Terahertz emission from gradient InGaAs surfaces

J.L. Regalado-de-la-Rosa,1 A. Belio-Manzano,2 V.H. Mendez-Garcia,2 and E. Castro-Camus1,3
1Centro de Investigaciones en Optica, A.C., Loma del Bosque 115, Lomas del Campestre, Leon, Guanajuato 37150, Mexico
2Center for the Innovation and Application of Science and Technology, Universidad Autónoma de San Luis Potosí, Sierra Leona 550, Lomas 2a Sección, San Luis Potosí 78210, Mexico
3Department of Physics, Philipps-Universität Marburg, Renthof 5, 35032, Marburg, Germany.

e-mail: enrique.castrocamus@physik.uni-marburg.de

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We present an experimental study of the terahertz emission from InxGa1−xAs epitaxial layers that were grown with varying the alloy fraction x. We observe a terahertz emission that is significantly different depending on the variation of the alloy fraction. We attribute the difference to the significant change of the band bending induced by the growth direction and to the position-dependent variation of the effective mass.

The emission of terahertz electromagnetic transients from semiconductor surfaces after ultrafast photoexcitation is a topic that has attracted enormous attention over the last couple of decades1–6. Not only because it provides a convenient and passive source of terahertz radiation7,8 but also because it is a potential tool to understand the carrier dynamics of the semiconductor material in the picosecond timescale9.

The generation of terahertz radiation at the surface of a semiconductor after photoexcitation has been attributed to two main effects. First, the ballistic transport of electrons that acquire most of the kinetic energy resulting from the difference between the photon and the bangap10. Secondly, the acceleration experienced by the electrons owing to the electric field caused by the band bending in the vicinity of the surface11,12. The relative weight of these two effects strongly depends on the wavelength of the laser pulse, the bandgap of the semiconductor and the effective mass of the carriers in the material10.

In this article we study the terahertz emission of gradient InxGa1−xAs structures. In other words, samples in which the alloy fraction x was varied monotonically during the growth process. The resulting bandgap variation in the direction of growth brings an additional electric field inbuilt in the sample, which can point either towards the surface or towards the bulk. This modifies the carrier dynamics, and therefore the terahertz emission of the samples.

When excited by 800 nm pulses it is well known that the THz emission of GaAs is mostly due to the carrier acceleration caused by the band-bending near the surface which is schematically represented in Fig. 1a. On the other hand, for InAs, the main contribution comes from ballistic transport, owing to its very low carrier effective mass, and the rather large excess of energy of the photoexcited carriers associated to its narrow bandgap, also schematically represented in Fig. 1b10. In this article we propose two gradient InxGa1−xAs structures in which the alloy fraction is continuously varied in the direction normal to the surface from 1 to 0 (Fig. 1c) and vice versa (Fig. 1d).

The following samples were grown by molecular beam epitaxy. The substrate temperature was set at 525 and 605°C for the growth of InAs and GaAs, respectively, and it was linearly changed between these values for the growth of the gradient structures. All samples were grown on SI-GaAs (100) substrates.

1. GaAs: A 1 μm thick GaAs layer with an estimated carrier density of 1 × 10^14 cm^{-3}.
2. InAs: A 1.5 μm thick InAs layer with a carrier density of 8 × 10^16 cm^{-3}.
3. Inx>Ga: A 0.8 μm-thick gradient layer which starts with an alloy fraction of x = 0.97 (~InAs) and ends with an alloy fraction of x = 0.12 (~GaAs) with an estimated carrier density of 7 × 10^16 cm^{-3}.
4. Ga>x>In: A 0.8 μm layer which started with an alloy fraction of x = 0.12 (~GaAs) and ended with an alloy fraction of x = 0.97 (~InAs) with an estimated carrier density of 8 × 10^16 cm^{-3}.

Each sample was cleaved into two pieces, one set was served as-grown and the other set was annealed by raising the temperature slowly until reaching 250°C where an Ar atmosphere was introduced, in order to avoid surface damage. The temperature was further increased until reaching 600°C and maintained for 10 minutes.

All samples were studied by x-ray diffraction14,15. GaAs and InAs samples show diffraction patterns typical...
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![Graphs showing x-ray diffraction intensity as function of the angle for as-grown In>Ga and Ga>In samples. Panels (c) and (d) are the corresponding ones for the annealed samples.]

The lattice constant of those two crystals and we will not discuss them here. The diffraction intensity for the gradient samples as a function of the angle is shown in Fig. 2a and b. What we observe is a strong peak at $\sim 33.03^\circ$, which corresponds to the GaAs lattice. In addition, a less prominent, but still large peak is observed at $\sim 30.53^\circ$, which corresponds to the 98% In lattice constant. However, these two peaks are continuously joined by a plateau of lower intensity, this demonstrates the presence of continuously varying lattice constant, consistent with the presence of a gradient. These features have been observed before in similar samples. The gradient reported here is rather large, going from $x\sim 0$ to $x\sim 1$, therefore plastic relaxation of the film at certain thickness must have occurred. Which leads to the low and continuous diffraction intensity plateau that corresponds to the graded film region. Additional subtle features can be seen in the curves of Fig. 2a and b. The small, but visible peak at around 32.6° in Fig. 2b for the Ga> In sample corresponds to the diffraction of a 16% indium concentration alloy, which is consistent within our error to the composition of 12% at which the gradient started. Similarly, In>Ga sample started with an In concentration of 98% explaining the peak located at 30.74° in Fig. 2a. The slight differences with the nominal concentration for both of the gradients are related to strain. The annealing process seems to have two important effects on the samples. The plateau appears higher for both samples, which suggests an improvement of the crystallinity along both gradients, most likely owing to the removal of lattice defects. In addition, as seen in Fig. 2d, the plateau of Ga> In seems to be more constant, suggesting a “softening” of the lattice discontinuity in the vicinity of the pure GaAs substrate.

In order to characterize the samples, a terahertz time-domain spectroscopy setup was built based on a Ti:sapphire mode-locked laser that emits pulses of $\sim 33$ fs duration centered at 800 nm at a repetition rate of 80 MHz. The pulses are split into two beams. The first one, with an average power of $\sim 50$ mW is sent through a motorized delay line and then is used to excite the semiconductor sample at an incidence angle of $45^\circ$. The THz radiation generated in the form of a single cycle electromagnetic transient, is collected at $45^\circ$ as seen in Fig. 1e by using an off axis parabolic mirror. A second parabolic mirror focuses the THz radiation onto a 1 mm thick ZnTe crystal. The second laser beam is also focused onto the ZnTe crystal and copropagates with the THz radiation, subsequently an achromatic $\lambda/4$ waveplate, a Wollaston prism and a pair of balanced photodiodes are used to detect the small changes in the relative intensity of the laser polarized components, which is proportional to the instantaneous electric field. By changing the relative delay between the first laser pulse and the second laser pulse it is possible to manipulate the time-dependent waveform of the THz electric field. Further details about electrooptic sampling of THz pulses can be found in Valdmanis, Mourou, and Gabel.

The time-domain signals for the four materials is shown in Fig. 3a and b for the as-grown and annealed samples respectively. Among the as-grown samples, InAs has the strongest emission, it is worth noticing that the as-grown InAs had excellent crystal quality according to in-situ high energy electron diffraction except for the first $\sim 2$ nm near the interface. The second strongest signal is the one produced by the annealed In>Ga gradient sample. We calculated the integrated power of the samples, which is shown in Fig. 3c. We can see that the emission of InAs is about 13 times higher than the power emitted by as-grown GaAs, while the power emitted by annealed In>Ga is about 10 times that of as-grown GaAs. Among the interesting things to point out in this plot is the significant reduction of the InAs power after annealing. It is also remarkable that except for the two strongly emitting materials already mentioned, the rest all have relatively similar emission powers. An exception is the annealed Ga> In sample, which shows the lowest emission of all.
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The annealed In\textgtr Ga sample, is a very strong emitter, almost comparable to InAs. In order to explain this, we will refer to the schematics in Fig. 1a-d. The In\textgtr Ga sample is composed of GaAs on the surface, which has a relatively broadbandgap and, owing to the alloy-variation, the bandgap narrows down as we move into the bulk, this produces a strong acceleration of the electrons which adds to the acceleration caused by the surface field. In other words, the force experienced by an electron is mostly given by the difference of potential energy between the initial position, near the surface, and the final position, away from the surface. Thus, the large band bending of the gradient contributes with a significant acceleration. Furthermore, the force acting on the carrier is given by

$$ F = \frac{dp}{dt} = m^* a + v \frac{d\delta m}{dt}. $$

where $p$ is the momentum, $m^*$ is the effective mass and $a$ is the acceleration. Therefore, the acceleration will also have an additional term given by $-\delta m^*/dt$, in the case of the In\textgtr Ga, the derivative of the mass is negative, resulting in an additional positive term that contributes to the acceleration. In order to do an “order-of-magnitude” estimation of the terahertz emission caused by the acceleration experienced by the electrons in all four semiconductor samples (the contribution of the holes is neglected here), we assumed that the average electron was accelerated from 0 to $v_\text{f} = \sqrt{2K/m}$, on average, over a period $\delta t \sim 100$ fs which is about 2 to 3 times the duration of the laser pulse, where $K$ is the kinetic energy, given by the difference between energy of the $\lambda=800$ nm photons and the bandgap energy at the surface $K = \hbar c/\lambda - E_{BG}(0)$. In addition, if we make the right-hand-side part of Eq. 1 equal to $-dU/dx$ the gradient of the potential, which we can approximate as a linear function that changes $\sim E_{BG}(0)/2$ about half the bandgap energy at the surface for all samples owing to surface pinning, plus the bandgap change owing to the alloy variation for the In\textgtr Ga and Ga\textgtr In samples. Therefore in total the acceleration experienced by the electrons will be approximately

$$ a = \frac{\bar{v}_i}{\delta t} - \frac{1}{m} \left( \frac{1}{2} \left( \frac{E_{BG}(0)}{\delta x} + \frac{\Delta E_{BG}}{\delta x} \right) \right) - \frac{1}{2} \frac{\bar{v}_i}{m} \frac{\Delta m}{\delta t}. $$

where $\delta x \sim 1 \mu m$ is an estimation of the surface deplanation, as well as the distance over which the alloy fraction is varied. In addition, $\Delta E_{BG}$ and $\Delta m$ are the total change of the bandgap and the change of effective mass over the alloy-fraction layer.

Using this simplistic approach and using the parameters show in Table I. Since the emission is proportional to the acceleration, in order to present the estimation, all the values obtained where normalized to that of GaAs, and are shown in Fig. 3c. The estimation shows qualitative resemblance, and predicts correctly the relative order of the strength of the materials, supporting the qualitative picture that we have presented. Of course, it is a rather approximate model that does not take into account many subtleties of the carrier dynamics, including the carrier screening of the fields, and the differences in absorption coefficient, among others.

We attribute the enormous difference observed between the annealed and the as-grown sample to the defects in the latter, which will contribute with both scattering centers and traps which will reduce the current transient responsivity and the emission. During the annealing process the lattice rearranges and this reduces the number of defects, increasing the mobility and reducing the number of traps, both in the bulk and those on the surface. This is consistent with the analysis of the X-ray diffraction presented earlier. In the case of the reverted gradient, namely Ga\textgtr In, the situation is reversed; the band bending caused by the alloy-variation produces an electric field that competes with the surface field, and points in the direction opposite to the ballistic motion of electrons into the bulk, therefore reducing the emission. This applies to the $-\delta m^*/dt$ term, which now points in the opposite direction since the effective mass increases. Finally, we want to point out that both InAs and Ga\textgtr In, which are samples that have Indium at the surface, experience a reduction of the emission with the annealing process. We believe this is because these two samples became semi-metal-like to the surface during the baking process, which lead to a reduction that we could observe by optical and atomic-force microscopy images of the annealed samples (not shown). This resulted in a very high carrier density, and therefore high conductivity, which in turn reduces the terahertz emission.

In summary, we have presented a terahertz emission of In\textgtr Ga\textgtr In samples. We observed that the emission strongly depends on the direction in which the alloy-fraction changes, producing strong emission when the bandgap narrow in the bulk and larger at the surface and a weak emission in the opposite case. The strong terahertz emission of the first case is comparable to that of InAs. This is expected by the electric field imposed by the variation of the bandgap energy, which promotes acceleration of electrons toward the bulk in the In\textgtr Ga sample, but inhibits the acceleration in Ga\textgtr In case.

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| Parameter | GaAs | InAs | In\textgtr Ga | Ga\textgtr In |
|-----------|------|------|--------------|--------------|
| $E_{BG}(0)$ | 1.42 | 0.35 | 1.42 | 0.35 |
| $\bar{v}_i$ | 0.066 | 0.022 | 0.041 | 0.041 |
| $\Delta E_{BG}$ | 0 | 0 | -1.08 | 1.08 |
| $\Delta m$ | 0 | 0 | -0.041 | 0.041 |
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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

CONFLICT OF INTERESTS

The authors have no conflicts to disclose.

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Growth direction

(a) GaAs

(b) InAs

(c) In$>$Ga

(d) Ga$>$In

(e) To electro-sampli...
