Low-Quantum-Defect Solid State Lasers: 2-, 3-, or 4-level?

by Jeffrey O. White

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**14. ABSTRACT**
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Low-Quantum-Defect Solid State Lasers: 2-, 3-, or 4-level?

Jeffrey O. White

The concept of three-level- and four-level-lasers was used early on to explain why it was easier to make the Nd$^{3+}$ ion lase than the Cr$^{2+}$ ion. Almost 50 years later, work on solid-state lasers has recently led to a quantitative definition of the concept. The new level parameters $\ell_0$ and $\ell_1$ defined below will provide an improved basis for comparing Nd$^{3+}$, Cr$^{2+}$, and other ions, e.g. Er$^{3+}$, Yb$^{3+}$, Ho$^{3+}$, and Tm$^{3+}$. Together with the cross section, lifetime, etc., they help in the design of a laser by providing a basis for choosing the operating temperature, pump wavelength, and laser wavelength.

The original distinction between three- and four-level-lasers addressed only the separation between the ground state and the lower laser level [1]. The terminology suffices if the levels are either well-separated or well-overlapped with respect to $k_B T$. The separation between the emitting levels was not an issue initially, because the upper pump levels were far above the upper laser level. This was because early solid-state lasers were pumped with arc lamps whose output spanned the visible spectrum. Most of the excess energy remained as heat, however, which resulted in thermal stresses and optical inhomogeneities. Now, the availability of high power diode lasers has made it possible to pump levels that are not so far above the upper laser level, and that relax to the upper laser level with high efficiency.

Even with today's more efficient diode laser pumping, however, the excess photon energy remains an issue. The recent development of high average power, high brightness solid-state-lasers has had to address this heat load (see sidebar). The 105 kW laser recently demonstrated by Northrop Grumman [2,3] uses a slab geometry to favor heat removal and minimize deleterious thermal gradients, and incorporates adaptive optics to compensate the residual aberrations and achieve a beam quality of three [4,5]. Efficient removal of heat due to a high surface to volume ratio has helped IPG Photonics develop the world's first 10 kW single-mode production fiber laser [6].

Another approach to minimizing the heat load is low-quantum-defect [7] pumping, where the separation between absorbing states should be as small as practical, and likewise for the emitting states. However, the search for systems with a low quantum defect inevitably leads from the ideal case of a four-level-system, down to a two-level system, unless the temperature drops accordingly. What are the implications for laser design? In a 2-level system (Fig. 1a), unless the upper energy level has a higher degeneracy, there can be no inversion in steady state, and thus no gain. In a 3-level laser where the ground state is shared, as in the case of 694.3 nm emission from ruby, ground state absorption (GSA) at the laser wavelength implies a higher laser threshold (Fig. 1b). If the emitting levels are too close, electrons can be thermally excited from the upper laser level to the upper pump level, reducing the gain at $\lambda_l$ and the absorption at $\lambda_a$. When the excited state is shared (Fig. 1c), one can expect to encounter saturation of the pump absorption. If the absorbing levels are too close, ions can be thermally excited from the ground state to the lower laser level, again reducing the gain at $\lambda_l$ and the absorption at $\lambda_a$.

In an ideal 4-level laser, these issues are absent (Fig. 3d), but real lasers fall somewhere in between 2-level and 4-level, because the separation between absorbing levels is comparable to $k_B T$, or the separation between emitting levels is comparable to $k_B T$, or because the level alignments are sub-optimal (Fig. 3e).

Extraneous levels also play a role in all of the above cases because they indirectly modify the absorption at $\lambda_a$ and the gain at $\lambda_l$. If an energy level is added in the middle of the absorbing states, it helps to depopulate the lower laser level, which increases the gain at $\lambda_l$, but it also repopulates the lower pump level, which reduces the absorption at $\lambda_a$. Similar issues arise when extraneous levels appear around the emitting levels.

The quasi-level terminology has arisen to describe intermediate situations that are unclear because of a combination of several levels that are directly involved in the optical transitions, extraneous levels that become populated but that are only indirectly involved, and multiple level spacings, some comparable to $k_B T$. In the last 10 years, there have been 280 papers that have used the terminology quasi-2 level, quasi-3, and quasi-4 in the title or abstract alone [8]. The interest in low-quantum-defect lasers has forced the issue of finding a figure of merit for a system of energy levels that (a) can compare 2-, 3-, and 4-level systems and everything in between, (b) is closely tied to physical quantities like gain, (c) is based on the level occupancies, taking into account thermal population of the lower laser level and of the upper pump level, and (d) helps to choose $\lambda_l$, $\lambda_a$, and the operating temperature. A well-defined figure of merit, e.g. a level scale that varies continuously between two and four, would be a useful intuitive guide to have in mind when thinking about a laser. Can all of that be accomplished with one figure of merit? I believe that it can be done with two closely related figures of merit, called level parameters, derived below.

Using Er:YAG as an example, the $I_{152}$ angular momentum state is split by the crystal field into a manifold of eight sublevels; the $I_{152}$ is split into seven (Fig. 2). To reduce the quantum defect, one can pump and lase between the lowest two manifolds. Referring to Fig. 2, $f_{\ell\ell'}$ is the probability that an electron
is in a state that can emit a laser photon, given that it’s in one of the emitting levels. \( f_{a} \) is the probability that an electron is in a state that can absorb a pump photon, given that it’s in one of the absorbing levels, etc. The optimum case would be where \( f_{a} = f_{r} = 1 \), and \( f_{a} = f_{r} = 0 \), however, the splittings are too small for that to be true at 300 K.

Consider a laser beam and a pump beam propagating through a medium e.g. Er:YAG, at temperature \( T \), characterized by cross sections at \( \lambda_{p} \) and \( \lambda_{L} \). The rate equation for the population density of the absorbing states, \( N_{1} \), includes the conventional terms for absorption at \( \lambda_{p} \), and emission at \( \lambda_{L} \). To these, we add terms for emission at \( \lambda_{p} \) and absorption at \( \lambda_{L} \).

\[
\frac{dN_{1}}{dt} = + \Phi_{p}\sigma_{p}(f_{a}N_{2} - f_{a}N_{1})
+ \Phi_{L}\sigma_{L}(f_{a}N_{2} - f_{a}N_{1}) + N_{2}W_{21}
\]

\[
\frac{dN_{2}}{dt} = - \frac{dN_{1}}{dt}
\tag{1}
\]

\( \Phi_{p}(\lambda_{p}) \) is the pump (laser) photon fluence, \( \sigma_{p}(\sigma_{L}) \) is the cross section at \( \lambda_{p}(\lambda_{L}) \), \( W_{21} \) is the relaxation rate from the emitting states \( N_{2} \) to \( N_{1} \), typically equal to the spontaneous emission rate. The propagation equations show gain for \( \Phi_{L}(\lambda_{L}) \) to the extent that the upper laser (pump) level is occupied, and loss to the extent that the lower laser (pump) level is occupied.

\[
\frac{d\Phi_{L}}{dz} = \sigma_{L}(f_{a}N_{2} - f_{a}N_{1}) \Phi_{L}
\]

\[
\frac{d\Phi_{p}}{dz} = \sigma_{L}(f_{a}N_{2} - f_{a}N_{1}) \Phi_{p}
\tag{2}
\]

Eqns. (1) can be easily solved for \( N_{1} \) and \( N_{2} \) in steady state, and inserted into (2). By neglecting spontaneous emission, and considering the case that \( \Phi_{L}\sigma_{L}(f_{a} + f_{a}) < \Phi_{p}\sigma_{P}(f_{a} + f_{p}) \) (transitions at \( \lambda_{p} \) much faster than transitions at \( \lambda_{L} \)), which we can call the small signal regime, the propagation equation simplifies to

\[
\frac{d\Phi_{L}}{dz} = \sigma_{L}N_{\text{tot}}f_{0}\Phi_{L}, \quad \text{where} \quad f_{0} = \frac{f_{a}f_{a}}{f_{a} + f_{r}}
\tag{3}
\]

\( N_{\text{tot}} \) is the sum of \( N_{1} \) and \( N_{2} \). The exponential gain coefficient for \( \Phi_{L} \) clearly has a factor involving level occupancies alone.

In the large signal regime, the transitions at \( \lambda_{L} \) are much faster than \( \lambda_{p} \), which could obtain inside a laser cavity. In this case, the pump fluence decreases exponentially, and the laser fluence increases asymptotically according to

\[
\frac{d\Phi_{L}}{dz} = \sigma_{L}N_{\text{tot}}f_{1}\Phi_{L}, \quad \text{where} \quad f_{1} = \frac{f_{a}f_{a}}{f_{a} + f_{r}}
\tag{4}
\]

The coefficient that couples \( \Phi_{L} \) and \( \Phi_{L} \) clearly has a factor involving level occupancies alone.

Eqns. (3) and (4) suggest the following definitions for level parameters \( \ell_{0} \) and \( \ell_{1} \).

\[
\ell_{0} = 2(f_{0} + 1) \quad \ell_{1} = 2(f_{1} + 1)
\tag{5}
\]
In Er:Sc$_2$O$_3$, one can see that the level parameters approach their optimum values below ~25 K (Fig. 4).

In the most common scenario for Nd:YAG, where $\lambda_p = 809$ nm and $\lambda_L = 1064$ nm, there is a 24% quantum defect. Although it has historically been considered a four-level laser, one can see that, at 300 K, the level parameters are closer to three (Fig. 5). This is because the occupation probability for the upper laser level is cut in half by the presence of the lower level of the $4F_{3/2}$ doublet, separated by only ~85 cm$^{-1}$. Based on this analysis, it appears that Nd does not deserve the 4-level ranking. Obviously, it does not mean that a good laser cannot be made from Nd, however, the mediocre level parameter has to be compensated by increased doping, or a higher cross section.

The Yb energy level structure is close to ideal because the level separations are good, the alignments are good, and there are no extraneous levels close to the upper laser level, or lower pump level. For $\lambda_p = 941$ nm, $\lambda_L = 1030$ nm, the quantum defect is 9%. One can see that $\ell_1 \sim 3.5$ at 300 K, and rises to close to four at 200 K (Fig. 6).

Using eleven pump and laser transitions in well-known rare-earth ions as examples, one can see that $\ell_0$ and $\ell_1$ span the range from two to four (Fig. 7). The systems that suffer from thermal population of the lower laser level or the upper pump level have higher level parameters at 80 K. The systems that rely on thermal excitation to populate the lower pump level or the upper laser level may have higher level parameters at 300 K.

The search for systems with a low quantum defect inevitably leads to a departure from the ideal case of a four-level-system. At some point, thermal population of either the upper pump level or the lower laser level will reduce the gain at $\lambda_L$, and reduce the absorption at $\lambda_p$. Ground state absorption at $\lambda_L$ will become a factor, or absorption saturation at $\lambda_p$, or both.
The importance of low-quantum-defect lasers to the development of high power solid-state lasers has motivated a quantitative definition of the two-, three-, and four-level laser concept. A simple definition is possible when a unique pump transition can be identified, e.g. when the pumping is by diode lasers. The level parameters $\ell_0$ and $\ell_1$ defined above should be

![Figure 7. $\ell_1$ vs $\ell_0$ for a variety of known lasers at (a) 80 K and (b) 300 K. The color is keyed to the quantum defect. The host material is YAG in all cases, except in the last case, where the host is Sc$_2$O$_3$.](image)

**Approaches to High-Average-Power Solid-State Lasers**

There are several other techniques for dealing with the heat load in solid-state lasers. The thin disk laser operates in reflection so that it can have one entire face in contact with a heat sink, therefore a short thermal path. The cavity propagation axis is nearly normal to the surface, and thus parallel to the thermal gradients [9]. Radiation balancing compensates the heat deposited as a result of every stimulated emission by pumping to the red side of the fluorescence peak, so that the average spontaneous emission removes heat [10]. Cryogenic lasers have attracted renewed interest because of improved thermal conductivity and an index less sensitive to temperature, as well as increased cross sections and reduced ground state absorption [11,12]. Fiber lasers can have efficient heat removal due to the high surface to volume ratio [6]. The heat capacity laser circumvents the heating problem by operating multiple slabs, each with a low duty cycle [13].

**Advances in Low-Quantum-Defect Pumping**

In the most common configuration for diode-pumped Nd:YAG, five different manifolds are populated, $\lambda_p = 808$ nm and $\lambda_L = 1064$ nm, therefore the QD is 24% (see Table I). If the pumping and lasing transitions are chosen to be between levels in the same two manifolds, e.g. with $\lambda_p = 869$ nm and $\lambda_L = 946$ nm, the quantum defect can be reduced to 8%. Changing to Yb$^{3+}$ can reduce the quantum defect to 6%. In these last two examples, the quantum defect is limited in part by the crystal field splitting.

In a host material with a low crystal field strength, e.g. GdVO$_4$, the QD can be reduced to 3%. In a recent experiment, a crystal of Yb:GdVO$_4$ lased at 1,015 nm when longitudinally pumped with a Ti:sapphire laser at 984 nm [14]. The pump beam entered the cavity through a dichroic mirror with high transmission at 984 nm and high reflectivity at 1015 nm. Although the slope efficiency could theoretically be as high as 97% in this case, the experimental result (output power vs incident power) was 32%. Use of CaGdAlO$_4$ and non-colinear pumping has brought the QD in the 1 µm region down to 0.8% [15,16].

In the eye-safer region, where Er:YAG lases at 1,645 nm, pumping at 1,470 nm reduces the quantum defect (QD) to 11% [17,18], compared to 41% when pumping at ~980 nm. Pumping at 1,532 nm further reduces the defect to 7%. A 1.5% QD laser recently demonstrated in Er:Sc$_2$O$_3$ at 77 K by pumping at 1,535 nm and lasing at 1,558 nm [19]. A volume Bragg grating was used for a dichroic input coupler.
particularly useful in these situations where (a) the quantum defect is comparable to $k_B T$, (b) where extraneous levels are present, and (c) where pumping is with narrow band light.

**References**

7. The quantum defect is the difference between the pump photon energy and the laser photon energy.
8. ISI Web of Knowledge, and INSPEC.

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Table I. Fractional quantum defect, and level parameters, calculated for various gain media, and wavelengths, at 300 K.
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