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To cite this version:
S. Rand, D. Sumida, L. Deshazer. ROOM TEMPERATURE LASER ACTION AND Q-SWITCHING OF F-AGGREGATE COLOR CENTERS IN LiF. Journal de Physique Colloques, 1985, 46 (C7), pp.C7-479-C7-482. <10.1051/jphyscol:1985785>. <jpa-00225115>

HAL Id: jpa-00225115
https://hal.archives-ouvertes.fr/jpa-00225115
Submitted on 1 Jan 1985

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ROOM TEMPERATURE LASER ACTION AND Q-SWITCHING OF F-AGGREGATE COLOR CENTERS IN LiF

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Abstract - Pulsed, room temperature laser action of \( F_2 \) and \( F_2^+ \) color centers was observed in gamma-irradiated LiF. Passive Q-switching by \( F_2^- \) centers also produced 30 ns Nd:YAG pulses.

I - INTRODUCTION

In recent years there have been several intriguing reports\(^1\),\(^2\),\(^3\) of tunable, color center lasers operating in LiF at room temperature with excellent stability and efficiency. Laser action has been ascribed to the aggregate centers \( F_2 \), \( F_2^+ \), and \( F_2^- \) created by gamma irradiation of this material and Q-switching by \( F_2^- \) centers was also described\(^4\). However, the reported operating characteristics of the color center lasers have been regarded by others as inconsistent with known properties\(^2\) of these centers and have been widely greeted with skepticism.

II - EXPERIMENT

We obtain room temperature operation at 10 Hz of \( F_2 \) and \( F_2^+ \) lasers in 2x10x10 mm crystals of LiF excited transversely by Q-switched 532 nm radiation and are able to explain some of the controversial aspects of earlier work. LiF containing only 50 ppm Mg\(^{2+}\) as the chief impurity was irradiated at room temperature with Co\(^{60}\) radiation to a total dose of 50 megarads at a 1.5 MRad/hr rate. Thermal annealing at 250°C for 12 hours in air resulted in an optical absorption coefficient of approximately 4.6 cm\(^{-1}\) at 532 nm in the wings of \( F_2 \), \( F_3^+ \) and \( F_4 \) center absorptions. A 15 cm cylindrical lens was then used to focus 30 mJ pulses of 8 ns duration at 532 nm to a line 5 cm beyond the sample. The laser cavity consisted of two plane mirrors 15 cm apart with reflectivities of 100% and 90% over the spectral region 650-950 nm. Two opposite 2x10 mm faces of the uncoated sample were polished plane parallel and oriented along the cavity axis.

Two RCA 7102 photomultipliers preceded by optical bandpass filters were used to observe that laser action due to \( F_2 \) centers\(^5\).
occurred briefly at 710 nm but diminished as progressive photoionization generated high concentrations of F$_2^+$ and led within two minutes to stable operation of the F$_2^+$ laser at a wavelength of 920 nm. Output occurred in 2 microjoule pulses of 16 ns duration with a beam divergence of 1.2 milliradians. Operation with no external cooling was tested at 10 Hz for periods of up to 10 hours and showed no evidence of degradation, provided excitation was maintained continuously. When operation was discontinued for periods as short as 5 minutes, laser action could not be reinitiated in the same active volume of the crystal. However, by moving to a new region of the crystal the photoionization process could be repeated and laser action re-established.

After laser operation and the disappearance of the photo-generated F$_2^+$ absorption, our samples reveal more permanent changes. These consist chiefly of a reduction in the width of the F$_2$ peak with a concomitant disappearance of the F$_4$ peak. The photoionized samples exhibit F$_2^-$ absorptions (960 nm) of 0.2 cm$^{-1}$, nearly identical to those measured prior to use in the cavity. Provided that samples are protected from strong ultraviolet light exposure, this remnant concentration of F$_2^-$ centers is very stable in contrast to the F$_3^-$ (790 nm) population, for example, which completely disappears. This unexpected stability of F$_2^-$ centers is not understood. Gellermann et al. have studied electron irradiated LiF and observed similar stability during cw F$_2^-$ laser experiments but found that impurities invariably lowered the F$_2^-$ yield. By contrast, with gamma irradiation we observe a small enhancement of F$_2^-$ yield by Li$_2$O impurities (Fig. 1) in material grown by the Czochralski technique in a reactive atmosphere which dehydrates the melt and scavenges CO$_2$ from the Li$_2$CO$_3$ additive. Still, maximum yields with low dose-rate gamma irradiation are invariably lower than those obtained using electron irradiation.

![Graph](image-url)

Figure 1. Absorption coefficient at 960 nm versus gamma ray dosage for different LiF samples.
Hence the earlier reports of stable $F_2^+$ laser action are confirmed although the cause of $F_2^+$ laser failure following interruption of the excitation needs clarification. Stimulated emission from $F_2$ centers fades rapidly due to photoionization at room temperature. Unexpected stability of $F_2^-$ centers is also observed, but the role of crystal composition appears to depend on the irradiation method and the stabilization mechanism in pure LiF remains puzzling.

![Figure 2](image_url)

**Figure 2.** Absorption versus incident intensity at 1.06 microns in a 5 cm LiF rod irradiated to a dose of 105 MRad. The dashed curve is a fit to low intensity data assuming a single transition with saturation intensity 0.93 MW/cm$^2$. The presence of a background absorption due to $F_2$ centers accounts for the difference in high intensity values.

We also obtained stable Q-switching action due to the saturable absorption of $F_2^-$ centers for over $10^6$ shots at room temperature. An uncoated 5 cm rod of commercially available LiF was irradiated with a total dose of 105 MRad. The low intensity transmission corrected for Fresnel loss at 1.06 microns was 39%. The absorption saturation curve was measured using a Nd:YAG laser and is shown in Fig. 2. When used intracavity in an oscillator consisting of 100% and 30% reflectors, passive Q-switching was observed. 21 mJ pulses were obtained for 11 J stored energy in a Q-switched pulse envelope of 30 ns duration. Partial mode-locking was evident (Fig. 3a) when a measurement bandwidth of 1 GHz was attained. The time interval between successive pulses in the figure corresponds accurately to the cavity round trip time, which is considerably less than the measured excited state lifetime (64 ns). Insertion of an uncoated 1 cm etalon in the cavity resulted in single mode operation (Fig. 3b).
Figure 3. (a) Partially mode-locked, Q-switched Nd:YAG pulse produced with the F$_2$:LiF switch of Fig. 2. (b) Single longitudinal and transverse mode operation obtained with 1 cm unocated, intracavity etalon.

In summary, we have demonstrated room temperature operation of F$_2$ and F$_2^+$ color center lasers in LiF heavily irradiated with gamma rays. Our results help explain the origin of unusual behavior reported previously for F$_2$ and F$_2^+$ lasers. Also, the F$_2^+$ centers in our samples exhibit exceptional stability and can be used as efficient passive Q-switches for Nd:YAG lasers. Larger doses of gamma radiation, the use of Brewster-cut crystals, and a longitudinal pumping geometry should provide higher efficiencies for the remarkably simple, tunable F$_2^+$ solid state laser.

Acknowledgement: The authors wish to thank M. Robinson of Hughes Research Laboratory for the growth of ultrahigh purity LiF crystals doped with Li$_2$O, Mg and Ni and the Crystal Growth Lab of the Dept. of Physics, University of Utah for providing zone-refined samples of MgF$_2$:LiF.

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