The aluminum gallium nitride (AlGaN) system has been used to boost the development of optoelectronics and electronics, as represented by deep-ultraviolet (DUV) light-emitting diodes (LDs) and high-electron-mobility transistors (HEMTs). In indium gallium nitride laser diodes (LDs) and vertical AlGaN/GaN HEMTs, free-standing GaN substrates with low threading dislocation densities (TDDs) strongly support device performance, including lifetime and reliability. However, the free-standing AlN substrate remains expensive, and epitaxial growth of high-crystallinity AlN on sapphire substrates is not yet well established. Therefore, the paucity of inexpensive and high-crystallinity AlN substrates inhibits further improvement in device performance. Our group recently proposed novel fabrication of high-crystallinity AlN templates on sapphire by combining sputter deposition of an AlN layer with a thickness of 170–340 nm with subsequent post-growth annealing at temperatures reaching 1700 °C. A key finding is that even a thickness of 340 nm sufficiently reduces the TDDs to on the order of 10⁹ cm⁻². Namely, this method can lower the production cost because few-micrometer-thick AlN buffer layers currently need to be grown by metal organic vapor phase epitaxy (MOVPE) to reduce the TDDs. X-ray rocking curve (XRC) measurements of the (1012) plane showed marked improvement in the full width at half-maximum (FWHM), where a value of 287 arcsec was successfully achieved, compared with 6031 arcsec before annealing. Although the detailed mechanism of the improvement in crystallinity remains unclear, high-temperature annealing is assumed to strongly enhance the coalescence of sputtered AlN (SPT-AlN) grains through a solid-phase reaction.

Direct wafer bonding (DWB), which plays an essential role in, for example, the packaging of micro-electro-mechanical systems (MEMS) and the fabrication of silicon-on-insulator wafers, is also based on a solid-phase reaction at the wafer surface in order to achieve an adhesion strength comparable to that of the bulk material. For common Si/Si hydrophilic bonding, an annealing temperature of 800–1000 °C is required to form strong covalent bonds at the Si/Si interfaces. Recent progress using plasma and an ion beam for surface treatment enabled low-temperature bonding at temperatures as low as room temperature (∼300 °C), which paved the way for novel device applications such as back-illuminated complementary metal oxide semiconductor sensors, multijunction solar cells, and III–V/Si LDs for Si photonics.

This similarity in the mechanism was taken into account when high-temperature annealing by face-to-face stacking was applied for direct bonding of SPT-AlN. Polarity inversion generated at the bonding interface is of considerable interest with respect to the epitaxial growth of N-polar materials, quasi-phase matching (QPM) for second harmonic generation (SHG), and two-dimensional electron gases induced at the heteropolar interface. In particular, transverse-mode QPM-SHG using direct bonding is expected to overcome the difficulty in longitudinal-mode QPM-SHG in the DUV region, because a grating periodicity of less than 1 μm and the roughness of the N-polar surface are still critical issues in the fabrication of periodically poled AlN waveguides.

In this paper, we report the novel fabrication and crystallographic evaluation of polarity-inverted AlN based on DWB (Fig. 1). To the best of our knowledge, this is the first demonstration of III/N polarity inversion through a single monolayer. Although much effort has been directed...
toward intentional polarity control based on epitaxial growth by MOVPE and molecular beam epitaxy, strict optimization of the growth conditions is still necessary to achieve a smooth morphology and low defect densities. As a completely different approach, our proposal using DWB provides abrupt polarity inversion without the need to insert intermediate layers or amorphous layers. A conceptual diagram of the fundamental wave $E_{2\omega}$ ($\lambda = 532$ nm, $TM_{00}$ mode) and second harmonic wave $E_{2\omega}$ ($\lambda = 266$ nm, $TM_{01}$ mode) in the polarity-inverted AlN waveguide are shown in Fig. 1 as an application of transverse-mode QPM-SHG. The blue and red sections indicate positive and negative amplitudes of the electric field, respectively. The abrupt polarity inversion is indeed appropriate for the SHG device because overlapping of the electric field and the interlayer can cause optical absorption or scattering. Therefore, the proposed approach is expected to provide new insight into previous studies and into device applications in the future.

Fabrication started with the preparation of a pair of 2 in. AlN samples by sputtering an AlN target onto vicinal c-plane sapphire substrates. The sputtering conditions were an RF power of 700 W, a chamber temperature of 600 °C, and an Ar/N$_2$ ratio of 1/4. Subsequently, the $+c$-oriented SPT-AlN wafers were stacked face-to-face and annealed at 1700 °C for 3 h under N$_2$ atmosphere. The sapphire substrates were orientated such that the (1120) planes were aligned in parallel. The annealing process simultaneously improved the crystallinity and the DWB of SPT-AlN. Subsequently, the atomic distance of Al extracted from the STEM image is 2.8 Å at the bonding interface and 2.5 Å at a position nine monolayers from the interface. The 12% larger lattice constant at the interface implies the existence of impurities. Additionally, the depth profile obtained by X-ray photoelectron spectroscopy showed that 11 atom % oxygen was detected at the bonding interface, whereas approximately 1 atom % was detected in the film. On the basis of these experimental findings, a structural model in which $+c$ AlN and $-c$ AlN are mediated by oxygen atoms is proposed on the basis of first-principle calculation and overlapped with the STEM image [Fig. 2(d)]. First-principle calculations of a (2×1) unit cell were carried out by the plane-wave pseudopotential approach using the generalized gradient approximation. To satisfy the electron counting rule, three nitrogen atoms in a unit cell were replaced by oxygen atoms. The proposed model successfully explains the entire atomic arrangement. Additionally, an 18% expansion in atomic distance is expected from this model, which agrees with the experimentally observed expansion. Therefore, the presence of oxygen is estimated to play an essential role in this bonding process, as it does in Si/Si hydrophilic bonding.

The crystallinity of the bonded sample was evaluated using the FWHM of the XRCs, as shown in Fig. 3, where the (0002) and (1012) XRCs of the bonded sample are compared with those of typical as-grown SPT-AlN with a thickness of...
200 nm. X-ray diffraction measurement was carried out using an asymmetric Ge(220) monochromator and Cu Kα radiation (0.154 nm). The bonded sample exhibited FWHMs of 137 arcsec in the (0002) plane and 302 arcsec in the (1012) plane, whereas those of the as-grown SPT-AlN were 64 and 6900 arcsec, respectively. For the (0002) plane, only the sharp peak remained after annealing, and the broad peak vanished. On the other hand, for the (1012) plane, the FWHM decreased markedly, by a factor of 23. Significant improvements in crystallinity are currently understood on the assumption that the coalescence of small grains is enhanced by a solid-phase reaction, and the TDDs are decreased to the order of $10^4 \text{cm}^{-2}$ through the elimination of grain boundaries.\(^{10}\)

Residual stress is problematic because cracking in the epilayer and sapphire substrate results in a low yield ratio. Using 2θ–ω scans of the (0002) and (1012) planes, the c-axis lattice constant was found to be 4.990 Å, and the a-axis lattice constant was found to be 3.098 Å. The out-of-plane lattice constant was found to be 4.982 Å. The bonded sample exhibited FWHMs of 6.1 cm\(^{-1}\) and an RMS surface roughness of 0.26 nm. Regarding the surface roughness, the RMS value was improved from the previously reported value of 0.47 to 0.26 nm.\(^{10}\)

A comprehensive evaluation of the bonded sample was carried out using Raman spectroscopy under the z(x, y) backscattering geometry with a 532 nm excitation light source, as shown in Fig. 4. The Raman shift of the $E_2$ (high) phonon mode shows a peak intensity of 664.2 cm\(^{-1}\), suggesting that in-plane compressive stress is induced compared with free-standing AlN (peak value of 657.4 cm\(^{-1}\)).\(^{31}\) An FWHM of 6.1 cm\(^{-1}\) indicates high crystallinity even after DWB and sapphire separation. Additionally, the $A_1$ (LO) peak clearly observed at 893.5 cm\(^{-1}\) indicates a low residual carrier density of less than $10^{17}$ cm\(^{-3}\).\(^{32}\) Note that the peaks located at 577 and 751 cm\(^{-1}\) resulted from the $E_g$ (int) phonon modes of the sapphire substrate.\(^{33}\)

![Fig. 3. X-ray rocking curves of (a) (0002) and (b) (1012) planes.](image)

![Fig. 4. Raman spectroscopy measurement of bonded AlN sample.](image)
of covalent bonds at the wafer surfaces. HAADF-STEM observation of the bonding interface revealed an abrupt transition from Al polarity to N polarity through a single monolayer. Additionally, an atomically flat interface and regular atomic arrangement were observed across the measured sample. The bonded sample exhibited XRC FWHMs of 137 arcsec in the (0002) plane and 302 arcsec in the (1012) plane. No additional stress caused by direct bonding was recognized. Such ideal polarity inversion of monolayer. Additionally, an atomically

Acknowledgments The authors gratefully acknowledge the contribution of Mr. E. Komatsu of Shinko Seiki Co., Ltd., to the preparation of SPT-AlN samples. This work was partially supported by JSPS KAKENHI (JP15H03556, JP16H06415, JP16H06418, JP17H05335, and JP17H06762), JST SICORP EU H2020 No. 720527 (InRel-NPower), JST SICORP with MOST in China, and the Strategic Foundational Technology Improvement Support Operation of the Kansai Bureau of Economy, Trade and Industry.

1) M. Shatalov, W. Sun, R. Jain, A. Lunev, X. Hu, A. Dobrinsky, Y. Bilenko, J. Yang, G. A. Garrett, and L. E. Rodak, Semicond. Sci. Technol. 29, 084007 (2014).
2) T. Takano, T. Mino, J. Sakai, N. Noguchi, K. Tsubaki, and H. Hirayama, Appl. Phys. Express 10, 031002 (2017).
3) H. Tokuda, M. Hatano, N. Yafune, S. Hashimoto, K. Akita, Y. Yamamoto, and M. Kuzuhara, Appl. Phys. Express 3, 121003 (2010).
4) N. Yafune, S. Hashimoto, K. Akita, Y. Yamamoto, H. Tokuda, and M. Kuzuhara, Electron. Lett. 50, 211 (2014).
5) S. Nagahama, T. Yamamoto, M. Sano, and T. Mukai, Appl. Phys. Lett. 79, 1948 (2001).
6) T. Miyoshi, S. Masui, T. Okada, T. Yamamoto, T. Kozaki, S. Nagahama, and T. Mukai, Appl. Phys. Express 2, 062201 (2009).
7) A. Avramescu, T. Lermer, J. Müller, C. Eichler, G. Brueederl, M. Sabathil, S. Lutgen, and U. Strauss, Appl. Phys. Express 3, 061003 (2010).
8) S. Chowdhury and U. K. Mishra, IEEE Trans. Electron Devices 60, 3060 (2013).
9) H. Nie, Q. Diduck, B. Alvarez, A. P. Edwards, B. M. Kayes, M. Zhang, G. Ye, T. Prunty, D. Bour, and I. C. Kizilyalli, IEEE Electron Device Lett. 35, 939 (2014).
10) H. Miyake, C.-H. Lin, K. Tokoro, and K. Hiramatsu, J. Cryst. Growth 456, 155 (2016).
11) H. Hemmi, S. Shoji, Y. Shoji, K. Yoshimi, and M. Esashi, Sens. Actuators A 43, 243 (1994).
12) M. Bruel, B. Aspar, and A.-J. Auberton-Herve, Jpn. J. Appl. Phys. 36, 1636 (1997).
13) U. Gösele and Q.-Y. Tong, Annu. Rev. Mater. Sci. 28, 215 (1998).
14) S. G. Wu, C. C. Wang, B. C. Hsieh, Y. L. Tu, C. H. Tseng, T. H. Hsu, R. S. Hsiao, S. Takahashi, R. J. Lin, C. S. Tsai, Y. P. Chao, K. Y. Chou, P. S. Chou, H. Y. Tu, F. L. Hsieh, and L. Tran, IEDM Tech. Dig., 2010, 14.1.1.
15) F. Dimroth, M. Grave, P. Beutel, U. Fiedeler, C. Karcher, T. N. Tibbits, E. Oliva, G. Siefer, M. Schachtner, and A. Wekkeli, Prog. Photovoltaics 22, 277 (2014).
16) A. W. Fang, H. Park, O. Cohen, R. Jones, M. J. Panicia, and J. E. Bowers, Opt. Express 14, 9203 (2006).
17) V. Ramachandran, R. Feenstra, W. Sarney, L. Salamanca-Riba, J. Northrup, L. Romanov, and D. Greve, Appl. Phys. Lett. 75, 808 (1999).
18) F. Liu, R. Collazo, S. Mitra, Z. Sitar, G. Duscher, and S. J. Pennycook, Appl. Phys. Lett. 91, 201115 (2007).
19) M. H. Wong, F. Wu, J. S. Speck, and U. K. Mishra, J. Appl. Phys. 108, 123710 (2010).
20) W. Wirges, S. Yilmaz, W. Brinker, S. Bauer-Gogonea, S. Bauer, M. Jäger, G. Stegeman, M. Allheim, M. Stähelin, and B. Zysset, Appl. Phys. Lett. 70, 3347 (1997).
21) J. Wright, C. Moe, A. Sampath, G. Garrett, and M. Wraback, Phys. Status Solidi C 8, 2331 (2011).
22) D. Alden, W. Guo, R. Kresse, F. Kaess, I. Bryan, T. Troha, A. Bagal, P. Reddy, L. H. Hernandez-Balderrama, and A. Franke, Appl. Phys. Lett. 108, 261106 (2016).
23) M. Stutzmann, O. Ambacher, M. Eickhoff, U. Karrer, A. Lima Pimenta, R. Neuberger, J. Schulwig, R. Dimitrov, P. Schuck, and R. Grober, Phys. Status Solidi B 228, 505 (2001).
24) A. Aleksov, R. Collazo, S. Mitra, R. Schlesser, and Z. Sitar, Appl. Phys. Lett. 89, 052117 (2006).
25) S. Mohn, N. Stolyarchuk, T. Markert, R. Kirse, M. P. Hoffmann, R. Collazo, A. Courville, R. Di Felice, Z. Sitar, and P. Vennéguès, Phys. Rev. Appl. 5, 034004 (2016).
26) T. Akyama, H. Nakane, K. Nakamura, and T. Ito, Phys. Rev. B 94, 115302 (2016), and references therein.
27) M. D. Pashley, K. W. Haberern, W. Friday, J. M. Woodall, and P. D. Kirchner, Phys. Rev. Lett. 60, 2176 (1988).
28) W. Yim, E. Stofo, P. Zanzucchi, J. Pankove, M. Ettensen, and S. Gilbert, J. Appl. Phys. 44, 292 (1973).
29) J. Wagner and F. Bechstedt, Appl. Phys. Lett. 77, 346 (2000).
30) J. Rouviere, K. Rousseau, F. Fournel, and H. Moriceau, Appl. Phys. Lett. 89, 052117 (2006).
31) T. Prokofyeva, M. Seon, J. Vanbuskirk, M. Holtz, S. Nakishin, N. Faleev, H. Temkin, and S. Zollner, Phys. Rev. B 63, 125313 (2001).
32) M. Gómez-Gómez, A. Cros, M. Hermann, M. Stutzmann, and M. Eickhoff, Phys. Status Solidi A 206, 1183 (2009).
33) T. Wermelinger, C. Borgia, C. Solenthaler, and R. Spolenak, Acta Mater. 55, 4657 (2007).