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Interface-mediated suppression of radiation damage in GaN

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Interfaces are often sinks for radiation-generated defects and could either promote defect recombination or cause detrimental disorder accumulation. Here, we study (0001) GaN irradiated with 500 keV Xe ions at room temperature. Results show that, when point defects are generated within \sim 50 nm from the surface, they experience efficient recombination without any measurable increase in the rate of surface amorphization. Our findings provide clear experimental evidence of efficient suppression of radiation damage by an interface in a non-metallic material.

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Radiation damage is a limiting factor for several technologies. These include ion implantation doping and etching of semiconductor devices, structural and fuel materials for nuclear power reactors, and electronics for use in a radiation environment [1]. Minimizing undesirable radiation damage is a common challenge in all these fields. A strategy that has recently been pursued by a number of research groups for designing radiation-resistant materials is to maximize the number of interfaces such as free surfaces, grain boundaries or interfaces between dissimilar materials [2]. This strategy is based on a well-known concept that interfaces are potential sinks for radiation-generated mobile defects and impurity atoms. If trapping of defects at interfaces leads to defect recombination, increasing the number of interfaces will improve radiation resistance. There is indeed some experimental evidence of an interface-mediated improvement of radiation resistance in several metallic systems. Examples include nanocrystalline metals (Pd [3], Au [4], Ni [5], Cu-0.5Al₂O₃ [5] and TiNi [6,7]) and oxide-dispersion-strengthened steels [8].

The consequences of the interaction of radiationgenerated mobile defects with interfaces, however, are presently not so optimistic for a large class of non-metallic inorganic materials. While interfaces could potentially promote defect recombination, as appears to be a case for some metals [2–8], they could also act as the primary nucleation site for detrimental disorder accumulation, stoichiometric imbalance and phase transformations. For example, the surface proximity in Si (arguably the most extensively studied non-metal) inhibits recombination of radiation-generated defects and hence severely deteriorates material's radiation resistance. Indeed, it has been known for a long time that the Si surface (more specifically, the interface between Si and the native surface oxide layer) is a defect sink and a preferential nucleation site for amorphization [9–12]. The fact that the Si surface also strongly inhibits defect recombination has recently been demonstrated by van den Berg et al. [13,14]. A similar effect of a decreased radiation resistance has also been reported for Si [15], Ge [16], SiC [17,18], SnO₂ [19] and ZrO₂ [20] nanocrystalline materials.

An improved resistance to complete lattice amorphization has been reported for nanocrystalline forms of complex oxides, MgGa₂O₄ [21] and Gd₂(Ti_{0.65}Zr_{0.35})₂O₇ [22], with grain sizes of \sim 8 and \sim 20 nm, respectively. Subsequent study [23], however, has pointed to a dominant role of stoichiometric disorder (rather defect recombination by an interface) in Gd₂(Ti_{0.65}Zr_{0.35})₂O₇ nanocrystals in determining their radiation resistance. Some mechanisms other than defect recombination at interfaces are also likely responsible for a high stability of MgGa₂O₄ nanocrystals to amorphization reported in Ref. [21] since, in the same report, the MgGa₂O₄ surface has been demonstrated to be a nucleation site for disorder accumulation. Hence, we must conclude that unambiguous experimental evidence of suppression of radiation damage by an interface in a non-metallic material is still to be demonstrated, while numerous

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recent reports have suggested adverse effects of interfaces in this class of materials [9–20].

For another extensively studied non-metallic material, GaN, previous experiments have shown that the GaN surface acts as a defect sink and is a nucleation site for amorphization [24]. Ion bombardment results in layer-by-layer amorphization proceeding from the sample surface. A recent experimental study [25] has further revealed that the surface-related interface, as compared to stable radiation damage in the crystal bulk, is a more efficient sink for mobile point defects with respect to both processes of point defect recombination and trapping. Such a surface-related interface is either the sample surface itself (i.e. the solid/vacuum interface) at low ion fluences or the amorphous/crystalline (a/c) interface of the surface amorphous layer (SAL) for fluences above a threshold of ~ 1 displacement per atom (DPA) for the SAL formation [24,25]. It is, however, not clear whether mobile defects trapped at the interface experience annihilation or contribute to detrimental surface amorphization. In other words, does the interface improve or deteriorate a material's radiation resistance?

In this letter, we demonstrate that the surface-related interface in GaN acts as an efficient sink for mobile defects, causing their recombination. Defects generated ballistically near the interface recombine without changing the rate of surface amorphization. To our knowledge, this is the first clear experimental demonstration of suppression of radiation-induced disorder by an interface in a non-metallic material. In addition to providing a strategy for minimizing radiation damage in GaN-based devices, our results have important implications for designing radiation-resistant materials.

About 2 μ m thick undoped wurtzite (0001) GaN epilayers, grown on c-plane sapphire substrates by metalorganic chemical vapor deposition, were used as irradiation targets. The 4 MV ion accelerator (National Electrostatics Corporation, model 4UH) at Lawrence Livermore National Laboratory was used for both ion irradiation and ion beam analysis. Implantation was done at room temperature with 500 keV Xe ions to various fluences between 1×10^{14} and 2×10^{15} cm⁻². During irradiation, the sample normal was tilted 7°, 48°, 60° or 72° from the ion beam incidence direction. This was done in order to form near-surface damage layers with a similar overall shape of the displacement profile but with different thickness while maintaining a constant average density of collision cascades (which is known to influence radiation damage in GaN) [26].

For a better comparison of data for different sample tilt angles, we express ion fluences in DPA at either the depth of the maximum nuclear energy loss (R_{pd}) or at the a/c interface. Quoted DPA values, calculated with the srim code (version SRIM-2008.04) [27] with effective threshold energies for atomic displacements of 25 eV for both Ga and N sublattices [26], are the concentrations of ion-beam-generated lattice vacancies, normalized to the atomic concentration of GaN (8.85×10^{22} atoms cm⁻³). In order to account for the fact that, with increasing fluence, the a/c interface moves through regions with different vacancy generation rates, we calculate DPA at the a/c interface following the algorithm from Ref. [26]. During irradiation, beam flux values were kept con-

stant to maintain a constant displacement generation rate of $\sim 6 \times 10^{-3}$ DPA/s at $R_{\rm pd}$.

The damage build-up was measured by Rutherford backscattering/channeling (RBS/C) spectrometry with a 2 MeV ⁴He⁺ beam incident along the [0001] channeling direction. Two detectors, located at 103° and 164° from the incident beam direction, were used to register backscattered particles. The glancing-angle detector at 103° provided enhanced depth resolution (~0.7 nm per channel) near the sample surface, which was required for characterization of SALs. The data measured with the detector at 164° (that had a larger solid angle and, hence, better sensitivity) were used for the analysis of bulk damage. All RBS/C spectra were analyzed with one of the conventional algorithms [28] for extracting the effective number of scattering centers (referred to below as "relative disorder"). Depth profiles of relative disorder were fitted with Gaussians to evaluate positions and intensities of bulk and surface damage peaks.

Figure 1(a) shows srim-predicted depth profiles of lattice vacancies generated in GaN by Xe-irradiation at 7°, 48°, 60° or 72°. These profiles are Gaussian-like (unimodal), with maxima at corresponding $R_{\rm pd}$'s that, as expected, get shallower with increasing sample tilt angle. Representative profiles of stable post-implantation lattice disorder measured by RBS/C are shown in Figure 1(b) and (c) for ion fluences resulting in ~2 and ~5 DPA at $R_{\rm pd}$, respectively. Such experimental profiles are bimodal, with a strong peak at the surface and a bulk damage peak (BDP), whose maximum ($h^{\rm max}$) is deeper than $R_{\rm pd}$. Both the bimodal nature of damage–depth profiles and a shift of $h^{\rm max}$ relative to $R_{\rm pd}$ are consistent with previous observations [24,29–31].

From a comparison of Figure 1(b) and (c), it is seen that, for all tilt angles, both the BDP magnitude and the SAL thickness grow with increasing fluence. More importantly, for both cases of \sim 2 and \sim 5 DPA, decreasing depth of the displacement generation profile (by increasing incident beam angle) dramatically reduces the size of BDP but has a negligible effect on the SAL growth rate. The fact that the SAL growth rate is independent of the proximity of the BDP to the a/c interface is better illustrated in Figure 2(a) (the right axis), showing the ion fluence dependence of the SAL thickness, which was calculated as the thickness measured by RBS/C plus the thickness of the sputtered layer predicted by ballistic calculations. The nonlinear growth of the SAL at fluences $\lesssim 5$ DPA (at the a/c interface) is consistent with previous reports [26,29]. The sputtering yield was calculated with the SRIM code [27] with values of surface binding energies adjusted to 3.6 eV for both Ga and N in order to match the ion-energy-dependent sputtering yield data for GaN reported in Ref. [32]. According to such calculations, the sputtering rates are 0.17, 0.39, 0.40 and 0.55 nm DPA⁻¹ (with fluences expressed in DPA at $R_{\rm pd}$) for 7°, 48°, 60° and 72° cases, respectively. The fact that shallower implants result in lower bulk damage is better illustrated in Figure 2(b), showing an ion fluence dependence of the level of relative disorder at the BDP maximum. For all four sample tilt angles, with increasing fluence, the BDP level increases, approaching a saturation value of \sim 45%. Such a behavior of the BDP maximum, including the damage

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