

Detection of freely propagating terahertz radiation by use of optical second-harmonic generation

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We report the application of electric-field-induced optical second-harmonic generation as a new technique for measuring the field of freely propagating terahertz radiation. Using silicon as the nonlinear medium, we demonstrate subpicosecond time resolution and a sampling signal that varies linearly with the terahertz electric field. This approach, which is attractive for centrosymmetric media, permits a significantly broadened class of materials to be exploited for free-space sampling measurements. © 1998 Optical Society of America

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The use of ultrafast laser sources to generate and detect freely propagating terahertz (THz) radiation has been a subject of considerable interest in recent years. At present, the most popular means for detecting this radiation is through the use of photoconductive detectors.¹ Although these devices can be highly sensitive, the frequency response is constrained by the time constants associated with carrier dynamics. With the widespread availability of lasers exhibiting pulse durations as short as 10 fs, there is strong incentive to translate this enormous optical bandwidth to the far infrared. The use of nonlinear-optical approaches seems well suited for this application. This is exemplified by the recent demonstration of electro-optic sampling²⁻⁴ as a sensitive probe of transient far-infrared pulses. Although the technique has a number of attractive features, including the capability for broadband phase-matched interaction,⁵ the existence of strong vibrational resonances in many electro-optic crystals can impose limits on the system response.

Optical second-harmonic generation (SHG) has been shown to be highly sensitive to electric fields.^{6,7} The process was first observed in calcite by Terhune *et al.*⁸ and extended to semiconductors and metals by Lee *et al.*⁹ Several further studies examined the effect of static electric fields in centrosymmetric¹⁰ and noncentrosymmetric¹¹ semiconductors. The phenomenon has also been exploited to probe rapidly varying electric fields guided by transmission lines. Using femtosecond laser pulses to sample the electric field in the silicon substrate, Ohlhoff *et al.*¹² were able to detect a high-frequency sine wave, and we¹³ were able to resolve subpicosecond pulses generated optoelectronically.

In this Letter we demonstrate the use of electric-field-induced SHG to characterize freely propagating electromagnetic radiation. In contrast with electro-optic sampling, which requires noncentrosymmetric materials, the SHG sampling technique is applicable to centrosymmetric media. This fact is important, since it creates the possibility of using a broader class of materials, including those with favorable properties at the frequency of the probed electromagnetic radiation. For example, it can be shown that for cer-

tain centrosymmetric media, such as silicon, the ionic contribution to the dielectric function is extremely weak.¹⁴ This weakness is due to the nonpolar nature of silicon. Indeed, the peak absorption coefficient of the lowest phonon resonance in silicon is $\sim 10 \text{ cm}^{-1}$, which is more than 2 orders of magnitude lower than the corresponding absorption in typical electro-optic crystals such as LiTaO_3 .¹⁵ Here we report the measurement of the electric field of THz radiation by means of SHG sampling in silicon. Subpicosecond time resolution is achieved, together with a linear response to the strength of the THz electric field.

The overall experimental setup is shown schematically in Fig. 1. A mode-locked Ti:sapphire laser producing optical pulses of 60-fs duration at 800 nm and a 100-MHz repetition rate was used to generate and detect the THz radiation. The THz pulses were produced by a large-aperture photoconductive emitter and focused onto the detection medium, which consisted of an ion-implanted silicon-on-sapphire wafer. The 0.6- μm thick epitaxial silicon film was amorphized during the implantation process and thus rendered nearly isotropic. Since the escape depth of the second-harmonic (SH) radiation is only 125 nm, the silicon film may be considered equivalent to a bulk sample for these measurements. The probe beam, with an average power as great as 75 mW, was incident upon

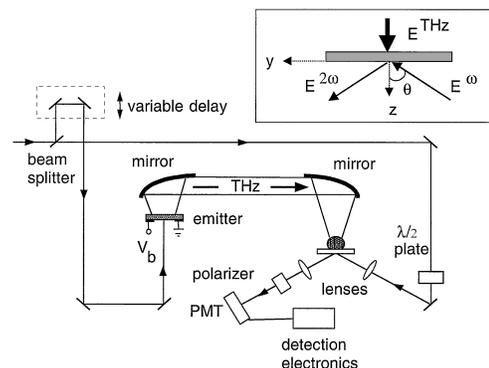


Fig. 1. Schematic of the experimental setup. PMT, photomultiplier tube. Inset: Detail of the electric-field-induced SHG configuration.

the silicon sample at a 45° angle and was focused to approximately 10 μm. The *s*-polarized SH radiation was detected in reflection with a photomultiplier tube and standard photon-counting techniques.

The field-induced SHG process may be described as a four-wave-mixing process in which radiation at frequency 2ω is produced from fields at frequencies ω, ω, and 0. The third-order susceptibility tensor, χ⁽³⁾(2ω = ω + ω + 0), describing this process has 21 nonvanishing elements for an isotropic medium, of which only three are independent.¹⁶ In the experimental configuration (Fig. 1, inset) the THz (i.e., the low-frequency) electric field **E**^{THz} is normally incident upon the Si detection element and the plane of incidence of the probe laser is perpendicular to the polarization of **E**^{THz}. For this arrangement the relevant tensor components are χ_{xxxx}⁽³⁾ and χ_{xyyx}⁽³⁾ = χ_{xzxx}⁽³⁾. Neglecting for the moment any background SHG in the absence of an applied THz field, we may write the radiation SH field as

$$\Delta \mathbf{E}_x^{2\omega} \propto [\chi_{xxxx}^{(3)} \cos^2 \varphi + \chi_{xyyx}^{(3)} \sin^2 \varphi] \mathbf{E}^\omega \mathbf{E}^\omega \mathbf{E}_x^{\text{THz}} \cdot l, \quad (1)$$

where φ denotes the polarization angle of the probe beam measured with respect to *s* polarization (the *x* axis), **E**^ω represents the electric field of the incident probe beam at frequency ω, and *l* is the escape length for the SH radiation. For simplicity here and below, we omit the Fresnel factors⁶ describing the influence of the linear-optical properties of the sample, since they do not significantly affect the present discussion.

Experimentally, we measure the SH intensity, which would vary quadratically with the strength of the THz field **E**^{THz}. To obtain a detected signal linear in the THz electric field, we use the SH radiation produced by the sample in the absence of an applied field. Since the silicon sample is centrosymmetric, no strong (dipole-allowed) SHG process occurs. A weaker response may arise, however, from the surface and bulk quadrupole terms.^{6,7} For an isotropic material with mirror symmetry, this contribution to the SH radiation can be characterized by a surface nonlinear susceptibility tensor χ_s⁽²⁾(2ω = ω + ω) with three independent elements: χ_{s,zzz}⁽²⁾, χ_{s,zxx}⁽²⁾ = χ_{s,zyy}⁽²⁾, and χ_{s,xzx}⁽²⁾ = χ_{s,yzy}⁽²⁾. For the purposes of this discussion these tensor elements may be taken to include contributions from bulk nonlocal terms and any static field-induced effects, since the angular dependence of these contributions is functionally identical to that of the surface contributions.⁶ For the particular case of detecting *s*-polarized SH radiation, we need consider only the element χ_{s,xzx}⁽²⁾ (see Fig. 1). The background contribution to the SH radiation, **E**_o^{2ω}, expected in the absence of the THz field is then given by

$$\mathbf{E}_o^{2\omega} \propto \sin \theta \sin 2\varphi \chi_{s,xzx}^{(2)} \mathbf{E}^\omega \mathbf{E}^\omega, \quad (2)$$

where θ is the angle of incidence. Thus, in the absence of a THz field, the corresponding SH intensity varies as $I_o^{2\omega} \propto |\chi_{s,xzx}^{(2)}|^2 \sin^2 \theta \sin^2 2\varphi (I^\omega)^2$, where *I*^ω is the optical intensity of the fundamental beam.

When a THz electric field is present and the probe beam is of mixed (*s/p*) polarization, both SH electric-field contributions, Δ**E**^{2ω} and **E**_o^{2ω}, are present. The resulting SH intensity is given by

$$I^{2\omega} \approx I_o^{2\omega} + a \mathbf{E}^{\text{THz}} (I^\omega)^2 \equiv I_o^{2\omega} + \Delta I^{2\omega}, \quad (3)$$

where $a \propto \{\text{Re}[\chi_{s,xzx}^{(2)*} \chi_{xxxx}^{(3)}] \cos^3 \varphi \sin \varphi \sin \theta + \text{Re}[\chi_{s,xzx}^{(2)*} \chi_{xyyx}^{(3)}] \cos \varphi \sin^3 \varphi \sin \theta\}$ contains the dependence on the polarization angle and the coefficients for the nonlinear material response. Here we neglect quadratic terms in Δ**E**^{2ω} under the assumption that the background signal is large compared with the field-induced signal. In this limit the SH intensity consists of a quiescent term, *I*_o^{2ω}, which is constant for a fixed experimental arrangement, and a signal term, Δ*I*^{2ω}, which varies linearly with the magnitude of the THz electric field.

We initially examined the SH signal variation in the absence of the THz field as a function of the polarization of the probe beam. The observed SH intensity versus the polarization angle of the fundamental beam, shown in Fig. 2, is found to follow the expected variation. In Fig. 2, φ = 0° corresponds to *s*-polarized light. We note that for a purely *s*- or *p*-polarized fundamental beam the background SH vanishes.

We measured the freely propagating THz electric field, using two different polarization angles for the linearly polarized fundamental beam. The time-resolved SH intensity *I*^{2ω} for polarization angles of φ₁ = 14° and φ₂ = 8° is shown in Fig. 3. In both cases we subtracted the dc offset, *I*_o^{2ω}, which is typically ~50 times as large as Δ*I*^{2ω}. As expected from expression (3), the two temporal waveforms look the same, aside from a scaling factor. For relatively small values of the polarization angle φ it can be shown that Δ*I*(φ₁)/Δ*I*(φ₂) ≅ [*I*_o(φ₁)/*I*_o(φ₂)]^{1/2}. For the two polarization angles used

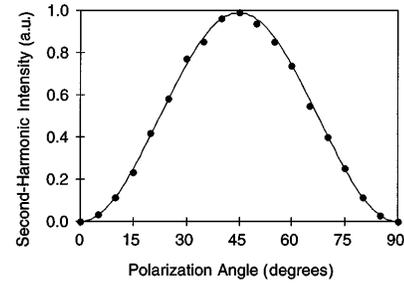


Fig. 2. Variation in the SH intensity from the silicon sample as a function of the polarization of the probe laser (φ = 0° corresponds to *s* polarization) in the absence of a THz electric field. The filled circles are experimental data, and the solid curve represents a fit to theory.

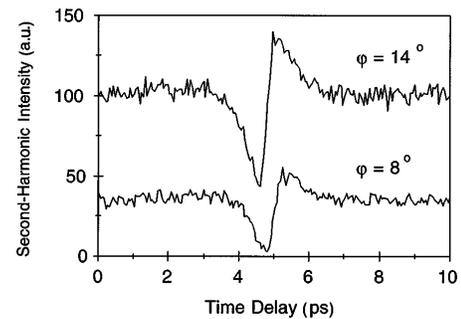


Fig. 3. Waveform of the THz electric field measured by the field-induced SHG sampling technique. Curves are shown for probe polarizations of φ = 14° and φ = 8°. In both cases the quiescent contribution to the SH intensity, *I*_o^{2ω}, was subtracted and the waveforms were offset from the origin for clarity.

above, this ratio is ~ 1.7 and is in good agreement with our data.

As we discussed previously,¹³ we anticipate an intrinsic response time of < 10 fs for this sampling technique. In the present experiment the time resolution of the SHG detection scheme should then reflect simply the finite (60-fs) duration of the laser pulse. In the waveforms shown in Fig. 3 the transition from the waveform minimum to the waveform maximum occurs in approximately 300 fs and is presumed to correspond to the finite bandwidth of the photoconductive emitter used to generate the THz pulse.

As noted above, the escape length of the SH radiation from silicon is extremely short. Thus there is no particular advantage in using a copropagating versus a counterpropagating beam geometry. However, with the appropriate choice of transparent media, we expect that a transmission geometry with appropriate phase matching will yield a significant enhancement in the detection sensitivity, without deleterious effects on the detection response time. We are currently investigating this approach.

In conclusion, we have demonstrated the detection of freely propagating THz radiation, using field-induced SHG in a centrosymmetric medium. By utilizing the SH radiation emanating from the surface of the nonlinear medium, we obtained a linear response in the amplitude of the THz electric field. The SHG sampling scheme offers the advantage of being applicable to a broad class of centrosymmetric media for which conventional electro-optic sampling techniques cannot be applied. This expanded class of materials may be particularly helpful in dealing with the complications encountered in electro-optic sampling as one attempts to probe time-varying fields at frequencies comparable with those of the phonons in the sampling medium.

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