Resonant Nonlinear Susceptibility near the GaAs Band Gap

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We report new results on subpicosecond optical rectification near the semiconductor GaAs band gap at room temperature. We discriminate between radiation produced by real carriers and that produced by optical rectification by using normal and off-normal incident optical illumination. Under normal incident optical illumination of the ⟨111⟩ GaAs surface, the amplitude of the nonlinear susceptibility varies dramatically (2 orders of magnitude) and the sign of the susceptibility reverses when the photon energy is tuned above the band gap. This resonance enhancement and the sign reversal are explained by the dispersion of the second-order nonlinear susceptibility at the band gap.

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When illuminated by femtosecond laser pulses at an oblique incident angle, a GaAs sample emits subpicosecond submillimeter-wave radiation [1]. This phenomenon was interpreted as radiation from accelerated photocarriers in the surface depletion field [1] and the dependence of the radiation on crystal rotation symmetry was explained as strong evidence of $\chi^{(3)}$ related optical rectification [2]. More recently, quantum well structures have been used to limit the carrier transport. This allows examination of the mechanism of optical rectification as well as the study of electron coherent oscillations in a double-well potential [3]. To distinguish between the contribution from carrier transport and that from optical rectification is crucial for a physical understanding of the origin of the radiation, as well as for realizing an intense THz source application. In this Letter we report new measurements of terahertz radiation from bulk GaAs and its superlattice structures. Our results clearly demonstrate, for the first time, the contribution from both the subpicosecond optical rectification (SOR) and the photocarrier transport effect. When the SOR was separated from the background radiation from carrier transport and the semiconductor was excited at resonance, we observed an enhancement of the submillimeter-wave nonlinear susceptibility. We also observed that when the laser energy was tuned above the band gap the change of the polarity of the radiated electromagnetic field was due to the resonant behavior of the subpicosecond nonlinear susceptibility.

The experimental setup was a time-resolved, optoelectronic, coherent sampling arrangement and it has been described elsewhere [3]. A cw Ar laser-pumped, mode-locked Ti:sapphire laser (Coherent MIRA) was used as the source of optical pulses. The MIRA laser produced an output pulse energy greater than 10 nJ at a repetition rate of 76 MHz with a pulse duration of less than 200 fs. The energy spectrum of the laser beam (full width at half maximum) is about 14 meV at a wavelength of about 820 nm. The laser beam was split into two parts by a beam splitter with a 0.05/0.95 reflection/transmission ratio. The stronger optical beam passed through a variable time delay stage and illuminated the semiconductor sample with a spot diameter of approximately 6 mm. The weaker optical beam, typically < 20 mW, was used for optical gating at a photoconducting detector. The submillimeter-wave beam, radiated in the forward direction, was focused on a photoconductor attached to a 50-μm dipole antenna. This dipole antenna is a submillimeter-wave polarization-sensitive detector. A photocurrent was developed at the antenna when the submillimeter-wave radiation was spatially and temporally overlapped with the optical gating pulse. The temporal measurements were achieved by varying the time delay between the excitation laser pulse illuminating the crystal (strong optical beam) and the trigger laser pulse focused on the detector (weak optical beam). The measurements were taken at room temperature.

Normal optical incidence on the GaAs surface was used to distinguish the subpicosecond optical rectification signal from the carrier transport effect. Under normal incidence the THz signal from the accelerated carriers driven by the surface depletion field cannot be measured in the forward direction because no radiation is emitted along the current flow direction. However, subpicosecond optical rectification can radiate in the forward direction under normal incident illumination. Figure 1 shows the THz radiation (in the forward direction) from a ⟨111⟩ GaAs bulk sample for three incident angles ($\phi = 0°$ and

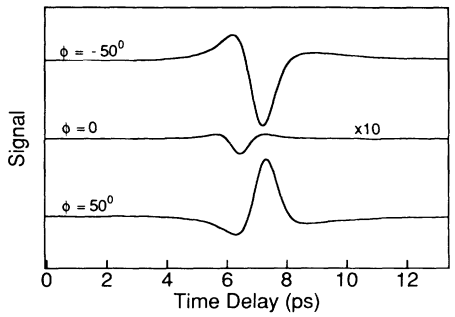

FIG. 1. THz radiation from a ⟨111⟩-oriented GaAs with normal (center trace) and $\pm 50°$ (lower and upper traces) optical incident angles. Temporal broadening of the THz radiation wave form is pronounced with the oblique optical beams.

$\pm 50°$). The photon energy is 1.52 eV. The center trace ($\phi = 0$) is due to subpicosecond optical rectification only, while the upper and lower traces ($\phi = \pm 50$) combine radiation from both the SOR and carrier transport effects. The timing shifts of the wave forms taken at oblique angles are due to an increase in the THz beam path from tilting the sample. Since the carrier transport process may last much longer than the polarization dephasing time, this relatively slow feature should be reflected in time-resolved measurements. The temporal broadening of the THz signal due to the transient photocurrent can be well resolved when oblique optical incident angles are used. We also investigated the dependence of the temporal broadening on photocarrier density (with the photocarrier densities ranging from $2.2\times10^{13}/\text{cm}^3$ to $4.4\times10^{16}/\text{cm}^3$), but did not observe any change in the temporal broadening within our temporal resolution. However, at higher optical fluence, a wave form change is expected, due to transition saturation, signal absorption, and the screening of the static electric field. In principle, if the optical pulse duration is longer (or shorter) than the polarization dephasing time the duration of the SOR should be close to the optical pulse duration (or polarization dephasing time) since the SOR is proportional to the negative second time derivative of the nonlinear polarization. It is possible to observe the polarization dephasing time if a much shorter optical pulse is used. Because of the finite bandwidth of the detector we currently cannot resolve the actual wave form of the SOR.

The SOR signal changes dramatically when the laser energy is tuned near the GaAs band gap. The rectified signal is nearly unchanged for small photon energy tuning around 1.35 eV (70 meV lower than the band gap). When the photon energy is tuned to the band gap the amplitude of the SOR signal increases, reaching a peak when the laser energy is just above the band gap. Tuning the laser energy further above the band gap results in a polarity change of the radiation signal. Figure 2(a) is a plot of the SOR signal from a ⟨111⟩ GaAs sample versus photon energy (normal incident angle). The energy resolution is limited by the laser spectrum which is about 14 meV. The resonance enhancement near the band gap is pronounced. We measured about a 106-fold increase in the rectified field when the laser energy was tuned from 1.40 to 1.46 eV. Figure 2(b) contains two temporal wave forms at photon energies of 1.43 and 1.49 eV, clearly demonstrating the polarity change of the rectified field. During laser energy tuning the temporal duration of the rectified field remained essentially unchanged. Resonance enhancement of the SOR signal from a ⟨100⟩ GaAs sample with off-normal optical illumination was observed previously, and the temporal wave form flip-over was explained as evidence of virtual photoconductivity [4]. However, the virtual-current model cannot generate forward radiation (longitudinal) under normal incidence. Our new result suggests that this polarity flip-over is due to a change in sign of the resonant susceptibility at the band gap.

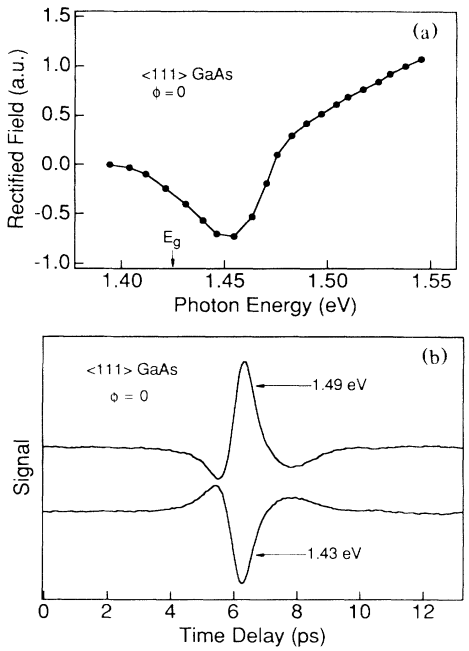

FIG. 2. (a) The peak value of the rectified signal vs laser energy at normal optical incident angle, from ⟨111⟩ GaAs. (b) Temporal wave forms of the radiated field at a photon energy of 1.49 eV (upper trace) and 1.43 eV (lower trace), respectively.

We measured the dependence of the SOR signal on crystallographic orientation. In this measurement a ⟨111⟩-oriented GaAs sample was rotated around its surface normal. Figures 3(a) and 3(b) show the SOR signal versus crystal azimuth angle at two optical energies (1.43 and 1.53 eV) and two optical polarizations (incident light polarized parallel and perpendicular to the detector axis), respectively. The solid and open circles are the experimental data. The SOR signal has opposite polarity at these two energies. A threefold rotation symmetry was observed at both photon energies. Since the submillimeter-wave radiation is proportional to the negative second time derivative of the low-frequency polarization, we can calculate the angular dependence of the nonlinear polarization (low frequency) to obtain information about the rectification. The optical rectification from a ⟨111⟩B-oriented GaAs under a normal incident angle can be calculated from the nonlinear polarizations P_p and P_s corresponding to different incident optical polarization geometries (light polarization parallel and perpendicular to the dipole detector axis, respectively). The nonlinear polarizations P_p and P_s (SI units) are given by

$$P_p = -P_s = (4/\sqrt{6})d_{14}\cos(3\theta)EE^*, \quad (1)$$

where the azimuthal angle θ is measured with respect to the $[11\bar{2}]$ direction and d_{14} is a nonlinear optical susceptibility tensor element having a frequency response from dc to several terahertz (mainly characterized by the optical pulse duration) [5]. Figures 3(a) and 3(b) also show the

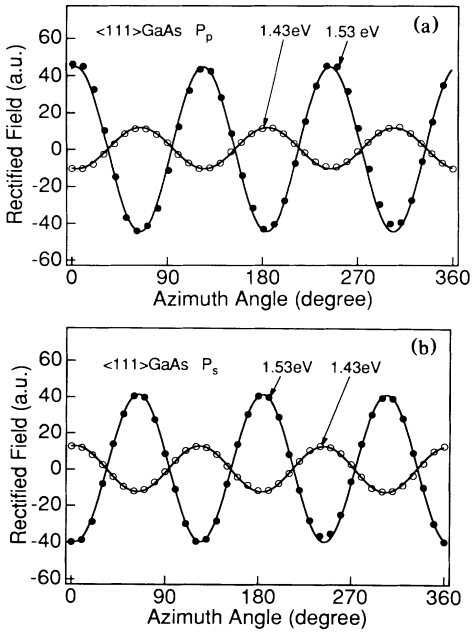

FIG. 3. Rectified field vs azimuth angle at two optical polarizations (a) parallel and (b) perpendicular to the detector axis. The open and solid circles are the experimental data at two photon energies (1.43 and 1.53 eV, respectively). The curves are plots of the rectified field calculated from the nonlinear polarizations (a) P_p and (b) P_s (the sign is reversed).

reversed rectification fields calculated from the nonlinear polarizations P_p and P_s, respectively (curves are fitted to the experimental data). The plots of the calculation from Eq. (1) fit the experimental data at 1.43 eV well. However, to fit the experimental data at 1.53 eV the sign of the susceptibility needs to be changed. In Fig. 2 the rectification signal shows a sign reversal at the band gap. After we changed the sign of d_{14} the data at 1.53 eV fit well with the calculations. We also calculated the nonlinear polarization from a ⟨110⟩-oriented GaAs. The calculation showed no SOR signal in the forward direction under normal incidence, regardless of the azimuthal rotation of the crystal. This calculated result has been verified in our experiments.

Besides the use of normal incident illumination of the bulk GaAs to confirm the subpicosecond optical rectification, we also examined the THz radiation from superlattice structures. The use of quantum barriers to limit the carrier transport leads to the observation of coherent submillimeter-wave emission from charge oscillations in a double-well potential [3]. Here we measured the THz radiation from a GaAs/$In_{0.1}Ga_{0.9}As$ ⟨111⟩-oriented superlattice with photon energies below and above the barrier potential. The GaAs/InGaAs sample has fifteen periods of quantum wells with a barrier width of 140 Å (GaAs) and a well width of 70 Å ($In_{0.1}Ga_{0.9}As$). We selected two laser energies (below and above the GaAs barrier potential) to induce THz radiation.

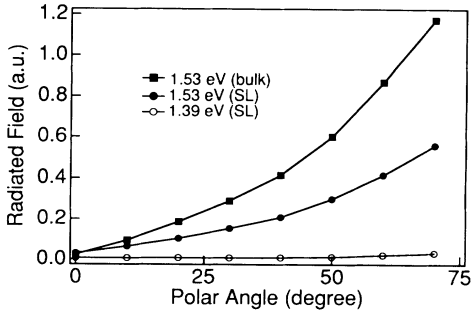

FIG. 4. Rectification signal vs optical incident angle (polar angle) from a ⟨111⟩-oriented GaAs bulk and GaAs/$In_{0.1}Ga_{0.9}As$ superlattice at three photon energies.

Figure 4 shows the THz signal versus optical incident angle (polar angle) from ⟨111⟩-oriented GaAs bulk and superlattice samples. When the photon energy (1.39 eV) is less than the barrier potential of GaAs, the radiation signal increases slowly with increasing polar angle (about 4 times). In this case the carriers are injected directly into the wells and quantum confinement will limit the photocurrent and the radiation is mainly from optical rectification. When the photon energy (1.53 eV) is above the barrier potential, the THz signal increases quickly with increasing polar angle (over 20 times). The "free" photocarriers (above the barrier) can accelerate in the static field to radiate a THz signal. We have measured the temporal wave forms at two energies. At the same oblique angle a higher-energy (1.53 eV) excitation leads to temporal broadening of the THz signal, while a much cleaner and shorter THz wave form is measured when a lower-energy (1.39 eV) excitation is used. When the superlattice is replaced by a GaAs sample (superlattice substrate), the THz signal is nearly doubled at a photon energy of 1.53 eV, but the same dependence on polar angle is observed. The rectified signal, even at resonance, is smaller than the contribution from the photocurrent (off-normal incident angle). For example, the ratio of the THz radiation, under oblique angle illumination (near the Brewster angle), to that at normal incidence, from a set of ten different ⟨111⟩ GaAs samples, ranges from 20 to 40. Similar to Fig. 2(a), we have measured the peak value of the radiation signal versus laser energy at normal optical incident angle from this GaAs/InGaAs superlattice sample. In addition to the resonance feature at 1.45 eV from the GaAs barrier, we also observed another resonance at 1.39 eV which is believed to be due to the confined state of the quantum wells. A detailed discussion of the superlattice-related results will be given in a subsequent paper.

The dispersion of the nonlinear susceptibility is more pronounced when the optical light enters the absorption region ($\hbar\omega \geq E_g$). The observation of amplitude enhancement and the sign reversal of the subpicosecond optical rectification is the signature of resonant behavior in

the nonlinear susceptibility. The sign reversal near the band gap can be understood directly through the classical anharmonic oscillator model of the optical rectification susceptibility, whose principal value changes sign at resonance [6]. Since the band states form a continuum for energies greater than E_g, the sign reversal must be at an energy greater than E_g, not at $\hbar\omega = E_g$ as in the case of a single resonance. In fact, we observed the reversal point at approximately 1.47 eV [Fig. 2(a)], about 50 meV above the band gap.

When the photon energy is below the band gap and the optical pulse duration is much longer than the crystal dielectric relaxation time, the nonlinear polarization process is purely electronic in origin. The rectified signal in this optically transparent region is comparable to submillimeter-wave generation by difference-frequency mixing [7]. When the photon energy is near or above the band gap, real photocarriers are involved in the enhancement of the nonlinear susceptibility [8]. One example is the observation of enhancement of the nonlinear susceptibility in second-harmonic generation when the optical beam enters the region of strong second-harmonic absorption [9]. Another example is the investigation of microwave nonlinear susceptibilities in acentric crystals. For some materials, due to the ionic contribution, the microwave susceptibility exceeds the largest values of the optical and electro-optic coefficients [10]. We used resonant excitation with a fundamental photon energy near the band gap and measured the dispersion of the nonlinear susceptibility in the coherent-transient regime where the optical pulse duration is comparable with the dephasing time.

In conclusion, we report the direct observation of subpicosecond optical rectification from GaAs bulk and quantum-well structures. The amplitude of the rectified signal varies dramatically and the sign of the rectified field reverses when the photon energy is tuned above the band gap. This behavior is due to the dispersion of the nonlinear susceptibility when the optical excitation is near the resonance.

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