is a serious disadvantage. Several researches on the Al alloy which the transition metals (Cr, Fe, Co, or Ni) or rare-earth elements (Y, Gd, or Nd) were added to pure Al were done to inhibit the hillock formation. Recently it was reported that the Al–(2–6 at.%) Nd alloy thin films have an excellent hillock resistance [2–5]. Arai et al. practically showed that both gate and data bus lines applied by Mo-capped Al–2 at.% Nd (neodymium) alloy could have excellent physical properties such as step coverage, hillock suppression, patternability, and mechanical strength [6].

With respect to the composition and crystal structure of Al–Nd intermetallic compounds, it was known to form an $Al_{11}Nd_3$, the Al-rich compound. While the $\alpha Al_{11}Nd_3$ phase has orthorhombic structure, the $\beta Al_{11}Nd_3$ phase has tetragonal structure of the Al-deficient Al_4Ba-type [7]. Onishi et al. reported that the tetragonal structured Al_4Nd having the Al_4Ba-prototype was precipitated during annealing process in Al–(2–6 at.%) Nd thin films. They inferred the excellent hillock resistance was caused by the stress relief due to the precipitation of Al_4Nd [5]. Apart from the formation of Al–Nd intermetallic compounds in Al-rich compositions, Kale et al. reported that Nd is soluble in Al with the solubility of about 2 at.% and suggested few modifications pertaining to Al-rich side of Al–Nd phase diagram [8].

A transmission electron microscopy (TEM) is a very useful analytical method not only for the microstructural characterization of materials but also for the failure analysis of LCD panels together with time-of-flight secondary ion mass spectrometry (TOF-SIMS), X-ray photoelectron spectroscopy (XPS), and so on [9,10]. Even though the Al–Nd thin films are currently used in

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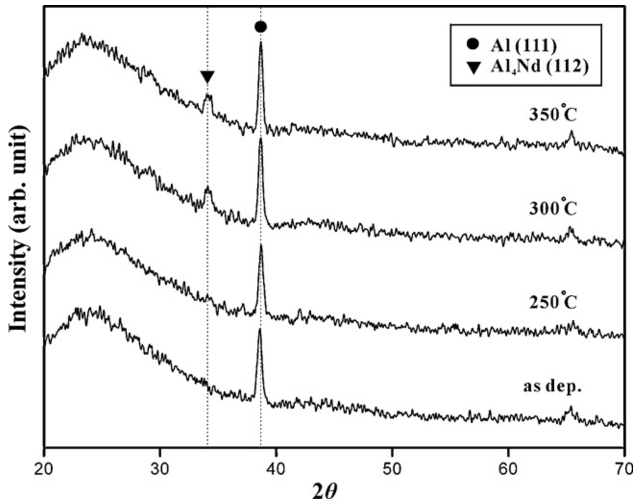

Fig. 1. XRD patterns of Al–2 at.% Nd thin films with the variation of annealing temperature. (●) The FCC and (▼) the peak conforming to the tetragonal structured Al_4Nd.

TFT-LCD as metal electrodes, researches on the microstructural characterization of these materials were somewhat uncommon. Especially, a high-resolution transmission electron microscopy (HRTEM) study on an atomic level for the investigation of the crystal structure and atomic arrangement of the Al–Nd thin films has never been reported yet.

We report the microstructural properties of the Al–2 at.% Nd thin films deposited on the glass substrates by sputtering method. The Al–Nd thin films were annealed and investigated by the HRTEM in order to reveal exactly the crystal structure and atomic arrangement on an atomic level.

2. Experiments

The Al–Nd thin films with the thickness of 200 nm were deposited on glass substrates, with the deposition rate of 8 nm/s, by sputtering a single target (2 at.% Nd) in Ar gas with the DC power and the gas pressure set to 30 kW and 0.25 Pa, respectively. A thermal annealing process was carried out under N_2 ambient conditions at 250, 300 and 350 °C for 10 min.

The crystal structure of overall Al–Nd thin films was determined by X-ray diffraction (XRD) (Rigaku-D/MAX-IIIC). The annealed Al–Nd thin films were analyzed by TEM to investigate the crystal structure and atomic arrangement of the Al–Nd intermetallic compound. The cross-sectional TEM specimen was prepared by a focused ion beam (FIB) method using Ga ions and followed by an ion milling with Ar ions. A FIB method was mainly done throughout the specimen preparation and the Ar ion milling process was conducted to remove the amorphized surface layer damaged by Ga ion beam, because the unavoidable and undesirable amorphization due to the surface damage by Ga ion may affect the HRTEM images. For a cross-sectional TEM specimen, bright field (BF) TEM images, selected area electron diffraction (SAED) patterns, high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) images, and the HRTEM images were obtained with FE-TEM (FEI, Tecnai G2 F30 S-Twin) operated at 300 kV with a high-resolution pole piece. The fast Fourier transform (FFT) and Fourier filtered HRTEM images were obtained with a Gatan Digital Micrograph (version 3.7.4).

3. Results and discussion

According to the binary phase diagram of Al–Nd alloy, the stable phases of Al–2 at.% Nd are Al and $Al_{11}Nd_3$ in their thermal equilibrium, and the ratio of each phase can be calculated with a lever rule in the phase diagram. As a metallurgical theory, the microstructure of Al–2 at.% Nd thin films can be constructed with Al matrix and Al–Nd intermetallic compounds. In general, Al–Nd thin films deposited by sputtering method consist of poly-crystalline including some defects such as vacancies. The expected intermetallic compound is not the orthorhombic structured $\alpha Al_{11}Nd_3$ phase, but the $\beta Al_{11}Nd_3$ phase, the tetragonal structure of the Al-deficient Al_4Ba-type. Considering the previous literature reported by Onishi et al., the existence of tetragonal structured Al_4Nd is expectable and reliable [5].

XRD experiments were conducted to identify the variation of the crystal structure of Al–2 at.% Nd thin films as the annealing temperature increased. Results were shown in Fig. 1. Only the Al

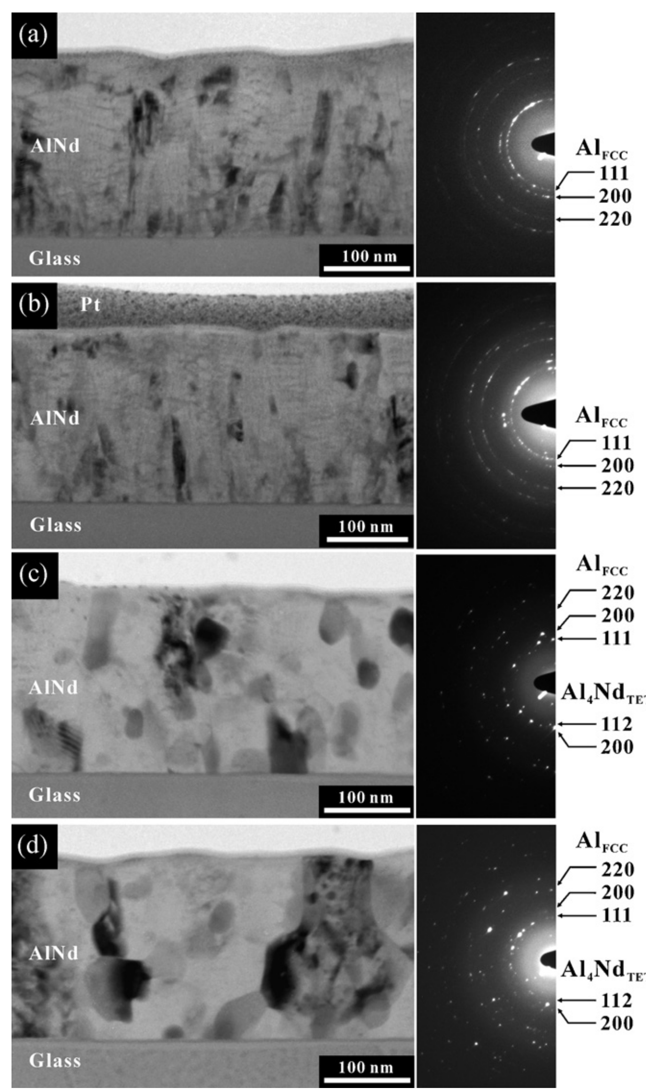

Fig. 2. Cross-sectional BF-TEM images and SAED patterns of Al–2 at.% Nd thin films annealed with various temperature for 10 min. (a) as-deposited, (b) 250 °C, (c) 300 °C, and (d) 350 °C.