**Terahertz optical rectification from \( \langle 110 \rangle \) zinc-blende crystals**

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We report the study of optically induced terahertz (THz) electromagnetic radiation from \( \langle 110 \rangle \) oriented zinc-blende crystals. This work extends our previous studies of \( \langle 100 \rangle \) and \( \langle 111 \rangle \) GaAs. Excellent agreement between calculated results and experimental data indicates that, under conditions of moderate optical fluence and normal incidence on the unbiased sample, second-order optical rectification is the major nonlinear process that generates THz radiation.

Several different optoelectronic THz techniques, utilizing ultrafast laser pulses, have been recently developed to generate broadband electromagnetic pulses in free space.  

Due to these newly developed techniques, the generation and application of subpicosecond electromagnetic waves is a fast growing field in the photonics and optoelectronics community. Currently there are two basic approaches for generating THz beams: photoconduction and optical rectification. In the photoconductive approach high-speed photoconductors are used as transient current sources for radiating antennas, which include elementary Hertzian dipoles, resonant dipoles, tapered antennas, transmission lines, and large-aperture photoconducting antennas. The optical rectification approach uses electro-optic crystals as a rectification medium and depending on the optical fluence the rectification is a second-order (difference frequency generation) or a higher order nonlinear optical process.

Previous studies of optically induced THz radiation from unbiased semiconductors indicate that no radiated signal is observed from \( \langle 100 \rangle \) GaAs at normal incidence and moderate optical fluence. However, for \( \langle 110 \rangle \) GaAs, a signal with a threefold rotation symmetry around the surface normal was observed.  

In this letter, we report recent results of THz optical rectification from \( \langle 110 \rangle \) oriented GaAs, CdTe, and InP crystals. We calculated the optically induced dielectric polarization in unbiased \( \langle 110 \rangle \) zinc-blende crystals as a function of the crystallographic azimuthal angle by second-order optical rectification. In the far field the electromagnetic radiated field is proportional to the second time derivative of the dielectric polarization. We then measured the peak value of the optically induced THz radiated field as a function of the crystallographic azimuthal angle. There is excellent agreement between the calculated results from the second nonlinear process and the experimental data. The good agreement indicates that, under conditions of moderate optical fluence and normal incidence, second-order optical rectification is the major nonlinear process that generates THz radiation.

When zinc-blende samples are illuminated by femtosecond laser pulses, they emit subpicosecond submillimeter wave radiation,  

generated by ultrafast photocarrier acceleration in the static field region and by nonlinear optical rectification.  

Conventional optical rectification, which was among the first nonlinear optical effects discovered,  

usually refers to the generation of dc electric polarization by an intense optical beam in a nonlinear medium.  

In the THz optical rectification process an ultrafast laser pulse in a nonlinear optical medium creates a beating polarization due to the spectral broadening of the laser pulse due to the uncertainty principle.  

Pulsed electromagnetic waves are then radiated by the time varying electric polarization (a transient dipole). The THz radiation contributed by the photocarrier effect can be eliminated with the use of normal incidence, since there is no radiation along the dipole axis in the direction of the surface field. In general, at normal incidence the forward THz radiation from zinc-blende crystals is purely due to optical rectification.

We have calculated the general expression for the nonlinear dielectric polarization

\[
P_\ell(\Omega) = \varepsilon_0 \int_{\omega_0 - \Delta \omega/2}^{\omega_0 + \Delta \omega/2} d_{j k}(\Omega, \omega) \times E_j(\omega + \Omega) E_k^*(\omega) d\omega,
\]

where \( \omega_0 \) is the central frequency of the incident light beam, \( \Delta \omega \) is the bandwidth of the incident light beam, and \( d_{j k} \) refers to the nonlinear optical susceptibility tensor element of the crystal samples. The frequency \( \Omega \) ranges from dc to \( \Delta \omega \). \( E_j(\Omega + \omega) \) and \( E_k^*(\omega) \) are the Fourier transforms of the electromagnetic field components of the incident light beam. Zinc-blende crystals (most of III-V and some II-VI semiconductors) have cubic structure with point group \( 43m \) and have only one independent nonvanishing second-order nonlinear optical coefficient, namely \( d_4 = d_{25} = d_{36} \).

Since the electromagnetic radiation field is proportional to the second time derivative of the optically induced dielectric polarization, the dielectric polarization is derived so that a comparison with the calculated results to our experimental results can be made. To derive light induced polarization from a \( \langle 110 \rangle \) crystal sample, we use three coordinate systems \( (x, y, z), (x', y', z'), \) and \( (x'', y'', z'') \) to represent the axes fixed in space, the crystallographic axes of the sample, and the principal axes of the sample dielectric tensor, respectively.

The following assumptions have been made: for \( P_\parallel \) (light

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polarized parallel to the dipole detector axis) the light is polarized along the x axis and the detector is aligned with the x axis and for $P_\perp$ (light polarized perpendicular to the dipole detector axis) the light is polarized along the y axis and the detector is aligned with the x axis. By definition the electric field of the optical beam in the $(x,y,z)$ system has the simple form

$$E = E[1,0,0].$$

The electric field can then be expressed in the crystallographic axis, $(x',y',z')$, by a simple mathematical transformation from the electric field in the fixed space coordinate system $(x,y,z)$. Through another transformation of the electric field from the $(x',y',z')$ coordinate system to $(x'',y'',z'')$ coordinate system, the electric field of the optical beam takes the form

$$E = E\left(-\frac{1}{\sqrt{2}} \sin \theta, \frac{1}{\sqrt{2}} \sin \theta, \cos \theta\right),$$

where $\theta$ is the azimuthal angle between the space coordinate, $(x,y,z)$, and the crystallographic coordinate, $(x',y',z')$, measured with respect to the $(-1,1,0)$ direction. By definition the dielectric polarization in the principal axis system, $(x'',y'',z'')$, has the form

$$P = (P_\parallel, P_\perp, P_z) = \epsilon_0 d_{14} E^2(E_{y''}E_{x''}, E_{x''}E_{x''}, E_{x''}E_{y''}).$$

By substitution of the components from Eq. (3) into Eq. (4) the polarization in the $(x'',y'',z'')$ coordinate system can be expressed as

$$P = \epsilon_0 d_{14} E^2\left(\frac{1}{\sqrt{2}} \sin \theta \cos \theta, -\frac{1}{\sqrt{2}} \sin \theta \cos \theta, -\frac{1}{2} \sin^2 \theta\right).$$

Converting the polarization from Eq. (5) in the $(x'',y'',z'')$ coordinate system into the fixed coordinate system $(x,y,z)$, the polarization takes the form

$$P = \epsilon_0 d_{14} E^2\left(\frac{1}{\sqrt{2}} \sin \theta \cos \theta, -\frac{1}{\sqrt{2}} \sin \theta \cos \theta, \frac{1}{2} \sin^2 \theta\right).$$

Hence, the component of the light induced polarization parallel to the incident beam can be expressed as

$$P_\parallel = P_x = \frac{1}{2} \epsilon_0 d_{14} E^2(\cos \theta - \cos \theta).$$

Similarly, when the light beam is polarized in the direction of $y$ and the detector is still aligned in the $x$ direction, the polarization has the form

$$P_\parallel = P_y = \frac{1}{2} \epsilon_0 d_{14} E^2(3 \cos \theta + \cos \theta).$$

Our configuration for generation and detection of broadband pulsed submillimeter waves consisted of a femtosecond laser and a submillimeter wave polarization sensitive detector. A cw mode-locked Ti:sapphire laser was used as the source of a continuous train of 200 fs duration optical pulses which is divided by a beam splitter into pump (optically stronger) and probe (optically weaker) pulses. The pulse repetition rate was 76 MHz and the peak optical fluence was 48 nJ/cm$^2$. As shown by Fig. 1 the probe pulse gates the photoconducting detector while the pump pulse passes through a time delay before illuminating the sample with a 4 mm spot. The radiated submillimeter beam was focused by a silicon lens on a photoconductor attached to a dipole antenna. When the pulsed submillimeter wave and the gating optical pulse simultaneously and spatially strike the detector a photocur-
rent will develop at the antenna detector. The photocurrent is then amplified by a lock-in amplifier and analyzed by a computer. In angle-dependent measurements the samples were placed in the laser beam path (in front of the dipole detector) and rotated around its surface normal. Measurements were performed at room temperature. In general, we measured THz radiation in the forward direction; however, the THz signal can radiate transversely as well.

We have measured the angular dependent radiation by measuring the transmitted electromagnetic wave through (110) oriented InP, GaAs, and CdTe wafers as a function of azimuthal angle. Figures 2 and 3 show the peak radiation from a (110) GaAs wafer versus azimuthal angle at normal optical incidence angle and 813 nm laser wavelength, with optical polarization parallel (Fig. 2) and perpendicular (Fig. 3) to the dipole detector axis. The solid dots are the experimental data while the curves are the plots from Eqs. (7) and (8). The amplitude was the only fitting parameter. In Figs. 2 and 3 the experimental data deviate slightly from the calculated curve, because the surface normal of the (110) GaAs sample is 6° off the (1,1,0) plane.

Figures 4 and 5 plot peak values of radiated signal from (110) CdTe versus the crystal azimuthal angle at 878 nm laser wavelength, with polarization of the detected signal parallel (Fig. 4) and perpendicular (Fig. 5) to that of the fundamental beam. The surface normal of the CdTe crystal was 2° off the (1,1,0) plane. Due to the smaller orientation deviation, better agreement between the calculated curve and the experimental data for CdTe is attained. Similar results were also observed from (110) InP samples. The results from the (110) oriented zinc-blende crystals confirm our previous conclusions from (100) and (111) oriented zinc-blende crystals and provide strong evidence that second-order optical rectification is the major nonlinear process under the condition of moderate optical fluence and normal incidence on the unbiased.

Under normal optical illumination on (110) CdTe and GaAs surfaces the amplitude of the nonlinear susceptibility varies dramatically and the sign of the susceptibility changes when the photon energy was tuned near the band gap. Similar results were observed for (111) GaAs. The amplitude enhancement and sign reversal of THz radiation is due to the dispersion of nonlinear susceptibility tensor, $d_{14}$, near the electron resonance state.

In conclusion, we have measured THz optically induced electromagnetic submillimeter waves from (110) oriented zinc-blende crystals as a function of azimuthal angle, with optical polarization parallel and perpendicular to the dipole detector axis. Excellent agreement between the experimental and theoretical results for the angular-dependent experiments provides evidence that second-order optical rectification is the major nonlinear process under the condition of moderate optical fluence and normal incidence on the unbiased sample.

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