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Amplified spontaneous emission of F_2^- color center in LiF crystal

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Abstract. Intense, directional, and narrow-bandwidth 1130-nm emission was generated from LiF crystal at room temperature by the pumping with radiation in the wavelength region of the absorption band of the F_2^- color center. The emission was observed by the pumping with radiation of pulse energy above 5 mJ, and the divergence angle of the beam was about 16 mrad. This emission is attributed to the amplified spontaneous emission by the F_2^- center.

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In a laser operating as an amplifier, population inversion is achieved in the active material. As the inversion is increased, a number of different processes begin to depopulate the upper level. For example, amplification by the process of not only stimulated emission but also spontaneous emission is generated. The latter process, which is called amplified spontaneous emission (ASE), is considered the randomly fluctuating noncoherent amplifier noise [1, 2]. The ASE is highly directional and narrow-bandwidth radiation of high intensity and has some spatial and temporal coherence when the population inversion is achieved in high-gain materials under strong pumping conditions. Therefore the ASE is useful because of application to mirrorless lasers (where a powerful laser-like beam is generated without installing the oscillator elements) as in the cases of conventional nitrogen and excimer lasers, although the coherent properties of the ASE-type laser are conceptually different from the usual laser radiation [3, 4].

Color centers are lattice vacancies trapping electrons or holes, which are produced mostly in ionic crystals [5]. The color centers used for laser oscillation have the homogeneously broadened emission bands with high oscillator strengths and high quantum efficiencies. Therefore a laser using color centers exhibits the characteristics of wide tuning range, high output power and low-threshold pump power [6–9]. There are various laser-active color centers, but the ASE of color centers has been observed in only the F_2 and F_3^+ color centers in LiF crystal, where two electrons are trapped by two and three negative-ion vacancies in the nearest neighbor positions, respectively [10–12]. We report the observation of ASE by the F_2^- color center, which has three electrons at two negative-ion vacancies in the nearest neighbor positions, in LiF crystal at room temperature.

A single crystal of LiF crystal (the size is $54.80 \times 10.04 \times 9.72$ mm) was colored by the irradiation with electron beam at room temperature. The colored LiF crystal was purchased from General Physics Institute Company, Moscow. Although this crystal is remarkably big, the coloration is made homogeneously throughout the whole crystal. This was checked by the measurement of the absorption spectrum at different places in the crystal.

The two crystal surfaces perpendicular to the 54.80-mm length were polished and longitudinally pumped with the laser radiation from a Surelite optical parametric oscillator (OPO). The OPO was pumped with the third harmonic (i.e. 355 nm radiation) of a Surelite I-10 pulsed YAG:Nd^{3+} laser operated at a repetition rate of 10 Hz. The pulse duration of the YAG laser is 6 ns, and the maximum output energy of the 355 nm laser is 100 mJ per pulse. The spotsize of the OPO laser beam is about 5 mm in diameter. The maximum output energy is 15 mJ per pulse at 930 nm laser radiation. The output emission from the LiF crystal was viewed using an IR viewer and an IR sensor card. The emission power was measured with a Gentec TPM-310 power meter. The emission spectrum was detected with an Advantest Q8381A spectral analyzer which has a sensitivity in a spectral region of 400–1750 nm. Absorption spectra of the crystal were measured with a Shimadzu UV-3100 spectrophotometer.

Figure 1 shows the absorption spectrum, at 296 K, of the LiF crystal used in the ASE experiment. Absorption bands due to the F_2^- and F_3^- color centers [13, 14] are observed at 960 and 800 nm, respectively, in addition to the strong absorption bands due to the F_2 and F_3^+ centers [15, 16] which have peaks at 445 and 448 nm. The 960 nm F_2^- absorption band has an absorption coefficient of $1.47\ \mathrm{cm}^{-1}$ at the peak position and has a half width of $1350\ \mathrm{cm}^{-1}$, which were estimated by subtracting the background. The concentration of color centers can be calculated from the absorption spectrum using Smakula's equation [17]. The oscillator strength of the 960-nm F_2^- absorption band has been estimated to be 6.5×10^{-3} [13]. Using this value and the observed 960-nm

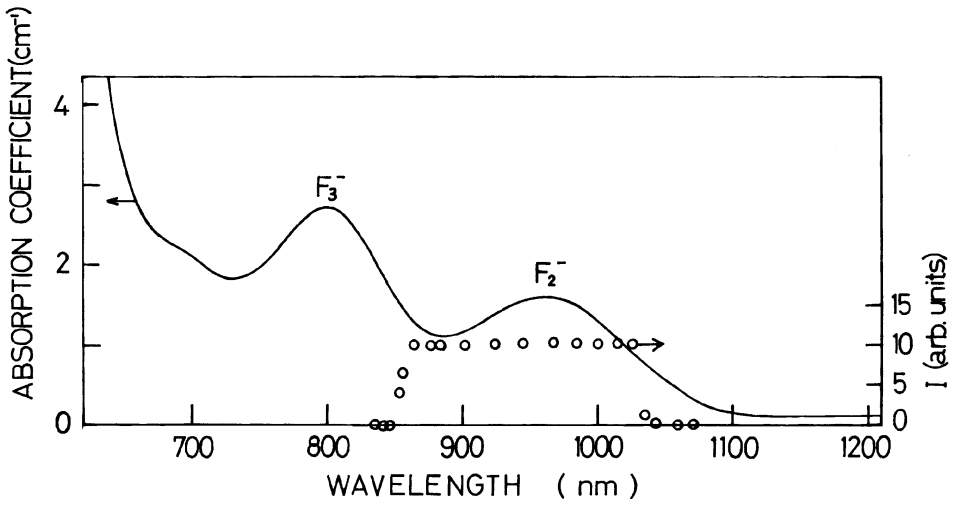

Fig. 1. Absorption spectrum (*solid line, left-hand scale*) of an electron-beam-irradiated LiF crystal at 296 K and the excitation spectrum (*open circle, right-hand scale*) for the amplified spontaneous emission of the F_2^- center where the pump radiation with pulse energy of 9 mJ was used. *I*: emission intensity

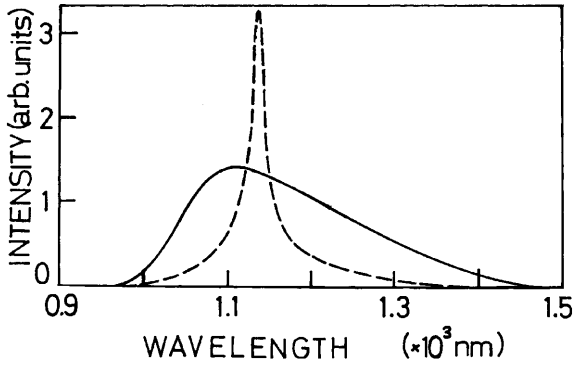

Fig. 2. Emission spectra of the F_2^- center in LiF crystal which was excited with the 930-nm laser radiation with pulse energies of 1.5 mJ (*solid line*) and 9 mJ (*broken line*) at 296 K. The amplified spontaneous emission (shown by *broken line*) was measured at the distance 12 cm away from the crystal

absorption spectrum, we estimated that the concentration of the F_2^- centers is 2.8×10^{17} cm^{-3}.

The F_2^- luminescence band with a peak at about 1120 nm [8, 9] was observed by exciting the crystal with the OPO radiation of various wavelengths of 840–1070 nm which correspond to the F_2^- absorption band region. The luminescence spectrum is shown in Fig. 2 (see the solid line). When the pulse energy of the OPO radiation was increased and beyond 5 mJ per pulse, strong emission of highly directional light (i.e. ASE) emerged from the output surface of the crystal. The spot of the ASE beam was observed even at 60 cm away from the crystal. The diameter of spot was about 8.0 mm at 17 cm away from the output surface of the crystal, which was obtained when 930-nm pump radiation with the energy of 6 mJ was used. The divergence angle of the ASE beam was about 16 mrad. This angle is smaller than in the cases of the F_3^+ center (20 mrad [11]) and the F_2 center (30 mrad [10]), but it is not so different from those of the F_3^+ and F_2 centers. However, the angle is considerably larger than in the case of laser radiation whose angle is less than 1 mrad. In the present ASE experiment, no care was taken to focus the beam inside the crystal.

The ASE was obtained by the pumping radiation whose wavelength is between 860 and 1030 nm as shown in Fig. 1 (see the open circles). This 860–1030-nm region corresponds to the F_2^- absorption band region. It is noted that the F_2^- ASE intensity does not reflect the F_2^- absorption spectrum, i.e. the intensity does not depend on the absorption coefficient of the F_2^- absorption band as shown in Fig. 1. The broken curve of Fig. 2 shows the spectrum of the ASE which was obtained using 930-nm pump radiation. The ASE has a peak at

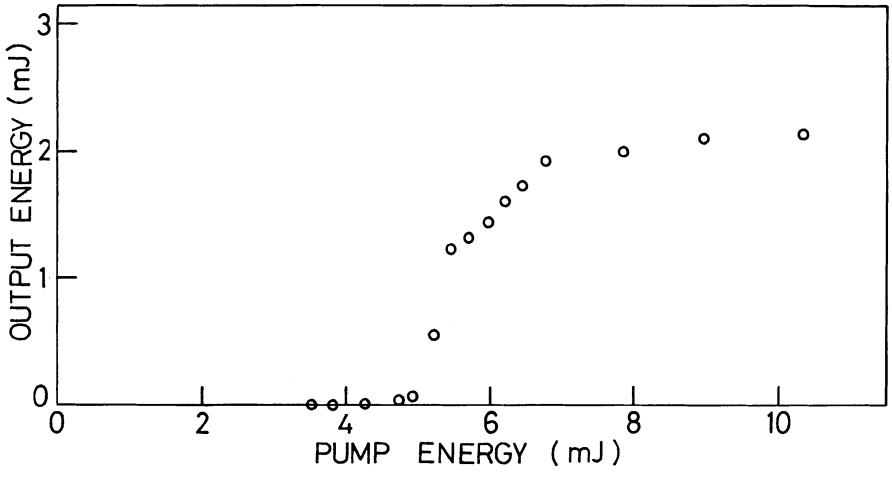

Fig. 3. The amplified spontaneous emission intensity of the F_2^- center plotted against the pulse energy of 936-nm pump beam

about 1130 nm and the half width is about 30 nm, which is much narrower than the half width of the luminescence band (180 nm); the ASE bandwidth is appreciably narrower than that of spontaneous emission. The ASE exhibits a Lorentzian-like line shape with a sharp peak and long tails at both short- and long-wavelength sides.

The favorable condition for strong ASE is a high gain combined with long path length in the active material [1]. The two end surfaces of the 54.80-mm-long LiF crystal are polished and almost parallel to each other. This indicates that not only the fluorescence but also the pump radiation can be reflected back into the active material from the two end surfaces, leading to the amplification of the fluorescence by the increase of path length. In fact, strong emission was observed throughout the whole of the 54.80-mm-long crystal although the effective penetration depth of the 930-nm pump beam is approximately 8 mm (because the absorption coefficient at the 930-nm pump wavelength is $1.23\,\mathrm{cm}^{-1}$). Therefore, taking into account that the F_2^- center is not a low-gain specimen since the fluorescence quantum efficiency at room temperature has been estimated to be 0.5 by Durarte et al. [14] and 0.22 by Gellermann et al. [15], it is understood that the ASE was easily obtained by the F_2^- center.

Figure 3 plots the ASE energy against the pulse energy of pump radiation. This figure shows that threshold pump energy for ASE is about 5 mJ and the saturation is observed at high pump energy. Unlike the case of usual laser oscillation, the ASE intensity was not observed to increase linearly with the pump energy when the energy is above the threshold. Neither bleaching effect nor output signal drop was observed after the continued operation of more than 2 h, indicating the F_2^- center has a high photo-stability and thermo-stability. This was also confirmed from the absorption spectra measured after the repeated ASE experiment.

In conclusion, the intense, directional and narrow-bandwidth 1130-nm emission was observed by the pumping with radiation in the wavelength region of the F_2^- absorption band and by the pumping with radiation of pulse energy above 5 mJ. This emission is attributed to the ASE by the F_2^- center in LiF crystal.

References

1. W. Koechner: *Solid-State Laser Engineering* (Springer, Berlin, Heidelberg 1996) pp. 182–186
2. B.A. Saleh, M.C. Teich: *Fundamentals of Photonics* (Wiley, New York 1991) pp. 488–489
3. U. Ganiel, A. Hardy, G. Neumann, D. Treves: IEEE J. Quantum. Electron. **QE-11**, 881 (1975)
4. O. Svelto: *Principles of Lasers* (Plenum Press, New York 1998) pp. 71–76
5. G. Baldacchini: *Spectroscopy and Dynamics of Collective Excitations in Solids*, ed. by B. DiBartolo (Plenum Press, New York 1977) pp. 495–517
6. W. Gellermann: J. Phys. Chem. Solids **52**, 249 (1991)
7. T.T. Basiev, S.B. Mirov, V.V. Osiko: IEEE J. Quantum. Electron. **QE-24**, 1052 (1988)
8. T.T. Basiev, S.B. Mirov: *Room Temperature Tunable Color Center Lasers* (Harwood, Chur, Switzerland 1994)
9. V.V. Ter-Mikirtychev, T. Tsuboi: Prog. Quantum. Electron. **20**, 219 (1996)
10. T. Kulinski, F. Kaczmarek: Opt. Commun. **28**, 101 (1979)
11. L. Zheng, S. Guo, L. Wan: Appl. Phys. Lett. **48**, 381 (1986)
12. G. Baldacchini: private communications
13. W. Gellermann, A. Muller, D. Wandt, S. Wilk, F. Luty: J. Appl. Phys. **61**, 1297 (1987)
14. M. Durarte, I.M. Ranieri, M.M.F. Veira: Opt. Mater. **3**, 269 (1994)
15. G. Baldacchini, M. Cremona, G. d'Auria, R.M. Montereali, V. Kalinov: Phys. Rev. B **54**, 17 508 (1996)
16. T. Tsuboi, V.V. Ter-Mikirtychev: Appl. Surf. Sci. **106**, 447 (1996)
17. W.B. Fowler: *Physics of Color Centers*, ed. by W.B. Fowler (Academic Press, New York 1968) Chapt. 2