Erbium 3- μ m Fiber Lasers

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Invited Paper

Abstract—With its recent breakthrough in terms of output power, the erbium 3- μ m fiber laser has become an object of intense scientific research and an increasingly attractive tool for medical applications. This paper reviews the research on the erbium 3- μ m fiber laser since its first demonstration. Its development is seen in relationship to the early success of the corresponding crystal laser system, to the foundations that were laid by the investigation of its spectroscopy and population mechanisms, and the recent technological developments in related fields.

Index Terms—Cascade laser, energy recycling, energy-transfer upconversion, erbium, excited-state absorption, fluoride fiber laser, laser efficiency, laser in surgery, mid-infrared laser, upconversion laser.

I. HISTORY OF ERBIUM 3- μ m LASERS

IN RECENT YEARS, there has been an increased interest in lasers emitting at 3 μ m, mainly because of their potential and partly already demonstrated applications in laser surgery [1]–[5]. Due to the high absorption of 3- μ m radiation in water, high-quality cutting or ablation has been demonstrated in biological tissue using erbium-doped solid-state lasers. The erbium 3- μ m laser can be used as a noncontact scalpel or drill, thus being aseptic and avoiding or reducing pressure at the incision site. The 3- μ m fiber laser can be of specific use in a number of applications, e.g., in endoscopy.

In this review paper, we will concentrate on the fundamental physics of the erbium 3- μ m fiber laser, i.e., its spectroscopy and operational regimes. A retrospective of the development of erbium 3- μ m lasers shows that its history was dominated by crystalline systems. Only recently, fiber lasers at 3 μ m have caught up with crystal lasers in terms of output power. This has several reasons. On the one hand, new technologies such as high-power diode lasers and double-clad fibers have become available only in the past decade. Especially the difficult development and high costs of fibers with sufficiently low losses in the mid-infrared spectral region have slowed down the necessary research efforts in this field. The fabrication of low-loss fluoride fibers with

rare-earth dopant concentrations comparable to those used in erbium 3- μ m crystal lasers still represents a major problem for the fiber suppliers. On the other hand, the early success of the erbium 3- μ m crystal laser has given rise to a significant amount of spectroscopic investigations mainly in Russia and later on also in Western Europe and the United States. This has led to a deep understanding of the population mechanisms of this laser system and to the development of a large number of suitable host materials. Compared to these tremendous research efforts, the spectroscopic accounts of the erbium 3- μ m fiber laser remained few, and it is characteristic that the first breakthrough [6] of the fiber laser in 1995 was based entirely on a better understanding [7], [8] of the spectroscopy and population mechanisms of this system.

In order to understand the failure of the first experimental approaches toward a high-power fiber laser and the subsequent underestimation and neglect of this system, it is important to understand the significant differences in the population mechanisms of the first fiber lasers as compared to the crystal lasers. The positive results obtained with the crystal laser system also anticipate the future direction of research in the fiber laser system.

A. Energy Recycling by Energy-Transfer Upconversion

The first observation [9] of coherent emission at 3 μ m from erbium ions was reported in 1967. Yttrium Aluminum garnet (YAG), today's most widely used solid-state laser material, entered the stage as a host for the erbium 3- μ m laser [10], [106] in 1975. Notably, it was this material with its high phonon energies and strong multiphonon quenching of the $^4I_{11/2}$ upper laser level (leading to an upper-to-lower level lifetime ratio of $\sim 1 : 50!$), in which the first continuous-wave (CW) lasing at 3 μ m was observed [11], [107] in 1983.

At about the same time, it was established [12]–[14], [108]–[110] that energy-transfer processes [15] between neighboring erbium ions in the host lattice govern the population mechanisms of this highly erbium-doped laser system. In Fig. 1, the energy-level scheme of erbium, a suitable transition for the ground-state absorption (GSA) of pump power, the laser transition at 3 μ m, and the important energy-transfer-upconversion (ETU) and cross-relaxation (CR) processes are introduced. The ETU process ($^4I_{13/2}, ^4I_{13/2} \rightarrow ^4I_{15/2}, ^4I_{9/2}$) leads to a fast depletion of the lower laser level and enables CW operation of a laser transition that, otherwise, could be self-terminating owing to the unfavorable lifetime ratio of the upper compared to the lower laser level (Fig. 1).

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Fig. 1. Partial energy-level scheme of erbium indicating the processes relevant to the energy-recycling regime of the 3- μm laser at high dopant concentration: Direct pumping of the $^4\text{I}_{11/2}$ upper laser level by GSA, 3- μm laser transition, energy-transfer processes such as ETU from the $^4\text{I}_{11/2}$ upper and $^4\text{I}_{13/2}$ lower laser levels and CR from the thermally coupled $^4\text{S}_{3/2}$ and $^2\text{H}_{11/2}$ levels, as well as subsequent multiphonon relaxations. Lifetimes are given for ZBLAN:Er $^{3+}$.

In particular, this ETU process offers another generous advantage. Half of the ions that undergo this process are upconverted to the $^4\text{I}_{9/2}$ level and, by subsequent multiphonon relaxation, are recycled to the $^4\text{I}_{11/2}$ upper laser level from where they can each emit a second laser photon, for a single pump-photon absorption. For a large number of ions participating in this process, a slope efficiency η_{sl} of twice the Stokes efficiency $\eta_{St} = \lambda_p/\lambda_l$ obtains [16], because the quantum efficiency $\eta_q = n_l/n_p$ of pump photons converted to laser photons increases from 1 to 2 (λ and n are the wavelengths and photon numbers of laser and pump transitions, respectively):

$$\eta_{sl} = \eta_q \times \eta_{St} = 2 \times \eta_{St}. \quad (1)$$

This is illustrated in Fig. 2.

The ETU process from $^4\text{I}_{13/2}$ can be so dominant that even under direct pumping of the $^4\text{I}_{13/2}$ lower laser level and subsequent excitation of the $^4\text{I}_{11/2}$ upper laser level by ETU, 3- μm laser operation was demonstrated in several host materials [17].

B. Optimum Slope Efficiency of Erbium 3- μm Lasers

Returning to Fig. 1, one can solve a simple rate-equation system [16], which includes the processes shown in this figure. This approach is based on the assumption that the erbium concentration and the absorbed pump intensity are high enough to establish a fast depletion of the two laser levels by the ETU processes. In this case, the intrinsic decay mechanisms characterized by the lifetime of each level are negligible. (A generalized solution including intrinsic decay can be found in [18]. At pump-power levels a few times above laser threshold, the generalized solution converges to the simplified solution presented here.) We further assume that the CR process depletes

Fig. 2. Partial energy-level scheme of erbium illustrating the process of energy recycling from the lower to the upper laser level by ETU. Indicated are the relative pump rate η_p of the upper laser level and the quantum efficiency η_q , which increases from 1 to 2 if a large number of ions participate in the process.

the $^4\text{S}_{3/2}$ level effectively and leads to a fast repopulation of the laser levels. The slope efficiency, in this case, is given by [16]

$$\eta_{sl} = \eta_{St} \eta_m \frac{\ln(1-T)}{\ln[(1-T)(1-L)]} \left[\frac{2 - \frac{b_1^2}{b_2^2} W_{22}}{W_{11}} \right] \quad (2)$$

with η_m the geometrical overlap of pump and laser modes, T the transmission of the outcoupling mirror, L the internal resonator losses, and b_i and W_{ii} the Boltzmann factors and ETU parameters of the upper and lower laser levels, respectively. If ETU occurs only from the lower laser level, i.e., $W_{22} = 0$, we obtain the same factor-of-two increase in slope efficiency from (2) as in the simple model of Fig. 2. The slope efficiency is reduced, however, by the resonator losses, the nonperfect mode overlap, and the ETU process from the upper laser level in the case of $W_{22} > 0$ (Fig. 1). One significant advantage of the fiber laser over the crystal laser is the fact that the mode overlap is $\eta_m \sim 1$.

The parameters W_{ii} of both ETU processes increase with increasing erbium concentration because of the influence of energy migration within the erbium $^4\text{I}_{11/2}$ and $^4\text{I}_{13/2}$ levels on ETU. For example, the measured concentration dependence of the ETU parameters [19] in ZBLAN bulk glasses for the currently available dopant concentrations is shown in Fig. 3. The slope efficiency of (2) is optimum for a maximum ratio W_{11}/W_{22} . Spectroscopy of these processes in crystal hosts revealed that the maximum ratio is obtained at dopant concentrations of $\sim 12\text{--}15\%$ in BaY_2F_8 ([20], [21]), $\sim 15\%$ in LiYF_4 ([22]), $\sim 30\%$ in YSGG ([23]), and $\sim 50\%$ in $\text{Y}_3\text{Al}_5\text{O}_{12}$ ([24]). A trend in this series is the increase of the optimum erbium concentration with phonon energy of the host material.

The pump wavelength that provides the highest Stokes efficiency is 980 nm, which corresponds to pumping directly into the upper laser level [25] (Fig. 1). For this pump wavelength, a Stokes efficiency of $\eta_{St} = \lambda_p/\lambda_l = 35\%$ is derived. The highest slope efficiency obtained experimentally [26] is currently $\eta_{sl} =$

Fig. 3. Macroscopic parameters (solid symbols) of ETU from $\text{Er}^{3+} {}^4\text{I}_{13/2}$ and ${}^4\text{I}_{11/2}$ as well as CR from $\text{Er}^{3+} {}^4\text{S}_{3/2/2} {}^2\text{H}_{11/2}$ versus Er^{3+} concentration in ZBLAN bulk glasses. Also shown are the macroscopic parameters (open symbols) of ET from $\text{Er}^{3+} {}^4\text{I}_{13/2}$ and ${}^4\text{I}_{11/2}$ to the Pr^{3+} codopant for a Pr^{3+} concentration of $8 \times 10^{19} \text{ cm}^{-3}$. (Data taken from [19].)

50% in $\text{LiYF}_4 : 15\% \text{ Er}^{3+}$. This result shows that energy recycling is indeed efficient and that slope efficiencies far above the Stokes efficiency can be obtained under CW pumping. Under quasi-CW excitation, the slope efficiency is strongly reduced [27], because the lower laser level is much less populated than in the steady-state regime and the ETU processes whose rates depend on the square of the population density are less efficient [28], [29].

Once it has been established that ETU from the lower laser level leads to energy recycling and an enhancement of the slope efficiency by a factor of two, it becomes clear that other operational regimes that deplete the lower laser level without recycling the energy to the upper laser level are less efficient. Consequently, neither colasing at the $1.6\text{-}\mu\text{m}$ transition from the lower laser level to the ground state [30] nor energy transfer from the erbium lower laser level to a rare-earth codopant [31] have reached the efficiency of the recycling regime. This statement extends to the fiber laser system without restriction.

A fact that concerns the pump threshold rather than the slope efficiency shall be mentioned here briefly. The lifetime of the ${}^4\text{I}_{11/2}$ upper laser level is quenched by multiphonon relaxation. According to the energy-gap law [32], this influence is stronger in oxide compared to fluoride host materials because of the larger maximum phonon energies in oxides. With an energy gap between the ${}^4\text{I}_{11/2}$ and the next lower lying ${}^4\text{I}_{13/2}$ levels of $\sim 3400\text{--}3500 \text{ cm}^{-1}$, the radiative decay becomes dominant for phonon energies below $\sim 550 \text{ cm}^{-1}$. A long lifetime of the ${}^4\text{I}_{11/2}$ upper laser level provides a small pump threshold. Fluorides are, therefore, preferable host materials [33] for this laser transition if the pump power is not many times above threshold.

C. Limitation of Erbium $3\text{-}\mu\text{m}$ Crystal Lasers

In recent years, researchers have obtained diode-pumped output-power levels exceeding 1 W at $3 \mu\text{m}$ from fluoride [22] and oxide [18], [34] crystalline host materials. However, as higher pump powers become available from laser diode

systems, it is generally recognized that thermal and thermo-optical issues set limitations to the power scalability of end-pumped bulk laser systems [35]–[37]. Owing to the unfavorable temperature dependence of thermal and thermo-optical parameters [38], the large heat load in the crystal leads, firstly, to a significant temperature increase in the rod, secondly, to strong thermal lensing with pronounced spherical aberrations and, ultimately, to rod fracture in a high-power end-pumped system. These effects are especially pronounced in highly erbium-doped crystals, as the multiphonon relaxations following the efficient ETU processes (Fig. 1) lead to significant extra heat generation compared to, e.g., $1\text{-}\mu\text{m}$ Nd^{3+} lasers under similar pump conditions [39].

A possible solution is diode side pumping, which leads to smaller excitation densities and correspondingly weaker ETU processes, as well as a better heat removal in the slab geometry. The highest output power of 1.8 W from an erbium $3\text{-}\mu\text{m}$ crystal laser [40] has been obtained in this way. A reduced erbium concentration with correspondingly smaller parameters of the ETU processes may aid this approach. However, the efficiency of the energy-recycling regime cannot be reached in this approach.

At this point it becomes clear that the erbium-doped fluoride fiber represents a promising alternative for the construction of a compact and efficient all-solid-state laser emitting at the transition at $3 \mu\text{m}$. Due to its geometry, the fiber provides large flexibility and potentially high pump- and signal-beam intensities without the drawbacks of thermal and thermo-optical effects.

II. EARLY INVESTIGATIONS OF ERBIUM $3\text{-}\mu\text{m}$ FIBER LASERS

A suitable host material for an erbium $3\text{-}\mu\text{m}$ fiber laser is the fluorozirconate glass ZBLAN (53 mol% ZrF_4 , 20 mol% BaF_2 , 4 mol% LaF_3 , 3 mol% AlF_3 , 20 mol% NaF). It was invented [41] in 1974. The Er^{3+} ion replaces the La^{3+} ion, i.e., the maximum possible Er^{3+} concentration in ZBLAN is 4 mol%. Any further increase in Er^{3+} concentration leads to a modification of the glass composition. Such highly erbium-doped fluorozirconate glasses have become available recently [42]. The maximum phonon energy in ZBLAN [43] is $\sim 500 \text{ cm}^{-1}$. ZBLAN is transparent in the mid-infrared spectral region. The loss at $2.7 \mu\text{m}$ in undoped ZBLAN glass is $\leq 10^{-1} \text{ dB/km}$ [44], [45]. Losses in fibers increase with the concentration of the rare-earth dopant. For $\sim 4 \text{ mol}\%$ Er^{3+} , the losses [46] specified by a fiber supplier are currently $< 50 \text{ dB/km}$. Its higher transparency and lower maximum phonon energy give ZBLAN a significant advantage over silica fibers for laser operation in the $3\text{-}\mu\text{m}$ spectral region.

However, ZBLAN also has some undesired properties. The fabrication of ZBLAN fibers is much more expensive than that of silica fibers. The material is hygroscopic, which leads to a degradation in quality of ZBLAN fibers in air within less than a year. ZBLAN fibers are very fragile as compared to silica fibers and have to be handled with extreme care. Last but not least, fluorozirconate materials are toxic, which represents a major disadvantage in medical applications. It can, therefore, be anticipated that an erbium $3\text{-}\mu\text{m}$ fiber laser based on ZBLAN will make its way to the market only if its performance (output power, slope efficiency, and beam quality) will be significantly better than

that achievable in 3- μm crystalline lasers. Recent results show that this might be the case in the near future.

The first erbium 3- μm fiber laser was demonstrated [47] in 1988. However, output powers remained at the submilliwatt level in the following years [48]–[50]. High dopant concentrations that could lead to efficient ETU and energy recycling (Section I-A) were unavailable at that time, and other ways to deplete the lower laser level in order to achieve higher output powers had to be found.

A. Continuous-Wave Inversion at a “Self-Terminating Transition”

Two important aspects were realized early in the investigation of this laser. Firstly, it is not solely the lifetime ratio that determines whether CW population inversion can be obtained between upper and lower laser level. Quimby and Miniscalco pointed out [51] that CW lasing can be obtained even at a so-called “self-terminating” four-level-laser transition, i.e., in a situation where the lifetime of the lower exceeds that of the upper laser level, if the lower laser level is not fed significantly by luminescent or multiphonon decay from the upper laser level. In addition, the Stark splittings of and Boltzmann distributions within the upper and lower laser levels contribute to population inversion, when the laser transition occurs between a low-lying Stark component of the upper and a high-lying Stark component of the lower laser level. Solution of a simple rate-equation system leads to the following condition for CW inversion [51], [52]:

$$\frac{b_2\tau_2}{\beta_{21}b_1\tau_1} > 1. \quad (3)$$

τ_i and b_i are the lifetimes and Boltzmann factors of, respectively, upper and lower laser levels and β_{21} is the branching ratio for decay from upper to lower laser level. For the ZBLAN:Er³⁺ laser transition at 3 μm , the measured lifetimes [19] of upper and lower laser levels are $\tau_2 = 6.9\text{ms}$ and $\tau_1 = 9.0\text{ms}$, respectively. With the decay rate $A_2 = 1/\tau_2 = 145\text{ s}^{-1}$ and the radiative rates $A_{21,\text{rad}} = 21\text{ s}^{-1}$ and $A_{20,\text{rad}} = 86\text{ s}^{-1}$ (from a Judd–Ofelt analysis [53]), a nonradiative rate of $A_{21,\text{nr}} = 38\text{ s}^{-1}$ and an overall rate of $A_{21} = 59\text{ s}^{-1}$ are derived, leading to $\beta_{21} = A_{21}/A_2 = 0.41$. These values fulfill the inequality (3), since $b_2 > b_1$ can be assumed (the Stark splitting cannot easily be measured in ZBLAN glass because of the strong broadening of the absorption and emission lines). CW laser operation can, therefore, be achieved in ZBLAN without employing any method to depopulate the $^4\text{I}_{13/2}$ lower laser level.

During the relaxation oscillations at the onset of lasing, a red-shift of the lasing wavelength is often observed in erbium 3- μm laser systems [54], because the excitation energy is accumulated in the long-lived $^4\text{I}_{13/2}$ lower laser level and the lasing process changes from four-level to three-level nature. At the beginning of laser emission, the lower multiplet is empty and the laser oscillates at short wavelength at a crystal-field transition with high emission cross section (four-level nature). As soon as the lower multiplet is populated during laser emission, reabsorption losses build up at the short-wavelength side of the fluorescence band, and the laser is forced to oscillate at longer wave-

Fig. 4. Effective cross sections of the absorption transitions of ZBLAN:Er³⁺ in the pump-wavelength region near 800 nm: Measured GSA transition $^4\text{I}_{15/2} \rightarrow ^4\text{I}_{9/2}$ (799 nm) and Gaussian fits of the ESA transitions $^4\text{I}_{13/2} \rightarrow ^2\text{H}_{11/2}$ (793 nm), $^4\text{I}_{11/2} \rightarrow ^4\text{F}_{3/2}$ (808 nm), and $^4\text{I}_{11/2} \rightarrow ^4\text{F}_{5/2}$ (831 nm). (Figure taken from [60].)

length at a crystal-field transition with lower emission cross section but also lower reabsorption losses (three-level nature). This effect was also reported for ZBLAN fiber lasers [55], [56]. For the same reason, the tuning range of a 3- μm CW laser [57] is narrowed and red-shifted with increasing pump power.

B. Influences of Excited-State Absorption

Secondly, it was realized that excited-state absorption (ESA) of pump light that had already been investigated [58] in other erbium-doped fibers for applications in the 1.5- μm telecommunications window has a major influence also on the population mechanisms of the low-doped, core-pumped erbium 3- μm ZBLAN fiber laser [59]. The measured cross sections [60] of the ESA transitions in ZBLAN in the region of the 800-nm pump GSA are shown in Fig. 4. The levels in which these transitions originate were determined from time-resolved ESA measurements [61]. At the short-wavelength side of GSA, an ESA transition originates in the $^4\text{I}_{13/2}$ lower laser level (Figs. 4 and 5), while at the long-wavelength side, two ESA transitions occur from the $^4\text{I}_{11/2}$ upper laser level (Fig. 4; not shown in Fig. 5). All these ESA transitions are sufficiently strong to populate the $^4\text{S}_{3/2}$ level and operate [62] the green upconversion laser transition $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ in a core-pumped ZBLAN fiber.

ESA from the $^4\text{I}_{11/2}$ upper laser level is detrimental to lasing and must be avoided. Experimentally, the best pump wavelength [59], [7] was near 792 nm, at the peak of ESA from the lower laser level (Fig. 4). ESA from the lower laser level was obviously important for the operation of this laser, despite the fact that the lifetime ratio and branching ratio (Section II-A) are generally sufficient for CW laser operation. The reason for the strong influence of ESA is the combination of a) the low dopant concentration, b) the high pump intensity in a core-pumped fiber, and c) the long lifetime of the lower laser level. This combination leads to a large fraction of ions being excited into the two laser levels and an accordingly large ground-state bleaching [7]. As a consequence, ESA from the laser levels is increased, GSA is decreased, and depletion of the lower laser level by ESA favorably results in a redistribution of its upconverted population, e.g., by green emission at the transition $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ to the ground state. This overcomes the bottleneck of the lower level lifetime.

Fig. 5. Partial energy-level scheme of erbium indicating the processes relevant to the cascade regime of the 3- μm laser at low dopant concentration. Two loops drive the 2.7- μm laser transition: Lower loop with GSA $^4I_{15/2} \rightarrow ^4I_{9/2}$, multiphonon relaxation to $^4I_{11/2}$, laser transition $^4I_{11/2} \rightarrow ^4I_{13/2}$ at 2.7 μm , fluorescence decay to $^4I_{15/2}$, and upper loop with ESA $^4I_{13/2} \rightarrow ^2H_{11/2}$, thermal relaxation to $^4S_{3/2}$, laser transition $^4S_{3/2} \rightarrow ^4I_{9/2}$ at 1.7 μm , multiphonon relaxation to $^4I_{11/2}$, laser transition $^4I_{11/2} \rightarrow ^4I_{13/2}$ at 2.7 μm . Competitive lasing at the transition $^4S_{3/2} \rightarrow ^4I_{13/2}$ at 850 nm, which is suppressed in the cascade regime, is also indicated.

Pumping near 792 nm provided the best results from the core-pumped fibers, although the Stokes efficiency at this wavelength is not optimal (Section I-B). Other pump transitions were operated less favorably [16]: Pumping near 650 nm [63] at the transition $^4I_{15/2} \rightarrow ^4F_{9/2}$ leads to a smaller Stokes efficiency than 800-nm pumping, and pumping near 980 nm [64] at the transition $^4I_{15/2} \rightarrow ^4I_{11/2}$ introduces ESA at the transition $^4I_{11/2} \rightarrow ^4F_{7/2}$ from the upper laser level [65]. This ESA transition is typically exploited for upconversion excitation of the green erbium laser transition $^4S_{3/2} \rightarrow ^4I_{15/2}$ in ZBLAN fibers [66], [67].

C. Output-Power Saturation at 3 μm

Despite the improvements in understanding the important spectroscopic processes, the performance of core-pumped erbium 3- μm fiber lasers remained unsatisfactory. The slope efficiencies obtained were <15%. Moreover, a saturation of the output power at 3 μm was observed (Fig. 6, circles), regardless of the pump wavelength, and the highest output powers reported were in the 20-mW region [68], [69].

The reason for the output-power saturation was not ground-state bleaching, as had been speculated earlier [70], because this effect was sufficiently reduced by ESA (Section II-B). However, another consequence of ESA, the excitation of the metastable $^4S_{3/2}$ level (lifetime $\sim 580 \mu\text{s}$ [19]), led to accumulation of excitation in this level and to inversion with respect to the $^4I_{13/2}$ level (Fig. 5). A second laser transition at 850 nm, which had been reported independently of the 3- μm research [71], repopulated the $^4I_{13/2}$ lower laser level of the 3- μm transition. Because of the high emission cross section at 850 nm and the high gain in the fiber, this transition lased even with only the fresnel

Fig. 6. Results for core-pumped low Er^{3+} doped fiber lasers in cascade and competitive lasing regimes: Output power at 2.7 μm versus launched pump power for cascade lasing at 2.7 and 1.7 μm (pump threshold for 1.7- μm lasing is 120 mW) compared to competitive lasing at 2.7 and 0.85 μm (pump threshold for 0.85- μm lasing is 500 mW). (Data taken from [6] and [7].)

reflections from the fiber ends, as did the 2.7- μm transition. Since the excitation density was large and the ESA cross section was higher than that of GSA (Fig. 4), ESA prevailed over GSA, and with increasing pump power, the system operated more and more at the 850-nm transition, causing the 3- μm laser to saturate at low output powers [7] (Fig. 6, circles).

III. OPERATIONAL REGIMES OF ERBIUM 3- μm FIBER LASERS

In this section, we will review how the knowledge that had been established on the basis of the spectroscopic investigations, the failure of the early fiber system to produce high efficiencies and output powers, and the success of the crystal system (Sections I and II) was exploited to operate erbium 3- μm fiber lasers with high efficiency and output power. The breakthrough in these recent years is related also to the progress in glass and fiber technologies.

A. Low Erbium Concentrations: High Slope Efficiencies from Cascade Lasing Schemes

Since ESA was important in order to reduce ground-state bleaching in a low-doped, core-pumped fiber (Section II-B), improvements in the performance of this laser system could only be expected when avoiding the competitive laser at 850 nm. The idea was to make appropriate use of the excitation energy that was unavoidably accumulated in the $^4S_{3/2}$ level (Fig. 5).

The solution was found [6] by deliberately operating a third laser transition $^4S_{3/2} \rightarrow ^4I_{9/2}$ at 1.7 μm , which had also been reported [72] independently of the 3- μm research. With suitable mirrors that were highly reflective at 1.7 μm and highly transmissive at 850 nm, the threshold of the former transition could be decreased to a value below that of the latter (Fig. 5). Under steady-state conditions, the $^4S_{3/2}$ population density was clamped to threshold inversion with respect to the short-lived $^4I_{9/2}$ level, which was depopulated by fast multiphonon relaxation to the $^4I_{11/2}$ upper laser level. In this way, the 850-nm transition was kept below threshold.

When the 1.7- μm laser reached threshold the energy was recycled into the upper laser level and the slope efficiency of the 2.7- μm transition increased significantly [8]. Since each GSA or ESA pump photon can at best produce one laser photon in this operational regime, i.e., the quantum efficiency becomes

unity in (1), the theoretical limit of the slope efficiency is given by the Stokes efficiency [16], 29% under 800-nm pumping. A slope efficiency of 23% was demonstrated experimentally [6]. The output power of the 3- μm fiber laser was improved by an order of magnitude to 150 mW, limited only by the available pump power [6] (Fig. 6, diamonds). This cascade-lasing regime represents the best option for dopant concentrations of typically 0.1 mol% ($\sim 1.6 \times 10^{19} \text{ cm}^{-3}$) at which ESA is important. Also a three-transition-cascade lasing regime with additional lasing at the transition $^4I_{13/2} \rightarrow ^4I_{15/2}$ near 1.6 μm was demonstrated [73], but no further improvement was obtained.

This first breakthrough, which was obtained in 1995, pointed out the potential of the erbium 3- μm fiber laser and initialized further research efforts in this field.

B. Medium Erbium Concentrations: Lifetime Quenching and Simple Four-Level Laser Operation

With the availability of low-loss ZBLAN fibers with higher dopant concentrations and with double-clad geometry, ESA became much less important, because the larger number of erbium ions led to a relative increase of the importance of GSA as compared to ESA, and the reduced pump intensity under cladding pumping with low-brightness diode lasers led to smaller excitation densities. The new geometry allowed for much higher pump powers to be coupled into the fiber, but it also meant that the cascade regime (Section III-A) was not applicable anymore, because this regime relied on upconversion by ESA.

In addition, the relatively high fabrication costs of ZBLAN double-clad fibers imposed limitations on the flexibility of the work and altered the way in which the research proceeded. Mistakes in the initial fiber design were intolerable, and pre-calculation and optimization of the expected device performance on the basis of the available spectroscopic parameters [74], [75], [43], [60], [65], [19] became an important tool.

In 1997, when we prepared our next step toward a high-power erbium 3- μm fiber laser by performing rate-equation analysis [76], the situation was still quite different from what it is today. The highest available dopant concentrations in ZBLAN fiber were typically 1 mol% ($\sim 1.6 \times 10^{20} \text{ cm}^{-3}$). One measurement of ETU in ZBLAN:Er³⁺ [43] suggested that ETU was inefficient even up to an erbium concentration of 4 mol%. Another paper [77] reported parameters of $W_{11} = 4.4 \times 10^{-18} \text{ cm}^3 \cdot \text{s}^{-1}$ and $W_{22} = 1.5 \times 10^{-17} \text{ cm}^3 \cdot \text{s}^{-1}$ for ETU from the $^4I_{13/2}$ and $^4I_{11/2}$ levels, respectively, for an erbium concentration of 2 mol%. These values are a factor-of-three smaller for ETU from $^4I_{13/2}$ and a factor-of-five larger for ETU from $^4I_{11/2}$ than the parameters determined in our recent measurements [19] (Fig. 3) and would lead to a ratio W_{11}/W_{22} of only 0.3 in (2). Based on the erbium concentrations in ZBLAN fibers available on the market and the reports on ETU published in the literature, the energy-recycling regime (Section I-A) was not an option for the fiber system, and still an eye had to be kept on avoiding the possibility of ESA and uncontrolled redistribution of the upconverted energy from $^4S_{3/2}$ at these dopant concentrations.

We chose an approach (reported earlier for erbium 3- μm lasers [78], [31], [68]) that can operate the erbium 3- μm transition as a simple four-level laser (Fig. 7): The $^4I_{13/2}$ lower laser

Fig. 7. Partial energy-level scheme of erbium indicating the processes relevant to the lifetime-quenching regime of the 3- μm laser at medium dopant concentration. The laser operates as a simple four-level laser: GSA to the $^4I_{9/2}$ pump level, multiphonon relaxation to the $^4I_{11/2}$ upper laser level, laser transition to the $^4I_{13/2}$ lower laser level, and relaxation to the ground state via energy transfer ET₁ to the Pr³⁺ codopant. The energy transfer ET₂ from the $^4I_{11/2}$ upper laser level to the Pr³⁺ codopant is weak.

level is depopulated by energy transfer to a Pr³⁺ codopant and fast decay to the ground state by multiphonon relaxation within Pr³⁺ (Fig. 7). As was suggested by lifetime measurements [79] and confirmed later when the corresponding parameters were determined [19] (Fig. 3), this energy-transfer process (ET₁ in Fig. 7) is much more efficient than the corresponding energy-transfer process (ET₂ in Fig. 7) from the $^4I_{11/2}$ upper laser level to the Pr³⁺ co-dopant, because the oscillator strength in Pr³⁺ is much higher for the transition involved in the former process [19]. The relatively weak lifetime quenching of the upper laser level affects the pump threshold, which is small anyhow, but it does not influence the slope efficiency. The strong lifetime quenching of the $^4I_{3/2}$ lower laser level and the fact that the $^4I_{11/2}$ population density is clamped to laser threshold significantly reduce ground-state bleaching and excitation of the laser levels, thus making negligible the influence of ESA, but similarly preventing energy recycling by ETU [80].

Like in the cascade-lasing regime (Section III-A), each pump photon can at best produce one laser photon in the Er³⁺, Pr³⁺-codoped system. The quantum efficiency becomes unity in (1), and the theoretical limit of the slope efficiency is given by the Stokes efficiency, 29% under 800-nm pumping. Experimentally, a slope efficiency of 17% and an output power of 1.7 W were obtained [46] (Fig. 8). This represented another order-of-magnitude improvement compared to the cascade-lasing regime (Fig. 6, diamonds). Other researchers [81] followed the same proposal [76] and reported output powers of 660 mW. Since ESA from both laser levels is negligible, the system can alternatively be pumped near 980 nm, which provides the highest possible Stokes efficiency of 35% (Section I-B). In this way, the experimental slope efficiency could be increased to 25% [82].

Fig. 8. Results for a cladding-pumped Er^{3+} , Pr^{3+} -codoped fiber laser: Output power at $2.7 \mu\text{m}$ versus launched pump power at 790 nm of the diode-pumped double-clad ZBLAN fiber laser. Pump threshold is 0.5 W and slope efficiency is 17% with respect to launched pump power. (Data taken from [46].)

The ZBLAN glass and fiber technologies had improved since the time of our initial calculations, and the fiber that was used in these experiments in 1999 contained already an erbium concentration of $3.5 \text{ mol}\%$.

C. High Erbium Concentrations: Energy Recycling by Energy-Transfer Upconversion

It was shown (Section I-A; Fig. 2) that the generally most efficient way for operation of the erbium $3\text{-}\mu\text{m}$ laser is energy recycling by ETU. This statement holds true also for $3\text{-}\mu\text{m}$ ZBLAN fiber lasers. The criterion for optimization of the slope efficiency in this operational regime (Section I-B; Fig. 1) is a maximum ratio W_{11}/W_{22} of the parameters of ETU from lower versus upper laser level. The parameters of ETU in ZBLAN bulk glasses [19] for currently available dopant concentrations are shown in Fig. 3. For Er^{3+} concentrations up to $8.75 \text{ mol}\%$, ETU has approximately a factor-of-three larger probability for the lower laser level as compared to the upper laser level. These values are more favorable than those reported [18] for $\text{LiYF}_4:\text{Er}^{3+}$. From the optimum erbium concentrations in different crystalline host materials (Section I-B) and the trend within this series with respect to maximum phonon energy of the host lattice, one can estimate that the optimum erbium concentration for energy recycling in a $3\text{-}\mu\text{m}$ ZBLAN fiber laser is probably $\sim 10\text{--}15 \text{ mol}\%$ ($1.4\text{--}2.0 \times 10^{21} \text{ cm}^{-3}$).

With the measured ETU parameters, we calculated the performances of singly Er^{3+} -doped and Er^{3+} , Pr^{3+} -codoped $3\text{-}\mu\text{m}$ ZBLAN fiber lasers versus erbium concentration [83]. The results (Fig. 9) show that the benefits of energy recycling by ETU versus lifetime quenching by energy transfer to Pr^{3+} should be present already at an erbium concentration of $4 \text{ mol}\%$. However, a number of authors reported on active-ion clusters in other fiber materials [84]–[89]. It is not clear at present [19] whether such clustering also occurs in ZBLAN bulk glasses and/or fibers and whether the ETU parameters measured in ZBLAN bulk glasses are also valid for ZBLAN fibers, i.e., whether the results presented in Fig. 9 are correct.

Two research groups tried to take advantage of energy recycling at high erbium concentrations by use of fibers without Pr^{3+} codoping. In the first case, the erbium concentration of $2 \text{ mol}\%$ was still quite low [90], and the reported slope efficiency of 16% versus absorbed pump power at 791 nm , which

Fig. 9. Future prospects for double-clad highly Er^{3+} doped fiber lasers: Calculated slope efficiencies for Er^{3+} singly-doped and Er^{3+} , Pr^{3+} -codoped ZBLAN fibers versus Er^{3+} concentration. (Figure taken from [83].)

was smaller than the 17% obtained in the Er^{3+} , Pr^{3+} -codoped fiber [46], does not allow for the conclusion that enhanced energy recycling by ETU was present in the singly erbium-doped fiber laser. In the second case, an M-profile fiber was used [91]. The main advantage of the M-profile geometry is its high pump absorption, resulting in short fiber length, while its main drawbacks are high fabrication costs and the poor beam quality of its output. The high pump absorption and, thus, higher excitation densities would support ETU. Pumping at 980 nm increased the slope efficiency (Section I-B) and further improved the pump absorption compared to 800-nm pumping. In addition, an erbium concentration as large as $5 \text{ mol}\%$ was used. The slope efficiency, however, did not exceed 25% , the same result as obtained in the Er^{3+} , Pr^{3+} -codoped fiber under 980-nm pumping [82].

The question at which erbium concentrations the energy-recycling regime can work efficiently in $3\text{-}\mu\text{m}$ ZBLAN fiber lasers has as yet not been answered, and the high slope efficiencies achieved in crystal laser systems remain to be demonstrated in fibers. Since higher erbium-doped fluorozirconate fibers are commercially available now, new results will probably be reported within the next 12 months. Output powers at $3 \mu\text{m}$ of 5 W have been predicted [83]. With suitable pump sources, highly erbium-doped low-loss fibers, and a proper design of the fiber geometry, even output powers of $>10 \text{ W}$ in a transverse single mode seem feasible in the future. Such high output powers may exceed the power levels that can possibly be achieved with bulk crystalline materials, if the thermal and thermo-optical disadvantages of the bulk geometry are not overcome.

IV. FUTURE PERSPECTIVES AND NEW DEVELOPMENTS

Besides the possibility to improve the output power from erbium $3\text{-}\mu\text{m}$ CW fiber lasers by yet another order of magnitude, further research directions will be pursued, with the aim to provide a $3\text{-}\mu\text{m}$ laser source that can be used in surgery. One obvious question concerns the effects that occur if CW powers of several watts are applied to biotissue. We do not know so far what will be the specific fields of this new laser source in surgery, because most of the previous investigations used pulsed laser sources at $3 \mu\text{m}$.

Pulsed output at $3 \mu\text{m}$ has been generated from erbium-doped crystalline materials in many configurations and regimes, e.g.,

under quasi-CW pumping [27], [29], active [92], [93] and passive [94] Q -switching, and mode-locking [94]. The first steps toward pulsed output from erbium 3- μm ZBLAN lasers [56], [95] were unsatisfactory in terms of output energies and average powers. Further investigations will be necessary. With the recent advances in nonlinear frequency conversion by use of periodically poled materials, 3- μm sources based on optical parametric oscillation [96] will be strong competitors in the pulsed regime.

New excitation schemes will be tested. A possible way to reduce fiber lengths and costs is codoping the ZBLAN:Er³⁺ fiber with Yb³⁺, which has a high absorption cross section at 975 nm, and sensitization of the erbium upper laser level by energy transfer. This approach works well for 1.5- μm laser systems [97] that use host materials with high phonon energies and, thus, reduced ⁴I_{11/2} lifetime. In ZBLAN, back transfer to Yb³⁺ and/or ETU from ⁴I_{11/2} by energy transfer from Yb³⁺ may turn out to be a problem.

Host geometries other than glass fiber or crystalline bulk geometries are under investigation. Whereas an approach that uses glass bulk materials [98] may suffer from the same thermal and thermo-optical drawbacks as the crystalline bulk materials, with even decreased thermal conductivity in the glass, crystalline waveguides [99] may show some prospects for high slope efficiencies at moderate output powers due to the confinement of pump and signal beams. The slab geometry [40] will lead to further increase of the output powers obtained from crystalline bulk materials due to its improved thermal management.

New glass and crystalline materials that possess very promising properties for laser operation at 3 μm have been developed. Gallium lanthanum sulfide glass fibers [100] have low phonon energies, are transparent up to the mid-infrared spectral region, and rare-earth ions doped into these glasses exhibit large oscillator strengths. Sesquioxide crystals [101] also have low phonon energies that are suitable for 3- μm lasing, but possess heat conductivities larger than that of YAG, which may help in reducing thermal and thermo-optical effects.

Last but not least, spectroscopy and laser oscillation in the 3- μm spectral region from other rare-earth ions such as Ho³⁺ [102], [103] and Dy³⁺ [104], [105] have been reported for crystals and glass fibers as host materials. Whereas the 3- μm transition in Dy³⁺ is a ground-state transition that suffers from re-absorption losses and an accordingly small slope efficiency, the Ho³⁺ transition may someday become a competitor for the erbium 3- μm CW fiber laser.

V. CONCLUSION

The variety of spectroscopic phenomena and its fundamentally different operational regimes that all have a single effort in mind, namely to decrease the population density in the lower laser level, have made the erbium 3- μm fiber laser one of the most complex and best investigated solid-state laser systems ever. It has been understood how the influences of individual spectroscopic processes on the population mechanisms change dramatically with dopant concentrations and pump geometries. Interestingly enough, the final step toward a high-power CW 3- μm fiber laser, which will probably take place within the next 12 months and which may surpass the powers obtained from

crystal lasers, will exploit exactly that knowledge that was established for the crystal laser system and, thus, point back to the early ideas that were developed by Russian scientists like Bagdasarov, Prokhorov, Zharikov, Zhekov, and co-workers. After more than 20 years of research, the circle of the spectroscopic investigations of the erbium 3- μm laser closes.

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