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Alexandrite laser pumped LiF:F_2^- laser

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Abstract

For the first time to our knowledge a direct alexandrite laser pumping of LiF:F_2^- crystals was suggested and realized by means of energy transfer from the F_3^- to the F_2^- color centers (CCs). Tunable laser oscillation of LiF:F_2^- in the 1000–1300 nm spectral range with a maximum efficiency of 10% was achieved. The process of two-step photo-ionization of the F_3^- CCs under 740 nm excitation was demonstrated experimentally on the basis of the measured decay rate of F_3^- centers. The photo-ionization cross-section from the first excited level of F_3^- centers was calculated to be 2×10^{-22} cm^2. Stable output lasing of the LiF:F_2^- laser at room temperature under 793 nm excitation was predicted and realized.

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Keywords: Color center laser; Tunable laser; Photo-ionization; Alexandrite laser

Recent progress in LiF color center (CC) crystals is essential for design of effective room temperature stable CC lasers tunable in the near-IR spectral region (Refs. [1–4], and references therein). LiF:F_2^- are the most widely used CC crystals in quantum electronics, specifically as passive Q-switchers of resonators of neodymium lasers and as active elements of near-IR tunable lasers [1–3]. The F_2^- CCs can be considered as three electrons trapped by two adjacent anion vacancies having axial symmetry along the (1 1 0) direction. F_2^- CCs feature wide near-IR absorption (0.85–1.1) and emission bands (1.0–1.3 μm) and a high (~50%) quantum efficiency of fluorescence at room temperature. LiF:F_2^- active elements combine unique spectroscopic, oscillation, thermo-optic, and operational properties [1,2].

Neodymium laser pumping, being very attractive, from the other hand limits the tuning range and efficiency of LiF:F_2^- CC lasers [5]. The fundamental harmonic of the neodymium laser falls into the region of overlapping of F_2^- CC absorption and emission bands (see Fig. 1). In this case, as it is shown in Ref. [5], the maximum of population density of the F_2^- CC exited state under short nanosecond excitation will be significantly limited due to the competition between simulated absorption and emission at pumping wavelengths:

$$N_{\text{excited}}^{\max} = N_{\text{total}}\sigma_{\text{ab}}(\lambda_{\text{p}})/[\sigma_{\text{ab}}(\lambda_{\text{p}}) + \sigma_{\text{em}}(\lambda_{\text{p}})] = N_{\text{total}}\xi^{\text{eff}}(\lambda_{\text{p}}), \quad (1)$$

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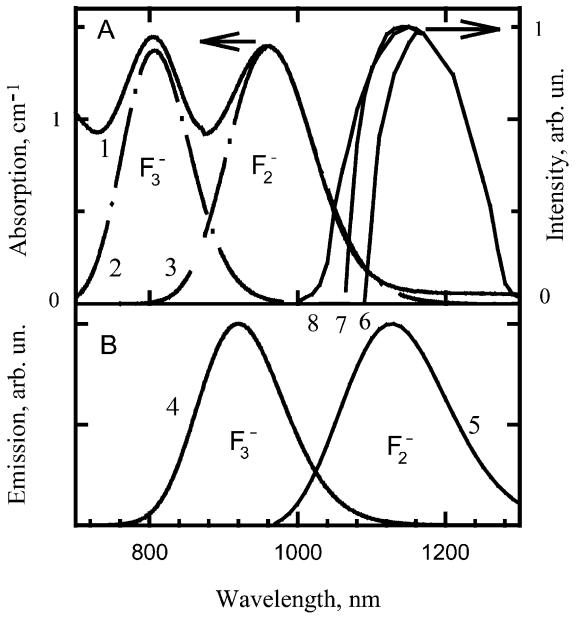

Fig. 1. Absorption (1–3) and fluorescence (4,5) bands of LiF:F_2^- crystal compared to normalized tuning curves of LiF:F_2^- laser (6–8). Curves 1–3 represent overall, F_3^-, and F_2^- absorption bands, respectively. Curves 4 and 5 are fluorescence bands of F_3^- (4) and F_2^- (5) CCs in LiF crystal. Curves 6–8 are normalized tuning curves of LiF:F_2^- CC laser pumped by the fundamental harmonic of YAG:Nd^{3+} laser [5], YLF:Nd^{3+} laser [5], and alexandrite laser (current study), respectively.

where N_{total} and $N_{\text{excited}}^{\max}$ are total and maximum concentrations of the excited state of F_2^- CCs; $\sigma_{ab}(\lambda_p)$ and $\sigma_{em}(\lambda_p)$ are absorption and emission cross-sections at the pumping wavelength. The effective partial density $\xi^{\text{eff}}(\lambda_p)$ depends strongly on pumping wavelength and its values are shown in Table 1 for several types of neodymium lasers. As one can see the shift of the pump wavelength from 1079 nm ($YAlO_3$:Nd^{3+} laser) to shorter than 980 nm wavelength results in 6.25 times increase of the excited CC concentration. Optimization of the F_2^- CC partial density plays a crucial role in effective CC laser operation since maximum of F_2^- CC concentration is limited to 2×10^{16} cm^{-3} even for advanced technological process of LiF crystal coloration [1]. After crystal γ-irradiation with a dosage of $\sim 2 \times 10^8$ rad the concentration of active CCs reaches saturation level accompanied by formation of colloidal particles responsible for parasitic losses at oscillation wavelength [1]. Thus, a shift of pumping wavelength to 1000 nm and shorter results in both enhancement of tuning range and better efficiency due to increase of effective concentration of active CCs. Indeed, when pumped near maximum of F_2^- CCs absorption band LiF:F_2^- laser yield a very high conversion efficiency (up to 60% for 10 ns pump pulse duration) and wide tuning over 1040–1300 nm spectral range [3].

Table 1
The effective partial density $\xi^{\text{eff}}(\lambda_p)$ of F_2^- CCs in LiF crystal under different pumping wavelength [5]

Pumping wavelength (laser)	ξ^{eff}
1079 nm (YAP:Nd^{3+})	0.16
1064 nm (YAG:Nd^{3+})	0.26
1047 nm (YLF:Nd^{3+})	0.43
<980 nm	~1

In this paper we report a study of LiF:F_2^- CC laser operation under alexandrite laser pumping (720–800 nm) when population inversion occurs due to a resonant absorption of pump photons by F_3^- centers with subsequent radiative energy transfer from F_3^- to lasing F_2^- CCs. Being very interesting from the physical point of view, this mechanism also holds promise for improvements in efficiency and tunability range of LiF:F_2^- laser.

Recently an efficient alexandrite laser pumped LiF:F_2^{+**} laser tunable in 796–1210 nm and stable at room temperature was demonstrated [4]. Therefore, design of LiF:F_2^- and LiF:F_2^{+**} CC lasers with alexandrite laser as a single pump source is also very promising for the development of an ultra-widely tunable (720–1300 nm) solid state laser system.

The F_3^- CC consists of two electrons trapped on three adjacent anionic vacancies. It is known that the F_3^- CCs in LiF possess an absorption band centered at 800 nm with FWHM of 1400 cm^{-1} and maximum of absorption cross-section of $\approx 10^{-17}$ cm^2 [1]. So, alexandrite laser radiation tunable from 720 to 800 nm may be used for F_3^- CCs excitation. The lifetime of the excited state is 10 ns at room temperature. The emission band of F_3^- CCs with a peak at 920 nm and a half width of 1600 cm^{-1} overlaps well with the broad (FWHM $\sim$ 1400 cm^{-1}) absorption band of F_2^- CCs centered at 960

nm. This overlap of F_3^- emission and F_2^- absorption bands may result in effective excitation of F_2^- through the following mechanism of F_3^- to F_2^- CCs energy transfer:

$$h\nu_{\text{pump}} + F_3^- + F_2^- \rightarrow F_3^{-*} + F_2^- \rightarrow F_3^- + h\nu_{920} + F_2^- \rightarrow F_3^- + F_2^{-*} \quad (2)$$

Crystals used in this study were grown by Kyropulos method from nominally pure raw material and featured 0.05 cm^{-1} coefficient of absorption of OH^- group at 2680 nm. LiF crystals were Brewster cut to a length of 4 cm and polished. To create an active medum with both F_3^- and F_2^- CCs, LiF crystals were γ-irradiated at 300 K with a dose of 10^8 rad using ^{60}Co source. The absorption spectrum of the studied crystal is shown in Fig. 1. Maximum absorption coefficients of F_2^- and F_3^- CCs were 1.2 cm^{-1} (at $\lambda = 960$ nm) and 1.3 cm^{-1} (at $\lambda = 800$ nm), respectively.

Initial laser experiments were performed in a conventional dispersive cavity formed by an input dichroic mirror and Littrow mount diffraction grating (1200 grooves/mm) with diffraction efficiency of 30%. A zero order of diffraction served as the output. The input mirror had a high-level transmittance at pumping wavelengths ($T > 93\%$) and high reflection ($R > 99\%$) in the oscillation range 1050–1300 nm. The non-focused alexandrite laser (PAL 101, Light Age, Inc.) radiation with a maximum pulse energy of 80 mJ at 750 nm, beam diameter of 1 mm (FWHM), pulse duration of 70 ns, and repetition rate of 24 Hz was used for pumping.

The LiF:F_2^- CC laser provided a near diffraction limited spatial beam with a spectral line width of output radiation 2 cm^{-1}. Experimental tuning curve extended from 1000 to 1300 nm with a maximum efficiency of 10% at 1130 nm (see Fig. 1, curve 8). For comparison, the normalized tuning curves of LiF:F_2^- CC laser pumped by the radiations of YAG:Nd^{3+} (1064 nm, curve 6) and YLF:Nd^{3+} (1047 nm) laser (curve 7) are also shown in Fig. 1 according to Ref. [5]. As one can see from Fig. 1 the using of such $F_3^- \rightarrow F_2^-$ excitation mechanism allows to extent the short wavelength tail of the tuning curve down to 1000 nm. However, the output radiation of the LiF:$F_3^- \rightarrow F_2^-$ laser under 100 MW/cm^2 of 750 nm pump power density was not stable and featured ~10-fold decrease (due to F_2^- CCs two-photon bleaching) after 30 min of operation at a repetition rate of 24 Hz.

Since CCs photo-stability under powerful pump radiation is one of the important parameters of CC lasers we performed a detailed study of the LiF:F_2^- and F_3^- photo-stability under the action of different power densities and wavelengths of the pump radiation. It is known that the F_2^- CCs and F_3^- CCs are photo-stable under excitation into the main absorption band [1]. The photo-ionization properties of F_2^- and F_3^- CCs were studied earlier in Ref. [6] and the probability of single-photon ionization under UV excitation according to this paper is depicted in Fig. 2. As it is seen in Fig. 2, the long wavelength edge of F_2^- CC photo-ionization is about 385 nm. However, under the laser excitation, possible multi-photon processes including excited state absorption should be taken into account, particularly in our case when two-photon tuning curve of pumping laser overlaps with photo-ionization bands (see Fig. 2, curve 3).

To study multi-photon photo-ionization a decrease of the F_3^- CCs concentration versus irradiation time was measured for different pump peak

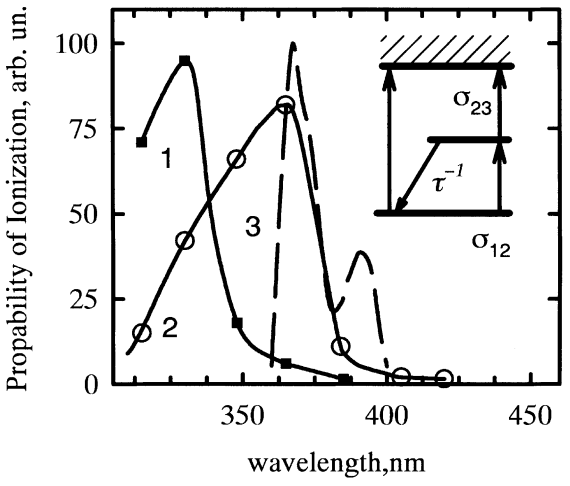

Fig. 2. Spectral dependencies of probability of F_3^- (1) and F_2^- (2) CCs single-photon ionization at room temperature [6] compared to the alexandrite laser two-phonon excitation tuning curve (3).

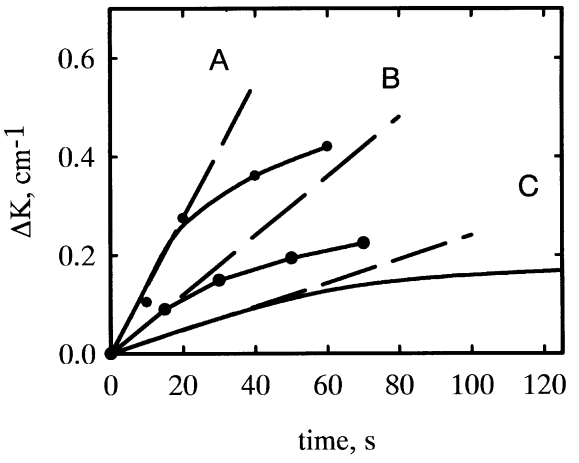

Fig. 3. The kinetics of F_3^- CC concentration changes (ΔK) under 740 nm irradiation with different power density: (A) 38 MW/cm^2, (B) 26 MW/cm^2, and (C) 16 MW/cm^2.

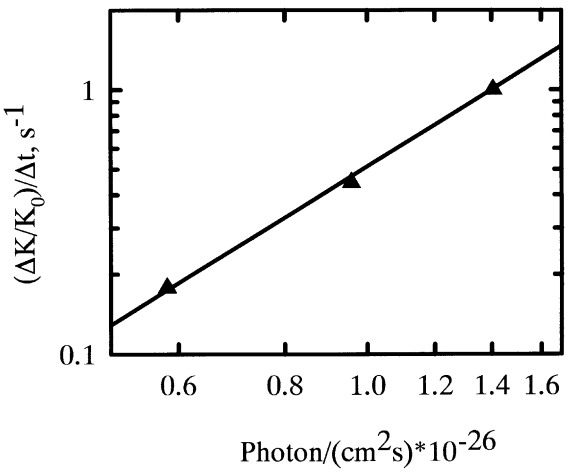

Fig. 4. The dependence of initial decay rate of F_3^- CC $(\Delta K/K_0)/(\Delta t)$ versus pump power density (at 740 nm).

powers. Fig. 3 demonstrates how F_3^- CCs absorption coefficient changes as a function of exposition time for three different levels of 750 nm pump peak power. The decrease of the pump power density results in a significant slowdown of F_3^- destruction. According to theoretical analysis [7] the initial rate of decay $(\Delta K/\Delta t)$ of the F_3^- centers in LiF in the case of two-step photo-ionization mechanism is proportional to square of the power density:

$$(\Delta N/N_0)/\Delta t = (\Delta K/K_0)/\Delta t \approx \sigma_{12}\sigma_{23}\tau I^2(m+1), \tag{3}$$

where σ_{12} and σ_{23} are absorption cross-sections from the ground and excited state levels, τ is the lifetime of excited level, I is pump power density, and the last factor $m+1$ equals to the product of pulse duration (t_{pump}) and the pulse repetition rate (f): $(m+1) = t_{\text{pump}} * f$.

Fig. 4 shows the experimental dependence of initial F_3^- centers destruction rate (solid dots) and the square dependence theoretical fit (straight line), measured from the slopes of the linear parts of kinetics in Fig. 3. As one can see the rate of the decay is proportional to the square of the excitation power density. This experimental data comes as evidence in support of the following two-step mechanism of F_3^- CCs destruction under 750 nm excitation:

$$2h\nu_{\text{pump}} + F_3^- \rightarrow F_3^{-*} + h\nu_{\text{pump}} \rightarrow F_3 + e \tag{4}$$

The excited state absorption cross-section (σ_{23}) calculated using Eq. (3) on the basis of experimental results in Fig. 4 was equal to 2×10^{-22} cm^2. It is close to the value of cross-section of two-step photo-ionization of F_2 CCs pumped by the radiation of the second harmonic of YAG:Nd^{3+} laser and measured in Ref. [7]. As discussed above, the long wavelength edge of photo-ionization of F_3^- CCs in LiF is near 380 nm (see Fig. 2, curve 1). Hence, two-step photo-ionization rate should have fallen as the pumping wavelength is increased to 790 nm and longer wavelengths.

To check this idea a long term stability of LiF:F_2^- CC laser under different pumping wavelengths of the alexandrite laser was investigated. Laser experiments were performed in a simple flat–flat resonator, set to a length of 15 cm. The output coupler has transmitted ~50% in the 1000–1300 nm range. An unfocused pumping beam with a power density of 100 MW/cm^2, and repetition rate of 24 Hz was introduced into 4 cm long Brewster cut LiF crystal through the input mirror. The temporal stability of the LiF:$F_3^- \rightarrow F_2^-$ CC laser output oscillation for different pump wavelengths is demonstrated in Fig. 5. As evident from the Fig. 5, the temporal stability of output oscillation of the LiF:$F_3^- \rightarrow F_2^-$ laser strongly depends on excitation wavelength. For shorter pump wavelengths

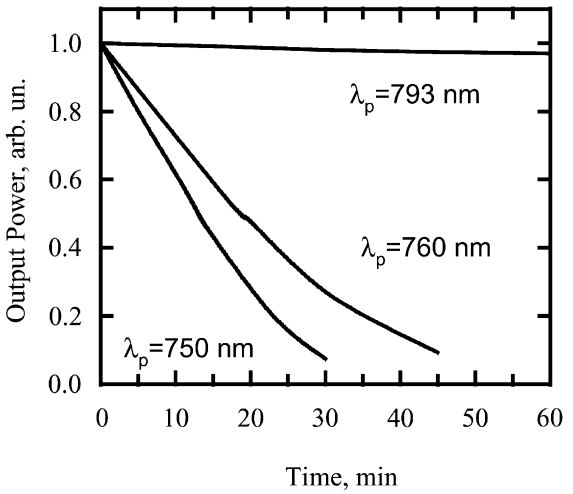

Fig. 5. Long term stability of LiF:F_2^- CC laser for different alexandrite laser pump wavelengths.

(e.g. 740–750 nm) there was a 10-fold fall of the output power after 30 min of operation. On the other hand, when pumped with 793 nm radiation the laser operated without any noticeable fading during 60 min. Presented results on stable operation of the laser under 790–800 nm pumping are in a good agreement with data shown in Ref. [5] for a single-photon ionization of F_3^- centers.

In conclusion, for the first time to our knowledge a direct alexandrite laser pumping of LiF:F_2^- crystals was suggested and realized by using energy transfer mechanism from F_3^- to F_2^- CCs. Tunable oscillation of LiF:F_2^- laser in 1000–1300 nm spectral range with maximum efficiency of 10% was achieved. The two-step photo-ionization of F_3^- CCs under $\lambda = 750$ nm excitation was demonstrated experimentally. The photo-ionization cross-section from excited state level of F_3^- CC was calculated to be 2×10^{-22} cm^2. A stable output oscillation of LiF:$F_3^- \rightarrow F_2^-$ laser at room temperature under $\lambda = 793$ nm excitation was predicted and shown experimentally. Thus, in combination with early reported alexandrite laser pumped LiF:F_2^{+**} CC laser [4] an all solid state laser system continuously tunable over 720–1300 nm spectral range (alexandrite laser—720–800 nm, LiF:F_2^{+**} CC laser—800–1200 nm, and LiF:$F_3^- \rightarrow F_2^-$ CC laser—1000–1300 nm) was demonstrated.

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