Cooperative luminescence and absorption in Ytterbium-doped silica fiber and the fiber nonlinear transmission coefficient at $\lambda = 980$ nm with a regard to the Ytterbium ion-pairs' effect: Reply

Alexander V. Kir'yanov and Yuri O. Barmenkov

Centro de Investigaciones en Optica, Loma del Bosque, 115, Col. Lomas del Campestre, León 37150, Gto, México
kiryanov@cio.mx

Abstract: We reply to the comment [R. Paschotta and A.C. Tropper, Opt. Express, to be published (2006)] on our recent work reporting a study of the cooperative absorption and emission in heavily-doped Ytterbium silica fibers and mechanisms of the fiber nonlinear transmission coefficient reduction due to the Ytterbium ion-pairs' effect [A.V. Kir'yanov et al., Opt. Express, 14 (9), 3981 (2006)]. We provide some additional evidences for that our work hypotheses and conclusions.

©2006 Optical Society of America

OCIS codes: (190.4370) Nonlinear optics, fibers; (190.4180) Multi-photon processes.

References and links

1. R. Paschotta and A.C. Tropper, "Cooperative luminescence and absorption in Ytterbium-doped silica fiber and the fiber nonlinear transmission coefficient at $\lambda = 980$ nm with a regard to the Ytterbium ion-pairs' effect: Comment," Opt. Express **14,** 6981 (2006).
2. A.V. Kir'yanov, Yu.O. Barmenkov, I.L. Martinez, A.S. Kurkov, and E.M. Dianov, "Cooperative luminescence and absorption in Ytterbium-doped silica fiber and the fiber nonlinear transmission coefficient at $\lambda=980$ nm with a regard to the Ytterbium ion-pairs effect," Opt. Express **14,** 3981-3992 (2006).
3. R. Paschotta, J. Nilsson, P.R. Barber, J.E. Caplen, A.C. Tropper, and D.C. Hanna, "Lifetime quenching in Yb-doped fibres," Opt. Commun. **136,** 375-378 (1997).
4. L.A. Diaz-Torres, E. De la Rosa, P. Salas, and H. Desirena, "Enhanced cooperative absorption and up-conversion in Yb^{3+} doped YAG nanophosphors," Opt. Mater. **27,** 1305-1310 (2005).
5. E. De la Rosa, P. Salas, and L.A. Diaz-Torres, "Strong visible cooperative up-conversion emission in $ZrO2$: Yb^{3+} nanocrystals," Journ. Nanoscience and Nonotechn. **5,** 1480-1486 (2005).
6. F. Sanchez, P. Le Boudec, P.L. Francois, and G.Stephan, "Effects of ion pairs on the dynamics of erbium-doped fiber lasers," Phys. Rev. A **48,** 2220-2229 (1993).
7. T. Ishii, "First-principles calculations for the cooperative transitions of Yb^{3+} dimer clusters in $Y_3Al_5O_{12}$ and Y_2O_3 crystals," J. Chem. Phys. **122,** 024705-1-024705-6 (2005).
8. V.Pilla, T. Catunda, H.P. Jenssen, and A. Cassanho, "Fluorescence quantum efficiency measurements in the presence of Auger upconversion by the thermal lens method", Opt. Lett., **28,** 239-241 (2003).
9. J.J. Kopponen, M.J. Soderlund, S.K.T. Tammela, and H. Po, "Photodarkening in ytterbium-doped silica fibers," in *Optically Based Materials and Optically Based Biological and Chemical Sensing for Defence II*, Proc. SPIE **5990,** 72-81 (2005).
10. T. Kitabayashi, M. Ikeda, M. Nakai, T. Sakai, K. Himeno, and K. Ohashi, "Population inversion factor dependence of photodarkening of Yb-doped fibers and its suppression by highly Aluminium doping," in *Techn. Digest (International OFC/NFOEC-2006 conference (5-10 March 2006, Anaheim, Cal., USA)*, Paper # OThC5.
11. H.J. Schugar, E.I. Solomon, W.L. Cleveland, and L. Goodman, "Simultaneous pair electronic transitions in Yb_2O_3," J. Am. Chem. Soc. **97:22,** 6442-6450 (1975).
12. A.V. Kir'yanov and Yu.O. Barmenkov, "Self-Q-switched all-fiber Ytterbium laser," Laser Phys. Lett., to be published (2006); available in www.interscience.wiley.com, **DOI** 10.1002/lapl.200610039.

As the authors of Ref. [1] on our recent work [2] note, there currently is in-question the reason of essentially non-ideal performance of Ytterbium-doped fibers (YFs) and Ytterbium fiber lasers (YFLs) on their base. Specifically, the efficiency of YFL is decreased and the YF

nonlinear transmission coefficient is reduced (that is demonstrated in our research [2]) if the active YF is heavily-doped. In Ref. [2], we assumed that a modification of the dependences of YF nonlinear transmission coefficient on 980-nm pump power and fiber length arises due to the presence in YF of Ytterbium ion-pairs (IPs). In our opinion, the reason for such an assumption is that in fibers heavy-doped with Yb^{3+} are experimentally detected strong "cooperative" visible emission and absorption (about the definition "cooperative" see our comments below). On one hand, notice that our YFs [2] were free from contaminating by any other un-wanted rare-earth dopants [2] or impurities [3]. On the other hand, their features are the mentioned "cooperative" effects. That's why we assumed that the two observations – the YF reduced nonlinear transmission at $\lambda = 980$ nm and YF "cooperative" processes – are interconnected. We have performed a modeling of the YF nonlinear transmission coefficient which incorporates as the excitation quenching mechanism the "cooperative" relaxation stemming from the presence of Ytterbium IPs in YFs. As the authors of Ref. [1] note, the modeling results are in reasonable agreement with the experimental data.

Meanwhile, the authors of Ref. [1] doubt concerning our interpretations. They notice that (i) the "cooperative" emission *"is orders of magnitude too weak"* for being responsible for strong IR power loss in the fibers and (ii) the multi-phonon relaxation, despite *"such a mechanism could exist if Ytterbium ion-pairs would have intermediate electronic levels"*, needs to be proved as the significant at longer (IR) wavelengths. Addressing the first point, notice that a true conclusion of our work [2] is that the extra-losses in YF were regarded by us to the effect of Ytterbium IPs, not to the "cooperative" emission; whereas the "cooperative" processes in the fibers were considered as the indicative features given by the Ytterbium IPs' presence. However, it is indeed the fact that the "cooperative" emission and absorption in our fibers are much more expressed than one might expect. However, these expectations may be misleading in the light of recent experimental results reporting similar facts (see, e.g., Refs. [4,5]). Addressing the second point of criticism [1], notice that the presence of Ytterbium IPs in a material implies the presence of absorption transitions not only in the visible (~ 0.5-μm), but in the IR (~ 1-μm) spectral range, too. The energy level diagram given in Fig. 9 [2] is a rough sketch for clarifying the main processes taken in that modeling. Actually, the effect of doping IPs can be treated in terms of the "quasi-molecular" [6] or "dimer-cluster" [7] models; in both cases the most reasonable energy level diagram for Ytterbium is shown in Fig. 1. In this diagram, the pair's ground state is $11> = (^2F_{7/2}, {}^2F_{7/2})$, the state within the 1-μm region is $12> = (^2F_{7/2}, {}^2F_{5/2})$, and the state within the 0.5-μm region is $22> = (^2F_{5/2}, {}^2F_{5/2})$. Once Ytterbium IPs are present in YF, they are to be involved (as single Ytterbium ions are) in the 1-μm resonant absorption and emission processes (evidently, the 1-μm transitions within Yb^{3+} - Yb^{3+} pairs are masked by the stronger 1-μm transitions within single Yb^{3+} ions). The IR Ytterbium IP band structure seems to be wider and complicated in comparison with those of Ytterbium single ions, because they are a composition of the multiplets of both the Ytterbium ions composing the pair. Thus, multi-phonon processes can be the case in silica YF that stems from a richer energy levels' structure of Ytterbium IPs [7,11] comparing the one of Ytterbium single ions.
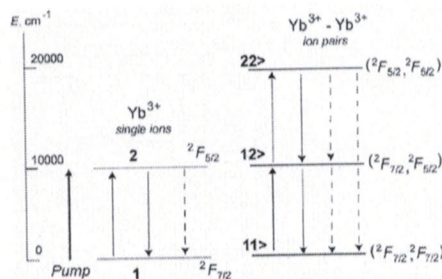

Fig. 1 Energy level diagram of Ytterbium in heavily-doped YF. Schematically are shown the resonant absorption and induced emission transitions (solid arrays) and spontaneous emission transitions (dashed arrays) under 1-μm excitation.

We should add here that excitation quenching, as it is addressed by the last term in Eq.(3) [2], was taken by us into consideration to explain the experimentally registered 1-μm emission decay (see Fig. 11, Ref. [2]). That figure clearly demonstrates the two-exponents' decay, with the longer and shorter ones being correspondingly related to the excitation relaxation of single Ytterbium ions and Ytterbium IPs. In our opinion, the last process inevitably results in a strong modification of YF nonlinear transmission at 980-nm excitation (see Fig. 8, Ref. [2]). However, notice that when deriving Eqs. (2,3) [2], we assumed that the induced transitions between the Yb^{3+} - Yb^{3+} IPs' levels have the same cross-sections as single Yb^{3+} ions have and the pairs' excited-state absorption is neglected. Only in such an approximation the problem of propagation of pump radiation along the fiber can be reduced to a couple of Eqs.(2,3) [2], where the Ytterbium IPs' effect is accounted for by the "cooperative" relaxation term, n_2^2/τ_p (like it is done at treating the Auger up-conversion; see, e.g., Ref. [8]).

The authors of Ref. [1] discuss further on the Ytterbium concentration dependence of the additional decay process (i.e., the process stemming from the IPs' effect), arguing that this dependence *"is not compatible with expectation that the density of Ytterbium ion pairs should grow at least in proportion to the square of the Ytterbium concentration"*, so the quantity $1/\tau_p$ *"should thus rise at least in proportion to the Ytterbium concentration"*. As for us, the last passage looks strange, since in our case, as it is seen from Fig. 10 (b) [2], τ_p decreases (and thus $1/\tau_p$ increases) with Ytterbium concentration (N_0) growth. Meanwhile, the parameter β (as it is introduced in Ref. [2] for addressing the binding stress of two ions composing a pair) is also proportional to N_0 (Fig. 10 (a) [2]). The latter is not surprising: As the evidence, Ref. [8] can be used, where the Auger up-conversion in a doped medium is characterized, as in our work [2], by the parameter $\beta \sim \tau_p N_0$ (see formula (2), Ref. [8]). Therefore, the higher concentration N_0, the stronger is the binding strength of ions in an IP.

At the last, the authors of Ref. [1] refer to another interesting phenomena observed in YFs and YFLs [3,9]. In Ref. [3], it is reported the presence in heavily-doped YFs of significant extra loss resulting in deteriorating of YFL efficiency. As the authors of Ref. [3] (and also of Ref. [1]) recognize, a source of the loss has not been identified; meanwhile, they reveal that this loss has a resonant character (a narrow 976-nm absorption peak located exactly at the maximum of Ytterbium resonant absorption line). To the best of our knowledge, a source of the extra loss in YFs [3] stays unknown to-date. On the other hand, its resonant character and spectral location allows one to hypothetically assume that its origin can be the absorption transitions of Ytterbium IPs ($^2F_{7/2}$, $^2F_{7/2}$) -> ($^2F_{7/2}$, $^2F_{5/2}$) in 1-μm region (see Fig. 1). Otherwise, it looks mysterious that *"unidentified color centers or structural defects"* [3], responsible for this loss, arise in YF exactly at the wavelength of Ytterbium resonant absorption maximum. Let us also comment on the referred work [9], which reports the observation in YFLs of the photo-darkening effect. Notice that the last effect, as it is treated at least by some authors, is closely related to the presence in YF of Ytterbium IPs [10]. The photo-darkening effect has been also detected by us in different types of YFs at their use as an active medium of self-Q-switched YFLs [12].

Finally, we would stress that a re-consideration of the nature of "cooperative" emission and absorption is a necessity of further studies. The deal is that the "cooperative" processes are usually attributed to the transitions between the virtual levels of a pair composed of two isolated doping ions. However, two isolated dopants, even though located at a very short distance each to another, cannot have real double energy levels. So, the "cooperative" ~0.5-μm absorption registered in many Ytterbium-doped materials could not be anything rather than an artifact, and the "cooperative" emission power should be a few orders weaker than it is frequently measured. The resolution of this general disagreement seems to be the hypothesis of a "third body", which assumes the existence in a heavily-doped material of true molecules ("dimer clusters") composed of two non-isolated, but bind by strong covalent forces, rare-earth ions with the third "body", which is an ion or groups of ions of the lattice. In the case of Ytterbium-doped materials, including YFs, these are meant to be two adjacent ions of Ytterbium and ions of Oxygen, respectively. The theories [7,11] undertaking such a model seems to be productive for reasonable explanation of the experimental results [2-5].