Evaluation of Laser-Induced–Damage Threshold in Saturated and Unsaturated Nematic Liquid Crystals Between 600 fs and 1.5 ns at 1053 nm

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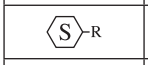
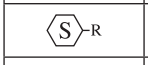
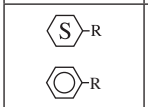
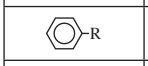
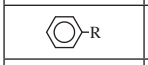
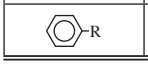
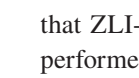
A wide-ranging series of liquid crystal (LC) materials, which included compounds with saturated (cyclohexane) and unsaturated (benzene) carbon rings (Table I), were selected to explore the effect of varying degrees of π-electron delocalization and electron density on their laser-induced–damage threshold (LIDT). This work provides baseline measurements on the damage threshold of LC's as a function of their chemical structure, and it extends the currently limited available knowledge at subnanosecond pulse lengths. This information provides insight into the damage-initiation mechanisms in LC's and guidance for the possible implementation of future applications in high-power and/or peak intensity systems. Furthermore, this work can provide insight into the design of new materials such as polymer and glassy LC materials.

Table I: Nematic liquid crystals used in this study.

	Name	Supplier	Absorption Edge
	1550C	Dabrowski^	294 nm
	MLC-2037	Merck	306 nm
	ZLI-1646	Merck	324 nm
	PPMeOB/PPPOB	LLE*	345 nm
	5CB	EMB	377 nm
	E7	EMB	385 nm

Materials are designated as saturated  and unsaturated in the first column. Note that ZLI-1646 has a mixture of compound types. Transmission measurements were performed on a Perkin Elmer Lambda-900 spectrophotometer. LC materials were in the isotropic state during measurement. Here, the absorption edge is defined at $T = 98\%$.

^Isothiocyanate compound synthesized by M. Dabrowski, University of Warsaw.

*A 60/40 mixture of two phenyl benzoate ester compounds used on the OMEGA laser and synthesized at LLE.

Both 1-on-1 (single shot per test site) and *N*-on-1 (up to ten shots at 0.1 Hz, with the fluence ramped until damage occurs) damage testing were performed to determine the corresponding LIDT. Because the LC mesophase is fluid, a test site is considered damaged upon any visual change, as observed with an *in-situ* microscope system. The difference between pre-exposure and post-

exposure images is used to identify such changes. Damage-induced changes (tiny spot or bubble-like features) were observed as a change in scattered light in a micron-scale area and typically redissolved or migrated on a time scale dependent on laser fluence and fluid viscosity. The number density and size of these features increased with increasing fluence above the damage threshold. Additional details on sample preparation and testing protocol are found in Ref. 1.

Damage-threshold data were acquired at six pulse lengths (τ): 600 fs, 2.5 ps, 10 ps, 50 ps, 100 ps, and 1.5 ns at 1053 nm. Both the 1-on-1 and *N*-on-1 LIDT values are plotted in Fig. 1 as a function of each material's UV-absorption edge. Brackets identify the saturated, unsaturated, and mixed materials. In this study, data for saturated and unsaturated materials are fairly easily differentiated, and the partially saturated material behaves more like a fully saturated material (at least under 1053-nm irradiation). Damage thresholds of three common LC materials (E7, 5CB, and ZLI-1646), which were reported in 1988 (using nanosecond laser pulses at 1053 nm) (Ref. 2), were remeasured and found to be higher. This increase is attributed to significant improvements in the chemical purity of commercial LC compounds. Early measurements of the conjugated compound 5CB and its saturated analog ZLI-S-1185 (Ref. 3) are extended in the present work by exploring additional materials and pulse durations from subpicosecond to nanosecond. Of special significance are the data at 1.5 ns, where the LIDT values of saturated LC's approach those of bare fused silica.[4]

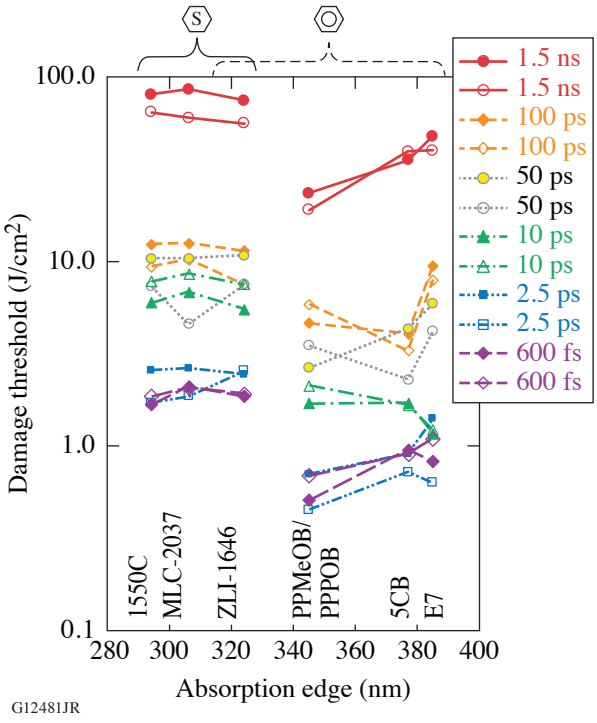


Figure 1
The 1-on-1 (open symbols) and *N*-on-1 (closed symbols) damage thresholds plotted as a function of the UV absorption edge. Materials with completely saturated or mostly saturated carbon rings have absorption edges <330 nm, with correspondingly higher damage thresholds.

The *N*-on-1 LIDT results exhibit a power dependence on the pulse length τ^x where $x \sim 0.5$, as shown in Fig. 2. A similar pulse-length dependence is observed in both dielectrics[4] and biological materials,[5] although in both cases the $\tau^{0.5}$ dependence extends only into the range of tens of picoseconds. This pulse-length dependence is attributed to thermal diffusion effects that govern the damage-initiation process, especially that of defects or defect states, which leads to free electrons and ionization of the material. In the range of tens of picoseconds, multiphoton ionization starts to contribute to electron production, and in the subpicosecond range, multiphoton ionization becomes the dominant process. At this time, we consider the fact that LC damage thresholds at pulse lengths <50 ps still follow the $\tau^{0.5}$ trend reasonably well as coincidental.

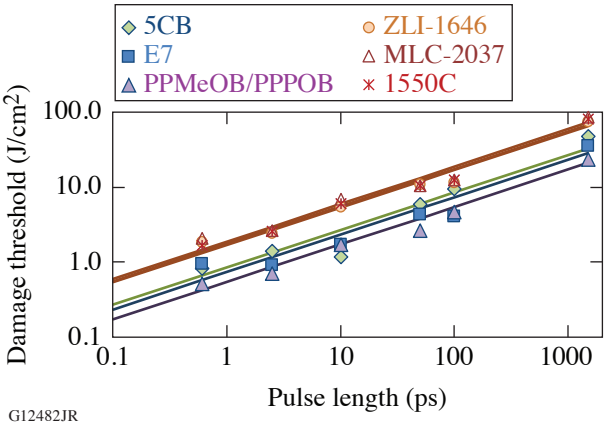


Figure 2
N-on-1 LIDT values for saturated and unsaturated LC materials are shown as a function of pulse length. A fit based on $\tau^{0.5}$ is shown for each material. R^2 values range between 0.91 (E7) and 0.97 (PPMeOB/PPPOB, 1550C, and ZLI-1646).

The absorption mechanisms, which lead to laser-induced damage, are largely dependent on the electronic structure of the material, intrinsic (LC orientation and domain boundaries) and extrinsic (impurities, substrate defects or inclusions) defects, and the laser parameters. The electronic structure in LC materials is generally known, involving a singlet ground state (S0) and excited singlet (S1, S2,…Sn) and triplet states.[6,7] The absorption spectra measurements suggest that, under 1053-nm laser irradiation, the unsaturated materials require three-photon absorption for the S0 ⟶ S1 transition, while the saturated materials require four-photon absorption. This difference in the order of the absorption process required to generate excited-state electrons is clearly captured by the difference in the damage threshold between the two types of materials, where the saturated materials have 2× to 3× higher damage threshold across all pulse lengths tested.

This research has reported damage-threshold fluences for a series of saturated and unsaturated nematic LC materials for pulse lengths between 600 fs and 1.5 ns at 1053 nm. Saturated materials always have higher damage resistance, although the pulse-length–dependent behavior varies somewhat for the two different types of material. Damage mechanisms are still under investigation, but current results point toward the presence of both multiphoton absorption and excited-state absorption.

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