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TEM observation of liquid-phase bonded aluminum–silicon/aluminum nitride hetero interface

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Abstract Liquid-phase bonded aluminum–silicon/aluminum nitride interface structure was investigated using high-resolution transmission electron microscopy. A textured layer of aluminum formed a stable orientation relationship with aluminum nitride, which showed Al(111) to be tilted by about 4° with respect to the AlN(0001) interface plane. The unique orientation relationship between Al and AlN was predicted as one of the stable orientation relationships using coincidence of reciprocal lattice point method, which surveys the degree of geometrical coherency between two crystals in three-dimensional space. A stable orientation relationship was found to be $(001)[1\bar{1}0]\mathrm{Al}//(2\bar{2}03)[11\bar{2}0]\mathrm{AlN}$.

Introduction

Aluminum nitride (AlN) is considered as a high performance dielectric substrate in laser diodes, semiconductor modules, and power electronics. In the development of new packaging technologies for improved heat dissipation in integrated circuits (IC), AlN ceramic substrate has the advantages of high thermal conductivity, coefficient of thermal expansion (CTE) near that of silicon, and non-toxic nature [1].

Joining of AlN ceramics to metals can be accomplished with brazing technique, which makes use of metal interlayers. Brazing process joins two materials with a filler metal in between the parent materials. The joint is heated at a temperature above the melting point of the filler metal and below that of the base materials [2]. In power electronics industry, Al–silicon (Si) interlayer is typically used to decrease the bonding temperature for direct brazing aluminum (DBA) with AlN substrate. Al–Si filler is also used to significantly increase the bond strength based on a study of some metal/ceramic joints but still without a detailed understanding on the bonding mechanism [3]. Previous studies have shown that some fixed orientation relationships (OR) between metal crystals and ceramic crystals are used to estimate interface adhesion and strength theoretically. In the case of Al/Al_2O_3, Ag/Al_2O_3 and Cu/Al_2O_3 interfaces, the common orientation of (111)FCC metal//(0001)Al_2O_3 was used to estimate theoretical adhesive energies, and these values were found to agree with the experimental measurements [4, 5]. A similar orientation of (111)Al//(0001)AlN was considered as the stable interface orientation where the ideal strength of Al/AlN model was calculated by uniaxial tensile simulation [6]. However, the previous studies lack experimental evidences for the stable orientation relationships on the Al/AlN interface. Since the orientation relationships may directly influence the mechanical and functional properties of the Al/AlN interface, it is important to experimentally determine the stable orientation relationships at the interface using selected process conditions.

In the present study, high-resolution transmission electron microscopy (HRTEM) was used to investigate the interface structure and stable orientation relationship of Al–Si/AlN interface formed by liquid-phase bonding method. Theoretical evaluation of stable crystallographic orientation relationships has been performed based on the concept of geometrical coherency between the two dissimilar crystals.

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Experimental procedure

A “sandwiched” structure of α-Al_2O_3(0001)/AlN(0001)/Al–10mass%Si/AlN(0001)/α-Al_2O_3(0001) was fabricated in vacuum ($\sim 10^{-5}$Pa) using a hot-press process with a constant load of 0.5 MPa. A 20-μm thick Al–Si foil with less than 0.01% impurity was used in this experiment. A commercially available epitaxial AlN/α-Al_2O_3(0001) (from DOWA Electronics Materials Co. Ltd.) was used as a starting ceramic substrate. AlN film is deposited by metal–organic chemical vapor deposition (MOCVD) on sapphire (α-Al_2O_3)(0001) substrate. The AlN film thickness is typically 1 μm and is relatively flat. The average roughness is about 0.2 nm with impurities of H, C, O, and Si at about $\times 10^{-16}$ or less. The orientation relationship is AlN(0001)//Al_2O_3(0001), $[11\bar{2}0]$AlN//$[1\bar{1}00]$$Al_2O_3$, which is similar to previous results of thin film deposition of AlN on Al_2O_3(0001) by pulsed laser deposition (PLD), molecular beam epitaxy (MBE), and MOCVD [7, 8]. Figure 1 shows the schematic of the sample fabricated in this study. The sandwiched structure was fabricated at a bonding temperature of 646 °C, which is above the Al–Si eutectic temperature (577 °C), for 8 h. The sandwiched structure was cooled down at a rate of −5 °C/min., which is within the range of what is commonly applied to liquid-phase bonding of metal/ceramic interfaces for heat sink structures in the power electronics industry.

Cross-sectional TEM samples were prepared by a standard procedure, using mechanical grinding followed by ion thinning method. A low angle ion milling and polishing system (Fischione 1010) was used to perforate a hole in the Al–Si/AlN interface. The argon beam angle, power and current were controlled together with the sample stage rotation of ±45° to preserve the thin metal from being sputtered away ahead of the ceramic at the interface. The cross-sectional interface structure observations and diffraction analyses were performed using TEMs (JEOL JEM-4010, JEM-2010) operated at 400 and 200 kV, respectively. Compositional analyses of the interfaces were performed by TEM energy dispersive X-ray (EDX) analysis.

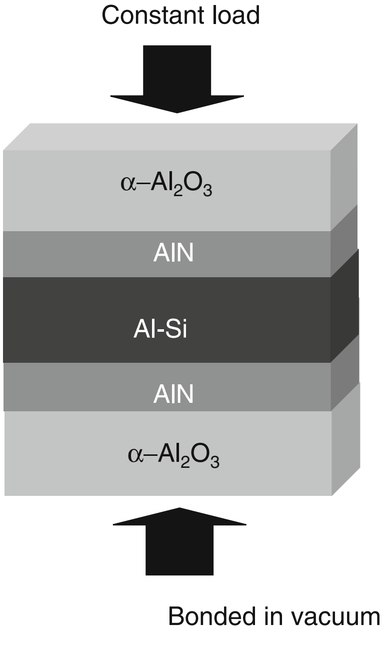


Fig. 1 Schematic of α-Al_2O_3(0001)/AlN/Al–Si/AlN/α-Al_2O_3(0001) sandwiched sample

Results and discussion

TEM observation of Al–Si/AlN interface

During liquid-phase bonding, the Al–Si foil becomes a one-phase region and wets the AlN substrate. Upon cooling, Al from the bulk hypoeutectic Al–Si alloy solidifies followed by the crystallization of Si. As the temperature decreases trace amount of dissolved Si is precipitated out of solid Al such that the solubility of Si in Al decreases. After bonding, no epitaxial relationship was observed for Al–Si on AlN except for a textured layer of Al (with small amount of dissolved Si) on some areas at the interface as shown in the low magnification TEM image in Fig. 2a.

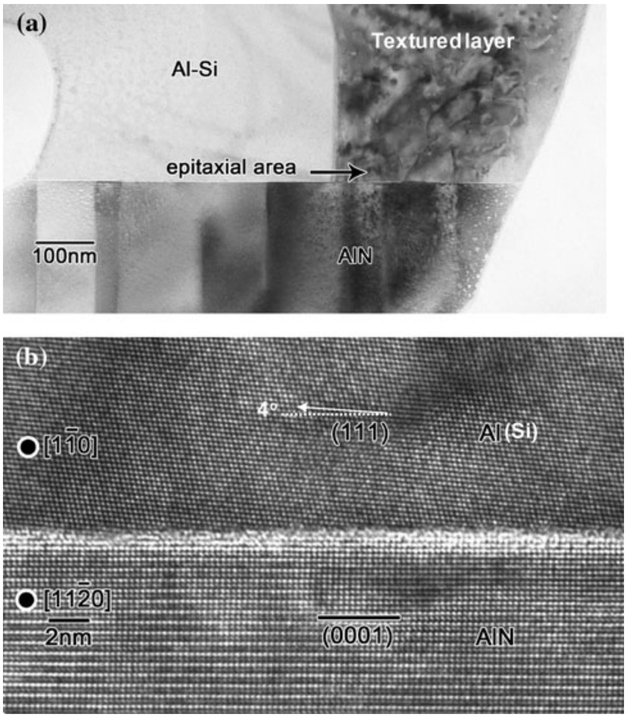


Fig. 2 **a** TEM image of textured layer of Al–Si/AlN interface. **b** HRTEM image of localized epitaxial area of Al (with trace amount of dissolved silicon), which shows the orientation relationship: $[1\bar{1}0]$Al//$[11\bar{2}0]$AlN, ~4° tilt (111)Al on (0001)AlN

It was found that the textured area has the OR of Al(111) plane being slightly tilted with respect to the AlN (0001). The tilting of Al(111) was also observed in the different textured areas, indicating that this tilt is reproducible. In Fig. 2b, the Al(111) lattice planes are tilted by approximately 4° as observed in the zone axes Al[10]//AlN[110] (Fig. 2b). On the contrary, it has been reported that no tilt in Al(111) on the AlN(0001) interface plane was observed using pulsed laser ablation technique nor plasma-alloy reaction [9, 10]. These results indicate that the stable OR should be dependent on the process conditions. In the HRTEM image of Fig. 2b, the interface appeared to be not very smooth. This is most likely due to atomic relaxation to accommodate the local lattice mismatch at the interface because of the inclined Al(111) plane. If Al(111) and AlN(0001) planes were exactly parallel then we may expect a sharp and abrupt interface. A similar contrast fluctuation of about 1 nm was also reported for Al/Al_2O_3 interface [11].

In this liquid-phase bonded Al–Si/AlN hetero interface, the Al textured area is expected to have dissolved silicon at a level of about 0.20 mass% after cooling an Al–10 mass%Si alloy based on the binary phase diagram. The TEM-EDS raw data show Si content of less than 1% on different areas of the aluminum matrix. Silicon segregation was observed at the interface in the textured and non-textured areas in the TEM-EDX analyses. Since silicon segregation was observed along the entire interface, silicon segregation within the textured area is likely to have no significant effect on the observed Al(111) tilt character on the interface plane. Previous study in liquid-phase bonded Al–Si/α-Al_2O_3 hetero interface showed that silicon segregates at the interface, but these interfaces show no preferential ORs between Al–Si and α-Al_2O_3 [12]. It has been shown through controlled wetting experiments that pure Al has higher adhesion on sintered AlN ($W_a = 1.421$ J/m^2, $\theta_F = 41°$ at 1100 °C) than on α-Al_2O_3 ($W_a = 0.950$ J/m^2, $\theta_F = 80°$ at 1100 °C) [13]. W_a is defined as the work of adhesion between Al and AlN while θ_F is the final contact angle, which was assumed to be equivalent to the intrinsic contact angle of the system according to the Young–Dupré equation. The higher work of adhesion and lower contact angle of Al on AlN suggest a stronger interaction of Al on AlN than on α-Al_2O_3. This may be one of the factors for the formation of preferential Al textured layer on Al–Si/AlN interfaces, which was not observed in the past study on liquid-phase bonded Al–Si/α-Al_2O_3 interfaces.

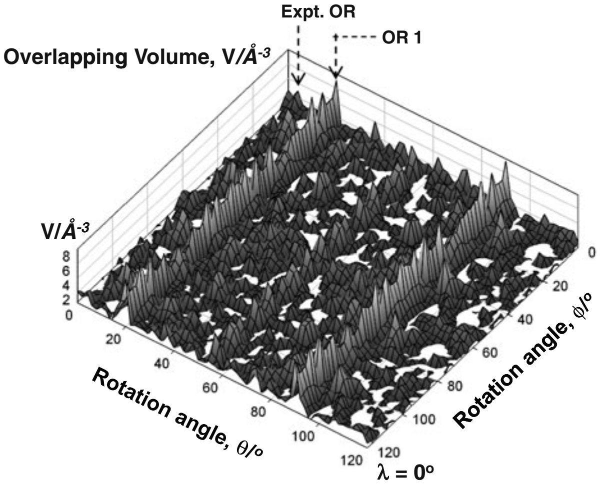
Coincidence of reciprocal lattice point calculation of Al/AlN hetero-system

Coincidence of reciprocal lattice point (CRLP) is an effective tool to predict the stable orientation relationships in hetero-systems based on three-dimensional geometrical coherency between two crystals [14–17]. The detailed calculation method is reported elsewhere [14, 15]. Calculations are performed in reciprocal space with each lattice point designated as a sphere. In the present study, the RLP radius was set at 10% of the reciprocal lattice parameter of each crystal and the maximum index planes were set at (333)Al and $(44\bar{8}8)$AlN. CRLP determines the preferentially stable orientation relationships using the maximum overlapping volume of the RLPs between two crystals. The Al reciprocal lattice is rotated inside the AlN reciprocal lattice around the rotation axes of [111], $[1\bar{1}0]$ and $[11\bar{2}]$ with the corresponding rotation angles ϕ, θ, and λ. The initial position of the lattices was set at (111), $(1\bar{1}0)$, and $(11\bar{2})$ planes of Al parallel to (0001), $(11\bar{2}0)$, and $(1\bar{1}00)$ planes of AlN.

Figure 3a shows the CRLP three-dimensional profile at the θ–ϕ rotation angles of the Al/AlN hetero system. The lower section of the three-dimensional profile was cut off to make the peaks appear more clearly. It shows the different peaks of the overlapping volume, which correspond to the preferentially stable ORs based on three-dimensional geometrical coherency. Considering the $[1\bar{1}0]$ rotational axis (θ-rotational angle), the most stable OR1 is described as: $(\bar{1}\bar{1}2)[1\bar{1}0]$Al//(0001)$[11\bar{2}0]$AlN and the experimentally observed OR as: Al (111) 3.7° tilt to AlN (0001) for Al/AlN hetero system around the same rotational axis of $[1\bar{1}0]$. The experimentally observed OR is calculated to be one of the preferentially stable secondary ORs in CRLP. The two-dimensional profile in Fig. 3b shows the cross-section of Fig. 3a with $\phi, \lambda = 0°$, which shows the experimentally observed OR at $\theta = 3.7°$. This secondary "expt. OR" peak is shown to have a higher stability than the case when the Al(111) is exactly parallel to the AlN (0001) ($\theta = 0°$). Equivalent orientation relationships for the Al(111) tilt to AlN(0001) and Al(111)//AlN(0001) can be found at $\theta = 105.7°$ and $\theta = 109.4°$, respectively because of the crystal symmetry.

A higher peak in the CRLP profile indicates the higher degree of parallelism of planes between two crystals in three-dimensional space. The unique orientation relationship of the experimental OR is calculated in CRLP as $(001)[1\bar{1}0]$Al//$(2\bar{2}03)[11\bar{2}0]$AlN. This suggests a potentially strong interaction between Al(001) and AlN$(2\bar{2}03)$ planes with the favorable high geometrical coherency in three-dimension. If the Al(111) plane were exactly parallel to the AlN(0001) plane, the Al(001) and AlN$(2\bar{2}03)$ planes would not be parallel as schematically drawn in Fig. 4. The angles in the schematics were exaggerated to emphasize the tilt angle and the corresponding parallel planes. The Al(111) tilt also favors the low interplanar misfit at the interface. The effect of the Al(111) tilt on the misfit parameter is calculated using the equation,

(a) CRLP Three-dimensional profile of overlapping volume



(b) Cross-sectional profile of overlapping volume

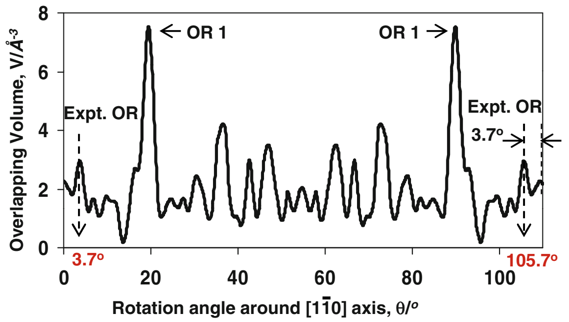


Fig. 3 **a** 3D and **b** 2D plots of the most stable OR1 and the experimentally observed OR between Al and AlN, which were predicted in CRLP calculations. A 3.7° rotation of Al(111) with respect to AlN(0001) is shown in (b) at $\theta = 3.7°$ and 105.7°

$\delta = [2(d_{AlN} - d_{Al})/(d_{Al} + d_{AlN})] \times 100\%$, where d is the interplanar spacing for the respective planes [15]. The interplanar misfit between Al{001} and AlN{$2\bar{2}03$} is about 4%. Here, the minimum misfit is considered such that three {$2\bar{2}03$} planes correspond to every {001} plane. In contrast, if Al(111) were exactly parallel to the (0001) interface plane then Al($11\bar{2}$) and AlN($1\bar{1}00$) would also be parallel with a larger misfit of 8% for 3 × {$11\bar{2}$}Al// 2 × {$1\bar{1}00$}AlN. A rotation of the Al lattice by 3.7° around the Al[10] axis establishes a more stable orientation relationship based on three-dimensional geometrical coherency and may suggest a more favorable chemical bonding at the interface at this orientation relationship. A similar tilt character was found on deposited vanadium (V) film on α-Al_2O_3 ($1\bar{1}02$) substrate, which showed a unique three-dimensional orientation relationship in V/Al_2O_3 system irrespective of the interface plane [16]. Normally, epitaxial thin films grow following the symmetry of the substrate surface. However, in some cases a small tilt is more favorable, such as in V/Al_2O_3 and in niobium (Nb)/Al_2O_3, which shows a tilt on (001) plane with respect to the ($01\bar{1}2$) interface plane in order to preserve its three-dimensional orientation relationship [18, 19].

Since the observed Al(111) tilt is predicted to be one of the preferentially stable secondary ORs in CRLP calculations of geometrical coherency then it strengthens the likelihood that this characteristic tilt as found experimentally in the Al/AlN liquid-phase bonding condition is not just an isolated event. During experimentation, there may be limitation in the evaluation of the consistency of the stable ORs by multiple observations of different samples. Thus, CRLP model is also a useful tool to indicate the likelihood that an experimentally observed OR will be repeatable under the same process conditions.

Conclusions

Al–Si/AlN hetero interface was fabricated by liquid-phase bonding method and characterized by HRTEM. We found that Al forms a textured (111) layer with about 4° tilt with

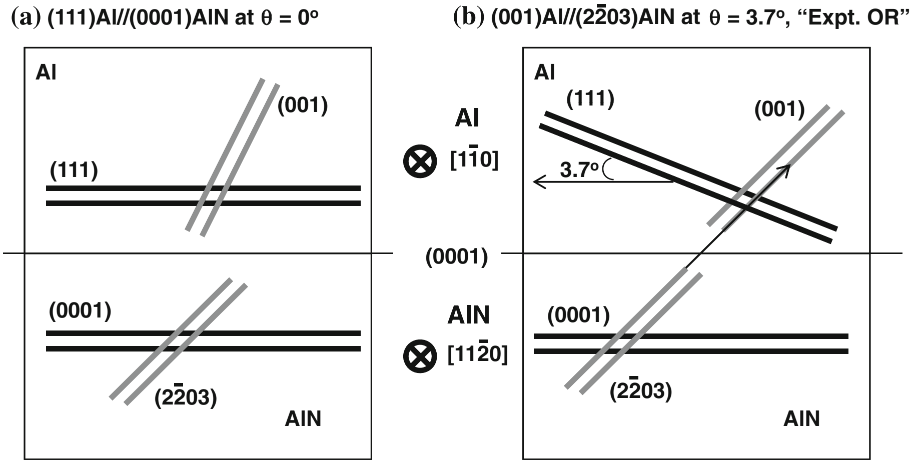


Fig. 4 Schematic of the parallelism of Al(111) with respect to the AlN(0001) in the [1$\bar{1}$0] rotation axis, **a** when Al(111) and AlN(0001) are exactly parallel and **b** when Al(111) is tilted by 3.7° with respect to AlN(0001) which results to (001)Al//($2\bar{2}03$)AlN

respect to the AlN(0001) interface plane under the present bonding conditions. The corresponding stable orientation relationship, $(001)[1\bar{1}0]$Al//$(2\bar{2}03)[11\bar{2}0]$AlN, was also predicted as one of the stable ORs by CRLP calculations. These results suggest that there may be a much stronger preference for aligning Al(001) and AlN$(2\bar{2}03)$ planes than for aligning the Al(111) and AlN(0001) planes when forming the hetero interface using the liquid-phase bonding conditions in this study.

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