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Recent advances in crystal optics/Avancées récentes en optique cristalline

Review of photorefractive materials: an application to laser beam cleanup

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Invited Paper

Abstract

Nonlinear photorefractive materials are well suited to record dynamic volume holograms using the wave mixing of coherent laser beams. The photo-induced index modulation is due to a space charge field which modulates the crystal refractive index. The basic phenomena and beam interactions are reviewed for different types of materials operating in the visible and near infrared. In particular, we outline their ability to amplify a low intensity signal beam due to the intensity transfer of a spatially multimode pump beam. We apply this interaction to the cleanup of a multimode large core fiber amplifier. This class of nonlinear materials contribute to extending the performances of laser sources for advanced applications of photonics. ***To cite this article: L. Lombard et al., C. R. Physique 8 (2007).***

Résumé

Revue sur les matériaux photoréfractifs : application à l'amélioration de la qualité spatiale de faisceau laser. Les matériaux nonlinéaires photoréfractifs sont bien adaptés à l'enregistrement d'hologrammes de volume en exploitant le mélange d'ondes de faisceaux laser cohérents. Le mécanisme de changement d'indice est dû à un effet de charge d'espace qui module l'indice du cristal électro optique. Les mécanismes de base et interactions entre faisceaux sont présentés pour différent types de matériaux fonctionnant dans le visible et proche infrarouge. En particulier on décrit la possibilité d'amplifier un signal de faible intensité par transfert de l'intensité d'un faisceau pompe spatialement multimode. On applique cette interaction pour obtenir un faisceau de haute qualité spatiale à partir d'un laser à fibre à gros cœur multimode. Cette classe de matériaux nonlinéaires contribue à améliorer les performances des sources laser destinées aux applications de la photonique. ***Pour citer cet article : L. Lombard et al., C. R. Physique 8 (2007).***

Keywords: Photorefractive materials; Laser beam cleanup

Mots-clés : Matériaux photoréfractifs ; Amélioration de faisceau laser

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1. Introduction to photorefractive effects and materials

The research and developments in laser physics and nonlinear optics have rapidly expanded over the last twenty years, and many applications of photonics are now relevant to industrial or consumer markets. Also, photonics will be disseminated in the near future into new important areas such as, medicine, biology or nanotechnologies. A major factor behind such a growth is the advent of high power and efficient solid state lasers, in combination with the use of materials that exhibit large second or third order nonlinearities. In such conditions, nonlinear optics is becoming an important technology in the design of new laser sources emitting in the visible or near infrared. Nonlinear optics also enables the attainment of new functionalities in laser systems and in optoelectronic signal transmission and processing. The class of nonlinear phenomena based on the photorefractive effects in electro-optic crystals will undoubtedly play a major role for these different applications of laser photonics.

Since its discovery by Ashkin et al. in 1966 [1], photorefraction has stimulated much basic research covering both fundamental materials studies and their applications in dynamic holography, laser beam control, optical processing, ... To briefly introduce this particular field of nonlinear optics let us recall in the following the basic of the physical mechanisms and main characteristics [2–4]. When coherent laser beams interfere and create a grating in the volume of the material, they generate a photoinduced space charge field which modulates the crystal or polymer refractive index through the linear Pockels electro-optic effects. The space charge field arises from the photoconductive properties of the crystal involving the diffusion (at zero applied field) or the drift (at large applied field) of the photoinduced charge carriers in the crystal volume: after charge trapping it generates the space charge field. It is the drift and the diffusion length of the charges which control the recording slope of the grating. In other words, photorefractive materials are very well suited to record dynamic volume holograms using two-wave or four-wave mixing beam interference with continuous wave or pulsed lasers. Since the magnitude of the nonlinear index modulation is also proportional to the absorbed incident energy, there is a trade off between response time and material photosensitivity. The result is that, in general, a photorefractive mechanism is not as fast as other nonlinearities such as those of Kerr, Raman and Brillouin.

For many years only the challenge to understand the basic mechanisms giving rise to this effect in different crystals, drove the research. More and more materials were found to be photorefractive. New electro-optic interactions were discovered. The fields of nonlinear optics, optical spectroscopy, electro-optics, ferroelectrics, electronic transport and Fourier optics were brought together to develop a complete understanding of the complex microscopic mechanism involved.

Another important aspect of photorefractive materials is their ability to perform efficient energy transfer between a signal and a reference beam interfering in the volume of the crystal. This property opens a wide range of applications, including image amplification, optical phase conjugation with gain and selfpumped optical cavities. The amplitude of the beam intensity transfer is governed by the gain coefficient whose value is deduced from the intrinsic material parameters (trapping center density, electro optic coefficient, diffusion length, quantum efficiency, ...). Remarkable properties of photorefractive crystals come from this gain coefficient. In particular, a fascinating example is the self-pumped phase conjugate mirror which provides a dynamic correction of beam aberrations after reflection on the nonlinear mirror. Thus the wavefront distortions and the thermal lensing effects can be compensated in a laser cavity in order to operate with optimum power and brightness. To achieve these results and to develop applications require us to optimize nonlinear material properties, and therefore the choice of the best material is a critical issue, as will be shown in the following.

Nonlinear photorefractive optics is now well established and it has reached scientific maturity. It contributes to stimulate basic research in solid state physics and to investigate with details the mechanisms of charge transport in different types of ferroelectric or semiconductor crystals such as $LiNbO_3$, $BaTiO_3$, $KNbO_3$, $Bi_{12}SiO_{20}$ or GaAs. The basis of the early works on the charge transport models and beam coupling phenomena in these materials were investigated in depth. It has thus resulted in great interest to grow new doped crystals whose photorefractive properties can be tailored to applications, encompassing materials that have no photorefraction to materials that can exhibit large photoinduced index modulations.

Materials showing photoinduced refractive index changes via space-charge driven electro-optical effects presently cover a wavelength range from 257 nm [5] to 1550 nm [6]. The short wavelength region from the near-ultraviolet to the blue–green spectral range is presently dominated by inorganic crystals. Organic crystals and photorefractive polymers [7,8] occupy mainly the red to near-infrared border region, while semiconductors and $Sn_2P_2S_6$ crystals [9] respond at infrared wavelengths larger than 800 nm. Multiple quantum well photorefractive devices (MQW) differ from the

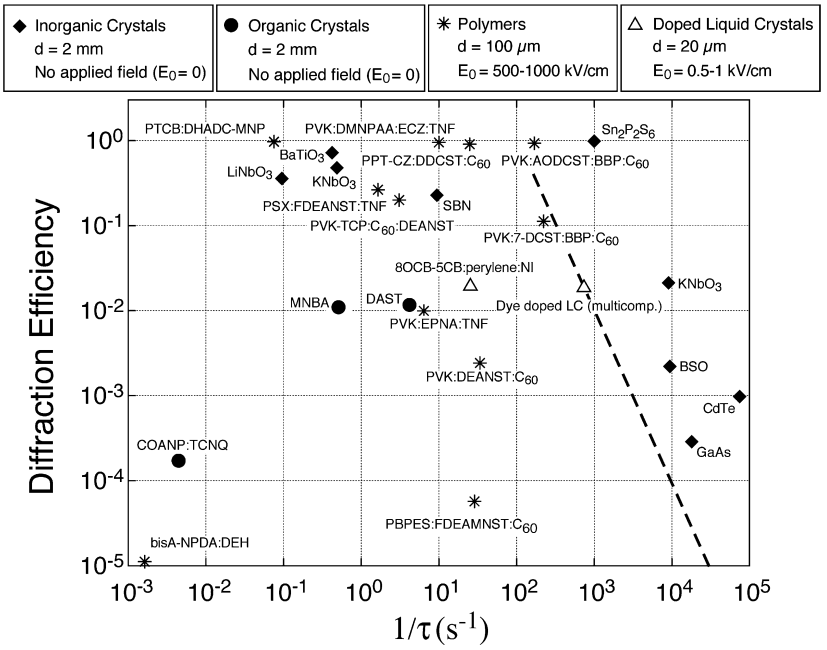

Fig. 1. Diffraction efficiency versus growth rate for a selection of photorefractive crystals, polymers and doped liquid crystals (abbreviations of organic crystals, polymer composites: see [8,10]. The values are scaled to 1 W/cm^2 light intensity.

other materials by the fact that thin holographic gratings can be recorded and read out with only a few nanometer-wide spectral band corresponding to the excitonic resonances.

For many applications the most important material parameters are the photoinduced refractive index changes (dynamic range), the two-wave mixing gain, the recording time or the photosensitivity. The refractive index changes mainly depends on the electro-optic figure of merit n^3r (n: refractive index; r: electro-optic coefficient) and on the effective density of trapped photomigrated charges.

A brief graphical overview of the performances of the most important materials (inorganic and organic crystals and polymers) is shown in Fig. 1 [8,10].

The diagram of Fig. 1 shows the steady state diffraction efficiency and the inverse response time obtained in a selection of inorganic and organic crystals, photorefractive polymers and dye-doped liquid crystals. The diffraction efficiencies are scaled to a typical thickness d of actual samples, i.e. $d = 100$ μm for polymers, $d = 20$ μm for liquid crystal cells and $d = 2$ mm for inorganic and organic crystals. The response time is scaled to a light intensity about 1 W/cm^2. The data for polymers are taken under an applied electric field of typically 500–1000 kV/cm, for liquid crystals under a field of 0.5–1 kV/cm, while those for inorganic and organic crystals are under zero-field conditions. The dashed line in the diagram connects points of constant material photosensitivity.The region of the best materials (the upper right corner in the diagram) is still occupied mainly by inorganic crystals; however the best polymers and liquid crystals are approaching this region. We also include a new inorganic crystal: $Sn_2P_2S_6$ (tin hypothiodisulphate), a wide bandgap semiconducting ferroelectric with a high sensitivity, combining fast recording times and large diffraction efficiencies in the near infrared as reported recently [9].

2. An application of photorefractive nonlinear optics for high power fiber lasers

The performance and the technology of fiber lasers capable of delivery high power have progressed very rapidly since early demonstrations more than ten years ago. Over the past two years, the level of CW power which can be produced at one micron wavelength with a good beam quality has increased from 100 W to nearly 1 kW [11]. In the ns pulse regime, output energies of a mJ are demonstrated. These lasers exhibit intrinsic advantages in comparison with classical solid state technology: fiber lasers can be made more compact, efficient, reliable and cost effective. These

qualities are of major interest in scientific applications, for industrial systems or material processing. It also provide very attractive coherent sources for metrology, imaging or atmospheric remote sensing.

To realize high power fiber lasers, it is required to use a large core area; it is the only practical way to reduce both the susceptibility of the fiber to optical damage and also the appearance of detrimental nonlinear effects (Brillouin, Kerr, Raman ...) which would limit the output power of the source and disturb the spectral qualities. The high numerical aperture of the double clad also permits to efficiently collect the pumping power issued from the diodes. In order to realize a large core fiber, the numerical aperture NA has to be decreased. This conclusion appears from the following simple formalism: a step index fiber is single mode when the normalized parameter $V = \frac{2\pi}{\lambda} R_c \sqrt{n_c^2 - n_g^2} = \frac{2\pi}{\lambda} R_c NA$ satisfies the condition $V < 2.4$. R_c is the core radius, n_c and n_g are respectively the core and clad index of refraction. Therefore, the recent availability of large mode area (LMA) fibers with a diameter of 20 to 30 μm and low NA of ≈0.06 has made possible new progress and performances. However, for a further increase of the current LMA fiber laser performance, in particular in the short pulse regime, it will be required to switch to very large mode area fibers (VLMA). Fulfilling the condition on V is no more possible: this would lead to weak guiding in the core and eventually makes very low NA fibers extremely sensitive to bending losses.

Therefore, there will be a need for step index fiber amplifiers with large enough NA and doped core diameters in the range of 50 to 500 μm. They would permit the delivery of output energies beyond several 10 mJ with several 10 ns pulse duration. Such a very large mode area will inevitably support many propagation modes, thus leading to a reduced beam quality due to mode coupling, after several meters of beam propagation. The resulting large value of the beam quality M^2 factor ($M^2 \gg 1$) will provide an important beam divergence and a complex speckle structure due to interference between the modes. In view of recovering a nearly diffraction limited beam quality (beam quality parameter $M^2 \approx 1$) in such a fiber, new concepts must be investigated [12–15]. An original approach in [16] is to implement a nonlinear mode conversion. In other words, the multimode beam issued from the very large core fiber amplifier is transformed into a single mode Gaussian beam. To achieved this function, two types of optical configurations are proposed: first, the beam cleanup method based on nonlinear two wave mixing [12], second optical, phase conjugation. These ideas which are issued from the research works on nonlinear wave mixing techniques applied to beam correction are shown in Figs. 2 and 3. According to Fig. 2, the multimode signal beam transfers its intensity (and not the phase) to a single mode reference beam after two beam interaction in a nonlinear media which exhibits a photoinduced index nonlinearity for beam cleanup. In Fig. 3, the output multimode beam is reflected by a nonlinear mirror which generates the phase conjugate replica of the complex incident wavefront. After a double pass through the amplifier, a nearly diffraction limited beam can be recovered. According to our previous experiences at Thales Research and Technology on these subjects we validate these original concepts with multimode fiber amplifiers operating

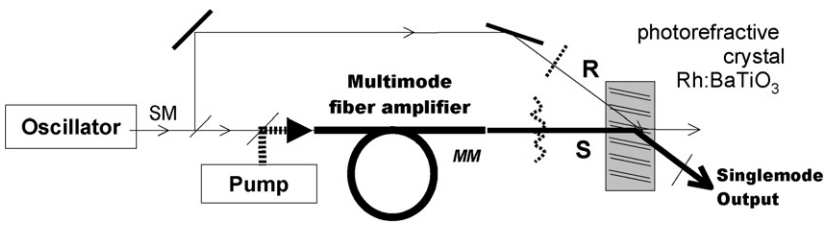

Fig. 2. Multimode amplifier with nonlinear beam cleanup by two wave mixing in a photorefractive crystal. SM: Single-mode input beam. MM: Amplified multimode beam.

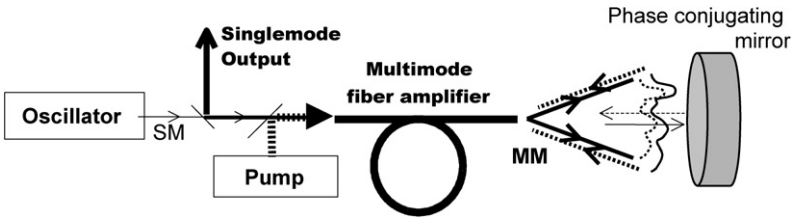

Fig. 3. Multimode amplifier with phase conjugating nonlinear mirror. SM: Single-mode input beam. MM: Amplified multimode beam. Output is single mode after two pass amplification and phase conjugation.

in CW at moderate output power levels. Single mode, single frequency and polarized beam emission is demonstrated using respectively a photorefractive crystal for beam cleanup and Stimulated Brillouin Scattering.

The approach firstly reported in [16] consists in correcting the beam profile after the multimode amplifier by a nonlinear beam cleanup method. The photorefractive two-wave mixing mode conversion has been validated in previous works with visible light but here we propose and demonstrate an improved version of this technique applied to a diode pumped multimode Yb-doped fiber amplifier operating in the near infrared one micron region.

3. Principle of photorefractive two-wave mixing beam cleanup

Diffusion-type (at zero field) photorefractive materials allow intensity transfer between the two coherent signal and reference beams writing a phase shifted dynamic phase volume hologram. The two-wave mixing interaction originates from self diffraction of the two incident beams when they propagate through the photorefractive crystal [17]. This effect is shown on Fig. 4 and it is expressed in the coupled wave equations (1), (2) when the shift between intensity and index grating is equal to a quarter of the grating period. I_R, I_S are respectively the incident reference and signal beam intensities, Γ is the exponential photorefractive gain coefficient (cm^{-1}) and α is the absorption coefficient (cm^{-1}):

$$\frac{\mathrm{d}I_S}{\mathrm{d}z} = -\Gamma \frac{I_S I_R}{I_S + I_R} - \alpha I_S \tag{1}$$

$$\frac{\mathrm{d}I_R}{\mathrm{d}z} = \Gamma \frac{I_S I_R}{I_S + I_R} - \alpha I_R \tag{2}$$

After propagation in a crystal with a thickness d, beam R experiences an intensity gain G given by relation (3) where β is the signal to reference intensity beam ratio:

$$G = \frac{I_R(d)}{I_R(0)} = \frac{\beta + 1}{\beta + \mathrm{e}^{\Gamma d}} \mathrm{e}^{(\Gamma - \alpha)d}, \quad \beta = \frac{I_S(0)}{I_R(0)} \tag{3}$$

The wavefront of the two interfering beams are unaffected in the interaction: S transfers its intensity to R and not its phase. This property is used for two-wave mixing beam cleanup. The high intensity and multimode beam S is diffracted into a single mode intense beam R. In other words, photorefractive two-wave mixing acts as a nonlinear spatial mode converter.

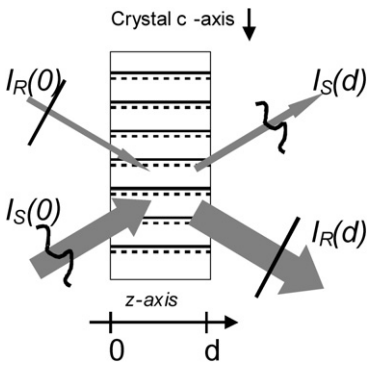

Fig. 4. Two wave mixing in a photorefractive crystal. $I_R(z)$, $I_S(z)$: intensities of beams R and S along the propagation. Continuous lines: maxima of the interference pattern. Dashed lines: maxima of the shifted index pattern.

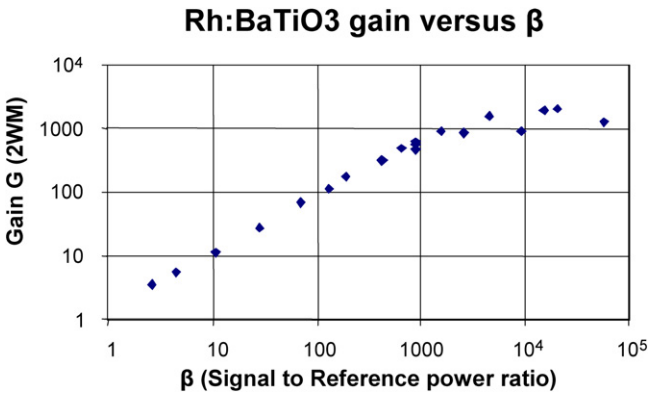

Fig. 5. Measured photorefractive gain of Rh $BaTiO_3$ in a two wave mixing experiment as a function on incident beam intensity ratio β.

4. A Rh:$BaTiO_3$ photorefractive crystal for beam cleanup of a multimode Yb doped fiber amplifier

The nonlinear material used in the experiment is a Rh:$BaTiO_3$ crystal This crystal is well known for efficient operation in the visible since it exhibits in the blue–green spectral range a very high photorefractive gain and a response time usually of the order of a fraction of second. The extension of the performances of the crystal in the near IR is not straightforward and significant material research works were pursued in order to control the spectral sensitivity. By growing crystals with Rh doping it is now possible to achieve higher absorption and a significant improvement of the photoconductive properties for longer wavelengths. In such conditions the crystal can now be used for efficient operation with fiber lasers. The photorefractive gain is nearly equivalent to the values measured in the visible, while the response time is longer compared to the visible. These considerations justify the use of the Rh doped crystal in the present experiment which demonstrates laser beam cleanup.This crystal has a 'roof shape' to prevent any parasitic oscillation arising from beam fanning [15]. The input and output faces are antireflection coated and are cut at 45° from the crystal c-axis in order to access large photorefractive gains. The crystal thickness is 3 mm. The measured gain on Fig. 5 is equal to $G_{\max} = 2000$, which corresponds to a rather large photorefractive gain coefficient Γ of 24.6 cm^{-1}.

5. Experimental setup

In this work, the photorefractive beam cleanup is applied to a diode pumped multimode-Yb-doped-fiber amplifier. A CW single frequency master oscillator (Nd:YAG laser, 500 mW at 1.06 μm) is amplified in a multimode fiber amplifier. The amplifier uses a double clad fiber with a 55 μm diameter (NA = 0.19) Yb-doped signal core and a 400 μm diameter D-shaped pump core. A picture of the fiber is shown on Fig. 6.

The 3.5 m long fiber absorbs a power of 60 W of the 940 nm pumping diode. The amplified signal beam S has a power of 18 W, is depolarized and bears many transverse modes visualized as speckle grains in Fig. 6. All the speckle grains have a different elliptical polarization but are coherent with each other.

To realize the beam cleanup interaction we have considered the experimental setup shown in Fig. 7. In this experiment the beam issued from the multimode fiber amplifier is used as the signal beam which interferes with a diffraction

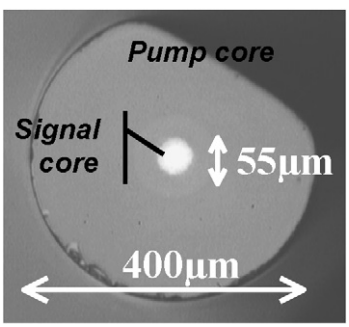

Fig. 6. Fiber used in the experiment, with a Yb-doped core and a D-shaped pump core.

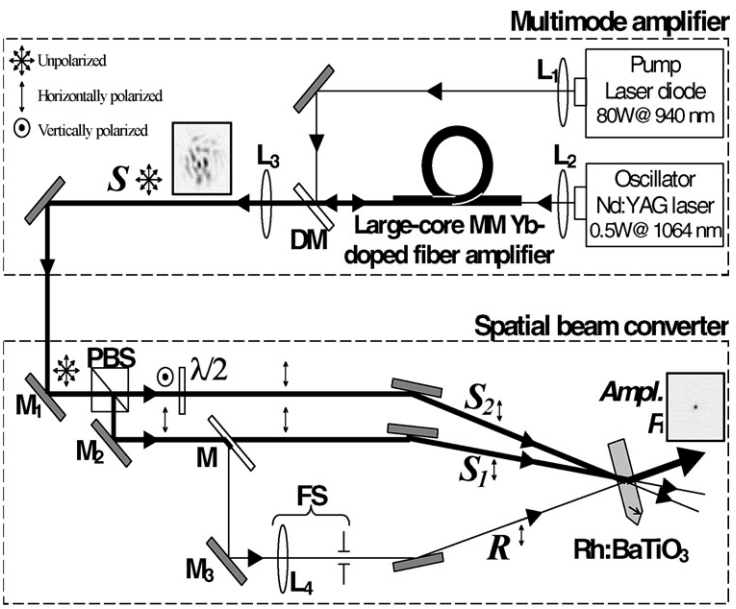

Fig. 7. Experimental setup of an Yb-doped multimode fiber amplifier with nonlinear mode converter. The amplified depolarized multimode signal beam S is split into its two linear polarizations and combined with single-mode beam R in a Rh:$BaTiO_3$ crystal. M: Spot mirror of the size of one speckle grain of beams S. FS: Spatial filter. Signal and amplified reference beam profiles are shown in the squares.

limited low intensity reference beam. After two wave mixing in the photorefractive crystal the high intensity multimode signal is transfered into the direction of the reference. The spot mirror in Fig. 7 acts a spatial filter to select the low intensity reference beam, and its phase is preserved after the nonlinear interaction. The polarizing beam splitter permits us to take account of the beam depolarization after propagation in the multimode amplifier. This setup, and the two wave mixing interaction, provide the required nonlinear mode conversion and beam cleanup: it amplifies the reference beam after propagation through the crystal but its phase profile is unchanged.

To operate in the optimum conditions the detailed description of the experimental setup can be found in [18]. To summarize, it takes account of the beam depolarization at the output of the fiber amplifier (beams S_1 and S_2 in Fig. 7) and also we introduce the self-referencing two wave mixing interferometer which is largely insensitive to any perturbation of the fiber amplifier (temperature, vibrations, ...). The spatial filter generates the diffraction limited reference beam of low intensity.

6. Experimental results

Under these conditions, we obtain after the photorefractive mode converter an amplified R output power of 11.6 W with 110 mW on the input reference beam. Fig. 8 shows output power I_R versus time as it is amplified by S_1 alone, by S_2 alone, and by both S_1 and S_2. The rise time of the nonlinear crystal is 2 to 3 s and the two-wave-mixing efficiency is close to 78%. With the same crystal a shorter response time can be obtained if the output power issued from the fiber amplifier is increased. Indeed, with a photorefractive nonlinearity, the speed scales as the inverse of the incident power density due to the signal beam. Note, however, that other crystals like the $Sn_2P_2S_6$ may exhibit a shorter response time, the millisecond range is possible. This new material is currently under study for improving crystal quality as well for optimizing the photorefractive properties in the near infrared by crystal doping and thermal treatment. Research on this subject are now in progress in several laboratories. The M^2 parameter has been measured for the different beams with the second order moment method and the results are shown in Fig. 9. As expected from the interaction, the amplified reference beam R after nonlinear beam cleanup is nearly diffraction limited with $M^2 = 1.2$ while beams S are multimode with $M^2 = 7.4$. Fig. 10 shows the intensity profiles in the far field.

7. Scalability

The proposed photorefractive beam correcting setup can convert any coherent depolarized multimode beam into a diffraction limited, linearly polarized beam with the spectral characteristics of the beam emitted by the oscillator. The input beam can be highly multimode or aberrated, since the angular acceptance of the crystal is several degrees and its surface can be 1 cm^2 (equivalent to a $M^2 > 1000$). The required coherence length for the multimode signal is several mm or more, corresponding to the interaction length in the nonlinear crystal. Furthermore, the concept is applicable to pulsed operation since the very large core fiber could sustain high peak power.

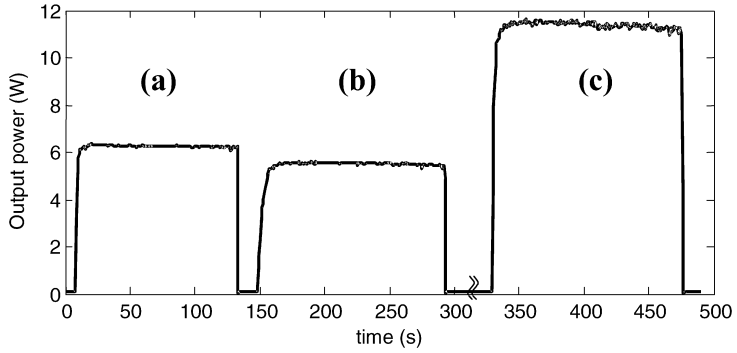

Fig. 8. Evolution of the reference beam power R after two wave mixing in the photorefractive crystal. Intensity transfer (a) with S_1, (b) with S_2 on, and (c) with both S_1 and S_2 on. The rise time is about 2–3 s.

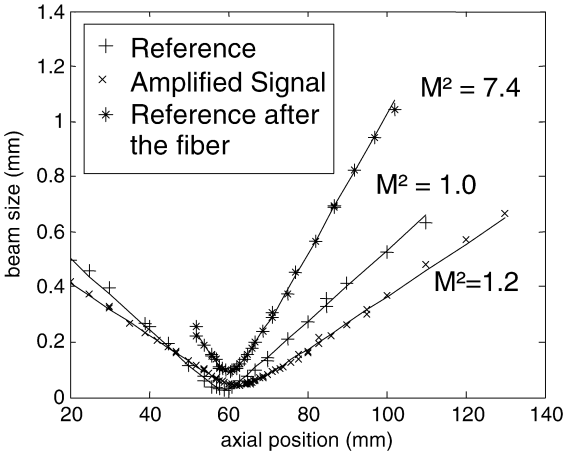

Fig. 9. The beam quality M^2 parameter for the different beams. The experimental points, and the solid lines correspond to fitted curves.

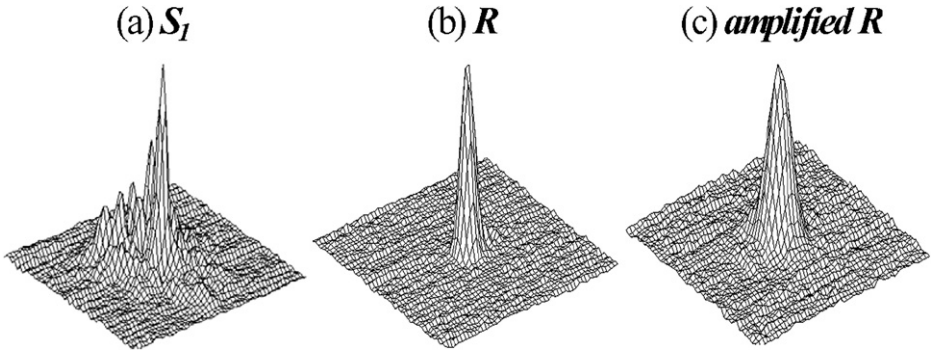

Fig. 10. Beam profiles: (a) signal S_1 at the output of fiber amplifier, (b) single mode 110 mW reference beam R, (c) single mode 11.6 W amplified reference R.

8. Conclusion

The presented original fiber laser architecture involves a nonlinear photorefractive mode converter. It permits beam cleanup by a self-referencing two wave mixing interaction. The emitted beam has the spatial and spectral qualities of the oscillator. The achieved output power with the components used in this experiment is 11.6 W with 78% photorefractive efficiency and the spatial beam quality is close to diffraction limit. The important feature is that the concept can be extended to very large core fiber amplifiers which will support high powers in the CW or pulsed regimes. Rh:$BaTiO_3$ has been used for proof of concept, but alternative photorefractive crystals or nonlinear mechanisms can also be appropriate for reliable operation at the high power levels required in applications. Therefore, it is expected that further progress on material research, will contribute to identify new efficient photorefractive materials in particular for operating in the near infrared. Results issued from these researches will contribute to largely extend the performances of laser sources and to propose innovative concepts for advanced applications of photonics.

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