Relationship between phase and generation mechanisms of THz waves in InAs

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1. Introduction

InAs has been considered one of the most intriguing materials for terahertz wave generations because it is the most efficient semiconductor surface emitter ever known and its efficiency is significantly enhanced under a magnetic field [1,2]. It has also been revealed that the transient diffusion current of electrons and holes with different diffusion velocities (photo-Dember effect) [3] and/or the second-order nonlinear processes (optical rectification) [4] particularly in the high excitation-power regime cause THz electromagnetic (EM) wave radiation from InAs.

To the underlying THz radiation mechanisms, optimal engineering factors including crystalline orientation, carrier type and doping density have been investigated in attempts to enhance the amplitude. Specifically, (111)-oriented InAs was demonstrated to utilize the optical rectification effect. It was noted that the angle between the crystallographic direction and excitation laser polarization should be properly chosen [5]. Moreover, an investigation into the carrier type and doping density dependence on the radiation intensity was conducted by Kai Liu et al. [1], where p-type doping with low carrier concentration (on the order of 10^{16} cm^{-3}) was considered the most favorable for intense THz radiation.

An important but much less investigated parameter is the light–matter interaction length, required to characterize and optimize the THz waves. Recently, the dependence of THz radiation intensity on material thickness was introduced for a limited range of thicknesses, from 150 to 520 nm [6]. However, investigations into the relationship between sample thickness and the concomitant THz wave generation mechanisms, encompassing both the diffusion length and optical absorption depth, has yet to be reported. Furthermore, the THz signal polarity change in InAs has not been observed in spite of material parameters changes to our knowledge, whereas in materials such as GaAs different doping type [1] or the external bias [7] can invert the polarity of the THz waves via the acceleration direction control. Such information could be useful for further optimizing scaling parameters in THz emission sources; e.g., periodic metal patterns were employed to create the lateral photo-Dember currents in GaAs and InGaAs layers [8].

In this paper, we measured the transient THz waves emitted from optically excited InAs layers, whose thicknesses were varied from 0.01 to 1.74 µm. Based on comparative analysis between phase information of THz waves and the carrier dynamics anticipated from the band diagram, we discuss the thickness-specific generation mechanisms of THz waves and the minimum thickness of InAs for optimized THz emission.

2. Experimental schemes and samples

We performed THz time-domain spectroscopy (TDS) measurements in optically excited InAs layers. We utilized two independently-tunable Ti:sapphire lasers having pulse durations of 160 fs, synchronized within a jitter of 113 fs at 800 nm. Each laser at

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a repetition rate of 76 MHz was then devoted to either the sample excitation or detection. The excitation beam, with ~0.5 W average optical power, was focused onto the samples within a spot size of 1 mm, such that the laser fluence remained below the saturation level [4]. Finally, the THz beam was collected and guided to the photo-conductive antenna (PCA), optimized for a broad range around 1 THz.

Six undoped InAs samples with different thicknesses were prepared: 0.01, 0.02, 0.07, 0.37, 0.90 and 1.74 µm. A 2.2 µm-thick AlAs$_{0.32}$Sb$_{0.68}$ buffer layer was adopted to reduce the lattice mismatch between the InAs (lattice constant: 6.06 Å) layer and (100) GaAs (lattice constant: 5.65 Å) substrate. The band gap of the AlAs$_{0.32}$Sb$_{0.68}$ was estimated to be about 1.8 eV [9], which is much higher than the excitation photon energy (1.55 eV). Each InAs layer was grown along a (100) crystalline orientation to reduce the optical rectification effect [1]. Consequently, the excited carriers were well confined within the InAs layers; thus, we could concentrate on the influence of the InAs layers thickness. From the AFM and X-ray diffraction experiments, we found that InAs film has considerably homogeneous surface. The root mean square roughness of the surface has a increasing tendency from 0.9 nm to 5.8 nm with InAs thickness increment in our range, which was on a much smaller scale than the absorption depth (142 nm) [10] and the excitation laser beam spot size.

3. Results and discussion

The two mechanisms of photo-Dember effect and the current-surge by photo-carriers around the tilted potential regions can independently contribute to the THz generations: At the surfaces of GaAs, GaSb, and InP, the THz generation was known to be originated from the carrier acceleration in the band-bending regions with n-type or p-type doping [11]. On the other hand, the emission from InAs has been reported to be dominated by the photo-Dember effect, mostly studied in a scale much larger than the diffusion length ($L_d$) [12]. Because both mechanisms are stemmed from the generation of transient electric dipoles, it is important to measure the electron mobility and the sheet carrier density prior to the THz measurements. We plot the electron mobility and electron sheet carrier density as a function of sample thickness in Fig. 1(a). The mobility monotonically increased with thickness, whereas the electron sheet carrier density increased in a group of thin samples (0.01–0.07 µm) followed by slight decrease in thick sample group (0.37–1.74 µm). Mobility change could be associated with the THz radiation intensity via the Einstein relation on the diffusion coefficient. The doping density, on the other hand, modifies the surface depletion region width with negligible screening effect in contrast to the high carrier density regime beyond $10^{16}$ cm$^{-2}$ [1].

In Fig. 1(b), we have estimated $L_d$ based on the mobility values measured in our samples, considering the excess energy of excitation laser above the band gap (~1.2 eV) [13] and relaxation time (≥0.6 ps), separately extracted from transient differential reflectivity measurements (not shown here). Samples could be classified into either thin sample group where the thickness (dotted line) was smaller than $L_d$ or thick sample group with thickness larger than $L_d$.

Fig. 2(a) shows the THz-TDS results in the reflection geometry. The signal-to-noise ratio was sufficient for measuring even the weakest signal in the thinnest sample, as magnified by ten times. As a reference, THz waves from GaAs substrate is displayed on top as being illuminated from the backside. In Fig. 2(b), we traced the amplitude with the InAs layer thickness. In contrast to the gradual change of the electron mobility and sheet carrier density, the THz intensity rapidly increased as the thickness became comparable to the absorption depth. The observed increment beyond the absorption depth is reasonable due to the efficient carrier diffusion and concomitant THz radiation (photo-Dember effect). Most interesting results were as follows: (1) With increasing InAs thickness, the amplitude was enhanced accompanying a phase inversion at about 0.37 µm. (2) In spite of the large amplitude change, there was negligible change within the sample groups.

To understand the unusual phase inversion and amplitude changes at 0.37 µm, we further inspected the energy band diagram, as simulated in Fig. 3. In particular, we noted the direction of the band bending and the range of the electric field, not only at the InAs surface but also at the InAs-AlAs$_{0.32}$Sb$_{0.68}$ interface; the band-bending region extended either for ~0.1 µm (at the InAs surface) or for ~0.07 µm (at the InAs-AlAs$_{0.32}$Sb$_{0.68}$ interface). We used the electron sheet carrier densities at the InAs surfaces as a parameter for the calculation whereas we ignored the strain effect at the InAs-AlAs$_{0.32}$Sb$_{0.68}$ interface.

In Fig. 3(a), the photo-excited carriers are swept away by the surface electric field, overriding the diffusion along the opposite direction in the thin sample group. In this case, the direction of the electric dipole is toward the substrate side. For the thick sample group, the THz emissions mainly occur by the photo-Dember effect with the dipole direction being inverted toward the surface of InAs layer. Therefore, we expect that the main THz radiation mechanisms transited from current surge to photo-Dember effect between 0.07 µm and 0.37 µm. In this regard, the stagnated signal amplitude at 0.07 µm (cf. Fig. 2(b)) possibly implies that the carrier acceleration effect was canceled out by the co-existing diffusion. In differential transient reflectivity measurements using Ti:sapphire lasers with 15 fs-pulse duration (not shown here), we have

![Fig. 1](image-url) (a) Electron sheet carrier densities and mobilities measured at 300 K as a function of thickness. (b) Estimated diffusion length based on mobility and relaxation time in each sample.
observed a pulse-form limited rapid decaying feature in samples thicker than 0.07 μm, which could substantiate the carrier swept-out dynamics from the surface due to the diffusion effects. In samples thicker than 0.37 μm, the calculated band-bending region was much smaller than $L_d$. Accordingly, the electron diffusion can dominate the transient dipole formation process, resulting in the inverted phase and enhanced amplitudes in the thick sample group (0.37–1.74 μm). Even with a larger mobility, thus with a larger diffusion length, signal amplitude was slightly reduced in 1.74 μm, possibly associated with lower sheet carrier density.

While we attribute the THz emission to the photo-Dember effect in thick samples, Kuznetsov et al. discussed the optical rectification effect even in isotropic plane due to virtual carriers [14]. This effect results from the creation of virtual carriers from femtosecond laser excitations just off resonance and below transition energy. These virtual electrons and holes have a dipole moment which adiabatically follows the laser pulse envelope in the signal (in contrast to rather slow response as seen in this paper), interestingly with a sign reversal. Since these virtual carriers are excited below the resonance whose density increases with the sample thickness, the band structure of InAs needs to be examined; the 800 nm excitation is quite close to the energy difference between the X-valley minimum in the conduction band and the $G$-valley maximum in the valence band with and excess energy of about 180 meV, which implies

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**Fig. 2.** (a) Measured THz waveform from InAs samples with different thicknesses, compared to backside illumination on GaAs. (b) The integrated amplitudes of THz waves.

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**Fig. 3.** Simulation results of the band diagrams for the (a) 0.07, (b) 0.37, (c) 0.90, (d) 1.74 μm-thick samples.
inter-valley scattering rather than the thickness-dependent virtual carrier excitation possibly influence our spectra. The inter-valley scattering could change the effective mass and diffusion constants which will be further discussed elsewhere.

Finally, we removed the GaAs substrate of the 0.9 μm-thick InAs sample using the AlAs0.32Sb0.68 as a etch-stop layer and the thin InAs layer was attached to a sapphire substrate for a THz measurement in transmission geometry. THz-TDS results in Fig. 4(a) shows that the thin InAs layer on the transparent substrate can now be used for the THz emission along the transmission direction (the same with excitation laser). When we compared the Fourier-transform spectra in Fig. 4(b), the transmission represented much narrower spectral width with multiple harmonics, implying the Fabry-Perot-type interference within the sapphire substrates. This extremely thin layer of InAs could be attached on various materials; e.g., we recently attached 0.9 μm-thick InAs layer on a tip of optical fiber for a THz imaging scheme beyond its diffraction limit [15]. Another application example could be the laser THz-emission microscopy (LTEM) [16], in which the resolution is limited by the optical spot size of the excitation laser overriding the diffraction limit of THz but has been applied to the THz-emitting (conducting) materials in reflection geometry. To apply such methodology to general non-conducting materials, an additional THz emitter should be attached together or be very close to the samples preferably in transmission geometry [15], where the thickness of emitter is now the decisive factor for obtaining the better resolution. This possibly exemplifies a novel methodology for engineering diffusive semiconductors for various THz applications.

4. Conclusion

In conclusion, we have carried out THz-TDS in InAs epilayers as a function of thickness, especially around the absorption depth and diffusion length. The phase inversion at 0.37 μm implies the transition of dominant THz generation mechanisms from the carrier acceleration into diffusion effect, which can be analyzed further in the viewpoint of thickness-dependent variation of dipole alignment in relevant band diagram. We found ~1 μm to be the optimum thickness for generating THz emissions among our sample structures. We further utilized the thin InAs layer to demonstrate a transmissive THz emitter whose thickness was negligibly small in THz region, such that the emitted THz beam spot size could possibly be maintained within the diffraction limit as the case may be [15].

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