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Electron-beam-induced damage in wurtzite InN

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Knock-on type damage with ejection of nitrogen atoms from a sample was observed in wurtzite InN during irradiation by 100 keV electron beam in scanning transmission electron microscope. Comparison of the measured integrated intensities of nitrogen K and indium $M_{4,5}$ edges with calculated mass-loss provided a method to measure the energy of vacancy-enhanced displacement in InN for nitrogen atoms, which was found to be 4.6 eV. The results were also applied to predict the rate of electron beam induced damage that will occur in InN specimens with different thicknesses. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1543642]

Before conducting experiments on a sample with electron microscopy or spectroscopy, an understanding of the nature and rate of the electron-beam-induced damage of the specimen occurring during observations is essential. This is especially important for materials such as InN that contain low atomic number elements. These suffer considerable energy transfer under irradiation by, for example, 100 keV electrons. InN is a promising candidate for fabrication of high-performance high electron mobility transistors or light-emitting diodes[1] as a result of the growth of high-quality wurtzite InN.[2,3] Electron energy-loss spectroscopy (EELS) with a scanning transmission electron microscope is a reliable method for measurement of basic electronic structure characteristics such as the densities of the unoccupied electronic states,[4] energies of the interband transitions, plasmon excitations, etc. However, useful quantitative EELS can only be achieved with great care in minimization of the radiation damage.

A common effect of the radiation-induced damage in materials for 100 keV beams is mass-loss[5,6] (sputtering of the atoms from the surface layer), while ionization, segregation, and lateral diffusion can also occur. Medlin and Howitt have developed a simple theoretical model to describe the dynamic changes in the specimen,[7] that was later generalized for multicomponent materials.[8] This model is adapted here to characterize damage in InN. In this letter, we report the results of EELS measurements of the electron-beam-induced damage in wurtzite InN that are compared with results of the theoretical model. This comparison allowed determination of the energy for vacancy-enhanced displacement of nitrogen atoms, i.e., the energy needed for displacement of a N atom into a nearby vacancy. At higher electron energies (e.g., >400 keV) dominant mechanism reflects atom displacements within bulk (see, for example, Regnier *et al.*).[9]

The wurtzite InN studied here was grown by conventional molecular beam epitaxy on a sapphire substrate with a 200 nm AlN buffer layer.[3] The tripod polishing technique[10] was used to prepare plan-view specimens for scanning transmission electron microscopy (STEM) from this wafer with the c axis oriented close to the incident beam. Details on the Cornell 100 keV UHV VG HB501 STEM with about 2 Å focused beam used for these measurements can be found elsewhere.[4]

Annular dark field (ADF) images of the InN taken right after 70 s exposure by 100 keV focused electron beam are shown in Figs. 1(a) and 1(b), where characteristic white spots of the damage can be seen. At first glance, composition sensitive ADF images indicate changes in local stoichiometry in the exposed areas.[11]

To understand the phenomena, several experiments under identical conditions were performed with simultaneous recording of the intensities of the nitrogen K and indium $M_{4,5}$ edges. The results are presented in Figs. 1(c) and 1(d), where decay of the intensity of the nitrogen K edge can be clearly seen. Unlike the nitrogen K edge, however, the indium $M_{4,5}$ edge loses fine structure but not intensity. The changes in the integrated intensities for both the N K edge

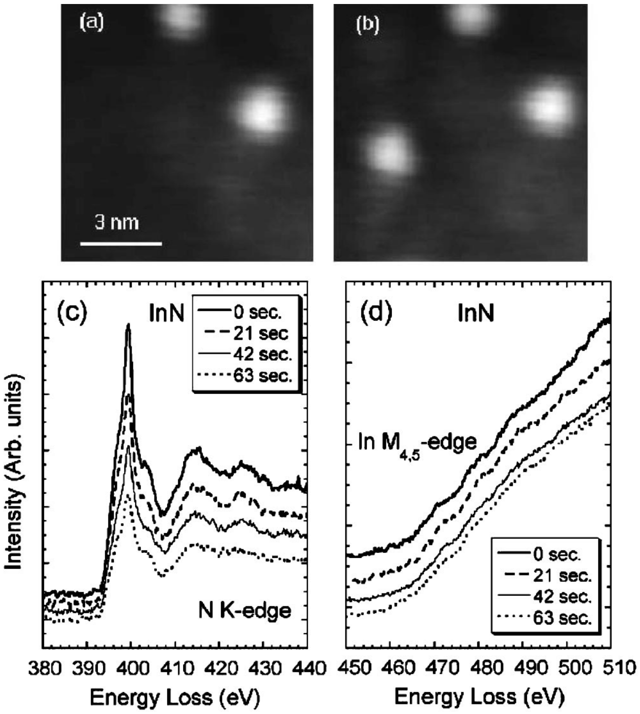

FIG. 1. (a) and (b) ADF images of wurtzite InN with characteristic damaged areas (bright spots). The evolution of the damage after the first 63 s of exposure: (c) effect on the N K edge and (d) on the In $M_{4,5}$ edge. Each spectrum was recorded with 5 s acquisition.

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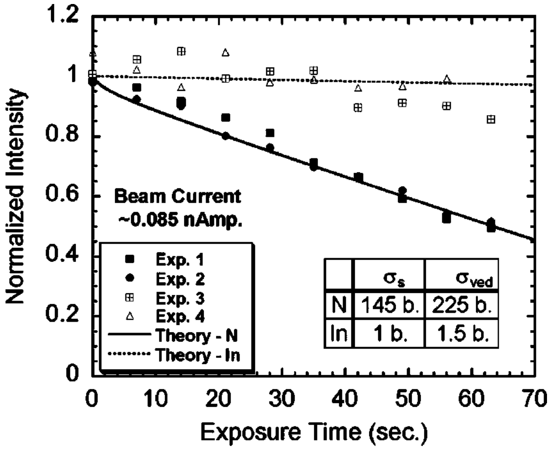

FIG. 2. Changes in the integrated intensities of the nitrogen K edge and indium $M_{4,5}$ edge with exposure time. Data from experiments 1 and 2 are for nitrogen K edge and 3 and 4 for indium $M_{4,5}$ edge obtained in spot mode. Solid and dashed lines are the solution of the coupled differential equations with the values of the cross-sections presented. The intensities are normalized to the first spectra.

(integrated from 390 to 420 eV) and the In $M_{4,5}$ edge (from 440 to 480 eV) during the first 70 s of exposure are measured. The results of four different experiments are presented in Fig. 2. In these experiments, measured beam current was about 8.5×10^{-2} nA. For these thicknesses, spectra do not show noticeable effects on fine structure until after at least 10 s exposure at these currents.

The intensity of the core-level EELS is proportional to the number of the corresponding atoms in the exposed area. Thus, these observations reflect the ejection of the nitrogen atoms out of the sample leaving behind indium-rich InN. Similar knock-on type damage has been reported for wurtzite GaN.[12]

According to the model[7,8] mentioned earlier, dynamic mass-loss in the InN irradiated by an electron beam can be described by the following coupled differential equations:

$$\frac{dN_j^i}{dt}=J\{\sigma_{\rm ved}^i[(N_{j+1}^i-S_{j+1}^i)f_j-(N_j^i-S_j^i)f_{j-1}]-\sigma_s^iS_j^i\}, \tag{1}$$

where N_j^i and S_j^i are the numbers of total and surface atoms of type i ($i=\mathrm{N}$ or In) in the layer j in the area of exposure, J is incident beam current density, $\sigma_{\rm ved}^i$ and σ_s^i are the cross-sections for vacancy-enhanced displacement (VED) and surface sputtering of the atoms, and f_j is the probability that a site in layer j is vacant.

All these parameters can be obtained from basic structural characteristics of the material except the VED and surface sputtering cross-sections, which can be calculated using Mott cross-sections for electron-atom knock-on collisions with relativistic electrons.[13–15] The electron-atom interaction may result in either sputtering of the latter from the surface if it is a surface atom, displacement to a neighboring vacancy, or bulk displacement. The calculated cross-sections[15] for surface sputtering and bulk displacement for nitrogen and indium atoms are presented in Fig. 3. The cohesive energy per bond of InN is about 1.9 eV,[16] which suggests that the surface bonding energy should be within a 5–6 eV energy range. The energy for bulk displacement is approximately five times bigger than the surface bonding energy.[15] These were the critical numbers used in the calculations of Mott cross-sections. As seen in Fig. 3, the cross-sections of bulk displacements are several orders of magnitude smaller than those for surface sputtering and, therefore, their contribution in mass-loss Eq. (1) is negligible.

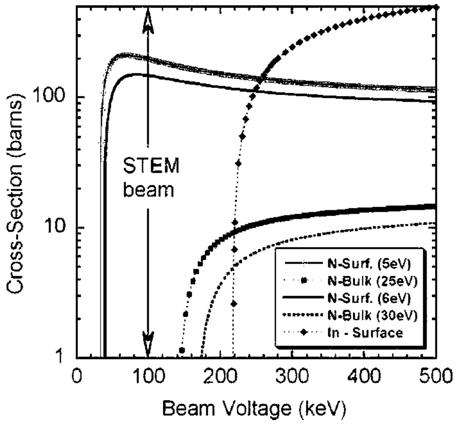

FIG. 3. Calculated Mott cross-sections of surface sputtering and bulk displacement for nitrogen and indium atoms in InN for different acceleration voltages of incident electrons.

The solution of the coupled Eq. (1) has two features. First, strong surface sputtering occurs ejecting nitrogen atoms from the exit surface and later, when the sputtering and VED rates balance, a steady state is reached and removal of the nitrogen from the exit surface takes place (see Fig. 2). At the same time, the In content remains almost unaltered. Thus, the damage model describes the N being swept out of the sample.

By comparing the results of the numerical solution of Eq. (1) with the measured integrated intensities of the nitrogen K and indium $M_{4,5}$ edges (see Fig. 2), the cross-sections for vacancy-enhanced displacement for the nitrogen atoms can be obtained. The best fit to measurements are shown in Fig. 2, which corresponds to the solution with a VED cross-section, $\sigma_{\rm ved}^N\approx225$ barns. The thickness of the specimen, used in these calculations, was estimated from low-loss EELS: $d\approx\lambda_{\rm Pl}\times I_{\rm PL}/I_{\rm ZL}=210\pm10$ Å, where the calculated[17] plasmon generation mean-free-path was $\lambda_{\rm Pl}\simeq122$ nm, and $I_{\rm PL}$ and $I_{\rm ZL}$ are intensities of plasmon-loss and zero-loss. Since there is some uncertainty in the value for energy of surface sputtering (5–6 eV), it is useful to estimate the sensitivity of the VED cross-section to this parameter. The calculations show that the VED cross-section should change only a few percent to accommodate the changes from 5 to 6 eV for surface sputtering energy, which correspond to changes in $\sigma_{\rm s}^N$ from 195 to 145 barns. Note here that the changes in $\sigma_{\rm s}^N$ and $\sigma_{\rm ved}^N$ are in opposite directions, e.g., a decrease in σ_s^N must be compensated by increase (but small) of $\sigma_{\rm ved}^N$ and vice versa.

To obtain the energy of the vacancy-enhanced displacement for nitrogen atoms in InN the dependence of the Mott cross-section on the energy transferred to an atom was used. For 100 keV incident electrons, it is presented in Fig. 4. From the measured cross-section (225 barns), Fig. 4 gives an estimate of 4.6 eV for the VED energy. We should note here that the accuracy in determination of the VED energy is primarily limited by the mass-loss model.[7,8] Simplifications, such as the assumption that only forward scattering of atoms

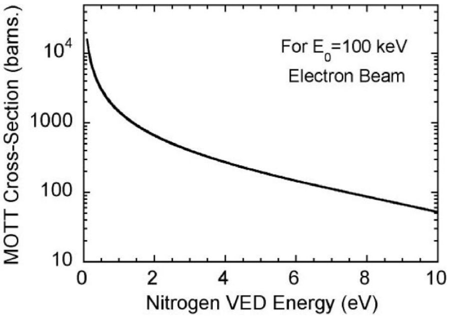

FIG. 4. Mott cross-sections calculated for 100 keV incident electrons as a function of energy transferred to atoms.

occur or ignorance of lateral diffusion and segregation, may introduce some error in the value of the VED energy.

The results of these observations were also used to predict the mass-loss in InN under the 100 keV electron beam for different thicknesses of the specimen. Calculations for the 100, 210, and 500 Å thicknesses based on the values obtained earlier were performed and results are presented in Fig. 5. As can be seen here for a 100 Å thick specimen 170 kC/cm^2 radiation is enough to remove all nitrogen from the sample.

Looking back to Figs. 1(a) and 1(b), the appearance of bright spots in ADF images of damaged areas can not be explained by simple Z-contrast (Z is an average atomic number of the material) when reduction in mass-thickness is observed. While the exact origin of the bright spots is not yet quite clear, the orientation of the matrix crystal can result a low intensity ADF signal if it is not perfectly aligned to the zone axis. Under these conditions, damaged regions of disorder can scatter more strongly than the crystalline matrix.

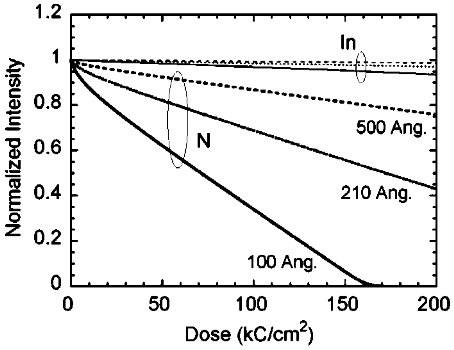

FIG. 5. Changes in amount of nitrogen and indium atoms for different specimen thicknesses irradiated by 100 keV electron beam. The results are normalized to original numbers of atoms.

In conclusion, knock-on type damage with ejection of primarily nitrogen atoms from the sample was observed in wurtzite InN during electron beam irradiation in STEM. Comparison of the measured integrated intensities of nitrogen K and indium $M_{4,5}$ edges with calculated mass-loss using Meldin and Howitt's model provided a nondirect method to measure the energy of the vacancy-enhanced displacement for nitrogen atoms in InN and the value of 4.6 eV was obtained. The results were also applied to predict the damage that will occur in InN specimens of different thicknesses.

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