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Single-cycle azimuthal angle dependence of terahertz radiation from (100) n-type InP

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Single-cycle azimuthal angle dependence of terahertz radiation from (100) *n*-type InP

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We have observed that the terahertz power emitted by (100) *n*-type InP exhibits a single maximum and a single minimum as the crystal is rotated through 360° about its surface normal. This stands in contrast to other semiconductor terahertz emitters for which two, three, or four maxima per rotation have been observed. We have investigated the terahertz emission as a function of sample doping, optical excitation fluence, and applied in-plane magnetic field. The data cannot be accounted for by bulk optical rectification. We suggest that the origin of the phenomenon may be related to crystal twinning. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3046287]

Terahertz-frequency electromagnetic radiation is emitted by semiconductors such as GaAs, InAs, and InSb when they are illuminated by ultrashort optical pulses. As the semiconductor crystal is rotated about its surface normal, a systematic variation in the radiated terahertz power is sometimes observed. Generally speaking, in such experiments *two* maxima per rotation are associated with a (100) face, as exemplified by (100) *n*-InSb, and *three* maxima per rotation are associated with a (111) face, as exemplified by (111) *n*-InAs.[1] In contrast, very recently, *four* maxima per rotation have been observed in the case of (100) *p*-InAs (Ref. 2) and in the case of *a*-plane *n*-InN.[3] No report has been given of *one* maxima per rotation for any material. This is what we report here.

Turning now to the subject of our study, InP, it has been stated that (111) *n*-InP displays three maxima per rotation under excitation fluence of 1000 $\mu J/cm^2$.[4] Semi-insulating (SI) (100) InP has also been investigated[5] (see also Ref. 6). Under the specific condition of "high-density" (11 $\mu J/cm^2$) excitation, SI (100) InP gives two maxima per rotation, but under "low-density" (0.11 $\mu J/cm^2$) excitation, almost no azimuthal angle dependence is observed.[7] There appears to be no data in literature on the azimuthal angle dependence of terahertz radiation from either *n*-type or *p*-type (100) InP.

In our experiments sub-12-fs pulses of *p*-polarized near-infrared radiation were directed at an angle of incidence of 45° onto the (100) face of InP crystals and the radiated terahertz field in the specular reflection direction was measured using free-space electro-optic detection in a (110) ZnTe wafer. The laser center wavelength of 790 nm corresponds to a center frequency of 379 THz and center photon energy of 1.57 eV. The pulse contains photons of energy both above and below the InP bandgap ($E_g = 1.35$ eV at 300 K).

In Fig. 1 we present the time-domain spectrum obtained from (100) *n*-InP together, by way of comparison, with the spectrum from (100) *p*-InAs under identical excitation conditions. Of the unbiased terahertz emitters investigated to date, *p*-InAs appears to be the strongest.[8,9] It may be seen that the terahertz electric field from the InP sample is about one-tenth of that from the InAs sample. Consequently, the power spectrum is about two orders of magnitude less (Fig. 1, inset). Nonetheless, the signal-to-noise ratios of the two emitters are similar. Interestingly, the InP emitter shows a relatively higher amount of spectral power at higher frequencies, which may make it a preferred emitter to InAs at higher frequencies.

In Fig. 2 a survey of terahertz time-domain spectra from eight (100) InP samples of different dopings is given. It may be seen that highly doped ($>10^{18}$ cm^{-3}) samples, whether *n*-type or *p*-type, are poor terahertz emitters. This may be accounted for in that the charge-carrier plasma will reduce the transmission of the terahertz signal produced within the crystal. The strongest emitters are lightly doped, of $\sim 10^{16}$ cm^{-3}. The polarity of the terahertz field reverses as the charge-carrier type is switched, which is expected if transient currents (TCs) driven by surface fields arising from band bending account for the terahertz signal.

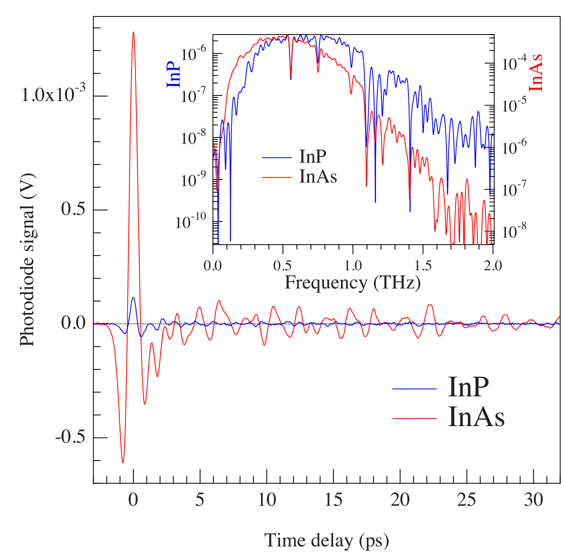

FIG. 1. (Color online) Comparison of terahertz emission by InP (blue curves) and InAs (red curves) unbiased samples under identical excitation conditions of angle of incidence 45° and excitation fluence of ~ 0.1 $\mu J/cm^2$. Main figure: time-domain spectra. Inset: frequency-domain spectra. The sharp lines in the frequency-domain spectra are due to atmospheric water vapor. The InP is nominally undoped but in fact is *n*-type with carrier concentration $\leq 10^{16}$ cm^{-3}. The InAs sample (Ref. 12) is *p*-type with concentration of $\sim 1.5 \times 10^{16}$ cm^{-3}. Both are (100) oriented.

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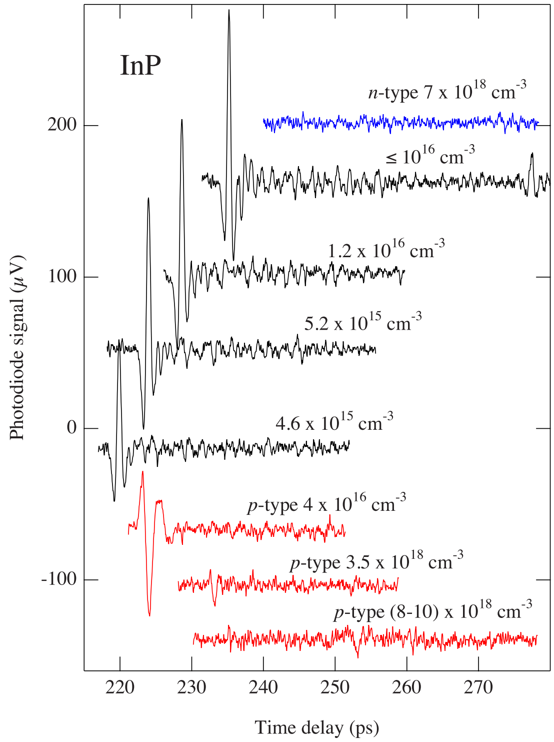

FIG. 2. (Color online) Survey of terahertz time-domain spectra of eight InP samples under identical excitation conditions of angle of incidence 45° and excitation fluence of $\sim 0.2\ \mu\text{J}/\text{cm}^2$. All samples are (100). Where the sample is deliberately doped, the carrier type is specified. (Red curves represent *p*-type samples; the blue curve is an *n*-type sample.) The nominally undoped samples (black curves) are all *n*-type. Successive spectra have been offset in the vertical and horizontal directions for ease of comparison but the vertical and horizontal scaling are the same for all samples.

To further explicate the mechanism responsible for the terahertz generation, a permanent magnet was placed behind the sample, with the magnetic field direction perpendicular to the sample normal. The magnetic field direction could be rotated about the sample surface normal independently of the crystal orientation. A single-cycle magnetic field angle dependence, as shown in Fig. 3, was obtained regardless of the crystal orientation. In addition, we measured the terahertz power as a function of excitation fluence (Fig. 3, inset).

Figure 4 gives our main result. For all of the (100) *n*-type InP samples that gave a measurable terahertz signal, we observed a single-cycle only azimuthal angle dependence. Using a wire-grid polarizer, we determined that the terahertz radiation was *p*-polarized. We have checked the azimuthal angle dependence also from the strongly emitting (100) *p*-type sample and from a SI (100) InP sample, and in those cases found no variation in terahertz intensity with angle.

We now discuss our results. In accordance with the time-bandwidth theorem, the short excitation pulses carry a range of Fourier components, including terahertz-frequency ones. The terahertz-frequency components may be extracted through interaction with the target. Established mechanisms include the nonlinear optical effect of optical rectification (OR) and TC effects. Within the TC effects may be distinguished those that rely on a current surge (CS) due to drift of charge carriers in the surface field and those that involve the photo-Dember (PD) effect,[10,11] due to the different diffusion rates of holes and electrons away from the surface. While the CS saturates at high excitation fluences, the PD effect does

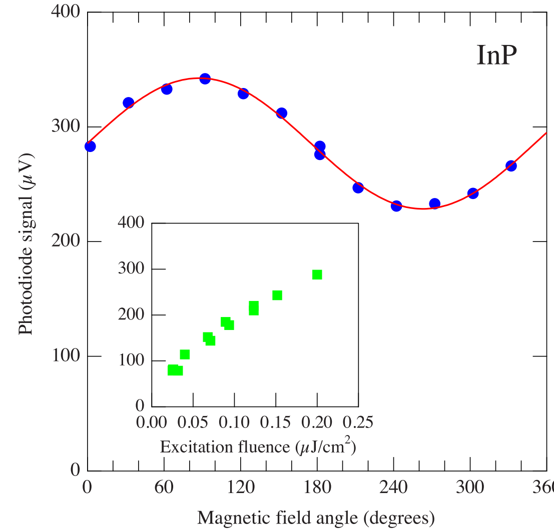

FIG. 3. (Color online) Terahertz emission from nominally undoped InP as a function of magnetic field (main figure) and of excitation fluence (inset). The magnetic field is provided by a permanent magnet behind the sample. The magnetic field direction is in the sample plane. The magnetic field angle is measured counterclockwise around the pump beam starting from the incidence-reflection plane. Excitation fluence is $\sim 0.2\ \mu\text{J}/\text{cm}^2$. Inset: terahertz emission in the absence of magnetic field as a function of excitation fluence.

not. CS effects dominate in wide-bandgap semiconductors, such as GaAs, where the surface field is large, whereas the PD effect is prominent in narrow-gap semiconductors, such as InAs, where the surface field is small.

The relatively low excitation fluence we employ suggests that TC, and not OR, will be the main origin of the terahertz emission. This suggestion is corroborated by the magnetic-field dependence (Fig. 3), which is not expected to be large for OR, as the magnetization contribution to the nonlinear optical susceptibility is expected to be small. Fur-

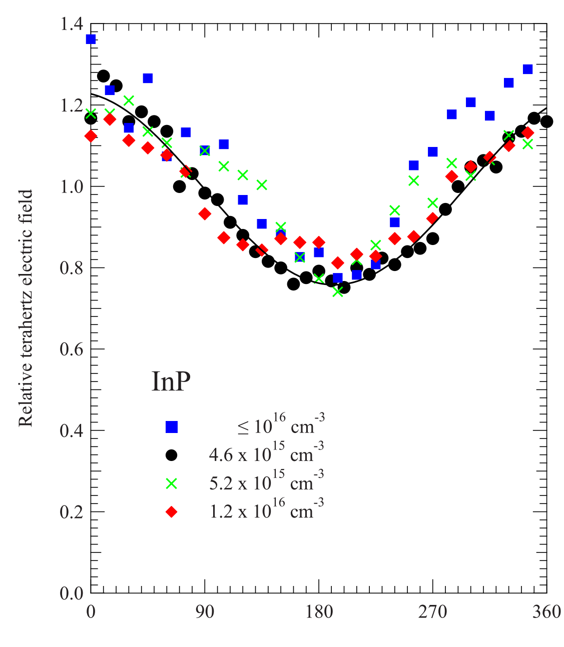

FIG. 4. (Color online) Azimuthal angle dependence of the terahertz field emitted by four (100) *n*-type InP samples. The full curve is a sinusoidal fit to the data for the sample with doping $4.6 \times 10^{15}\ \text{cm}^{-3}$.

ther, the nonlinear excitation fluence dependence (Fig. 3, inset) suggests that CS, which saturates with fluence, rather than PD, which does not, is the specific TC phenomenon contributing most of the terahertz field detected.

The occurrence of an azimuthal angle dependence is often interpreted as a signature of OR. We may rule out bulk OR as an explanation of our data on several grounds. First, the effect is expected to be small in (100) samples and much smaller than that from (111) samples, as has been illustrated in InAs.[9] Indeed, for normal incidence, there is in theory no effect for (100) samples, in contrast to the case of (111) faces, where an effect still occurs. The effect for (100) samples relies on the incident beam being at an angle to the surface normal. Quantitatively, we use the expressions of Gu *et al.*[1] to determine the magnitude of the terahertz field induced by OR in InP. Taking the static and optical dielectric constants to be 12.61 and 9.61, respectively, and so the terahertz and optical indices to be 3.55 and 3.10, respectively, yields the expected terahertz electric field dependence for the two orientations to be

$$E_{111} \propto [0.764 \cos(3\theta) - 0.157] \quad (1)$$

and

$$E_{100} \propto 0.161 \sin(2\theta). \quad (2)$$

Thus the (100) modulation is about five times smaller than the (111) modulation. Second, the modulation we observe is much larger (see Fig. 4; about $\pm 24\%$) than the effects observed previously [for example, $\pm 4\%$ in p-InAs (Ref. 12)]. Third, our effect depends on the doping, having been observed only for n-type and not for p-type or for SI InP. Finally, we observe 1 cycle, not the 2 cycle expected dependence from Eq. (2). No combination of refractive indices or incidence angles will yield other than a 2 (or 0) cycle modulation from the expressions of Gu *et al.*[1]

The origin of the effect appears to be associated with internal fields, which reduce the crystal symmetry and so influence the CS/TC. A form of

$$E_{100} \propto a \sin(\theta) + b \sin(4\theta) \quad (3)$$

has been proposed for n-InN,[3] but the physical origin of the $a \sin(\theta)$ term is not explained. While we cannot give a detailed microscopic account without further investigation, we suggest that the physical mechanism may be related to crystal twinning, which is known to be common in (100) InP.[13] Recently it has been observed that the far-field second-harmonic generation differs between twinned and twin-free ZnO rods, which is accounted for by a dipole wire model.[14] It is possible that an extension of this mechanism to difference-frequency generation may account for the present results.

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