Cherenkov Radiation from Femtosecond Optical Pulses in Electro-Optic Media

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The propagation of femtosecond-duration optical pulses in electro-optic materials is observed to produce a Cherenkov cone of pulsed far-infrared radiation in the terahertz spectral range. Coherent detection of the electric field of this far-infrared pulse by electro-optic sampling shows it to have a duration of approximately one cycle and an amplitude of 10 V/cm making it a potentially attractive source for transient far-infrared spectroscopy.

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It was recently suggested that under suitable conditions, the propagation of femtosecond optical pulses in electro-optic materials should be accompanied by the radiation of an extremely fast electromagnetic transient.[1,2] This phenomenon, which arises from the inverse electro-optic effect,[3] produces a Cherenkov cone of pulsed radiation having a duration of approximately one cycle and a frequency in the terahertz range. Although closely resembling the classical Cherenkov radiation from relativistic charged particles in dielectric media, the physical basis for this effect has some unique distinguishing features. Most important, it is a nonlinear optical effect, arising from a second-order nonlinear polarization, and consequently requires a lack of inversion symmetry in the host material. Also, the charge state of the effective source is neutral, being a dipole moment, rather than a point charge. The velocity of the source, however, does exceed the radiation velocity as a result of the additional contribution to the low-frequency dielectric permittivity from the infrared lattice vibrations. As indicated in Fig. 1, this is expected to produce a characteristic cone of radiation in the form of a shock wave. Unlike classical Cherenkov radiation, however, the radiation source is spatially extended, being proportional to the intensity envelope of the optical pulse. Consequently the details of the radiation field are expected to depend sensitively on both the duration and the beam waist of the optical pulse. A discussion of these effects, based on an analysis of this phenomenon, is given in Refs. 1 and 2. Our approach differs from previous work[4] on the use of the inverse electro-optic effect for microwave and far-infrared generation by its emphasis on the use of extremely short optical pulses and the consequent relaxation of phase-matching requirements, and the coherent detection of the generated wave form.

We report here the direct experimental observation of this phenomenon. Our approach is illustrated schematically in Fig. 1. Lithium tantalate was chosen as the electro-optic medium for the following specific reasons. First, it has a relatively large electro-optic coefficient along its polar axis, equal to 2.8×10^{-11} m/V.[5] Second, it has a very low anisotropy of both its optical and low-frequency dielectric permittivities (0.2% and 0.5%, respectively). Third, it has a relatively large region of transparency in the far infrared, extending from dc to the first lattice vibrations at approximately 6 THz.[6]

Femtosecond optical pulses were obtained from a colliding-pulse mode-locked ring dye laser.[7] Their duration, measured by second-harmonic autocorrelation, was 100 fs (full width at half maximum), and their center wavelength was 625 nm. A rela-

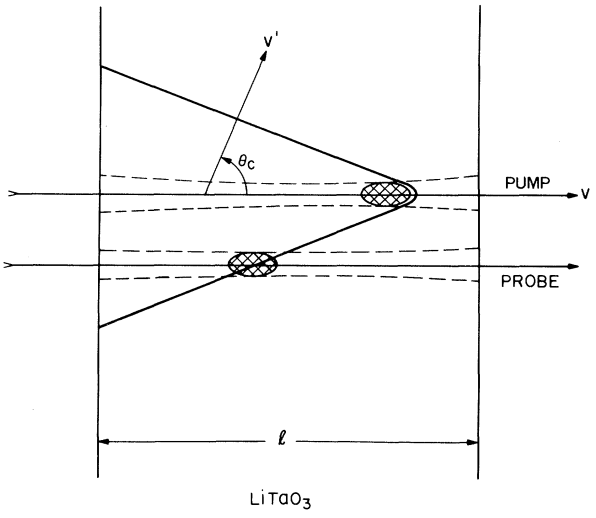

FIG. 1. Schematic of the experiment used to generate and detect short bursts of far-infrared Cherenkov radiation from femtosecond optical pulses in lithium tantalate. The Cherenkov cone of radiation propagates away from the "pump" pulse in a direction θ_c with a velocity which is determined by the ratio of the low-frequency index of refraction (6.53) to the optical group velocity (c/2.33). A "probe" pulse measures the small birefringence due to the electro-optic effect induced by the electric field of the far-infrared transient as it moves in synchronism with the Cherenkov wave front.

tively low pulse energy of only 10^{-10} J at a repetition rate of 150 MHz was sufficient for the experiment.

As indicated in Fig. 1, two optical pulses were used, one to generate the radiation field, and the other to detect it. This configuration has a novel symmetry arising from the use of the electro-optic effect for both generation and detection. In the case of the generating pulse, it is the inverse electro-optic effect that produces the nonlinear polarization responsible for the radiation field, and in the detection process, it is the direct electro-optic effect that is used to measure the small birefringence produced by the electric field of the radiation pulse. This latter technique, known as electro-optic sampling,[8] has previously been used to measure subpicosecond electrical transients in traveling-wave electro-optic transmission lines.

Both optical pulses were focused on a sample of lithium tantalate through a common lens and were carefully aligned to propagate parallel through the crystal. The generating pulse was polarized parallel to the c axis of the crystal (out of the plane of Fig. 1), to produce a radiation field polarized in the same direction by the r_{33} electro-optic coefficient.

The method of detection is based on Pockel's effect suitably modified for transient measurements with femtosecond pulses as described in Ref. 8. The probing pulse was polarized 45° to the c axis of the lithium tantalate, and the static birefringence was compensated by a Soleil-Babinet compensator placed after the crystal. A calcite Glan-Thompson polarizing prism was used as an analyzer to separate the two orthogonally polarized components of the transmitted pulse. Differential detection of these two signals was used to measure the rotation of the axis of polarization of the transmitted optical pulse arising from the birefringence produced by the radiation field. The compensator was used to bias the detection system at the point of maximum incremental sensitivity where the response is linearly proportional to the electric field of the radiated wave.

A unique feature of the use of optical pulses for both generation and detection of the radiation field is the automatic synchronism of the velocities of the radiation field and the probing pulse. As indicated in Fig. 1, the probing pulse "surfs" along the Cherenkov wave front enabling it to measure the electric field at a stationary point in the wave form by integrating the birefringence along the entire path through the crystal. To plot out the shape of the wave form, the timing of the probing pulse was delayed or advanced relative to the generating pulse by introduction of a variable path length between them.

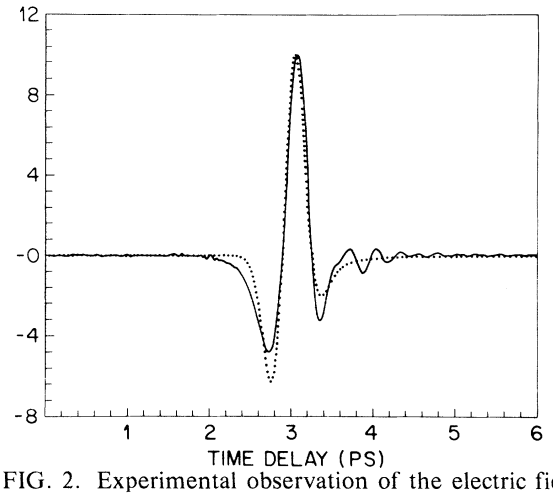

FIG. 2. Experimental observation of the electric field of the far-infrared transient (solid line) measured by the method illustrated in Fig. 1. For comparison, the theoretical wave form is also plotted (dotted line), for an optical pulse duration of 60 fs and an optical beam waist of 3.8 μm ($1/e$ half-widths for Gaussian profiles). The peak amplitude of the electric field is approximately 10 V/cm.

The observed wave form for our particular experimental configuration is shown in Fig. 2. It is extremely fast and approximates a single cycle of a frequency of 1.5 THz. With use of the known electro-optic coefficient for lithium tantalate, the amplitude of the electric field was estimated from the measured birefringence to be 10 V/cm. The cone angle could also be measured by changing the lateral spacing between the optical pulses and observing the corresponding optical delay necessary to restore synchronism with the Cherenkov wave front. This angle, θ_c, as defined in Fig. 1, was found to be 69° equal to the theoretical value given by the inverse cosine of the ratio of the low-frequency radiation velocity ($0.153c$) to group velocity of the optical pulse ($0.433c$). The frequency spectrum of the wave form, determined by a numerical transform is shown in Fig. 3. The spectral content of the pulse is extremeley broad, extending from dc to 4 THz.

An analysis of the radiation process[2] predicts the following expression for the time variation of the electric field:

$$E(t)=\frac{n_0^2\epsilon_3 r_{33}\mathscr{E}_p}{c^2\tau^2}\left[\frac{\sqrt{2}\cot\theta_c}{v\tau r_\perp}\right]^{1/2}U\left(-2,-\frac{\sqrt{2}t}{\tau}\right)\exp\left(-\frac{t^2}{2\tau^2}\right), \quad (1)$$

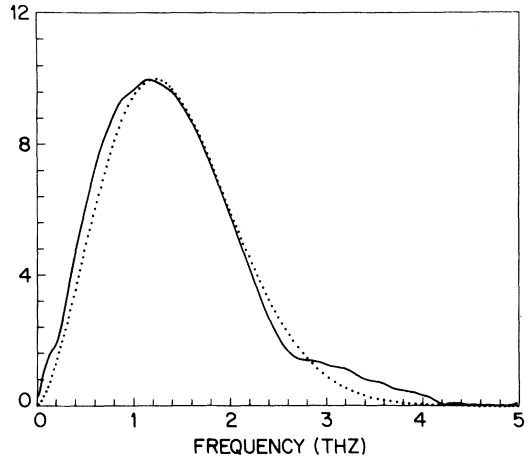

FIG. 3. Fourier spectra of the experimental (solid line) and theoretical (dotted line) wave forms in Fig. 2.

where $\mathscr{E}_p$ is the optical pulse energy, n_0 and v are the optical index of refraction and group velocity, $r_\perp$ is the radial distance from the beam axis, $U(-2,x)$ is the parabolic cylinder function of order -2, and the parameter τ is defined by the relationship

$$\tau=\left(\tau_p^2+\frac{w^2\tan^2\theta_c}{v^2}\right)^{1/2}, \tag{2}$$

where τ_p is the $1/e$ half-width of the duration of the optical pulse and w is the $1/e$ beam radius (both assumed to have Gaussian profiles). The theory neglects dispersion, and assumes that the point of observation is much greater than the optical beam waist. A unique feature of this result is the variation of the electric field amplitude with the inverse $\frac{5}{2}$ power of the pulse width parameter τ (the field intensity varies as the inverse fifth power of τ). This makes the efficiency of this process as well as its speed much more attractive when extremely short optical pulses are used. A second aspect of the theory is the contribution to the infrared pulse duration from the beam waist of the optical pulse as illustrated by expression (2). This means that tight focusing is necessary to produce extremely fast pulses.

The theoretical wave form is plotted in Fig. 2 by choosing the width parameter τ to fit the zero-crossing points of the experimental wave form ($\tau=225$ fs). This is equivalent to a choice of optical pulse duration and beam waist of 60 fs and 3.8 μm, respectively, in good agreement with the experimental values. Aside from the imbalance of the two negative lobes, the most dramatic difference between the theoretical and experimental wave forms is the sustained high-frequency ringing following the experimental pulse. This feature is also evident in the spectra of the theoretical and experimental wave forms as shown in Fig. 3, and is probably due to the strong dispersion and absorption associated with the infrared lattice vibrations in lithium tantalate at 6 THz. The measurement technique introduces an additional broadening to the observed wave form due to the spatial and temporal convolution between the electric field and the probing pulse. If the generating and probing pulses are identical, and no other sources of broadening are present, this would introduce an additional factor of $\sqrt{2}$ into the parameter τ in expression (2). This is equivalent to a reduction in the time scale in Fig. 2 by $\sqrt{2}$ and a corresponding expansion of the frequency scale in Fig. 3 by the same factor. In our experiment, however, there is a substantial broadening due to dispersion, which makes this simple deconvolution scheme inappropriate.

To determine more accurately the influence of absorption and dispersion we have measured the decay and broadening of the wave form as the distance between detector and generator is increased over the range from 100 to 650 μm. Fourier analysis of these wave forms gives the complete variation of the absorbance over the spectral range spanned by the far-infrared wave form. We find that the absorbance varies as the square of the frequency and has the value of 43 cm^{-1} at 1 THz. This value is almost a factor of 2 smaller than that reported in Ref. 6. From this result we deduce a damping time of 143 fs for the lattice vibration at 6 THz.

The coherent nature of the detection process makes this approach a novel method of measuring the far-infrared properties of materials by observing the change in wave form in the time domain due to dispersion in the frequency domain. Since the time reference is accurately known, phase information is retained, and consequently both the real and the imaginary parts of the dielectric function can be obtained over the entire spectral range spanned by the pulse. For example, we are currently exploring the use of this method for the direct observation of lattice vibrations in the time domain, and for the determination of the momentum relaxation times of free carriers in bulk and multi-quantum-well semiconductors. The use of more intense optical pulses would result in substantially larger electric fields of a few tens of kilovolts per centimeter making possible the direct observation of transient nonequilibrium phenomena such as velocity overshoot and ballistic propagation. The details of

these and other applications will be described in subsequent publications.

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