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Intense terahertz emission from *a*-plane InN surface

H. Ahn,[1,a)] Y.-P. Ku,[1] C.-H. Chuang,[1] C.-L. Pan,[1] H.-W. Lin,[2] Y.-L. Hong,[2] and S. Gwo[2]
[1]*Department of Photonics and Institute of Electro-optical Engineering, National Chiao Tung University, Hsinchu 30010, Taiwan*
[2]*Department of Physics, National Tsing Hua University, Hsinchu 30013, Taiwan*

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We report a significant enhancement in terahertz emission from the indium nitride (InN) films grown along the *a* axis (*a*-plane InN), relative to the InN films grown along the *c* axis. The primary radiation mechanism of the *a*-plane InN film is found to be due to the acceleration of photoexcited carriers under the polarization-induced in-plane electric field perpendicular to the *a* axis, which effectively enhances the geometrical coupling of the radiation out of semiconductor. In addition, azimuthal angle dependence measurement shows that the *p*-polarized terahertz output consists of a large angularly independent component and a weak component with a distinctive fourfold rotation symmetry. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2892655]

The performance of short-wavelength optoelectronic devices realized by growing III-nitrides along the *c* axis closely depends on the polarization-induced internal electric fields. These fields are due to piezoelectric and spontaneous polarizations and the strain-dependent piezoelectric polarization along the *c* axis ([0001]) of the wurtzite crystals increases with the lattice mismatch in the nitride layers. For the layers grown along *a*- ($\langle 11\bar{2}0\rangle$) or *m*-axis ($\langle \bar{1}100\rangle$) direction, on the other hand, polarization-induced electric field perpendicular to the layer interface can be minimized and the efficiency of the devices can be increased. Recently, due to its narrow bandgap, indium nitride (InN) has received much attention in terahertz range applications, including as an efficient terahertz emitter.[1–5] Up to date, research on InN grown along the *a* axis (*a*-plane InN) were rarely reported mainly due to the technical difficulty in growing high crystalline quality *a*-plane InN films and none was reported for their terahertz emission properties. In addition, for *a*-plane InN, the absence of electron accumulation layer has been predicted and this issue is still under current debate.[6]

Previously, we have reported the enhancement of terahertz radiation (greater than ten times in intensity) from InN nanorod arrays compared to the InN films grown along the *c* axis (*c*-plane InN) and the dominant emission mechanism was proposed to be the photo-Dember effect.[5] Electron accumulation layer at the surface of *c*-plane InN is very thin (<10 nm) and its contribution to terahertz generation is negligibly small. The nonlinear-process-based radiation intensity is less than few percents of total emission for *c*-plane InN. In the present letter, we present even more significant enhancement (>100 times in intensity) from the *a*-plane InN surface excited at a moderate pump fluence. The azimuthal angle dependence measurement shows that terahertz emission from *a*-plane InN has a strong angle-independent response superimposed with a relatively weak angle-dependent component with a fourfold rotation symmetry. We propose that the dominant azimuthal-angle-independent response is due to the accelerated photocarriers in the in-plane electric field of the *a*-plane InN film, while angle-dependent radiation might be due to nonlinear optical processes.

The *a*-plane InN epitaxial film (~ 1.2 μm) was grown by plasma-assisted molecular beam epitaxy (PA-MBE) on *r*-plane $\{1\bar{1}02\}$ sapphire wafer, while the *c*-plane $(000\bar{1})$ InN epitaxial film (~ 2.5 μm) was grown on Si(111) using a double-buffer layer technique.[7] The back side of *r*-plane sapphire wafer was coated with a Ti layer for efficient and uniform heating during the PA-MBE growth. The growth direction of the InN film was determined using a 2θ-ω x-ray diffraction scan. The in-plane epitaxial relationship between *a*-plane InN and *r*-plane sapphire is identical to that reported by Lu *et al.*,[8] such that $[0001]_{InN} \| [\bar{1}100]_{Al_2O_3}$ and $[\bar{1}100]_{InN} \| [11\bar{2}0]_{Al_2O_3}$. Near-infrared photoluminescence was detected from the as-grown *a*- and *c*-plane InN films at room temperature. Unintentionally doped *n*-type carrier concentrations of 7.0×10^{18} and 3.1×10^{18} cm^{-3} and electron mobilities (μ) of 298 and 1036 cm^2/V s were determined by room-temperature Hall effect measurements for *a*- and *c*-plane InN films, respectively.

Terahertz emission from InN epilayer was investigated using a Ti:sapphire regenerative amplifier laser system, which delivers ~ 50 fs optical pulses at a center wavelength of 800 nm with a repetition rate of 1 kHz. For this experiment, the pump laser beam is collimated on the samples with a spot size of ~ 2 mm at the angle of incidence of 70°. The terahertz pulses were detected by free-space electro-optic sampling in a 2-mm-thick ZnTe crystal as a function of delay time with respect to the optical pump pulse.

Figure 1(a) shows the maximum amplitude of *p*-polarized terahertz field from the *a*-plane InN film, compared to that from the *c*-plane InN film. Under the same *p*-polarized pump fluence at ~ 0.24 mJ/cm^2, the amplitude of *p*-polarized radiation component from *a*-plane InN is at least ten times stronger than that from *c*-plane InN. To evaluate our result quantitatively, the peak values of terahertz field from the *c*- and *a*-plane InN films are compared to that from an *n*-type InAs(100) film, one of the strongest semiconductor terahertz emitters, measured under identical experimental conditions [inset of Fig. 1(a)]. Optically excited at the similar pump fluence with our experiment, the terahertz field generated from InAs is typically much larger than that from

a)Author to whom correspondence should be addressed. Electronic mail: hyahn@mail.nctu.edu.tw.

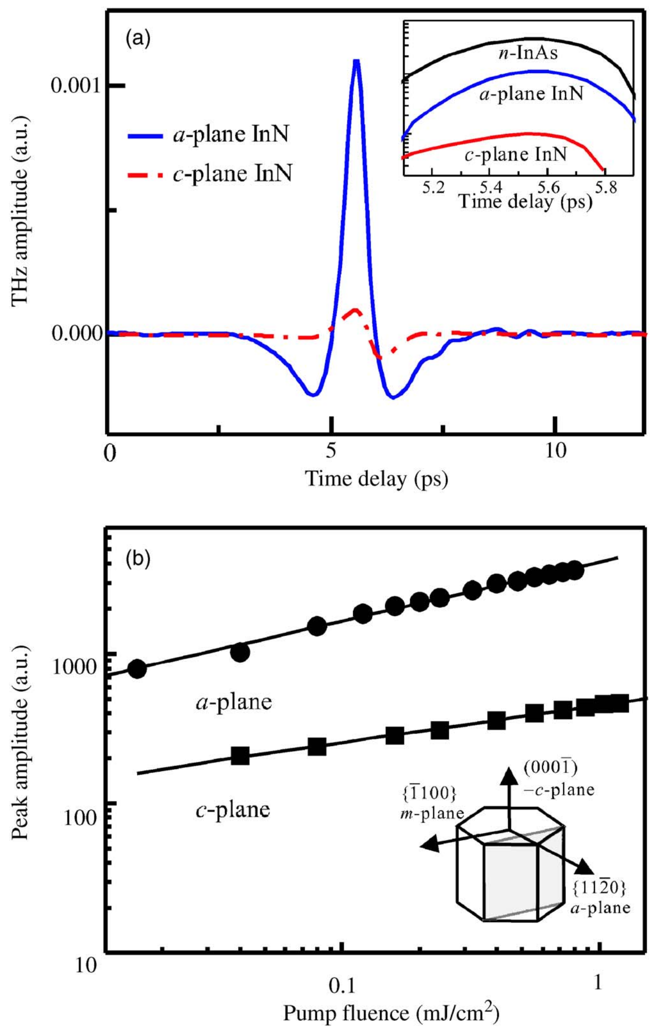

FIG. 1. (Color online) (a) The amplitude of the terahertz emission from *a*-plane InN film (black solid line) compared to that from *c*-plane InN film (red dashed line). The excitation fluence is 0.24 mJ/cm^2. Inset: comparison between *a*-plane InN and *n*-type InAs(100) measured under identical experimental conditions. (b) The amplitude of the *p*-polarized emission field vs pump fluence. The solid line is a linear fit to the data.

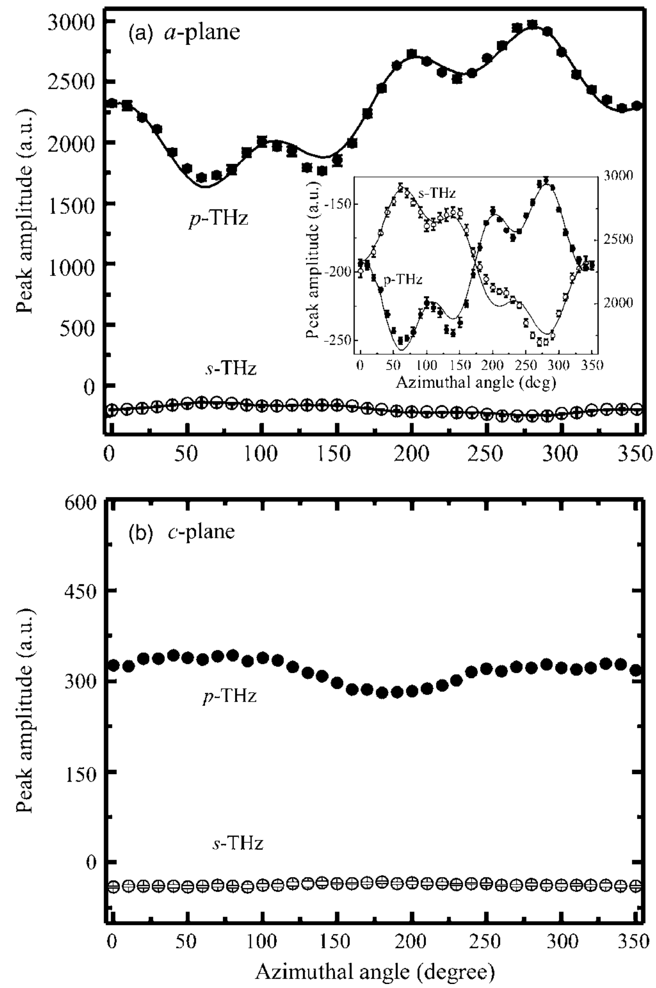

FIG. 2. Peak amplitudes of the *p*- and *s*-polarized terahertz fields vs azimuthal angle rotation of (a) *a*-plane and (b) *c*-plane InN excited at 0.32 mJ/cm^2. Inset in (a) shows the same features with different scales of axis.

c-plane InN (at least 30 times larger[4]). In contrast, the inset shows that the terahertz field generated from *a*-plane InN is about the same order of magnitude as that from InAs. The pump fluence dependence measured at the crystal orientation which gives the maximum emission is plotted in Fig. 1(b) for *a*- and *c*-plane InN. The measured *p*-polarized emission increases linearly with the pump fluence and the saturation of the emission is not observed for pump fluence of up to 1 mJ/cm^2 for both samples.

The azimuthal angle dependence of *p*- and *s*-polarized terahertz fields is measured as the InN samples are rotated about the surface normal (see Fig. 2). Each sample is excited at the pump fluence of ~0.32 mJ/cm^2. The *p*-polarized terahertz field (solid circles) of *a*-plane InN in Fig. 2(a) shows two main features; a large angle-independent field and a weak angle-dependent field with a fourfold rotational symmetry. The apparent angular dependence of the terahertz signal from *a*-plane InN is strikingly different from the terahertz emission from *c*-plane InN, which exhibits no significant angular dependence (<5% of total amplitude), as is shown in Fig. 2(b). The angle-dependent terahertz signals of *a*-plane InN can be qualitatively fitted by a phenomenological form, $E_{\mathrm{THz}}=a\sin(\phi)+b\sin(4\phi)$ and the fitting results are plotted as solid lines in Fig. 2(a). Despite of its small amplitude, *s*-polarized component (open circles) has the same rotation symmetry but has the opposite polarity relative to the *p*-polarized component (see the inset of Fig. 2).

Although angle-dependent radiation is clearly observed for the *a*-plane InN, its contribution to the radiation is less than 20% of the total amplitude. Usually, azimuthal angle-dependent terahertz emission is attributed to nonlinear optical mechanisms. However, nonlinear optical mechanisms, such as the surface nonlinear optical response observed for the (100) InAs (Ref. 9) and resonance-enhanced optical rectification for the *c*-plane InN (Ref. 10) under very high excitation fluence (>2 mJ/cm^2), may not have significant contribution for our measurement since the observed pump fluence dependence of terahertz radiation is linear up to 1 mJ/cm^2, as shown in Fig. 1(b). It reveals that our pump fluence (0.2–0.35 mJ/cm^2) is well below the regime of photocarrier saturation and the onset of the large nonlinear optical mechanisms. Slightly larger linear slope of the *a*-plane InN compared to that of the *c*-plane InN in Fig. 1(b) might be due to the slow increase of nonlinear effects with the increase of pump fluence. A similar fourfold azimuthal angle dependence has been observed for weakly excited (100) InAs under an external magnetic field, which is proposed to be due to an anisotropic intervalley scattering in four equivalent directions.[11] However, the intervalley scattering for InN should be quite small because the excess energy of ~ 1.55 eV pump laser over its bandgap energy is much smaller than the energy gap between the conduction band minimum and the next local minimum (2.8 eV).[2] Therefore, we can also rule out this mechanism. Nonetheless, the clarification of azimuthal angle dependence needs further study, including the detailed knowledge of the nonlinear tensor elements of the sample, which has not yet been known for the *a*-plane InN. Therefore, at this point, we will mainly focus on the large angle-independent component of *p*-polarized terahertz emission from the *a*-plane InN.

It is known that photoexcited carriers generated close to the surface of semiconductors can be accelerated by an appropriate electric field and the resultant transient electric dipole can lead to generation of terahertz pulses. The proper electric field can be provided either externally by separate electrodes in photoconductive antennas or internally by the photo-Dember field or by the electron accumulation field. The contribution of electron accumulation field to terahertz emission can be very small for both *c*- and *a*-plane InN due to the narrow thickness of the electron accumulation layer. For the photo-Dember effect, which is proportional to electron mobility and is independent to the crystal growth direction, its contribution to the radiation from *a*-plane InN ($\mu = 298\ \mathrm{cm}^2/\mathrm{V}$) can be even smaller than that from *c*-plane InN ($\mu = 1036\ \mathrm{cm}^2/\mathrm{V}$). Therefore, the drastic power enhancement observed for *a*-plane InN cannot be explained by either the electron accumulation or the photo-Dember fields. This enhancement, however, is comparable or even larger than that observed by applying a magnetic field parallel to a planar semiconductor surface.[12–14] For a dipole oriented perpendicular to the surface, only $\leq 1\%$ of the radiated terahertz power from this dipole can escape the surface because of the small emission cone limited by the total reflection within a material of high refractive index.[15] A magnetic field can rotate the direction of moving charges with respect to semiconductor surface and results as much as two orders of magnitude of power enhancement. Therefore, if the terahertz dipole is formed in the favorable in-plane direction, the same order of power enhancement can be expected.

The stacking sequence of *c*-plane wurtzite InN is $ABABAB\cdots$ along the wurtzite *c*-axis direction so that the surface layers of *c*-plane InN have either an In- or a N-terminated polar surfaces. Therefore, the electric field generated by these In–N bilayers directs perpendicular to the surface and the resultant out-of-surface radiation can be significantly limited by the geometrical reason mentioned above. On the other hand, the layers of *a*-plane InN have the same number of In and N atoms in a plane (typically called as a "nonpolar" film) and these in-plane In–N dimers form in-plane intrinsic electric field perpendicular to the *a* axis. The highly photoexcited carriers can then be efficiently coupled to the in-plane electric field such that a more favorable part of emission can escape the emission cone. Despite of the same order of magnitude of power enhancement, the magnetic-field-assisted method requires a cumbersome experimental setup of external magnets. In comparison, the power enhancement from *a*-plane InN only depends on the growth direction so that it can be a universal phenomenon for semiconductors grown in the nonpolar direction.

In summary, it has been found that the *a*-plane InN film radiates about 100 times more intense terahertz pulses compared to that of the *c*-plane InN film. We found a distinctive azimuthal angle dependence of the terahertz field from the *a*-plane InN film and it may be due to the nonlinear optical processes. The effective geometrical coupling between the in-plane electric field and the out of plane terahertz radiation is proposed to be responsible for the drastic power enhancement and it can provide a promising solution to the search of effective terahertz emitters.

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