Accurate method for the measurement of absorption cross sections of solid-state saturable absorbers

1 Centre Interdisciplinaire de Recherche Ions Lasers (CIRIL), UMR 6637 CNRS/CEA/ISMRA, Université de Caen, 6 Blvd Maréchal Juin, 14050 Caen Cedex, France
2 International Laser Center (ILC), 65 F. Scoriny av., Minsk, Belarus
3 Institute of Solid-State and Semiconductor Physics, National Academy of Science, 17 P. Brovki str., Minsk, Belarus
4 LETI-CEA Techniques Avancées, CENG 85X, 38041 Grenoble, France

ABSTRACT A rigorous approach taking into account both the temporal and spatial distributions of the pulsed probe beam used for transmission measurements of solid-state saturable absorbers is applied to characterise these absorbers accurately. The use of this method can significantly influence the resulting value for the ground-state absorption cross section. It also permits us to avoid the introduction of an artificial excited-state absorption in systems where, according to energy-level diagrams, no such loss should occur. This is illustrated in the case of passive Q-switchers for 1.5-µm lasers such as Co2+-doped LaMgAl11O19 and MgAl2O4 and Co2+- or Cr2+-doped ZnSe and ZnS.

PACS 42.55.Rz; 42.60.Jf; 42.60.Gd; 78.20-e

1 Introduction

During the last few years, Q-switched Er:glass lasers operating at around 1.54 µm and characterised by laser pulses in the nanosecond regime with high peak power have been widely investigated both in bulk – see as recent examples [1, 2] – and in microchip [3] laser geometries. Indeed, such a pulsed laser emission around 1.5 µm has been mainly used for many technological applications such as time-of-flight range-finding, target designation, reflectometry in optical fibres, surgery, etc. It comes from the fact that the considered emission wavelength well coincides with the so-called eye-safe spectral region, which is commonly defined as the spectral domain (1.53–1.55 µm) within which the human eye presents the highest damage threshold (the radiation is totally absorbed by the cornea and the aqueous humour before it reaches the retina, thus preventing any irreversible damage). It also corresponds to one of the atmospheric transmission windows (also called the ‘zero water absorption’ window) where the absorption coefficients of the gases composing the atmosphere remain very low, and to the third transmission window of the silica fibres used in optical telecommunication networks, with attenuation coefficients down to 0.2 dB/km in high-quality single-mode fibres.

The simplest way to obtain ns laser pulses with high peak powers in a compact and reliable all-solid-state laser system consists in passively Q-switching the laser by inserting a saturable absorber (SA) inside the resonator. Among all the different materials already used as SAs (organic dyes impregnated into thin sheets of cellulose acetate, colour centres in alkali halides, bulk or multiple-quantum-well semiconductors, etc.) the highest robustness and the best performance were obtained so far with single-crystalline hosts doped with transition-metal or actinide ions. So, to passively Q-switch Er:glass lasers emitting at 1.5 µm, the most promising SAs were the U2+-doped fluorides SrF2 and CaF2 [4], the Co2+-doped garnets YAG and YSGG [5] and, more recently, the Co2+-doped magnetoplumbite LaMgAl11O19 (LMA) [6], the Co2+-doped spinel MgAl2O4 (MALO) [7] and the Co2+ and Cr2+-doped chalcogenides ZnSe and ZnS [2, 8]. However, other materials such as a Co3+-doped glass ceramic [9] or PbS quantum dots in some glasses [10] were also used with success.

Many authors have estimated important parameters such as the ground-state absorption (GSA) and the excited-state absorption (ESA) cross sections σGSA and σESA, respectively, by measuring the non-linear transmission of the materials versus the incident fluence and by applying classical models such as the Frantz–Nodvik model and the modified Avizonis–Grotbeck approach to fit the measured data. Unfortunately, though most of the authors have used the same kind of experimental technique (single-pass saturation measurements) and similar models for data analysis, very different values – especially for ESA – were reported until now. Moreover, in the case of Co2+- or Cr2+-doped single crystals, the energy-level diagrams indicate that no higher-lying energy level can be found above the metastable level of the considered ions, which could lead to an ESA transition around 1.5 µm. Therefore, the weak but non-zero ESA which was reported for the Co2+- and Cr2+-doped crystals by different authors – Yumashev et al. [7], Podlipensky et al. [2] or Tsai and Birnbaum [8] for example – might be simply due to some invalid approximations introduced in their fitting procedure.

Indeed, when using analytical models such as the Frantz–Nodvik model [11] and the modified Avizonis–Grotbeck technique [12], one of the possible error sources is that these...
models assume a long metastable-level lifetime compared to the pulse duration of the probe beam. This assumption of a slow saturable absorber can be invalid for Co-doped oxide crystals, for example, in which the relaxation lifetime was measured in the order of only a few hundred of ns [7]. The second error, as shown theoretically by Rudolph and Weber [13] and by Burshtein et al. [14], can be attributed to the fact that most of the authors do not take into account the spatial distribution of the probe beam used for the bleaching experiments.

In this paper, we present accurate saturation measurements made under similar experimental conditions for a complete series of SAs based on Co²⁺-doped oxide crystals and Co²⁺- and Cr²⁺-doped chalcogenide crystals. Section 2 describes the experimental setup used for single-pass bleaching experiments. Sections 3 and 4 contain a brief review of basic spectroscopic properties of the different SAs investigated. Section 5 presents the different models used to analyse the bleaching curves in order to determine which kinds of approximations are valid for the analysis of such experiments. In particular, it is shown that the spatial distribution is a very important characteristic that must be taken into account explicitly. As results, we report in Sect. 6 new values for GSA and ESA cross sections and may be useful for scientists involved in the modelling of passively Q-switched Er:glass lasers using these Co²⁺- and Cr²⁺-based saturable absorbers. Some other measured parameters such as the lifetime of the metastable level and the SA damage threshold for the different studied materials will also be reported. Finally, the conclusions in Sect. 7 are devoted to some comments on the possible extension of this analysis to others SAs.

2 Experimental procedure

The experimental setup is represented in Fig. 1. It is a very classical scheme in which a single beam from a pulsed laser is focused onto the SA and the transmitted energy is measured versus the incident fluence. It is very similar to a pump–probe technique but, in this case, the same pulse plays both roles in order first to saturate the absorption optical transition and simultaneously to measure the modified transmission. The laser source was based on a homemade flashlamp-pumped and passively Q-switched Er:glass laser. The resonator was a standard plano-concave cavity of 310 mm in length. The highly reflecting mirror (R > 99.5% at 1.534 µm) was a concave mirror with a radius of curvature ROC = 2 m and the output coupler a plane mirror with a transmission T_out = 21%. The Cr–Yb–Er-doped QE-7S phosphate-glass laser rod from Kigre Inc. was a rod of size 3-mm diameter by 49-mm long. The laser was passively Q-switched with the aid of a 0.27-mm-thick plate of Co:LMA crystal oriented at the Brewster angle. The Co:LMA sample was cut so that E || c (π polarisation) and thus with the c crystallographic axis perpendicular to the direction of propagation into the crystal. The Q-switched Er:glass laser source was optimised for producing 1.534-µm light pulses with good pulse-to-pulse energy stability. Moreover, great care was taken during the alignment procedure to favour a single transverse mode with nearly Gaussian spatial intensity distribution. The laser output parameters remained almost constant during all the experiments. The pulse duration (FWHM) was measured to be equal to 75 ns with a fast InGaAs photodiode (rise time < 5 ns); the maximum pulse energy in the TEM₀₀ mode was $E = 11.5$ mJ at 1-Hz repetition rate with typical pulse-to-pulse instabilities of ±2%. To our knowledge, this is one of the best performances obtained so far for a passively Q-switched Er:glass laser emitting in the TEM₀₀ mode. Moreover, the output energy and the temporal pulse shape appear very stable compared to other passively Q-switched laser sources, and therefore the laser could be very easily used as a reference beam to probe the saturation behaviour of different SAs at 1.5 µm.

The Gaussian beam profile was checked first by using an IR vidicon tube from Hamamatsu (model C1000) interfaced with a frame grabber and beam-profiler software from Coherent. Then, the TV camera was placed at different distances from the focus point to measure the $M^2$ propagation beam factor. In all cases, the intensity distribution was found to be close to a purely Gaussian one and the $M^2$ factor was close to unity.

The pulse energy was adjusted thanks to a combination of two Glan–Taylor polarising prisms. Because the laser emission was linearly polarised, a variation of the first Glan–Taylor orientation would have already permitted us to adjust the output energy. The role of the second prism with a fixed orientation was to keep constant the polarisation state when an anisotropic SA such as LMA is studied. Moreover, it is well known that non-linear absorption with a polarised beam can