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Generation of High-Power, Subpicosecond, Submillimeter Radiation for Applications in Novel Device Development and Materials Research

Antoinette J. Taylor*, Jeffrey P. Roberts, Norman A. Kurnit, Pamela K. Benicewicz, George Rodriguez, Antonio Redondo, Darryl L. G. Smith, and Timothy J. Carrig

Abstract

This research exploits the high-energy, ultrafast laser technology and high voltage expertise at Los Alamos National Laboratory (LANL) to scale submillimeter-pulse generation with photoconducting antennas to large aperture sizes and high output powers. An experimental and theoretical approach was undertaken with a view towards optimizing the radiated output and determining the technology's ultimate scalability. This is the final report of a three-year Laboratory-Directed Research and Development (LDRD) project at LANL.

1. Background and Research Objectives

A practical technique has only recently been demonstrated for generating single-cycle pulses in the submillimeter-wave region. By exposing a large-aperture photoconductor (biased at high voltage) to a femtosecond visible laser pulse, submillimeter-wave radiation is generated by the impulsively driven photocurrent developed in the sample as the bias voltage is shorted to ground. Since from elementary electrodynamics a radiation field is produced only by a time-varying current, the far-field radiated pulse is comparable in duration to the optical excitation pulse, regardless of the carrier lifetime in the photoconductor. This radiated terahertz-bandwidth pulse is directional, diffraction-limited, and steerable.

The purpose of this research is to exploit the high-energy, ultrafast laser technology and high voltage expertise present at LANL to scale submillimeter-pulse generation with photoconducting antennas to high output powers. An experimental and theoretical approach resulting in a thorough understanding of the properties of this unique source of radiation in mobility of different electronic materials, particularly in the high field regime. Generation of both the near and far fields was undertaken with a view towards optimizing the radiated output and determining the technology's ultimate scalability. The characteristics of the generated submillimeter radiation was studied to determine the transient charge-carrier terahertz radiation

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through nonlinear processes in unbiased materials was investigated. The efficiency and scalability of terahertz radiation from these materials was compared to terahertz generation from biased photoconductors and large aperture unbiased emitters were developed.

2. Importance to LANL's Science and Technology Base and National R&D Needs

a. Applications in Electronic Materials Research

Single-cycle submillimeter pulses can reveal both the transient and steady-state response characteristics of electronic materials such as semiconductors, superlattices, superconductors, polymers, and quasi-one-dimensional MX electronic materials (transition metal ions bridged by halogen ions) in the frequency range from 0.1 to 2 THz. By comparing the shape of the incident pulse with a pulse transmitted through the sample, the frequency-dependent absorption and dispersion of the material can be deduced. The advantages of this technique over current methods include its enormous spectral bandwidth, the ability to use coherent detection (which allows the extraction of both the imaginary and real parts of the dielectric response), and a sensitivity which exceeds that of conventional far-infrared spectrometers operating in the same spectral range.

High-power THz pulses can also be used as *nonlinear* pump pulses to investigate the ultrafast dynamics of high-field interactions in materials. A THz pulse is focused into the sample and its effect on an optical probe pulse is measured as a function of the temporal delay between the pump and the probe. Applications include studies of the dynamic-Stark and Franz-Keldysh effects.

The characteristics of the generated submillimeter radiation can provide insights into the microscopic physics of the photoconducting antenna itself. In the near field the radiated pulse is proportional to the transient photocurrent, while in the far field it is proportional to the time-derivative of the transient photocurrent. By analyzing the temporal shape and magnitude of the radiated pulse in both regimes, such important quantities as the transient carrier mobility and its relaxation rate can be determined as a function of the carrier excitation energy. By varying the fluence of the optical excitation pulse, the effect of carrier-carrier interactions on the transient mobility can be measured. It is even possible to access the ballistic transport regime by increasing the bias voltage up to the breakdown voltage of the material.

Critical experimental features intrinsic to the THz-pulse generation technique and relevant to all of the applications described above are that the temporal resolution is limited only by the optical-pulse duration (< 100 fs), and that problematic high-speed electrical contacts are never needed. The techniques can therefore be easily applied not only to conventional

semiconductors, but also to more exotic materials such as superlattices, photoconducting polymers, and quasi-one-dimensional MX electronic materials.

b. Applications in Novel Device Development

Areas for potential device applications include satellite communications, laser-particle acceleration, ultrafast electro-optic switching, fast-scanning radar for sensor development and surveillance, and battlefield electronic sensor negation.

Increased bandwidth of satellite communications links is absolutely necessary to handle increasing traffic and to reduce costs. Time-division multiplexing at THz bandwidths has already been demonstrated using an array of photoconducting antennas in which an electronically controlled spatial-dependence of the bias field is translated into a time-dependence in the radiated waveform. Demultiplexing of this signal with an array of photodetectors requires scaling of the technology to high output power levels.

The electric-field strengths possible with a high-power THz pulse far exceed those available with conventional approaches to charged-particle acceleration. In particular, the development of a high-brightness electron gun using this technology seems feasible. This application would use a high power, ultrashort optical pulse to eject electrons from a small cathode as well as to produce the high-field electromagnetic pulse required to accelerate the ejected electrons to high energy. Extension of the technique to compact accelerating structures could also be pursued.

The large transient electric field of a THz pulse can be exploited to give one or two orders of magnitude improvement in the risetime of current electro-optic switches. High field strengths also enable the use of significantly thinner electro-optic crystals, hence minimizing pulse broadening by group-velocity dispersion. Such devices have applications in laser systems for pulse shortening and prepulse suppression.

Electrical steerability of THz pulses has been demonstrated at low power levels using an array of photoconducting antennas. With applications in air traffic control or military surveillance, this technique can be developed into scanning radar devices for fast tracking of moving objects. Again, for these applications high output powers must be demonstrated.

Finally, there are several military applications for high-power THz pulses. For example, they may be useful tools for developing and testing EMP-hardened electronics, and conversely, may help determine the feasibility of electronic sensor negation. The demand for submillimeter wavelengths in these applications is driven by the decreasing size of electronic components; submillimeter waves couple more strongly to critical electronic microstructures, thus enhancing their capacity to disrupt sensitive circuits.

3. Scientific Approach and Results to Date

A study of the submillimeter radiation generated from GaAs and InP emitters over a large range of optical fluences and bias fields was completed. The saturation behavior, which is essential for the optimal design of efficient emitters, was determined and related to the carrier mobility in the material. The dependence on the emitted radiation from GaAs and InP emitters on crystal orientation was examined. No orientational effects were observed, validating the current surge model for biased emitters.

Saturation of the radiated THz output from a large-aperture, biased photoconductor at high optical fluence effectively limits the terahertz output from biased photoconductors. Previously, this process has been explained in terms of screening of the applied electric field by the THz radiation. Space charge screening of the applied bias field induced by separating carriers should also contribute to saturation, as previously observed in dipole antennas. To elucidate the role of these two mechanisms in the saturation process, we have performed pump-probe measurements in large-aperture, biased photoconductors. We measured the peak radiated THz output from the probe versus pump-probe delay for a variety of bias voltages (1 to 10 kV/cm) and probe fluences well below saturation. The pump fluence is varied over fluences corresponding to a photoexcited carrier densities from 0.084 to $2.1 \times 10^{18} \text{ cm}^{-3}$. For carrier densities as high as $5 \times 10^{17} \text{ cm}^{-3}$, the decrease in THz radiation from the probe is small near zero delay, implying that space charge screening does not significantly affect the THz output in this regime. For carrier densities in excess of 10^{18} cm^{-3} , the decrease in THz radiation near zero delay is significant, implying that space-charge screening affects THz output in this range. For bias fields above 2.5 kV/cm, the proportional decrease in THz radiation at zero delay is roughly independent of bias, implying that over that range of bias fields, space-charge screening does not depend on bias field.

A theoretical model describing the far-field radiation from biased large-aperture photoconducting emitters has been developed. The radiated fluence and intensity, as well as waveforms of the radiated electric fields have been calculated as a function of optical excitation energy, bias field and fluence. Transient velocity effects, including velocity overshoot, as well as the effects of electron-hole and electron-phonon scattering have further been incorporated in the model.

We have also modeled the effects of carrier screening at high excitation densities. For large aperture emitters, modeling of this saturation process to date has included only the screening of the applied electric field by the generated THz radiation. However, the applied bias field will also be screened by the space-charge induced by separating carriers, also contributing to saturation of the terahertz output. To model these saturation mechanisms we

determined the time-dependent response of a photoconductor to ultrashort pulse excitation. This required the solution of a system of nonlinearly coupled partial differential equations. One component of the system consisted of the continuity equations for the electron density and hole density. Current transport equations were used to account for drift of electrons and holes. Finally Poisson's equation was used to determine the effect of charge separation on the electric field. The results were in agreement with both the experimentally measured saturation curves and the pump-probe space-charge screening measurements.

Optical rectification of femtosecond pulses in electro-optic crystals also produces terahertz radiation. Such radiation should be linearly scalable with excitation fluence until higher-order nonlinear effects become important. We have compared the performance of three electro-optic crystals, LiNbO₃, LiTaO₃, and the organic salt dimethyl amino 4-N-methylstilbazolium tosylate (DAST) as THz emitters. Our results extend previous studies by increasing the optical excitation energies by six orders of magnitude. The generated peak electric field radiated by LiNbO₃ and LiTaO₃ emitters increases linearly with increasing optical fluence for fluences up to 60 mJ/cm², while for DAST emitters the peak field begins to saturate at fluences greater than 20 mJ/cm². At excitation fluences below 20 mJ/cm², DAST is roughly fourteen times more efficient at THz generation than LiNbO₃, while at a fluence of 60 mJ/cm² DAST is only nine times more efficient. This decrease in efficiency of THz generation from DAST was accompanied by strong fluorescence, indicating that two-photon absorption may be occurring.

To avoid the saturation phenomena in DAST, while still generating high energy THz radiation, it is necessary to use large-aperture emitters. The problem with this approach is that large, high optical quality crystals cannot be easily grown. Our solution is to use a multicrystal DAST mosaic as a THz emitter. To investigate this concept we fabricated a 1-cm² mosaic, consisting of four identically oriented DAST crystals. We characterized this crystal in terms of THz output, beam quality, and temporal waveform and found no degradation in performance when compared to a single crystal. Therefore, for a given optical energy, an optimum size emitter may be chosen to maximize THz output. We have applied for a patent on this mosaic emitter. These emitters are being used in the characterization of novel electronic materials, the development of fast electro-optic switches and the development of terahertz imaging systems for nondestructive inspection.