Investigation of Parameters Governing Damage Resistance of Nematic Liquid Crystals for High-Power or Peak-Intensity Laser Applications

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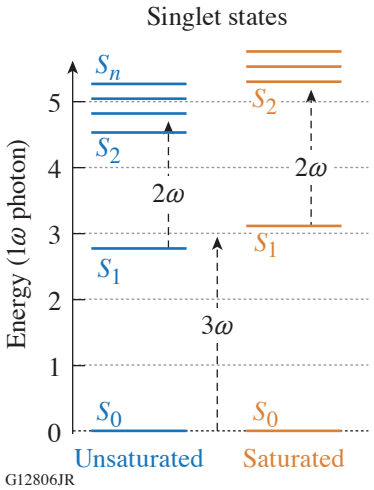
The damage resistance of saturated and unsaturated liquid crystals (LC's) under a wide range of laser excitation conditions, including 1053-nm pulse durations between 600 fs and 1.5 ns and nanosecond pulse excitation at 351 nm and 532 nm, has been investigated. This multiwavelength investigation probed the correlation between the electronic structure of each material and its laser-induced damage behavior by altering the excitation photon energy. The laser-induced damage threshold at all wavelengths and pulse durations was consistently higher in saturated materials than in their unsaturated counterparts.

The electronic excitation pathways in LC materials are generally known and involve a singlet ground state (S_0) and excited singlet (S_1, S_2,…S_n) and triplet states. The time scale of the transition from the singlet states to the corresponding triplet states during relaxation, or intersystem crossing, is typically >1 ns, which has been confirmed for several unsaturated LC compounds.[1,2] Because the excitation leading to laser-induced damage (breakdown) occurs during the laser pulse, transitions with lifetimes longer that the pulse duration (in our case ~1 ns) have no (or minimal) effect on laser-damage mechanisms. Consequently, we consider only the transitions between the singlet states. The accordingly modified Jablonski energy diagram in Fig. 1 describes the electronic structure in LC materials involving singlet ground state (S_0) and excited singlet (S_1, S_2,…S_n), where the energy levels are defined as multiples of the energy of a 1ω photon used in this study.



	Number of photons required to bridge the lowest excited state S_1		
Wavelength (photon energy)	351 nm (3.35 eV)	527 nm (2.36 eV)	1053 nm (1.18 eV)
Unsaturated	1	2	3
Saturated	2	2	4

Figure 1
A schematic depiction of the electronic transitions leading to laser-induced breakdown in LC materials is presented by a modified Jablonski energy diagram involving a singlet ground state (S_0) and excited singlet states (S_1, S_2,…S_n). The energy levels are referenced as multiples of a 1053-nm photon (1ω). The UV transmission edge for each material (saturated < 351 nm < unsaturated) provides insight into the order of photon absorption required to bridge the energy gap from $S_0 \longrightarrow S_1$.

The relative difference in the measured laser-induced–damage threshold (LIDT) at different wavelengths is illustrated in Fig. 2 showing the *N*-on-1 LIDT results for both saturated and unsaturated materials at all wavelengths. Comparison of the 351-nm and 527-nm results shows a difference between LIDT values of ~5× for unsaturated materials and only of ~1.5× for saturated materials. This behavior can be anticipated from the order of absorption required for electrons to undergo the $S_0 \longrightarrow S_1$ transition. Specifically, unsaturated materials require both linear absorption at 351 nm and two photon absorption at 527 nm, while for saturated materials, two-photon absorption is necessary to populate the first excited state at both wavelengths. This key difference in the electronic excitation process is reflected in the corresponding difference in the LIDT values.

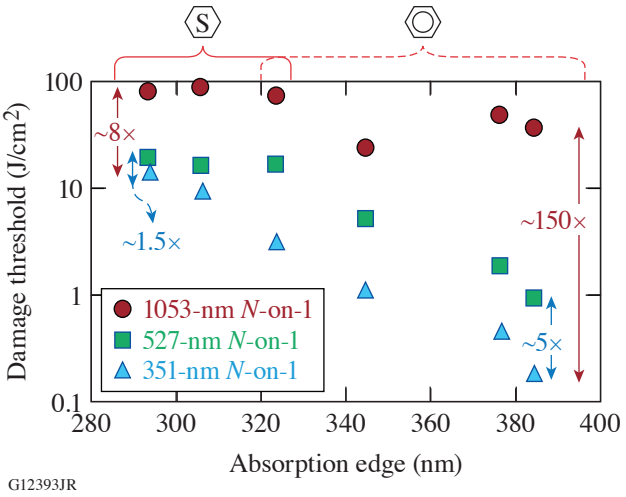


Figure 2
The *N*-on-1 LIDT values for nanosecond pulses at all three wavelengths (and 1-ns pulses) as a function of each material's absorption edge allows direct comparison of the relative differences in LIDT. The LC's investigated and their apsorption edge, as defined by $T = 98\%$, include: 1550C (294 nm), MLC-2037 (306 nm), ZLI-1646 (324 nm)PPMeOB/PPPOB (345 nm), 5CB (377 nm), and E7 (385 nm). Brackets identify saturated, unsaturated, and mixed materials. The symbols S and ○ are used to designate saturated and unsaturated materials, respectively.

Comparing LIDT results obtained under 351-nm and 1053-nm excitation, the difference in LIDT for unsaturated materials is ~150×, but only ~8× for saturated materials. The dramatic variation in LIDT differences for the two material types is arguably related to the different order of the absorption process required for the $S_0 \longrightarrow S_1$ transition. The order changes from linear absorption to a three-photon absorption process in unsaturated materials, while for saturated materials, a nonlinear process is required at both wavelengths (two-photon and four-photon processes for 351-nm and 1053-nm excitation, respectively). These results demonstrate the importance of the electronic structure of each material on the observed damage threshold and as a function of photon energy.

In summary, the experimental data suggest that key components in the laser-induced damage mechanisms in LC's involve a complex interplay of both multiphoton absorption and excited-state absorption, where their relative contributions vary with wavelength. Future work will concentrate on extending and applying these findings to both glassy and polymer LC materials systems and developing improved passive and active devices that offer polarization, phase, and intensity control for high-peak-power and average-power laser applications.

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2. G. E. O'Keefe *et al.*, Liq. Cryst. **21**, 225 (1996).