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Dedicated to Academician Professor Dr. Emil Burzo on His 80th Anniversary

CONCENTRATION AND PUMP INTENSITY EFFECTS IN THE EMISSION OF Nd LASER MATERIALS

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ABSTRACT. High doping concentrations useful for granting efficient absorption of the pump radiation can be hampered by the concentration-dependent self-quenching of emission due to energy transfer inside the system of doping ions. The paper discusses the pump intensity-dependent balance between the down- and upconversion processes in the Nd laser materials, based on their effect on emission dynamics. Reduction of emission quantum efficiency by self-quenching and its influence on the laser emission and heat generation is analysed.

Keywords**:** *Nd lasers, energy transfer, down-conversion, upconversion, heat generation*

INTRODUCTION

The non-radiative de-excitation of the emitting level in the pumped laser materials is major factor in limitation of the performances of the solid-state lasers. Basically there are two major mechanisms, the electron-phonon (EP) interaction that transforms the excitation into heat and the non-radiative energy transfer ET to another ion. The final levels of the donor D ion and of the acceptor A ion involved in this process could de-excite further by EP interaction and then the joint action of ET and EP interaction contributes to heat generation. The non-radiative processes accelerate the dynamics of emission of the excited level and the competition of radiative de-excitation with the non-radiative processes, is characterized by the emission quantum efficiency η_{qe}. The efficiency of the ET processes depends on their nature, which defines a microparameter of transfer and on the concentration of acceptors.

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Of major relevance for the laser ions are the ET processes inside the system of doping ions since these could determine the selfquenching of emission. In case of Nd^{3+} the self-quenching could involve both down-conversion (DC) cross-relaxation ($^4F_{3/2}$, $^4I_{9/2}$) → ($^4I_{15/2}$, $^4I_{15/2}$) and several upconversion (UPC) processes, ($^4F_{3/2}$, $^4I_{9/2}$) → ($^4I_{13/2}$, $^4G_{7/2}$) or/and ($^4I_{11/2}$, $^4G_{9/2}$) or/and ($^4I_{9/2}$, $^2P_{1/2}$). In many Nd laser materials, including $Y_3Al_5O_{12}$ (YAG) the richer variety of processes and the better matching of D emission with the A absorption could determine larger ET microparameter for UPC than that for DC. The final D and A state $^4I_{15/2}$ for down-conversion is de-excited rapidly by fast EP relaxation along the ladder of ^4I_J levels to the ground level $^4I_9$ and thus the whole excitation involved in process is finally transformed into heat. However, in case of UPC the EP interaction relaxes the final state of the donor to the ground state and that of the acceptor back to the emitting level, and thus only one of the two excitations involved in ET is transformed into heat; physically UPC+EPI transforms the pair of two interacting excited Nd ions into a pair (excited ion)-(non-excited ion) that could be suitable for subsequent DC. A special case of ET involves the complete transfer of excitation from an excited to a unexcited ion which is thus promoted to the initial excited state of the donor (excitation-conserving D-D ET). If this process repeats it can migrate the excitation to large distances from the originally-excited donor, till is lost either by emission or by transfer in an energy converting DC or UPC process (migration-assisted ET). Both DC and UPC could involve direct (static) D-A ET as well as migration-assisted transfer. The relative contributions of DC and UPC at any moment of decay will depend not only on the specific ET microparameters but also on the relative instantaneous populations of the acceptors for each of these processes, i.e. excited ions for direct UPC and non-excited ions for direct DC and for the energy migration and thus it will depend both on the initial density of excited ions and on its temporal evolution. Reduction of emission quantum efficiency by self-quenching could alter the laser parameters, whereas the various effects of heat generation could contribute to reduction of the laser beam quality and of the abilities of power scaling. Most early experiments on the emission decay of Nd^{3+} in YAG used weak excitation, which determines low initial density of excited ions and thus the effect of ET on decay could be described quite accurately assuming only DC processes [1,2], with constant concentrations of acceptors during decay. However, with stronger excitation the presence of UPC became evident [3-5] and many subsequent studies invoked UPC as the only ET effect, without considering the time-dependent balance between UPC and DC. Moreover, the UPC process was generally discussed in terms of a constant rate and without any account of existence of direct and migration-assisted processes and of distribution of the Nd ions in the crystalline lattice; moreover, the ET parameters reported by various authors differ in large limits.

The contribution of the ET to heat generation is determined by the competition with other de-excitation processes, i.e. it will be different for the ions participating or non-participating to lasing: whereas for the first of these the stimulated emission dominates all other de-excitation and the global heat generation is determined solely by the laser quantum defect, with no non-radiative de-excitation of the emitting level, for the second category additional heat generation from this level results from the competition of the luminescence with non-radiative ET de-excitation, characterized by the emission quantum efficiency η_{qe}. As consequence, the heat generation parameter η_h, which expresses the fraction of absorbed power transformed into heat, will be different for these two categories of de-excitation, $\eta_h^{(l)}$ and respectively $\eta_h^{(f)}$, and in presence of lasing the contribution of these two categories of ions, delineated by the laser emission efficiency η_l, to the intensity and distribution of heat generation must be considered.

This paper addresses some of the problems described above by considering the joint action of DC and UPC processes on the emission decay and on quantum efficiency for various relative strengths of these processes and different fractions of Nd ions excited by pump. Additionally, the effects of the spatial distribution of the pump and laser mode on the generated heat field are discussed.

THE EFFECT OF ENERGY TRANSFER ON EMISSION DECAY

General account

The non-radiative energy transfer is determined by the static interactions between the D and A ions [6] and it leads to de-excitation of donor with rates with specific dependence on D-A distance: $W_{DA}^{(s)} = C_{DA}^{(s)} / R_{DA}^6$ with *s*=6, 8 or 10 for electric dipole-dipole (d-d), dipole-quadrupole (d-q) and quadrupole-quadrupole (q-q) interactions, whereas the strong short-range superexchange coupling leads to very fast rates. The ET microparameters $C_{DA}^{(s)}$ reflect the electronic structure of the ions and although this process is non-radiative, they depend on the superposition integral of donor emission and acceptor absorption in the spectral range of transfer, $S_{DA} = \int \sigma_e^D(\nu)\sigma_a^A(\nu)d\nu$. The multipolarities of the electric ET interactions are thus determined by the selection rules for the transitions involved in the donor and acceptor act. The electric and superexchange interactions are cumulative and $W_{DA} = \sum_s (W_{DA}^{(s)}) + W_{DA}^{ex}$.

The D-A ET competes with other de-excitation processes and the transfer to a unique acceptor ion preserves the exponential decay but it reduces the lifetime τ_D of the donor to $\left(\tau_D^{-1} + W_{DA}\right)^{-1}$; however, in solids each donor is surrounded by a particular configuration of acceptors at distances R_j and its lifetime becomes $\left(\tau_D^{-1} + \sum_j W_j\right)^{-1}$. The emission decay of the whole ensemble of donors would assume the impossible task of summing the individual contributions, but this could be replaced by proper averaging procedures based on the distribution of the doping ions in the laser material. The averaging procedure will then enable definition of an ET function $P(t)$ as the survival probability of the donor in its excited state in presence of transfer is $\exp[-P(t)]$. With this transfer function the emission decay becomes

$$I(t) = I(0)\exp(-t/\tau_D)\exp[-P(t)] \tag{1}$$

and a similar equation can be written for the evolution of the donor population in excited state n_D, which can be regarded as solution of population evolution equation

$$\frac{dn_D}{dt} = -\frac{1}{\tau_D}n_D - \frac{dP(t)}{dt}n_D . \tag{2}$$

The form of transfer function depends on the model of distribution of the ions in lattice used in the averaging procedure. In the almost general situation of absence of attractive or repulsive forces between the doping ions the most popular models of distribution are:

-*the average distance model* assumes that the doping ions are placed evenly in crystal at distances $\sim(n_{Nd})^{-3}$. In this model the configurations of acceptor ions around any donor ion and thus all ET rates are identical and the emission decay remains exponential, but with Nd concentration dependence [7]: for d-d coupling $W_{DA} \propto (n_{Nd})^2$. This model is very simple but not realistic and does not account for the structure of the crystal.

-*the uniform continuous model* assumes that the density of acceptors around donor is the same in each geometrical point of space [6,8,9]. The transfer function is non-linear in time, $P(t) = \gamma^{(s)}t^{3/s}$, and $\gamma^{(s)}$ depends on C_{DA}: for d-d coupling $P(t) = \gamma^{d-d}t^{1/2}$, with $\gamma^{(d-d)} = (4/3)\pi^{3/2}n_{Nd}(C_{DA})^{1/2}$. The decay predicted by this model is not exponential and the fit with experimental data enables

identification of the multipolarity *s* of ion-ion interaction and evaluation of C_{DA}. The model is restricted to unique type of interaction, it does not account for the structure of the crystal and predicts finite density of acceptors at the donor position;

-*the discrete random distribution* assumes discrete occupation by acceptors of any available lattice site *i* with same probability [10], equal to relative acceptor concentration C_A,

$$P(t) = \sum_i \ln\left[1 - C_A + C_A \exp(-W_i t)\right] \tag{3}$$

This model is the most realistic since it accounts for the crystal structure (the positions *i*), and the rates W_i can include any type of interaction; it predicts complex decay, with departures from exponential. In case of Nd^{3+} the microparameter C_{DA} for the direct ET is larger than that of the D-D energy-conserving transfer C_{DD}, and the migration of excitation inside the system of Nd^{3+} ions takes place by the hopping mechanism [11]. In this case the migration-assisted self-quenching can be characterized by an ensemble-averaged transfer function linear in time $P_{ma} = \overline{W}t$, with the rate depending on the acceptor concentrations specific to both of these successive processes, $\overline{W} = \overline{W}_0 C_D C_A$ and with the microparameter $\overline{W}_0$ dependent on both C_{DA} and C_{DD}. In presence of the direct and of migration-assisted transfer processes

$$I(t) = I(0)\exp(-t/\tau_D)\exp\left[-P(t)\right]\exp(-\overline{W}t) = I(0)\exp(-t/\tau_D)\exp\left[-P'(t)\right]. \tag{4}$$

Although the direct and migration-assisted ET act together over the entire decay, due to the different dependences on time the migration-assisted transfer is manifested as a new quasi-linear dependence of the global transfer function $P'(t) = P(t) + \overline{W}t$ at the end of decay.

The ET processes could involve a unique or several types of acceptors and in this case the transfer function $P(t)$ can be written as sum of the transfer functions to each acceptor species. Such situation holds for the general case of ET self-quenching of Nd laser materials, due to the co-existence of the two types of processes, down-conversion and upconversion, but it is a special case, since the concentrations of acceptors for these processes are correlated and dependent of time [12-14]. Thus, by denoting the fraction of Nd excited ions by $r(t) = r(0)\exp(-t/\tau_D)\exp\left[-P'(t)\right]$, the relative instantaneous concentrations of acceptors is $\left[1 - r(t)\right]C_{Nd}$ for down-conversion and $r(t)C_{Nd}$ for upconversion.

The effect of down-conversion self-quenching on decay

For very small $r(0)$ fractions the down-conversion dominates the self-quenching and the influence of upconversion can be neglected to a quite good approximation; in fact, most of measurements of decay of Nd^{3+} fulfill this condition. For this case C_A in Eq. (3) could be considered constant during decay and equal to the relative Nd concentration C_{Nd}. Function (3) is difficult to use directly; however, over definite intervals of decay it can be approximated by more simple functions [1, 2]: at early times it could show linear dependence on time, $P(t) \approx \sum_i C_A W_i t = W_{lin} t$, whereas at longer times it could be approximated by the function corresponding to the uniform continuous distribution. When a very strong short-range interaction, such as superexchange is active in the transfer to the nearest acceptors a fast drop of the emission takes place at beginning of decay, and is followed by the linear approximation with the positions influenced by the strong interaction excluded from summ, the passage between these stages being gradual. This is precisely what it happens in the high resolution decay of Nd:YAG under weak excitation [15,16] where the almost sudden initial drop ~$4C_{Nd}$ of $P(t)$ evidences strong coupling between the Nd^{3+} ions inside the n.n. pair with transfer rate larger than 10^6 s^{-1}. The subsequent portion quasi-linear in time corresponds to the linear approximation of $P(t)$, and this is followed by a portion with $t^{1/2}$ dependence corresponding to d-d coupling with $C_{DA} \approx 1.85 \times 10^{-40} cm^6 s^{-1}$ [2]. Calculation of rate W_{lin} with this microprameter gave satisfactory fit to experiment only by excluding the first four sites from summation, in accord with the dominance of the faster superexchange coupling inside the n.n. pair. Such behavior was observed for many other Nd laser materials, but it requires high temporal resolution of detection, tens of ns, and pulse excitation in the ten ns range. In case of down-conversion migration-assisted ET both the concentrations of ions able to fill the role of donor or acceptor are equal to the Nd concentration and thus the averaged transfer function becomes $\overline{W} = \overline{W}_0 C_{Nd}^2$: in Nd:YAG $\overline{W}_0 = 240$ s^{-1}(%Nd)$^{-2}$.

The effect of upconversion on the emission decay

The upconversion was usually considered as unique source of self-quenching of Nd^{3+} at strong excitation and the effect on decay was described by the population evolution equation

$$\frac{dn_D}{dt} = -\frac{t}{\tau_D} n_D - W^{(up)} n_D^2 \tag{5}$$

This equation is convenient since it has analytical solution and generally it gives acceptable formal description of the observed decay. However, the physical meaning of this approach is highly questionable since the upconversion is described by a constant rate W^{up}, which does not account for the nature of process (static, migration-assisted), for type of ion-ion interaction or for the real distributions of acceptors in crystal, and the down-conversion is either disregarded or included as a constant contribution in the lifetime τ_D. The upconversion transfer rates reported by various authors for Nd:YAG show large spread: for 1 at.% Nd the rate inferred from emission decay of a crystal was (2.8±1)×10^{-16} cm^3s^{-1} [3], whereas in case of thin films this was about 5 times smaller [4], and this last value was confirmed by the subsequent measurements on crystals [5]. The upconversion rate W^{up} inferred from these studies is much higher than that for down-conversion: this is not surprising, since the upconversion includes four processes and the overlap integral *S* could be much larger.

As discussed above, the upconversion cannot exclude the simultaneous presence of down-conversion and in the model of random distribution of the doping ion the ET function $P(t) = P^{dw}(t) + P^{up}(t)$, with

$$P^{dw}(t) = \sum_i \ln\left\{1 - [1 - r(t)]C_{Nd} + [1 - r(t)]C_{Nd} \exp(-W_i^{dw} t\right\} \text{ and}$$
$$P^{up}(t) = \sum_i \ln\left[1 - r(t)C_{Nd} + r(t)C_{Nd} \exp(-W_i^{up} t)\right] \text{ [13,14],}$$

whereas for the migration-assisted ET $\overline{W} = \overline{W}^{dw} + \overline{W}^{up}$,

with $\overline{W}^{dw} = \overline{W}_0^{dw}[1 - r(t)]^2 C_{Nd}^2$ and $\overline{W}^{up} = \overline{W}_0^{up} r(t)[1 - r(t)]C_{Nd}^2$

The ET microparameters for down-conversion can be measured independently at low excitation, whereas those for upconversion could be inferred from the fit at higher excitation by using these down-conversion parameters and usually a general ratio R of the microparameters for upconversion to those for down-conversion could be considered. Under these assumptions, it was found [12] that the decays for 1.7 at.% Nd-YAG thin films [4] can be described by assuming R in the range 20-30.

THE EFFECT OF SELF-QUENCHING ON THE EMISSION QUANTUM EFFICIENCY

The emission quantum efficiency η_{qe} expresses the fraction of the excited ions fed into the emitting level that de-excite by radiative processes. It can be measured directly by the total amount of radiated energy or indirectly, by the thermal effects of the heat generated by the non-radiative processes, such as the rise of temperature, photoacoustic spectrometry, calorimetric interferometry, laser thermal depolarization, thermal lensing, thermal line broadening, and so on. These measurements give quite large spread of data and thus a valuable test of validity could be the η_{qe} calculated from the emission decay $I(t)$,

$$\eta_{qe} = \frac{1}{\tau_{rad}} \int_0^\infty \frac{I(t)}{I(0)} dt \tag{6}$$

This equation requires knowledge of τ_{rad} and accurate description of decay. For exponential decay with lifetime τ_f, $\eta_{qe} = \tau_f / \tau_{rad}$. The calculated η_{qe} enables definition of an effective lifetime of the non-exponential decay as $\tau_{eff} = \eta_{qe} \tau_{rad}$. Evaluation of η_{qe} for complex decays would require numerical calculation. However, in certain cases η_{qe} can be expressed by closed formula, dependent on the distribution model. Thus, in case of down-conversion ET,

-in the average-distance model $\eta_{qe} = \left[1 + (C_{Nd}/C_0)^2\right]^{-1}$, where C_0 is the Nd concentration that reduces the lifetime to $0.5\tau_D$ [8]; despite of the lack of reality of this model, this equation is largely used to predict the Nd concentration dependence of η_{qe};

-for the uniform continuous distribution $\eta_{qe} = 1 - \pi^{1/2} x \exp(x^2)\left[1 - erf(x)\right]$, where $x = \; x = (1/2)\pi^{1/2} C_{Nd} / C_0$ [9].;

-in the random discrete model for direct ET $\eta_{qe} \approx \exp(-bC_{Nd})$ with

$b = \sum_i \frac{W_i}{\tau_D^{-1} + W_i}$ [14].; in presence of migration-assisted ET

$$\eta_{qe} = \frac{1}{1 + \tau_D \overline{W}_0 C_{Nd}^2} \exp\left(- \sum_i \frac{W_i}{\tau_D^{-1} + W_i + \overline{W}_0 C_{Nd}^2} C_{Nd} \right)$$ [13,16]

and the calculated η_{qe} for Nd:YAG crystals or ceramics (Figure 1) gives very good description of experimental values: for 1 at.% Nd it predicts $\eta_{qe} \approx 0.80$ and $\tau_{eff} = 208$ μs.

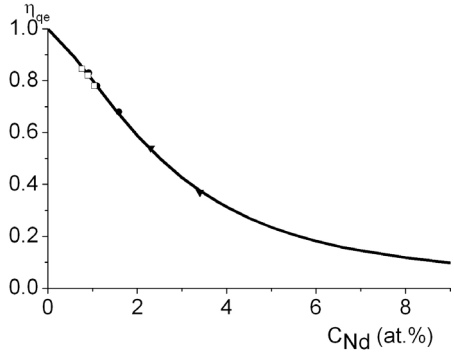

Figure 1. Emission quantum efficiency of Nd:YAG at weak excitation

Evaluation of η_{qe} in presence of both down-conversion (dc) and upconversion (uc) requires numerical calculation. For the low C_{Nd} used in the Nd lasers this corresponds well to the product of efficiencies of each of these conversion processes calculated with the transfer functions $P^{dw}(t)$ and respectively $P^{up}(t)$, i.e. $\eta_{qe}^{(dw+up)} \approx \eta_{qe}^{(dw)}\eta_{qe}^{(up)}$. The numerical calculation in case of 0.6, 1 and 1.5 at.%Nd in YAG under 809 nm pumping for different ratios R between the upconversion and down-conversion ET parameters (1 to 50) and for different initial fractions of excited ions $r(0)$ (0 to 1) reveals that starting from the C_{Nd}-dependent values for weak excitation (0.875, 0.800 and 0.680 for 0.6, 1 and 1.5 at.%Nd) at $r(0)=0$, η_{qe}^{dw} increases quasi-linearly with $r(0)$, with C_{Nd}-dependent slope, whereas η_{qe}^{up} starts from the value 1 for all C_{Nd} and decreases non-linearly with $r(0)$, more accentuated at higher C_{Nd} and R ratios. Thus, η_{qe}^{up} could be larger than η_{qe}^{dw} even for large R ratios, up to quite large $r(0)$ fractions, and the range extends for higher C_{Nd}; however, $\eta_{qe}^{(dw+up)}$ is always smaller than either η_{qe}^{dw} or η_{qe}^{up} and this situation is illustrated in Fig. 2 for R =30.

In a CW laser the threshold $P_{th} \propto \left(\eta_a \eta_{qe} \eta_{qd}^{(l)}\right)^{-1}$ and the slope efficiency $\eta_{sl} \propto \eta_a \eta_{qd}^{(l)}$, where η_a is the pump absorption efficiency and the Stokes ratio between the pump and laser wavelengths characterizes the laser quantum defect, $\eta_{qd}^{(l)} = \lambda_p / \lambda_l$. Reduction of η_{qe} at high C_{Nd} increases P_{th} but it has no effect on η_{sl};

moreover this could be compensated over a certain range of C_{Nd} by the enhancement of η_a, which could also enhance η_{sl}. Of major importance could be reduction of laser quantum defect: for the Nd lasers, where traditionally the diode laser pumping is made around 808 nm in the strongly absorbing level $^4F_{5/2}$ this can be made by direct ~880 nm pumping into the emitting level $^4F_{3/2}$ [17,18]: the reduction of 9-10% of the quantum defect will determine similar improvement of the laser parameters P_{th} and η_{sl}. Unfortunately, for most Nd materials the absorption cross-section for direct pumping could be considerably smaller than for traditional 800 nm pumping and this could impose special care to improve the absorption efficiency, such as higher C_{Nd} or longer pump radiation path inside the laser material using large active components or multipass pumping.

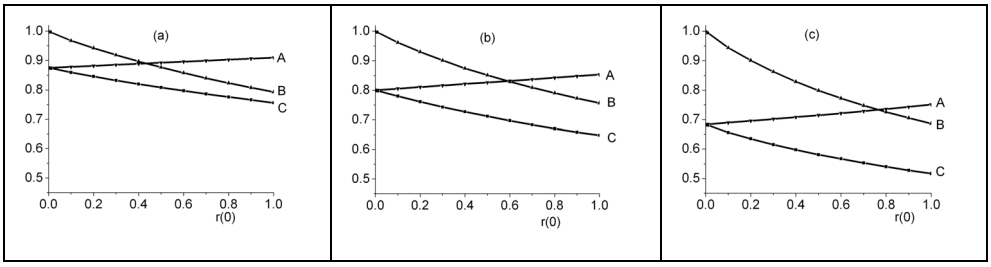

Figure 2. Calculated emission quantum efficiencies function on initial fraction of excited ions r(0) for Nd concentrations 0.6 at% (a), 1 at.% (b) and 1.5 at.% (c) and *R*=30: A - dc, B – uc, C – dc+uc

THE EFFECT OF SELF-QUENCHING ON HEAT GENERATION

By the involvement of EP interaction the self-quenching could contribute jointly with the quantum defect to heat generation in the pumped laser materials. The heat generation can be characterized by the heat load coefficient η_h that expresses the fraction of the absorbed power transformed into heat by non-radiative de-excitation: in CW emission processes this parameter is constant. In absence of laser emission both the emission quantum defect and the self-quenching of the emitting level are active and $\eta_h^{(f)} = 1 - \eta_{qe}\eta_{qd}^{(f)}$.

Under weak excitation the down-conversion makes η_{qe} dependent only on C_{Nd} and thus $\eta_h^{(f)}$ depends only on C_{Nd} and on λ_p. However, under strong excitation the joint action of down- and upconversion makes η_{qe} and $\eta_h^{(f)}$ dependent also on the fraction $r(0)$ and on ratio *R*. Calculation of $\eta_h^{(f)}$ for different C_{Nd} s in YAG using the calculated η_{qe} in presence of only down-conversion $\eta_h^{(f,dw)}$, only upconversion $\eta_h^{(f,up)}$ and of both down- and upconversion $\eta_h^{(f,dw+up)}$ shows that $\eta_h^{(f,dw)}$ starts from the C_{Nd}-dependent weak excitation values (0.318 for 0.6, 0.377 for 1 and 0.470 for 1.5 at.% Nd) and decrease quasi-linearly with *r*(0), whereas $\eta_h^{(f,up)}$ starts from the C_{Nd}-independent quantum-defect limited value for luminescence (~0.22 in case of 809 nm excitation) and increases with $r(0)$, faster for large C_{Nd}. However, for definite $r(0)$ ranges $\eta_h^{(f,up)}$ is smaller than $\eta_h^{(f,dw)}$ and well under the values measured for $\eta_h^{(f)}$ below threshold for the Nd:YAG lasers. Thus, $\eta_h^{(f)}$ around 0.38 was currently measured for 1 at.% Nd:YAG under weak excitation in absence of laser emission, in agreement with the calculated value of $\eta_h^{(f,dw)}$, but much above $\eta_h^{(f,up)}$. At the same time, the calculated $\eta_h^{(f,dw+up)}$ starts from the value corresponding to $\eta_h^{(f,dw)}$ but increases with $r(0)$ and it is always larger than both $\eta_h^{(f,dw)}$ and $\eta_h^{(f,up)}$. This is illustrated in Figure 2 with the calculated values of $\eta_h^{(f)}$ for 0.6, 1 and 1.5 at.% Nd:YAG with R =30 under 809 nm excitation, using the η_{qe} values presented in Fig. 2. These data show that upconversion alone cannot explain the heat generation and the assistance of down-conversion should be always accounted for.

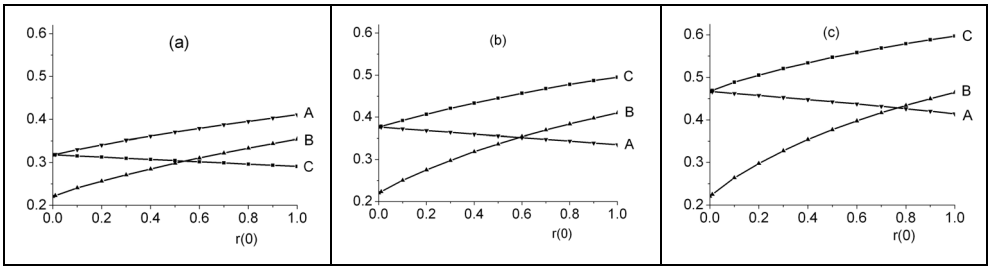

Figure 3. The calculated heat load parameter for 0.6 at.% Nd, (a), 1 at.% Nd (b) and 1.5 at.% Nd (c) in YAG for R=30: A – dc, B – uc, C – dc+uc.

In a pumped laser material the modal structure of the laser resonator, which determines the spatial configuration of excited ions that can be de-excited by stimulated emission could differ from the configuration of excited ions, determined by that of absorbed pump radiation; the relation between these two spatial configurations is characterized by the volume superposition integral η_v. Generally, it is considered that η_v =1 when the laser mode volume is larger than the absorbed pump volume and <1 in the opposite situation. Additionally, the laser process needs a threshold population which is reached for a given absorbed power P_{th}. As consequence not all excited ions participate to lasing and these can de-excite by luminescence and competing non-radiative de-excitation; these two classes of excited ions are delineated by laser emission efficiency $\eta_l = \eta_v\left[1-(P_{th}/P)\right]$. The stimulated emission is faster than any other de-excitation process and thus the only contribution of the lasing ions to the heat generation comes from the laser quantum defect and their heat load coefficient is $\eta_h^{(l)} = 1-\eta_{qd}^{(l)}$. The relation between the heat generated by the lasing ions and those that do not lase is governed by relation between $\eta_{qd}^{(l)}$ and $\eta_{qe}\eta_{qd}^{(f)}$ and for a given laser emission scheme in a given material it can be controlled by the doping concentration. The global contribution to heating of the excited ions is then

$$\eta_h = \eta_l\eta_h^{(l)} + (1-\eta_l)\eta_h^{(f)} = 1-\eta_l\eta_{qd}^{(l)} - (1-\eta_l)\eta_{qe}\eta_{qd}^{(f)} \tag{7}$$

Reduction of the quantum defect by direct pumping around 880 nm could reduce the heat generation for the one-micron CW Nd lasers by ~30% compared with 808 nm pumping and this could increase the power scaling ability by more than 55% [19].

SPATIAL DISTRIBUTION OF LASER, HEAT GENERATION AND LUMINESCENCE

The spatial distribution and not only the global value of heat generation could be of major relevance. Most papers consider the configuration of heat generation similar to that of the absorbed power. Based on differences in heat generation between the lasing and non-lasing ions it can be shown that the laser mode-pump volume superposition plays a major role and can distort such picture. Figure 4 shows the calculated radial distribution of heat and luminescence for 1 at.% Nd:YAG under uniform or Gauss 809 nm pumping for Gaussian one-micron laser emission with waist 60% from the absorbed pump waist and for P/P_{th} =10.

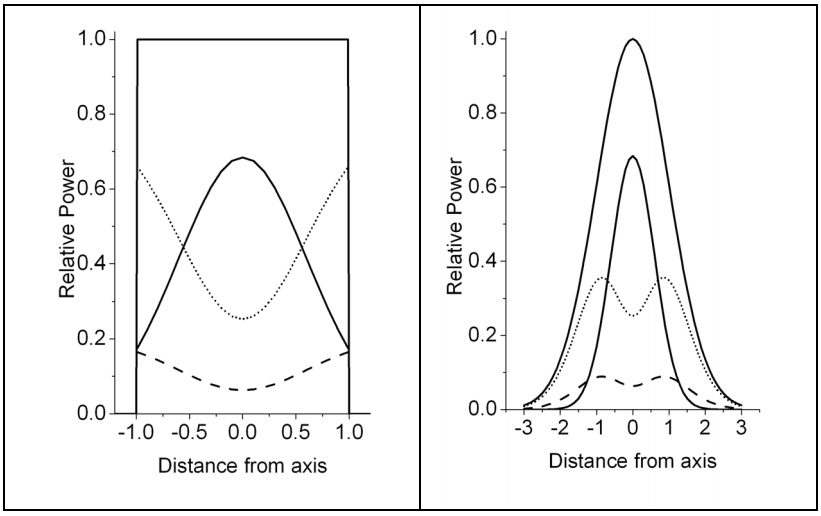

Figure 4. The calculated radial distribution of one-micron laser (full lines), luminescence (dashed lines) and heat generation for 1 at.% Nd:YAG under uniform (left side) or Gauss (right side) 809 nm pumping (external contours)

These distributions are calculated assuming weak pumping, with neglect of upconversion and show that even in these conditions, the excited ions outside the laser mode distort the distribution of heat generation; upconversion will accentuate these effects.

CONCLUSION

Concentration self-quenching in the Nd laser materials is a complex process which influences the dynamics of emission and reduces the emission quantum efficiency, leading to enhancement of laser threshold and contributing to generation of heat. Limitation of these effects requires detailed knowledge of the self-quenching mechanism: this paper shows that in case of Nd the down- and upconversion self-quenching act together in a correlated manner and, whereas down-conversion dominates at weak excitation, upconversion at higher excitation cannot be considered alone. The differences in the heat generation of the lasing and non-lasing ions could influence the thermal field inside the pumped laser material.

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REFERENCES

1. A.G. Avvanesov, B.I. Denker, V.V. Osiko et al., *Sov. J. Quantum Electron*. 12, 744 (1982)
2. V. Lupei, A. Lupei, S. Georgescu, C. Ionescu, *Opt. Commun*. 60, 59 (1986)
3. Y. Guyot, H. Manaa, J. Y. Rivoire, R. Moncorge et al, *Phys. Rev.* B51, 784 (1995)
4. S. Guy, C. L. Bonner, D. P. Shepherd, D. C. Hanna et al, *IEEE J. Quant. Electron*. 34, 900 (1998)
5. M. Pollnau, P. J. Hardman, M. Kern, W. A. Clarkson, D. C. Hanna, *Phys. Rev.* B58, 16076 (1998) 1
6. D. L. Dexter, *J. Chem. Phys*. 21, 836 (1953)
7. H. G. Danielmeyer, M. Blatte, P. Palmer, *Appl. Phys.* 1, 269 (1973)
8. Th. Forster, *Ann. Phys.* (Leipzig) 2, 55 (1948)
9. M. Inokuti, F. Hirayama, *J. Chem. Phys*. 43, 1978 (1965)
10. S. I. Golubov, Yu. V. Konobeev, *Sov. Phys. Sol. State* 13, 2679 (1972)
11. A. I. Burshtein, *Sov*. *Phys. Uspekhi* 27, 579 (1984)
12. V. Lupei, B. Diaconescu, A. Lupei, *Proc. SPIE* 4430, 88 (2001)
13. V. Lupei, *Opt. Mat.* 19, 95 (2002)
14. A. Ikesue, N. L, Aung, V. Lupei, Ceramic Lasers, Cambridge Univ. Press 2013
15. V. Lupei, A. Lupei, *Phys. Rev*. B61, 8087 (2000)
16. V. Lupei, A. Lupei, S. Georgescu, T. Taira, Y. Sato, A. Ikesue, *Phys. Rev*. B64, 092102 (2001)
17. R. Lavi, S. Jackel, *Appl. Opt*. 39, 3093 (2000)
18. V. Lupei, A. Lupei, N. Pavel, T. Taira, I. Shoji, A. Ikesue, *Appl. Phys. Lett*. 79, 590 (2001)
19. V. Lupei, *Opt. Mat*. 31, 701 (2009)