Terahertz-wave generation in quasi-phase-matched GaAs

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The authors demonstrate an efficient room temperature source of terahertz radiation using femtosecond laser pulses as a pump and GaAs structures with periodically inverted crystalline orientation, such as diffusion-bonded stacked GaAs and epitaxially grown orientation-patterned GaAs, as a nonlinear optical medium. By changing the GaAs orientation-reversal period (504–1277 μm), or the pump wavelength (2–4.4 μm), we were able to generate narrow-bandwidth (~100 GHz) terahertz wave packets, tunable between 0.9 and 3 THz, with the optical-to-terahertz photon conversion efficiency of 3.3%. © 2006 American Institute of Physics.

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In the optical rectification (OR) process, terahertz output is produced in an electro-optic medium via self-difference-frequency mixing between Fourier components of the optical pulse. First demonstrated with picosecond pulses in LiNbO₃,¹,² ZnTe, ZnSe, CdS, and quartz' crystals, this technique was later extended to femtosecond pulses³ and became an established way of generating single-cycle broadband terahertz radiation. Typically, thin (~1 mm or less) electro-optic crystals are used because of phase matching constraints as well as high absorption in conventional crystals (LiNbO₃ and ZnTe) at terahertz frequencies. Optical-to-terahertz conversion efficiencies achieved so far by optical rectification methods are low, typically 10⁻₆–10⁻⁹.⁴ Recently, conversion efficiency of 5 × 10⁻⁴ was reported.⁵ The authors used optical rectification in LiNbO₃, in a noncollinear geometry, with 150 fs 300–500 μJ optical pump pulses at 800 nm with a tilted intensity front. Lee et al.⁶ demonstrated that in a quasi-phase-matched (QPM) structure, the interaction length between terahertz and optical pulses can be dramatically extended; terahertz output in this case is produced in the form of narrow-band wave packets. With 200 fs pulses at 800 nm and periodically poled lithium niobate crystal, cryogenically cooled (18 K) to reduce terahertz absorption, the authors achieved 10⁻⁵ conversion efficiency.⁷

Zinc blende semiconductors (GaAs and GaP) show great potential for QPM terahertz generation because of their small, as compared to lithium niobate, terahertz absorption coefficient αₜₙ₉ [in GaAs, for example, αₜₙ₉ = 0.5–4.5 cm⁻¹ at 1–3 THz (Ref. 8)], small mismatch between the optical group and terahertz phase velocities, and high thermal conductivity. Terahertz-wave generation was demonstrated recently in QPM GaAs using optical rectification of femtosecond pulses⁸ and in QPM GaP using difference frequency generation (DFG) with nanosecond pulses.⁹ In this letter, we report on an efficient widely tunable source of terahertz radiation based on OR in quasi-phase-matched GaAs structures.

In our experiment, tunable pump pulses centered at 2–4.4 μm were produced using a parametric amplifier (OPera, Coherent Inc.) system pumped by 800 nm Ti:sapphire pulses after a regenerative amplifier (Legend, Coherent Inc.). To achieve λ > 3 μm wavelengths, an additional difference frequency generation stage was used. Typical pulse durations were ~100 fs, repetition rate 1 kHz, and pulse energy up to 3 μJ. The >2 μm pump wavelength range was chosen to avoid two-photon absorption (2PA) in GaAs (2PA edge at 1.74 μm), which creates additional losses both at pump wavelength and at terahertz frequencies (in the latter case because of induced absorption due to generated free carriers).

We have used two types of QPM GaAs samples: (i) diffusion-bonded GaAs (DB-GaAs) produced by stacking and bonding together alternately rotated (110) GaAs plates,¹⁰ wafer fusion in this case creates a monolithic body with periodic change in the nonlinear coefficient, and (ii) orientation-patterned GaAs (OP-GaAs) grown by a combination of molecular beam epitaxy (MBE), photolithography, and hydride vapor phase epitaxy (HVPE),¹² where periodic inversions of the crystallographic orientation are grown into the material. While DB-GaAs provides larger apertures, OP-GaAs has more reproducible technology and allows lithographic definition of QPM gratings. The samples were not anti-reflection coated and their main parameters are listed in Table I.

The optical pump beam with a beam size (1/e² intensity radius) ranging between w = 300 μm and 1.5 mm propagated along the (110) direction of GaAs. A Picarin lens was used to collect the terahertz radiation to the liquid-He-cooled silicon...
TABLE I. Parameters of the QPM GaAs samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>QPM type</th>
<th>Aperture (mm²)</th>
<th>Length (mm)</th>
<th>QPM period (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DB-77</td>
<td>DB-GaAs</td>
<td>10 × 10</td>
<td>6</td>
<td>504</td>
</tr>
<tr>
<td>A3</td>
<td>OP-GaAs</td>
<td>0.4 × 3</td>
<td>3</td>
<td>1277</td>
</tr>
<tr>
<td>A10</td>
<td>OP-GaAs</td>
<td>0.4 × 3</td>
<td>10</td>
<td>1277</td>
</tr>
<tr>
<td>B5</td>
<td>OP-GaAs</td>
<td>0.4 × 3</td>
<td>5</td>
<td>759</td>
</tr>
<tr>
<td>B10</td>
<td>OP-GaAs</td>
<td>0.4 × 3</td>
<td>10</td>
<td>759</td>
</tr>
<tr>
<td>C5</td>
<td>OP-GaAs</td>
<td>0.4 × 3</td>
<td>5</td>
<td>564</td>
</tr>
</tbody>
</table>

bolometer which measured the average power of terahertz pulses. A black polyethylene filter was used to block optical radiation. To measure the spectral properties of terahertz radiation (Fig. 1), we have used a Michelson interferometer, composed of two 2-in.-diameter flat gold mirrors and a 25-µm-thick Mylar beam splitter.

Theoretically, the frequency of terahertz radiation produced by the QPM optical rectification is centered\(^1\) at \(\nu_{\text{THz}} = c/\Delta n\), corresponding to the zero wave-vector mismatch condition; \(c\) is the speed of light, \(\Delta n = n_{\text{THz}} - n_{\text{opt}}\) is the mismatch between the terahertz refractive index and the optical group refractive index, and \(\Lambda\) is the QPM period. For GaAs, \(n_{\text{THz}}\) is nearly constant\(^2\) \(\approx 3.6\) at frequencies well below the lowest phonon resonance\(^3\) (8.1 THz) and \(n_{\text{opt}}\) varies\(^4\) between 3.43 (\(\lambda = 2\) µm) and 3.33 (4.4 µm). The spectral width of terahertz wave packets is determined by the QPM acceptance bandwidth \(\Delta \nu_{\text{THz}} = c/L\Delta n\), where \(L\) is the length of the crystal.

The power spectra of terahertz pulses were extracted in our experiment by computing the amplitudes of the Fourier transforms of Michelson interferograms. Figure 2 shows both original interferograms and computed spectra for different samples and different pump wavelengths. The spectra were noticeably distorted by water vapor absorption (also shown in Fig. 2). Interestingly, for sample A10 with the largest QPM period, 1277 µm, we observed [Fig. 2(d)] a second peak at \(\sim 2.6\) THz which is likely to be the third order QPM peak, at approximately three times the frequency of the main peak. The amplitudes of both peaks are comparable: efficiency reduction (1/3\(^2\)) due to the third order QPM is offset by the \(\nu_{\text{THz}}^2\) factor which appears in the expression for the efficiency of any DFG-like process.\(^5\)

Experimentally observed central frequencies and bandwidths of terahertz pulses are in good agreement with theoretical predictions based on known GaAs dispersion relations.\(^6\) By changing the pump wavelength or GaAs QPM period, we generated terahertz wave packets with central frequencies between 0.9 and 3 THz. The tuning curve for the 4.4 µm optical pump is shown in Fig. 3.

We found that the terahertz beam propagated collinearly with respect to the optical pump and was close to diffraction limited (a pinhole method was used to measure the far-field terahertz beam size). Figure 4 shows optical-to-terahertz conversion efficiency \(\eta_{\text{THz}}\) as a function of peak pump intensity \(I_0\) inside the sample (DB-77, pump at \(\lambda = 3.5–4.4\) µm). The pump beam size varied in this case between 810 µm (open circles), 520 µm (closed circles), and 300 µm (crossed circles). One can see that the linear dependence of \(\eta_{\text{THz}}\) expected by theory, rolls off for \(I_0 > 2\) GW/cm\(^2\). This roll-off behavior was also observed at similar intensities at shorter, \(\lambda \approx 2\) µm pump. The onset of saturation is most likely due to nonlinear refraction (\(n_2\)) in GaAs which induces self-phase modulation and self-focusing of the optical pulses. Indeed, we have measured \(n_2^2 = 1.5 \times 10^{-3}\) cm\(^2\)/GW for GaAs at 3.5 µm and estimated that at \(I_0 = 2\) GW/cm\(^2\) and \(L(\text{GaAs}) = 6\) mm, the nonlinear phase shift at beam center reaches \(\sim \pi\).

Terahertz conversion efficiencies for OP-GaAs samples A3 (also shown on Fig. 4) and B5 are very similar to each other and are smaller, by a factor of \(\sim 3.5\), than that of the DB-77 sample (in both cases the pump was at 3.5 µm, cen-
efficiency of 2.9% which corresponds to optical-to-terahertz conversion efficiency of 1.1% based on the calculated values, based on the known nonlinear optical coefficient for optical rectification $d_{33} = 2/\pi \times d_{14} = 2/\pi \times 47 \text{ pm/V}$, derived from the electro-optical coefficient $r_{14}(\text{GaAs}) = 1.5 \text{ pm/V}$.

In the DB-77 sample, we generated 0.66 nJ of output at 2.2 THz, with 2.3 \mu J of pump pulse energy ($w=300 \mu m$), which corresponds to optical-to-terahertz conversion efficiency of $2.9 \times 10^{-4}$, internal efficiency of $8.7 \times 10^{-4}$ (the samples were uncoated), and photon conversion efficiency of 1.1% (internal photon efficiency of 3.3%). These efficiencies are in accord with the calculated values, based on the known nonlinear optical coefficient for optical rectification $d_{33} = 2/\pi \times d_{14} = 2/\pi \times 47 \text{ pm/V}$, derived from the electro-optical coefficient $r_{14}(\text{GaAs}) = 1.5 \text{ pm/V}$.

In conclusion, we demonstrated efficient terahertz generation with 3.3% internal photon efficiency and with only 2.3 \mu J of pump pulse energy. The limit for efficiency was set by parasitic $\chi^{(3)}$-effects that can be avoided by using DFG with longer (picosecond) pulses. Thus, periodic GaAs structures show promise for optical terahertz generation in terms of robustness, wide tuning range, high conversion efficiency, and potential for scalability of the terahertz output power. Such source of terahertz radiation might be suitable for many applications including terahertz imaging and spectroscopy.

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