Table S1. XRD-derived phase contents, lattice parameters, and average crystallite sizes. MG – manual grinding, SBM – short ball milling, LBM – long ball milling. n/d – not determined.

<table>
<thead>
<tr>
<th>Powder (temperature, time/grinding)</th>
<th>Phase, content [%]</th>
<th>Lattice parameters a[Å], c[Å]</th>
<th>Av. crystallite size [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>650 °C, 36 h/LBM</td>
<td>h-GaN, 100</td>
<td>a=3.18, c=5.19</td>
<td>12</td>
</tr>
<tr>
<td>700 °C, 90 h/MG</td>
<td>h-GaN (small), 50</td>
<td>a=3.17, c=5.19</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>h-GaN (large), 33</td>
<td>a=3.20, c=5.23</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td>c-GaN, 14</td>
<td>a=4.52</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td>c-GaAs, 3</td>
<td>n/d</td>
<td>n/d</td>
</tr>
<tr>
<td>700 °C, 90 h/SBM</td>
<td>h-GaN, 99.8</td>
<td>a=3.19, c=5.21</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>c-GaAs, 0.2</td>
<td>n/d</td>
<td>n/d</td>
</tr>
<tr>
<td>700 °C, 90 h/LBM</td>
<td>h-GaN, 100</td>
<td>a=3.19, c=5.20</td>
<td>15</td>
</tr>
<tr>
<td>800 °C, 90 h/MG</td>
<td>h-GaN (small), 32</td>
<td>a=3.16, c=5.22</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>h-GaN (large), 25</td>
<td>a=3.19, c=5.19</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>c-GaN, 42.5</td>
<td>a=4.51</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td>c-GaAs, 0.5</td>
<td>n/d</td>
<td>n/d</td>
</tr>
<tr>
<td>800 °C, 90 h/SBM</td>
<td>h-GaN, 71</td>
<td>a=3.19, c=5.22</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td>c-GaN, 29</td>
<td>a=4.51</td>
<td>15</td>
</tr>
<tr>
<td>800 °C, 90 h/LBM</td>
<td>h-GaN, 62</td>
<td>a=3.20, c=5.20</td>
<td>14</td>
</tr>
<tr>
<td></td>
<td>c-GaN, 38</td>
<td>a=4.51</td>
<td>16</td>
</tr>
<tr>
<td>900 °C, 6 h/MG</td>
<td>h-GaN, 33.5</td>
<td>a=3.18, c=5.29</td>
<td>26</td>
</tr>
<tr>
<td></td>
<td>c-GaN, 65</td>
<td>a=4.51</td>
<td>55</td>
</tr>
<tr>
<td></td>
<td>c-GaAs, 1.5</td>
<td>n/d</td>
<td>n/d</td>
</tr>
<tr>
<td>900 °C, 6 h/SBM</td>
<td>h-GaN, 68</td>
<td>a=3.19, c=5.21</td>
<td>16</td>
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<tr>
<td></td>
<td>c-GaN, 32</td>
<td>a=4.51</td>
<td>23</td>
</tr>
<tr>
<td>900 °C, 6 h/LBM</td>
<td>h-GaN, 72</td>
<td>a=3.19, c=5.21</td>
<td>17</td>
</tr>
<tr>
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<td>c-GaN, 28</td>
<td>a=4.51</td>
<td>16</td>
</tr>
<tr>
<td>900 °C, 6 h/bulk wafer</td>
<td>h-GaN, 76</td>
<td>a=3.19, c=5.20</td>
<td>30</td>
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<tr>
<td></td>
<td>c-GaN, 24</td>
<td>a=4.51</td>
<td>33</td>
</tr>
</tbody>
</table>

Table S2. Positions of deconvoluted peaks in micro-Raman determinations for GaAs substrates and nitrided products. MG – manual grinding, LBM – long ball milling, am – amorphous, cryst – crystalline, br – broad. References in square brackets are listed as such in the text.
<table>
<thead>
<tr>
<th>shift [cm(^{-1})]/mode (assignment) [Ref.]</th>
<th>GaAs_MG</th>
<th>GaAs_LBM</th>
<th>700_MG</th>
<th>700_LBM</th>
<th>800_MG</th>
<th>800_LBM</th>
<th>900_MG</th>
<th>900_LBM</th>
</tr>
</thead>
<tbody>
<tr>
<td>150/LA (am GaAs) [20a]; 144/E(_1)(low) (h-GaN) [20g]</td>
<td>156</td>
<td>ca.160</td>
<td>151</td>
<td></td>
<td>161</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>210-240, br (am As); 198 (cryst As) [20b]; 205, br (am As) [20c]; ca. 200/DALA in GaAsN [20d]</td>
<td>197</td>
<td>195, 222</td>
<td>204, 213</td>
<td>197, 221</td>
<td>202, 221</td>
<td>196, 216</td>
<td>199, 226</td>
<td>200, 224</td>
</tr>
<tr>
<td>231-245'/TO, br (am GaAs) [20a,20c]; 255 (cryst As) [20b]; 246 (Ga metal) [20l]</td>
<td>239</td>
<td>243</td>
<td>238</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>264-265'/TO (cryst GaAs) [20c]</td>
<td>264</td>
<td>261</td>
<td>255</td>
<td>249</td>
<td>248</td>
<td>249</td>
<td>250</td>
<td></td>
</tr>
<tr>
<td>285-292'/LO (cryst GaAs) [20a,20c]; red shift in GaAsN [20e]</td>
<td>285</td>
<td>283</td>
<td>279</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>2(^{nd}) order GaAsN [20e]; B(_1)(low)silent (h-GaN) [20i]; nitrogen vacancies (h-GaN) [20j]</td>
<td></td>
<td></td>
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<td>299, 315</td>
<td>297</td>
<td>304</td>
<td>296</td>
</tr>
<tr>
<td>nitrogen vacancies (h-GaN) [20j], octahedrally bonded Ga [20k]</td>
<td></td>
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<td></td>
<td></td>
<td>411</td>
<td>411</td>
<td>417</td>
<td>410</td>
</tr>
<tr>
<td>480/2(^{nd}) order in GaAs [20a] or 470 in GaAsN [20e]; 467-475/N-related LVM [20f]</td>
<td>480</td>
<td></td>
<td></td>
<td></td>
<td>460</td>
<td>460</td>
<td>513</td>
<td></td>
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<tr>
<td>554-558/E(_1)(TO) (h-GaN); 552/TO (c-GaN) [20h]; 532/A(_1)(TO) (h-GaN); 559/E(_1)(TO) (h-GaN); 568/E(_1)(high) (h-GaN); 555/TO (c-GaN) [20g]</td>
<td></td>
<td></td>
<td>550</td>
<td>558</td>
<td>555, 561</td>
<td>544</td>
<td>541</td>
<td>560</td>
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<tr>
<td>B(_1)(low)silent (h-GaN) [20i]; nitrogen vacancies (h-GaN) [20j]</td>
<td></td>
<td></td>
<td>601, 663</td>
<td>689</td>
<td>612, 684</td>
<td>602, 652</td>
<td>599, 668</td>
<td>607, 645</td>
</tr>
<tr>
<td>735/A(_1)(LO) (h-GaN), 739/LO (c-GaN) [20h]; 734/A(_1)(LO) (h-GaN); 741/E(_1)(LO) (h-GaN) [20g]</td>
<td>700, 756</td>
<td>713</td>
<td>718, 756</td>
<td>708, 754</td>
<td>704, 755</td>
<td>714, 757</td>
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</tbody>
</table>