Emission of terahertz-frequency electromagnetic radiation from bulk Ga$_{1-x}$In$_x$As crystals

Youngok Ko,1 Suranjana Sengupta,2 Stephanie Tomasulo,2 Partha Dutta,1 and Ingrid Wilke2

1Electrical, Computer and System Engineering Department, Rensselaer Polytechnic Institute, 110 8th Street, Troy, New York 12180, USA
2Department of Physics, Applied Physics and Astronomy, Rensselaer Polytechnic Institute, 110 8th Street, Troy, New York 12180, USA

(Received 29 April 2008; published 1 July 2008)

We report an experimental study of femtosecond optically excited emission of terahertz frequency electromagnetic radiation from Ga$_{1-x}$In$_x$As bulk crystals with alloy composition in the range between 0 < x < 0.65. The terahertz-emission mechanisms in bulk Ga$_{1-x}$In$_x$As were studied as a function of carrier mobility, carrier concentrations, band gap as well as polycrystal grain size. Our experiments and analysis demonstrate that optical rectification is the dominant terahertz-emission mechanism for Ga$_{1-x}$In$_x$As with 0.01 ≤ x ≤ 0.43 while surface-field acceleration is dominant for Ga$_{1-x}$In$_x$As with 0.43 ≤ x ≤ 0.64. The magnitude of terahertz emission due to optical rectification is a function of the polycrystal grain size of the Ga$_{1-x}$In$_x$As samples. Terahertz emission from Ga$_{1-x}$In$_x$As due to optical rectification is due to a $\chi^{(2)}$ nonlinear optical process. Overall, terahertz emission from Ga$_{1-x}$In$_x$As is maximized for $x = 0.1 - 0.3$.

DOI: 10.1103/PhysRevB.78.035201 PACS number(s): 78.47.J–, 71.28.+d, 78.20.Jq, 95.85.Gn

I. INTRODUCTION

Binary III-V compound semiconductor materials have received great attention as potential sources of terahertz frequency electromagnetic radiation. GaAs, GaSb, InN, InP, InAs, InAlAs, InGaAs, and InSb have demonstrated emission of subpicosecond terahertz-radiation pulses upon irradiation with femtosecond near-infrared laser pulses. This type of terahertz radiation source has enabled the development of time-domain terahertz spectroscopy and terahertz imaging over the last decade. However, important applications of these techniques in basic and applied science such as nonlinear terahertz spectroscopy, nondestructive testing, or biomedical imaging are still limited by the power of the available sources. The development of bright, high bandwidth terahertz-radiation sources is important in order to expand the applications of these techniques.

Terahertz-radiation emission from binary III-V compound semiconductors exposed to femtosecond near-infrared laser pulses originates either from a nonlinear optical process or ultrafast photocurrents. Nonlinear optical processes in semiconductors resulting in terahertz-radiation emission are bulk, or surface-field induced optical rectification of the incident femtosecond near-infrared laser pulses. Terahertz-radiation emission due to ultrafast photocurrents can be achieved through acceleration of photocarriers by intrinsic or extrinsic electric fields. Intrinsic electric fields occurring at a semiconductor surface are surface depletion/accumulation fields or a Photo-Dember field. Extrinsic electric fields are generated by applying a voltage laterally across a gap between two metal electrodes deposited on the semiconductor surface (photoconducting switch). InAs, a narrow band-gap semiconductor ($E_g = 0.36$ eV) has demonstrated the strongest terahertz-radiation emission of all III-V semiconductor systems characterized to date.

The origin of terahertz-radiation emission from InAs is primarily attributed to the Photo-Dember effect and surface-field induced optical rectification. Conversely, GaAs, a semiconductor with a wider band gap ($E_g = 1.42$ eV) has shown terahertz-radiation emission dominated by different physical phenomena. GaAs, dominant terahertz-radiation mechanisms are acceleration of carriers by a surface depletion field and bulk optical rectification.

The III-V ternary alloy semiconductor Ga$_{1-x}$In$_x$As is a very interesting terahertz materials system. It is expected to exhibit properties physically related to both binary systems InAs and GaAs. Moreover, the band gap of Ga$_{1-x}$In$_x$As can be tuned from 0.36 to 1.42 eV by variation of the Ga mole fraction from x = 0 to x = 1. Therefore, Ga$_{1-x}$In$_x$As is an attractive material system for compact and lightweight time-domain terahertz spectroscopy and imaging systems powered by femtosecond fiber lasers with emission wavelengths between 0.75 and 1.55 μm.

However, ternary compound semiconductors such as Ga$_{1-x}$In$_x$As are extremely difficult to grow as bulk crystals and are not commercially available. Hence, very limited research has been carried out on terahertz emission from this material. Only a few compositions of Ga$_{1-x}$In$_x$As thin films were researched previously. All previous research was limited to Ga$_{1-x}$In$_x$As thin films grown by molecular-beam epitaxy on binary substrates. Furthermore, research was restricted to study terahertz emission from Ga$_{1-x}$In$_x$As photovoltaic switches.

In this article, we present an experimental study of femtosecond near-infrared excited terahertz emission of Ga$_{1-x}$In$_x$As. In contrast to previous work, we investigate bulk Ga$_{1-x}$In$_x$As crystals grown by the vertical Bridgman method over a wide alloy composition range 0 ≤ x ≤ 1. Furthermore, we focus on the investigation of terahertz-radiation emission by the bare Ga$_{1-x}$In$_x$As surface. We investigate in-depth the relationship between electrical and structural properties of bulk Ga$_{1-x}$In$_x$As and terahertz-radiation emission. We demonstrate that primarily optical rectification and surface-field acceleration contribute to terahertz emission from Ga$_{1-x}$In$_x$As depending on the Ga mole fraction x. We show that emission of terahertz radiation from Ga$_{1-x}$In$_x$As is maximized for x in the range of 0.1–0.3.
II. EXPERIMENTAL ARRANGEMENTS

Ga\textsubscript{x}In\textsubscript{1-x}As ingots were grown by the vertical Bridgman technique and the gradient freezing methods, namely vertical and horizontal gradient freeze techniques.\textsuperscript{40,45} After crystal-growth Ga\textsubscript{x}In\textsubscript{1-x}As, ingots were sliced into wafers of 1 mm thickness by a Low Speed Diamond Wheel Saw 660 from South Bay Technology. An Omnilap 2000 wafer polishing unit from South Bay Technology was used for lapping and polishing of the wafers. The lapped wafers were first polished with 1 \textmu m alumina slurry on a nylon pad. Mirrorlike shining surfaces were achieved by a second polishing with 0.01 \textmu m alumina slurry on a velvet pad.

Electrical characterization of our Ga\textsubscript{x}In\textsubscript{1-x}As samples was performed using a HEM–2000 EGK Hall Measurement System from EGK Co. Hall measurements were performed at 77 and 300 K with 0.37 or 0.51 T of magnetic flux and 1 mA of input current. The percentage errors for the measured electrical parameters were 3% for mobility, 2% for resistivity, and 2% for carrier concentrations.

An JEOL-733 electron probe microanalyzer (EPMA) was used to characterize the atomic composition of the Ga\textsubscript{x}In\textsubscript{1-x}As crystals. Samples were coated with approximately 200 Å of carbon using a Denton Vacuum DV–502A high-vacuum carbon evaporator. Calibration data was obtained using Probe Laboratory standards of GaAs and InAs for Ga, In and As. Data analysis was performed using GELLER ANALYTICAL DQUANT32 software. The Henrich model was used for the ZAF correction. The band gap of our Ga\textsubscript{x}In\textsubscript{1-x}As crystals were measured by Fourier-transform infrared spectroscopy (FTIR).

Electron beam backscattering diffraction (EBSD) was used to determine the crystallinity of our Ga\textsubscript{x}In\textsubscript{1-x}As samples and the crystallographic orientations of the surface of the grains. EBSD measurements were performed by an environmental scanning electron microscope FEI/Phillips XL30 ESEM-FEG, a Nordlys I EBSD detector by HKL Technology Inc. and with an HKL CHANNEL 5 software package.

The time-domain terahertz-emission measurement setup that was used for this work is an optical pump-probe arrangement.\textsuperscript{46} A commercial diode-pumped titanium-sapphire laser delivers pulses with a duration of 130 fs at a wavelength of 800 nm. The repetition rate is 82 MHz and the maximum average power is 700 mW. The laser beam is split into a pump beam and a probe beam using an uncoated glass as a beam splitter. The power ratio is typically 95% on the pump beam and 5% on the probe beam. Emission from unbiased semiconductor surfaces is achieved by exposing the surface to the pump light beam. The pump beam is slightly focused at the semiconductor surface with a laser spot size of 1 mm\textsuperscript{2} and an injected carrier density of approximately 10\textsuperscript{17} cm\textsuperscript{-3}. Measurements were performed setting the angle between the laser beam and the surface’s normal to 45\textdegree. The pulsed terahertz radiation is detected through electro-optic sampling\textsuperscript{47} using a (110) ZnTe crystal of 1 mm thickness. The bandwidth of our electro-optic terahertz detector is limited to 3 THz.

III. RESULTS AND DISCUSSIONS

The alloy compositions of our Ga\textsubscript{x}In\textsubscript{1-x}As samples as obtained by electron probe microanalysis (EPMA) are illu-
trated in Fig. 1. In our samples, the Ga mole fraction varies between $x=0.01$ to $x=0.64$. The band gap of the Ga$_{1-x}$In$_x$As samples was determined by FTIR absorption measurements and range from $E_g=0.36$ eV to $E_g=0.94$ eV (Fig. 2). The measured values of the Ga$_{1-x}$In$_x$As band gap agree very well with the empirical relation between band gap and electron concentrations and electron mobilities of all Ga$_{1-x}$In$_x$As and InAs are determined by Hall-effect measurements. Table II provides a summary of the alloy compositions, conductivity type, carrier concentrations, mobilities, and band gaps for these samples at 300 K. Table I. Electrical properties of Ga$_{1-x}$In$_x$As [Ref. 39]

<table>
<thead>
<tr>
<th>Sample</th>
<th>Ga (x)</th>
<th>In (1-x)</th>
<th>Band gap (eV)</th>
<th>Carrier concentration (cm$^{-3}$)</th>
<th>Hall mobility (cm$^2$V$^{-1}$s$^{-1}$)</th>
<th>Domain size (mm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>W1</td>
<td>0.01</td>
<td>0.99</td>
<td>0.37</td>
<td>$3.25 \times 10^{16}$</td>
<td>$n$ type</td>
<td>20700</td>
</tr>
<tr>
<td>W5</td>
<td>0.09</td>
<td>0.91</td>
<td>0.37</td>
<td>$2.10 \times 10^{16}$</td>
<td>$n$ type</td>
<td>16174</td>
</tr>
<tr>
<td>W6</td>
<td>0.29</td>
<td>0.71</td>
<td>0.59</td>
<td>$9.49 \times 10^{15}$</td>
<td>$n$ type</td>
<td>11601</td>
</tr>
<tr>
<td>W7</td>
<td>0.30</td>
<td>0.70</td>
<td>0.59</td>
<td>$9.08 \times 10^{15}$</td>
<td>$n$ type</td>
<td>10100</td>
</tr>
<tr>
<td>W8</td>
<td>0.44</td>
<td>0.56</td>
<td>0.72</td>
<td>$5.30 \times 10^{15}$</td>
<td>$n$ type</td>
<td>6566</td>
</tr>
<tr>
<td>W12</td>
<td>0.64</td>
<td>0.36</td>
<td>0.94</td>
<td>$6.74 \times 10^{14}$</td>
<td>$n$ type</td>
<td>3320</td>
</tr>
</tbody>
</table>

Full crystallographic orientation data of the domains are obtained by EBSD. The orientation of a domain is expressed by three Euler angles $\phi$, $\theta$, $\psi$ which describe the relationship between sample reference coordinate ($x'$, $y'$, and $z'$) and crystallographic coordinate ($a$, $b$, and $c$). The measured Euler angles demonstrate that the overall disorientations among the domains are small. We observed emission of terahertz-frequency electromagnetic radiation from all investigated Ga$_{1-x}$In$_x$As samples with $0.01 \leq x \leq 0.64$. The terahertz transients and frequency spectra emitted by Ga$_{0.01}$In$_{0.99}$As and Ga$_{0.64}$In$_{0.36}$As are displayed as examples in Fig. 5. We observe that Ga$_{0.01}$In$_{0.99}$As exhibits stronger terahertz emission than Ga$_{0.64}$In$_{0.36}$As. Normalized terahertz frequency spectra for Ga$_{0.01}$In$_{0.99}$As and Ga$_{0.64}$In$_{0.36}$As are illustrated in Fig. 6. We observe that Ga$_{0.01}$In$_{0.99}$As emits terahertz radiation with a slightly broader bandwidth than Ga$_{0.64}$In$_{0.36}$As.

Furthermore, we have investigated the terahertz emission from Ga$_{1-x}$In$_x$As as a function of the angle between the linear polarization of the excitation laser beam and the crystallographic orientation of the Ga$_{1-x}$In$_x$As surface for the entire compositional range ($0.01 \leq x \leq 0.64$). For this experiment, the Ga$_{1-x}$In$_x$As sample was rotated with the surface normal as axis of rotation. The terahertz emission from Ga$_{1-x}$In$_x$As was measured in reflection as a function of azimuth angle $\phi$. The results of these measurements for Ga$_{0.01}$In$_{0.99}$As and Ga$_{0.64}$In$_{0.36}$As are illustrated as examples in Fig. 7.
served that the magnitude of terahertz emission is a function of the angle $\phi$ between the polarization of the laser beam and Ga$_x$In$_{1-x}$As surface orientation. The magnitude of terahertz emission changes periodically as a function of angle $\phi$. The amplitude of the oscillations is larger for Ga$_{0.01}$In$_{0.99}$As than Ga$_{0.64}$In$_{0.36}$As. For Ga$_{0.01}$In$_{0.99}$As, the oscillation minima are close to zero. For Ga$_{0.64}$In$_{0.36}$As, the oscillation minima are significantly larger than zero. Our measurements of the whole compositional range reveal that the oscillation amplitude shrinks with increasing Ga mole fraction $x$ whereas the oscillation minima rise with increasing Ga mole fraction $x$.

We attribute the origin of the oscillating part of the observed terahertz signal to optical rectification of the incident femtosecond near-infrared laser pulses at the Ga$_x$In$_{1-x}$As surface. Terahertz emission at the oscillation minima is explained by transient photocurrents. Our results demonstrate that terahertz generation in Ga$_x$In$_{1-x}$As is maximized for $x=0.1–0.3$.

Next, we discuss why terahertz generation in Ga$_x$In$_{1-x}$As due to optical rectification decreases with increasing Ga mole fraction $x$. The amplitude of a terahertz radiation pulse generated by optical rectification in a nonlinear optical crystal depends on the following parameters: (i) amount of optical laser radiation transmitted into the crystal, (ii) crystallographic orientation of the crystal, (iii) the nonlinear electric susceptibility of the crystal, and (iv) microcrystal grain size.

We have examined all these parameters for Ga$_x$In$_{1-x}$As. The amount of laser radiation transmitted into the crystal depends on the index of refraction. The indices of refraction of both InAs and GaAs at 800 nm differ only by $\sim 1\%$ (Table III). Consequently, the amount of laser radiation transmitted into the Ga$_x$In$_{1-x}$As crystals basically does not change as function of Ga mole fraction $x$. Therefore, it is ruled out that terahertz generation in Ga$_x$In$_{1-x}$As due to optical rectification decreases with increasing Ga mole fraction because the amount of laser radiation transmitted into the material decreases.

Our EBSD data demonstrate that the domains in the Ga$_x$In$_{1-x}$As crystals exhibit very similar crystallographic orientations versus the direction of incidence and polarization of the near-infrared laser beam. Therefore, the variations in terahertz generation from our Ga$_x$In$_{1-x}$As crystals as function of Ga mole fraction $x$ cannot be explained by a change of the crystallographic orientation of the crystal.

![FIG. 4. Orientation contrast image of W1. The sample exhibits large area domains with good crystalline quality.](image)

![FIG. 5. Time-domain measurements of terahertz radiation pulses emitted by Ga$_x$In$_{1-x}$As.](image)

![FIG. 6. Frequency spectra of radiation pulses emitted by Ga$_x$In$_{1-x}$As.](image)

![FIG. 7. For this experiment, the Ga$_x$In$_{1-x}$As sample was rotated with the surface normal as axis of rotation. The terahertz emission from Ga$_x$In$_{1-x}$As was measured in reflection as a function of azimuth angle $\phi$. The terahertz signal oscillates as a function of the angle $\Phi$.](image)
Terahertz radiation generated by optical rectification in InAs and GaAs has been investigated experimentally previously. Terahertz generation in InAs was attributed to surface-field induced optical rectification whereas terahertz generation in GaAs was explained by bulk optical rectification. The magnitude of terahertz generation due to surface-field induced optical rectification is determined by the third order nonlinear electric susceptibility of the material and terahertz generation due to bulk optical rectification depends on the second-order nonlinear electric susceptibility. The explanation of strong terahertz emission from InAs by surface-field optical rectification was previously supported by the argument that the third order susceptibility of a narrow band-gap semiconductor such as InAs is several orders of magnitude larger than the third order susceptibility of a wider band-gap semiconductor such as GaAs is 1. $\times$ $10^{-10}$ esu. Based on the magnitudes of $\chi^{(2)}$ and $\chi^{(3)}$ for GaAs and InAs at 800 nm, we explain terahertz generation due to optical rectification in Ga$_{1-x}$In$_x$As to be dominated by the $\chi^{(2)}$ nonlinear optical process. Since $\chi^{(2)}$ of InAs and GaAs have approximately the same magnitude $\chi^{(2)}$ $\approx$ $10^{-6}$ esu, we expect that the amplitude of terahertz radiation emitted by In-rich Ga$_{1-x}$In$_x$As samples is the same order of magnitude than terahertz radiation emitted by Ga-rich Ga$_{1-x}$In$_x$As samples. This expectation is in agreement with our experimental observations.

Next, we discuss the influence of crystal size on terahertz generation by optical rectification. The efficacy of a nonlinear optical process depends on the coherence length.\textsuperscript{51} If the refractive indices of the near-infrared laser excitation frequency and the terahertz radiation are identical, then the strength of terahertz emission would increase similarly for all frequencies within the bandwidth with increasing crystal thickness. However, the refractive indices for the near-infrared laser frequency and terahertz frequency are generally not identical in Ga$_{1-x}$In$_x$As (Table III). Consequently, electromagnetic waves at terahertz- and near-infrared-frequencies travel at slightly different speeds through the crystal. The efficacy of nonlinear optical terahertz generation decreases if the mismatch between the velocities becomes too large. The distance over which the slight velocity mismatch can be tolerated is the coherence length. As a result, the efficacy of terahertz generation due to optical rectification will decrease if the crystal thickness is less than the coherence length.

We have measured the domain size $A$ of our Ga$_{1-x}$In$_x$As crystals (Table II). We define $l=A^{1/2}$ as a characteristic length of the domain. We observe that $l$ decreases with increasing Ga mole fraction 0.01 $\leq x \leq$ 0.64. We have plotted the terahertz signal, due to optical rectification, as a function of characteristic domain size $l$ (Fig. 9) and observe a positive linear correlation between terahertz signal due to optical rectification and characteristic domain size $l$. We have performed a linear regression analysis of the data displayed in Fig. 9 and found a coefficient of correlation $r=0.96$. The decrease of terahertz emission due to optical rectification is explained by a reduced efficacy of the nonlinear optical process. The efficacy of optical rectification decreases in Ga-rich Ga$_{1-x}$In$_x$As because of a smaller polycrystal grain size.

![Figure 8](image_url)

**TABLE III.** Optical properties of InAs and GaAs

<table>
<thead>
<tr>
<th>Optical properties</th>
<th>InAs</th>
<th>GaAs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Index of refraction at 800 nm $n_{800}$ $^48$</td>
<td>3.729</td>
<td>3.679</td>
</tr>
<tr>
<td>Index of refraction at 1 THz $n_{THz}$ $^48$</td>
<td>3.778</td>
<td>3.60</td>
</tr>
<tr>
<td>Second order susceptibility $</td>
<td>\chi^{(2)}</td>
<td>$ at 800 nm (esu)$^{49}$</td>
</tr>
<tr>
<td>Third order susceptibility $</td>
<td>\chi^{(3)}</td>
<td>$ at 800 nm (esu)$^{50}$</td>
</tr>
</tbody>
</table>
Terahertz generation proceeds by optical rectification or by the transport of photoexcited free carriers. The current surge induced by photoexcitation has two sources: (1) the acceleration of photoexcited carriers by the surface depletion field caused by Fermi-level pinning, and (2) the photo-Dember effect originating from the difference between the diffusion constants of electrons and holes. The origin of the surface-field effect is band bending due to Fermi-level pinning by surface states. A charge distribution at the surface forms a built-in electric field directed normal to the surface. When an ultrafast laser pulse illuminates a semiconductor surface with photon energy greater than the band gap of the material, electron-hole pairs are created at the semiconductor surface. The photogenerated carriers are accelerated by the built-in electric field to form a transient photocurrent which in turn generates terahertz radiation. The other mechanism known as photo-Dember effect originates from the charge-carrier density gradient caused by the photogenerated carriers near the surface which drives diffusion of electrons and holes into the material. A charge-carrier density gradient is formed due to the higher mobility of electrons compared to the mobility of holes. This diffusion current in turn generates terahertz radiation. In a narrow band-gap III–V compound semiconductors such as InAs, the origin of current-surge induced terahertz emission is the photo-Dember effect whereas in wide band-gap semiconductor such as GaAs, the surface-field acceleration effect is the responsible physical mechanism.

We have calculated for Ga<sub>1−x</sub>In<sub>x</sub>As as a function of Ga mole fraction <i>x</i>. The calculations are performed according to Eq. (1) (photo-Dember effect) and Eq. (2) (surface-field acceleration).

\[ E_{\text{far}} = \frac{S}{4\pi e c^2 \tau} \left[ \frac{b(n_b - p_b)}{1 + b} \ln \left( \frac{G(1 + b)}{p_b + b n_b} \right) \right] \]  

*(1)*

\[ E_{\text{far}} = \frac{Se^2 \mu_b G|N_p^0 - N_n^0|}{4\pi e c^2 \tau c \kappa c} |W\alpha - 1 + e^{-\alpha W}| \]  

*(2)*

As derived previously, the magnitude of terahertz radiation emitted due to the photo-Dember effect [Eq. (1)] depends on the electron and hole concentrations <i>n_b</i> and <i>p_b</i>, the ratio of electron and hole mobilities \(b = \mu_e / \mu_h\) and the temperature T<sub>e</sub>

The terahertz generation proceeds by optical rectification or by the transport of photoexcited free carriers. The current surge induced by photoexcitation has two sources: (1) the acceleration of photoexcited carriers by the surface depletion field caused by Fermi-level pinning, and (2) the photo-Dember effect originating from the difference between the diffusion constants of electrons and holes. The origin of the surface-field effect is band bending due to Fermi-level pinning by surface states. A charge distribution at the surface forms a built-in electric field directed normal to the surface. When an ultrafast laser pulse illuminates a semiconductor surface with photon energy greater than the band gap of the material, electron-hole pairs are created at the semiconductor surface. The photogenerated carriers are accelerated by the built-in electric field to form a transient photocurrent which in turn generates terahertz radiation. The other mechanism known as photo-Dember effect originates from the charge-carrier density gradient caused by the photogenerated carriers near the surface which drives diffusion of electrons and holes into the material. A charge-carrier density gradient is formed due to the higher mobility of electrons compared to the mobility of holes. This diffusion current in turn generates terahertz radiation. In a narrow band-gap III–V compound semiconductors such as InAs, the origin of current-surge induced terahertz emission is the photo-Dember effect whereas in wide band-gap semiconductor such as GaAs, the surface-field acceleration effect is the responsible physical mechanism.

We have calculated for Ga<sub>1−x</sub>In<sub>x</sub>As as a function of Ga mole fraction <i>x</i>. The calculations are performed according to Eq. (1) (photo-Dember effect) and Eq. (2) (surface-field acceleration).

\[ E_{\text{far}} = \frac{S}{4\pi e c^2 \tau} \left[ \frac{b(n_b - p_b)}{1 + b} \ln \left( \frac{G(1 + b)}{p_b + b n_b} \right) \right] \]  

*(1)*

\[ E_{\text{far}} = \frac{Se^2 \mu_b G|N_p^0 - N_n^0|}{4\pi e c^2 \tau c \kappa c} |W\alpha - 1 + e^{-\alpha W}| \]  

*(2)*

As derived previously, the magnitude of terahertz radiation emitted due to the photo-Dember effect [Eq. (1)] depends on the electron and hole concentrations <i>n_b</i> and <i>p_b</i>, the ratio of electron and hole mobilities \(b = \mu_e / \mu_h\) and the temperature T<sub>e</sub>.

The magnitude of terahertz radiation emitted due to surface-field acceleration [Eq. (2)] depends on depletion width \(W = (2\varepsilon_0\varepsilon_0\Phi_p / \rho)^{1/2}\), surface potential \(\Phi_p\), charge in the depletion region \(\rho = q(p - n + N_p^0 - N_n^0) = q(N_p^0 - N_n^0)\), donor and acceptor concentrations and absorption depth \(\alpha\). Furthermore, in Eqs. (1) and (2), \(S\) is the laser focal spot size, \(\tau\) is the laser-pulse duration, \(r\) is the distance in the far field, and \(G\) is the photocarrier generation rate. Also, in Eqs. (1) and (2), \(s_0 = 8.85 \times 10^{-12}\) As/Vm, \(c = 3.00 \times 10^8\) m/s and \(\varepsilon_r\) is the permittivity of the semiconductor.

The terahertz emission due to Ga<sub>1−x</sub>In<sub>x</sub>As as a function of Ga mole fraction <i>x</i> is calculated using the intrinsic parameters \(\mu_e(x), \mu_h(x), n_b(x), p_b(x), k_bT(x) = 1.55\) eV \(-E_g(x) = (1.19 + 0.63x + 0.43x^2)\) eV and \(G(x) = (2.45 + 30(1 - x)) \times 10^8\) cm<sup>−3</sup> in Eq. (1). We justify using intrinsic parameters by the observation that our measured electron carrier concentrations and electron mobilities exhibit the same functional dependence on Ga mole fraction <i>x</i> as the intrinsic properties. (Fig. 3)

The terahertz emission due to the photo-Dember mechanism and surface-field acceleration for Ga<sub>1−x</sub>In<sub>x</sub>As is illustrated in Fig. 10 as a function of the Ga mole fraction <i>x</i>. Our calculations demonstrate that terahertz emission is expected to be dominated by the photo-Dember effect for \(x \leq 0.2\) whereas terahertz emission due to surface-field acceleration is insignificant in this compositional range. Terahertz emission due to the Photo-Dember effect becomes negligible for \(x \geq 0.2\). Terahertz emission due to surface-field acceleration increases for \(x \geq 0.2\) and reaches a maximum at \(x = 0.45\).

The photo-Dember effect dominates terahertz-emission for \(0 < x < 0.2\) because the surface depletion field is negligible for these alloy compositions in Ga<sub>1−x</sub>In<sub>x</sub>As. Surface Fermi-level pinning in III–V compound semiconductors is caused by surface states associated with defects present at the surface of the semiconductor. The dominant types of surface state for most semiconductors are acceptors located within the band gap [71]. However, surface defect states in
InAs are donors and are located above the minimum of the conduction-band edge.\textsuperscript{53} This leads to an electron accumulation layer at the surface of InAs. The sheet electron concentration of the accumulation layer is typically about $1 \times 10^{12}$ cm\textsuperscript{-2} and is unaffected by bulk doping of InAs.\textsuperscript{53} Fermi-level pinning in InAs occurs for any InAs surface orientation. The energy difference between the conduction-band minimum and the Fermi-level energy in Ga\textsubscript{In}\textsubscript{1-x}As can be expressed as $E_F = (0.43x^2 + 0.502x - 0.10)^2$ eV. According to this equation, the Fermi-level reaches the minimum of the conduction band in Ga\textsubscript{In}\textsubscript{1-x}As for $x \approx 0.2$. The corresponding surface field $E_s = \rho(W + z)/(e_0\epsilon_0)$ and depletion width $W$ are calculated for Ga\textsubscript{In}\textsubscript{1-x}As as a function of Ga mole fraction $x$ and illustrated in Fig. 11. Again, our calculations demonstrate that surface depletion field is negligible for $0 < x < 0.2$. The surface field exhibits a maximum at $x \approx 0.35$. The depletion width $W$ monotonically increases with Ga mole fraction $x$ for $x > 0.2$. The terahertz emission due to surface-field acceleration depends on the integral over the photocurrent $E_{ph} \approx \int_{0}^{x} j(z) dz$ with $j(z) \approx G(z)E_s(z)$. This relationship results in a maximum of terahertz emission at $x \approx 0.45$. We explain that terahertz emission from Ga\textsubscript{In}\textsubscript{1-x}As is due to ultrafast currents being dominated by surface-field acceleration for $x > 0.2$.

We do not observe a strong contribution to terahertz emission from ultrafast currents in Ga\textsubscript{0.01}In\textsubscript{0.99}As. For this composition, terahertz emission appears to be entirely due to optical rectification. This is a surprising observation considering the results of previous measurements of terahertz emission from InAs. In experiments performed on (100) oriented InAs crystals, the terahertz emission was found to be due to the photo-Dember effect. This is in agreement with predictions by nonlinear optics which rule out optical rectification for this orientation. In previous experiments on (110) and (111) oriented InAs crystals the observation was made that terahertz emission is due to optical rectification as well as ultrafast currents. However, the terahertz generation due to optical rectification was generally weaker than terahertz generation due to ultrafast currents. In addition, optical rectification was generally observed for excitation by amplified titanium-sapphire laser pulses which provide much stronger optical electric fields than our titanium-sapphire oscillator. Our observation suggests that the mechanism and magnitude of terahertz emission from InAs depends on the microstructure of the InAs crystal. The microstructure of InAs crystals is influenced by the crystal-growth process. We suggest that further research on terahertz emission from InAs should focus on the effect of crystal growth and microstructure of the material on terahertz emission.

IV. CONCLUSIONS

We have studied femtosecond optically excited emission of terahertz radiation from bulk Ga\textsubscript{In}\textsubscript{1-x}As crystals over a large compositional range. We identified optical rectification of the incident femtosecond near-infrared laser pulses and acceleration of photocarriers by a surface depletion field as terahertz emission mechanisms in our Ga\textsubscript{In}\textsubscript{1-x}As crystals. Terahertz emission from In-rich Ga\textsubscript{In}\textsubscript{1-x}As is dominated by optical rectification of the femtosecond laser pulses. Terahertz emission from Ga-rich Ga\textsubscript{In}\textsubscript{1-x}As is primarily due to surface-field acceleration of photocarriers. The magnitude of the terahertz emission due to optical rectification increases with polycrystal grain size in our Ga\textsubscript{In}\textsubscript{1-x}As samples. According to our measurements and analysis, terahertz emission from In-rich as well as Ga-rich Ga\textsubscript{In}\textsubscript{1-x}As is due to a $\chi^{(2)}$ nonlinear optical process. We found that terahertz emission from Ga\textsubscript{0.01}In\textsubscript{0.99}As is caused entirely by optical rectification. Terahertz emission due to surface-field acceleration is maximized in Ga\textsubscript{In}\textsubscript{1-x}As with $x \approx 0.45$. This is explained by a maximum of the surface depletion field for this composition of Ga\textsubscript{In}\textsubscript{1-x}As. The overall terahertz emission from bulk Ga\textsubscript{In}\textsubscript{1-x}As crystals is the strongest in the $x \approx 0.1–0.3$ compositional range.

ACKNOWLEDGMENTS

We would like to thank E. B. Watson and J. Thomas for suggesting EBSD for structural analysis of our samples. Furthermore, we acknowledge technical support of EBSD measurements by Zhenting Jiang at Yale University. This material is based upon work supported by the National Science Foundation under Grant No. 0333314.

\textsuperscript{5}J. N. Heyman, P. Neocleous, D. Hebert, P. A. Crowell, T. Muller,
KO et al.

PHYSICAL REVIEW B 78, 035201 (2008)