Comment on "Structures of GaN(0001)-(2×2), -(4×4), and -(5×5) Surface Reconstructions"

In a recent Letter, Xue *et al.* [1] present scanning tunneling microscopy (STM) images of various surface reconstructions on the Ga-polar (0001) face of GaN, prepared by molecular beam epitaxy (MBE) using a plasma source for nitrogen. They present structural models for 2×2, 4×4, and 5×5 reconstructions and report the observation of other reconstructions as well. On the basis of our own STM/MBE studies [2,3], we argue here that the 2×2, 4×4, 5×5, and other reconstructions discussed by Xue *et al.* involve the presence of unintentional arsenic contamination of the GaN surface.

The 2×2 and 4×4 reconstructions of GaN(0001) have been reported by many groups performing MBE growth of GaN [2]. The 2×2 in particular is seen in reflection high-energy electron diffraction (RHEED) during growth and is often quite intense. These reconstructions have been used as indicators of both the Ga polarity of the film and the high quality nature of the growth [2]. Nevertheless, a number of other groups, including our own, have been unable to observe a 2×2 RHEED pattern *during* growth (although all groups can achieve it when the growth is interrupted). We have previously discussed this apparent discrepancy between the results of the various groups and have suggested that the 2×2 (and 4×4) may be the result of unintentional contamination in the vacuum chamber used for the growth. Arsenic is a prime candidate for such contamination, since many of the growth chambers used for GaN have previously been used for GaAs growth, and the high vapor pressure of arsenic ensures that any trace amounts present near the growth sources will produce a significant partial pressure during growth. For our prior studies, neither the growth chamber nor any of its components had ever been exposed to any arsenic.

To test the sensitivity of the 2×2 reconstruction to the presence of arsenic, we have purposely introduced it through resistive heating of a GaAs wafer located in the growth chamber about 5 cm from the substrate holder. For a beam equivalent pressure of the arsenic of $>1 \times 10^{-9}$ Torr during MBE growth, we clearly observe the development of a 2×2 RHEED pattern during growth under N-rich conditions. This pattern is consistently observed in the presence of arsenic, whereas we have *never* seen it during growth in the absence of arsenic. The presence of arsenic on this 2×2 surface was confirmed by Auger spectroscopy; a clear arsenic signal with As/Ga peak ratio of 0.022 is obtained from surfaces which show the 2×2 reconstruction, from which the arsenic coverage is estimated to be about 0.18 monolayer (ML).

Theoretically, arsenic on the GaN(0001) surface is expected to form a very stable 2×2 adatom structure [4]. The surface coverage of 0.25 ML expected for this structure is somewhat more than the 0.18 ML observed above, but the difference is within the uncertainty of our Auger evaluation. Upon adding small amounts of Ga to our 2×2 surface we observe the formation of 4×4 and 5×5 reconstructions, in agreement to that reported by Xue *et al.* and thus providing additional evidence that the 2×2 structure they observe is identical to that reported here. A simple model for the 4×4 structure would consist of 3 As adatoms and 1 Ga adatom per 4×4 cell, yielding STM contrast consistent with that reported by Xue *et al.* (dangling bonds are filled on As adatoms and empty on Ga adatoms).

In conclusion, we argue that the 2×2, 4×4, 5×5, and other reconstructions reported by Xue *et al.* on the GaN(0001) surface involve the unintentional presence of arsenic on the surface. We note that arsenic is not the only species which can produce a 2×2 reconstruction on GaN(0001); we have observed a 2×2 arrangement in the presence of Mg, [5] and 2×2 patterns during growth with ammonia have been reported. Nevertheless, because of the similarity of our results with arsenic compared to those of Xue *et al.*, we feel that their results, as well as those reported by other groups performing plasma-MBE growth of GaN, do indeed arise from the presence of arsenic on the surface.

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