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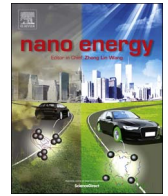
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Full paper

# High-energy terahertz surface optical rectification

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## ABSTRACT

The interest in surface terahertz emitters lies in their extremely thin active region, typically hundreds of atomic layers, and the agile surface scalability. The ultimate limit in the achievable emission is determined by the saturation of the several different mechanisms concurring to the THz frequency conversion. Although there is a very prolific debate about the contribution of each process, surface optical rectification has been highlighted as the dominant process at high excitation, but the effective limits in the conversion are largely unknown.

The current state of the art suggests that in field-induced optical rectification a maximum limit of the emission may exist and it is ruled by the photocarrier induced neutralisation of the medium's surface field. This would represent the most important impediment to the application of surface optical rectification in high-energy THz emitters.

We experimentally unveil novel physical insights in the THz conversion at high excitation energies mediated by the ultrafast surface optical rectification process. The main finding is that the expected total saturation of the Terahertz emission vs pump energy does not actually occur. At high energy, the surface field region contracts towards the surface. We argue that this mechanism weakens the main saturation process, re-establishing a clearly observable quadratic dependence between the emitted THz energy and the excitation. This is relevant in enabling access to intense generation at high fluences.

## 1. Introduction

The improvement of the generation efficiency of Terahertz pulses (THz) from intense ultrafast optical sources is presently a very hot topic, with a wide spectrum of solutions ranging from the development of novel materials [1–3] with a higher nonlinear response to efficient non-trivial field-matter interaction geometries [4–7]. Few options are left when ultrathin emitters are desired. Common quadratic nonlinear crystals have a low absolute conversion efficiency for effective thicknesses within the fraction of micrometres or less. In such scales the nonlinear field-matter interaction is only marginally governed by the bulk properties of the medium. Conversely, it is fundamentally ruled by a series of processes that belong to the domain of surface physics, responsible for THz conversion efficiencies per unit of thickness orders of magnitude larger. Although the literature on surface THz generation is quite vast, the generation at very high excitation and its physical limitations are quite untackled.

A significant fraction of the pioneering works on THz surface emission focuses on narrow band-gap III-V semiconductors, such as InAs and InSb, which exhibit surface terahertz (THz) emission when

excited with ultrashort optical pulses. In most scenarios, the generation is driven by the kinetic carrier dynamics [8–10] and surface field induced Optical Rectification (OR) [11–13], upon excitation with photon energies well above the energy bandgap [14].

The interest is certainly driven by the surprisingly high conversion efficiencies. For materials possessing a direct band structure, the optical absorption depth for photons with energy exceeding the bandgap is typically very low (e.g. approximately  $140\text{nm}$  at  $\lambda = 800\text{nm}$  in InAs [15]). A significant and macroscopic THz emission is then obtained in a very thin interaction region, with very high energy conversion per unit thickness [16,17].

At low impinging fluences (below  $100\text{nJ}/\text{cm}^2$ ), InAs is perhaps one of the most important benchmarks for surface THz emission driven by the photo-Dember effect [18,19]. However it has been argued that THz generation by carrier migration is quite saturated at high excitation energies such that the generation is dominated by surface OR [10]. This interpretation reconciles the observed dependence of the emission from the polarisation and crystal orientation in InAs and seeded through works about the microscopic mechanisms responsible for the non-linearity [20–22].

Abbreviations: THz, Terahertz; OR, Optical Rectification; InAs, Indium Arsenide; InSb, Indium Antimonide; OPTH, Optical Pump Terahertz Probe; OPRE, Optical Pump Rectification Emission; SP, Screening Pump; TP, Terahertz Pump

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A saturation fluence for surface OR has been identified within the order of tens of  $\mu\text{J}/\text{cm}^2$  by Reid et al. [23]. The specific mechanism has not been investigated. Yet, its understanding is clearly fundamental in boosting the THz conversion efficiency at semiconductor surfaces. The basic model of the surface Optical Rectification process suggests that the main saturation mechanism in OR is driven by the accumulation of photoinduced carriers that screens the static surface field of the semiconductor responsible for the surface nonlinearity [11].

In this paper we explore the emission limits, with a novel methodology. Our experimental approach involves the THz generation with an ultrafast generating pump and the simultaneous injection of photocarriers in the surface field region (within  $100\text{nm}$ – $200\text{nm}$  of the surface) with a second optical screening pump. The underlying idea is to connect the change of the emitted field to the modulation of the surface field as it is neutralised by the photo-carrier drift. The photo-carriers frustrate the surface potential, changing its contribution to the third-order nonlinearity in surface OR, in a process that we can define as Optical Pump Rectification Emission (OPRE) [24] (by analogy with the popular Optical Pump Terahertz Probe (OPTP)). Krotkus et al. [25] argued that at moderate exciting peak powers the surface static field could be significantly augmented by the carrier diffusion but that this does not affect the OR frustration mechanism. With specific reference to our experimental campaign, the signature of this specific contribution is not present at high energy excitations (we present our complete characterisation set in the [Supplementary materials](#)). It is worth noting that the THz emission through surface OR is a third-order nonlinear phenomenon and is driven by the cubic relation [11]

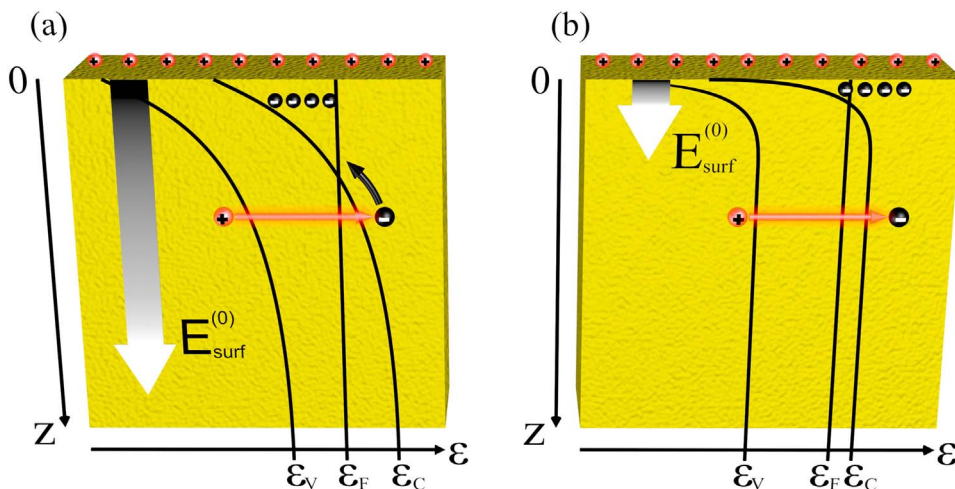
$$E_{\text{THz}} \propto \chi^{(3)}(\omega_{\text{THz}}; -\omega; \omega, 0); \mathbf{E}_G(-\omega); \mathbf{E}_G(\omega); \mathbf{E}_{\text{surf}}^{(0)} \hat{\mathbf{z}}. \quad (1)$$

As such it is not related to the bulk OR which is driven by the second-order medium nonlinear response ( $\chi^{(2)}$ ) and is not relevant in the physics at semiconductor surfaces.

In this investigation, we argue that because of the very small penetration depth of the optical NIR excitation, the change of the THz generation is also significantly connected to the weakening and contraction of the surface field region upon carrier screening. Hence, at high pumping energies most of the photocarriers are generated outside of the surface field region and therefore they no longer contribute to the saturation. At this point the system converges again to a quadratic conversion efficiency because the progress of the screening of the surface field is no-longer significant. In different words, a real flattening of the pump vs emission trend is not observed.

## 2. The field-matter interaction mechanism

The field-matter interaction at the semiconductor band bending



**Fig. 1.** A representation of the bending of the boundary of the valence ( $\epsilon_V$ ) and conduction ( $\epsilon_C$ ) bands at the InAs surface for (a) Undoped InAs and (b) n-Type InAs, where the positive surface states pin the Fermi level ( $\epsilon_F$ ) above the conduction band minimum at the surface.  $E_{\text{surf}}^{(0)}$  is depicted by the graduated arrow where darker shades represent a higher field magnitude. In the latter case, photo-excited electron-hole pairs are mostly generated outside the surface field region, weakening their role in surface field screening.

region is sketched in Fig. 1. In Indium Arsenide, our benchmark model, the surface states result in a positive charged surface density regardless of the doping type [26]. This pins the position of the Fermi-level above the conduction band minimum at the surface such that an electron accumulation layer is formed immediately below the surface. When photoexcited electron-hole pairs are generated, the electrons dominate the surface potential dynamics in light of their much higher mobility.

To infer the contraction, we start observing that in InAs the surface field region is known to weaken and contract as the concentration of free electrons increases [27–29]. Interestingly, because of the large difference in mobility, moderately doped p-type InAs exhibits lower conduction and a thicker surface field region than intrinsic and n-doped InAs. At significant n-doping levels, at room temperature, the surface field region thickness (Fig. 1(b)) can be much smaller (tens of nm) than the optical penetration depth.

We show here that the typical generation vs excitation trend observed at any n-doping concentration can be reproduced by photo-exciting a p-type semiconductor, corroborating the hypothesis that even in this latter case the field region weakens and thus as the concentration of photocarriers increases. Hence, we argue that upon intense optical pumping the fraction of photocarriers neutralising the surface-field decays and that this is a relevant mechanism in re-establishing a quadratic dependence between emission and excitation.

## 3. The experimental setup

The experimental setup is described in Peters et al. [24]. It is supplied with an ultrashort  $100\text{fs}$  optical pulse train with  $1\text{kHz}$  repetition rate and centre wavelength  $\lambda = 800\text{nm}$ , generated by a mJ-class Ti:Sa regenerative amplifier (Coherent Libra-He). The beam diameter (intensity at  $1/e^2$ ) is  $d = 9\text{mm}$ .

A Time-Domain Spectroscopy detection is implemented using a probe beam originating from a 3%-beam sampler. A motorised delay line controls the relative delay between the probe and the THz waves which are overlapped in a thick nonlinear ZnTe  $<110>$  crystal which functions as a THz electro-optical sampler [30]. The THz-emitting semiconductors are illuminated at an impinging angle of  $45^\circ$ .

For the implementation of the OPRE measurements a beamsplitter was placed to obtain two separate pump lines. A screening pump (SP) was directed towards a  $<100>$  InAs sample with an impinging angle of  $11.9^\circ$  relative to the THz-generating pump (TP). The SP is delayed by  $\tau_s$  using a second independent motorised translation stage.  $\tau_s = 0$  represents the synchronisation of the two optical pulses.

We verified that the screening does not induce an appreciable change in the emitted THz pulse field profile (which is expected for a DC surface field modulation). In addition, the actual THz-generating

pump beam size is reduced with an iris in order to mitigate the temporal smearing (down to the ps scale) in the synchronisation with the screening pump because of their slightly different impinging angle. Nevertheless, by changing the iris size, we verified that our observations are marginally affected by the beam profile.

Central in this investigation, in order to isolate the contribution of the OR from any parasitic contribution of carrier-mediated generation mechanisms, we rotated the detection crystal to detect the s-polarised (perpendicular to the impinging plane) Terahertz emission and we maximised it by changing the generating pump polarisation. We argue that for  $< 100 >$  InAs under the given illumination condition the potential out-of-plane (s) carrier dynamics induced by structural anisotropies for both the field-driven surge-current and photo-Dember mechanisms should be negligible. We also verified that no Terahertz contamination from the screening pump (p-polarised) is perceived in the detection. We stress here that a typical advantage in using large collimated pumps, made possible thanks to the availability of millijoule pulses, is the absence of cross-talk with the screening pump (for comparison, examples of OPRE with focused generation are presented in the [Supplementary materials](#)). Finally, no cross-frequency mixing between screening and generating optical fields can be present for  $\tau_s$  larger than a few ps, as both pulses and the nonlinear response of the medium are shorter.

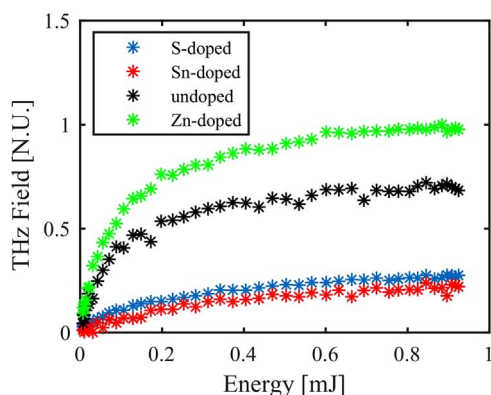
For pump photons with energy much larger than the energy bandgap, we can assume that the density of photocarriers is proportional to the impinging TP pulse energy  $W_G$  at any  $z$ .

$$n_{ph}(z) \propto W_G \quad (2)$$

The surface accumulation layer is essentially a 2-dimensional distribution of electrons centred up to tens of nanometres below the surface because of the quantisation of motion of the electrons in the  $z$ -direction [31]. We assume that a fraction of the photo-electrons drift to screen the surface field  $E_{surf}^{(0)}$  and approximated that as equivalent to an effective 2-D layer of density  $n_{ph}$ .

$$E_{surf}(z=0) = E_{surf}^{(0)} - n_{ph} \frac{e}{\epsilon} \quad (3)$$

However, as the field weakens at high excitations,  $n_{ph}$  becomes a progressively smaller fraction of the total population of photoexcited carriers, mitigating any further increment of  $n_{ph}$ , i.e. weakening the main saturation mechanism. This is a potential source of the trend observed in [Fig. 2](#), where a linear dependence between THz peak field and pump energy (i.e. a quadratic conversion) is re-established after a ‘knee’. Quite interestingly, in the case of highly n-doped materials (blue and red plots), in which the surface field region is known to be very thin, this *high-energy* linear dependence between peak field and energy



**Fig. 2.** The peak THz field has been plotted for various excitation energies, for a number of different freestanding  $< 100 >$  InAs substrates, two n-type samples doped with Sn and S donors, one undoped sample and one p-type sample doped with Zn acceptors. The peak THz field has been normalised across all substrates with respect to the maximum field achieved with the Zn-doped InAs.

(i.e. a quadratic conversion) almost covers the entire generation pump energy range.

In Reid et al. [23], the saturation curve in the OR process is fitted with a typical saturation equation

$$E_{THz} = \frac{AW_G}{W_G + W_{sat}} \quad (4)$$

Where  $A$  represents the maximum THz peak field,  $E_{THz}$  is the THz field and  $W_{sat}$  is the saturation energy. The best fit values for  $W_{sat}$  for each of the curves are displayed in [Table 1](#). In light of the sufficient accuracy of our experimental measurement, we can observe in [Fig. 2](#), that at high energy  $E_{THz}$  does not exhibit the plateau predicted by Eq. (4). This results in a very poor fit quality at high excitation energy [23]. The observed linear trend corroborates the hypothesis that the main saturation mechanism weakens at high energy.

It is worth noting that because of the much higher mobility of electrons, the undoped substrate (that possesses a process-induced n-doping around  $2 \times 10^{16} \text{cm}^{-3}$ ) exhibits a weaker surface field and a lower conversion efficiency when compared to the p-doped sample.

In [Fig. 3](#), the OPRE measurement in the different samples is presented: it is clear that the free electron concentration has an important impact on these dynamics. Following a similar approach in literature we fit the normalised recovery trend as [32]

$$\bar{E}_{THz} = 1 - k_1 \times e^{-\frac{t}{\tau_1}} - k_2 \times e^{-\frac{t}{\tau_2}} \quad (5)$$

where  $k_1$ ,  $k_2$ ,  $\tau_1$ ,  $\tau_2$  are the fitting parameters and  $\tau_s$  is the time delay between the screening pump and the THz emission pump.  $\tau_1$  represents a fast recovery time. It is potentially consistent with the time scale of the migration of photocarriers away from the surface-field region as its thickness can be a very small fraction of the pump wavelength. Although we should expect the carrier diffusion to be a dominant process in the carrier dynamics (as recognized in a number of studies as the source of the photo-Dember mechanism) we cannot exclude a contribution from quasi-ballistic photo-electron dynamics [33] and from the electron scattering into other conduction band valleys.  $\tau_2$  is consistent with the slow recovery time typically associated with the bulk recombination of photoexcited carriers.

For the n-doped sample we consider  $k_2 = 0$  as there is no appreciable slow decay in the traces: this appears consistent with the assumption that only the decay of the photocarrier density in a very thin region close to the surface dominates the dynamics. As soon as the concentration of carriers becomes comparable to the inherent free carrier concentration (from doping) the THz emission returns to its original level. Therefore, in n-doped substrates the bulk carrier recombination time does not appear to be relevant in the OPRE measurements. The best-fitting parameters can be appreciated in [Table 1](#).

We compared the doping of the substrates along with the calculated values for the saturation power ( $W_{sat}$ ) and surface field recovery times in [Table 1](#). For the sake of comparison,  $W_{sat}$  was extracted from the data fitted using Eq. (4). From this we observe that the apparent saturation of n-type samples occurs at significantly larger powers when we best fit the curve.

To test our hypotheses, we reproduced the conversion trend of [Fig. 2](#) for the undoped and n-doped media by artificially pumping photocarriers within the surface field regions in the p-type InAs, using the SP with appropriate energy. We fixed the delay of the SP to obtain the maximum screening. The expected effect is an increase in the electron-mediated screening of the surface field. The acquired data are presented in [Fig. 4](#).

We could adjust the average screening pulse energy  $W_s$  to reproduce fairly accurately all the observed conversion trends in the undoped and n-doped cases of [Fig. 2](#). Hence, we argue that the photo-carriers produce the same contraction of the surface field region as occurs in n-doped materials.

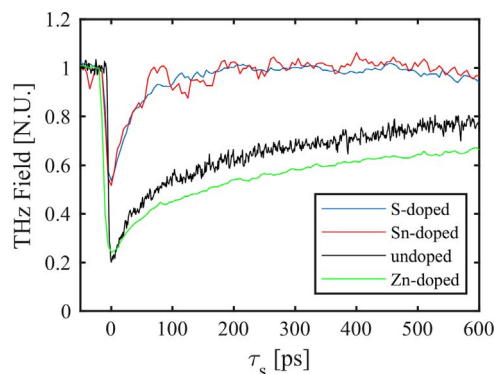
In addition, it is worth observing that the photo-excited carrier



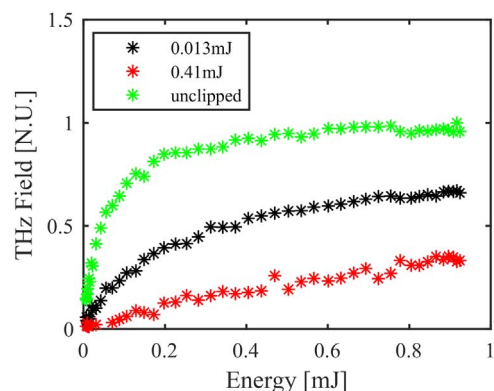
**Table 1**

Shows the known carrier concentration, saturation power and THz recovery times for a number of different substrates. The  $R^2$  (statistical coefficient of determination) values show the goodness of the fit used to retrieve recovery times. Considering the very different accuracies in the doping specified by the different sample suppliers, we also report the measured dark conductivity, relative to the undoped sample.

Substrate	Doping [ $cm^{-3}$ ]	$W_{sat}$ [ $\mu J$ ]	$\tau_1$ [ps]	$\tau_2$ [ns]	$R^2$ [N.U.]	Relative Dark Conductivity [N.U.]
n-InAs S-doped < 100 >	$n = 1 \times 10^{18}$	201	40	N/A	0.817	$2.83 \pm 0.01$
n-InAs Sn-doped < 100 >	$n = 3 - 10 \times 10^{17}$	371	31	N/A	0.846	$3.40 \pm 0.01$
n-InAs undoped < 100 >	$n < 3 \times 10^{16}$	88	54	> 1.0	0.947	$1.00 \pm 0.01$
p-InAs Zn-doped < 100 >	$p = 5.3 \times 10^{18}$	86	37	> 1.0	0.982	$0.28 \pm 0.01$



**Fig. 3.** OPRE measurements of the peak THz field vs the screening pump delay for the four different freestanding < 100 > InAs substrates of Table 1. Each curve is normalised with respect to the unscreened peak THz field. Each measurement was performed with a generating pump energy  $W_G$  of  $0.1mJ$  and a screening pump energy  $W_S$  of  $0.1mJ$ .



**Fig. 4.** Peak THz field plotted against  $W_G$ , for a Zn-doped < 100 > p-InAs substrate, for various screening energies  $W_S$  fixed at  $\tau_s = 0$ . Each measurement was performed with a generating pump energy  $W_G$  of  $0.1mJ$ . The peak THz field has been normalised across with respect to the maximum peak field generated by Zn-doped InAs.

distribution in the example of Fig. 4 is certainly very different from the almost homogenous free-carrier density of the n-type sample. Then, the close matching of the trends of Fig. 2 and Fig. 4 suggests that all the screening dynamics must occur within depths much smaller than the optical skin depth (so that the carrier profile plays a minor role), which also corroborates the main hypothesis. This also suggests that the carrier induced screening of the THz propagating field plays a minor role (the typical THz skin-depth in the used n-doped materials lies in the range of hundreds of nm to micrometre scales).

In conclusion, a physical process for the saturation of surface optical rectification at high fluences has been proposed. We argue that as the injection of photo-carriers increases, the thickness of the surface field region contracts and a progressively larger fraction of the photocarriers are excited outside of the surface field region. As those carriers do not contribute to the surface field screening, a quadratic conversion trend (i.e. a linear dependence of the peak THz field from the generating pump energy) is eventually re-established at high excitation energies.

As confirmation of our hypothesis, we verified that the depth of this surface field layer, and therefore the saturation trend of the substrates, can be controlled with either doping of the material or by injecting an equivalent quantity of photocarriers. We believe this to be an advancement in the understanding of the THz surface generation physics at very high excitation and a step forward in the realisation of high energy surface THz emitters.

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### Data availability

The data sets for all the experimental figures are freely accessible at <https://doi.org/10.25377/sussex.5777712> (<http://dx.doi.org/10.25377/sussex.5777712>).

### Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.nanoen.2018.01.027>.

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