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Optics Communications 281 (2008) 2526–2530
OPTICS COMMUNICATIONS
www.elsevier.com/locate/optcom

Origin of athermal refractive index changes observed in Yb^{3+} doped YAG and KGW

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Received 28 August 2007; received in revised form 7 November 2007; accepted 14 December 2007

Abstract

Time-resolved excited-state absorption and fluorescence measurements were performed in the Yb^{3+} doped YAG and KGW laser crystals. The results indicate that the athermal refractive index changes observed in previous transient interferometric and diffraction grating experiments can be related to polarisability changes caused by the existence of strong UV charge transfer absorption bands.

PACS: 42.55R; 42.70

Keywords: Ytterbium; Laser material; Refractive index; Charge transfer

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1. Introduction

Relatively strong refractive index changes of purely electronic origin have been reported and analysed recently in the case of the well-known laser material Nd:YAG when it is intensively pumped with diodes or flash lamps [1,2]. It was demonstrated rather unambiguously, both by performing time-resolved excited-state absorption measurements and crystal-field calculations [2] that the $4f^3 \rightarrow 4f^25d$ interconfigurational UV transitions of the Nd^{3+} ions were indeed responsible for polarisability changes which could account for most of the observed athermal refractive index variations in this material. It was noticed however that these $4f^3 \rightarrow 4f^25d$ bands may not be the only cause and that other types of strong absorption bands, such as ligand to metal charge transfer (LMCT) absorption bands, may also contribute. Such a possibility could not be checked in the case of Nd:YAG. Indeed, according to the estimation made in Ref. [2] or the well-known Jorgensen relation [3] $E_{\mathrm{LMCT}}(O^{2-} \rightarrow Nd^{3+}) \approx 30{,}000\ [\chi(O^{2-}) - \chi(Nd^{3+})]$ giving the position of such $O^{2-} \rightarrow Nd^{3+}$ LMCT absorption as a function of the ion electronegativities $\chi(O^{2-}) \approx 3.2$ and $\chi(Nd^{3+}) \approx 1.2$ (see in Appendix and Refs. [4–11]), the LMCT absorption band in Nd:YAG would be located above 60,000 cm^{-1}, hence at shorter wavelengths than 160 nm. Consequently, this band would be obscured by the $4f^25d$ bands which extend from about 250 to 110 nm [2].

We thus turned our attention to the other well-known and important Yb^{3+} laser crystals, Yb:YAG and Yb:KGW in which relatively large athermal refractive index changes were also recently reported [12,13], while the $4f^{13} \rightarrow 4f^{12}5d$ interconfigurational transitions of Yb^{3+} in these materials should be lying at much higher energies than in the case of Nd^{3+} and thus should less contribute to polarisability changes. Indeed, as reported in Appendix, the $4f^{13} \rightarrow 4f^{12}5d$ absorption bands in the case of Yb:YAG should be located above 59,000 cm^{-1} (170 nm) against 44,500 cm^{-1} (225 nm) for the $4f^3 \rightarrow 4f^25d$ bands in the case of Nd:YAG. On the other hand, in the case of the Yb^{3+} ions, with an electronegativity $\chi(Yb^{3+}) = 1.61$ (see in Appendix), and according again to the above mentioned Jorgensen relation, the $O^{2-} \rightarrow Yb^{3+}$ LMCT absorption

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doi:10.1016/j.optcom.2007.12.093

band should occur around 47,600 cm^{-1} (thus 211 nm). In fact, the existence of this band in Yb:YAG has been already evidenced and characterized in the past both via emission and excitation measurements made at low temperature and using synchrotron radiation in the near UV spectral domain [6,7]. According to these data, an absorption/excitation band occurs between about 190 and 230 nm, thus in good agreement with the above estimation, and two emission bands peaking around 340 and 490 nm and associated with optical transitions from this charge transfer state to the two spin-orbit levels $^2F_{7/2}$ and $^2F_{5/2}$ of the Yb^{3+} ions are observed. Namely, this band should clearly contribute, eventually more efficiently than the 4f → 5d bands, to the photo-induced polarisability changes and resulting refractive index variations observed in Yb:YAG.

On the other hand, in the case of Yb:KGW, the host matrix is made of molecular groupments WO_4^{2-} having their own luminescence properties and a third type of contribution originating from these absorption species may also contribute.

Namely, to confirm the above hypothesis in the case of Yb:YAG and to clarify the situation in the case of Yb:KGW, both time-resolved excited-state absorption (ESA) and fluorescence measurements have been performed and analysed.

2. Experimental results and analysis

ESA spectra were registered in the near UV spectral domain, between about 200 and 400 nm, by using a standard experimental technique [2] in which the probe beam was provided by a pulsed Xe flashlamp with 5 μs pulse duration and the pump by a 12 ns Nd:YAG pumped optical parametric oscillator (OPO). The pump wavelength was tuned, in the case of Yb:YAG and Yb:KGW at the $^2F_{7/2} \rightarrow {}^2F_{5/2}$ absorption wavelengths of 940 and 982 nm, respectively. Subsequently, UV fluorescence measurements were performed first by pumping the crystals at these infrared absorption wavelengths, then at the wavelengths corresponding to the positions of the UV absorption bands revealed by the previously evidenced ESA bands.

The ESA spectrum obtained in the case of Yb:YAG is shown in Fig. 1. This spectrum is given for three pump–probe time-delays of 3, 220 and 620 μs. It reveals the existence of only one strong ESA band peaking around 265 nm with an absorption cross-section σ_{ESA} (as derived from an estimated effective excited ion density N_{exc} of about 4.45×10^{18} cm^{-3}) of the order of 1.1×10^{-18} cm^2. Its temporal evolution first confirms that ESA really takes place in the excited metastable level $^2F_{5/2}$ with a fluorescence lifetime of about 1 ms. The energy position (~37,750 cm^{-1}) of the ESA band maximum added to the energy position of the metastable level (~10,250 cm^{-1}) then allow us to conclude (see in Figs. 2a and 3a) to the existence of a ground-state absorption band peaking around 48,000 cm^{-1}, thus 208 nm, in good agreement with the position (190–230 nm) of the LMCT absorption band derived from the low temperature UV excitation data [6,7] mentioned above.

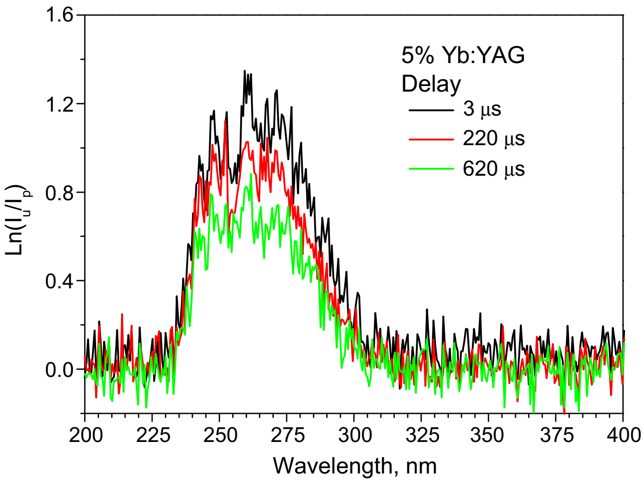

Fig. 1. ESA difference spectrum of Yb:YAG.

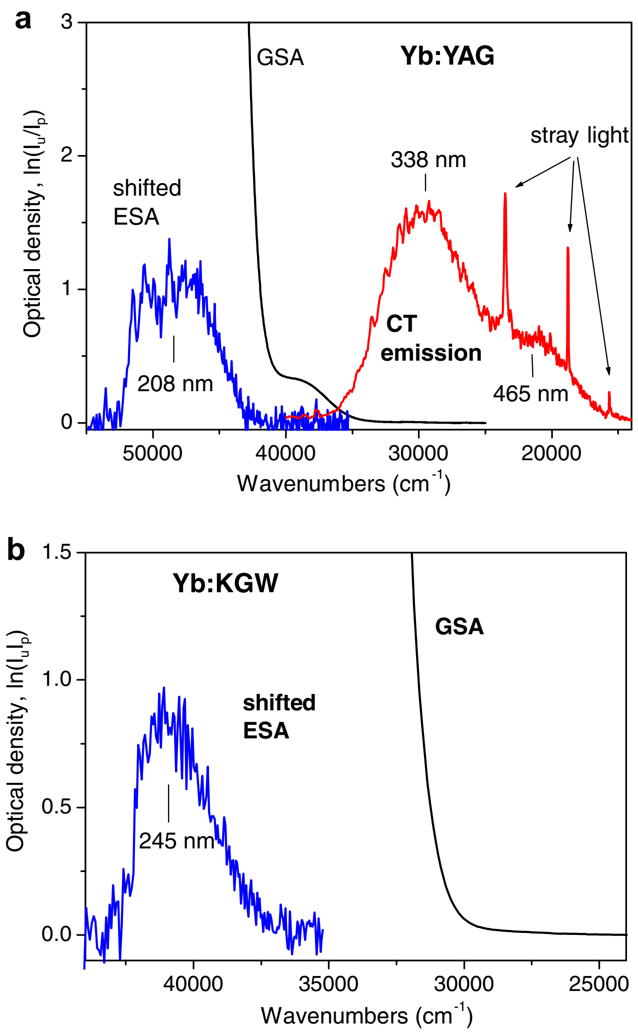

Fig. 2. GSA, fluorescence and ESA spectra (the latter shifted by the $^2F_{5/2}$ metastable level energy) of Yb:YAG (a) and Yb:KGW (b).