Ref: SPIE Proceedings Vol. 7601: Terahertz Technology and Applications III, Laurence P. Sadwick; Creidhe M. M. O'Sullivan, Editors, 76010C Date: 18 February 2010

Stimulated Emission of Terahertz Radiation from Electro-optic Dendrimer

Anis Rahman
Applied Research and Photonics, Inc., 470 Friendship Road, Suite 10, Harrisburg, PA 17111
*e-mail: a.rahman@arphotonics.net

ABSTRACT

Dendrimers are a polymeric nanomaterial whose molecular size varies from ~4 nm to ~13 nm based on its generation; within a generation these molecules are highly monodispersed. As the molecular size increases, its number of end groups increase too, resulting in an increased number of available sites for attaching foreign molecules. Generation 3 dendrimer was doped with a commercially available chromophore and formed thin film via spin coating. The cured dendrimer was corona poled. The resulting stand alone film exhibits an electro-optic coefficient r_{33} ~130 pm/V leading to a high $\chi^{(2)}$-dendrimer. This EO dendrimer was used for terahertz generation via difference-frequency technique (DFG). Two CW diode lasers were used to pump the EO dendrimer emitter to provide a combined pump power of 5.5W that generated a continuous wave DFG power of ~3.4 mW.

Keywords: Electro-optic Dendrimer, Difference-Frequency Generation, Terahertz Radiation, Corona Poling, Electro-optic Rectification, Milliwatt CW terahertz.

1. INTRODUCTION

A compact source of terahertz (THz) radiation that can deliver relatively high power is important for many practical applications including THz spectrometry (TS) and imaging. Of the several methods that can be employed for THz generation, the electro-optic route is the most promising. An advantage of using electro-optic (EO) route is the inherent power scalability, because, pump-THz conversion is not limited either by emission saturation or by heat dissipation. Also, both continuous wave (CW) and pulsed terahertz radiation may be obtained via EO route.

Currently photoconductors, quantum cascade lasers, as well as polymers are being researched for compact terahertz source. These sources are often limited to microwatts of power [1]. Reported data from a *GaP* based scalable source [2] shows that ~6.7 μW terahertz power is obtainable at a pump power of ~10W that has a quadratic dependence on the pump power from a femto second pulsed laser. While some technology such as free electron lasers [3] and particle accelerator [4] have been used for higher terahertz power, these systems are huge in cost and size, and require a dedicated facility.

The electro-optic rectification (EOR) process has been used to generate terahertz radiation from inorganic [5]–[7] and polymeric materials [8]–[11]. Most of the polymer based approaches use side chain nonlinear optical (NLO) polymers. Sinyukov et al. [8–9] used PMMA with a dye (Lemke) that generated radiation over ~0–3 THz window. This material's EO coefficient is 25–30 pm/V. The authors found that, because of low dielectric constant, it exhibits a better phase matching and generates higher power compared to *ZnTe* crystal. However, they also point out a low glass transition temperature (~85°C) of this polymer and photo-degradation of the dye molecules from continuous exposure to 510 nm laser leading to a short duration of stable power. Cao et al. [11] reported waveguide geometry with metal and acrylate clad, and disperse red 1-methyl methacrylate (DR1-MMA) core, for a phase-matched terahertz generation by EOR. They found that both the pump beam and the generated terahertz radiation propagate in the fundamental mode of their waveguide, extending out to a length of 3 mm at a relatively low optical pump power and better coherence length compared to *ZnTe*. However, in all cases the average power has been at a low level that may not be able to probe many molecular interactions involved in practical applications. Also, a femto-second pulsed laser must be used for EOR process. Such pulsed lasers are expensive and bulky, thus hindering the compactness of the source.

Difference-Frequency Generation (DFG). Simon et al. [12] have investigated the feasibility of mixing two single-mode diode lasers in $AgGaS_2$ to generate tunable terahertz radiation. They reported a scheme where the radiation from each diode-laser was collimated, spatially overlapped and focused into $AgGaS_2$ crystal. Their first diode-laser was emitting at 690 nm with power of 10.1 mW and second diode-laser had a power of 1.93 mW emitting at 808 nm. The combined pump generated 3.3 nW of far infrared radiation. Shi et al. [13] have reported terahertz generation by DFG method in GaSe crystal. These authors generated an average terahertz power of 0.43 μW corresponding to a peak power of 0.53 mW when the pump average power is 895 mW and the peak power is 1.12 kW.

2. DENDRIMER AS A TERAHERTZ MATERIAL

Unlike side chain polymers, dendrimer [14] is a class of star burst polymers with nearly spherical, monodispersed molecular architecture. Dendrimer has a lower dielectric constant (ε~2.5) that favors phase matching of the pump and generated THz radiation. Dendrimer also has a higher EO coefficient (EOC); chromophore doped dendrimer film exhibits r_{33} ~130 pm/V [15], but may reach to 300 pm/V or higher by optimizing the doping and poling protocol. Dendrimer offers a superior combination of the factors ε and r_{33} that are also important for optimizing pump-THz conversion. Being a non-absorbing system, dendrimer does not suffer from photo-degradation at the pump wavelengths, leading to a stable source of terahertz radiation.

In the present work, dendrimer is poled to align the dipole moments for optimum electro-optic properties by creating a uniaxial polar material. Poling of dendrimer and the electro-optic measurement of poly-(amidoamine organosilicon) (PAMAMOS) dendrimer (Dendritech, Inc., Midland, Michigan) has been previously reported [15]. Chromophore doped PAMAMOS dendrimer film was prepared on a glass slide coated with aluminum and subsequently corona poled [16] using a setup described elsewhere [15]. Essentially, a needle electrode was placed above the film to apply a high voltage across the thickness of the film while the poling current was monitored as a function of applied voltage at an elevated temperature near the glass transition point. The dendrimer was doped with alizarin (Alfa Aeser) chromophore. The average film thickness was 100 μm and corresponding poling field strength was 6.6×10^5 V/cm. Completion of dipole alignment was indicated by a saturation of the poling current (see ref. 15 for details). Under these conditions, the dipole alignment angle, θ is ~0, and the second order susceptibility is given by $\chi^{(2)} = n\beta f\langle Cos(\theta)\rangle = n\beta f$, where, n is the density of active molecules, β is the average hyperbolarizability, and f is the local field factor [17].

Refractive index (RI) of both poled and unpoled films was measured with a prism coupler (Metricon 2010) as a function of wavelength at room temperature. The measurements were done in the TE mode so that the propagation of probing wavelength was perpendicular to the dipole alignment. A systematic difference in refractive index, $n(\lambda)$, resulted due to poling [15]. The index difference $|\Delta n|$ is utilized to compute r_{33} from the Pockels effect, $|\Delta n| = \frac{1}{2} n^3 r_{33} E_p$ where E_p is the poling field strength; r_{33} value of ~130 pm/V was obtained at 633 nm falling to ~90 pm/V at 1553 nm. Thus dendrimer's EOC is significantly higher than any inorganic material used in terahertz generation such as $LiNbO_3$ (~32 pm/V) and GaP (~1.1 pm/V). This is also higher than many reported NLO organic materials such as DR1-MMA (~5–30 pm/V [8]). The NLO dendrimer is, therefore, expected to be a suitable nanomaterial for terahertz generation.

3. DIFFERENCE-FREQUENCY GENERATION

The difference frequency generation (DFG) is a stimulated emission process, described by a non-linear polarization [18],

$$P(\omega_1 - \omega_2) = 2\chi^{(2)} E_1 E_2^*, \quad (1)$$

where, E_1 and E_2 are the field strengths and ω_1 and ω_2 are the frequencies of the input lasers. Here, the generated frequency is the difference of those of the applied pump-fields, thus is tunable (see Fig. 1). Difference-frequency generation can be described in terms of the photon energy-level diagram of Fig. 2. In order to satisfy the conservation of energy requirements, a photon at the higher input frequency ω_1 must be destroyed and a photon at the lower input frequency ω_2 must be created for every photon that is created at the difference-frequency $\omega_3 = \omega_1 - \omega_2$. The lower-frequency field in effect is amplified by the process of difference-frequency generation, thus, it is a stimulated amplification process. According to the photon energy-level description of the DFG, the atom (molecule) first absorbs a photon of frequency ω_1 and jumps to the highest virtual (unstable) level. This level decays by a two-photon emission

process that is stimulated by the presence of the ω_2 field. However, two-photon emission can occur even if ω_2 was not applied. Therefore, ω_2 input field acts similar to an idler frequency of a parametric oscillator that is in effect filtered out (see Fig. 4).

For the input pump, two fiber-coupled single-mode diode lasers beam were overlapped in a beam-combiner. Two Amrel PPS-10710 power supplies were used to provide the input current at fixed bias to the diode lasers. Fig. 3 shows the measured output power of the overlapped beam as a function of the input current. Fig. 4 shows an experimental setup to measure the average (CW) terahertz power from DFG. Here, overlapped beam was normally incident on the dendrimer emitter. The detector (Astral AC2500) was fitted with a spectral filter that truncates wavelength up to 1500 nm and also a 3.3 mm thick polyethylene filter such that no light can enter the detector without passing through the filter combination.

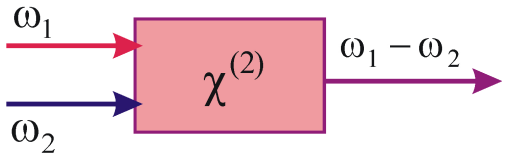

Fig. 1. Interaction geometry for difference frequency generation

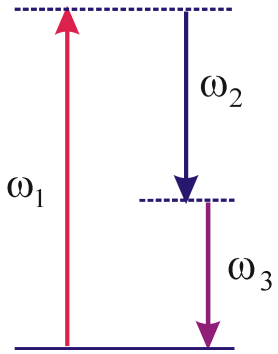

Fig. 2. Energy-level description of difference frequency generation.

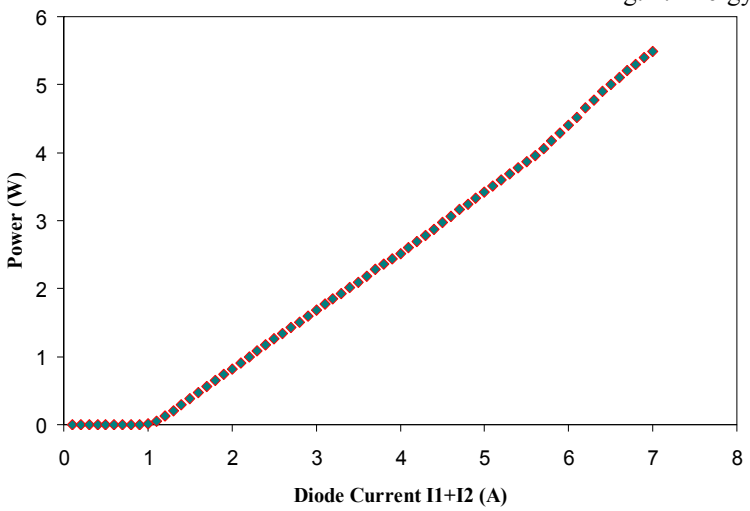

Fig. 3. Measured power of overlapped beam vs. combined input current to diode1 + diode2.

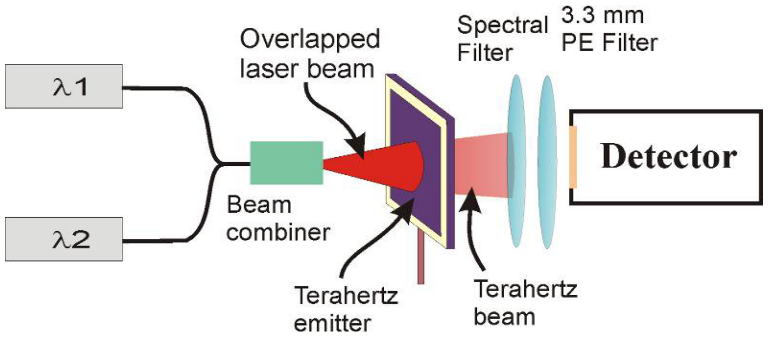

Fig. 4. Experimental setup for terahertz measurement via difference-frequency generation.

Fig. 5 shows the measured DFG average power as a function of the total pump power from diode1+diode2. As seen from Fig. 5, 3.4 mW of terahertz power was obtained from the dendrimer emitter at a combined pump power of 5.5 W. To our knowledge, this is the highest average CW power from any compact terahertz sources, both polymeric and inorganic. This is easily tunable and higher terahertz power may be obtained by tuning the pump power. In Fig. 4, the center wavelength of diode1 is λ1 = 975 nm, and the center wavelength of diode2 is λ2 = 940 nm. Therefore, it is expected that the source of Fig. 4 should yield a broadband terahertz radiation, $\nu_{THz} \propto \nu_1 - \nu_2 \sim 30$ THz.

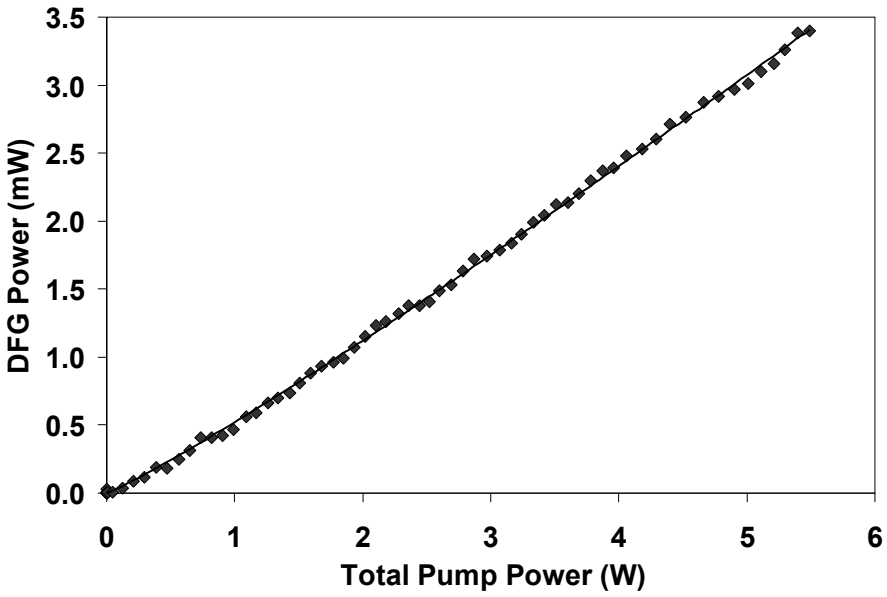

Fig. 5. Measured DFG average power vs. total pump power from (diode1+diode2).

SUMMARY

A high EOC dendrimer has been used to generate terahertz radiation via difference-frequency technique. The dendrimer emitter was pumped by two fiber coupled single-mode diode lasers. The diodes beams were overlapped that was tuned by input current to each diode, thus producing a tunable terahertz source. Combined total pump power from both diodes was varied from zero to a maximum of 5.5 W that generated a CW average terahertz power of 3.4 mW. Since the center wavelength of diode1 is λ1 = 975 nm, and the center wavelength of diode2 is λ2 = 940 nm, it is expected that the source should yield a broadband terahertz radiation of up to 30 THz. This source is suitable for practical terahertz applications such as time-resolved terahertz spectrometry and terahertz imaging.

REFERENCES

[1] Stuart, R.A., C.C. Wright, and A. Al-Shammaa. "Present Status of the Compact EM THz Source," *3rd EMRS DTC Technical Conference* – Edinburgh, A11 (2006).

[2] Chang, G., C.J. Divin, C-H. Liu, S.L. Williamson, A. Galvanauskas, and T.B. Norris, "Power scalable compact THz system based on an ultrafast Yb-doped fiber amplifier," *OPTICS EXPRESS*, 14(17), 7909 (2006).

[3] Ramian, G., "The new UCSB free-electron lasers," *Nucl. Instrum. Methods Phys. Rev*. A 318, 225 (1992).

[4] Carr, G. L., M.C. Martin, W.R. McKinney, K. Jordan, G.R. Neil, and G.P. Williams, "High-power terahertz radiation from relativistic electrons," *Nature*, 420, 153 (2002).

[5] Yang, K.H., P.L. Richards, and Y.R. Shen, "Generation of Far-Infrared Radiation by Picosecond Light Pulses in LiNbO3," *Appl. Phys. Lett*. 19, 320 (1971).

[6] Auston, D.H., A.M. Glass, and A.A. Ballman, "Optical Rectification by Impurities in Polar Crystals," *Phys. Rev. Lett.*, 28, 897 (1972).

[7] Gordon, D.F., A. Ting, I. Alexeev, R. Fischer, P. Sprangle, C.A. Kapetenakos, and A. Zigler, "Tunable, high peak power terahertz radiation from optical rectification of a short modulated laser pulse," *OPTICS EXPRESS*, 14(15), 6813 (2006).

[8] Sinyukov, A.M. and L.M. Hayden, "Generation and detection of terahertz radiation with multilayered electro-optic polymer films," *Optic Letts*., 27(1), 55 (2002).

[9] Sinyukov, A.M., M.R. Leahy, L.M. Hayden, M. Haller, J. Luo, A.K-Y. Jen, and L.R. Dalton, "Resonance enhanced THz generation in electro-optic polymers near the absorption maximum," *App. Phys. Letts*., 85(24), 5827 (2004).

[10] Hayden, L.M., A.M. Sinyukov, M.R. Leahy, J. French, P. Lindahl, W.N. Herman, R.J. Twieg, and M. He, "New Materials for Optical Rectification and Electrooptic Sampling of Ultrashort Pulses in the Terahertz Regime," *Journal of Polymer Science*: Part B: Polymer Physics, 41, 2492 (2003).

[11] Cao, H., R.A. Linke, and A. Nahata, "Broadband generation of terahertz radiation in a waveguide," Optics Letts., 29(15), 1751 (2004).

[12] Simon, U., Miller, C.E., Bradley, C.C., Hulet, R.G., Curl, R.F., and Tittel, F.K., "Difference-frequency generation in $AgGaS_2$ by use of single-mode diode-laser pump sources," Optics Letters, 18(13), 1062 (1993).

[13] Shi, W., Leigh, M., Zong, J., and Jiang, S., "Single-frequency terahertz source pumped by Q-switched fiber lasers based on difference-frequency generation in GaSe crystal," Optics Letters, 32(8), 949 (2007).

[14] Tomalia, D.A., "Birth of a new macromolecular architecture: Dendrimers as quantized building blocks for nanoscale synthetic organic chemistry," *Aldrichimica Acta*, 37(2), 39 (2004).

[15] Rahman, A. "Dendrimer waveguide based high-efficiency terahertz source," *Terahertz Technology and Applications*, edited by Kurt J. Linden, Laurence P. Sadwick, Proc. of SPIE Vol. 6893, 689302 (2008).

[16] Mortazavi, M.A., A. Knoesen, S.T. Kowel, B.G. Higgins, and A. Dienes, "Second-harmonic generation and absorption studies of polymer-dye films oriented by corona-onset poling at elevated temperatures," *J. Opt. Soc. Am*. B, 6, 733 (1989).

[17] Gadret, G., Kajzar, F. and Raimond, P., "Nonlinear Optical Properties of Poled Polymers," SPIE Vol. 1560 *Nonlinear Optical Properties of Organic Materials* IV, 226 (1991).

[18] Boyd, R.W., "Nonlinear Optics," 2nd Ed., *Academic Press*, New York, (1992).

[19] Chen, Q., M. Tani, Z. Jiang, and X.-C. Zhang, "Electro-optic transceivers for terahertz-wave Applications," J. *Opt. Soc. Am*. B, 18(6), 823 (2002).

[20] Ahn, J., A.V. Efimov, R.D. Averitt, and A.J. Taylor, "Terahertz waveform synthesis via optical rectification of shaped ultrafast laser pulses," *OPTICS EXPRESS*, 11(20), 2486 (2003).

[21] Reid M. and R. Fedosejevs, "Quantitative comparison of terahertz emission from (100) InAs surfaces and a GaAs large-aperture photoconductive switch at high fluences," *Applied Optics*, 44(1), 149 (2005).

[22] Xu, J.Z. and X-C. Zhang, "Optical rectification in an area with a diameter comparable to or smaller than the center wavelength of terahertz radiation," *Optics Letts*., 27(12), 1067 (2002).