Efficient Terahertz Generation Within InGaN/GaN Multiple Quantum Wells

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***Abstract*—We have investigated terahertz (THz) generation from InGaN/GaN multiple quantum wells (QWs). For the laser pump power of 400 mW at 391 nm, the highest THz output power is nearly 1 μW. Assuming that the output power quadratically scales up with the interaction length, such an output power corresponds to a normalized output power of 1.7 nW/nm^2. The normalized output power measured on the InGaN/GaN multiple quantum-well structures correspond to probably one of the highest values ever reported among all different semiconductor and nonlinear materials. Following our measurements of the output spectrum, power, and polarization angle as functions of average pump intensity, incident angle, and pump polarization angle, respectively, we have attributed the mechanism for the THz generation from the InGaN/GaN QWs to the radiation of the dipoles, following the generation of the spatially separated electrons and holes under the strong built-in electric fields inherently present in the nitride-based quantum-well structures.**

***Index Terms*—Broadband terahertz (THz) wave, built-in field, dipole radiation, InGaN/GaN quantum wells (QWs), spontaneous and piezoelectric polarizations.**

I. Introduction

BROADBANDterahertz (THz) pulses were generated primarily based on three mechanisms, i.e., optical rectification from second-order nonlinear materials [1], [2], photoconduction or photocurrent surge from electrooptic materials [3], and Cherenkov radiation from ferroelectric materials [4]. It was demonstrated that based on optical rectification in ZnTe [5] and $LiNbO_3$ [6], high peak intensities on the order of 10 MW/cm^2 can be reached. Such intensities are sufficiently high for investigating nonlinear fundamental phenomena in the THz region, such as harmonic generation [7], [8] and THz-enhanced UV emission from gas plasma [9]. It appears to us that semiconductor materials are more promising than ferroelectric materials, such as $LiNbO_3$ for scaling up the output powers. This is due to the fact that semiconductor materials do not suffer from photorefractive damage. In addition, since index of refraction in THz is significantly lower for semiconductors, the THz output power can be inherently higher [10]. Moreover, broadband phase matching is feasible for certain materials, such as ZnTe, GaP, and GaAs under the collinear configuration [10]. Besides optical rectification, photoconduction or photocurrent surge is another primary mechanism for THz generation from semiconductor materials [11]. When semiconductor quantum wells (QWs) were under a bias, the mechanism for THz generation was attributed to the creation of polarized electron-hole pairs [12]. However, in GaAs/AlGaAs QWs, an electric field must be externally applied. It was also demonstrated that THz pulses were generated due to the coherent oscillation of electrons in asymmetric-coupled QWs [13]. In nitride-based heterostructures, however, a strong built-in field originates from spontaneous and piezoelectric polarizations with its magnitude up to 3.1 MV/cm for InGaN/GaN QWs [14] and 9.2 MV/cm for GaN/AlN QWs [15]. The large polarization fields result in significant charge-separation effect in the nitride-based QWs (i.e., InGaN/GaN QWs). It is worth noting that by employing novel QW structures with improved electron-hole wavefunction overlaps, the charge-separation effect can be engineered [16]–[19]. In the past, THz emission from InGaN/GaN QWs was observed without the presence of an external electric field [20]. The mechanism for the THz generation was attributed to dynamical screening of a strong built-in electric field [21]. Such a built-in electric field is inherently present in the InGaN/GaN QWs grown on the [0 0 0 1] direction [14], as mentioned earlier.

In this paper, we report our result following our investigation of efficient THz generation from InGaN/GaN QWs achieved at room temperature. We have directly measured the average THz output power to be as high as 0.96 μW, generated by eight periods of the $In_{0.25}Ga_{0.75}N$/GaN QWs. The characteristics measured on the InGaN/GaN QWs, such as THz output spectrum and power versus incident angle, THz output power and polarization versus pump polarization angle, and THz output power and polarization versus azimuth angle, are essentially the same as those due to the radiation originating from the instantaneous generation of dipoles (i.e., photocurrent surge). Within the range of the pump intensities used for investigating the THz generation, the energy for the band-to-band transition peak exhibits a small amount of blue shift as the pump intensity is increased. Following our analysis, we have concluded that the mechanism for THz generation is primarily the instantaneous generation of spatially separated electron-hole pairs resulting in dipole radiation. Our comparison with the results obtained on the InGaN thin film reveals that the output power from the InGaN/GaN multiple QWs is dramatically scaled up.

II. InGaN/GaN QWs

The growth of InGaN/GaN QWs structures was carried out on a 2.8-μm-thick unintentionally doped GaN (background electron density of $\sim 4 \times 10^{16}$ cm^{-3}) template on a c-plane sapphire substrate by metal–organic chemical vapor deposition

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(MOCVD). The growth of the GaN template was performed at 1080 °C by employing 35-nm-thick low-temperature-grown (T_g = 525 °C) GaN buffer layer. Subsequently, eight periods of $In_{0.25}Ga_{0.75}N$/GaN QWs were deposited on the GaN template by MOCVD at 700 °C. The thicknesses of the InGaN well and GaN barrier layers are 3 and 12 nm, respectively. In addition to the InGaN/GaN QWs, samples of 180 nm $In_{0.36}Ga_{0.64}N$ and 2.8 μm GaN/sapphire thin films were grown for making the comparison with the InGaN/GaN QWs.

III. Experimental Setup

Broadband THz pulses were generated by a coherent radiation beam at the wavelength of 391 nm after frequency-doubling the output beam from a Ti:sapphire regenerative amplifier. Such an output beam can be used to generate electrons and holes in the conduction and valence bands of the $In_{0.25}Ga_{0.75}N$/GaN QWs, respectively. The pulse duration is ∼210 fs and repetition rate is 250 kHz. The excitation beam was focused onto each sample by a positive lens with the focal length of 10 cm. The THz radiation was collimated, and then, focused onto a bolometer or a pyroelectric detector by a pair of gold-coated parabolic mirrors.The first mirror was used to collimate the THz output beam, whereas the second one was used to focus it onto each detector. We measured the THz output beams in both the transmission and reflection geometries. In order to measure the energy of the band-to-band transition peak from the $In_{0.25}Ga_{0.75}N$/GaN QWs, we used the output beam from the same amplifier to generate the luminescence signal.

IV. THz Generation

In this section, we present our result on the THz generation from the $In_{0.25}Ga_{0.75}N$/GaN QWs, as well as the comparison studies with the InGaN and GaN thin-film samples.

Based on our measurements of the output powers from the $In_{0.25}Ga_{0.75}N$/GaN QWs, the highest output power is 0.956 μW for an average pump power of 400 mW at 391 nm. The densities of the photogenerated carriers following the absorption of the pump pulses are expected to increase linearly with the period of the QWs. By assuming that the output power quadratically increases with increasing the carrier densities, we have estimated the output power from a single period of the QW with the well thickness of 3 nm to be about 120 nW. Since the noise level of our pyroelectric detector operating at room temperature is 230 pW, such an output power provides us with a dynamic range as high as 520. We can also determine the output power normalized by the square of the effective interaction length (i.e., 576 nm^2) to be 1.66 nW/nm^2. We believe that the normalized output power measured on the InGaN/GaN multiple QWs is probably one of the highest values ever reported among all different materials. For comparison, let us consider a GaSe crystal. According to our recent result [22], the average output power generated was 5.4 μW for a GaSe thickness of 145 μm. Such an output power corresponds to the normalized output power of 2.6×10^{-7} nW/nm^2 (i.e., the output power normalized by the square of the thickness). Therefore, the output power normalized by the square of the effective interaction length, obtained by us on the InGaN/GaN multiple QWs, is seven orders of magnitude higher than that from the GaSe crystal. On the other hand, the average output power of 0.956 μW is a factor of about 1900 (i.e., three orders of magnitude) higher than that measured by us on a 25-nm-thick InN film sample [23].

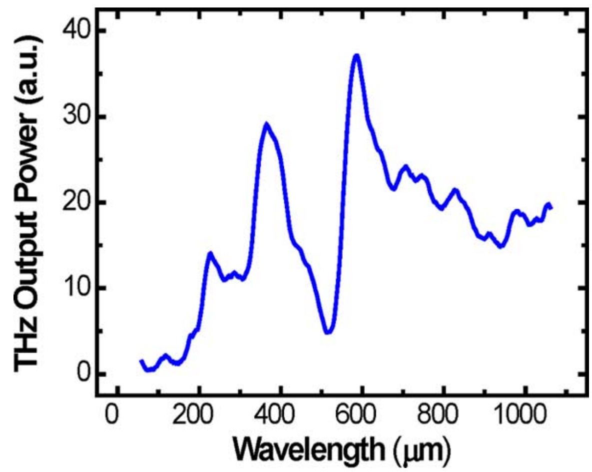

Fig. 1. Typical spectrum of THz wave emitted by $In_{0.25}Ga_{0.75}N$/GaN QWs. The huge dip at 520 μm and other modulations were caused by water-vapor absorption.

In comparison, the output power from the 180-nm $In_{0.36}Ga_{0.64}N$ thin-film sample was measured to be 112 nW. Assuming the quadratic dependence of the output power on the thickness, the output power from a 3-nm $In_{0.36}Ga_{0.64}N$ film only is expected to be 31.1 pW, which is a factor of about 3800 lower than that deduced from a single InGaN/GaN QW. On the other hand, for the 2.8-μm GaN film, the THz signal was buried by the noise of the bolometer. Such comparisons among the output powers generated from three samples reveal that the mechanism for the THz generation from the InGaN/GaN QWs must be fundamentally different from that for the InGaN thin-film sample.

We have measured the spectrum of the THz output beam generated by the $In_{0.25}Ga_{0.75}N$/GaN QWs by directly studying the diffraction of the THz beam by a set of the rotating gratings. Based on Fig. 1, we can see that the frequencies of the THz output roughly span the frequency range from 300 GHz to 1.5 THz. The huge dip located at 520 μm (577 GHz) is caused by water-vapor absorption. Based on our measurements of the THz output spectra at different pump intensities, the spectral features illustrated in Fig. 1 are independent of the pump intensities. Therefore, the dynamical screening of the built-in electric field in our QWs is negligible, unlike the results obtained in [20] and [21]. Such a conclusion is supported by the measurement of the dependence of the THz output power on the pump intensity, as shown by Fig. 2. One can see from Fig. 2 that for the pump intensities below 13.6 W/cm^2, the power dependence exhibited a nearly square law. Therefore, within such a range of the pump intensities, we are certain that the dynamical screening of the built-in field is insignificant.

Above 13.6 W/cm^2, the power dependence significantly deviates from the quadratic dependence. We can rule out the screening of the built-in electric field [20], [24], [25], similar to screening the surface field in InP observed in the past [11], as a possible mechanism for causing such a deviation. Indeed, according to our measurement of the energy for the band-to-band transition peak in the QWs, the shift of the peak is roughly 6 meV, as the pump intensity is increased from 1.5 to 30 W/cm^2, as illustrated by Fig. 3. Since the linewidth of the photoluminescence peak is about 100 meV, such a small amount

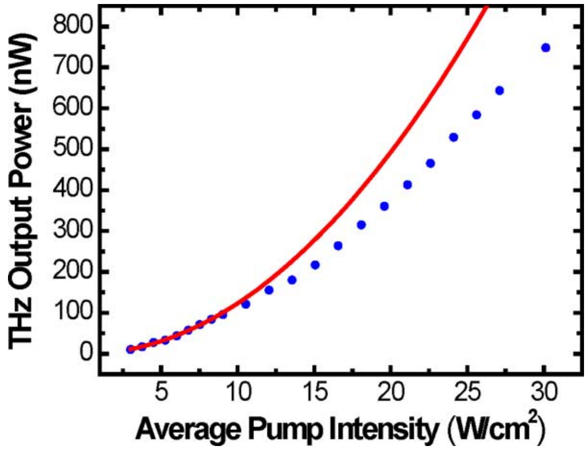

Fig. 2. Average THz output power generated from $In_{0.25}Ga_{0.75}N$/GaN multiple QWs was measured as a function of average pump intensity. Dots correspond to our data. Solid curve correspond to quadratic fit to the first nine pump powers from the low-intensity end.

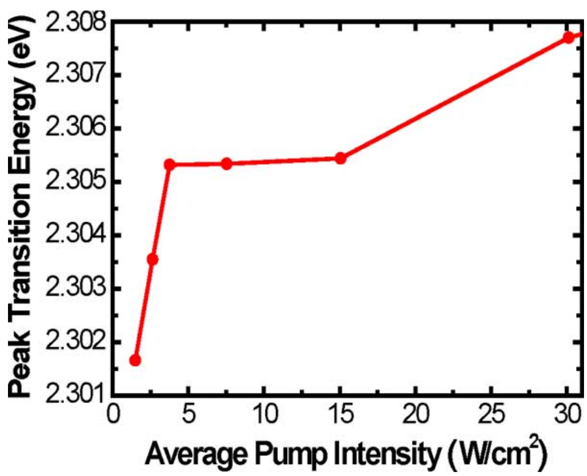

Fig. 3. Transition energy of $In_{0.25}Ga_{0.75}N$/GaN QWs as a function of average pump intensity at room temperature.

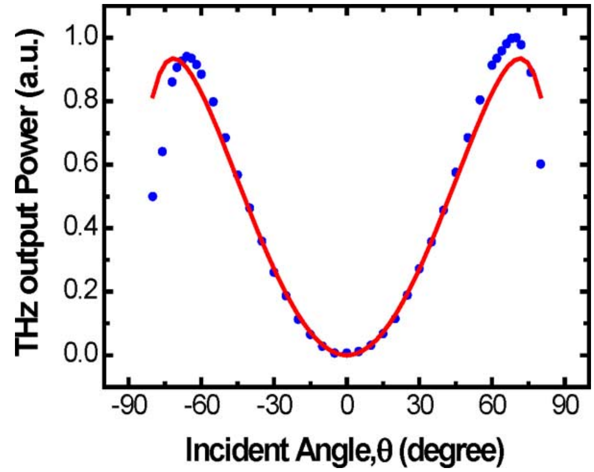

Fig. 4. THz output power generated by $In_{0.25}Ga_{0.75}N$/GaN multiple QWs was measured as a function of incident angle defined as the angle of surface normal being formed with the pump beam. Dots correspond to data points. Solid curve correspond to data fitting using (1). During our measurement, the pump beam was always *p*-polarized.

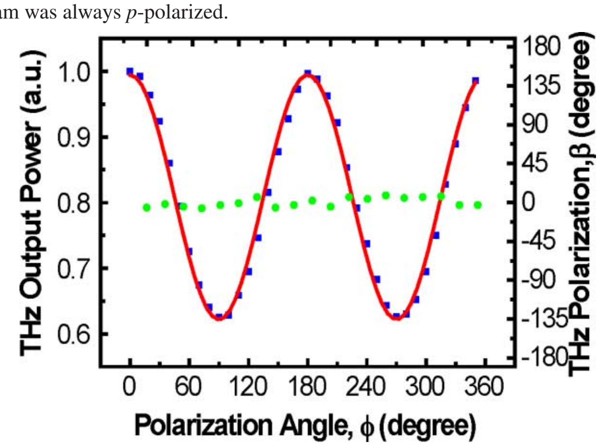

Fig. 5. Polarization dependences of THz output power generated by $In_{0.25}Ga_{0.75}N$/GaN multiple QWs and THz polarization angle. Polarization angle of pump beam is defined as the angle between the pump polarization and the incident plane. Polarization angle of THz wave, defined as the angle between the THz polarization and the incident plane, was determined by wire-grid polarizer. Squares designate average output power as function of pump polarization. Dots correspond to polarization angle of THz beam measured versus pump polarization. Solid curve is a theoretical result after taking into consideration Fresnel reflection.

of the shift determined from the photoluminescence peaks is only 6% of the photoluminescence linewidth, and therefore, it may be subject to a large error. Moreover, compared with the previous result [21], [24], the amount of the blue shift deduced by us is much lower. As we increased the pump intensity above 30 W/cm^2, the photoluminescence peak was further blue-shifted. However, the THz output power was significantly reduced. Such a trend is completely different from that observed in [20]. Therefore, the dynamical screening of the built-in field did not play a significant role within the pump intensities of 13.6–30 W/cm^2. A plausible mechanism causing the deviation observed in Fig. 2 can be the increase in the absorption of the THz wave by the photogenerated carriers, which was previously evidenced and investigated for pulsed-MOCVD-grown InN alloy [25]–[27].

In order to further understand the mechanism for the THz generation in our InGaN/GaN QWs, we first measured the output power of the THz beam propagating in the transmission direction as a function of the incident angle for the pump beam, as shown in Fig. 4. According to Fig. 4, when the incident angle for the pump beam was zero, the THz output power was close to zero. The highest THz output power occurred at an incident angle of 72°, which is close to the Brewster angle for the pump beam. After taking into consideration the Fresnel reflection for the pump beam, the THz output power (P_{THz}) can be approximated by the following expression:

$$P_{\mathrm{THz}} \approx f(\theta)\sin^2(\theta) \quad (1)$$

where *f*(θ) describes the contribution originating from the Fresnel reflection of the pump beam and $\sin^2(\theta)$ represents a typical angle distribution of the dipole radiation. Our data can be well fitted by using (1), which is illustrated by Fig. 4. Thus, we conclude that the angular distribution of the THz radiation is consistent with that of the THz generation due to the dipole radiation.

Besides the angular distribution of the THz radiation, we also measured the dependences of the THz output power and polarization on the polarization of the pump beam and azimuth angle. Fig. 5 illustrates our result following the measurement of the THz output power as a function of the pump polarization

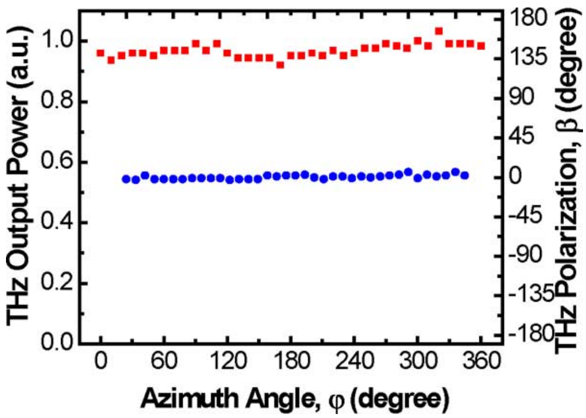

Fig. 6. Dependences of THz output power and THz polarization on azimuth angle for $In_{0.25}Ga_{0.75}N$/GaN multiple QWs. Azimuth angle is defined as the angle between the projection of the pump polarization onto the sample surface and a direction within the sample surface. Squares correspond to average output powers whereas dots represent polarization angles of THz beam.

angle in the reflection geometry. In such a case, we set the incident angle to be around 72° in order to collect the maximum amount of the THz output power. One can see from Fig. 5 that the THz output power periodically oscillates as a function of the pump polarization angle. After taking into consideration the Fresnel reflection of the pump beam, our data can be well fitted by our theoretical curve, as shown by Fig. 5. This implies that the oscillation of the THz output power as a function of the pump polarization angle is primarily caused by the dependence of the Fresnel refraction on the polarization angle of the pump beam. Therefore, the dependence of the THz output power on the pump polarization angle is consistent with that of the THz generation due to the dipole radiation. Furthermore, one can see from Fig. 5 that the THz polarization angle is always around zero, i.e., the THz output beam was p-polarized with its polarization lying in the incidence plane. This is again consistent with our claim that the mechanism of THz generation is the dipole radiation.

We measured the dependences of the THz output power and polarization angle on the azimuth angle from the $In_{0.25}Ga_{0.75}N$/GaN QWs in the reflection geometry. During these measurements, we set the incident and polarization angles of the pump beam to 35° and 0°, respectively. According to Fig. 6, we can see that both the THz output power and the polarization angle do not change when increasing the azimuth angle. Such results are also consistent with the THz generation due to the dipole radiation. Indeed, different azimuth angles would not affect the densities of the photogenerated electrons and holes, and therefore, they would not affect the THz output power and polarization.

After analyzing the data obtained from our measurements, we conclude that the mechanism for the THz generation from the InGaN/GaN QWs is the radiation of the dipoles, following the creation of the spatially separated electrons and holes. Such a mechanism was used to explain the THz radiation from GaAs/AlGaAs QWs under an external field [12]. According to [12], the spectrum of the THz radiation was more or less independent of the external electric field. Therefore, the THz spectrum was primarily determined by the frequency spectrum of the pump beam. On the other hand, according to [10], the low and high frequencies defining the bandwidth of the THz output are inversely proportional to the pulsewidth of the pump laser. Since the pulsewidth for the laser beam used in [12] was 100 fs, the high-frequency components extended well beyond 2.5 THz. In our case, however, since we set the pulsewidth to 210 fs, the high-frequency components were expected to extend well beyond 1.2 THz. According to Fig. 1, this frequency is close to the value estimated from the spectrum. However, according to [21], the spectrum of the THz output was expected to strongly depend on the pump intensity due to dynamical screening of the built-in fields. Such a dependence is absent from our experiment. Moreover, according to [12], the electron-hole dipole moment is proportional to the built-in electric field. Therefore, due to the presence of the extremely high built-in electric fields in our InGaN/GaN QWs originating from the spontaneous and piezoelectric polarizations, we generated sufficiently high output powers. On the other hand, for the 180-nm-thick InGaN thin-film sample, photocurrent surge due to the diffusion and drift of the photogenerated carriers is the dominant mechanism for the THz generation. In such a case, however, the surface field is much lower than the built-in fields inherently present in the InGaN/GaN QWs. Such a huge difference in electric fields is the main reason for a dramatically reduced output power achieved from the 180-nm-thick InGaN thin-film sample.

V. Conclusion

In conclusion, we have efficiently generated broadband THz signals from InGaN/GaN multiple QWs. The THz emission from such a structure is attributed to the radiation of dipoles consisting of the photogenerated and spatially separated electrons and holes upon the illumination of the pump pulses at 391 nm. Considering the fact that the total thickness of the eight periods of the InGaN/GaN QWs is about 24 nm, the measured output power normalized by the square of the total thickness reaches 1.7 nW/nm^2, which is probably one of the highest values ever reported. Such a high normalized output power was achieved by utilizing the very high built-in electric fields inherently present in the QWs to spatially separate electrons and holes in order to instantaneously create large dipoles. The proposed mechanism for the THz generation, i.e., the radiation of the dipoles due to the instantaneous generation of spatially separated electrons and holes, is consistent with our measurements of the spectrum of the THz output as well as the dependences of the THz output power and polarization angle on pump intensity, incident angle, polarization angle of the pump beam, and azimuth angle.

It was predicted in the past that wurtzite GaN/AlGaN QWs possess very high second-order nonlinear coefficients enhanced by strong built-in fields [28]. We believe that these second-order nonlinearities may play an important role under the pump intensities much higher than those used under our investigation. At such a pumping condition, it may be feasible for us to use InGaN/GaN QWs to detect THz waves based on frequency upconversion in second-order nonlinear materials [29], [30], as previously demonstrated. According to [22] and [23], these second-order nonlinear coefficients can be dramatically enhanced under the condition of the resonant excitation.

The unique aspect of nitride-based semiconductor heterostructures, such as QWs offers an opportunity for us to engineer the structure of the QWs in order to further increase the THz output power. For example, for the InGaN/GaN QWs, the THz output power may be further increased by optimizing the widths of the wells and barriers, or by employing novel nitride-based QW structures [16]–[19], [31]. In addition, according to [10], the average output power can be increased to about 40 μW, i.e., by a factor of 40, by reducing the pulsewidth of the pump beam to 60 fs from 210 fs. In the future, we are going to investigate how the THz output power will scale with the temperature and external electric field.

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