Appl. Phys. B 62, 437–442 (1996)

Applied Physics B Lasers and Optics

Diode-pumped colour-centre lasers tunable in the 1.5 μm range

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Received: 1 June 1995 / Accepted: 18 December 1995

Abstract. Under pumping with a 990 nm, 1 W laser diode, continuous-wave (cw) tunable laser emission in the 1.5 μm wavelength range was obtained from two different colour centres: $Tl^0(1)$ in $NaCl:Tl^+$ and $(F_2^+)_H$ in $NaCl:OH^-$. The results are compared to those recently obtained with a similar apparatus and $Tl^0(1)$ centres in $KCl:Tl^+$. The highest output power (30 mW) and the broadest tuning range (1.48–1.68 μm) were achieved with $(F_2^+)_H$ centres.

PACS: 42.55; 42.60

Colour-Centre Lasers (CCLs) are useful sources of tunable coherent radiation in the 0.8–4.0 μm range [1]. They are usually pumped by other lasers such as Ar^+, Kr^+, $YAG:Nd^{3+}$. These are, however, cumbersome pump sources with a high consumption of electric power. It is therefore of interest to replace them by Laser Diodes (LDs), which have both a very small volume and a good electrical-to-optical conversion efficiency. This mode of pumping is indeed very popular nowadays for rare-earth or transition-metal lasers [2]. On the other hand, it met, until now, with little success for pumping CCLs, probably because of the technological difficulty to cope simultaneously with the poor geometrical quality of LD beams and with the cryogenic requirements of CCLs. The first positive result in this field was an $NaF:Mg^{2+}$, $(F_2^+)^*$ centre laser which our laboratory successfully operated a few years ago [3, 4]. Pumped by a 500 mW, 825 nm LD, it emitted up to 35 mW and could be tuned in the 1.03–1.12 μm range. However, $(F_2^+)^*$ centres suffer from a slow but unrelenting fading under the influence of the pump light [5]. Very recently, Gubin and co-workers [6] reported single-frequency laser operation in the 2.9 μm range using $F_A(II)$ centres in $RbCl:Li^+$ pumped by a red laser diode. It is also attractive to apply LD pumping to CCLs which emit around 1.5 μm, because:

(*i*) This spectral domain is of great interest both for telecommunications with optical fibres and for applications which require an "eye-safe" emission (like atmospheric lidars). Widely tunable cw sources, such as CCLs, are valuable for diagnostics both of passive devices (optical fibers [7]) and laser-active samples (for instance erbium-doped materials [8]) operating in this wavelength region.

(*ii*) Some of the best CCLs precisely operate around 1.5 μm. They use either $Tl^0(1)$ centres in $KCl:Tl^+$ [9] or $(F_2^+)_H$ centres in $NaCl;OH^-$ [10]; they are usually pumped at 1.064 μm by a $YAG:Nd^{3+}$ laser and, when properly operated, their active materials have an effective lifetime of several months.

The commercial availability of powerful InGaAs LDs in the 960 nm domain opened the possibility to pump $Tl^0(1)$ and $(F_2^+)_H$ lasers by diodes. Last year, we announced our first achievement in this domain [11]: a CCL using $Tl^0(1)$ centres in $KCl:Tl^+$ pumped by a 962 nm SDL diode. In the present paper, we report the results obtained more recently with a 990 nm LD pumping $Tl^0(1)$ or $(F_2^+)_H$ centres in NaCl. Section 1 describes the apparatus and experimental techniques, Sect. 2 deals with the performance of our LD-pumped CCLs and, finally, Sect. 3 contains a brief discussion.

1 Apparatus and experimental techniques

Our active crystals are home made. $NaCl:OH^-$ is obtained by the Czochralski technique from a melt containing 500 ppm (molar) NaOH. $KCl:Tl^+$ and $NaCl:Tl^+$ are grown by the Bridgman technique in a sealed silica tube. In the case of $NaCl:Tl^+$, the starting TlCl doping is 5960 ppm (molar), but there is a Tl^+ concentration gradient in the crystal due to segregation. In order to avoid "parasitic" absorption bands which are noxious for laser operation [1], we select slices with a thallium doping corresponding to a room-temperature absorption coefficient of 4–9 cm^{-1} at 280 nm (on the long-wavelength wing of the $^1S_0$–$^3P_1$ band of Tl^+).

Coloration of samples is achieved by the standard methods fully described in [1]. Figure 1 shows the longest wavelength (i.e., pump) absorption bands for the three materials, along with the corresponding emission bands.

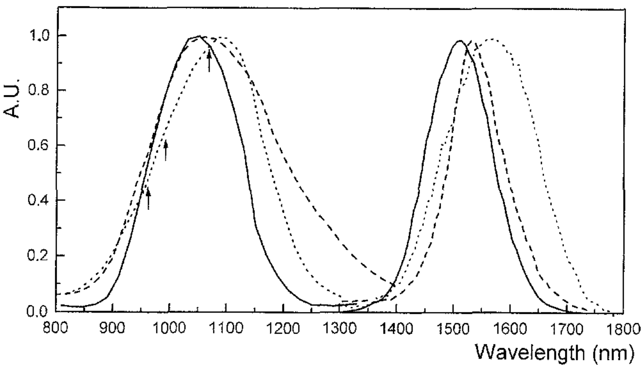

Fig. 1. Transition between the two lowest states in emission and absorption for $Tl^0(1)$ centres in $KCl:Tl^+$ *(full line)*, $Tl^0(1)$ centres in $NaCl:Tl^+$ *(dashed line)* and $(F_2^+)_H$ centres in $NaCl:OH^-$ *(dotted line)*. All curves are normalized to maximum intensity. *Arrows* show the locations of 962, 990 and 1064 nm pump wavelengths

Figure 2 schematically represents the CCL. The optical cavity is linear, astigmatically compensated, thanks to an intracavity, antireflection coated lens with 30 mm focal length, titled by 9.9° in order to correct for the astigmatism induced by the 2 mm thick sample at Brewster angle. The input mirror is a neutral meniscus, with 18 and 15 mm external and interval curvature radii, respectively. It transmits approximately 87% at 990 nm; it is coated for maximum reflection from 1.38 to 1.68 μm. Flat output mirrors of 6.5 and 13% transmission at 1.50 μm have been used[1]. The sample, with one of its [1 0 0]-axes horizontal, is located at the waist, clamped at the end of a copper cold finger cooled by a nitrogen cryostat. A mechanical device, not shown in the figure, allows the sample to be accurately positioned from outside, both horizontally, along the laser beam, and vertically. The cryostat vacuum chamber is closed by the input mirror and the intracavity lens; the position of both these elements can be finely adjusted by micrometric screws.

The LD is a SDL 6362 P1 from Spectra Diode Laboratories, a 10 element device delivering up to 1 W cw. In our previous experiments with $KCl:Tl^+$ [11], we used a diode emitting at 962 nm; later, a 990 nm diode became available and we used it for the present work. Indeed, Fig. 1 shows that 990 nm is a more favorable pump wavelength than 962 nm for the three colour centres we have studied. The LD beam is polarized in the plane of the junction, i.e., horizontally. According to the manufacturer, its divergence is 16° in the plane of the junction and 36° in the perpendicular plane. We use standard Melles-Griot coupling optics: a 6.5 mm focal length collimation lens, a 4 × magnification pair of anamorphic prisms and a 30 mm focal length focusing lens. The resulting minimum cross section of the pump beam is $90 \times 10\ \mu m^2$, to be compared with a waist "radius" for the CCL cavity of 21 μm (vertically) and 31 μm (horizontally), within the sample. The overlap between pump and laser beam is

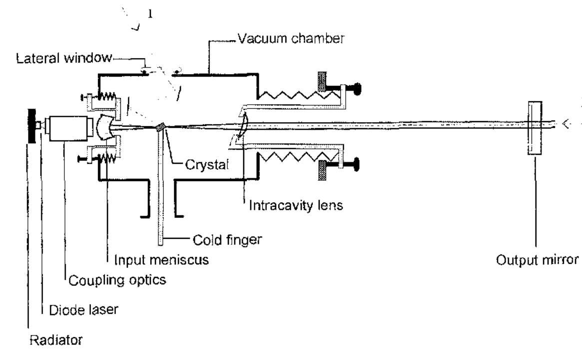

Fig. 2. Schematic of the CCL. The *dashed lines* labelled "1" and "2" indicate two possible paths for the regeneration auxiliary beam (Sect. 2.2)

therefore far from perfect, which is probably the main limitation of our CCL performance. Moreover, because of the diaphragm introduced by the entrance and exit holes in the anamorphic prisms case, maximum pump power after the focusing lens is measured to be 690 mW only for 1 W output of the LD. This power is further reduced to 600 mW inside the optical cavity due to the transmission coefficient of the input mirror. In the whole paper (including figures), "input pump powers" will be those measured at the exit of the focusing lens.

In the case of $(F_2^+)_H$ centres (Sect. 2.2.), a visible auxiliary irradiation is necessary [1, 10]: its role is to prevent the anisotropic centres to orientate themselves under the influence of the intense infrared pump beam into the directions which have the smallest absorption for this beam. For this regeneration, we use either the light from an Osram HBO 100 W mercury lamp or the 476, 482.5 and 488 nm lines of a Kr^+ laser[2]. This laser can be operated either with an intracavity prism in order to select one of the blue–green lines or without prism, in which case it emits them simultaneously. In the latter case, we shall speak below of "multiline Kr^+ laser". The auxiliary beam can be introduced into the vacuum enclosure through a lateral window and sent onto the sample by two small plane mirrors inside this enclosure (dashed line 1 in Fig. 2). With the Kr^+ laser, the regeneration beam can also be introduced through the output mirror[3] and the intracavity lens (dashed line 2). Below, the quoted auxiliary beam power will be the one measured at the location of numbers "1" or "2" of Fig. 2.

2 Performance of the LD-pumped CCL

With the above-described apparatus, we obtain LD pumped, cw laser emission from the two tested colour centres. In both cases, the emission at liquid nitrogen temperature remains stable for days (no detectable "fading"), which was expected a priori from the known laser behaviour of

[1] At 1.52 and 1.56 μm, which are, respectively, the free running wavelength of the $Tl^{(0)}$ (1) and $(F_2^+)_H$ lasers, the transmissions of these mirrors are 5.5, 11.0 and 5.0, 11.5%. However, for simplicity, we shall call them below by their 1.50 μm transmissions

[2] This "krypton" laser also emits some argon lines, such as 488 nm

[3] For 476 and 488 nm, the transmission of our two output mirrors is, respectively, 78 and 58% "6.5% mirror", 82 and 73% "13% mirror"

Table 1. Characteristics of LD-pumped CCLs. All data at 77 K, with the exception of the last line

Crystal and active centre	$NaCl:Tl^+$ $Tl^0(1)$		$NaCl:OH^-$ $(F_2^+)_H$	
Colouration mode	1.6 MeV electrons		Additive colouration	
Peak absorption of active band[a]	5.9		6.6	
Pump laser	Laser diode 993 nm	Nd:YAG laser 1064 nm	Laser diode 993 nm	Nd:YAG laser 1064 nm
Threshold [mW]	255[b]	180[c]	125[b,d]	48[c,g]
Slope efficiency[c] [%]	3.5	7.3	7.0[e]	14.55[g]
Peak output [mW] For 690 mW pumping	11	33	30[e]	74[g]
For 1200 mW pumping		70.5		152[g]
Free-running wavelength [µm]	1.520	1.520	1.560	1.560
Tuning range [µm]	1.493–1.540[b]	1.464–1.590[c]	1.480–1.680[b,f]	1.479–1.705[c,g]
Maximum operating temperature [K]			140[b,d]	185[c,g]

[a] absorption coefficient (cm^{-1})
[b] 6.5% transmission output mirror
[c] 13% transmission output mirror
[d] regeneration beam: 41 mW multiline Kr^+ laser, injected along path 2
[e] regeneration beam: 37 mW multiline Kr^+ laser, injected along path 2
[f] regeneration beam: 33 mW multiline Kr^+ laser, injected through the intracavity prism
[g] regeneration beam: 23 mW of the 476 nm krypton line injected through the input mirror

$Tl^0(1)$ and $(F_2^+)_H$ centres under the more conventional $YAG:Nd^{3+}$ pumping [9, 10]. Table 1 summarizes the main characteristics of the two LD-pumped CCLs. Let us now turn to the distinctive features of each of them.

2.1 *$Tl^0(1)$ centres in $NaCl:Tl^+$*

The spectroscopic properties of these centres are described in [9, 12]. Their laser emission under pumping by a $YAG:Nd^{3+}$ laser has been obtained by Gellermann [13], but does not seem to have been reported in the literature. As preliminary to LD pumping, we reproduced Gellermann's experiment with the apparatus of Fig. 2, expect that the LD and coupling optics were replaced by a cw Spectron SL 903–2000 $YAG:Nd^{3+}$ laser. We observed that the laser behaviour of $Tl^0(1)$ centres is indeed very similar in NaCl and in KCl. With NaCl, we obtain without careful optimization a threshold of 180 mW and a slope efficiency of 7.3% (for 13% transmission output mirror). The free-running wavelength is 1.52 µm, very close to the one obtained with homologous centres in KCl, which is not surprising in view of the fluorescence spectra (Fig. 1). Under 1.5 W $YAG:Nd^{3+}$ laser pumping, we tuned the laser emission from 1.464 to 1.590 µm.

Subsequently, a $NaCl:Tl^+$ sample has been used as the active element of our LD-pumped CCL. We obtained thresholds of 225 and 300 mW and slope efficiencies of 2.2 and 3.5% for output couplers of 6.5 and 13% transmission, respectively (Fig. 3a). These values are significantly worse than those observed with $YAG:Nd^{3+}$ pumping. The difference arises from two causes, the second being by far prevailing:

1) At 77 K, the absorption band peaks at 1.065 µm (Fig. 1). Therefore, the pump wavelength 990 nm is 75 nm too low, which corresponds to an absorption-coefficient decrease by a factor of ≈ 0.8 (4.7 cm^{-1} at 77 K, instead of 5.9 cm^{-1} at band maximum). Thus, for a 2 mm thick sample at Brewster angle, 67.5% of the incident power is absorbed, instead of 75.5% (for low incident intensities).

2) As remarked in Sect. 1, the overlap between pump and laser beams is not good even in the waist plane (front face of the sample). Moreover, the laser diode emits two lobes which can be made coincident only on a depth of ca. 0.5 mm, so that the rear part of the sample is quite poorly pumped.

The CCL output beam is TEM_{00} at moderate pump powers, it becomes TEM_{10} at high pump powers. This probably arises from the poor geometrical quality of the LD emission. With an intracavity prism and a 5.5% output mirror, the CCL is tunable from 1.493 to 1.540 µm (Fig. 3b), i.e., nearly in the same range as the LD-pumped $KCl:Tl^+$ CCL we described in [11]. Other characteristics (threshold, slope efficiency) are also very close with both crystals. Let us finally mention that in $NaCl:Tl^+$, like in $KCl:Tl^+$, it is difficult to obtain a

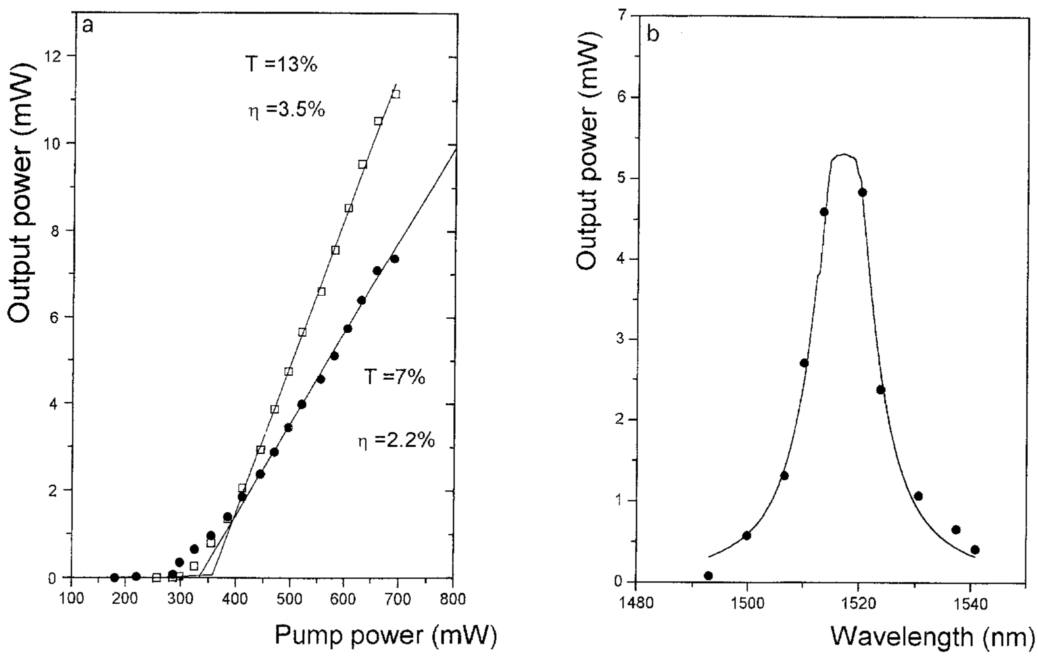

Fig. 3a,b. Performance of the NaCl: Tl^{+}, $Tl^{0}(1)$ laser: (**a**) output power vs input pump power; (**b**) tuning curve for 690 mW input pump power and 6.5% transmission output mirror

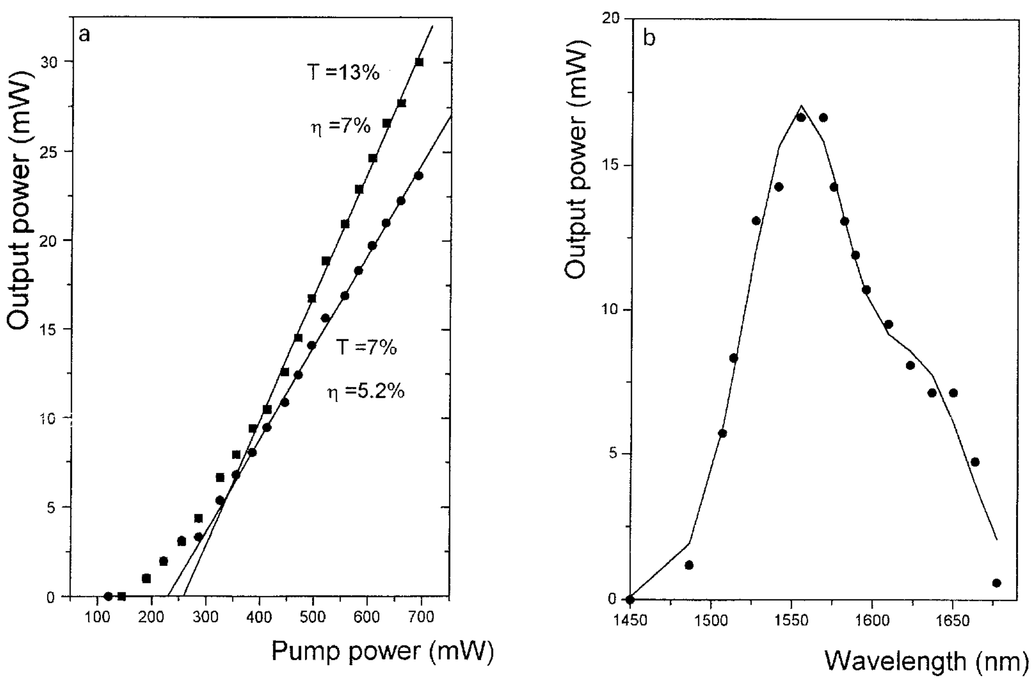

Fig. 4a,b. Performance of the NaCl: OH^{-} $(F_2^{+})_H$ laser: (**a**) output power vs input pump power for ≈40 mW multiline regenerating power; (**b**) tuning curve for 690 mW input pump power, 33 mW multiline regenerating power and 6.5% transmission output mirror

large concentration of $Tl^{0}(1)$ centres without producing simultaneously several other defects which have overlapping absorptions [1] and which are therefore noxious for laser operation. A ticklish compromise must therefore be found between the intensities of the $Tl^{0}(1)$ band and of its "parasites".

2.2 $(F_2^{+})_H$ centres in NaCl: OH^{-}

Using for the regeneration a ≈40 mW multiline Kr^{+}-laser beam injected through path 2 in Fig. 2, we obtained the data of Fig. 4a and of the last but one column of Table 1: thresholds of 125 and 150 mW, slope efficiencies of 5.2

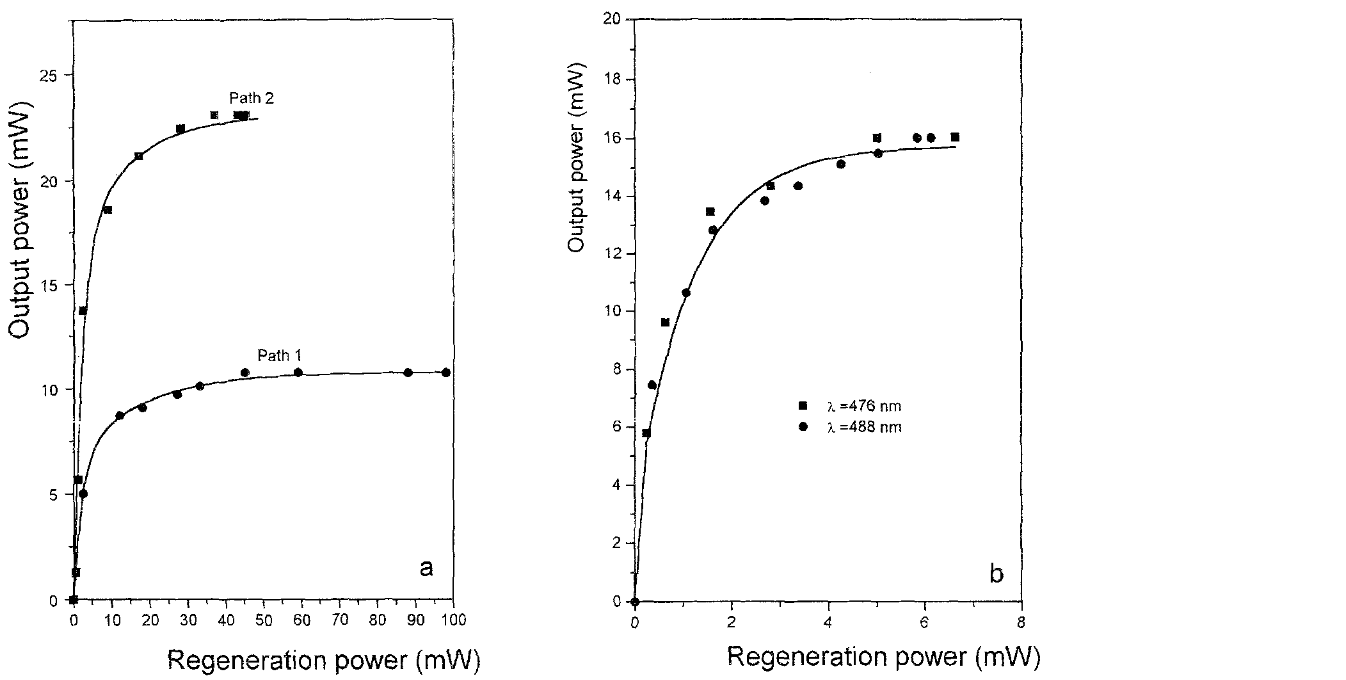

Fig. 5a,b. Output power of the NaCl:OH$^-$ $(F_2^+)_H$ laser vs injected auxiliary beam power for different methods of regeneration: (**a**) Influence of the geometry of the regeneration beam (paths 1 and 2 of Fig. 2). The experiment is performed with the multiline Kr^+ laser. (**b**) Influence of the regenerating beam wavelength when path 2 of Fig. 2 is used

and 7.0%, respectively, for 6.5 and 13% transmissions of the output mirror. In order to help the discussion in the next section, we also include in Table 1 the characteristics obtained when the **same** NaCl:OH$^-$ laser is pumped in the conventional manner with a YAG:Nd^{3+} laser[4]. With an intracavity prism (and injecting a 33 mW Kr^+ beam through this prism), we can continuously tune the diode-pumped CCL from 1.48 to 1.68 μm (Fig. 4b), which is close to the range obtained with the YAG:Nd^{3+}-pumped device. Thus, the characteristics of the $(F_2^+)_H$ laser are markedly better than those of the $Tl^0(1)$ lasers ([11] and Sect. 2.1): lower threshold, higher output and much broader tuning range. These favourable results are obtained in spite of an even poorer adequacy of the pump wavelength (990 nm) to the material absorption spectrum (peak absorption at 1.080 μm, i.e., 90 nm higher). The absorbance is thus reduced from 6.6 to 4.3 cm^{-1} (Fig. 1); in the case of a 2 mm thick sample at Brewster angle, the percentage of absorbed pump power falls from 79 to 64% (for low incident intensities). The better results obtained with $(F_2^+)_H$ centres are perhaps due to the larger facility to obtain suitable optical densities without overlapping of other, detrimental absorption bands.

The losses of the CCL cavity have been evaluated by a variant of the Findlay-Clay method [15] in which we used only one output mirror but introduced an adjustable supplementary intracavity loss of known magnitude. We thus found a logarithmic loss of 0.140 for a round trip in the cavity. Since 0.051 is due to the output mirror, useless losses of the cavity amount to 0.089, out of which the contribution of the intracavity lens has been estimated from an auxiliary experiment to be 0.040 $\pm$ 0.005 and the remainder is probably due to diffusion by the faces of the active material. As in the case of NaCl:Tl^+, and probably for the same reason, the $(F_2^+)_H$ CCL output beam is TEM_{00} or TEM_{10} depending on the pump power. We also studied the behaviour of the untuned $(F_2^+)_H$ CCL when the sample temperature was risen above 77 K with a small electric resistor coiled on the cryostat cold finger. We observed the threshold to increase and the output to decrease until the CCL completely stopped emitting at 140 K, when the threshold reached the available pump power, i.e., 690 mW.

The main problem with $(F_2^+)_H$ centres is the need for an auxiliary reorientating beam [1, 10] already mentioned at the end of Sect. 1. In the case where this beam is provided by the Kr^+ laser, the best results are obtained, as theoretically predicted, with polarizations of the regeneration and pump beams parallel to one another. Figure 5a shows the CCL output vs regeneration power for both auxiliary-beam geometries in Fig. 2 (in the case of multiline Kr^+ laser). One observes, as expected from the literature, that regeneration powers of a few tens of milliwatts are sufficient to saturate the emission of the CCL. Injection of the regeneration beam through the lateral window (path 1 in Fig. 2) is approximately half efficient as injection through the output mirror (path 2), a fact which we ascribe to a worse overlapping between pump and regeneration beams in the first case. The smaller asymptotic value of the CCL output power for regeneration scheme 1 probably arises from the fact that, even if the pump and auxiliary

[4] The current laser cavity is designed for LD pumping. Therefore, the characteristics in the last column of Table 1 are far below those which can be obtained from the same centres with YAG:Nd^{3+} pumping in an optimized CCL[14]

beams do coincide at the front face of the sample, they do not overlap at all in its rear portion, because they are at a noticeable angle θ to one another, with a corresponding reduction of the laser output.

Let us now turn to the wavelength dependence of the regeneration-beam efficiency. Figure 5b shows data obtained with geometry 2, using two different lines of the Kr^+ laser. Within experimental uncertainties, 476 and 488 nm are found to be equally effective. On the other hand, the HBO mercury lamp is significantly less active than the Kr^+ laser. In a comparative experiment, we obtain 5.2% slope efficiency for Kr^+ regeneration along path 2 of Fig. 2, 3.6% for the Kr^+ beam along path 1 and 1.9% only when the regeneration is with the unfiltered Hg lamp (also injected through path 1). Part of the difference may arise from the geometrical difficulty to focus efficiently the 4π steradians emitting lamp on our sample inside the vacuum enclosure of the CCL. Clearly, the problem of the regeneration in LD-pumped $(F_2^+)_H$ CCLs requires further study. As mentioned at the beginning of this section, the data of Fig. 4 and of the fourth column of Table 1 have been obtained using the currently most favourable regeneration scheme.

3 Discussion

Compared to $YAG:Nd^{3+}$ laser pumping, the results obtained when pumping with the LD are between two and three times poorer. The main reason is the bad overlapping between the pumping light and the transverse mode TEM_{00} of the laser cavity, especially because the mode pattern emitted by the LD is formed of two separated lobes which cannot be made coincident in the whole depth of the sample.

LD-pumped CCLs are potentially very useful tools for producing tunable coherent emission in the vicinity of 1.5 µm. According to Sect. 2, $(F_2{}^+)_H$ centres seem much more promising for this purpose than $Tl^0(1)$ centres. However, in order to take full advantage of LD pumping, the apparatus should be as compact as possible and have the smallest possible consumption of electrical energy. One should therefore get rid of the liquid nitrogen cryostat and the auxiliary Kr^+ laser.

Concerning the latter point, we may hope to obtain an efficient reorientating light from a green LED inserted inside the CCL vacuum chamber in the immediate vicinity of the input mirror. One would thus obtain an all-solid-state CCL with a satisfactory overall electrical-to-optical efficiency. The sample-cooling problem, however, seems much more serious. When we started this investigation, we hoped that CCL operation would be possible at high enough temperatures to allow replacement of the liquid nitrogen cryostat by a Peltier cooler[5]. The results of Sect. 2 (last line of Table 1) show that such is unfortunately not the case. The three investigated centres do not allow laser emission under LD pumping above 140 K[6] (with $YAG:Nd^{3+}$ pumping, laser operation is possible up to 185 K in $NaCl:OH^-$ and $KCl:Tl^+$, probably because of higher available pump power and better geometry of the pump beam).

In summary, we have operated the first LD-pumped CCLs which are continuously tunable in the 1.5–1.7 µm range. But we feel that this achievement is not fully satisfactory, since we could not get rid of the cumbersome and inconvenient liquid nitrogen cryostat.

Acknowledgements. We would like to thank Mr Bernard Rocher of our Laboratory for crystal growth and Mr Vidal of the "Groupe de Physique des Solides" at Paris VII University for his helpful suggestions during colouration of samples by electron irradiation.

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[5] Multistage Peltier coolers are commercially available for temperatures down to 153 K

[6] For $NaCl:Tl^+$, the maximum operating temperature was not measured, but it is probably comparable to the one of $KCl:Tl^+$ (120 K with LD pumping) in view of the great similarities of other laser properties of both crystals (Sect. 2.1)