Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl17

Application of Nonlinear Organic Materials to Femtosecond Light-Pulse Technology

M. Yamashita [a] & K. Torizuka [a]

[a] Laser Section, Optoelectronics Division, Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba, Ibaraki, Japan, 305

Version of record first published: 04 Oct 2006.

To cite this article: M. Yamashita & K. Torizuka (1990): Application of Nonlinear Organic Materials to Femtosecond Light-Pulse Technology, Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics, 182:1, 157-162

To link to this article: http://dx.doi.org/10.1080/00268949008047797

Mol. Cryst. Liq. Cryst., 1990, Vol. 182A, pp. 157–162
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

Application of Nonlinear Organic Materials to Femtosecond Light-Pulse Technology

M. YAMASHITA and K. TORIZUKA

Laser Section, Optoelectronics Division, Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba, Ibaraki, Japan, 305

(*Received May 15, 1989; in final form June 30, 1989*)

Two experimental studies are presented illustrating applications of organic materials of high nonlinear refractive index to femtosecond light-pulse technology. One is intracavity pulse-compression of a colliding-pulse mode-locked dye laser with the addition of nonlinear organic materials into a saturable absorber solution. Another is femtosecond pulse compression external to the laser using nonlinear organic fibers. On the basis of the experimental results, the present problems and the optical properties that nonlinear organic materials should satisfy are discussed.

Keywords: nonlinear organic materials, femtosecond pulse compression

1. INTRODUCTION

Recent studies of highly nonlinear organic materials permit their use as optical devices (crystals, fibers, waveguides and thin films for harmonic generators, modulators and optical switches).[1] In the field of ultrashort-pulse laser technology, nonlinear inorganic materials such as fused-silica glass fibers of relatively low nonlinear refractive index $n_2 (= 5.5 \times 10^{-16}\ \mathrm{cm^2/W})$ are utilized for femtosecond pulse compression.[2] In a femtosecond pulsed dye laser, self phase-modulation (SPM) due to the nonlinear refractive index (NRI) in a dye solvent of low n_2 plays an important role in the generation of shorter pulses[3] and of soliton-like pulses.[4]

Therefore, it is expected that the application of ultrafast, highly nonlinear organic materials (with third-order nonlinearities originating from π-electron hyperpolarizability) to ultrashort pulse technology can permit more effective control of femtosecond pulses of low peak power (pulse compression, continuum generation, soliton propagation, sub-terahertz modulation, and optical switching[5]). In this paper, two experimental studies of femtosecond pulse compression utilizing nonlinear organic materials are presented.[6,7]

2. INTRACAVITY PULSE COMPRESSION

First the principle of femtosecond-pulse compression in a laser cavity is briefly described.[8,9] When a nonlinear organic material is inserted into the cavity of a

femtosecond, colliding-pulse, mode-locked dye laser (CPM laser), intracavity SPM occurs and consequently the width of the laser spectrum is broadened by up-chirp. Subsequently, when the up-chirp is compensated by negative group-velocity dispersive (GVD; $\ddot{k}(\omega) \equiv \partial^2 k(\omega)/\partial\omega^2$, where $k(\omega)$ is the wavelength number) elements such as $\lambda_0/4$ multilayer dielectric mirrors of double stacking[10] or Brewster-prism sequences,[3] the pulse becomes quasi-transform limited and the pulse duration is shortened to near the reciprocal of the spectral width. Furthermore, when the values of SPM and GVD are changed, N = 2 or 3 soliton-like pulses with a period of several thousands cavity-round-trips may appear.[4,11]

The experiment was carried out using a CPM (R6G + DODCI) laser with a simple cavity configuration composed of seven $\lambda_0/4$-multilayer dielectric mirrors without any additional elements for chirp compensation (Figure 1-a). Up-chirp was compensated by the negative GVD of two of the mirrors, M_1 and M_6[12]. The dispersion was fine tuned by adjusting the incident angle to the mirrors M_6 and M_7. M_6 was an angle-dependent dispersive mirror, while M_7 was an angle-independent mirror of negligible dispersion.

Measurements of the pulse duration and of the spectrum were initially carried out for the CPM laser with only 7×10^{-3} mol/l of DODCI absorber. The cavity-mirror dispersion, cavity alignment, and pump power were carefully adjusted, while the fast-scanned autocorrelation trace and pulse spectrum were monitored on oscilloscopes. The duration of the generated pulse, which had an asymmetrical spectrum, was 73 fs, as shown in Figure 1-b. Next, 1.1×10^{-2} mol/l of paradimethylamino-β

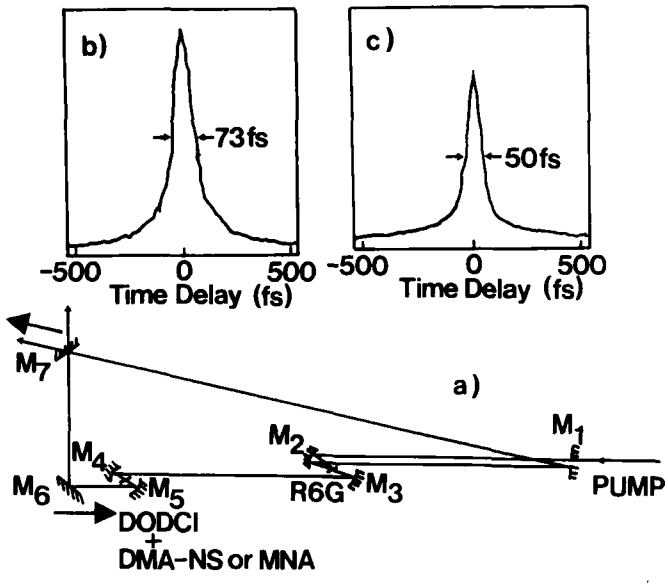

FIGURE 1 a) Cavity configuration of the cavity-mirror chirp-compensated CPM laser. The two arrows represent the directions of the simultaneous movement of mirrors M6 and M7 for dispersion adjustment. b) and c) are autocorrelation traces from the CPM laser without and with addition of 1.1×10^{-2} mol/l DMA-NS, respectively.

nitrostyrene (DMA-NS)[13], serving as the nonlinear organic material, was dissolved in the DODCI solution. At this concentration n_2 of the DMA-NS solution is 5.3×10^{-15} cm^2/W, 18 times larger than that of the solvent, ethylene glycol (EG). Consequently, the pulse duration was reduced to 59 fs. After adjustment of the cavity-mirror dispersion, the pulse duration was further reduced to 50 fs (Figure 1-c) and the spectral width was broadened from its initial value of 9.1 nm to 14.1 nm.

The intracavity dispersion (second-order phase dispersion $\ddot{\phi}(\omega) \equiv \partial^2\phi(\omega)/\partial\omega^2$, where $\phi(\omega) = -k(\omega)l$) dependence of the pulse duration generated from the CPM laser with the DMA-NS addition was measured. The value of $\ddot{\phi}(\omega)$ necessary for minimum pulse duration increased from 1.8×10^{-28} s^2 to 2.6×10^{-28} s^2 when the DMA-NS was added.[10,12] This difference is due to upchirp by the addition of the concentrated DMA-NS, and is in agreement with a theoretical result.[8]

The same intracavity pulse-compression experiment was carried out for the CPM laser with a thinner film (flared-stream jet nozzle) of a solution of a mixture of DODCI and 3.8×10^{-2} mol/l 2′-4′-methyl nitroaniline (MNA; $n_2 = 3.0 \times 10^{-15}$ cm^2/W). The results showed that pulses as short as 53 fs were shortened to 48 fs.

For generation of shorter pulses, the optical properties that the nonlinear organic material should satisfy are as follows: 1) an n_2 greater than about 100 times that of EG ($n_2 = 3.0 \times 10^{-16}$ cm^2/W),[1,9], 2) ultrafast time-dependent index changes by electronic hyperpolarizability, 3) high solubility for the EG or for a mixture of EG and benzylalcohol, 4) no absorption around the lasing wavelength region, and 5) good photochemical stability in the solution.

Computer calculations[7] of nonlinear propagation of femtosecond pulses predict that as the n_2 of a nonlinear organic material becomes larger[1,14] and the input pulse duration becomes shorter, the nonlinear organic material must be made thinner, i.e. in the form of a thin film or thin crystal. In addition, the positive GVD of its material causes the induced up-chirp to become quasi-linear. Therefore, it is assumed that utilization of such a highly nonlinear thin element at the Brewster angle in the cavity, independent of a saturable absorber, gives interesting results not only for generation of shorter pulses, but also for studies of formation processes of higher-order soliton-like pulses.[4,11]

3. ORGANIC FIBERS FOR PULSE COMPRESSION

The conventional technique of femtosecond-pulse compression by dispersive SPM in a single-mode glass fiber external to a laser has some disadvantages. Because glass fibers have a small n_2, high amplification of femtosecond-laser pulses by a factor of $\sim 10^5$ by means of a complex amplifier system is needed. This leads to a worsening of the temporal amplitude stability and spatial beam homogenity of the pulses as well as to a remarkable reduction in the pulse repetition rate. In addition, this lowering of the pulse repetition rate makes it difficult to adjust precisely the pulse duration measuring apparatus and each optical component of the pulse compressor. Utilization of a nonlinear organic material with a large n_2 and a small $\ddot{k}(\omega)$ as the fiber core allows these disadvantages to be overcome. It should be

noted that the fiber length for compression of femtosecond input-pulses is shorter than a few centimeters. Thus, it is easy to fabricate the organic fiber.

The pulse compression ratio for 100 fs input pulse passing through a liquid organic-fiber plus grating pair was numerically calculated using a nonlinear Schrödinger equation.[15] The results are shown in Figure 2. The calculated result for the case of glass fibers is also shown. The single-mode liquid organic-fiber consists of a 1 cm-long fused-silica capillary tube with a 4 μm-radius MNA-solution core. The MNA is dissolved in a mixture of nitrobenzene (NB) and ethanol (ET). The single-mode condition is satisfied by adjustment of the volume mixture ratio of those solvents (MNA:NB:ET = 0.077 : 0.415 : 0.508). The n_2 and positive $\ddot{k}(\omega)$ are $65.1 \times n_{2\cdot GF}$ and $5.1 \times \ddot{k}(\omega)_{GF}$ at 640 nm, respectively ($n_{2\cdot GF}$ and $\ddot{k}(\omega)_{GF}$ are the NRI and GVD of glass fibers). Figure 2 suggests that the same pulse-compression as in the glass fiber case is achieved in the case of liquid organic fiber at 1/10 the input peak-power.

The experiment using the liquid organic fiber was carried out for both amplified pulses of the CPM laser as well as for its direct output pulses. Measured spectra of the output pulses from the liquid organic fiber and of the 4 kW, 70 fs input pulses, respectively, are shown by solid and dotted lines in Figure 3-a. Similar spectra for glass fibers for 70 kW, 70 fs incident pulses are shown in Figure 3-b. Comparison between them shows that spectral broadening of the femtosecond pulses due to dispersive SPM in the liquid organic fiber occurs efficiently at a peak-power 10 times lower than that of the glass fiber case. This agrees with the calculated results.

Similar results were also measured for an MNA crystal multi-mode fiber. In

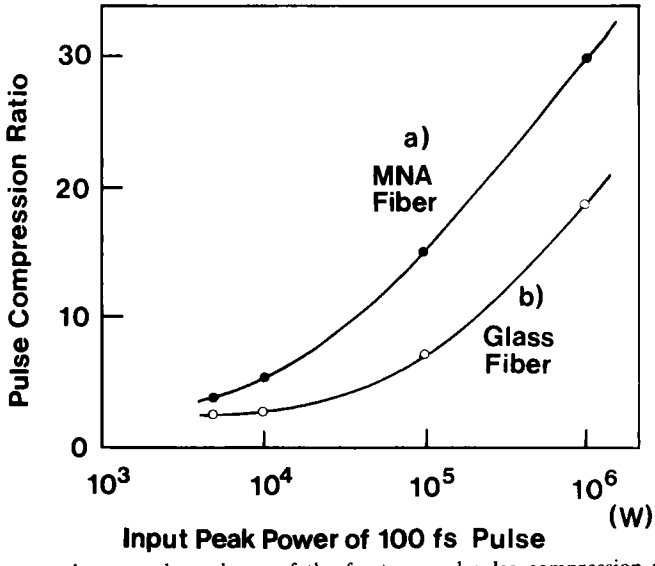

FIGURE 2 Input peak-power dependence of the femtosecond-pulse compression ratio for a) the single-mode MNA liquid fiber and b) the single-mode fused-silica glass fiber. The duration of the input pulse at 630 nm was 100 fs.

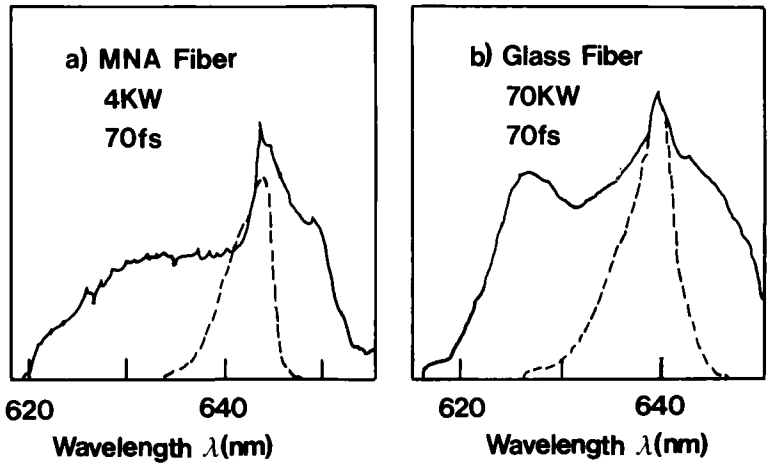

FIGURE 3 Output spectra from a) the single-mode MNA liquid fiber and b) the single-mode fused-silica glass fiber (solid lines). Spectra of the 70 fs incident pulse with 4 and 70 kW peak-powers are also shown, respectively, by dotted lines, for the liquid fiber (a) and the glass fiber (b).

those cases, unfortunately, it was impossible to measure the duration of the pulses after passing through the chirp compensation grating pair because the low pulse repetition rate of 10 Hz yielded an inadequate signal to noise ratio for the auto-correlation measurement.

Pulse durations after liquid organic-fiber propagation and chirp compensation by the grating pair were measured to be 210 and 60 fs, respectively, for the 500 W, 75 fs pulses directly output by the laser (92.5 MHz repetition rate). This result is shown in Figure 4. It can be seen that these pulses were too weak to achieve significant compression. The corresponding calculated durations were 149 and 74 fs. The small difference between the experimental and calculated results may be

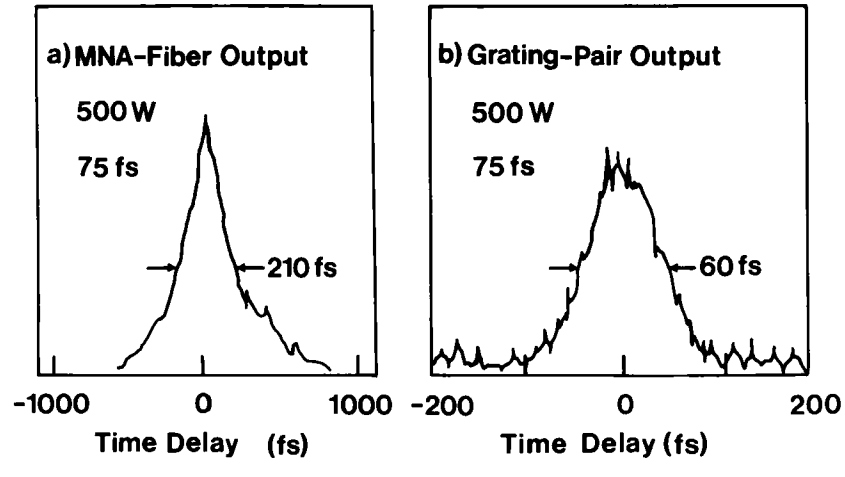

FIGURE 4 Autocorrelation traces after passing through (a) the single-mode MNA liquid fiber and (b) the grating pair for the 500 W, 75 fs incident pulse from the CPM laser.

due to the fact that the input pulse in the experiment was slightly up-chirped, but further experimentation is necessary. We are presently improving the fiber-coupling efficiency and laser output power.

From these results it is concluded that, for further pulse compression, nonlinear organic materials should satisfy the following properties: 1) In addition to the properties 2), 4) and 5) mentioned in the previous section, n_2 should be larger than that of glass fibers by a factor of $\sim 10^3$,[1,14,16], 2) $\ddot{k}(\omega)$ should be no more than five times that of glass fibers, and 3) Crystalline nonlinear organic materials should be used with the end face of the crystal fiber being in a cleavage plane[16] so as to reduce coupling losses. In addition, a low linear refractive index n_0 is better for fabrication of single-mode organic fibers.

In conclusion, the utility of the application of nonlinear organic materials to femtosecond-pulse compression internal and external to a laser has been demonstrated. In addition the optical properties that the nonlinear organic materials should satisfy have been presented.

References

1. For example, D. S. Chemla and J. Zyss, "Nonlinear Optical Properties of Organic Molecules and Crystals," Academic Press, Inc., Tokyo, 1987.
2. R. L. Fork, C. H. Brito Cruz, P. C. Becker and C. V. Shank, *Opt. Lett.*, **12**, 483 (1987).
3. J. A. Valdmanis, R. L. Fork and J. P. Gordon, *Opt. Lett.*, **10**, 131 (1985).
4. F. Salin, P. Grangier, G. Roger and A. Brun, *Phys. Rev. Lett.*, **56**, 1132 (1986).
5. For example, W. Kaiser, "Ultrashort Laser Pulses and Applications," Springer-Verlag, Berlin, 1988.
6. M. Yamashita, K. Torizuka and T. Sato, *Opt. Lett.*, **13**, 24 (1988).
7. M. Yamashita, K. Torizuka, T. Shiota and T. Sato, in "Ultrafast Phenomena VI," (ed. T. Yajima, K. Yoshihara, C. B. Harris and S. Shionoya), Springer-Verlag, Berlin, 1988.
8. H. A. Haus and Y. Silberberg, *IEEE J. Quantum Electron.*, **QE-22**, 325 (1986).
9. O. E. Martinez, R. L. Fork and J. P. Gordon, *J. Opt. Soc. Am.*, **B2**, 753 (1985).
10. M. Yamashita, M. Ishikawa, K. Torizuka and T. Sato, *Opt. Lett.*, **11**, 504 (1986).
11. F. Salin, P. Grangier, G. Roger and A. Brun, *Phys. Rev. Lett.*, **60**, 569 (1988).
12. M. Yamashita, K. Torizuka and T. Sato, *IEEE J. Quantum Electron.*, **QE-23**, 2005 (1987).
13. J. L. Oudar, *J. Chem. Phys.*, **67**, 446 (1977).
14. T. Kobayashi, H. Ohtani and K. Kurokawa, *Chem. Phys. Lett.*, **121**, 356 (1985).
15. W. J. Tomlinson, R. H. Stolen and C. V. Shank, *J. Opt. Soc. Am.*, **B1**, 139 (1984).
16. P. Kerkoc, Ch. Bosshard, H. Arend and P. Günter, *Appl. Phys. Lett.*, **54**, 487 (1989).