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J. Goodfellow

Stanford University, SIMES Institute for Materials and Energy Sciences and Stanford PULSE Institute, SLAC National Accelerator Laboratory

M. Fuchs

SIMES Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, University of Nebraska - Lincoln, mfuchs@unl.edu

D. Daranciang

Stanford University

S. Ghimire

Stanford PULSE Institute, SLAC National Accelerator Laboratory

F. Chen

Stanford University

See next page for additional authors

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Authors

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Below gap optical absorption in GaAs driven by intense, single-cycle coherent transition radiation

J. Goodfellow,^{1,2,3} M. Fuchs,^{2,7} D. Daranciang,⁶ S. Ghimire,³ F. Chen,⁸ H. Loos,⁴
D. A. Reis,^{2,3,5} A. S. Fisher,⁴ and A. M. Lindenberg^{1,2,3,*}

¹Department of Materials Science and Engineering, Stanford University, Stanford, California 94305, USA

²SIMES Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA

³Stanford PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA

⁴Accelerator Directorate, SLAC National Accelerator Laboratory, Stanford University, Menlo Park, California 94025, USA

⁵Department of Applied Physics, Stanford, California 94305, USA

⁶Department of Chemistry, Stanford University, Stanford, California 94305, USA

⁷University of Nebraska - Lincoln, Lincoln, Nebraska 68588, USA

⁸Department of Electrical Engineering, Stanford University, Stanford, California 94305, USA
[*aaronl@stanford.edu](mailto:aaronl@stanford.edu)

Abstract: Single-cycle terahertz fields generated by coherent transition radiation from a relativistic electron beam are used to study the high field optical response of single crystal GaAs. Large amplitude changes in the sub-band-gap optical absorption are induced and probed dynamically by measuring the absorption of a broad-band optical beam generated by transition radiation from the same electron bunch, providing an absolutely synchronized pump and probe geometry. This modification of the optical properties is consistent with strong-field-induced electroabsorption. These processes are pertinent to a wide range of nonlinear terahertz-driven light-matter interactions anticipated at accelerator-based sources.

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1. Introduction

Intense single-cycle light pulses at terahertz (THz) frequencies enable new means for engineering materials properties through direct manipulation of atomic and electronic degrees of freedom. In the past several years, significant progress has occurred utilizing both laser-

based and accelerator-based approaches for generating high fields in the 1-10 THz frequency range. Laser-based sources have enabled pulses with peak fields of order 1 MV/cm and energies of ~10-100 μ J at frequencies near 1 THz [1–9]. Nevertheless, many field-driven processes remain out of reach at these field strengths, and accelerator based sources, based on coherent radiation from relativistic electron beams, represent an alternative pathway towards the generation of extreme fields, reaching sizable fractions of interatomic fields in matter [10–23]. Previously, we have reported the measurement of quasi-half cycle pulses with peak fields exceeding 20 MV/cm at a center frequency of 10 THz via coherent transition radiation (CTR) at the Linac Coherent Light Source (LCLS) free-electron laser [13,24]. Here, we present first pump-probe, time-resolved measurements of dynamics induced by a CTR-based source. At these field strengths, THz pulses are intense enough to strongly modify the band structure in a wide range of solid-state materials. Accordingly, characterization of this effect is a prerequisite for subsequent observations of dynamics induced by strong fields with these characteristics. Probing below band-gap in intrinsic GaAs, we observe large amplitude (~80%) transmission decreases consistent with previous observations of strong-field-induced electroabsorption [25–27], but occurring within a largely unexplored region of light-matter interaction at frequencies ~10 THz and fields ~1 MV/cm, where the ponderomotive energy is comparable both to the band gap and to the conduction band width, in the single-cycle limit. This work additionally demonstrates a new experimental capability to carry out THz pump / broad-band spectroscopic probe measurements where both pump and probe are generated via transition radiation and therefore absolutely synchronized with respect to each other.

The ionization of solids in an AC electromagnetic field was first treated comprehensively by Keldysh [28], following previous studies in DC fields [29,30]. He showed that the familiar cases of multiphoton ionization and field induced tunneling are in fact limiting cases of a unified physical process. He also introduced the adiabaticity parameter $\gamma = \omega(m\varepsilon_g)^{1/2} / (eE)$ (later known as the Keldysh parameter) as an indicator of the dominant ionization regime for strong field measurements. Here ω and E are the frequency and amplitude of the driving field, m is the effective mass, and ε_g is the band gap of the material. This parameter is the ratio of the tunneling time to the optical period. For $\gamma \ll 1$, electrons follow the driving field adiabatically and the process reduces to the familiar case of tunneling in a quasi-static field. Conversely, for $\gamma \gg 1$ the ionization is well-described by a multiphoton absorption process in which the incident field can be treated perturbatively. The condition $\gamma \sim 1$ thus defines a transition between classical and quantum regimes, in which both field and photon effects must be considered.

2. Results

Terahertz radiation for these experiments was generated via coherent transition radiation (CTR) from 50 fs FWHM, 13 GeV, 350 pC electron bunches at 120 Hz generated by the LCLS. As each bunch passes through a thin metal foil, the discontinuity in dielectric constant leads to emission of broadband radiation at frequencies related to the temporal charge distribution of the bunch [31]. The short duration of the bunch leads to coherent enhancement of the emitted radiation in the THz regime with peak frequencies centered near 10 THz (30 μ m) and a quasi-half-cycle pulse shape of duration ~100 fs [13,24]. The radiated spectrum extends to higher frequencies through the visible range, with some optical frequencies also experiencing coherent enhancement due to microbunching in the electron beam [32]. This optical transition radiation (OTR) serves as a fully synchronized probe for time resolved experiments, and its broadband spectral content allows access to the band-edge electronic structure in a variety of semiconducting and insulating materials. Both the CTR and OTR beams are radially polarized due to the symmetry of the electron bunch. In the following we use CTR and THz interchangeably to refer to the single-cycle THz-frequency pump beam.

The transition radiation is extracted from the electron beam pipe and diverted into the experimental setup as shown in Fig. 1. The electron beam traverses through a 10 μ m thick Be foil at 45° angle of incidence, emitting transition radiation in the forward and downward (i.e.

reflected) direction. The downward radiation exits the evacuated beam pipe via a 250 μm thick, 25 mm

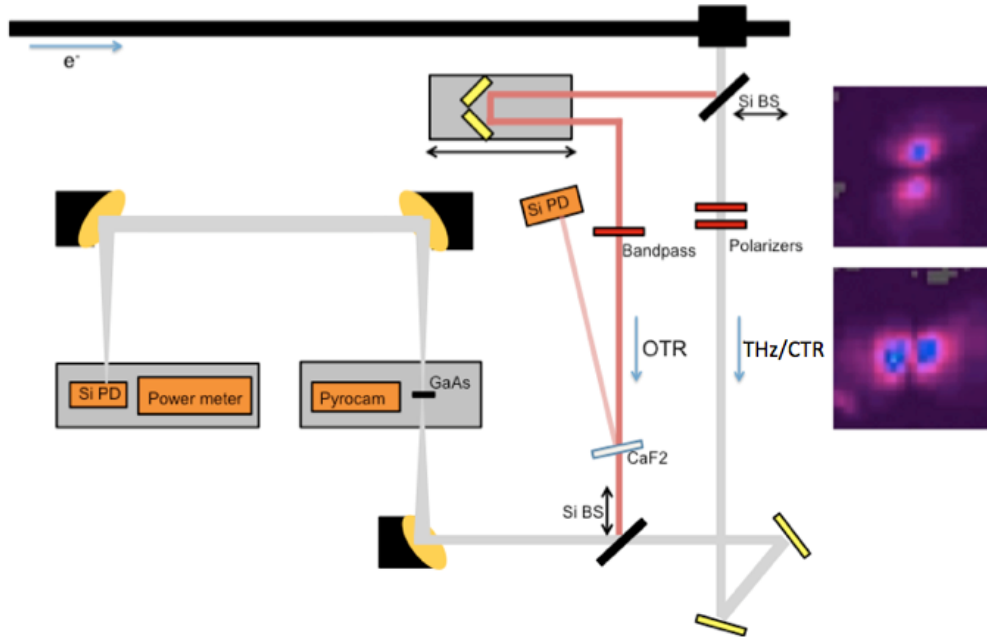


Fig. 1. Experimental setup showing extraction of terahertz pulses from electron beam and pump-probe setup. At right shows images of transmitted THz fields through a polarizer oriented vertically (top) and horizontally (bottom) showing the radial polarization of the extracted beams. Pump (THz) and probe (OTR) light are spatially and temporally overlapped as shown in a 1 mm thick GaAs crystal.

diameter diamond window and is collimated by a gold off-axis parabolic mirror with a 190 mm effective focal length. The beams are propagated through the remainder of the setup via free space optics in an enclosure purged with dry air. The pump probe setup takes the form of a Mach-Zehnder interferometer, with 1 mm thick, intrinsic Si wafers used as quasi-dichroic beamsplitters. Nonlinear THz interactions in the Si can be neglected due to the large beam size there. To perform diagnostics on the full CTR beam, the Mach-Zehnder can be bypassed by removing these beamsplitters. In this configuration the transition radiation can be routed to a thermopile power meter or a pyroelectric camera for imaging. A pair of broadband THz polarizers can be inserted into the CTR path for independent control of the polarization and field amplitude. Due to the radial polarization, the first polarizer spatially truncates the CTR profile as shown in Fig. 1. The peak field is not significantly reduced by the insertion of the polarizers when optimally aligned. For measuring the field dependence of pump probe signals, the second polarizer is rotated in order to prevent any changes to the spatial overlap of the pump and probe beams. When the silicon beamsplitters are inserted, the OTR is spectrally separated from the CTR and recombined collinearly after traversing a mechanical delay stage. The OTR passes through a thick calcium fluoride window to remove the terahertz radiation from the probe line. The reflection from the window also serves as normalization for the transmission studies. The overlap of the two beams is obtained using a pyroelectric camera.

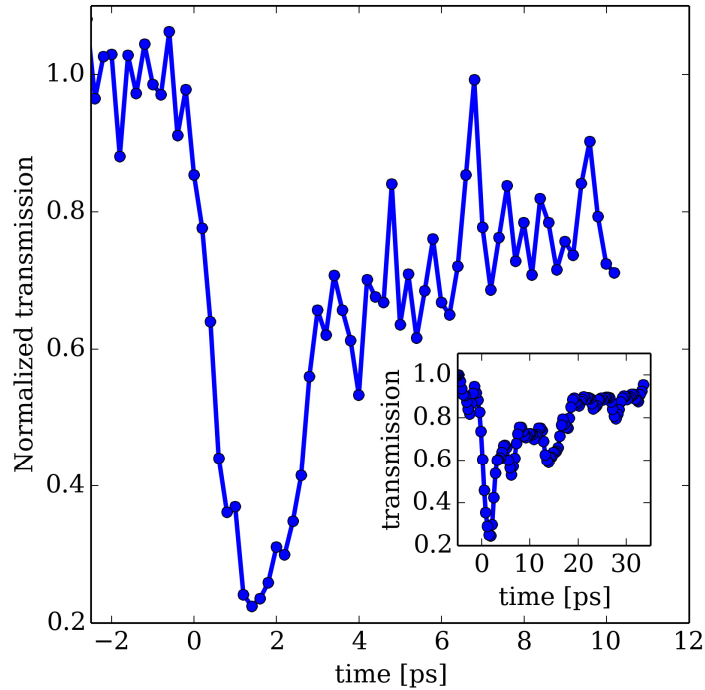


Fig. 2. Normalized THz pump / below-band-gap optical probe signal in GaAs. Inset shows long-time response.

As a first pump probe experiment, we investigated the time resolved transmission through a GaAs wafer excited by a high-field THz CTR pulse. A 1 mm thick, intrinsic GaAs sample cut in the (110) orientation is positioned in the focus of the CTR. The OTR and CTR have similar focused spot sizes of $\sim 400 \mu\text{m}$. THz polarizers in the CTR line are used to fix the spatial profile of the pump and modify the incident intensity. A bandpass filter in the OTR path restricts the probe wavelengths to a range of 750-1000 nm with energy $< 1 \mu\text{J}$ to prevent any modification of the sample's optical properties due to strong visible or IR radiation. The transmitted OTR intensity is monitored with a Si photodiode. Because light with wavelength below approximately 900 nm is strongly absorbed by the sample, we effectively probe the region from about 900-1000 nm in this measurement, limited in the long wavelength range by the band pass filter and the cut-off of the silicon diode response. A calcium fluoride window in front of the photodiode prevents any anomalous response due to the strong collinear CTR.

The results of the measurement are shown in Figs. 2 and 3. We observe an 80% reduction in the transmitted OTR intensity when the temporal overlap with the CTR pulse is optimized. Adjusting our previous field estimates for transmission through two Si wafers, different focusing conditions, and the reflectivity of the GaAs wafer, we estimate peak fields in the experiment of $\sim 1 \text{ MV/cm}$ centered at 10 THz within the GaAs sample. Previous measurements have characterized the spectral bandwidth of these pulses using a Michelson interferometer [24]. For these fields, taking the GaAs band gap of 1.43 eV and electron effective mass of $0.062m_e$, γ is approximately 0.5 and the ionization behavior is expected to be described by the dynamical Franz-Keldysh effect (DFKE) with the transmission decrease attributable to a transient redshift of the absorption edge. In the absence of the THz field, the bandwidth of the transmitted probe is approximately 0.2 eV. Since the sample is thick, we treat the transmission coefficient as a step function which goes from 100% transmissive to

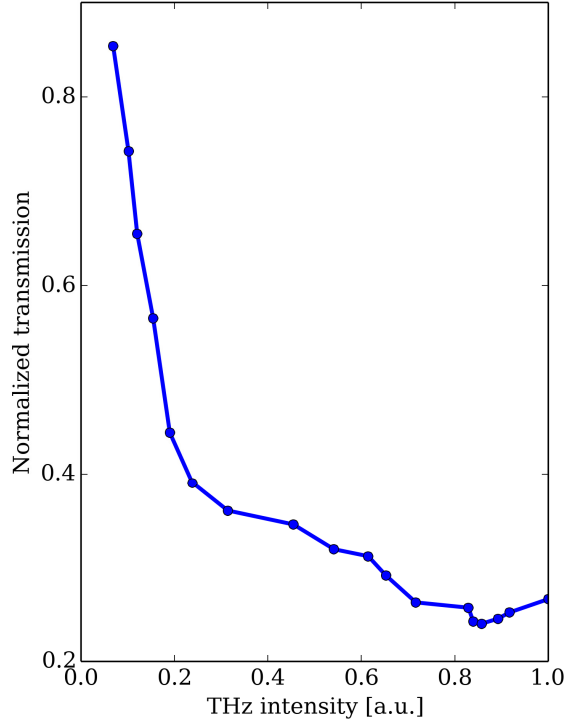


Fig. 3. Fluence dependence of nonlinear THz induced modulation at $t \sim 2$ ps after excitation.

opaque at the onset of the absorption edge. Under this approximation, a redshift of ~ 0.1 eV would be required to explain a $\sim 50\%$ transmission decrease. The induced redshift can be approximately estimated from the tunneling transition rate for photon energies below the band gap [30]:

$$\omega(E, \Omega) \propto \exp \left[-\frac{4\sqrt{2m}}{3e\hbar E} (\epsilon_g - \hbar\Omega)^{3/2} \right] \quad (1)$$

where Ω is the photon frequency. Identifying the edge position as the energy for which the exponent becomes -1 , we obtain a redshift of 0.15 eV for the peak field of 1 MV/cm in rough agreement with observations. At high intensities, the transmission decrease saturates as previously observed (Fig. 3) [25] without evidence for sample damage. Deviations in the effect at high field are not surprising given the ponderomotive energy of 1.8 eV for our experimental conditions, a value comparable to the conduction band width, and a regime in which the Bloch frequency is comparable to the probe frequency [25,33,34]. We note that the broad temporal signature of the effect is consistent with the group velocity dispersion between the THz and the optical radiation, which leads to a few picosecond difference in propagation times through the 1 mm GaAs crystal, consistent with observations. Additionally, the THz field is back-reflected at the back interface of the sample, leading to Fabry-Perot effects and replicas of the THz-driven effect at longer times (>16 ps), as shown in the inset to Fig. 2, explaining the long-lived nature of the observed signal [25].

3. Conclusion

In summary, we have studied the nonlinear THz response of intrinsic GaAs under intense THz-fields generated by coherent transition radiation. The broadband OTR from the same electron bunch was used as a dynamical probe in order to obtain jitter-free time resolution in a

pump-probe geometry. While our results are consistent with previous work at moderate field strengths, we observe novel phenomena such as the saturation of the THz effects in semiconductors at higher peak fields. Future investigations into these effects will be improved by spectrally resolving the transmitted probe in order to directly measure the field-induced distortions in the optical absorption, by measurement of thin film samples, and by extensions of this work to even higher fields, now achievable at accelerator-based sources.

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