Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary information:

Direct visualization of N impurity state in dilute GaNAs using scanning tunneling microscopy

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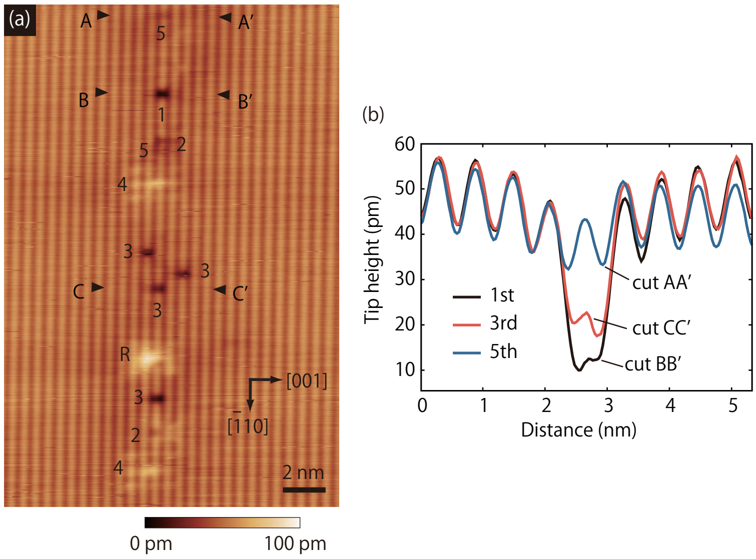

FIG. S1: (a) Topographic image of GaAs(110) surface obtained along N-doped layer ($V_S = -2.5$, $I_T = 100$ pA). (b) Cross sections taken along line AA', BB', and CC'. The positions of N atoms were determined to be the first, third, and fifth plane in terms of the depth of depressions.

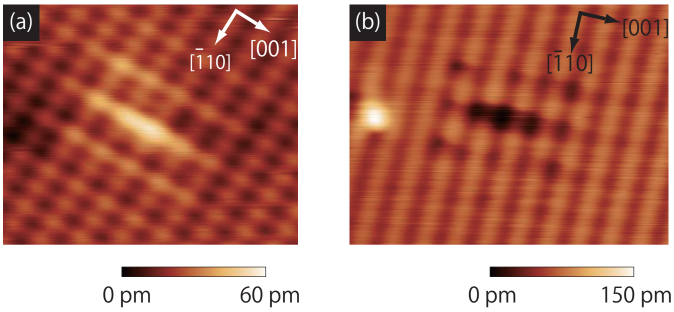

FIG. S2: Topographic images of N impurities in fourth plane obtained at (a) $V_S = -2.5$ V and (b) $V_S = -2.3$ V. N impurities showed bright or dark features superimposed on the As sub-lattice, depending on the tip condition and/or lateral position at the surface. A typical feature was three lines elongated along the [001] direction.

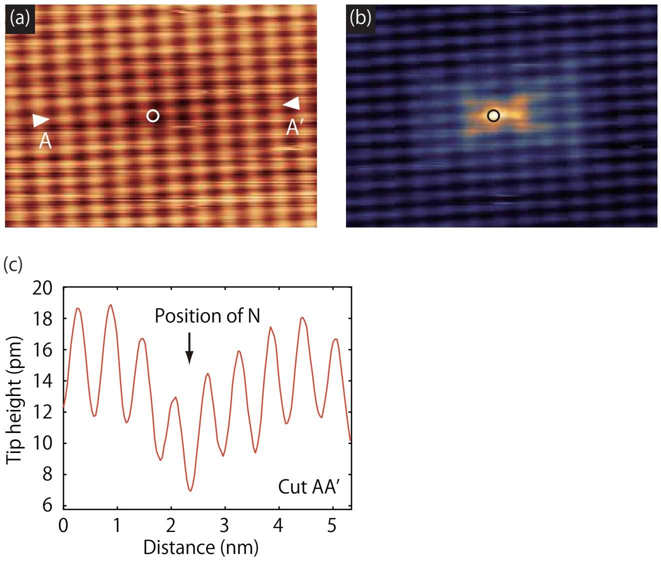

FIG. S3: (a) Topographic image ($V_S = -2.5$ V) and (b) current image ($V_S = 1.4$ V) of single N impurity located in the sixth plane below the surface. The projected position of N atom is indicated by a circle. (c) Line profile taken along the line AA'. The topographic height around the position where the N impurity exist underneath was slightly lower by several picometers than that at surroundings.

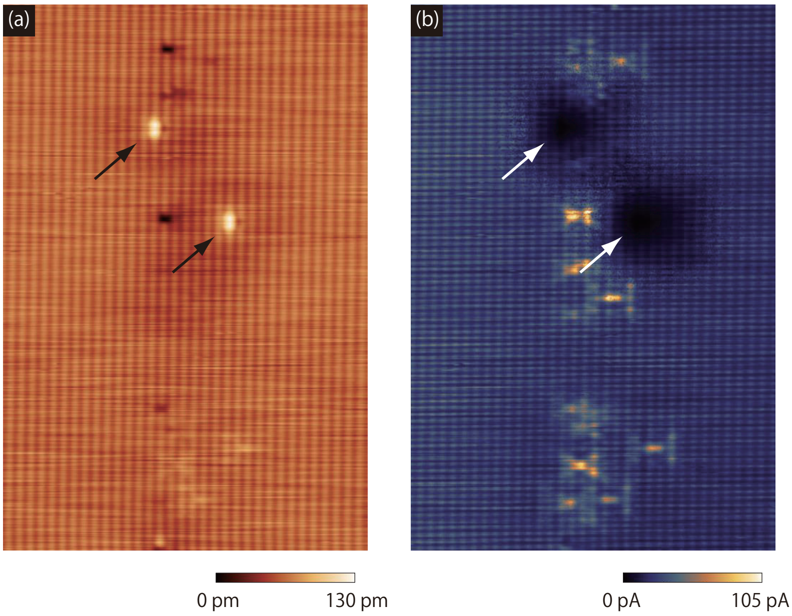

FIG. S4: (a) Topographic image of GaAs(110) surface obtained along N-doped layer ($V_S = -2.5$ V, $I_T = 100$ pA). Surface adsorbates are indicated by arrows. (b) Current image ($V_S = 1.9$ V) taken simultaneously with (a) at identical surface position. Tunneling current was strongly suppressed around surface adsorbates, as indicated by arrows.

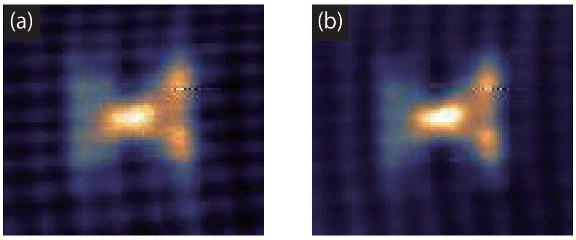

FIG. S5: (a) Current and (b) dI/dV images of N impurity in the third plane at $V_S = 1.5$ V. These images are extracted from a single set of current imaging tunneling spectroscopy data. The imaged dI/dV values for (b) were numerically calculated from the I-V spectra.

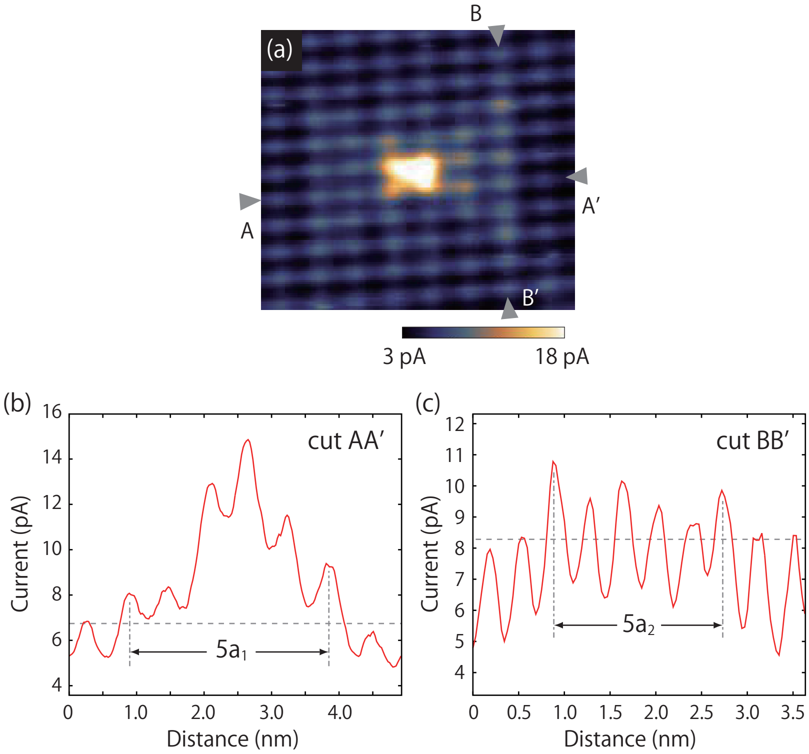

FIG. S6: (a) High resolution current image ($V_S = 1.4$ V) of single N impurity located in the fifth plane without the height correction (raw data). (b) and (c) show the cross sections along line AA' and BB' in (a), respectively.

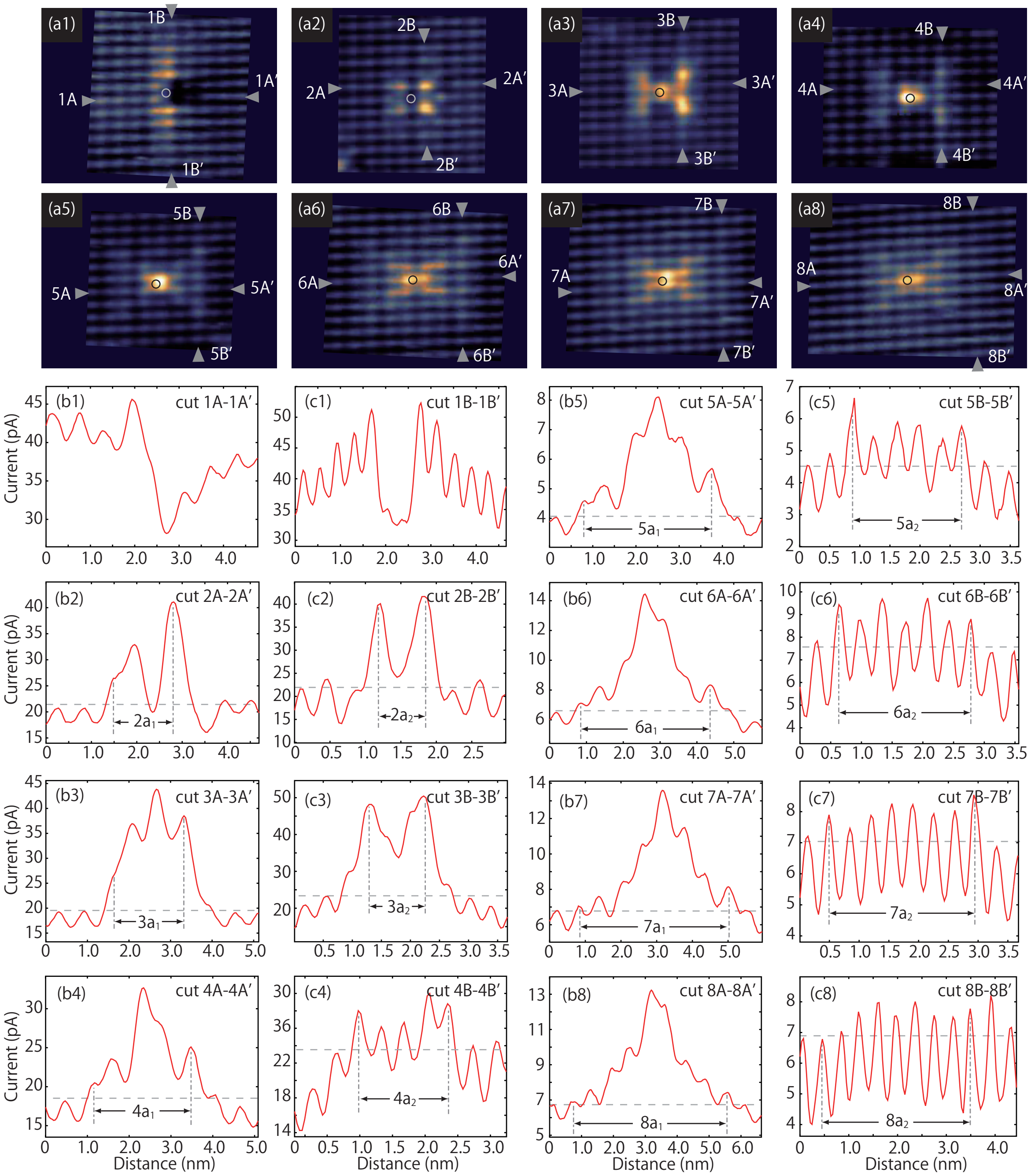

FIG. S7: (a1-a8) Corrected current images of N impurities located in first to eighth planes. (b1-b8) and (c1-c8) show the cross sections taken along lines indicated in (a1-a8). The size of N impurity features increase by a lattice constant of GaAs(110), i.e., 0.565 nm in the [001] direction and 0.4 nm in the $[1\bar{1}0]$ direction, step by step from (a2) to (a8).

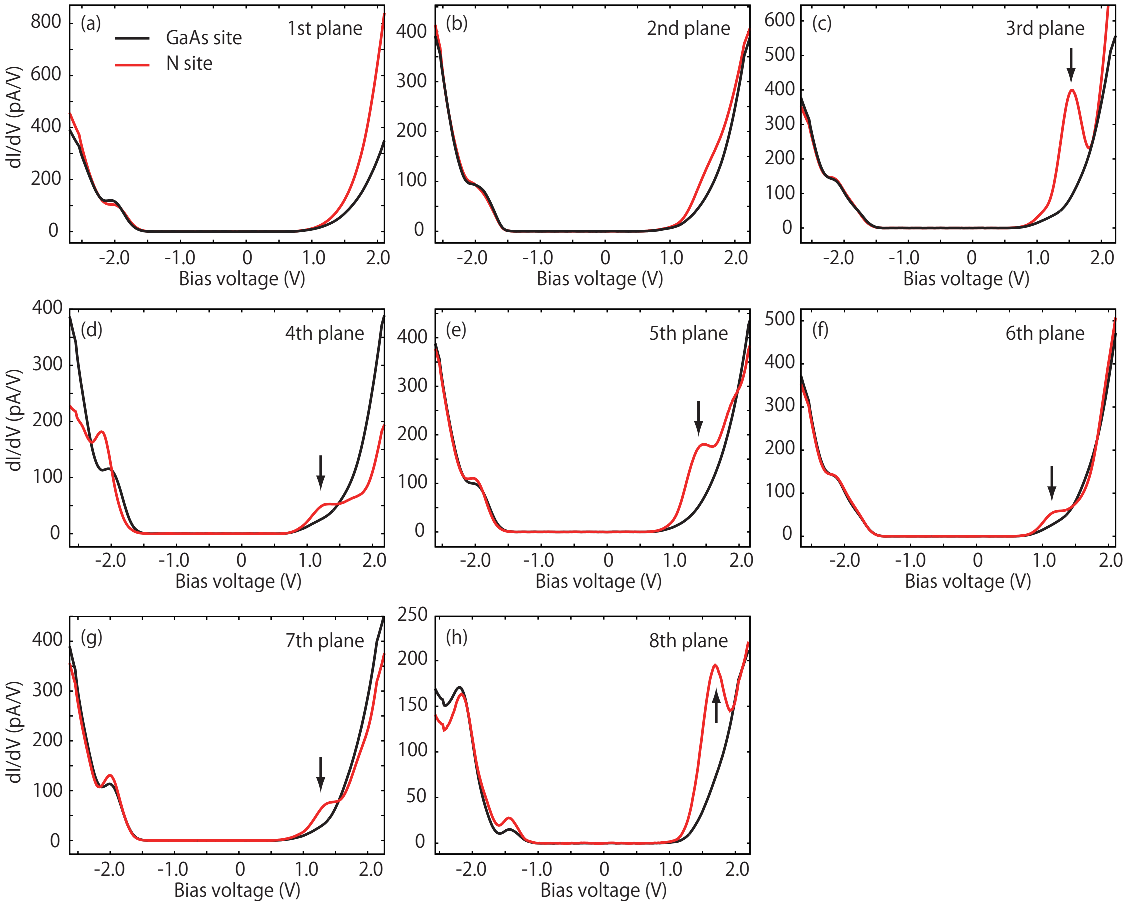

FIG. S8: Differential tunneling conductance spectra obtained above N impurities in (a) first to (h) eighth planes (corresponding to the position of circles in Fig. S7(a1-a8)). For an N atom in each plane, we obtained spectra both above the N atom and at the bare GaAs site with an identical tip state.

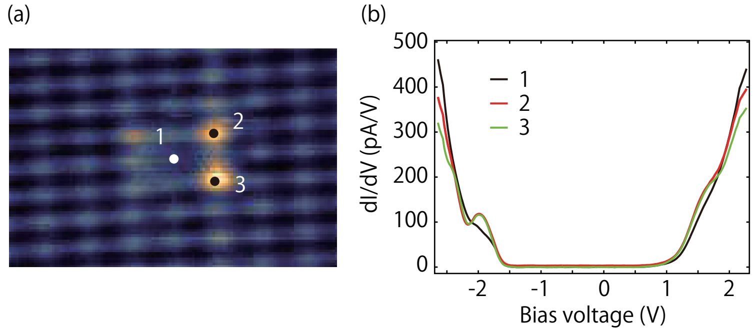

FIG. S9: (a) Current image ($V_S = 1.4$ V) of N impurity located in second plane. (b) Differential tunneling conductance spectra obtained at positions 1-3 shown in (a). At position 2 and 3, a shoulder feature (increase of signals) around 1.5 V is slightly enhanced than that at position 1.

Sample preparation

We fabricated ten N-doped GaAs layers (from layers 1 to 10) with different doping densities on the n-type GaAs(001) substrate by a molecular beam epitaxy. The followings are the detailed growth conditions in each layer. From layers 1 to 4, N atoms were δ-doped by supplying atomic N and As to the growth-interrupted GaAs surface at a substrate temperature of 580 °C.[1,2] A cell shutter for N was closed during the δ-doping, thus only small amount of the atomic N were able to reach the substrate surface. The duration of the N supply was changed to control the amount of δ-doped N atoms. The values were 30, 60, 120, and 240 s for layer 1, 2, 3, and 4, respectively. For layer 5 and 6, N atoms were uniformly doped during the growth of 4 nm-GaAs without the growth interruption (growth of GaNAs alloy). The cell shutter for N was opened during the N supply. The substrate temperature for the growth of GaNAs was 580 °C for layer 5 and 520 °C for layer 6. From layers 7 to 10, N atoms were doped in a similar way as for layer 5 and 6, but additional N atoms were also supplied using the growth-interrupted method:[3] the growth of GaNAs was stopped after every 1 nm growth and the atomic N and As were supplied to the surface for a certain time. The durations of the N supply were 2, 5, 10, and 20 s for layers 7, 8, 9, and 10, respectively. The substrate temperature during the growth was 520 °C.

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[1] T. Kita and O. Wada, Phys. Rev. B **74**, 035213 (2006).

[2] M. Ikezawa, Y. Sakuma, and Y. Masumoto, Jpn. J. Appl. Phys. **46**, L871 (2007).

[3] T. Mano, M. Jo, K. Mitsuishi, M. Elborg, Y. Sugimoto, T. Noda, Y. Sakuma, and K. Sakoda, Appl. Phys. Express **4**, 125001 (2011).