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

by Jeffrey O. White

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14. ABSTRACT
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 ℓ_0 and ℓ_1 defined here 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+}. ℓ_0 and ℓ_1 assign to a system of discrete energy levels a number, with a physical basis, that varies continuously between 2 and 4, in the same way that the Hausdorff and Rényi definitions yield non-integer dimensions for fractals. 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 wave-length, and laser wavelength.

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Research Highlights

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

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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 ℓ_0 and ℓ_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 λ_L and the absorption at λ_P. 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 λ_L and the absorption at λ_P.

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 λ_P and the gain at λ_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 λ_L, but it also depopulates the lower pump level, which reduces the absorption at λ_P. 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 λ_L, λ_P, 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 $^4I_{15/2}$ angular momentum state is split by the crystal field into a manifold of eight sublevels; the $^4I_{13/2}$ 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_{eL} 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_{aP} 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 $f_{aP} = f_{eL} = 1$, and $f_{aL} = f_{eP} = 0$, 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 λ_P and λ_L. The rate equation for the population density of the absorbing states, N_1, includes the conventional terms for absorption at λ_P, and emission at λ_L. To these, we add terms for emission at λ_P and absorption at λ_L.

$$
\begin{aligned}
\frac{dN_1}{dt} &= +\Phi_P\sigma_P(f_{eP}N_2 - f_{aP}N_1) \\
&\quad + \Phi_L\sigma_L(f_{eL}N_2 - f_{aL}N_1) + N_2W_{21} \\
\frac{dN_2}{dt} &= -\frac{dN_1}{dt}
\end{aligned} \tag{1}
$$

$\Phi_P(\Phi_L)$ 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 $\Phi_L(\Phi_P)$ to the extent that the upper laser (pump) level is occupied, and loss to the extent that the lower laser (pump) level is occupied.

$$
\begin{aligned}
\frac{d\Phi_L}{dz} &= \sigma_L(f_{eL}N_2 - f_{aL}N_1)\,\Phi_L \\
\frac{d\Phi_P}{dz} &= \sigma_P(f_{eP}N_2 - f_{aP}N_1)\,\Phi_P
\end{aligned} \tag{2}
$$

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 $\Phi_L\sigma_L(f_{aL} + f_{eL}) \ll \Phi_P\sigma_P(f_{eP} + f_{aP})$ (transitions at λ_P much faster than transitions at λ_L), which we can call the small signal regime, the propagation equation simplifies to

$$
\frac{d\Phi_L}{dz} = \sigma_L N_{tot} f_0 \Phi_L, \qquad \text{where } f_0 \equiv \frac{f_{eL}f_{aP} - f_{aL}f_{eP}}{f_{aP} + f_{eP}}. \tag{3}
$$

N_{tot} is the sum of N_1 and N_2. The exponential gain coefficient for Φ_L clearly has a factor involving level occupancies alone.

In the large signal regime, the transitions at λ_L are much faster than λ_P, which could obtain inside a laser cavity. In this case, the pump fluence decreases exponentially, and the laser fluence increases asymptotically according to

$$
\frac{d\Phi_L}{dz} = \sigma_P N_{tot} f_1 \Phi_P, \quad \text{where } f_1 \equiv \frac{f_{eL}f_{aP} - f_{aL}f_{eP}}{f_{eL} + f_{aL}}. \tag{4}
$$

The coefficient that couples Φ_P and Φ_L clearly has a factor involving level occupancies alone.

Eqns. (3) and (4) suggest the following definitions for level parameters ℓ_0 and ℓ_1.

$$
\ell_0 = 2(f_0 + 1) \qquad \ell_1 = 2(f_1 + 1) \tag{5}
$$

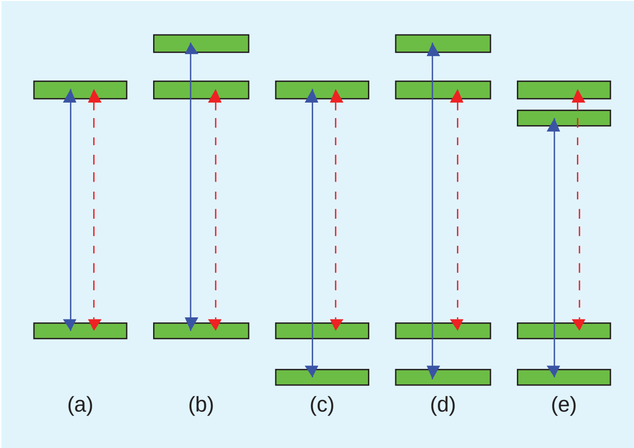

Figure 1. Different possibilities for two-, three-, and four-level lasers, showing pump (solid) and laser (dashed) transitions.

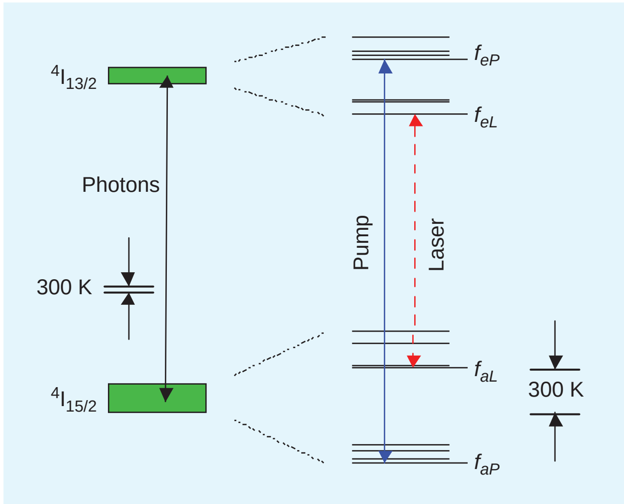

Figure 2. Er^{3+} ground state manifold and first excited state manifold. The scale on the right is magnified.

The level parameters are so-named because in the optimum 4-level case, $\ell_0 = \ell_1 = 4$. In the optimum 3-level case, $\ell_0 = \ell_1 = 3$, and in the 2-level case, $\ell_0 = \ell_1 = 2$. Other things being equal, in the small signal (amplifier) regime, a system with $\ell_0 = 4$ will have twice the gain of a system with $\ell_0 = 3$. In the large signal (laser) regime, a system with $\ell_1 = 4$ will have twice the coupling coefficient of a system with $\ell_1 = 3$.

Given the electronic energy levels of Er:YAG, and assuming a Boltzmann distribution in the absorbing states and the emitting states, one can easily calculate ℓ_0 and ℓ_1 as a function of temperature. For $\lambda_P = 1470$ nm and $\lambda_L = 1645$ nm, there is an 11% quantum defect and the system behaves like a 2.46-level laser at 300 K (Fig. 3). At high temperature, ℓ_1 approaches two, as one would expect. At low temperature, ℓ_1 increases to four. ℓ_0, however, reaches an optimum at 130 K, and then goes to zero because the upper laser level is not the lowest in its manifold, and freezes out at low temperature. Of course, the absorption and emission cross sections will also change with temperature. However, other things being equal, 130 K would be the optimum temperature for an Er:YAG amplifier operating at these wavelengths.

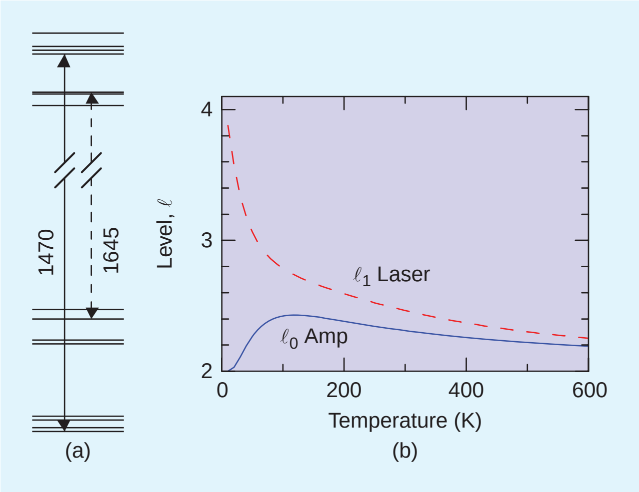

Figure 3. Er:YAG, λ_p = 1470 nm, λ_L = 1645 nm: (a) energy levels, pump transition (solid line), and laser transition (dashed line), (b) temperature dependence of the level parameters.

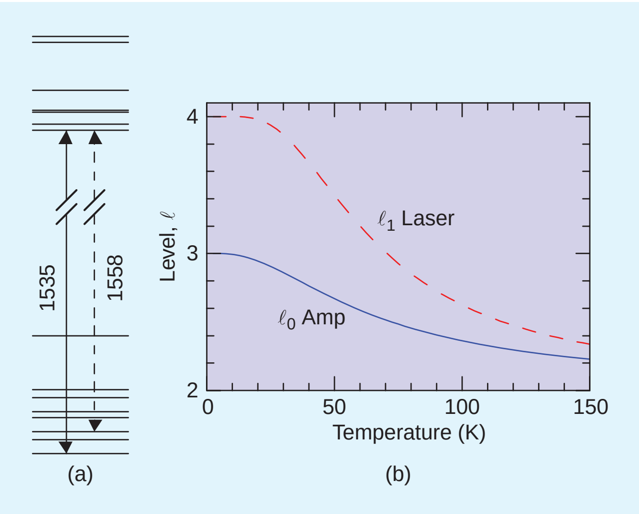

Figure 4. Er:Sc_2O_3, λ_p = 1535 nm, λ_L = 1558 nm: (a) energy levels, pump and laser transitions, (b) temperature dependence of the level parameters.

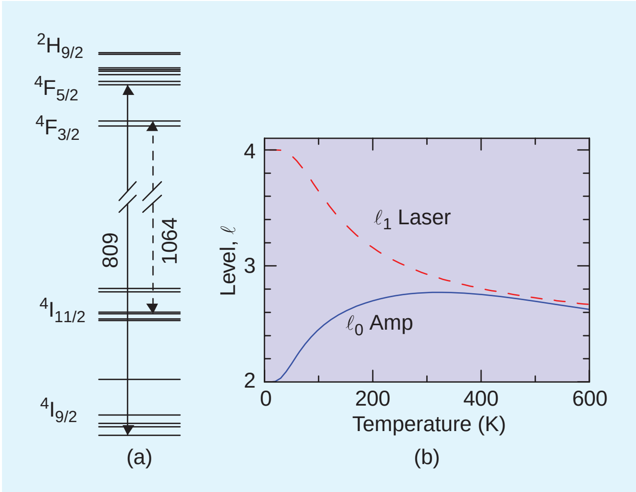

Figure 5. Nd:YAG, λ_p = 809 nm, λ_L = 1064.1 nm: (a) energy levels, pump and laser transitions, (b) temperature dependence of the level parameters.

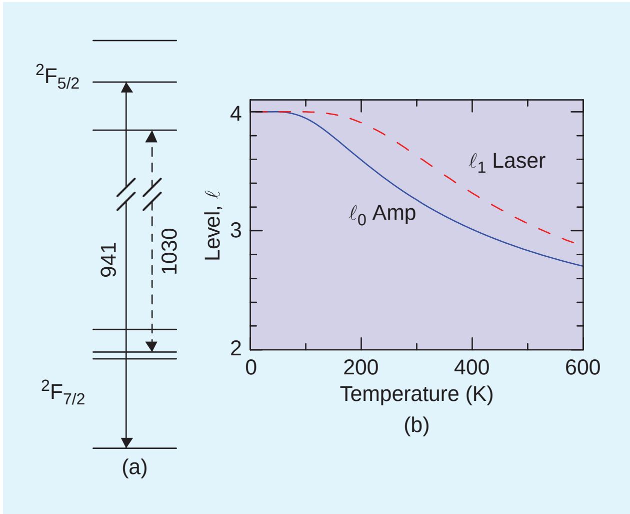

Figure 6. Yb:YAG, λ_p = 941 nm, λ_L = 1030 nm: (a) energy levels, pump and laser transitions, (b) temperature dependence of the level parameters.

In Er:Sc_2O_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 λ_P = 809 nm and λ_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 λ_P = 941 nm, λ_L = 1030 nm, the quantum defect is 9%. One can see that ℓ_1 ~ 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 ℓ_0 and ℓ_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 λ_L, and reduce the absorption at λ_P. Ground state absorption at λ_L will become a factor, or absorption saturation at λ_P, or both.

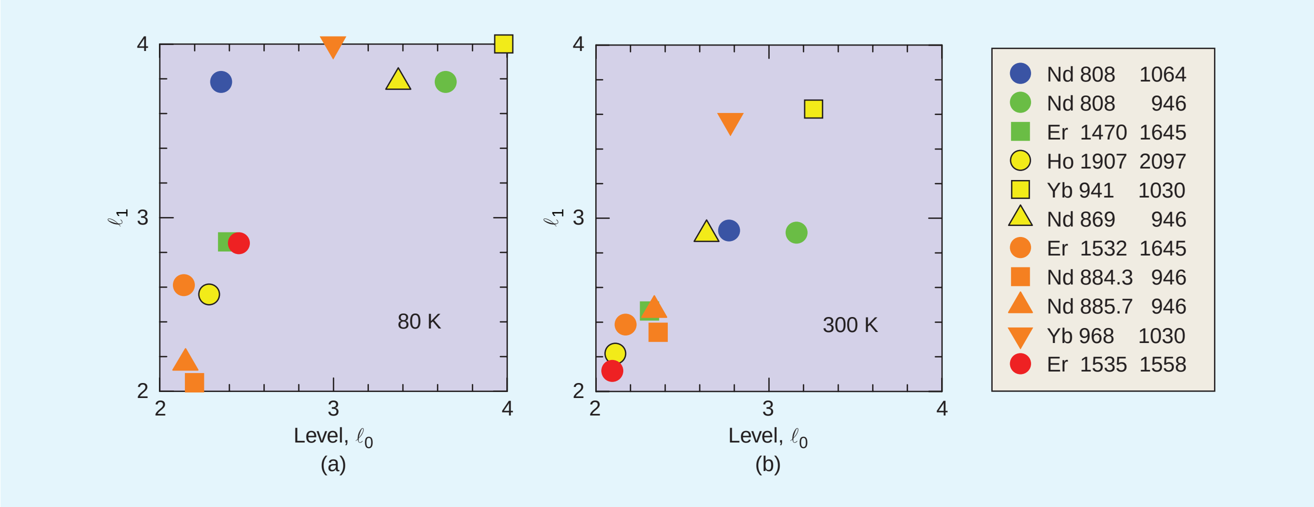

Figure 7. ℓ_1 vs ℓ_0 for a variety of known lasers at (a) 80 K and (b) 300 K. The color is keyed to the quantum defect. The host material is YAG in all cases, except in the last case, where the host is Sc_2O_3.

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 1645 nm, pumping at 1470 nm reduces the quantum defect (QD) to 11% [17,18], compared to 41% when pumping at ~980 nm. Pumping at 1532 nm further reduces the defect to 7%. A 1.5% QD laser recently demonstrated in Er:Sc_2O_3 at 77 K by pumping at 1535 nm and lasing at 1558 nm [19]. A volume Bragg grating was used for a dichroic input coupler.

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 ℓ_0 and ℓ_1 defined above should be

	λ_P (nm)	λ_L (nm)	Fractional QD	ℓ_0	ℓ_1
Er:YAG	1470	1645	0.106	2.2	2.5
“	1532	1645	0.069	2.2	2.4
Er:Sc_2O_3	1535	1558	0.015	2.1	2.1
Nd:YAG	808	1064.1	0.241	2.8	2.9
“	808	946	0.146	3.2	2.9
“	869	946	0.081	2.6	2.9
“	884	946	0.065	2.4	2.3
“	886	946	0.064	2.3	2.5
Yb:YAG	941	1030	0.086	3.3	3.6
“	968	1030	0.060	2.8	3.6
Yb:$GdVO_4$	984	1015	0.029	*	*
Yb:$CaGdAlO_4$	979	987.6	0.009	*	*
Ho:YAG	1907	2097	0.091	2.1	2.2

* to be calculated

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

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.

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