Efficient, wideband THz emission from thin electro-optic polymer films

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Abstract: We use a 3 μm thick poled polymer film to generate THz radiation that is greater than that emitted from a 1000 μm thick crystal of ZnTe. The resonantly enhanced EO coefficient is estimated to be above 350 pm/V. In a non-resonant experiment, we demonstrate THz emission up to 20 THz in an electro-optic polymer film.

OCIS codes: (190.4710) Optical nonlinearities in organic materials; (190.7110) Ultrafast nonlinear optics

1. Introduction

Terahertz applications, ranging from medical[1] and near-field imaging[2] to spectroscopy of biological and chemical agents[3], are becoming more widespread. Efficient emitters and sensors are needed to improve the performance of all of these THz systems. Photoconductive dipole antennae, electro-optic (EO) crystals, and EO polymers can be used as THz emitters and sensors. Due to their good phase matching conditions and high electro-optic coefficients, EO polymers are promising materials to increase the spectral response as well as the signal to noise ratio of THz systems.

2. Materials

We are studying organic guest-host polymer composites consisting of various kinds of chromophores and polymer matrices. Chromophores with extended π-electron systems and strong electron donors and acceptors exhibit large molecular hyperpolarizabilities and large dipole moments. These molecular features help to contribute to large EO coefficients in poled composites of these materials. Previously, we reported efficient EO polymers for THz applications in guest-host mixtures of 40% DCDHF-6-V and 60% Poly(bisphenol A carbonate-co-4,4'-(3,3,5-trimethylcyclohexylidene)diphenol carbonate) (APC)[4]. We refer to this composite as DAPC. Here we report resonance enhanced THz generation near the absorption maximum in a polymer composite composed of 20% CF_3FTC and 80% APC (Figure 1) and wideband emission from DAPC films.

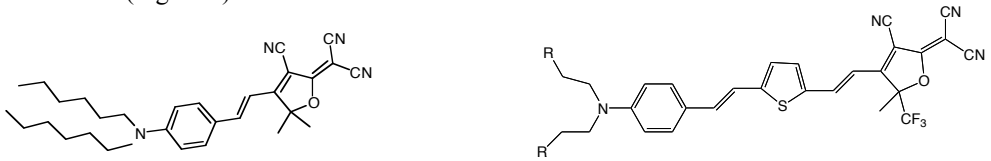
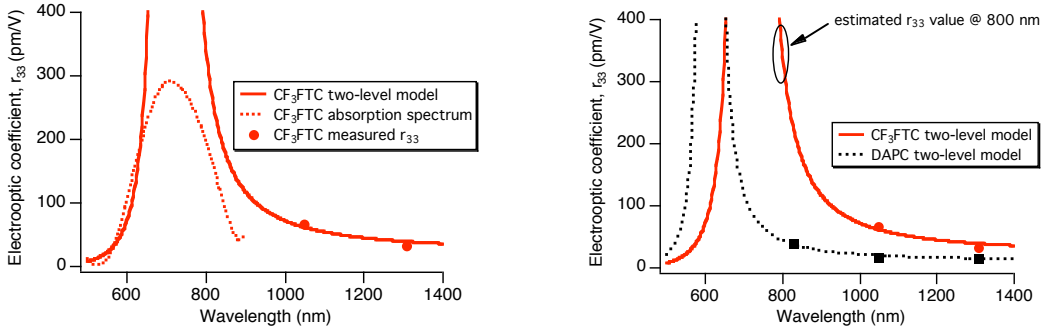


Fig. 1. Chromophores: DCDHF-6-V, CF_3FTC.

In molecular nonlinear materials, the macroscopic second order nonlinear susceptibility is directly proportional to the second order molecular polarizability. By applying time dependent perturbation theory in the two-level model approximation[5], the dispersion of the second order susceptibility can be expressed as: $\chi^{(2)}_{333}(\lambda) \propto \lambda^2(3\lambda^2 - \lambda^2_{ag})/3(\lambda^2 - \lambda^2_{ag})^2$, where λ_{ag} is the wavelength of the transition from the ground state *g* to the 1st excited state *a*. A sharp resonance in the material nonlinear properties is expected at the transition wavelength λ_{ag}. In Fig. 2, the dispersion of the EO coefficient of a polymer composite together with its absorption is presented.

3. Results and Discussion

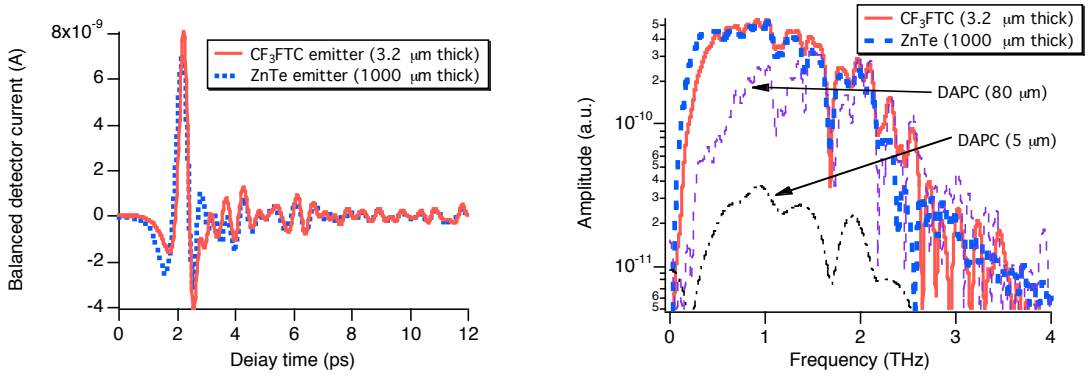
The EO coefficient in a 3 μm thick film of 20% CF_3FTC/80% APC was measured using a reflection geometry[6] at 1310 nm and 1047 nm. This material has an absorption maximum near 710 nm; therefore, in the THz experiment, with the pump beam at 800 nm, we are working near the absorption maximum causing a resonant increase of the EO coefficient.



(a) (b)

Fig. 2. (a) Absorption spectrum (arbitrary units) and the two-level model prediction of the dispersion in the electrooptic coefficient of the CF_3FTC/APC composite. (b) Two level model predictions along with the measured r_{33} for CF_3FTC/APC (circles) and DAPC (squares). The ellipse represents our best estimate of r_{33} for the CF_3FTC/APC composite.

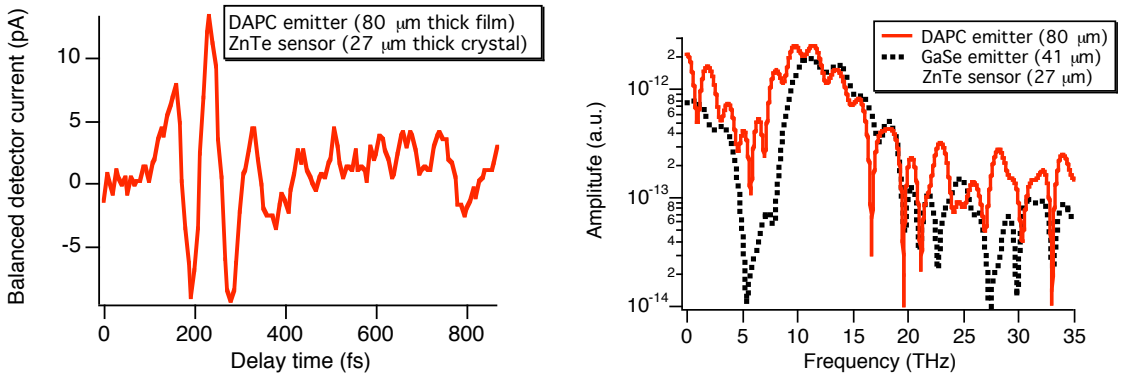
In Fig.3, we make a direct comparison of the THz signal generated via optical rectification from a 1000 μm thick ZnTe crystal and thin films of two different EO polymer composites. The amplitude of the THz field generated from a 3.2 μm thick film of the 20%CF_3FTC/80%APC composite is greater than the corresponding field from a 1000 μm thick crystal of ZnTe, which is over 300 times thicker.



(a) (b)

Fig. 3. (a) Time domain THz signals generated via optical rectification in a 3.2 μm thick polymer film of CF_3FTC/APC (solid line) compared with a signal from a 1000 μm thick crystal of (110) ZnTe (dashed line). (b) Corresponding frequency domain signals. In addition, signals from 5 μm and 80 μm thick DAPC polymer films are shown for comparison. All experiments were performed in air and the sensor for all the experiments was a 2000 μm thick crystal of (110) ZnTe.

Due to the strong material absorption of CF_3FTC at 710 nm, we cannot measure the EO coefficient of this composite at 800 nm, however, with the two-level model approximation, we estimate this coefficient to be near 350 pm/V at 800 nm. In contrast, since the DAPC composite has an absorption maximum near 600 nm, its electrooptic coefficient is not as enhanced (see dotted curve in Fig. 2b) as that from the CF_3FTC material. The THz performance for equivalent thickness of DAPC and CF_3FTC shown in Fig. 3b are proportional to their respective electrooptic coefficients. Even though we are unable to directly measure the electrooptic coefficient of the CF_3FTC composite at 800 nm due to its absorption, we can use the THz data in Fig. 3b to estimate its electrooptic coefficient at the THz pumping wavelength of 800 nm by comparing the THz signals from the 5 μm DAPC film to that from the 3.2 μm CF_3FTC film. Comparing those two THz amplitudes at 1.47 THz (4.1/0.25), scaling the amplitude ratio to account for the film thickness ratio (3.2/5), and using the directly measured value of 38 pm/V for the DAPC, we estimate that the electrooptic coefficient of CF_3FTC/APC is 400 pm/V, in good agreement with the prediction from the two-level model (solid curve in Fig. 2b).



(a) (b)

Fig. 4. (a) Time domain THz signals generated via optical rectification in a 80 μm thick polymer film of DAPC and detected by a 27 μm thick crystal of (110) ZnTe. (b) Corresponding frequency domain signal (solid line). For comparison, a THz signal generated from a 41 μm thick crystal of GaSe (dotted line) is also shown. The GaSe trace has been linearly scaled to normalize the two traces to the same incident pumping power. The experiment was performed in air.

In Fig. 4 we show some recent results that explore the wide bandwidth potential of these polymer composites. In these experiments we used a 14 fs Ti:Sapphire oscillator as the optical rectification pumping source and a 27 μm thick crystal of ZnTe as the THz sensor. We compared the THz emission from an 80 μm film of DAPC with that from a 41 μm thick crystal of GaSe. The signal-to-noise in this experiment (Fig. 4a) is low due to the need to reduce the input optical power to ~ 10 mW to prevent optical damage to the polymer films in the tight focusing geometry of the experiment. In Fig. 4b, the GaSe amplitude is scaled accordingly so that we might compare the amplitudes of the two materials for the same pumping powers. Both materials appear to have similar spectra although the polymer does not have the strong absorption near 5-8 THz associated with phonons in GaSe, but does show some absorption in that region presumably due to the 5.3 THz phonon in ZnTe. The polymer does show a strong absorption near 17 THz and another near 19 THz. The structural basis for these absorptions is being modeled using molecular dynamics and will be reported elsewhere.

In summary, we have shown that two different electrooptic polymer composites can efficiently emit THz radiation. On a per pump photon basis, our CF_3FTC material is nearly 300 times as efficient as ZnTe based on a comparison of their thicknesses. However, pumping these films near the absorption maximum causes bleaching of the material resulting in a diminished THz signal upon successive scans. In addition, we must operate at low pumping power densities to prevent damage to some of our polymer films. The issue of optical damage is certainly an impediment to application development for these materials at 800 nm. However, we believe that widely available and affordable THz systems for commercial applications may need to operate at 1.0 – 1.55 μm where more compact and potentially cheaper, ultra-fast fiber lasers operate. In this wavelength range, our materials show great promise as their electrooptic coefficients are still very large but the chromophores experience no linear absorption. Our near term goal is to develop efficient THz emitters and sensors suitable for use with 1.0-1.55 μm compact, ultra-fast fiber lasers[7].

4. References

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