Carrier and photon dynamics in a topological insulator Bi2Te3/GaN type II staggered heterostructure

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We have demonstrated a type-II band-aligned heterostructure between pulsed laser deposited topological insulator bismuth telluride and metal organic-chemical-vapour deposited GaN on a sapphire substrate. The heterostructure shows a large valence band-offset of 3.27 eV as determined from x-ray photoelectron spectroscopy, which is close to the bandgap of GaN (3.4 eV). Further investigation using x-ray diffraction, Raman spectroscopy, and energy-dispersive x-ray spectrum reveals the stoichiometric and material properties of bismuth telluride on GaN. Steady state photon emission from GaN is found to be modulated by the charge transfer process due to diffusion across the junction. The time constant involved with the charge transfer process is found to be 0.6 ns by transient absorption spectroscopy. The heterostructure can be used for designing devices with different functionalities and improving the performance of the existing devices on GaN.

Research on (In,Al)GaN materials and devices has received a lot of thrust because of its immense potential for various device applications including light emitting diodes, laser diodes, heterojunction bipolar transistors, and high electron mobility transistors. Investigations of both electronic and optoelectronic devices have gained a lot of momentum on GaN based material systems. The spontaneous and piezoelectric polarizations along with the potential to tune the bandgap from infrared (0.8 eV) to ultraviolet (6.1 eV) region make this material system very attractive. Device structures with various degrees of quantum confinement are also being investigated aggressively for quantum effect devices such as single-phonon emitters and other nanophotonic devices. Efforts are now being directed to grow materials of other families on GaN to develop multifunctional devices on the same substrate. Growth of ferromagnetic and other functional materials on GaN is being investigated in this respect. Here, we have reported pulsed laser deposition (PLD) of bismuth telluride (Bi$_2$Te$_3$) on GaN. Bi$_2$Te$_3$ is a V-VI narrow band-gap (0.18 eV) semiconductor and it is best known for its room temperature thermoelectric properties. Bi$_2$Te$_3$ is also a topological insulator and it is used for its exotic properties like bulk insulating gap and metallic graphene-like surface states and spin-momentum locking.

While both Bi$_2$Te$_3$ and GaN are attractive choices as materials for various applications, we have observed a type-II band alignment for Bi$_2$Te$_3$/GaN heterostructure with large valence band offset. The near conduction band edge alignment and large valence band offset offer advantages for various devices with potential applications in intra sub-band devices due to the spatial separation of the electrons and holes, heterojunction bipolar transistors for large gain, and bias-tunable sources of radiation of energy less than the bandgaps. Photo-generated electrons and holes can diffuse across the junction and the process can enhance or deplete the carrier densities depending upon the choice of the GaN doping. In turn, the photon generation dynamics in GaN can be modulated as a result of the carrier transport.
which may be used as an advantage to improve the efficiency of light emitting devices. The time constant involved with the carrier transfer process is determined by using transient absorption spectroscopy.\textsuperscript{19} The Bi$_2$Te$_3$/GaN type-II heterostructure enriches GaN as a choice for material for various electronic and optoelectronic devices.

The GaN is grown on a c-plane sapphire substrate by metal organic-chemical-vapour deposition (MOCVD) technique. The sample is then transferred to a PLD chamber for the growth of Bi$_2$Te$_3$ from a single crystal source prepared by self-flux method. The details of the growth conditions are mentioned in the supplementary material.\textsuperscript{20} Samples with 50 nm of Bi$_2$Te$_3$ are grown on the un-doped n-GaN ($n = 10^{18} \text{cm}^{-3}$) and p-GaN ($p = 10^{17} \text{cm}^{-3}$) substrates. The carrier densities are determined through Hall measurements in the Van der Pauw geometry. A sample with a thin layer (2–3 nm) of Bi$_2$Te$_3$ is also grown on an un-doped GaN sample for x-ray photoelectron spectroscopy (XPS). The thickness of Bi$_2$Te$_3$ is confirmed through x-ray reflectometry (XRR) measurements. Hall measurements on Bi$_2$Te$_3$ indicates that it is n-type in nature with an electron density of $\sim 10^{20} \text{cm}^{-3}$.

Figure 1(a) shows the x-ray diffraction (XRD) pattern which indicates a rhombohedral crystal structure for Bi$_2$Te$_3$. The absence of peaks other than (001) indicates c-axis orientation of the film. The mean size of the crystallite ($\langle d \rangle$) is estimated to be 14.7 nm from Scherrer’s equation $\Gamma = K\beta \cos(\theta)/\lambda$ where $K = 0.9$, $\beta (=1.54 \text{Å})$ is the X-Ray wavelength, $\beta$ is the FWHM, and $\theta$ is the angle of diffraction. The Raman spectra (Fig. 1(b)) shows the characteristic in-plane ($E_g^2$) and out-of-plane ($A_{1g}^2$) vibrational modes at 104.1 and 135.7 cm$^{-1}$, respectively.\textsuperscript{21} The presence of sharp peaks confirms the good crystallinity of the film. The root mean square surface roughness of the film is found to be 9 Å for the 50 nm Bi$_2$Te$_3$ film as determined from atomic force microscopy (AFM) shown in Fig. 1(c). The film is found to be uniform over large area and free of any large ablated particulate, which is generally a problem in PLD grown samples. This can be attributed to the deposition using single crystal target instead of pelletized source. Figure 1(d) shows the energy dispersive spectrum (EDS) of the film over a large area of 60 $\mu$m $\times$ 60 $\mu$m to determine the stoichiometry. The elemental atomic ratio of Bi to Te is found to be 0.67, which is the same as the ideal stoichiometry for Bi$_2$Te$_3$. The electron density and mobility for Bi$_2$Te$_3$ are experimentally determined to be $10^{20} \text{cm}^{-3}$ and 8.3 cm$^2$/V s, respectively, from AC hall measurements which are more suitable for low mobility and thermoelectric materials like Bi$_2$Te$_3$. Figure 1(e) shows the transmission electron microscopy (TEM) image for the Bi$_2$Te$_3$ film grown on GaN. The lattice spacing between adjacent planes is found to be 0.22 nm which corresponds to the d-spacing of (1 1 0) planes of the

![Figure 2](https://example.com/figure2.png)

**FIG. 2.** (a) The XPS spectra of GaN/Sapphire with and without Bi$_2$Te$_3$ thin film. The valence band maxima are determined from linear fitting; (b) a schematic flat-band diagram at the Bi$_2$Te$_3$/GaN interface showing a type-II band alignment; (c) current density versus voltage for the Bi$_2$Te$_3$/n-GaN heterojunction. The current flow is bidirectional due to electrons; (d) current density versus voltage for the Bi$_2$Te$_3$/p-GaN heterojunction. The current flow is unidirectional as holes can only flow from p-GaN to Bi$_2$Te$_3$ at a high enough bias; equilibrium band diagrams for (e) Bi$_2$Te$_3$/n-GaN and (f) Bi$_2$Te$_3$/p-GaN heterostructures. The arrows in red color show the direction of carrier flow.
Bi$_2$Te$_3$. A selected area electron diffraction (SAED) pattern, shown in the inset, indicates the hexagonal lattice of Bi$_2$Te$_3$ and single crystalline growth of the film in this region. The cross-sectional TEM in Fig. 1(f) shows an ordered growth of Bi$_2$Te$_3$ on GaN.

XPS measurements are done to determine the band alignment at the Bi$_2$Te$_3$/GaN heterojunctions. Figure 2(a) shows the XPS spectra of GaN with and without 2 nm of Bi$_2$Te$_3$. The position of the valence band maxima with respect to the Fermi level is determined by extrapolating the leading edge of the valence band spectra to the base line for both the cases. The valence band offset ($\Delta E_v$) is found to be 3.27 eV. Considering the room temperature band gaps for Bi$_2$Te$_3$ (0.18 eV) and GaN (3.4 eV), the conduction band offset ($\Delta E_c = \Delta E_v - \Delta E_h$) is estimated to be 50 meV. Figure 2(b) shows the conduction and valence band alignment for the Bi$_2$Te$_3$/GaN heterojunction, which is of type-II staggered in nature.

The carrier transport across the junction is investigated by fabricating diodes on both Bi$_2$Te$_3$/n-GaN and Bi$_2$Te$_3$/p-GaN heterostructures. The ohmic contacts to n-GaN, p-GaN, and Bi$_2$Te$_3$ are formed by depositing metal stacks Ti/Al/Ni/Au, Ni/Au, and Ti/Au, respectively, in an electron-beam evaporator under a high vacuum (10$^{-7}$ Torr). The metal stacks on GaN are further annealed at high temperature in a rapid thermal processing chamber.22 Figures 2(c) and 2(d) show the room temperature current-voltage characteristics for both the heterostructures. While the Bi$_2$Te$_3$/n-GaN shows bi-directional current flow for positive and negative bias voltages, Bi$_2$Te$_3$/p-GaN shows a rectifying nature. This can be understood from the equilibrium band-diagrams shown in Figs. 2(e) and 2(f) as determined by solving the Schrödinger and Poisson equations self-consistently with experimentally determined band-offsets and carrier concentrations. The majority carrier electrons can flow bi-directionally in the Bi$_2$Te$_3$/n-GaN heterostructure as the barrier height is only 50 meV. The effect of the small barrier is also manifested as a small change in the slope near zero bias. The Bi$_2$Te$_3$/p-GaN shows a large depletion region with a barrier height of 3.27 eV, which is approximately equal to the bandgap of GaN (3.4 eV). The holes flow over the barrier from p-GaN with a large cut-in voltage, and the current flow is unidirectional. A type-II band alignment can therefore explain the observed experimental characteristics.

Type-II nature between Bi$_2$Te$_3$ and GaN brings in additional degree of freedom in carrier transport and heterostructure design for various electronic and optoelectronic devices. The opposite directions for electrons and holes transport, across the junction is corroborated through temperature dependent photoluminescence (PL) measurements. While the PL energy spectra looks similar to that of GaN (see supplementary material), Fig. 3(a) shows the integrated intensity around 3.4 eV for all four samples as a function of temperature. It is observed that the PL intensity increases for Bi$_2$Te$_3$/p-GaN in comparison to that of p-GaN. This is due to the diffusion of electrons from Bi$_2$Te$_3$ to p-GaN, which is the minority carrier in p-GaN and limits the photon generation. The photo-generated carriers in Bi$_2$Te$_3$ act as a reservoir for electrons supplying extra carriers to p-GaN. Though p-GaN loses holes due to diffusion in the other direction, it does not affect the photon generation much as it is the majority carrier in p-GaN and abundant in supply. The opposite trend in intensity is observed between Bi$_2$Te$_3$/n-GaN and n-GaN, which is due to the loss of the minority carrier holes from n-GaN. The activation energy ($E_a$) is determined by using the relation $I(T) = I_0/[1 + \exp(-E_a/(k_BT))]$, where $I_0$ is the intensity at the lowest temperature, $k_B$ is the Boltzmann constant, and $T$ is the temperature.23 The activation energy is found to increase for Bi$_2$Te$_3$/p-GaN to 40 meV in comparison to 15 meV for the case of p-GaN. This can be attributed to the non-radiative loss of photo-generated carriers in Bi$_2$Te$_3$ at higher temperatures and hence less number of electrons diffuse across the junction at higher temperature. Similarly, the depletion of minority carrier holes across the junction explains the decrease in $E_a$ for Bi$_2$Te$_3$/n-GaN ($E_a = 8.1$ meV) in comparison to that of n-GaN ($E_a = 9.6$ meV).

The diffusion of electrons and holes across the type-II junction can explain both the temperature dependent photoluminescence and electrical characteristics of the heterostructure. The time constants involved with the carrier transfer and photon generation is determined by transient absorption spectroscopy in the transmittance mode, which can capture both radiative and non-radiative processes. The
560 nm is shown in Figs. 4(c) and 4(d), respectively. The GaN as a function of time for a typical probe wavelength for longer time (for optical delays up to 3.3 ns. It may be noted that no fs white light continuum (350–800 nm) at a very low power (3.64 eV) and 2 mW power. The system is probed with a 50 fs pump at 340 nm then explored in the presence of a 50 fs pump at 340 nm with the theoretically predicted value. The heterostructure is the inset of Fig. 3(b). The experimental data matches closely to the simulated data. The interference pattern only slightly. Figure 3(c) shows a magnified view of the simulated and measured characteristics as a function of wavelength. The normalized power spectral density as obtained by fast Fourier transform (FFT) is shown in the inset. The origin of interference pattern is confirmed through measurements using transient absorption spectroscopy details of Bi2Te3/GaN heterostructures. The dynamics is largely dominated by the band-to-band recombination process. An additional time constant of 0.6 ns is required for matching the experimental data for the Bi2Te3/p-GaN heterostructure, which is associated with the carrier transfer process across the junction.

In summary, we have demonstrated a type-II heterostructure between Bi2Te3 and GaN. Electrical and optical characterizations corroborate observations from the physical measurements using XPS. The electron and hole transport across the junction can explain experimental observations. Measurements using transient absorption spectroscopy show the charge transfer process across the junction, which leads to the observed change in photon dynamics. While type-II band alignment opens up additional possibilities, Bi2Te3 on the alloys of (In,Al)GaN may provide additional opportunities through tuneable band-alignments.

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