# Enhanced terahertz emission from porous InP (111) membranes

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Bulk *n*-InP wafers and porous membrances with (111) crystallographic orientation have been illuminated with 120 fs pulses of 800 nm radiation from a Ti:Sapphire amplified laser system. Terahertz (THz) emission from samples was measured as a function of excitation fluence in the reflection geometry. It was established that the THz emission from both bulk and porous InP (111) saturates at high excitation fluence, emitting comparable levels of far-infrared radiation. Below saturation, however, the emission from the porous InP (111) membrane was found to be approximately an order of magnitude greater in radiated electric field or approximately two orders of magnitude in power relative to the bulk sample. The observed increase in efficiency from the porous, relative to the bulk samples, can be attributed either to the local field enhancement in the porous network for the nonlinear contribution to the radiated THz fields, or to modifications of the transient currents resulting in enhanced THz radiation. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1849813]

Over the last years, terahertz (THz) sources have been developed by using ultrafast laser sources to excite photoconductive switches,[1] semiconductor surfaces generating transient currents,[2] or to excite nonlinear processes in materials.[3,4] As application areas for THz technologies are rapidly expanding, the need for efficient THz sources is growing.

THz emission from semiconductor surfaces was investigated previously in connection with transient photocurrents radiating THz fields,[2] as well as in the context of nonlinear optics.[5–7] At high excitation fluence, there is a strong contribution to the radiated THz field from optical rectification in the surface region within the optical penetration depth of the pump beam.[6] It is therefore worth examining approaches for increasing the nonlinear interaction of the pump beam in semiconductor materials to enhance the conversion efficiency to the far infrared.

It has been recently reported that electrochemical etching of GaP can introduce a large birefringence into the semiconductor, allowing phase matching for optical second-harmonic generation.[8] In addition to affecting nonlinear optical processes, changes in the sample surface architecture affect transient currents generated in semiconductors that may modify the THz emission. In the present letter, we study the impact of electrochemically introduced porosity upon the characteristics of far-infrared emission from InP (111) samples.

(111)-oriented substrates of *n*-InP single crystals with 500 μm thickness and free electron concentration of $1.3 \times 10^{18}$ $cm^{-3}$ were used. The electrochemical etching was carried out in 5% HC1 aqueous solution in potentiostatic regime as described elsewhere,[9] the area of the samples exposed to the electrolyte was 1 $cm^{-2}$. A scanning elctron microscope (SEM) (Tescan SEM) equipped with an energy dispersive x-ray (EDX) (Oxford Instruments INCA EDX) system was used to analyze the morphology and chemical composition of the porous samples. Figure 1 shows a SEM image taken from a porous InP membrane in cross section. Note that the pores stretch perpendicular to the initial surface (±5° tolerance), leaving InP walls with an average thickness of about 50 nm (see inset in Fig. 1), and extend 65 μm into the sample, which is deeper than the optical penetration depth of the pump beam used in the experiments (~300 nm for InP at $\lambda = 800$ nm). The EDX microanalysis confirmed the stoichiometric composition of the InP skeleton.

The experimental setup for investigating the THz emission from bulk and porous InP (111) is shown in Fig. 2. A regeneratively amplified Ti:Sapphire laser system (Spectra Physics Hurricane) is used as a source, operating at a center wavelength of 800 nm, at 1 kHz repetition rate, with a maximum pulse energy of 750 μJ and a pulsewidth of 120 fs (Gaussian full width at half maximum). The probe pulse is

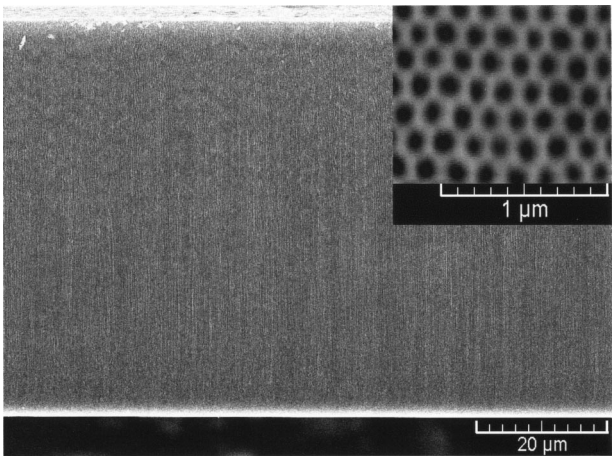


FIG. 1. SEM micrograph of the porous InP (111) membrane. The inset shows a top view of the pores.

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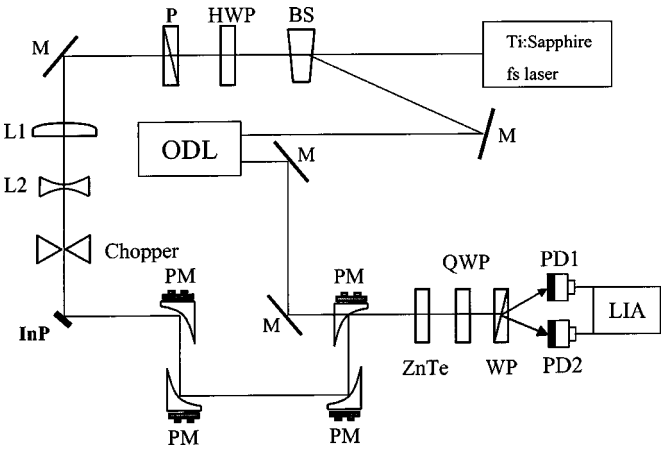


FIG. 2. Experimental setup for investigating THz emission from InP. The InP samples are oriented at an angle of incidence of 45°. BS is a beam splitter, HWP is a half-wave plate, P is a polarizer, M are mirrors, ODL is an optical delay line, L1 and L2 are lenses, PM are parabolic mirrors, QWP is a quarter-wave plate, WP is a Wollaston prism, PD are photodiodes, and LIA is a lock-in amplifier.

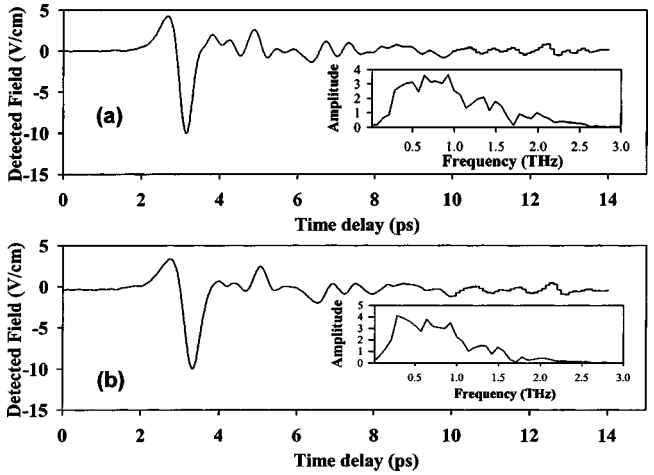


FIG. 3. Measured THz wave forms in the time domain from bulk (a) and porous (b) InP samples. Insets show the frequency spectrum, taken as the Fourier transform of the time domain signals.

delayed with respect to the pump using a scanning optical delay line. A variable attenuator ($\lambda/2$ plate and polarizer) is used in the pump beam to vary the fluence. The THz radiation from the surface of the sample oriented at 45° angle of incidence is collected in the specular direction and imaged onto the ZnTe detector using four $F/2$ parabolic mirrors. A 1 mm thick ZnTe (110) electro-optic crystal is used as detector, oriented for sensitivity to *p*-polarized THz emission.[10]

Generally, THz generation mechanism from semiconductor surfaces can be classified into two distinct categories: (i) Nonlinear-optical response of the material (optical rectification), and (ii) transient photocarrier related effects. Nonlinear contributions may come from bulk[6] or surface second-order nonlinearity of the semiconductor,[5] or through higher-order nonlinear effects.[7] Photocarrier related effects arise as a consequence of transient photocurrents, resulting from either acceleration of carriers in the surface depletion field[2] or from diffusion of carriers into the sample away from the surface.[11] At high excitation fluence, there is typically a large contribution from the nonlinear response of the material.[6]

A typical wave form and spectrum from bulk and porous InP (111) is presented in Fig. 3. As can be seen, the wave forms are similar in both frequency and time, with the bulk semiconductor exhibiting slightly higher field amplitudes at frequencies above 1.75 THz than the porous membrane. The THz signals reported here are not corrected for Fresnel reflection at the detector, and suffer attenuation in the opaque beam block used to remove the fundamental beam. The beam block has a flat frequency response over the bandwidth of the detected THz signals reported here.

In order to determine if there is a measurable difference between the radiated THz fields from the bulk and porous InP (111) samples, the fluence was varied using a variable attenuator. Figure 4 shows the peak detected THz field as a function of pump fluence for both bulk and porous InP samples. The measurements were made with the *p*-polarized pump beam parallel to the $[11\bar{2}]$ crystallographic axis in the (111) plane for both samples. It should be noted that the measured dependencies (not shown here) of the *p*-polarized THz radiation on azimuthal angle for a *p*-polarized excitation, at a fluence of 1 $mJ/cm^2$, revealed three-fold rotational symmetry for both bulk and porous InP, reflecting a contribution from optical rectification to the radiated field, proving the high crystalline quality of the porous sample. The peak THz field is seen to saturate for both bulk and porous InP. At lower excitation fluences, however, the peak radiated field from the porous sample proves to be substantially larger than that inherent to bulk InP. At the excitation fluence of 2 $\mu J/cm^2$, the peak radiated THz field from the porous membrane is approximately ten times as large as that from the bulk InP. The increased radiated field amounts to a relative power increase of approximately two orders of magnitude. Note that for an index of refraction of 3.46 at 800 nm, the transmission of the pump beam into the bulk InP is $T_p = 0.82$ (45 ° incident angle, *p*-polarized pump). The reduced Fresnel reflectivity of the porous InP due to the reduced effective refractive index could lead to enhancements of emission from porous InP on the order of 10% relative to bulk, far smaller than the differences observed here. It is difficult to determine the origin of the increase in conversion efficiency from optical to far-infrared between porous and bulk InP (111) without knowing the exact contributions from the various processes to the radiated THz field. If one assumes that the nonlinear response is the dominant THz emission mecha-

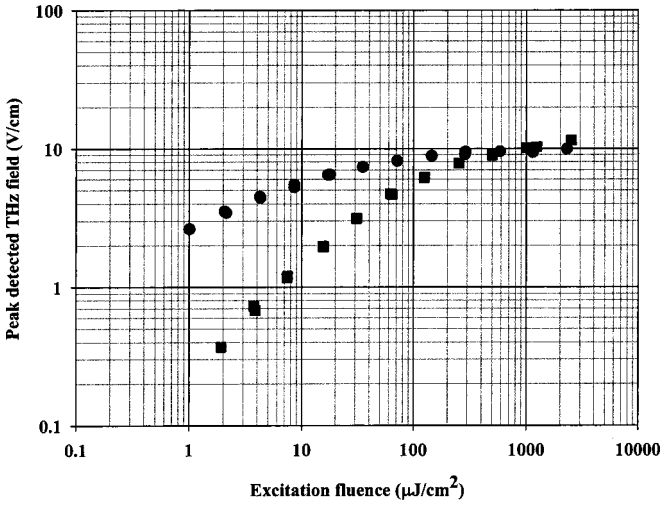


FIG. 4. Peak detected THz field as a function of fluence for porous (filled circles) and bulk (filled squares) InP samples.

nism for the range of excitation fluences considered in this letter, then the increase in conversion efficiency could result from the local field enhancement in the porous network of the membrane, as was proposed for the increase in second-harmonic generation from nanostructured GaP reported in Ref. 8.

In contrast to this, if the emission were predominantly a result of photocarrier-related effects, then the observed increase of the THz radiation from porous membrane relative to the bulk can be explained by the fact that, due to the high density of pores, most of the volume of the porous InP skeleton is subjected to the surface depletion field which may contribute to the acceleration of carriers and, as a consequence, to the intensification of transient photocurrent. However, for the depletion field within the walls, the lateral photocurrents from both sides of the side wall would be expected to be equal and opposite under uniform illumination, and therefore, cancel. However, asymmetry may arise as a consequence of the angled illumination of the pores from a single direction. The resultant net lateral photocurrent in the surface plane of the substrate could radiate more efficiently than the transient current generated normal to the surface for the bulk sample. In addition, the portion of the transient current resulting from the diffusion of photogenerated carriers will predominantly flow down the pore walls. In this case, because the side walls are open to free space, the dipole would couple efficiently out through the membrane walls. Similar arguments for enhanced emission by redirection of photocarrier generated currents along the surface plane have been used to explain enhanced emission when an external magnetic field is applied to the system.[12,13]

At very low excitation fluences (1–100 $nJ/cm^2$),[14] the emission from InP is due to transient photocurrents, either diffusion of carriers at low temperatures or acceleration of carriers in the surface depletion field at room temperature.[14] At higher fluences ($\gtrsim 100\ \mu J/cm^2$), optical rectification contributes significantly to the emission.[5–7] For fluences of 1–1000 $\mu J/cm^2$ as studied here, it is expected that there is a transition in the dominant emission mechanism. The clarification of the detailed balance of emission mechanisms in this fluence range will require further study.

In conclusion, THz emission from bulk and porous InP (111) was examined as a function of pump fluence in a reflection geometry. It was shown that at fluences of $\sim 1\ mJ/cm^2$, there is a three-fold rotational symmetry in the radiated THz field for both samples. At the excitation fluence involved, the peak radiated THz fields prove to be approximately the same for both bulk and porous InP. As the excitation fluence is decreased to as low as $\sim 2\ \mu J/cm^2$, the conversion efficiency from the porous InP (111) exceeds that of bulk InP by almost two orders of magnitude. The observed increase in THz power from porous relative to bulk InP is possibly related to the impact of local field enhancement in the porous network, as well as to the modification of the flow of the transient photocurrent and orientation of the resultant dipole emission.

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