Powerful ultrabroadly tunable LiF:F$_2$$^{+**}$ laser

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ABSTRACT

The LiF:F$_2$$^{+**}$ laser has been shown to provide high power (hundreds of mJ), efficient (tens of %), room temperature stable laser operation tunable from 800 nm to nearly 1210 nm. However, no reasonable explanations were provided for such an ultrabroadband tuning capability using this active medium. This work describes the mechanism of the ultrabroadband lasing from LiF:F$_2$$^{+**}$ under Alexandrite laser pumping. Alexandrite laser pump radiation corresponds to the region of spectral overlapping in the absorption bands of two color center in LiF, namely F$_2$$^{+**}$ and a new F$_2$$^{+**}$-like center, conclusively discovered in this work. Using the Alexandrite laser's excitation, we can achieve simultaneous population inversion in both centers making F$_2$$^{+**}$ and the F$_2$$^{+**}$-like centers, the primary color centers responsible for the ultrabroadband tunability. This work details the temperature-dependent spectroscopic measurements of these two color centers and the resulting laser characteristics. Absorption and fluorescence measurements were performed from 300K to 13K. The room temperature (RT) absorption data shows that the F$_2$$^{+**}$ center has maximum absorption at $\lambda_{abs} = 614.5$ nm and FWHM, $\Delta \nu = 3819$ cm$^{-1}$; the F$_2$$^{+**}$-like center has RT absorption at $\lambda_{abs} = 813.4$ nm and FWHM, $\Delta \nu = 3592$ cm$^{-1}$. The fluorescence of both centers was measured using several excitation wavelengths from 532 nm to 778 nm. F$_2$$^{+**}$ demonstrated fluorescence emission from 765-1080 nm, corresponding to 10% of maximum, and F$_2$$^{+**}$-like from roughly 890-1400 nm, with fluorescence maxima at $\lambda_0 = 905$ nm and $\lambda_0 = 1098$ nm, respectively.

Keywords: LiF:F$_2$$^{+**}$, Color Centers, Solid-State laser, Alexandrite pumping, Tunable laser, Spectroscopy

1. INTRODUCTION

Many applications exist in scientific experimentation for broadband laser sources including multi-element intra-cavity spectroscopy, multi-element laser atomic fluorescence (requires multiline regime of operation — only selected, pre-assigned composition of wavelengths), color-holography, and others. Also, broadband sources are widely used in telecommunications as well as military applications, e.g., objective secure information processing and coding.

We discuss here an ultrabroadband infrared laser source based on the color center crystal LiF:F$_2$$^{+**}$, which is both thermal- and photo-stable. Ultrabroadband lasing is the ability to provide laser radiation consisting of spectral output that coincides to nearly the entire fluorescence spectrum of the medium. Previous results have realized non-selective cavity lasing in this medium from 800 nm-1210 nm. During this work, we have realized superbroadband lasing (SBL) from ~880-1080 nm. The current experiments indicate that this range can be extended from 800-1300 nm and also realized as an UBL system. To our knowledge, no other single laser medium has such a broad tuning range.

2. PRINCIPLES OF ULTRABROADBAND LASING (UBL)

This section and sub-sections are adapted from a previous work by Basiev, Zverev and two of this paper's authors. That work investigated the SB lasing of F$_2$ color centers in LiF; a logical adaptation of that work to F$_2$$^{+**}$ is provided here to mathematically explain the broadband lasing we have realized.

The LiF:F$_2$$^{+**}$ UB laser system is made possible by the broad absorption/emission bands of the active medium and the novel laser cavity, shown in Figure 1, which essentially eliminates mode-competition between the various frequencies generated. This cavity was patented by Basiev, et. al and effectively demonstrated for superbroadband lasing of LiF:F$_2$ crystal (see and references herein).
In our initial experiments where SBL was achieved, the active medium was quasi-longitudinally pumped by the Raman shifted (D₂) second harmonic of a Q-switched Nd:YAG laser at 632 nm. The circular profile with diameter of 10 cm is incident on a beam-reshaping cylindrical lens, which creates an elliptical beam profile with major axis in the direction of the diffraction grating dispersion. The reshaped beam is then incident on the input mirror (85% transmission at 632 nm and >99% reflective for 800-1200 nm) of the laser cavity. The pump beam then strikes the active medium, which then undergoes spontaneous emission. A fraction of the total spontaneously emitted radiation will continue through the cavity and is incident on the intracavity cylindrical lens. The portion of the spontaneous emission, which was essentially parallel to the optical axis prior to refraction by the lens, passes through the off-axis mode-suppression element (vertical slit). These primary laser modes encounter the corner reflector assembly, which consists of a diffraction grating, in Littrow configuration, which serves as the output coupler, and highly reflecting dielectric mirror. The Littrow mount implies that the first-order diffraction is retro-reflected back into the laser cavity and the zero-order is the output of the system. The retro-reflected radiation consisting of many diffracted primary laser modes, each with its own wavelength, again encounters the mode-suppression element; the slit extracts from the diffracted radiation only the main laser modes, all other secondary modes that are divergent from the optical axis, are therefore blocked by the slit. The remaining radiation passes back through the cylindrical lens and active medium. This radiation and the stimulated emission produced by it are then reflected back by the input mirror. This cavity gives rise to the ultrabroadband oscillation: independent oscillation from different parts of the active medium, each with a different wavelength, and identical output direction. The output radiation from the UBL is comprised of a continuous number of beams in the active medium, each parallel to the other and the optical axis. Each beam trajectory in the cavity has a unique angle of incidence on the grating and, consequently, a unique oscillation wavelength, which is determined by the diffraction grating equation when operating in the autocollimation regime: \( \lambda = 2t \sin \theta \), where \( t \) is the grating spacing and \( \theta \) is the angle of incidence. Because we have essentially eliminated mode competition, it is possible to make the statement that each part of the active medium, which is parallel to the optical axis, operates as an active element of independent laser, each with a unique wavelength. The total UBL output from the system is a spatially spread "rainbow" spectra that is divergent in the direction of the grating dispersion; this means that each wavelength has a unique angular direction in the total output.

### 2.1. Spectral resolution \(^2\)

From Figure 1, we can state that each beam (wavelength) passing through the active medium has a coordinate "y" in the transverse direction and the value "\( y_0 \)" corresponds to the "radius" of the excited region from the edge of the pump beam to the laser axis. The intracavity cylindrical lens has focal length, \( f \). Also, the intersection of the focused radiation (between "y" and "\( y_0 \)" is measured as angle, \( \alpha \). From this geometry, we have:

\[
\alpha = \tan^{-1} \left( \frac{y_0 - y}{f} \right)
\]
Also from Figure 1, the angle β is assigned to be the angle between the grating normal and the optical axis. This implies that each beam "y" has incident angle "β+α" to the grating. In autocollimation regime, the grating equation is given as

\[ m\lambda = 2t \sin(\beta + \alpha) \]  

(2)

where \( m \) is the order of the reflected beam (here, \( m=1 \)), and \( t \) is the grating spacing (1200 grooves/mm).

The possibility of establishing oscillations of a certain wavelength without interfering with another wavelength is the spectral resolution of the UBL and is determined by the spectral resolution of the laser cavity itself. An approximation can be made from the angular selectivity of the cavity established by the ratio of the intra-cavity aperture width and the focal length of the intra-cavity cylindrical lens:

\[ \Delta\alpha = \frac{h}{f} \]  

(3)

By applying this angular selectivity into Equation (2) we can show that the spectral resolution is given as:

\[ \Delta\lambda = 2\frac{h}{f} \cos(\beta + \alpha) \]  

(4)

The width of the aperture must be such that the laser obtains high conversion efficiency and spectral resolution while at the same instant maintains operation as a single transverse mode with low diffraction losses. This implies choosing the Fresnel number for each individual laser ≈ 1.

\[ F = \frac{h^2}{L \cdot \lambda} \approx 1 \]  

(5)

where \( L \) is the length of the cavity.

Spectral resolution on a similar super broadband crystal, \( F_1 \), with similar cavity dimensions obtained spectral resolution on the order of 6nm. This tells us that the entire UBL system operates as dozens of independently operating standard narrowband lasers. With proper spatial filtering of the pump beam, which subsequently blocks specific regions of the active medium, the output can consist of any smaller quantity, e.g., 1-10, independent lasers with pre-assigned spectral composition.

### 2.2. Pump system

The pumping system for the UBL system plays an integral part in the generation of ultrabroadband oscillations. The pumping system for the UBL plays a direct role in determining the bandwidth of the laser emission, \( \Delta\lambda \). The UBL pump system is a combination of a pump laser (initial experiments used a Raman shifted (D2) second harmonic of a Q-switched Nd:YAG laser, subsequent experiments use Alexandrite laser excitation) and a cylindrical lens used for beam profile reshaping.

The following equations (see reference 2) illustrate the interrelated nature of the beam width, \( d \), of the pumped zone in the active medium, the dispersion, \( D \), of the diffraction grating, the focal length, \( f \), of the intracavity cylindrical lens, and the bandwidth of the produced laser radiation.

\[ d \approx f \cdot \tan(\Delta\lambda \cdot D) \]  

(6)

where \( D = \frac{\partial(\beta + \alpha)}{\partial\lambda} \).

The provided value for the dispersion and the schematic geometry in Figure 1 leads to the following equation:

\[ D = \frac{\tan(\beta + \alpha)}{\lambda} = 0.67 \text{nm/mr} \]  

(7)

Our particular system incorporates a lens with focal length, \( f = 25 \text{ mm} \), the dimension of the pump beam is \( d = 10 \text{ mm} \), and from these value we can determine the spectral output bandwidth, \( \Delta\lambda = 250 \text{ nm} \).
3. SPECTROSCOPY OF THE ACTIVE MEDIUM

A crystal sample from a single boule prepared by the method described by Dergachev, et al., and was used to perform spectroscopic measurements from room temperature (RT) to 13 K. The crystal thickness was 0.53 cm and it was co-doped with oxygen ($-2 \times 10^{17}$ cm$^{-3}$), hydroxyl groups ($-2 \times 10^{18}$ cm$^{-3}$), and magnesium ($-10^{17}$ cm$^{-3}$).

Initial measurements determined the transmission spectrum of the sample. A Shimadzu UV3101-PC spectrophotometer was used to perform these measurements. Once RT measurements were completed, a closed cycle refrigerator system was employed for measuring transmission at 13 K.

Figure 2 depicts the RT absorption spectrum of this sample with corresponding absorption coefficients and spectral deconvolution of the known color centers$^6$ in the crystal. Similar $F_2^{**}$-like centers were described by Ter-Mikirtychev$^8$, however, the fluorescence properties described in that work are completely different from those depicted here.

![Absorption Spectrum](image-url)

**Figure 2** Overall absorption spectrum of the sample measured at RT.

Figure 3 illustrates the 13 K absorption spectra of the sample and corresponding deconvolution. As expected, the centers show significant narrowing with decreased temperature and corresponding increase in absorption coefficients. Also, zero-phonon lines are present for other centers in the sample; however, no zero-phonon lines were evident at 13 K for either of the $F_2^{**}$-like centers.

Fluorescence measurements of the sample were also performed at RT and 13 K using several different excitation wavelengths. The wavelengths include 532 nm, 632 nm, 747 nm, 760 nm, and 780.35 nm provided by the second harmonic of a Q-switched Nd:YAG, the Raman shifted ($D_2$) second harmonic of a Q-switched Nd:YAG, Alexandrite (747 and 760 nm), and the second harmonic of a Raman shifted ($D_2$) fundamental frequency of the Q-switched Nd:YAG laser, respectively. The detection system included a fiber optic bundle for collection of fluorescent photons, an Acton Research 750 spectrometer, InGaAs detector (for spectral region 800-1700 nm), and a PMT detector (for spectral region 400-1100 nm). The entire system was calibrated using an Oriel Instruments tungsten halogen lamp of known spectral output.
Figure 3 Absorption spectrum at 13 K.

Figure 4 depicts the RT fluorescence of the sample under 532 and 633 nm excitation. Inspection of the RT absorption curves shows that excitation at these wavelengths primarily excites only the standard $F_2^{**}$ center; the result is the fluorescence curve of only that center.

Figure 4 RT fluorescence under 532 nm & 632 nm excitation.

Figure 5, also at RT, illustrates the same sample under 760 nm excitation from the Alexandrite laser. From Fig. 2 it is clear that excitation at 760 nm is absorbed by both $F_2^{***}$-like centers, which leads to fluorescence emission from both, as is clear from the deconvolution of the resulting fluorescence spectrum.
Figure 5 RT fluorescence under 760 nm excitation. (A) F2+** center, (B) F2+**-like center, (C) superposition of deconvolution.

Low temperature fluorescence of the sample is depicted in Figure 6 under 747 nm and 760 nm. From Fig. 3, excitation at 747 nm will continue to be absorbed by both centers; while under 760 nm excitation, the standard F2+** center is no longer absorbing. It is clear that under the 747 nm excitation, both types of centers are present in the fluorescence emission; however under 760 nm excitation, the fluorescence is only that of the F2+**-like center. Again, no zero-phonon lines were evident for these centers at low temperatures.

Figure 6 Low-T fluorescence under (A) 747 nm and (B) 760 nm excitation.

Finally, measurements of the kinetics lifetimes of these two centers were performed. Figure 7 illustrates the RT and 13 K lifetime of the standard center (under 532 and 632 nm excitation). The exponential curves describe the lifetimes as (A) $\tau = 32\text{ns}$ and (B)&(C) $\tau = 21\text{ns}$, which results in a quantum efficiency of $\eta=66\%$. 

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The kinetics measurements for the F$_2$**-like color center, measured at 1100 nm using an amplified silicon detector with temporal response $\sim$400 ps, are shown in Figure 8 along with the excitation pulse from the second harmonic of the Raman shifted (D$_2$) fundamental frequency of a Q-switched Nd:YAG laser at $\lambda$=780.35 nm. Due to the two nonlinear processes involved in the formation of the excitation pulse, it exhibits very fast rise and decay time (FWHM of 4 ns), as well as some fringe effects at its top. The figure clearly shows that the pulse duration of the excitation pulse (A) is not a factor when calculating the lifetime of the center because the excitation pulse has already terminated as the fluorescence signal (B) is still in the developing process.

All of the spectroscopic results are shown in Table 1.
Table 1 Spectroscopic data summary

<table>
<thead>
<tr>
<th>Property</th>
<th>$F_2^{**}$ Centers</th>
<th>$F_2^{**}$-like Centers</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT Absorption Maximum</td>
<td>4.86 x 10^14 Hz</td>
<td>3.68 x 10^14 Hz</td>
</tr>
<tr>
<td>RT Absorption FWHM</td>
<td>3819 cm⁻¹</td>
<td>3591 cm⁻¹</td>
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<tr>
<td>Max. Absorption Coef.</td>
<td>1.49 cm⁻¹</td>
<td>0.38 cm⁻¹</td>
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<tr>
<td>13 K Absorption Maximum</td>
<td>4.91 x 10^14 Hz</td>
<td>3.84 x 10^14 Hz</td>
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<tr>
<td>13 K Absorption FWHM</td>
<td>3183 cm⁻¹</td>
<td>2834 cm⁻¹</td>
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<tr>
<td>Max. Absorption Coef.</td>
<td>1.66 cm⁻¹</td>
<td>0.61 cm⁻¹</td>
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<tr>
<td>RT Fluorescence Max.</td>
<td>3.30 x 10^14 Hz</td>
<td>2.74 x 10^14 Hz</td>
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<tr>
<td>RT Fluorescence FWHM</td>
<td>2174 cm⁻¹</td>
<td>2274 cm⁻¹</td>
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<tr>
<td>13 K Fluorescence Max.</td>
<td>3.35 x 10^14 Hz</td>
<td>2.75 x 10^14 Hz</td>
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<tr>
<td>13 K Fluorescence FWHM</td>
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<td>RT Lifetime</td>
<td>21 ns</td>
<td>To Be Measured</td>
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<tr>
<td>13 K Lifetime</td>
<td>32 ns</td>
<td>~ 5 ns</td>
</tr>
</tbody>
</table>

4. LASER OPERATION

Figure 9 illustrates the RT tuning range of the LiF:F$_2^{**}$ under 740 nm excitation from an Alexandrite laser (~145 mJ pulse energy and $t_{\text{pulse}}$ ~ 50 ns).

![Figure 9 Tuning curve of LiF:F$_2^{**}$ laser at 300 K under 740 nm excitation](image)

Figure 10 depicts the input/output dependencies of the LiF:F$_2^{**}$ laser at RT using excitation from (1) 740 nm, (2) 683 nm, and (3) 633 nm. Line (4) is simply a linear fit. The laser radiation at 683 nm is provided by the Raman shifted (H$_2$) second harmonic of a Q-switched Nd:YAG laser.

![Figure 10 Laser output dependencies](image)
The data represented in Figures 9 and 10 were published by Dergachev and Mirov in [5]. The results in this previously published work described the available tuning curve and input/output dependencies of the laser under non-selective cavity oscillation. The spectroscopic information provided in the previous section describes the means by which this broad tuning range can be obtained: simultaneous excitation of two F$_2$$^{**}$-like color centers in the active medium using proper pumping wavelength.

To obtain simultaneous Second Harmonic Generation (SHG) of ultrabroadband radiation in a single nonlinear crystal, one needs to satisfy the phase matching conditions for all oscillating wavelengths and to fit angular distribution of oscillating wavelengths to the angular dependence of phase matching condition in nonlinear crystal \(^4\). Calculations of dependencies of phase matching angles outside nonlinear medium for LiIO$_3$ and BBO crystals have been performed.
Analysis shows that the spectral angular dependencies for these crystals have a small slope and can be fitted to ultrabroadband output angular-wavelength distribution. Hence, these crystals can be used for white light lasing by means of efficient SHG of continuous spectral distribution (800-1300 nm). The angular dependency in LiIO₃ is shown in Figure 11 as well as the approximation of the fit with a single spherical lens.

Figure 12 illustrates a current laser cavity, whose schematic is shown in Figure 1, by which superbroadband laser operation has been successfully realized under 633 nm excitation. As we have shown, under 633 nm excitation we will excite only the standard F₂⁺⁺ color center in the material. Initial superbroadband laser experiments have been successfully realized with this excitation wavelength. The spectral region of F₂⁺⁺ (~880-1080 nm) was achieved and directed into a lithium iodate (LiIO₃) non-linear crystal for subsequent second harmonic generation into the visible spectral region: ~440-540 nm.

Finally, we are able to demonstrate in Figure 13 superbroadband laser output, from 633 nm pumping, after frequency doubling into the visible spectral region using LiIO₃.
5. SUMMARY

Previous results using a non-selective cavity, the LiF:F$_2^{**}$ active medium, and Alexandrite laser excitation have shown that this crystal is tunable from ~820-1210 nm; however, no reasonable explanations of such a broad tuning range were provided.

We have demonstrated here the mechanism for an ultrabroadband tuning range in a single solid state laser crystal, LiF:F$_2^{**}$, using a single pump laser source. The spectroscopic results indicate the presence of two overlapping F$_2^{**}$-like color centers present in the medium. The spectral region of overlapping corresponds to the Alexandrite pump wavelength ~740 nm at RT. With proper selection of the input mirror and diffraction grating, it should be possible to realize ultrabroadband lasing from both centers that will correspond to nearly the entire (superposition) emission spectra, namely ~800-1300 nm. This research is currently underway.

REFERENCES