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## **FINAL REPORT**

**Title: "Electro-optic modulation and mirrorless lasing using unique organic materials and single-crystal films" (4/00 - 4/03)**

Our accomplishments in this project include: i) preparation of single crystal films of specific organic nonlinear optical materials including molecular salts such as DAST, ii) measurement of electro-optic coefficients, iii) demonstration of electro-optic modulation with a modulation depth of 80 % at a field of 4 V/  $\mu\text{m}$  for a single-pass device (using a 3  $\mu\text{m}$  thick film), iv) demonstration of high-speed modulation with a speed up to 1.5 GHz. v) demonstration of highly efficient (with 40% conversion) mirrorless lasing, vi) measurement and use of the short pulses (40 ps) generated by mirrorless lasing and vii) second harmonic generation in specific single crystal films to determine d-coefficients. These results show that the original objectives of the project have been accomplished.

## FINAL REPORT

**Title: "Electro-optic modulation and mirrorless lasing using unique organic materials and single-crystal films" (4/00 - 4/03)**

Our accomplishments within the past three years include: i) preparation of single crystal films of organic nonlinear optical materials including molecular salts such as DAST, ii) fabrication of specific single-pass electro-optic devices based on these films (~ 3 microns thick), iii) demonstration of electro-optic modulation with a modulation depth of 80 % at a field of 4 V/  $\mu\text{m}$  for a single-pass device, iv) demonstration of high-speed modulation with a speed up to 1.5 GHz. v) demonstration of highly efficient mirrorless lasing, vi) measurement and use of the short pulses generated by mirrorless lasing and vii) second harmonic generation using single crystal films to determine d-coefficients. More details of these accomplishments are discussed in the following.

Larger area (~ 1  $\text{cm}^2$ ) single-crystal films of DAST have been prepared by a modified shear method at a higher temperature. Electro-optic modulation at 4 kHz using single-crystal film of DAST has been reported earlier. The measurement was made in the transverse configuration with the light beam propagating perpendicular to the film while electric field was applied in the plane of the film - along the dipole axis. This was a field-induced birefringence measurement. A large modulation ~ 20% was observed at a low field of 1 V/ $\mu\text{m}$  for a 3  $\mu\text{m}$  thick film ( $\lambda = 720 \text{ nm}$ ). The magnitude of  $r_{11}$  at 720 nm is 530 pm/V.

Modulation at 100 MHz has been demonstrated using such a thin film using the same configuration. The set-up essentially involves a polarizer, a thin film, a compensator, an analyzer, and then a detector in series. The device is simple in construction. The 100 MHz results have shown exceptional signal-to-noise ratio, comparable to that of waveguide devices.

Recently, measurements on DAST films have been extended to a higher frequency - up to 1.5 GHz. We have observed excellent signal-to-noise ratio at these high frequencies as well. The voltage applied was only ~ 1 volt and the observed signal-to-noise ratio was comparable to that of

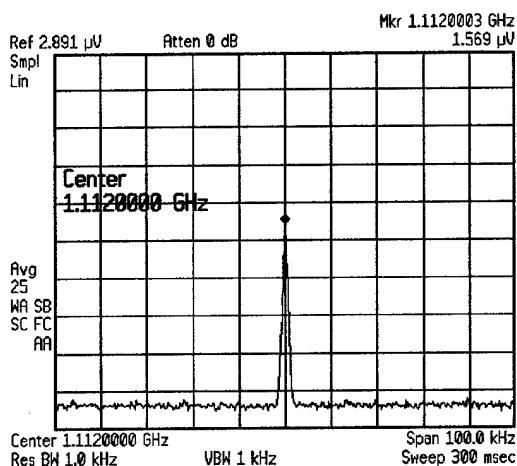


Fig.1 Electro-optic modulation in a 3  $\mu\text{m}$  thick DAST film at 1.11 GHz

guided-wave electro-optic modulators. The measurement principle was the same as before (field-induced birefringence), with special care taken to reduce radiation noise at higher frequencies. Results of such a measurement at 1.11 GHz are shown in Fig.1. The wavelength at which these results were obtained is 750 nm. These are unique results, can be obtained only for single-crystal films with excellent optical quality. Loss is low ( $\leq 1$  dB) in such a device configuration. The magnitude of  $r_{11}$  at 750 nm is 445 pm/V.

Single-crystal films have been prepared by modified shear method for two important organic materials that include: 3-methyl-4-methoxy-4'-nitrostilbene (MMONS) and 8-(4'-acetylphenyl)-1,4-dioxo-8-azaspiro[4.5]decane (APDA). Films of areas larger than  $1\text{cm}^2$  have been prepared. Characterization of the films was performed using polarized optical microscopy, optical absorption measurements and x-ray diffraction. The optical microscopic studies have shown excellent optical uniformity of the films. The crystallographic orientations of the MMONS and APDA films as determined by x-ray diffraction are [100] and [001] respectively. Therefore, in the case of MMONS, the dipole axis lies parallel to the film plane. This orientation is because of the interaction (hydrogen bonding) between the polar groups at both ends of the molecule with the hydrophilic substrate surface. In the case of APDA the molecules orient perpendicular to the substrate surface. This is because APDA is an amphiphilic molecule with only one polar group at the end that interacts with the substrate surface. Measurement of the

second order susceptibilities (d-coefficients) of the MMONS films has been completed. This was performed by detailed polarization selective SHG

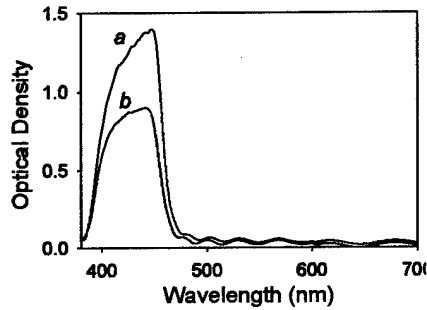


Fig. 2

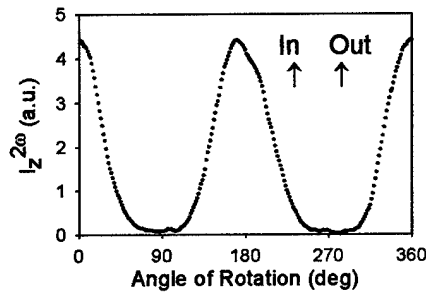


Fig. 3(a)

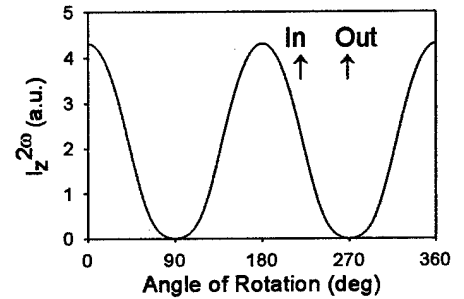


Fig. 3(b)

Fig. 2 Absorption spectra with polarization parallel (a) and perpendicular (b) to the dipole axis in the single-crystal thin-film of MMONS.

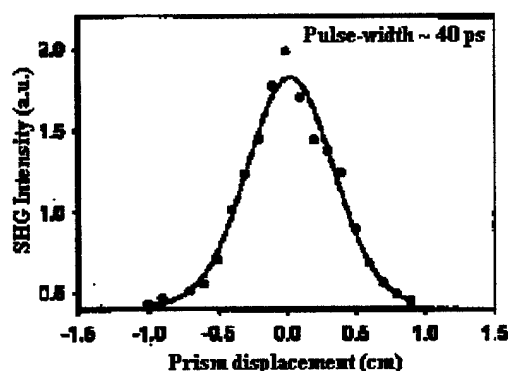
Fig. 3 Second-harmonic intensity as a function of the angular rotation of the film about the beam axis with parallel polarization configuration: (a) experimental results, and (b) calculated results.

using 100 ps pulses at  $1.06\mu\text{m}$  as the fundamental. The magnitudes of the d-coefficients are:  $d_{33} = 195 \pm 10 \text{ pm/V}$  and  $d_{24} = 75 \pm 5 \text{ pm/V}$ . These are very large magnitudes of d-coefficients considering the fact that MMONS is transparent down to 480 nm. Electro-optic measurements of the films have shown that  $r_{33}$  is about 50 pm/V at  $1.06 \mu\text{m}$  – the magnitude is consistent with the d-coefficient. Both type I and type II phase-matched propagation directions have been identified in the crystal and the type II phase-matching direction lies parallel to the film. Therefore, guided-wave phase-matched SHG should be possible using these films.

For APDA thin films, the standard transverse configuration of the SHG measurements that we have developed for thin-films has not been effective since the dipole-axis is perpendicular to the substrate. Therefore, measurements with the fundamental beam incident at an angle has been utilized. The second order optical measurements have provided further evidence supporting that the molecules of APDA

are oriented perpendicular to the film surface because of the amphiphilicity of the molecules. The measured  $d$ -coefficient at  $1.06 \mu\text{m}$  is  $d_{33} = 54 \text{ pm/V}$ . The material has a broad window of transparency (down to  $400 \text{ nm}$  in wavelength).

Laserlike emission without mirrors with the highest known efficiency (40%) has been reported for an organic molecular salt (SPCD). Such high efficiency occurs even though the photoluminescence efficiency is very low ( $< 0.3\%$ ). The excitation wavelength was at  $532 \text{ nm}$  and the emission occurred at  $620 \text{ nm}$ . Temporal measurement on and using pulses of the laserlike emission from styrylpyridinium cyanine dye (SPCD) has been made. The pulse-width of the laserlike emission measured by autocorrelation is  $40 \text{ ps}$ . These pulses at  $620 \text{ nm}$  were used as a probe in the time-resolved measurement of the spectrally narrowed emission in SPCD. The results show that the gain decays faster than the pulse-width of the probe ( $40 \text{ ps}$ ). The results also show that this emission can be used as inexpensive short-pulse lasers for specific applications. The pulse-width of the emitted beam at  $620 \text{ nm}$  was measured by background-free autocorrelation technique. A BBO crystal was used for this measurement. The fundamental pulses at  $620 \text{ nm}$  divided into two were focused at an angle on the BBO crystal. The intensity of the second harmonic at  $310 \text{ nm}$  that appeared at the center was measured after filtering as a function of the delay between the two fundamental pulses leading to the autocorrelation signal. The measured pulse-width of the emitted beam is  $40 \text{ ps}$ . These pulses were subsequently used as a probe in a pump-probe measurement of the SPCD solution. In that experiment, a pump beam ( $80 \text{ ps}$ ) at  $532 \text{ nm}$  was used along with  $40 \text{ ps}$  laserlike emission pulses as the probe. The experiment with the delay between these two pulses provided information about how fast the gain decays subsequent to excitation. The results show that the gain decays faster than the pulse-width of the probe ( $40 \text{ ps}$ ). As these results show, the mirrorless laserlike emission can produce picosecond pulses that are useful in conducting optics experiments.



Autocorrelation trace of mirrorless laser-like emission from SPCD solution for pumping at  $532 \text{ nm}$ .

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