Electron Holography Studies of the Charge on Dislocations in GaN

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(Received 4 May 2001; published 29 October 2001)

Off-axis electron holography in a transmission electron microscope is used to examine the charge on threading edge dislocations in n-GaN (0001). It is shown that the crystal inner potential is reduced within 10 nm of the dislocation consistent with a negatively charged core. The results can be explained by a simple unscreened potential due to a core charge of about 4×10^7 electrons cm^{-1}. The origin of this charge is discussed. The application of the method to other types of dislocation is also considered.

DOI: 10.1103/PhysRevLett.87.205504 PACS numbers: 61.72.Ff, 61.14.Nm, 73.20.–r

In the last few years, GaN/InGaN/AlGaN layers grown in the hexagonal-wurtzite structure have been used for commercial light emitting diodes (LEDs) and laser diodes (LDs) working at blue-green wavelengths [1]. There is also great interest in their application to high speed, high power electronic devices [2]. A major issue, however, is why these devices, particularly LEDs and LDs, work efficiently given that threading defect densities are generally extremely high, up to 10^{11} cm^{-2}. In comparison, fabrication of light emitting devices based on GaAs requires threading defect densities less than about 10^5 cm^{-2}. In the GaN heterostructures, threading defects arise through the, so-far unavoidable, use of highly lattice-mismatched substrates, the most common being (0001) sapphire (Al_2O_3) where the mismatch is 14%. The resulting (0001)-oriented layers have threading dislocations which align closely with the [0001] growth direction. These dislocations are of three types, with Burgers vectors of $\frac{1}{3}\langle 1\,1\,-\,2\,0\rangle$ (**a**, or edge type), $\langle 0001\rangle$ (**c**, or screw type), and $\frac{1}{3}\langle 1\,1\,-\,2\,3\rangle$ (**c** + **a**, or mixed type) [3].

The tolerance of GaN devices to threading dislocations might, of course, be explained if dislocations did not give rise to electronic states in the band gap. The evidence is conflicting on this point. Calculations have suggested that, for closed core stoichiometric configurations, screw and mixed dislocations should give rise to band-gap states, but edge dislocations, which usually predominate in device structures, should be electrically inactive [4,5]. In contrast, calculations for edge dislocations with open core configurations, or with Ga vacancies in the core, suggest that band-gap acceptor-type states should be present [5]. Experimentally there is indirect evidence that dislocations in n-GaN can be charged. Look and Sizelove [6] correlated transport data with dislocation densities and concluded that edge dislocations are scattering centers consistent with a negatively charged core. A combination of atomic force microscopy and scanning capacitance microscopy has been used to demonstrate an accumulation of negative charge near surface pits, believed to be due to threading edge dislocations [7]. Observations of differential photoelectrochemical etching around dislocations also suggest that dislocations are charged [8].

In this Letter, we present direct evidence for the charging of dislocations in n-GaN. Electron holography carried out in the transmission electron microscope (TEM) is used, in a novel application, to examine the crystal inner potential around endon edge dislocations. It is shown that the inner potential is reduced by up to 2.5 V within a few nanometers of the dislocation core. The results, which can be fitted with an unscreened potential, are consistent with a line charge of about 2 electrons/c, where $c = 0.52$ nm is the unit cell parameter. The origin of this line charge is discussed. The application of electron holography to screw and mixed dislocations is also considered.

The sample examined here was a 1.2 μm n-GaN layer (Si doped at $N_d = 6 \times 10^{17}$ cm^{-3}, mobility = 360 $cm^2\,V^{-1}\,s^{-1}$) grown by metal-organic chemical vapor deposition on (0001) sapphire at Hewlett-Packard Laboratories, California. TEM specimens were prepared by backthinning using mechanical polishing, dimpling, and finally ion thinning using 5 keV Ar ions at 12° incidence. Specimens were examined in a Hitachi HF2000 field emission gun TEM operating at 200 kV. Off-axis electron holography was carried out using a Moellenstedt biprism located in the image plane of the intermediate lens. Holographic interference patterns were recorded on a 1024 × 1024 CCD array, compared with reference empty space holograms, and separated into phase and amplitude maps by standard means [9]. Considering just the phase variations, the relative phase between a reference (vacuum) wave and the specimen wave is given by

$$\varphi(x, y) = \int_{-\infty}^{\infty} C_E V(x, y, z)\, dz \tag{1}$$

where $C_E = (2\pi/\lambda)(E + E_0)/[E(E + 2E_0)]$, E = kinetic energy of the electrons of wavelength λ, E_0 is the rest mass energy of the electron, and $V(x, y, z)$ is the potential at depth z. In the absence of fringing fields, and under conditions where electron channeling can be neglected, we can write

$$\varphi = C_E V_0 t\,, \tag{2}$$

where V_0 is the average inner potential and t is the foil thickness.

Electron holography was carried out on edge dislocations viewed in an endon geometry. To satisfy the conditions for Eq. (2), dislocations were not examined exactly endon, i.e., down [0001], where strong beam diffracting conditions and diffraction contrast from the dislocations are likely to influence phase changes [9]. Instead, orientations 3°–4° off [0001] were chosen to achieve both minimal dislocation contrast and near kinematical diffracting conditions. The Burgers vectors of the dislocations were analyzed using two-beam images taken in reflections **g**, and applying the $\mathbf{g} \cdot \mathbf{b} = 0$ invisibility criterion [10]. Film thicknesses were measured adjacent to dislocations using thickness fringes in convergent beam electron diffraction patterns taken under two-beam conditions [10].

Figures 1 and 2 illustrate results for the same dislocation. Figure 1 shows images confirming the edge character of a group of dislocations arrowed. Figure 2 shows electron holography results with the dislocation A close to endon. In the hologram of Fig. 2(a), the dislocation is approximately in the center of the outlined box. Figure 2(b) shows a map of the phase φ in the boxed region, in which a constant phase ramp has been applied to remove coarse variations due to thickness gradients (N. B., in this case both the "specimen" and "reference" beams pass through the sample). This shows a clear reduction in φ close to the dislocation core. Figure 2(c) shows an averaged line profile across the core; to reduce effects due to the dislocation deviating from endon orientation, this profile is taken left to right across the phase map, approximately along the line of greatest phase gradient, which we expect to be perpendicular to the trace of the dislocation. For convenience, the phase variation has been converted to a change in the inner potential ΔV_0 assuming the measured foil thickness, $t = 67$ nm, remains constant across the core.

The solid curve in Fig. 2(c) is a theoretical fit assuming that the dislocation is negatively charged and that the excess electrons are associated with deep acceptor states on the core, and are thus relatively localized. For simplicity the dislocation has been assumed to be exactly endon. Assuming a line charge of N_L electrons per unit distance along the core and neglecting screening by bound or free charges, the field $E(r)$ at distance r from the core is given by

$$E = -N_L e/2\pi\varepsilon\varepsilon_0 r\,, \tag{3}$$

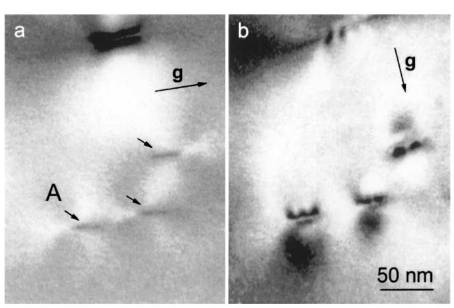

FIG. 1. Two beam bright field electron micrographs in (a) $\mathbf{g} = 1\ -1\ 0\ -1$ and (b) $\mathbf{g} = -1\ \ -1\ 2\ 0$ showing a group of threading edge dislocations (arrowed). The plan-view GaN sample is tilted to show the dislocations obliquely inclined. The edge dislocations show only residual contrast in (a) consistent with $\mathbf{g} \cdot \mathbf{b} = 0$.

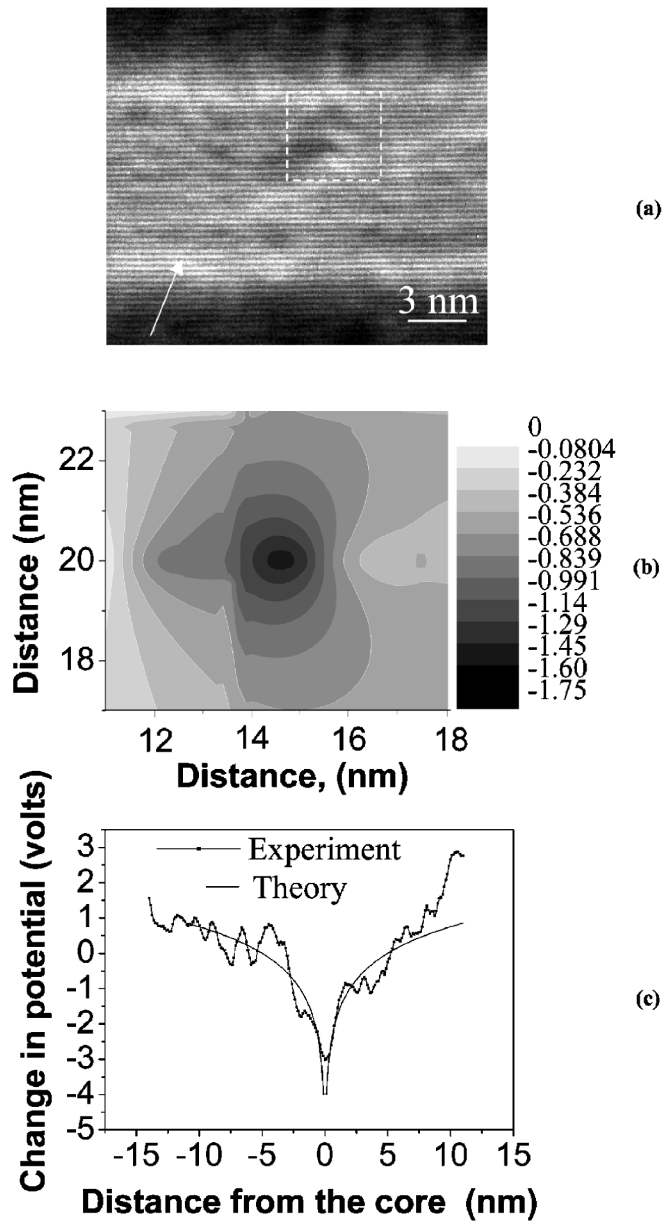

FIG. 2. Electron holography studies of dislocation A in Fig. 1, (a) electron hologram with a fringe spacing of about 0.3 nm; the dislocation is in the center of the outlined box and the arrow indicates the projection of [0001] and (b) phase map from the boxed region showing a reduced phase ϕ near the dislocation core (measured in radians from an arbitrary zero), (c) the dotted line shows a line scan across the map in (b) taken in the horizontal direction where the phase gradient is near maximum. The phase is plotted as an equivalent change in inner potential assuming no pitting at the dislocation and is compared with theory (solid line).

where e is the electronic charge, ε is the dielectric constant ($= 10.4$ for bulk GaN), and ε_0 is the permittivity of free space. Thus taking $E = \partial V_0/\partial r$ (V_0 is conventionally positive) and integrating gives

$$V_0 = (N_L e/2\pi\varepsilon\varepsilon_0)\ln(r) + A, \tag{4}$$

where A is a constant. The curve in Fig. 2(c) is from Eq. (4) using $N_L = 4 \times 10^7\ \mathrm{cm}^{-1}$, equivalent to 2 electrons$/c$, and fitting the experimental profile asymptotically at $|r| > 10$ nm. Close to the dislocation core, where the potential is rapidly varying, we expect Eq. (4) to overestimate the magnitude of ΔV_0. This is partly since the dislocation is not exactly endon, although this problem has been reduced by the choice of profiling direction (see earlier). However, assuming a dislocation line direction of [0001] (which is not inconsistent with the results; see Fig. 2) and an estimated tilt of 7×10^{-2} rad from [0001], the projected dislocation line length is ± 2.5 nm about its center, which provides an upper bound to the spatial resolution. Within atomic distances of the core, the assumptions that the dielectric constant takes its bulk value and that the line charge is localized are also questionable. At large distances from the core, screening due to free and fixed charges should be considered. Although free carriers will be produced by scattering of the incoming electron beam, we have no evidence that beam intensity affected our results. Thus, assuming that screening is due solely to ionized donors, which are both uniformly distributed and 100% ionized, complete screening should be achieved within a cylinder, radius R, where $R = (N_L/\pi N_D)^{1/2} = 47$ nm. Incorporating the effects of screening reduces ΔV_0, although the effect is minimal in Fig. 2(c) ($<0.01\%$ correction for $r < 10$ nm). The effects of fringing fields may also be ignored for $r \ll t$, which is the case here [11].

A further uncertainty is whether surface pitting occurs in the vicinity of the dislocation core, since a reduction in t would also cause a reduction in φ [Eq. (2)]. While substantial pitting is not expected at edge dislocations (0.5 nm as reported by Hansen *et al.* [7]), we have carried out a range of experiments to eliminate this factor. For example, Fig. 3 shows the phase change $\Delta\varphi$ plotted for dislocations in a range of foil thicknesses. Whereas the phase change due solely to pitting is expected to be independent of foil thickness, there is clearly a linear decrease with foil thickness, with no evidence of an offset due to pitting.

The results in Figs. 2 and 3 confirm that edge dislocations in n-GaN are negatively charged. The estimated charge of 2 electrons$/c$ is, of course, extremely high. This is, however, comparable to the estimate of 1 electron$/c$ required to fit transport data [6]. It is also worth noting that electron energy loss spectroscopy (EELS) studies of edgeon $\{10\bar{1}0\}$ boundaries in GaN films suggested an excess charge of 1.5 electrons/boundary atom [12]. In both cases the authors ascribed the charge to the presence of Ga vacancies in the defect core. This appears at odds with the

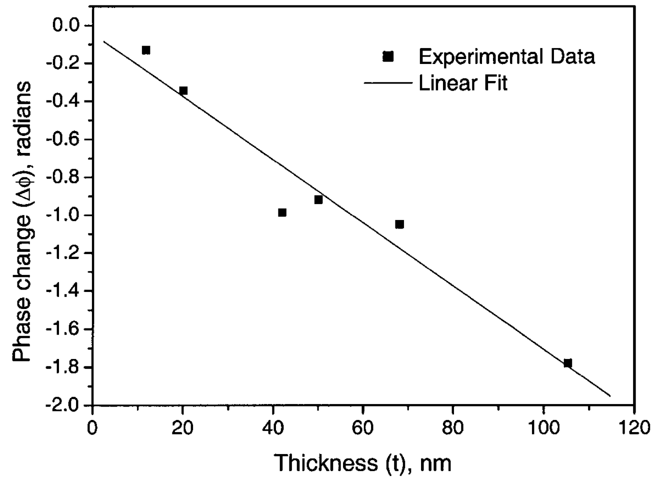

FIG. 3. Phase change, $\Delta\varphi$, for different edge dislocations as a function of foil thickness t; $\Delta\varphi$ was estimated by comparing the background phase, extrapolating from regions far from the dislocation, with a phase average within 2 nm of the core.

work of Xin *et al.* [13] who reported EELS and atomic resolution Z contrast from endon edge dislocations in n-GaN. The maximum Ga vacancy concentration in the core column was estimated at $0.15/c$. The corresponding EELS data for the N K-edge showed no evidence of absorption below the band edge, indicating that any band-gap states were fully occupied. This is consistent with a negatively charged core but, even assuming a charge of three electrons/vacancy [12], the maximum line charge should not exceed $0.45/c$. The origin of our line charge, therefore, remains uncertain.

In conclusion, we have demonstrated, for the first time, that electron holography can be used to measure the charge on dislocations in semiconductors. Our results confirm that edge dislocations in n-GaN are negatively charged and that a simple model applies, giving a charge of about 2 electrons$/c$, or 4×10^7 electrons cm^{-1}. The method could be extended to screw and mixed dislocations in GaN. However, these dislocations show much stronger contrast than edge dislocations in the near endon geometry, due to surface relaxation associated with the Eshelby twist [14]. This is expected to influence the phase profiles; however, provided the strain fields and diffracting conditions are known, the effects on the phase profiles could, in principle, be determined accurately (e.g., see Ref. [9]).

We are grateful to the U.K. Engineering and Physical Sciences Research Council for supporting this work under Grant No. GR/M85371.

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