Supplementary Material:

*Understanding the Nature of Crystallographic Bonds by Establishing the Correlation between Ion Pair Chemistry and their Separation in Detector Space*

Distributions were taken from 10 nm thick representative regions within the central region of interest. The layer (AlGaN or GaN) and growth method (MBE or MOCVD) are indicated above each set of figures.

Ga+-N+ evaporated from AlGaN

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 2.1 mm  Median = 1.4 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 1.9 mm  Median = 1.4 mm |

Ga+-N+ evaporated from GaN

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.5 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 1.9 mm  Median = 1.4 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.7 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 1.8 mm  Median = 1.3 mm |

Ga+-N2+ evaporated from AlGaN

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.0 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.7 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 2.3 mm  Median = 1.6 mm |

Ga+-N2+ evaporated from GaN

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.0 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.7 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.6 mm | Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.6 mm |

Ga+-Ga+ evaporated from AlGaN

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 1.6 mm  Median = 1.3 mm | Chart, histogram  Description automatically generated  FWHM = 2.0 mm  Median = 1.5 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.0 mm  Median = 1.3 mm | Chart, histogram  Description automatically generated  FWHM = 1.9 mm  Median = 1.4 mm |

Ga+-Ga+ evaporated from GaN

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 1.8 mm  Median = 1.5 mm | Chart, histogram  Description automatically generated  FWHM = 1.7 mm  Median = 1.5 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 1.8 mm  Median = 1.4 mm | Chart, histogram  Description automatically generated  FWHM = 1.9 mm  Median = 1.5 mm |

Al+-N+

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 3.3 mm  Median = 2.1 mm | Chart, histogram  Description automatically generated  FWHM = 3.5 mm  Median = 2.6 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 3.5 mm  Median = 2.2 mm | Chart, histogram  Description automatically generated  FWHM = 3.3 mm  Median = 2.2 mm |

Al+-N2+

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.8 mm  Median = 2.8 mm | Chart, histogram  Description automatically generated  FWHM = 2.7 mm  Median = 2.8 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.7 mm  Median = 2.5 mm | Chart, histogram  Description automatically generated  FWHM = 2.7 mm  Median = 2.7 mm |

Al++-N+

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.7 mm  Median = 1.9 mm | Chart, histogram  Description automatically generated  FWHM = 2.8 mm  Median = 1.9 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.6 mm  Median = 1.9 mm | Chart, histogram  Description automatically generated  FWHM = 2.9 mm  Median = 1.9 mm |

Al++-N2+

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.7 mm  Median = 2.5 mm | Chart, histogram  Description automatically generated  FWHM = 2.8 mm  Median = 2.4 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.6 mm  Median = 2.3 mm | Chart, histogram  Description automatically generated  FWHM = 2.9 mm  Median = 2.3 mm |

Al+++-N+

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.6 mm  Median = 1.6 mm | Chart, histogram  Description automatically generated  FWHM = 2.3 mm  Median = 1.5 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.7 mm  Median = 1.6 mm | Chart, histogram  Description automatically generated  FWHM = 2.5 mm  Median = 1.6 mm |

Al+++-N2+

|  |  |
| --- | --- |
| MBE-1 | MOCVD-1 |
| Chart, histogram  Description automatically generated  FWHM = 2.0 mm  Median = 1.6 mm | Chart, histogram  Description automatically generated  FWHM = 1.9 mm  Median = 1.5 mm |
| MBE-2 | MOCVD-2 |
| Chart, histogram  Description automatically generated  FWHM = 2.2 mm  Median = 1.6 mm | Chart, histogram  Description automatically generated  FWHM = 1.8 mm  Median = 1.6 mm |