STABLE LiF:F_2^+ COLOR CENTER LASER OSCILLATION AT ROOM TEMPERATURE*

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An LiF:F_2^+ color center laser using a two-mirror cavity is reported. The LiF:F_2^+ laser was achieved by creating the F_2^+ centers through the photoionization of the F_2 color centers at room temperature and simultaneously by exciting the F_2^+ centers optically. We obtained the F_2^+ broadband laser oscillation with a peak at 930 nm, and it was more powerful than the room-temperature broadband LiF:F_3^+ color center laser.

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1. Introduction

Color centers are one or more anion and cation vacancies which are trapping one or more electrons and holes in ionic crystals, respectively [1, 2]. They are produced when crystals are exposed to ionizing radiation such as X-rays, γ-rays and electron beams, or heated to a high temperature in the alkali or halide atom vapor (called additive coloration), or applied to an electric field at a high temperature (electrolytic coloration). Various color centers (for example, F, F_2 and F_3) have been observed in not only alkali halides such as KCl but also other ionic crystals such as CaF_2. The F center is formed when an electron is trapped in an anion vacancy, while the F_2 and F_3 centers are formed when two and three electrons are trapped in two and three anion vacancies adjacent to each other, respectively.

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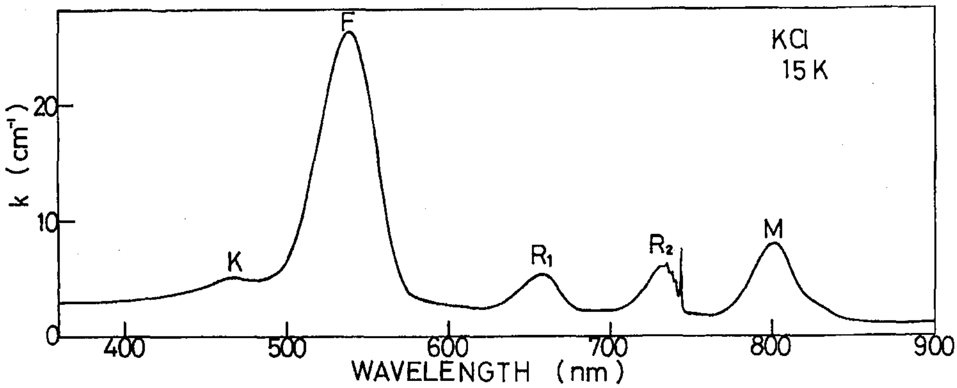

Fig. 1. Optical absorption spectrum, measured at 15 K, of additively colored KCl crystal. k — absorption coefficient.

Most color centers give rise to absorption in the near-infrared to ultraviolet spectral region. An example is shown in Fig. 1. In this figure, absorption bands named K, F, R_1, R_2 and M are observed in a visible spectral region, which were produced in an additively colored KCl crystal. The F and K bands are the lowest- and first higher-energy bands of the F center, respectively, while the R_1 and R_2 bands are due to the F_3 center and the M band is due to the F_2 center. The R_2 band is observed to have a zero-phonon line and vibronic structure at low temperature such as 15 K [2]. Figure 2 shows the case of LiF crystal where the F, F_2 and F_3 color centers are produced by the neutron-irradiation. The relative positions of these color centers are similar between LiF and KCl.

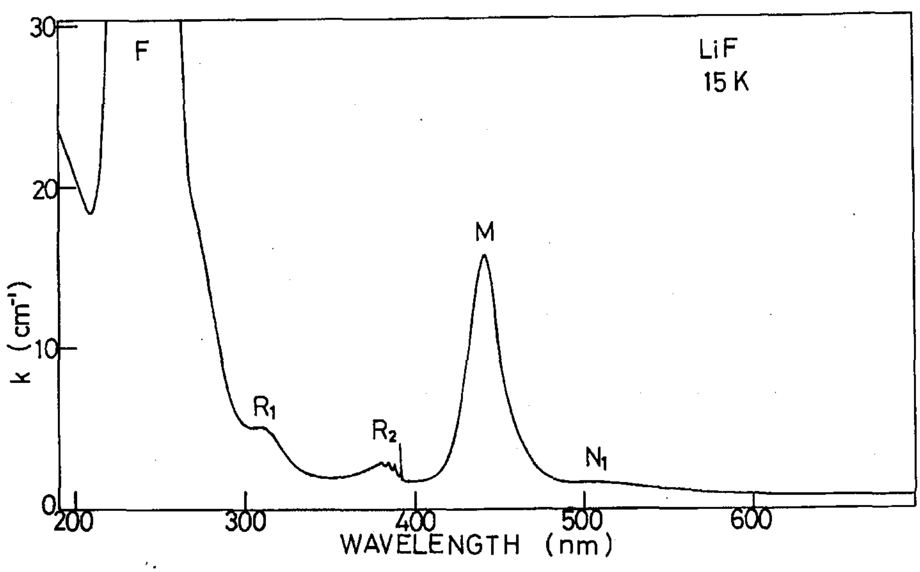

Fig. 2. Absorption spectrum of a neutron-irradiated LiF crystal at 15 K. k — absorption coefficient.

Color centers give rise to a broad luminescence band in the near-infrared and visible regions by the excitation in their absorption bands. As a result, some color centers are laser-active (known as color center lasers) [3, 4]. The F center has a very low gain coefficient in the stimulated emission and additionally it has a self-absorption of the luminescence. Therefore, the F center is not suitable as laser-active media [1]. When the F center is associated with a substitutional alkali ion in a nearest-neighbor cation position, F_A center is formed. There are two types of F_A centers, F_A(I) and F_A(II). The laser action has been observed in the F_A(II) center but not in the F_A(I) center [3].

Although the color centers have been known to be high-gain active materials in tunable solid state lasers, most color center lasers (including the F_A(II) laser) operate when the laser-active crystals are cooled with liquid nitrogen. The cooling is necessary to avoid thermal quenching of luminescence [3, 4]. For practical use, it is requested to be able to operate the laser at room temperature because it is more easy to adjust the optical alignment and because it is unnecessary to maintain the crystals at liquid nitrogen temperature not only during operation but also after stopping the operation to avoid the thermal bleaching of the color centers.

The room-temperature stable color center lasers have been achieved predominantly using LiF crystals [5, 6]. Recently we achieved the room-temperature LiF color center laser operation using F_3^+ color centers, together with the optical phase conjugation using the laser-active LiF:F_3^+ crystals [7, 8]. The LiF:F_3^+ color center laser has a tuning range in the green region of 510–570 nm. We obtained a free-running mode laser of a broadband with a peak at 543 nm. In the present paper we describe a room-temperature stable color center laser oscillation using the F_2^+ center in LiF crystals and we compare it with the case of LiF:F_3^+ color center laser. The F_2^+ color center is formed when an electron is trapped between two anion vacancies adjacent to each other along ⟨110⟩ direction.

2. Formation of F_2^+ centers

In LiF crystals the color centers of F, F_2 and F_3^+ are created by the ionizing radiation and they are stable at room temperature. However, the F_2^+ color centers are thermally unstable at room temperature [4–6]. The F_2^+ centers are created after warming the crystal, which was irradiated with ionizing radiation at temperature below liquid nitrogen temperature (during this irradiation both the anion vacancies and F centers are created), to a temperature near room temperature for a short time (in this process the vacancies become mobile to move to the F centers, resulting in the formation of the F_2^+ centers) [4].

One way to create high concentrations of the F_2^+ centers, which are sufficient to the laser oscillation at room temperature, is to ionize the F_2 centers. The ionization is made using light whose wavelength corresponds to the spectral region of the F_2 absorption band [4–6]. The second harmonic 532 nm radiation from the YAG:Nd^{3+} laser can be used for the photoionization of the F_2 centers. The F_2^+ absorption band has a peak at about 630 nm and a half-width of about 160 nm [9]. Its absorption band tail extends to 532 nm. This indicates that the 532 nm light can be used for the pumping of the F_2^+ centers. Therefore, the 532 nm light is also used to excite the created F_2^+ centers.

3. Laser cavity

For the practical use, it is requested to construct the laser that is simple and easy to adjust the optical alignment. An astigmatically compensated three-mirror cavity is widely used for the color center lasers [4]. The three-mirror cavity, however, is difficult to achieve the optical alignment. Therefore, we have constructed the two-mirror cavity. The two flat mirrors in the laser cavity were separated by 100–150 mm. The used input mirror is dichroic, with 90% transmission for pumping light of $\lambda = 532$ nm and 99.7% reflectivity for light of $\lambda = 900 \div 1000$ nm (corresponding to the wavelength of the LiF:F_2^+ color center laser radiation), as shown in Fig. 3. On the other hand, the output mirror has a transmission of 91% for light of $\lambda > 700$ nm and almost zero transmission for light $\lambda < 640$ nm, indicating that the pumping light of $\lambda = 532$ nm is cut by the output mirror.

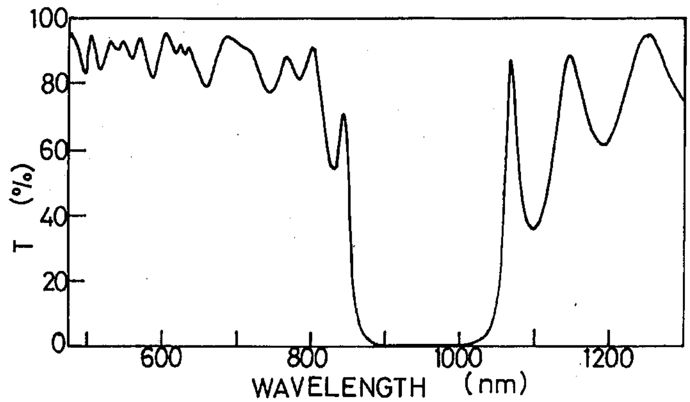

Fig. 3. Transmission spectrum of the input mirror used in the two-mirror cavity for LiF:F_2^+ color center laser operation.

The LiF crystal containing high concentrations of F_2 centers, with a thickness of 8 mm and surface of 40 x 20 mm^2, was Brewster-angle-cut. The presence of F_2 color centers in LiF crystal was checked by the absorption spectra of crystals. The LiF crystal was excited by the 532 nm radiation of a Surelite I-10 pulsed YAG:Nd^{3+} laser with a repetition rate of 10 Hz and a pulse duration of 6 ns. The output laser beam was viewed by eyes using an IR viewer. The laser spectrum was detected with an Advantest Q8381A spectral analyzer. The color center laser beam was introduced to an optical fiber attached with the spectral analyzer. The laser power was measured with a Gentec TPM-310 powermeter.

4. Laser oscillation

An intense broadband (i.e., free-running mode) laser oscillation with a peak at 930 nm and a half-width of 40 nm was observed at room temperature. Although we did not measure how long the stable laser oscillation continues if we never change the conditions of laser action, we obtained the laser oscillation without a significant decrease in power until we stop the pumping after about 2 hours. The 930 nm laser emission is confirmed to arise from the F_2^+ centers, because the F_2^+

luminescence has a peak at 910 nm and a half width of about 200 nm at room temperature [5, 9]. The laser output power increases with the pumping power as shown in Fig. 4. The laser threshold is 10 mW (i.e., about 1.1 mJ as the pulse energy of pump laser) and the slope efficiency is approximately 8%. When the output mirror of the laser cavity was replaced by a reflective diffraction grating, the wavelength tuning was obtained by rotating the grating.

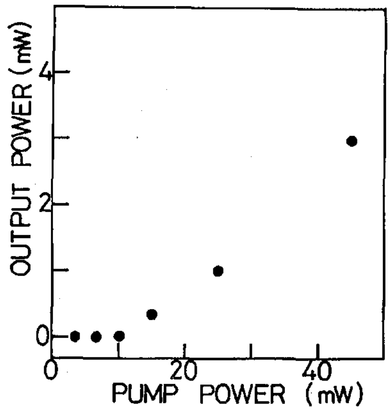

Fig. 4. Input/output power characteristic of the broadband LiF:F_2^+ color center laser. The 532 nm radiation of a pulsed YAG: Nd^{3+} laser was used as the pumping source.

The obtained slope efficiency of 8% is lower than the 13% efficiency which was obtained using the F_2 color centers in LiF crystal at room temperature [10]. However this efficiency is much higher than a slope efficiency of 0.2% which was obtained using the F_3^+ color centers in LiF crystal at room temperature. It is expected that the LiF:F_2^+ color center laser efficiency will increase if we use a dichroic mirror, which has a higher transmission than the present flat mirror, as the output mirror and a concave mirror as the input mirror. This means that the LiF:F_3^+ color center laser has a rather weaker efficiency and lower power than the LiF:F_2^+ laser. The threshold of the green LiF:F_3^+ laser was about 75 mW [7], while the near-infrared LiF:F_2^+ laser has a much low threshold. Therefore, it is confirmed that the LiF:F_2^+ color center laser is more efficient than the LiF:F_3^+ color center laser. Taking into account that the optically excited F_3^+ center has a possibility of transition from the radiative singlet-state into the non-radiative triplet-state [8], it seems that such a low efficiency and high threshold of the LiF:F_3^+ color center laser is reasonable.

5. Conclusion

In conclusion the LiF:F_2^+ color center laser was achieved by creating the F_2^+ centers through the photoionization of the F_2 color centers at room temperature and simultaneously by exciting the room-temperature unstable F_2^+ centers optically. It is confirmed that the laser is more powerful than the LiF:F_3^+ color center laser.

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