FORMATION OF $F_2^-$ COLOR CENTERS IN LiF MONOCRYSTALS BY ELECTRON IRRADIATION

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Abstract

Formation conditions, optical non-linear properties and some applications of $F_2^-$ color centers in LiF monocystals are investigated.

1. INTRODUCTION

The first reports on the application of $F_2^-$ color centers generated by irradiation of LiF monocystals were made in 1983 by Chebotaev and his co-workers [1].

The most important applications of LiF:$F_2^-$ crystals are based on their non-linear optical properties. LiF crystals with $F_2^-$ color centers can operate as passive Q-switches or can be used as optical isolators in high power laser amplifier systems [2,3]. They also can be used for the generation of ultrashort pulses and for the generation of difference frequencies in the i.r. range.

The LiF:$F_2^-$ crystals can be obtained by electron, gamma or neutron irradiation. The optical properties and thermal stability depend on the irradiation dose, doping impurities of the crystal and the method of irradiation.

In spite of their excellent optical properties and thermal stability of $F_2^-$ color centers, the use of LiF:$F_2^-$ crystals is prohibited by the high costs of irradiation, unless an efficient irradiation method is applied [4].

The present paper presents the results obtained when irradiation was made with electrons generated by a linear accelerator with a 7 MeV average energy and with gamma radiation generated by a 5 x 10^5 Gy/h Co-60 source.

Centers can be optically excited at high intensities without damaging them. They are evidencing a very good stability in the -40°C to 60°C temperature range.

2. COLOR CENTERS FORMATION BY GAMMA AND ELECTRON IRRADIATION

$F_2^-$ color centers in LiF crystals, created by gamma or electron irradiation are characterised by broad absorption and emission bands centered at 300K, at 960 nm and 1130 nm. $F_2^-$ centers are two anion vacancies located along the diagonal face of anion cell with three captured electrons. At room temperatures transitions are of vibrational type. The system can work as four level laser.

LiF:$F_2^-$ crystal has a high absorption cross section at Nd³⁺ laser wavelength of 1.5-2 x 10⁻¹⁷ cm². Excited level lifetime is 70-100ns.

When carrying out irradiations in gamma radiation field, more Co-60 radiation sources, circularly arranged to achieve an uniform irradiation field in centre, with 5 x 10⁵ Gy/h dose rate were employed.

Electron irradiation was carried out by a linear accelerator which can generate irradiation dose rates up to 5 x 10⁵ Gy/h at 7 MeV electron average energies. The electron energies decrease was accomplished using some aluminium attenuators.

The irradiation of the crystals was made at room temperature. After irradiation at different doses the crystals were stored for 48 hours. During this period all samples evidenced a fast decay of $F_2^-$ concentration. The remaining $F_2^-$ centers presented a much higher thermal stability.

Centers stabilisation can be accelerated if the crystals are kept at 100°C in a controlled temperature chamber.

After stabilising the centers, optical transmission measurement were made on the crystals with a Q-switched Nd:YAG laser and a laser energimeter-ratiometer system. The laser beam was attenuated to avoid bleaching the crystals.

Figure 1 illustrates the variation of the absorption coefficient for LiF:$F_2^-$ crystals at 1.064μm wavelength, function of the irradiation dose in gamma radiation field and by electron irradiation.

The crystals were identical having 38 mm length and 8 mm diameter, grown in air.

When irradiated by electrons, their average energy at the crystal level was 3 MeV and the electron beam direction was perpendicular on the crystal optical axis. In order to obtain an uniform irradiation, the crystals were rotated into the beam.
When irradiated by gamma radiations, one may observe a much lower irradiation efficiency.

At the dose of irradiation was performed the growth of the F\textsubscript{2} color centers density was linear. A saturation is expected to appear at higher doses.

When irradiated by gamma radiations a 0.45 cm\textsuperscript{−1} absorption coefficient was obtained for 6x10\textsuperscript{6} Gy integrated dose while for 3 MeV electron irradiation, the same absorption coefficient was obtained at 1.4x10\textsuperscript{4} Gy.

Due to more powerful irradiation, the final transmission for the gamma irradiated crystal was 72% while for electron irradiated crystal it was 80%. Final transmission was measured with a Q-switched Nd:YAG laser. The power density of the beam was higher than 10 MW/cm\textsuperscript{2}, so the color centers were completely bleached. At saturable absorbers, the final transmission characterises optical residual losses. Residual losses are function of the LiF:F\textsubscript{2} crystal preparation technology, nature and impurities content.

The efficiency of electron irradiation is much better, the irradiation time being only 4 hours as compared to 300 hours necessary for the irradiation in the gamma radiation field.

3. OPTICAL PROPERTIES

One of the most important applications of the LiF:F\textsubscript{2} crystals is laser cavity Q-switching, based on the non-linear optical effect of absorption saturation.

Figure 2 shows the non-linear variation of transmission in a 38 mm long LiF:F\textsubscript{2} crystal function of the incident laser pulse power density, having 1.064 \mu m wavelength and 10 ns pulse width.

![Figure 2](image_url)

The non-linear absorption is caused by changes in the population of electron states under high power density radiation.

The experimental results are well described by the expression:

\[
T = \frac{w}{w_s} \ln \left[ 1 + T_0 \left( e^{w/w_s} - 1 \right) \right]
\]

where \(T_0\) is the crystal initial transmission at \(w=0\), \(w\) is the power density, and \(w_s\) is the saturation parameter: \(w_s = 1.2\) MW/cm\textsuperscript{2}.

Figure 3 presents the transmission spectral data of a 38 mm long LiF monocrystal, irradiated by electrons...
at various irradiation doses. The electron average energy was 7 MeV. The crystal growth was performed in air presenting OH− impurities in the crystal network which resulted in a decrease of the F−2 color centers concentration for the same irradiation dose as compared to the irradiation of a pure crystal.

A significant improvement of the efficiency in generating F−2 color centers in LiF monocrystals impurified by OH− was obtained by decreasing the electron average energy from 7 MeV down to 3 MeV.

In spite of all these differences between the two crystals, when employed for passive Q-switching a Nd:YAG laser cavity, close results were obtained. The initial transmission at 1.064 μm laser wavelength was 20% for the OH− impurified crystal and 16% for the pure one. The final transmission measured with a ratiometric system, at a power density of 15 MW/cm² and a pulse width of 10 ns, was 80% and 72% respectively. Nd:YAG laser rod had dimensions of Φ4x60 mm. The output mirror reflectivity was 30% and the resonator length was 15 cm.

When employed in laser Q-switching output energies obtained were of 23 mJ for the impure crystal and 26 mJ for the pure one. The pulse width was 10 ns for both crystals.

4. CONCLUSIONS

As a conclusion it should be noted that LiF:F−2 crystals can be obtained from optically machined LiF crystals irradiated with a very good efficiency in electron beam.

One of the most important sources of accelerated electrons at energies of about 3 MeV can be the linear accelerators that allows high irradiation doses. This irradiation technique provides the possibility to obtain very good quality crystals at acceptable prices.

Nd:YAG lasers Q-switched with LiF:F−2 crystals can be used in applications that require extended range of temperatures.

REFERENCES