All Solid State Laser System, Continuously Tunable Over 0.2-10 Micron Spectral Range

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ABSTRACT

An efficient room temperature all-solid-state laser system continuously tunable in the 0.2-10 μm spectral range has been developed. It is based on the alexandrite laser pumped $LiF:F_2^{+**}$ color center laser system. The alexandrite - $LiF:F_2^{+**}$ color center laser combination system has been shown to be a suitable drive source for a number of efficient nonlinear processes, including harmonic, sum-frequency and difference-frequency generation.

Keywords: laser, color center, tunable laser, alexandrite laser, nonlinear up- and downconversion, sum frequency and difference frequency generation.

I. INTRODUCTION

Tunable solid state lasers have always been of interest to the scientific community. Application areas include analytical chemistry and environmental monitoring, atmospheric remote sensing, medicine, remote laser induced emission spectroscopy, optical communications, and different wavelength specific military operations.

The following desirable properties, such as high thermal conductivity, high optical damage threshold, compactness and operational reliability, make these sources very attractive for practical applications. At the present time, the most widely used lasers are Ti:sapphire, Alexandrite, Cr:LiSAF, Cr:LiCAF, Cr:Forsterite, and Cr:YAG, which provide laser radiation in the 0.7-1.6 μm range. The main drawbacks and disadvantages of existing solid state lasers include: (1) a limited tuning range, (2) difficulty in obtaining crystals of high optical quality, and (3) relatively low emission cross sections as compared with dye lasers.

We hope that our recent research [1,2,3,4,5] demonstrates that LiF color center lasers (CCLs) can serve as a reliable alternative to either dye, Ti and Cr activated crystalline laser systems, or optical parametric oscillators (OPOs). The advantages over dye lasers include wider wavelength tuning range, compactness, long operational lifetime, rigidity and ease of handling. Compared to OPOs and impurity based lasers, CCLs are relatively insensitive to the quality of the cavity's optical elements, and to the spatial, angular and spectral characteristics of the pump source.

LiF color center active elements combine unique spectroscopic, oscillation, thermooptic, and operational properties [4,5]. Room temperature LiF color center lasers yield very high conversion efficiencies (tens of % for nanosecond pump pulse duration). Due to the quasi-homogeneous broadening of color center gain profile the CCL have narrow spectral widths, achievable virtually without power loss while preserving a wide region of continuous tuning (up to 2500 cm^{-1}).

LiF crystals are moisture resistant, mechanically durable, and easy to handle. The crystals are isotropic with a face centered cubic symmetry. High-quality crystals can be grown using the Kyropulos technique, which is a simple, fast and convenient technique for producing large (for instance, ∅ 400x200 mm) commercial-grade crystal boule. In addition, LiF solubility in water of 0.1 g/100g H_2O is acceptable for most practical applications. LiF also has good thermooptic characteristics, with a temperature-derivative refractive index, $dn/dT = -1.2 \cdot 10^{-5}$ °C^{-1} and a high thermal conductivity

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coefficient α = 14 W/m°C (at 300 K). This is even higher than that of YAG crystal (13 W/m°C). The critical thermal power of LiF (thermal power at which the value of thermal aberration becomes comparable with the laser wavelength) is $P_{cr} = \lambda \cdot \alpha \cdot |dn/dT|^{-1}$ = 1.25 W at λ = 1.06 μm. This value is three to four orders of magnitude higher that of dye solutions [4,5], and very close to that of YAG (1.3 W).

In our recent studies of LiF:F_2^{+**} color center lasers [1,3] we showed that the LiF:F_2^{+**} laser exhibits excellent photo- and thermostable operation at room temperature when pumped by the radiation of an alexandrite laser and can provide efficient high power lasing tunable in 800-1200 nm spectral range. These results allowed us to build a reliable LiF:F_2^{+**} - alexandrite laser system which can be continuously tuned in 720-1200 nm range and serve as a suitable drive source for a number of efficient nonlinear processes, including harmonic, sum frequency and difference frequency generation.

II. SPECTROSCOPIC CHARACTERIZATION OF LiF:F_2^{+**} CRYSTALS

Basic spectroscopic characterization of LiF:F_2^{+**} crystals was performed elsewere.[1,3] The spectroscopic data have been refined in this article by thorough application of instrumental correction.

Absorption measurements were made with a Shimadzu 3401UVPC spectrophotometer. Deconvolution of the overlapped absorption spectra was performed with Peakfit software.

For measurement of luminescence spectra we used a spectroscopic set-up based on Acton Research 750 monochromator with R406 Hamamatsu photomultiplier or InGaAs detectors. Spectral sensitivity of the spectroscopic detection system in 700-1500 nm was measured with the help of calibrated tungsten halogen lamp (Oriel). These data were used for correction of measured luminescence spectra.

F_2^{+**} color centers feature quasi-homogeneously broadened absorption and luminescence bands (see Fig.1). Absorption and luminescence spectra are described by Gaussian profiles with central frequencies at ν_o^a=5.00x10^{14} and ν_o^e=3.31x10^{14} Hz, FWHM $\Delta\nu_a$ =1.65x10^{14} Hz and $\Delta\nu_e$ =6.4x10^{13} Hz, and absorption, (σ_o^a=7x10^{-17}) and emission (σ_o^e=1.2x10^{-16} cm^2) cross sections, respectively. Developed LiF:F_2^{+**} crystals feature a high concentration of the active F_2^{+**} centers ~4-8x10^{16} cm^{-3} at a low level of losses (contrast ~40).

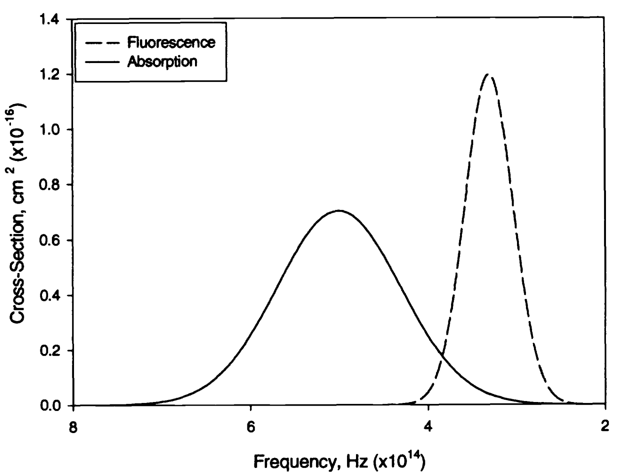

Fig.1 Absorption and luminescence cross-section of LiF:F_2^{+**} versus frequency

III. PROPERTIES OF THE ALEXANDRITE LASER PUMPED LiF:F_2^{+**} LASER

In all the frequency conversion experiments described in this article, the fundamental harmonic is obtained from a LiF:F_2^{+**} oscillator described in reference[1]. It is based on an input dichroic mirror and a corner reflector consisting of a Littrow mount plane diffraction grating (1200 g/mm) and totally reflective mirror installed orthogonally to the grating. The efficiency of diffraction in the first order was 30%. A zero order of diffraction served for output. Frequency tuning was performed by turning the corner reflector about the axis perpendicular to the plane of grating dispersion, providing unchanged direction of propagation of the output radiation. Experimental tuning curve extended from 800 to 1210 nm. Three-four different input mirrors with a transparency of 80-90% at pump wavelength (730-750 nm) and reflectivity of 100% at oscillation wavelength were necessary to cover the whole region of tunability. The LiF:F_2^{+**} active element was Brewster cut with a length of 4 cm. Its absorption coefficient at 740 nm was 1.2 cm^{-1} and passive losses in the maximum of emission band were 0.01 cm^{-1}. The pulse energy of alexandrite pump laser was up to 80 mJ. The pump pulse duration was about 70 ns. The LiF:F_2^{+**} resonator provided a near-diffraction-limited spatial beam with a Gaussian intensity distribution, horizontal polarization and propagation factor, M^2 of about 1.7. The spectral linewidth of the output radiation was about 1 cm^{-1}.

The temporal dynamic of the LiF:F_2^{+**} oscillation was investigated. Two avalanche germanium photodiodes detected alexandrite and LiF output pulses that were measured by TEKTRONIX 2430A digital oscilloscope. The rise time of the system was 3-5 ns. Figure 2 shows typical temporal shapes of the pump and output pulses for different wavelengths of LiF laser.

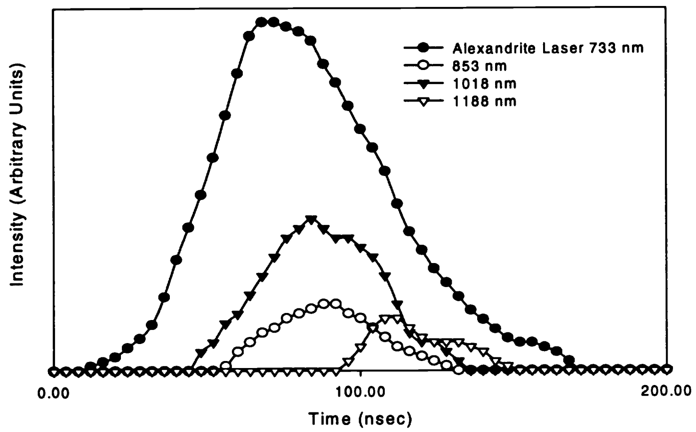

Fig.2 Typical temporal shapes and delays between the pump and LiF:F_2^{+**} oscillating pulses.

One can see that there is a small temporal delay of 30-65 ns (with respect to the 50 mJ pump pulse) of tunable laser pulse depending on the oscillating wavelength. The minimum temporal delay corresponds to the central frequency of the tuning curve featuring the highest cross section of F_2^{+**} center emission. The delay is increased at the edges of the tuning curve and reaches 65 ns for the long wavelength tail of emission around 1200 nm. It is important to note that these temporal delays are less than the pump pulse duration. This temporal overlapping of the pump and oscillation pulses within the whole region of LiF:F_2^{+**} laser tunability allows observing efficient sum- and difference frequency generation. The proposed alexandrite-color center laser combination has many positive features of a solid state dye like laser system. One of the most important features relates to the fact that the system exhibits virtually no temporal delay between pump and output pulses. This temporal overlap of pump and generated CCL pulses for the entire range of tunability provides an easy extension to the middle IR region through difference frequency generation of the pump and CCL output.

IV. LASER SYSTEM DESIGN

A diagram of the all-solid-state laser system continuously tunable in the 0.2-10 μm spectral range is shown in Fig.3. It is based on the tunable alexandrite laser. Radiation of the alexandrite laser is split into 3 channels.

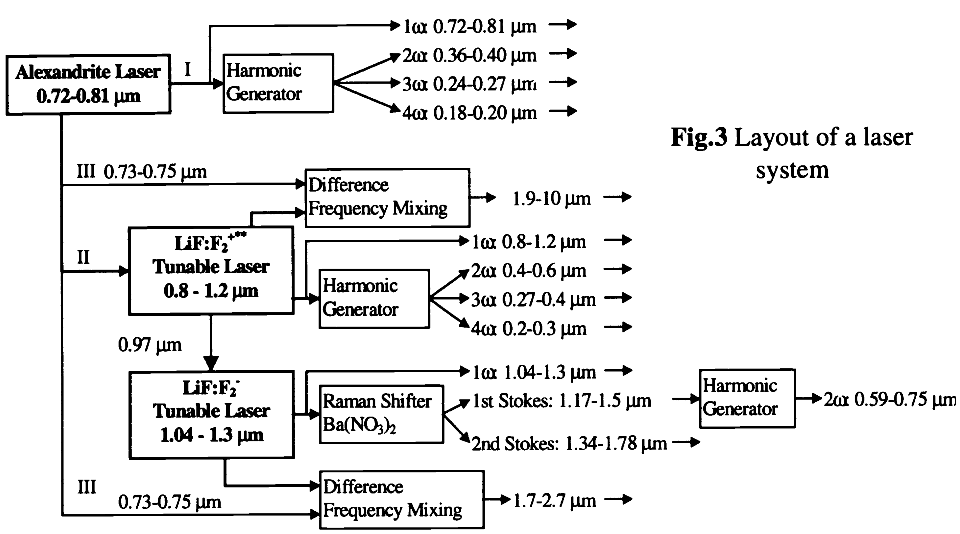

Fig.3 Layout of a laser system

Channel 1 provides the spectral output of alexandrite laser tunable in the 0.72-0.81 μm range. It is based on the Light Age PAL101 laser system.

Channel 2 represents efficient extension of the tunability of alexandrite laser into 0.8-1.2 μm spectral region. It is based on the tunable LiF:F_2^{+**} laser pumped by the radiation of the alexandrite laser within a spectral interval 0.73-0.75 μm. LiF:F_2^{+**} laser spectral output is further converted to the UV-visible spectral region by means of frequency doubling, tripling, and quadrupling – sum frequency generation of the fundamental and third harmonic.

Range of tunability is also extended in the 1.04-1.3 μm spectral range by means of $LiF:F_2^-$ laser, pumped by the monochromatic or broadband radiation of $LiF:F_2^{+**}$ laser (about 60% efficiency with respect to $LiF:F_2^{+**}$ output & 0.96-0.97 μm). Further efficient extension of the $LiF:F_2^-$ tuning region into the near-IR spectral (1.17-1.5 μm - 1 Stokes, and 1.34-1.78 μm - 2 Stokes) region is achieved by using solid state Raman shifters based on $Ba(NO_3)_2$ crystals. Tunability in the yellow green spectral range 0.59-0.65 μm is provided by means of frequency doubling of the first Stokes component of the $LiF:F_2^-$ radiation.

Channel 3 provides spectral output of alexandrite laser in the region 0.73-0.75 μm and is used for frequency downconversion - difference frequency generation of this radiation with a tunable radiation of the $LiF:F_2^{+**}$ laser (1.9-10 μm) and $LiF:F_2^-$ laser (1.7-2.7μm).

V. FREQUENCY UPCONVERSION

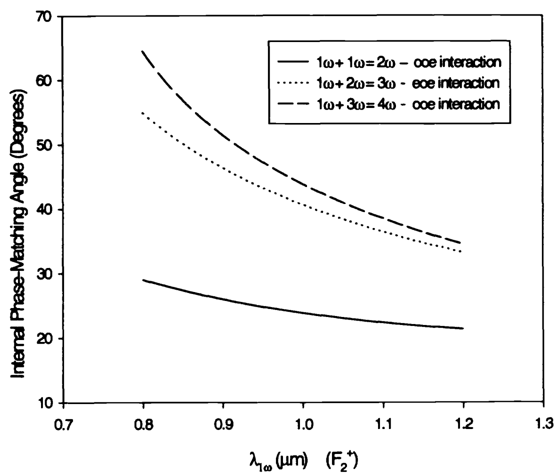

Fig.4 Type I SHG, type II THG, and type I SFM phase matching angles as a function of $LiF:F_2^{+**}$ laser wavelength for BBO crystals.

At the present time, a number of efficient nonlinear crystals for harmonics generation are available. In our studies we concentrated only on BBO crystals. These crystals are readily available commercially. The main advantages of these crystals are a very high optical damage threshold and high nonlinearity, which allows for attainment of high conversion efficiency.

Results of our calculations of phase matching angle θ versus wavelength λ of the light wave propagating in nonlinear crystal for second harmonic generation (SHG, type I), third harmonic generation (THG, type II), and sum frequency mixing (SFM, type I, 1ω + 3ω = 4ω) are illustrated in Fig.4 (solid, dotted, and dashed curves, correspondingly). The relatively small change in phase-matching angles with wavelength allows one crystal to be used over the entire LiF tuning range, a valuable property in many applications. One BBO crystal 6x4x7 mm, cut at angle θ_c=25° with respect to Z axis of the BBO crystal can be angle tuned over the entire 800-1200 nm tuning range for SHG. For THG and SFM processes BBO crystals were 10x4x7 mm in size and were cut at angles 45° and 49°, respectively.

The experimental set-up for SHG, THG, and SFM is shown in Fig.5. Parameters of alexandrite and LiF CCL are described in section II. We used two different schemes of the CCL cavity. In the first one the dispersive element was based on a corner reflector described below in section IV and shown in Fig.7. The second scheme utilized only Littrow mount diffraction grating. A zero order of diffraction served for output. Tuning was arranged by rotation of the grating about the axis normal to the plane of grating dispersion and spatial positioning of the beam was preserved by adjusting of mirror M_2 and directing the beam through the set of two apertures A_1 and A_2. After collimating the beam with Galilean beam collimator, incorporating a negative input lens L_1 followed by a positive collimating lens L_2, and compressing with 3x beam compressor the output beam featured a propagation factor of about 1.2. A CCD camera (Princeton Instruments) measured wavelength of the laser radiation through 750-mm Acton Research Corporation monochromator SpectraPro-750. Output power of the laser radiation after nonlinear upconversion was measured by Molectron PM10/V1 powermeter and J4-09 joulemeter-Tectronix digital oscilloscope combination.

The parallelepiped shaped BBO nonlinear crystal (6x4x7mm, θ_c=25°) with antireflection coating was used for SHG. The output beam of the $LiF:F_2^{+**}$ laser after collimation was focused into the nonlinear crystal by using a spherical lens L_3 (see Fig.5). The filter at the output of the nonlinear crystal blocked tunable pumping radiation. The lens focal length value was optimized and chosen f=100 mm for higher conversion efficiency to SHG and to prevent nonlinear crystal laser damage. Fig.6 (2ω) shows a typical wavelength dependence of the SHG output power versus wavelength. SHG was achieved in 400-600 nm spectral range with diffraction grating and 420-600 nm - with a corner reflector dispersive element. It was due to the low reflectivity of the corner reflector mirror in the 800-840 nm spectral region. Power measurements yielded pulses with a peak power of 80 kW at 500 nm and pulse duration of about 50 ns. This corresponds to an average power of about 80 mW. The SHG efficiency with respect to the fundamental $LiF:F_2^{+**}$ radiation reached 38% at the maximum of the tuning curve (~500 nm).

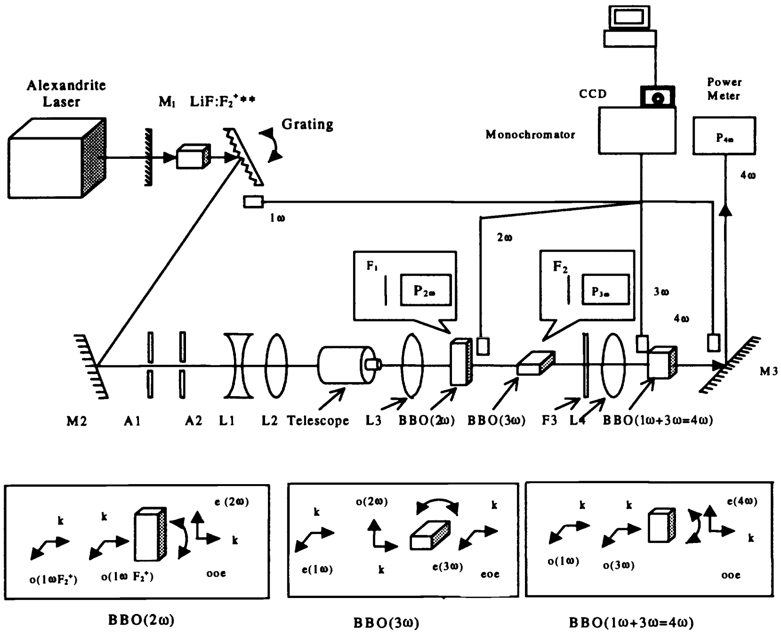

Fig. 5. Schematics for 2nd, 3rd harmonic generation, and 1ω and 3ω sum frequency mixing of LiF:F_2^{+**} laser

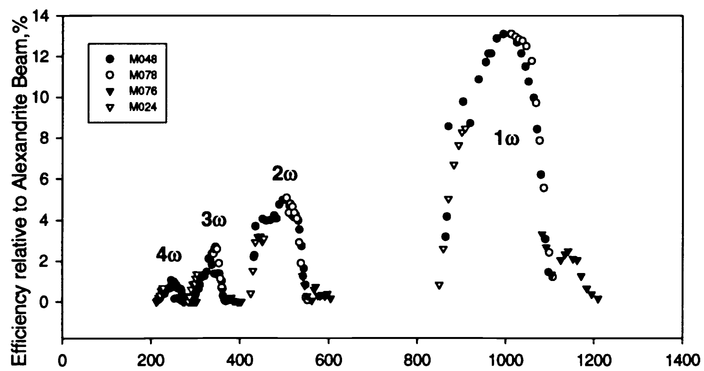

Fig.6 Fundamental, SHG, THG, and SFG conversion efficiency with respect to alexandrite laser pump energy as a function of wavelength for LiF:F_2^{+**} laser (corner reflector dispersive element). Inset shows notation of experimental points for four different input mirrors.

BBO nonlinear crystal (10x4x7mm, θ_c=45°) was used for THG of LiF:F_2^{+**} tunable radiation. A type II (eoe) interaction was tested. The reason why we used a type II THG crystal following a type I SHG crystal relates to the fact that the orthogonal polarization states exiting the SHG stage are correctly oriented for the type II mixing process in the THG crystal. The use of a type I THG BBO crystal is not convenient since one must rotate the polarization of the fundamental and second harmonic, such that the two are parallel.

THG is realized as sum frequency mixing of the fundamental (e) and second harmonic (o) oscillation of the LiF:F_2^{+**} laser. The tunable fundamental radiation was focused into SHG and THG BBO crystals using a spherical lens L_3 (Fig.5) with f=1 m. The input and output facets of crystals had antireflection coatings for visible and ultraviolet spectral ranges, respectively. The filter F2 (see Fig.5) placed after nonlinear crystal blocked the visible radiation and transmitted UV THG radiation. Fig. 6 (3ω) demonstrates wavelength dependence of the UV radiation output power as a function of THG wavelength. Tuning within a spectral region 270-400 nm was achieved. The maximum conversion efficiency with respect to fundamental LiF:F_2^{+**} radiation is close to 16% for 330-350nm spectral range. The maximum pulse energy ~1.5 mJ corresponds to pulse peak power ~3x10^4W.

Another BBO nonlinear crystal (10x4x7mm, θ_c=49°) was used for frequency quadrupling of LiF:F_2^{+**} tunable radiation. LiF:F_2^{+**} frequency quadrupling is realised as sum frequency generation (SFG) of the fundamental (o) and third harmonic (o) oscillation of the LiF:F_2^{+**} laser (ooe interaction). A type I (ooe) interaction was tested (see Fig. 5). The reason why we used a type I crystal following a type I SHG and type II THG crystals relates to the fact that the orthogonal polarization states exiting the THG stage are correctly oriented for the type I mixing process in the BBO frequency quadrupling crystal.

1ω and 3ω SFG approach for frequency quadrupling of the laser radiation is more advantageous then direct SHG of the frequency doubled radiation of the LiF:F_2^{+**} laser. The reason is a ~210 nm short wavelength limit for fourth harmonic generation due to vanishing of SHG interaction for wavelengths shorter than 420-410 nm (phase matching angle θ>90°).

Horizontally polarized fundamental and third harmonic outputs of the LiF:F_2^{+**} laser were focused into the BBO crystal using a spherical lens L_4 (see Fig.5). SHG output was blocked by filter F_3. The dichroic mirror M_3 was used for separation fundamental, tripled, and quadrupled radiations of the laser. It was transparent for 1ω and 3ω frequencies and reflective for 4ω. The lens focal length value was optimized and chosen f=100 mm for higher conversion efficiency to SFM. Fig.6 (4ω) shows a typical wavelength dependence of the SFG output power versus wavelength. SFG was achieved in 200-300 nm spectral range. Power measurements yielded pulses of peak power of about 15 kW and average power of 15 mW. Maximum conversion efficiency was up 50% with respect to the THG output of the LiF:F_2^{+**} laser.

VI. FREQUENCY DOWNCONVERSION

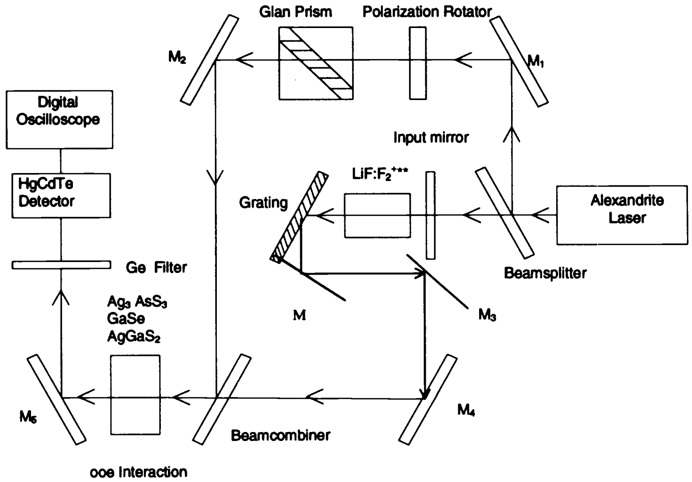

Fig.7 Experimental Set Up for Difference Frequency Generation

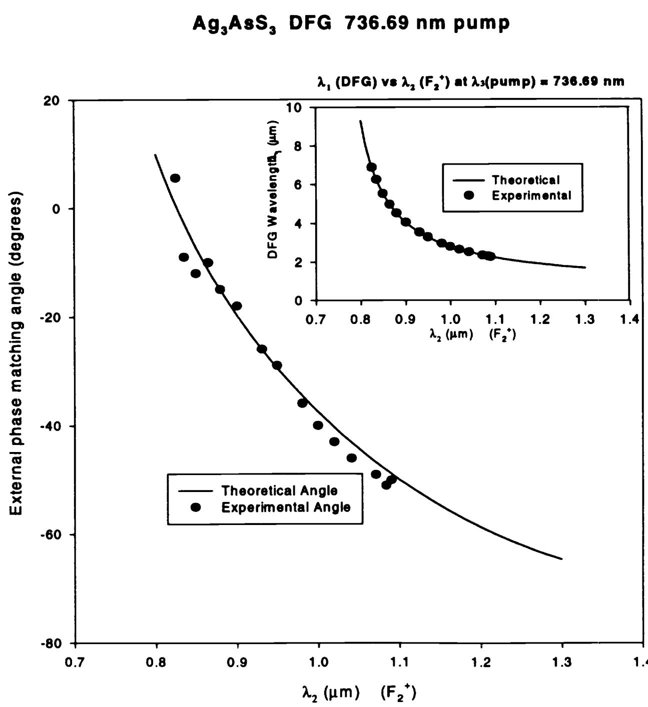

Fig.8 The external phase matching angle for Ag_3AsS_3. Inset: DFG wavelengths as a function of $LiF:F_2^{+**}$ wavelengths at a pump wavelength, λ_3 of 736.7 nm.

The extension of the $LiF:F_2^{+**}$ laser tuning region into the middle infrared region (channel 3, figure 3) was made possible via difference frequency generation (DFG) in GaSe, $AgGaS_2$ and Ag_3AsS_3. Fig. 7 depicts the setup used for the collinear DFG in the above crystals, between the outputs of the alexandrite and the alexandrite-pumped $LiF:F_2^{+**}$ lasers.

The interaction was of type I (ooe) phase matching (where the first symbol refers to the wave with the lowest frequency). The $LiF:F_2^{+**}$ laser is pumped with the fundamental output of the alexandrite laser PAL 101 (Light Age Inc.) with an energy of 70-100 mJ, a pulsewidth of 50 ns and a repetition rate of 20 Hz. The $LiF:F_2^{+**}$ laser output energies of up to 20 mJ (measured, using a Molectron, PM10V1 power meter) were obtained in the 0.8-1.2 μm spectral region. The rest of the alexandrite laser radiation was directed into the nonlinear crystal (GaSe, $AgGaS_2$ or Ag_3AsS_3), where it is mixed with the tunable near IR $LiF:F_2^{+**}$ laser radiation.

The polarization of the alexandrite laser beam (diameter ~ 3mm) was rotated (from horizontal to vertical), and the output power, simultaneously attenuated, using a polarization rotator-Glan prism combination. The alexandrite laser had a power density of about 3 MW/cm^2.

The generated middle IR radiation was detected by means of a cryogenically cooled HgCdTe detector or Joulemeter (J3S-10) combined with a digital oscilloscope. A Ge plate was used to filter out near IR input radiations. The IR wavelength verification was performed with the use of LiF, sapphire, MgF_2 and CaF_2 plates as transmission filters. Results of the experiments with GaSe and $AgGaS_2$ nonlinear crystals are presented elsewhere [6].

Figure 8 shows the theoretical (solid line, calculated for type I DFG, using the dispersion relation [7]) and the experimental (dots) angular tuning curves in Ag_3AsS_3, for the pump radiation at a wavelength, λ_3 of 736.69 nm versus the output of the alexandrite-pumped $LiF:F_2^{+**}$ laser. Inset, depicts the wavelengths of the infrared DFG signal, λ_1 versus the wavelengths of the tunable $LiF:F_2^{+**}$ radiation, λ_2. The theoretical and experimental phase matching curves are in a good agreement over the entire range of the middle IR output. Inset, also shows that with the use of the Ag_3AsS_3 (proustite) crystal, we experimentally obtained DFG signals over the spectral region 1.9 – 7 μm. Output energies, up to 250 μJ at a wavelength of 3 μm, and energy efficiency of 3% were obtained with a very good pulse-to-pulse stability.

In principle, by mixing the radiations of the alexandrite and the alexandrite-pumped $LiF:F_2^{+**}$ color center lasers, one can cover the spectral range, 1.9 – 10 μm. In our experiment practical realization of DFG in the spectral region 7-10 μm was not possible due to properties of the beamcombiner (Fig.7) that didn't transmit LiF laser radiation shorter than 0.83 μm.

VII. CONCLUSIONS

The proposed alexandrite-color center laser combination has many positive features of a solid state dye like laser system. These advantages include high gain coefficients, broad homogeneous gain profile, low threshold and highly efficient single mode operation with extremely narrow spectral outputs, wide wavelength tunability, compactness, long operational lifetime, rigidity, ease of handling, and insensitivity to the quality of the cavity's optical elements, and to the spatial angular and spectral characteristics of the pump source. The system also exhibits virtually no temporal delay between pump and output pulses. This temporal overlap of pump and generated color center laser pulses for the entire range of tunability provides an easy extension to the middle IR region through frequency difference mixing of the pump and CCL output.

The proposed all solid state alexandrite laser based system continuously tunable in 0.2-10 μm spectral range offer the simplest and most economical route to tunable narrow-linewidth performance for many practical applications.

VIII. ACKNOWLEDGEMENTS

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