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Krunal Radhanpura
University of Wollongong, krunal@uow.edu.au

Stuart Hargreaves
University of Wollongong, sh64@uow.edu.au

Roger A. Lewis
University of Wollongong, roger@uow.edu.au

L Sirbu
Academy of Sciences, Moldova

Ion M. Tiginyanu
Technical University of Moldova

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Heavy noble gas (Kr, Xe) irradiated (111) InP nanoporous honeycomb membranes with enhanced ultrafast all-optical terahertz emission

K. Radhanpura,¹ S. Hargreaves,¹ R. A. Lewis,^{1,a)} L. Sirbu,² and I. M. Tiginyanu³

¹*Institute for Superconducting and Electronic Materials, University of Wollongong, Wollongong, New South Wales 2522, Australia*

²*Institute of Electronic Engineering and Nanotechnologies, Academy of Sciences of Moldova, Chisinau MD-2028, Moldova*

³*National Center for Materials Study and Testing, Technical University of Moldova, Chisinau MD-2004, Moldova*

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Nanoporous honeycomb membranes on InP (111) surfaces emit ultrafast coherent terahertz pulses under near-infrared optical excitation. Irradiating the membranes with heavy noble gas Kr or Xe ions enhances the terahertz emission. The emission does not vary with in-plane magnetic field rotation and exhibits three-cycle dependence on azimuthal-angle rotation. Both suggest the terahertz source is not transient currents but optical rectification enhanced by the heavy-ion irradiation.

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Materials with nonlinear optical (NLO) properties under intense optical excitation exhibit difference-frequency mixing. This may result in emission of electromagnetic radiation of frequencies in the “terahertz gap” lying between electronics and photonics.¹ Semiconductors may be used thus as terahertz emitters. We focus here on InP. Terahertz emission from bulk InP has been found to depend on whether it is *n*- or *p*-type, with lightly doped ($<10^{17}$ cm⁻³) crystals being the best emitters.² A single-cycle in-plane magnetic field dependence on the terahertz emission as well as a saturation of the emission at relatively low excitation fluences (<0.20 μJ/cm²) indicates the primary mechanism is due to a transient current (TC) induced by the photoexcited carriers.² (Conversely, an absence of magnetic field dependence and nonsaturation with excitation fluence, as exemplified by GaBiAs,³ rule out the influence of TC.) Porous membranes on (100) InP surfaces give an enhanced NLO response for both second-harmonic and terahertz generation, related to optical rectification (OR) rather than to TC effects, and attributed to strong local fields in the porous network.⁴ Similar enhancements to the terahertz emission in porous samples relative to the bulk precursors have been observed for (111) InP.⁵ Measurements and modeling on charge carriers in porous InP membranes indicate a lower density and lower mobility but much longer recombination lifetime than for bulk InP, attributed to upward bandbending at the pore surfaces.⁶ Here we extend the modification of the base material by the creation of radiation defects through Kr and Xe noble-gas ion irradiation of InP nanoporous membranes. Swift heavy-ion irradiation studies in GaN and ZnO have shown that nanostructuring the surface improves radiation hardness.^{7,8} While enhanced terahertz emission has been noted for As implantation into GaAs,⁹ no studies are known of the effect on the terahertz emission of irradiating InP with any ion, or of any ion being used to irradiate a nanoporous membrane.

(111)-oriented substrates of 500-μm thick *n*-InP single crystals of free electron concentration 5×10^{18} cm⁻³ (Crys-

Tec GmbH, Germany) were anodically etched in the dark at room temperature in a 5% HCl aqueous solution in the potentiostatic regime using an electrochemical double cell.¹⁰ The sample, of exposed area 1 cm², was mounted between the cells. The electrolyte was continuously pumped through both cells. Four Pt electrodes were used: a reference electrode in the electrolyte, a sense electrode on the sample, a counter electrode, and a working electrode. After pore growth a shock voltage was applied for a few seconds to detach the nanoporous layer from the substrate. The top nucleation layer was removed by isotropic wet etching.

Both bulk and nanoporous samples were irradiated at room temperature by 85 MeV Kr⁺¹⁵ ions at doses of 10^{12} and 10^{13} cm⁻², or by 130 MeV Xe⁺²³ ions at doses of 5×10^{11} and 5×10^{12} cm⁻², at the IC-100 cyclotron of the Joint Institute for Nuclear Research, Dubna, Russia.

A VEGA TESCAN TS 5130MM scanning electron microscope with an Oxford Instruments energy-dispersive x-ray (EDX) system was used to analyze the morphology and chemical composition of the samples. The EDX microanalysis confirmed the stoichiometric composition of the InP nanoporous skeleton both before and after ion irradiation. Figure 1 is micrograph of a typical nanoporous structure. The

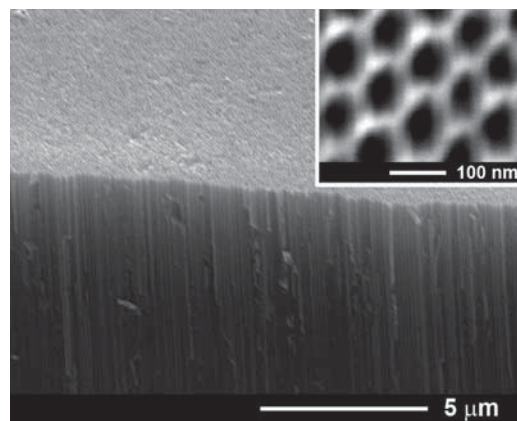


FIG. 1. Scanning electron micrograph of a nanoporous InP (111) membrane. The inset gives a top view of the hexagonal arrangement of the pores.

^{a)}Electronic mail: roger@uow.edu.au.

TABLE I. Relative terahertz (THz) electric field amplitude from nine different InP (111) samples.

	Bulk, unirradiated sample: THz emission ~ 0			
	Kr irradiated		Xe irradiated	
	10^{12} cm^{-2}	10^{13} cm^{-2}	5×10^{11} cm^{-2}	5×10^{12} cm^{-2}
Bulk	~ 0	1	~ 0	1
Nanoporous	4	14	5	12

hexagonal pores have a mean diameter of 60 nm and an interpore spacing of ~ 100 nm. The total thickness of the nanoporous membrane is ~ 30 μm .

Terahertz time-domain spectroscopy was carried out using the membranes as terahertz emitters. This produced subpicosecond, broadband pulses of terahertz radiation. No electrodes or electrical biasing of the emitters was employed. All measurements were at room pressure and temperature. Horizontally polarized ultrashort (< 12 fs) pulses of near-infrared (NIR; center-frequency 790 nm) radiation were pro-

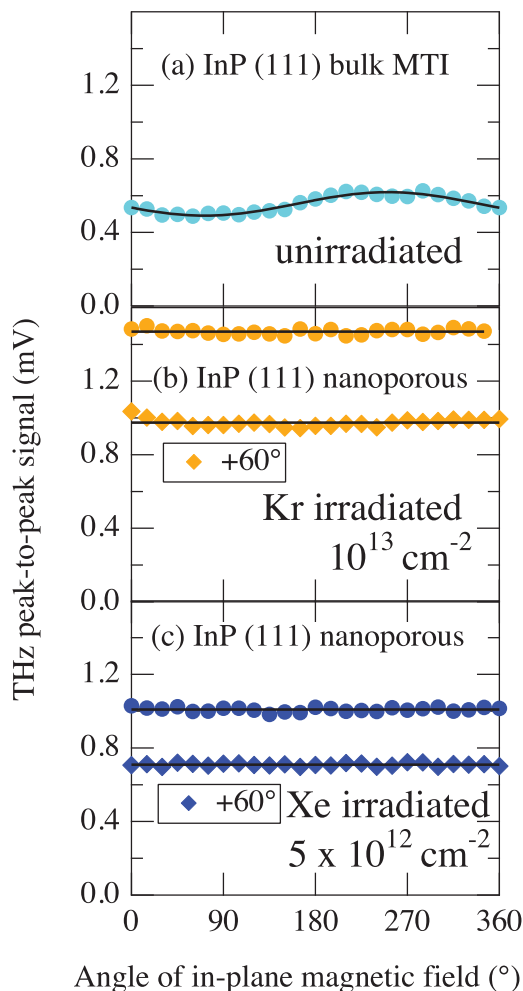


FIG. 2. (Color online) Effect of rotating a magnetic field in the plane of the sample on the peak-to-peak terahertz field emitted. (a) A bulk InP sample without ion irradiation. (b) A nanoporous InP sample with Kr ion irradiation at a level of 10^{13} cm^{-2} (circles) and the same sample, now rotated through an azimuthal angle of 60° (diamonds). (c) A nanoporous InP sample with Xe ion irradiation at a level of 5×10^{12} cm^{-2} (circles) and the same sample, now rotated through an azimuthal angle of 60° (diamonds). The data illustrate that transient current effects are present in the bulk material but not in the irradiated nanoporous material.

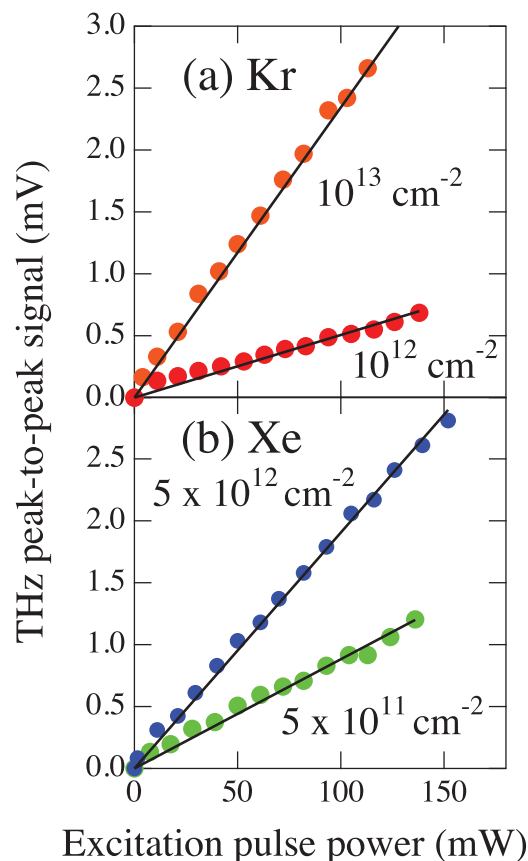


FIG. 3. (Color online) Effect of varying the excitation pump fluence on the terahertz emission from (111) nanoporous InP ion-irradiated in four different ways. (a) Kr-irradiated samples. (b) Xe-irradiated samples. The terahertz emission in all cases is directly proportional to the pump power. There is no evidence for saturation in the relationship (as is often observed in the case of charge-carrier screening in transient-current emitters).

vided at an 80 MHz repetition rate by a mode-locked Ti:sapphire laser (Femtolasers, Vienna, Austria). The power on the emitter (maximum 160 mW) was adjusted by a continuously variable attenuator, allowing control of the excitation optical fluence. A small (0.15 T) permanent magnet was rotated behind the emitter so that the in-plane magnetic field advanced through 360° . The emitter was rotated about the surface normal, or through the azimuthal angle, θ , by 360° . The horizontally (p) and vertically (s) polarized terahertz components were selected by a wire-grid polarizer. Reflection geometry was used with 45° angle of incidence of the NIR beam and 45° angle of terahertz detection. The terahertz radiation was collected, focused, collimated, and refocused onto a ZnTe detector crystal by off-axis paraboloidal mirrors. Time-domain spectroscopy was effected by conventional electro-optic sampling with a NIR probe beam, split off from the NIR pump beam, being coincident with the terahertz beam on the ZnTe crystal. The output of a balanced photodiode pair is recorded as a function of the time delay between the coherent terahertz pulse and the probe.

A survey of nine bulk and nanoporous samples as terahertz emitters is given in Table I. The unirradiated sample gave negligible terahertz radiation. Increasing the ion dose increases the intensity of terahertz emission. This holds for both ion species (Kr, Xe) for both bulk and nanoporous samples. For both bulk and nanoporous specimens, Xe ion irradiation is more effective than Kr ion irradiation in en-

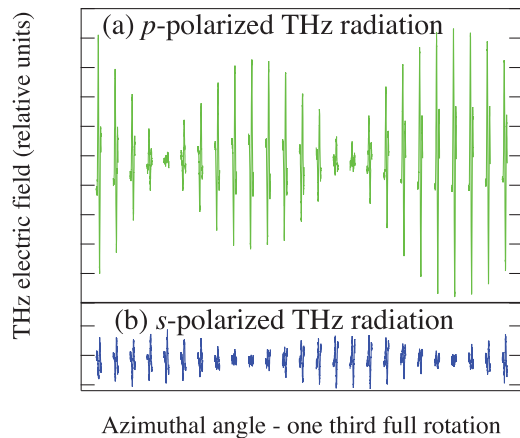


FIG. 4. (Color online) Azimuthal angle dependence of the terahertz emission from a Xe-irradiated ($5 \times 10^{12} \text{ cm}^{-2}$) nanoporous InP (111) membrane. The central 3 ps of the terahertz time-domain spectrum is plotted for each separate azimuthal angle, spaced at 5° intervals. (a) Magnitude of the horizontally (p) polarized terahertz field, over one-third of a full rotation, or 120° . (b) Magnitude of the vertically (s) polarized terahertz field, over one-third of a full rotation, or 120° . The same relative units for terahertz field are used in each panel.

hancing terahertz emission. The nanoporous structures yield more terahertz radiation than the original surfaces. Overall, the best emitters are the nanoporous samples with the highest levels of Kr and Xe ion irradiation.

Figure 2 shows the effect of an in-plane magnetic field on the terahertz emission. Only samples identified as having a strong emission (see Table I) were investigated in this way. None of these InP samples show a strong angular dependence on the magnetic field direction, in contrast to work on InP from other sources² (see also the data from a commercial sample in Fig. 2), where a single-cycle dependence on the magnetic field is observed. A single-cycle dependence on magnetic field is interpreted as being due to the Lorentz force on moving charge carriers adding to or subtracting from the radiating dipole as the magnetic field sweeps through 360° . That in the present data no variation in terahertz signal is evident on varying the direction of the in-plane magnetic field implies that no role is played by TC mechanisms in the generation of terahertz radiation in the samples under investigation here.

Figure 3 gives the effect of the excitation pump fluence on the terahertz emission. Only those samples identified as having a strong terahertz emission (see Table I) were investigated in this way. In all the cases in Fig. 3 the detected terahertz power varies linearly with the pump fluence. There is no evidence of saturation, which would serve as a signa-

ture of the TC mechanism.² It may be that the fluence is too small for TC effects to be observed but the present data give no support to a TC mechanism contributing to the terahertz emission detected.

Figure 4 shows the azimuthal angle dependence of terahertz emission from a (111) ion-irradiated, nanoporous sample. Over a complete (360°) rotation we observe a 3 θ dependence, as expected for a (111) face.¹¹ In Fig. 4 we only show the behavior over the one-third (120°) repeat unit. In Figs. 4(a) and 4(b) the horizontally (p) and vertically (s) polarized components are shown, respectively. The azimuthal angle dependence shows most of the radiation is originating in OR, consistently with a recent general model of OR.^{11,12}

To conclude, we have demonstrated that irradiating nanoporous membranes of (111) InP with heavy noble-gas ions enhances terahertz emission. Systematic investigation of the dependence of the generated terahertz electric field on excitation pump power, in-plane magnetic field, and azimuthal angle indicate that the underlying physical mechanism is OR rather than TC flow. These results may lead to further improvements in terahertz emitters.

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