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Wideband 15 THz response using organic electro-optic polymer emitter-sensor pairs at telecommunication wavelengths

Colin V. McLaughlin,¹ L. Michael Hayden,^{1,a)} Brent Polishak,² Su Huang,³ Jingdong Luo,^{3,4} Tae-Dong Kim,³ and Alex K.-Y. Jen^{2,3,4}

¹Department of Physics, University of Maryland, Baltimore County 1000 Hilltop Circle, Baltimore, Maryland 21250, USA

²Department of Chemistry, University of Washington, Seattle, Washington 98195, USA

³Department of Materials Science and Engineering, University of Washington, Seattle, Washington 98195, USA

⁴Institute of Advanced Materials and Technology, University of Washington, Seattle, Washington 98195, USA

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Poled polymer films are employed in terahertz emission and detection using telecommunication wavelength femtosecond pulses. Films 10–15 μm thick with electro-optic coefficients as high as 160 pm/V at 1300 nm are used to generate and sense subpicosecond pulses with continuous bandwidth up to 15 THz. Terahertz emission from a poled polymer film is compared to terahertz emission from the organic crystal DAST. The use of a poled polymer as the terahertz sensor allows the identification of DAST phonons at 1.1, 3.0, 5.3, 8.5, and 12.5 THz. © 2008 American Institute of Physics. [DOI: 10.1063/1.2909599]

The development of terahertz generation and sensing methods over the past two decades has led to techniques such as terahertz time-domain spectroscopy,¹ terahertz imaging,² and optical-pump/terahertz-probe spectroscopy³ with applications in medical, biological, and fundamental materials sciences.⁴ Many of these applications require bright, broad-bandwidth terahertz emission paired with sensors responsive to broad terahertz bandwidth. An effective scheme for obtaining bright, ultrabroadband terahertz radiation is the use of organic electro-optic (EO) polymers for terahertz emission via optical rectification and terahertz detection through EO sampling.⁵

Although emitters made from organic crystalline EO materials such as 4'-dimethylamino-N-methyl-4-stilbazolium tosylate (DAST) coupled with EO polymers can be used for broadband terahertz systems,⁶ employing EO polymer films for both terahertz emission and detection offer several advantages. Recent demonstrations of polymer materials with EO coefficients >300 pm/V provide a path to enhanced terahertz performance.⁷ High EO coefficients lead to brighter terahertz generation and more sensitive terahertz detection.⁶ When used as terahertz emitters and sensors, amorphous EO polymers do not suffer the strong dispersion and absorption associated with the lattice resonance (phonon) effect observed in EO crystals,⁸ allowing a gap-free⁵ terahertz response. In addition, molecular engineering of the EO polymer material can minimize the effects of group velocity mismatch,⁹ thereby increasing the coherence length l_c leading to bright, broadband terahertz emission. Recently, compact, cost-effective fiber laser systems generating femtosecond pulses between 1–1.5 μm have been developed.¹⁰ With the availability of compact ultrafast light sources, the need to study terahertz emitter-sensor pairs operating at telecommunication wavelengths is important for their potential use in mobile terahertz systems. In this letter, we present a broadband terahertz system utilizing an emitter-sensor pair based

on poled EO polymers operating at 1300 nm. We compare this system to one based on DAST as an emitter using our EO polymer as a sensor.

The EO chromophore AJTB203, shown in Fig. 1, was dissolved in a solution of cyclopentanone and amorphous polycarbonate (APC) with a total mass density of 24% and an EO chromophore to APC weight ratio of 28/72. The solution was cast through a 0.2 μm polytetra fluoroethylene filter onto indium tin oxide coated glass substrates and spun at 700 rpm for 1 min. Solvent removal was achieved through baking under a vacuum at 85 °C for 24 h followed by another 24 h under vacuum at 100 °C. A gold electrode was sputtered on the top surface of the polymer to allow poling of the film.¹¹ After poling, the gold was removed by a potassium iodide–iodine etch. The resulting 10–15 μm thick films have an absorption maximum at 803 nm and yield EO coefficients¹² between 100–160 pm/V at a wavelength of 1300 nm.

The AJTB203 chromophore has been specifically designed for use in terahertz applications at communication wavelengths. To prevent crystallization at the chromophore

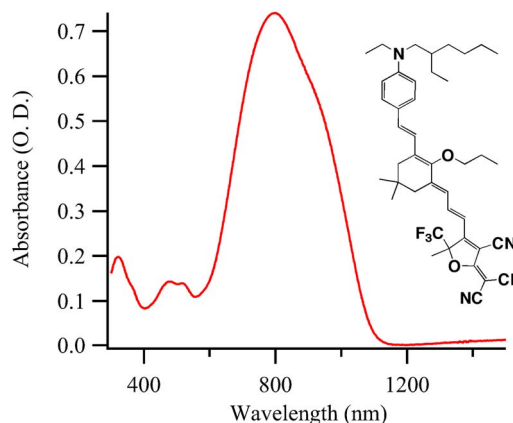


FIG. 1. (Color online) Molecular structure of the AJTB203 chromophore and absorption spectra of an AJTB203/APC thin film.

^{a)}Electronic mail: hayden@umbc.edu.

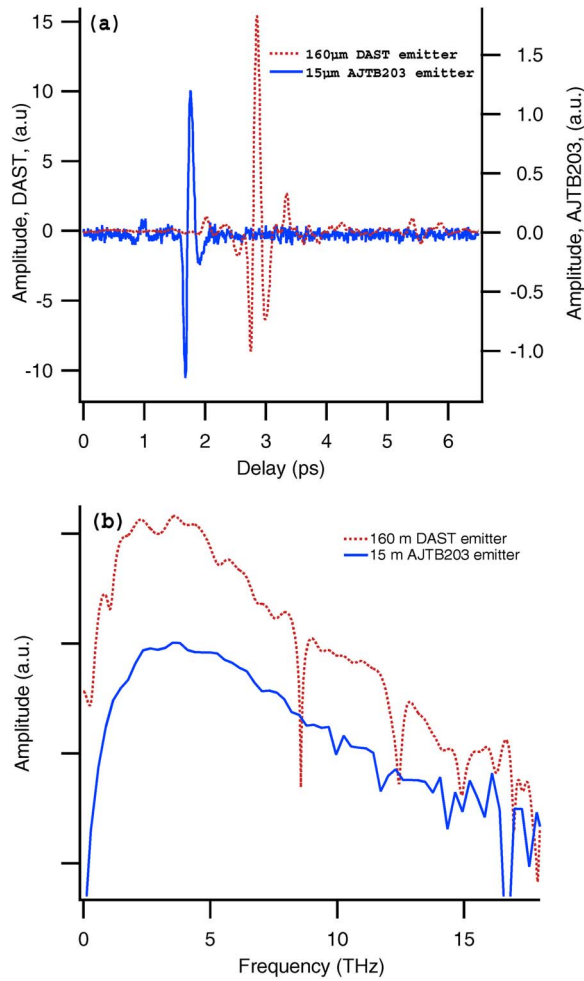


FIG. 2. (Color online) (a) Terahertz waveform from a 160 μm thick DAST emitter and a 15 μm thick AJTB203 emitter pumped at ~1300 nm and (b) their corresponding field amplitude spectra. Both waveforms were detected using an 11 μm thick AJTB203 sensor.

concentration required for large EO coefficients, the acceptor group contains variable moieties and the donor group contains an ethylhexyl group to improve solubility and prevent phase separation and chromophore aggregation. The alkoxy group attached to the isophorone portion of the conjugated bridge sterically hinders the rotation of the isophorone group within the conjugated bridge. This structurally-induced rigidity improves the planarity of the polyene bridge and thereby enhances the chromophore's nonlinearity and photochemical stability.¹³ The near-IR location of the absorption band ($\lambda_{\text{max}}=803$ nm) in the polymer allows our tunable (1200–1600 nm) optical parametric amplifier (OPA) laser to be a suitable light source for the nonresonant¹⁴ generation and detection of terahertz frequency radiation within our films. The OPA provides laser pulses with ~50 fs duration, ~30 nm bandwidth centered at a wavelength of 1300 nm with a 1 kHz repetition rate. To preserve the short pulse width of the pump laser, we use reflective optics to collect and focus the terahertz beam in our experimental setup.⁷

Figure 2(b) shows the terahertz spectra obtained from the time-domain signals in Fig. 2(a). The polymer and DAST terahertz emissions were both detected by using an 11 μm thick AJTB203/APC film having an EO coefficient of ~100 pm/V at a wavelength of 1300 nm. The signals for both emitters extend up to 15 THz and have a peak at

~3.5 THz. The amorphous polymer films, which do not possess a crystalline structure, produce broad-bandwidth, gap-free terahertz signals. In contrast, the effects of a crystalline structure are displayed in the DAST terahertz spectra, where lattice resonance-related spectral dips are visible at 1.1, 3.0, 5.3, and 8.5 THz. The broadband characteristics associated with our EO polymer sensor allow us to also identify a lattice resonance in the DAST spectrum at 12.5 THz.

The terahertz emission from a 15 μm thick polymer film is approximately an order of magnitude smaller than the emission from a 160 μm thick DAST crystal [Fig. 2(a)]. The EO coefficients of the EO polymer and DAST crystal emitters at 1300 nm are $r_{33} \sim 100$ pm/V and $r_{11} = 53$ pm/V, respectively. However, the effective nonlinear coefficient for optical rectification for terahertz generation in the polymer film,

$$d_{\text{eff}}^{\text{OR}} = \frac{2}{3} d_{33} \cos \theta_{\text{THz}} \sin \theta_{\text{opt}} \cos \theta_{\text{opt}} + \sin \theta_{\text{THz}} \left(\frac{1}{3} d_{33} \cos^2 \theta_{\text{opt}} + d_{33} \sin^2 \theta_{\text{opt}} \right), \quad (1)$$

leads to a corresponding $r_{\text{eff}} \sim 52$ pm/V because the polymer emitter must be rotated with respect to the incident pump beam in order to project a suitable component of the pump beam polarization along the poling direction of the film.¹¹ For film thicknesses less than the coherence length, the emitted terahertz electric field scales linearly with the product of the EO coefficient and film thickness.¹⁵ Increasing the thickness of the polymer will improve the terahertz amplitude since this material is reasonably well phase matched at 1300 nm. The optical group index n_g for AJTB203/APC at communication wavelengths [Fig. 3(b)] is close to the terahertz index ($n_{\text{THz}} \approx 1.65-1.75$) in our polymer systems,⁶ resulting in long coherence lengths given by $l_c = c / (2\nu_{\text{THz}}[n_g(\lambda) - n_{\text{THz}}])$. As shown in Fig. 3, we estimate l_c in the AJTB203/APC system to be 65–120 μm at 10 THz and 35–60 μm at 20 THz. Polymer film stacking and waveguiding will take advantage of the long coherence length and are topics of future research. Since both terahertz emission and sensing scale linearly with thicknesses up to the coherence length, the overall system response scales as the product of the increase in emitter-sensor thickness. By increasing the thickness of our emitter and sensor from 15 and 11 μm, respectively, to 100 μm each, we should realize an increase in the terahertz response by a factor of over 60 over a bandwidth up to 10 THz. The absorption of the optical pump beam in a 100 μm thick film will not be significant. As Gunter and co-workers^{16,17} pointed out, the effective generation and sensing lengths will be reduced as the terahertz absorption increases. For the thicknesses considered here and for terahertz absorption ~15 cm⁻¹ in the polymer films,¹⁸ terahertz absorption will result in <10% reduction of the absorption-free predicted enhancement in terahertz system response. The rapid change in group index [Fig. 3(b)] between 1300 and 1600 nm in an AJTB203/APC film will allow us to maximize coherence length by changing the pump beam wavelength to match n_g and n_{THz} . Taking $n_{\text{THz}} = 1.7$, the calculated coherence lengths [Fig. 3(c)] show that a 1300 nm pump is not optimally index matched. With a 1500 nm pump, we estimate that $l_c > 240$ μm at 10 THz and > 120 μm at 20 THz. Thus, by increasing the thickness of the emitter and sensor to 240 μm and using a 1500 nm pump, we predict an increase in response by a factor of over 300 over a bandwidth of up to 10 THz as compared to the

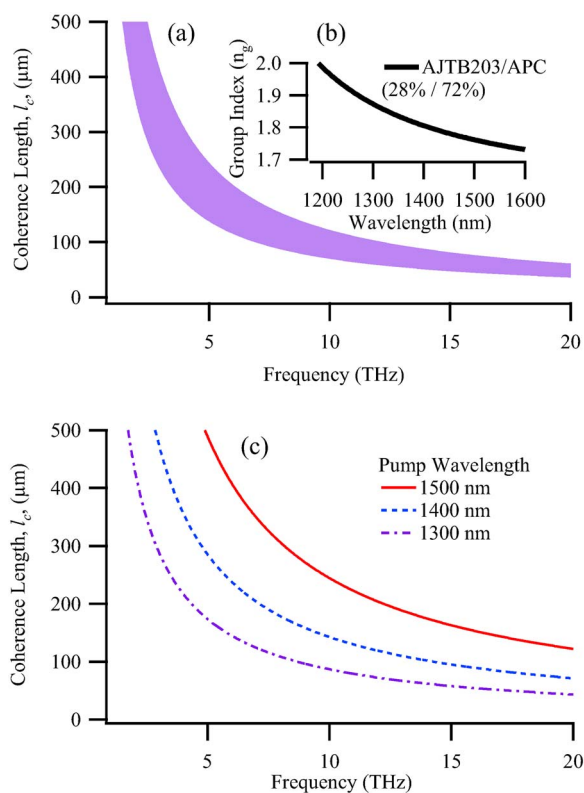


FIG. 3. (Color online) (a) Terahertz-pump coherence length in the AJTB203/APC system using a 1300 nm pump beam. The shaded area is bounded by THz indices of 1.65 and 1.75. (b) The experimentally determined optical group index (n_g) yields a value of 1.87, 1.80, and 1.76 at 1300, 1400, and 1500 nm, respectively. (c) The predicted coherence length ($n_{\text{THz}}=1.7$) increases at longer pump wavelengths.

current case. An additional factor of 10 improvement should be possible by utilizing the latest EO polymers with $r_{33} > 300$ pm/V.⁷

In conclusion, using a 1300 nm laser, we report broad, gap-free 15 THz emission and detection from an AJTB203/

APC emitter-sensor pair and a 15 THz response with multiple spectral gaps from a DAST crystal emitter utilizing an EO polymer sensor.

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