Glass in energy

Laser glass

MAT 498

Lehigh University

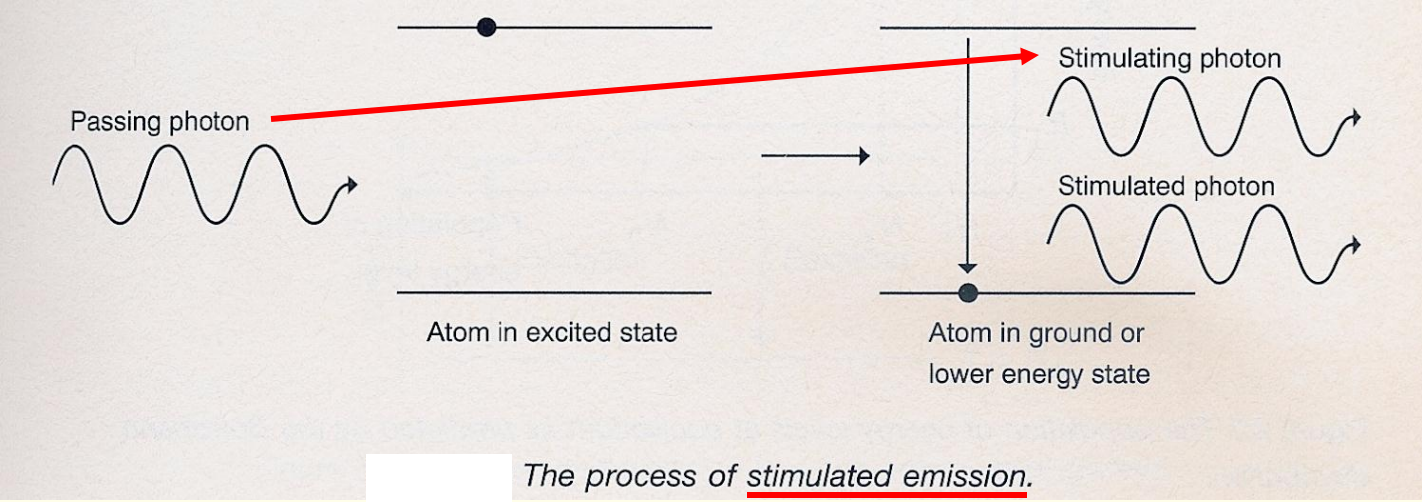
IMI NFG International Materials Institute for New Functionality in Glass

Laser fundamentals

In ***spontaneous*** photoluminescence (PL) processes, light emission is ***incoherent***, that is, there is no phase relation between the different photons emitted.

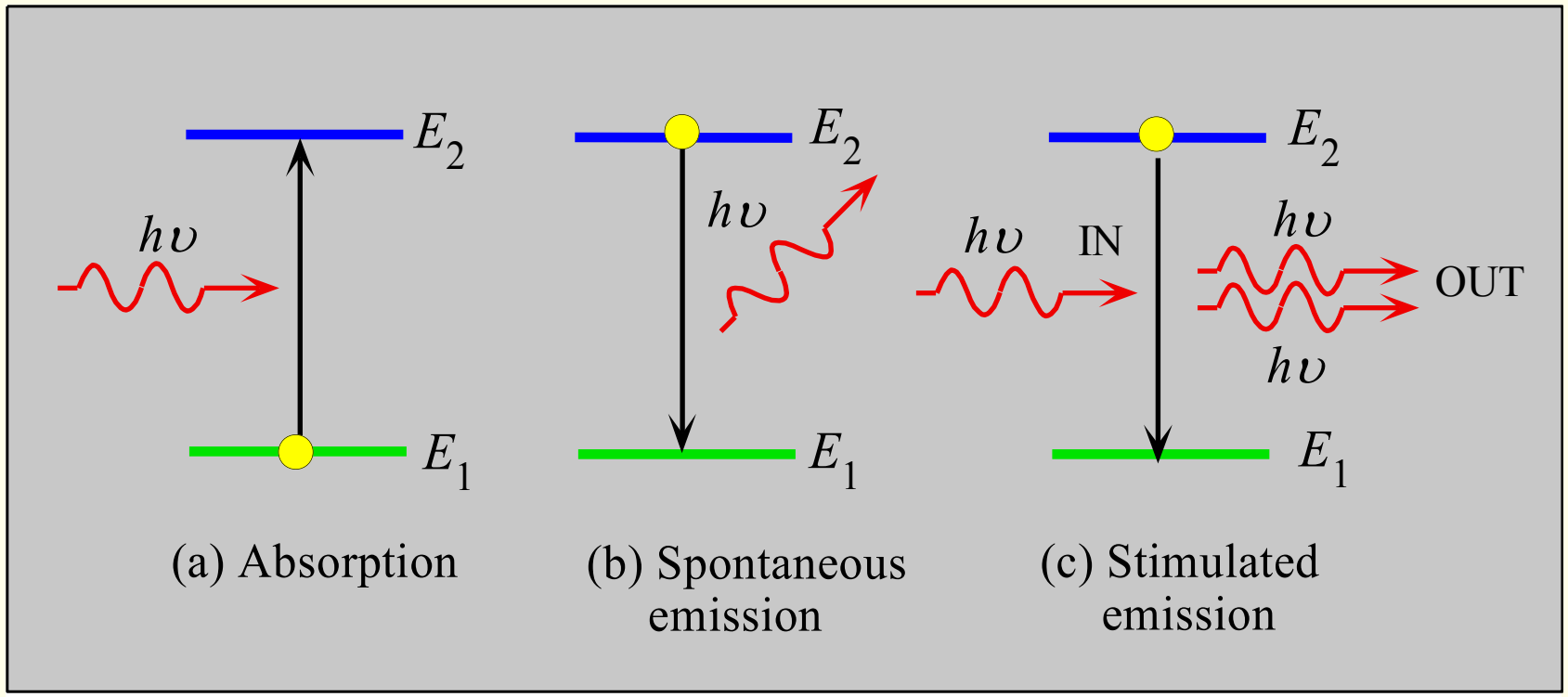
On the contrary, in a **LASER** (**Light Amplification by Stimulated Emission of Radiation**), the (**stimulated**) emitted radiation is ***coherent***, that is, all photons are *in phase* with each other and have the same frequency and polarization, in addition to being highly collimated along the direction of propagation.

In the **stimulated emission** process, an excited atom or ion is *persuaded* to emit a photon by the passage of another photon of the correct energy near the excited atom:



The process of stimulated emission.

(Adapted from: *The essence of optoelectronics*, K. Booth and S. Hill, Prentice hall, 1998)

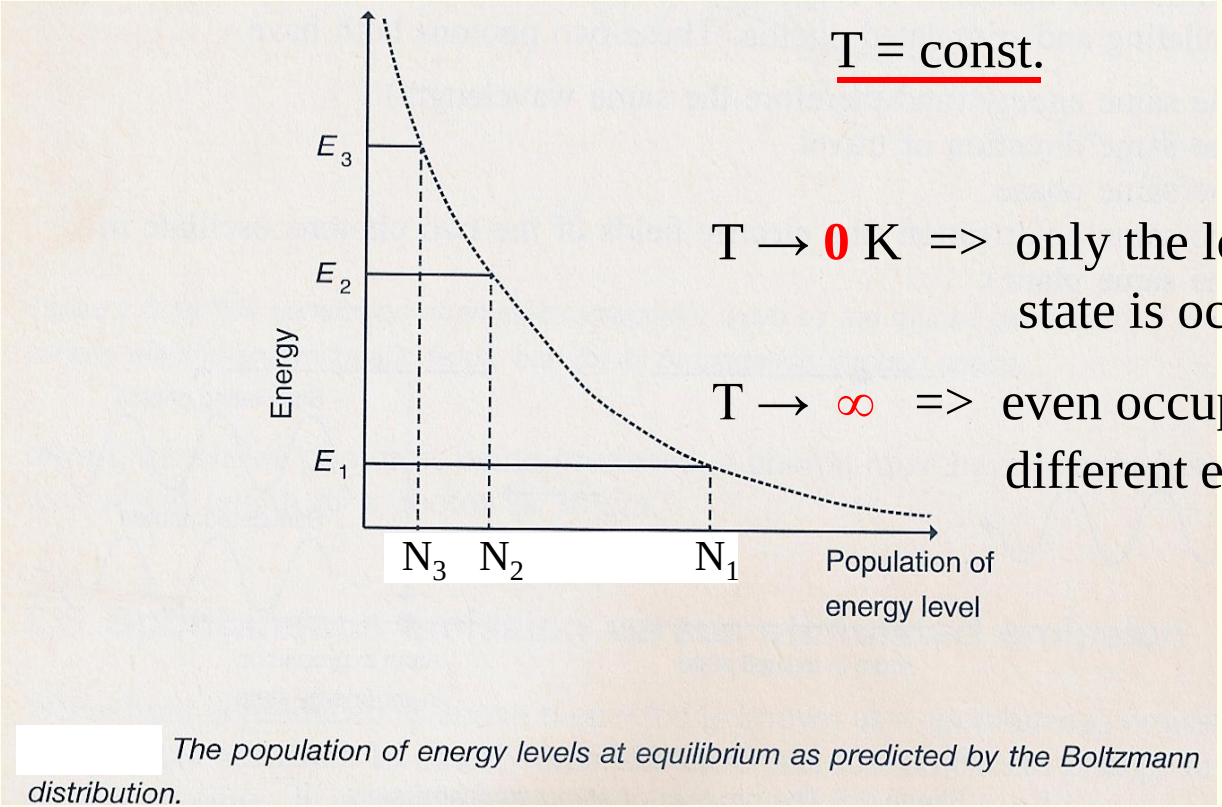


Absorption, spontaneous emission and stimulated emission

From: *Principles of Electronic Materials and Devices*, *Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

The **distribution of the atoms or ions among the different electronic energy levels** has an exponential shape, according to the **Maxwell-Boltzmann** formula. Thus, the **population** of the ith energy level, for a system in equilibrium is a function of the energy and temperature (N_o being a normalization constant):

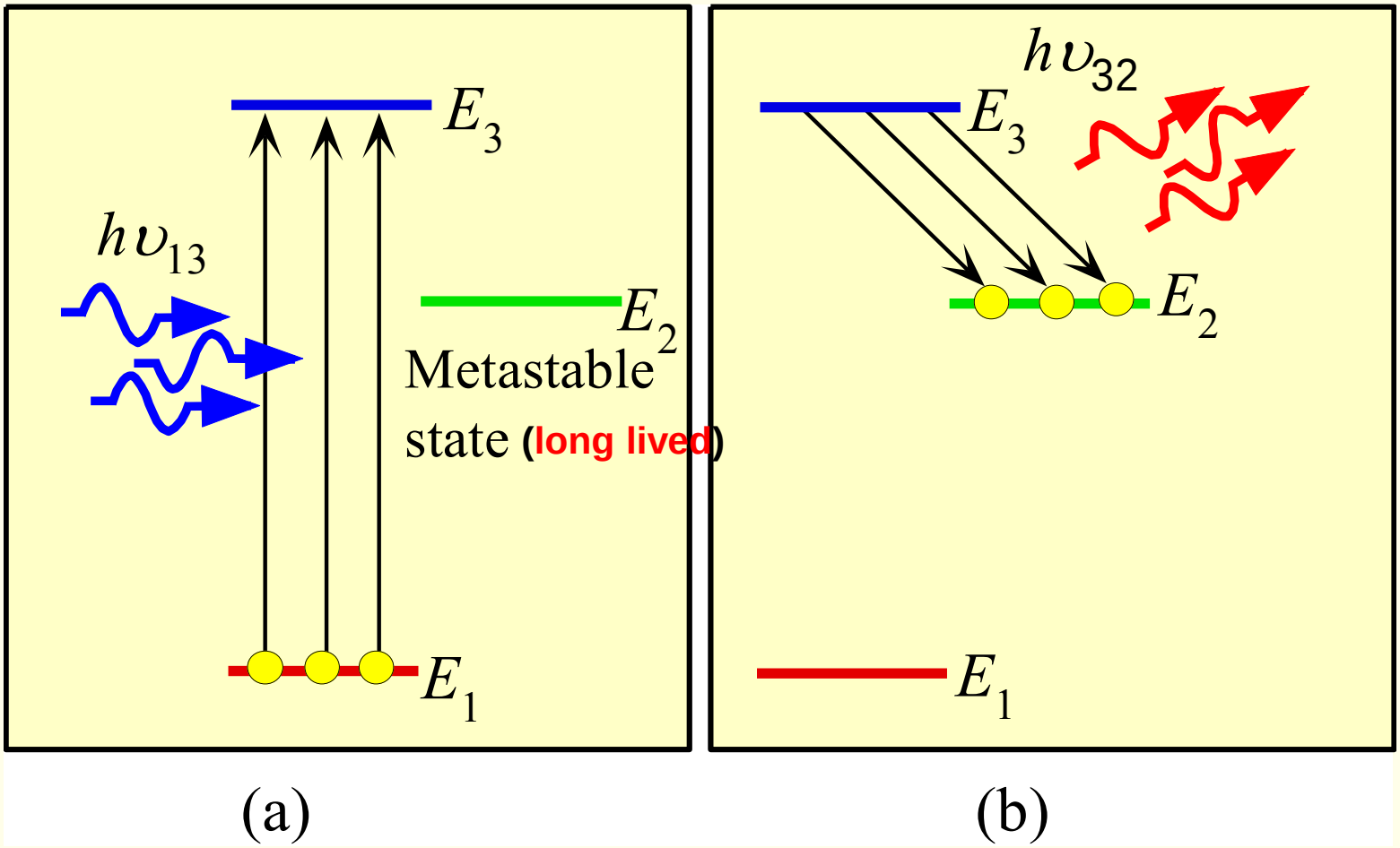
$$N_i = N_o \exp(-E_i / k_B T) \quad <=> \quad N_i / N_j = \exp[-(E_i - E_j) / k_B T]$$



The population of energy levels at equilibrium as predicted by the Boltzmann distribution.

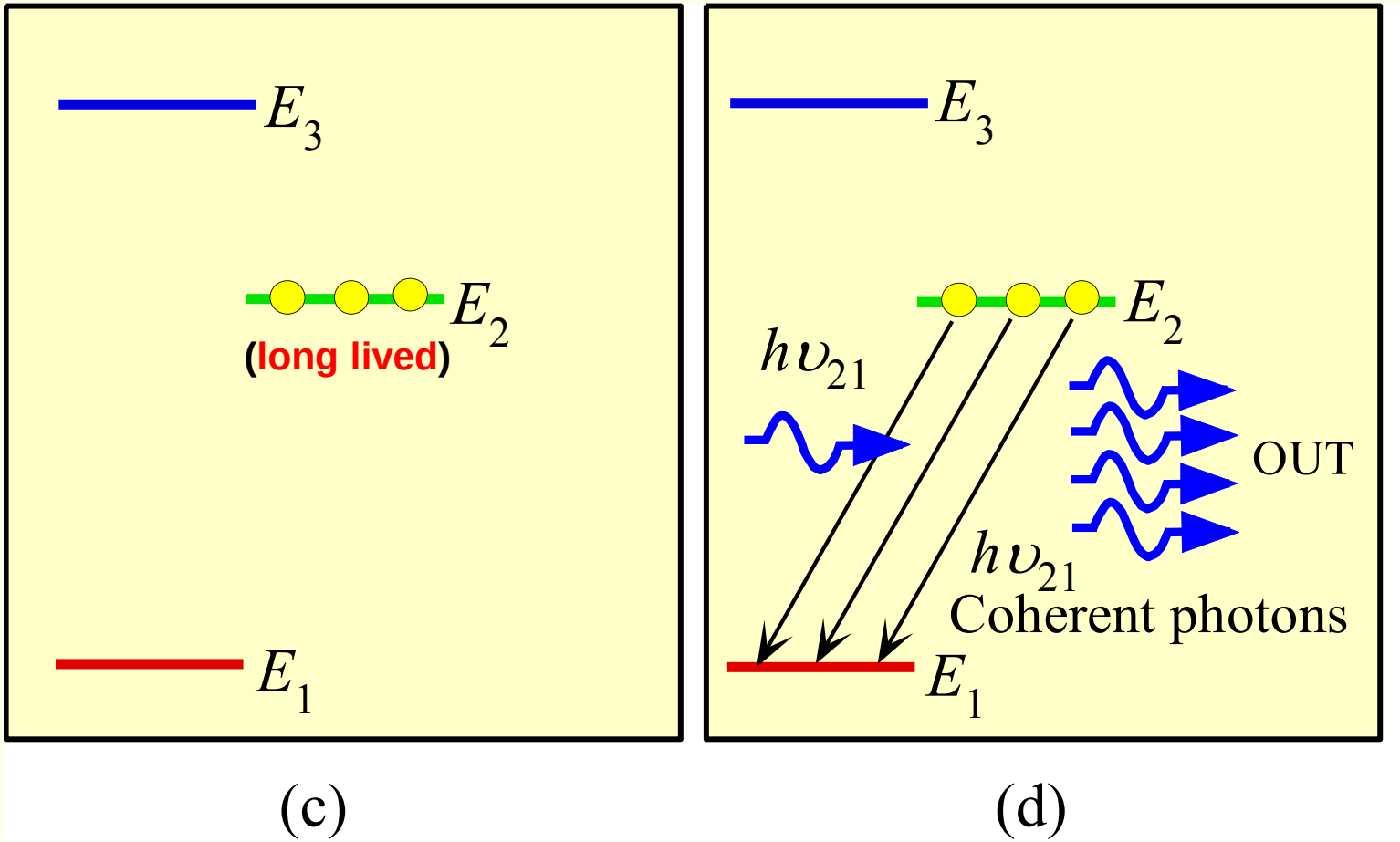
(Adapted from: *The essence of optoelectronics*, K. Booth and S. Hill, Prentice hall, 1998)

The principle of the LASER



(a) Atoms in the ground state are pumped up to the energy level E_3 by incoming photons of energy $h\nu_{13} = E_3-E_1$. (b) Atoms at E_3 rapidly decay to the metastable state at energy level E_2 by emitting photons or emitting lattice vibrations (phonons) with $h\nu_{32} = E_3 - E_2$.

From: *Principles of Electronic Materials and Devices*, *Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)



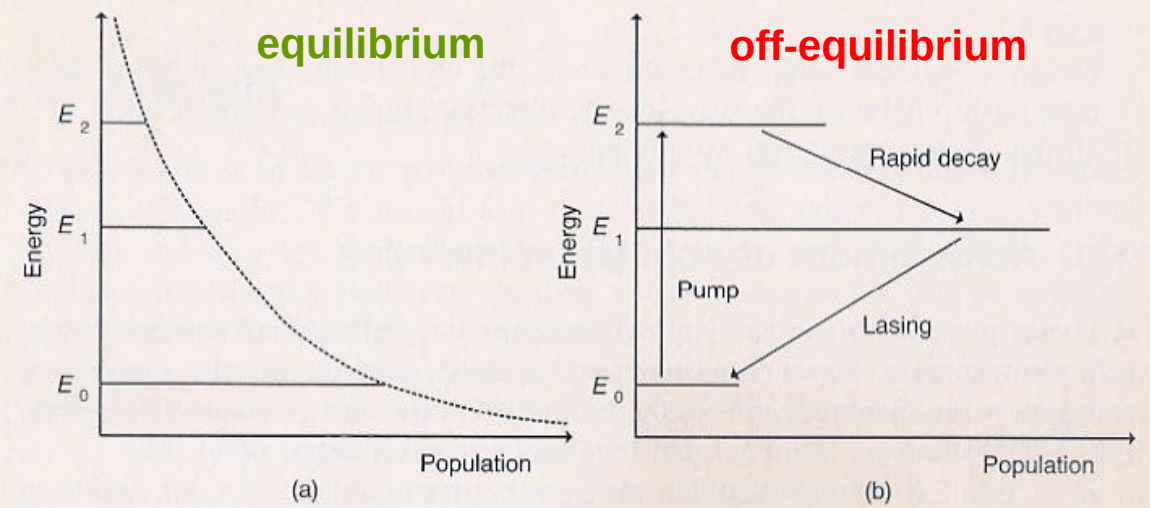
(c) As the states at E_2 are metastable, they quickly become populated and there is a **population inversion** between E_2 and E_1. (d) A random photon of energy $h\nu_{21} = E_2\text{-}E_1$ can initiate stimulated emission. Photons from this stimulated emission can themselves further stimulate emissions leading to an avalanche of stimulated emissions and coherent photons being emitted.

From: *Principles of Electronic Materials and Devices*, *Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Achievement of population inversion

With a simple two-level system, it is impossible to achieve population inversion and thus light amplification, even in the presence of stimulated emission. For that, **at least 3 levels are needed**. **Most lasers are 3- or 4-level systems**. To achieve a significant population inversion, **it is desirable that the excited level be a long-lived, or metastable level**, so that atoms can be excited to this state faster than they leave it.

In the example below (3-level), after pumping, the equilibrium population, in (a), changes to a non-equilibrium one, in (b). Ideally, the transition from E_2 to E_1 (the metastable level) should be as rapid as possible and E_2 should consist of a band of energy levels, to use the most pump power, which usually contains a range of energies.

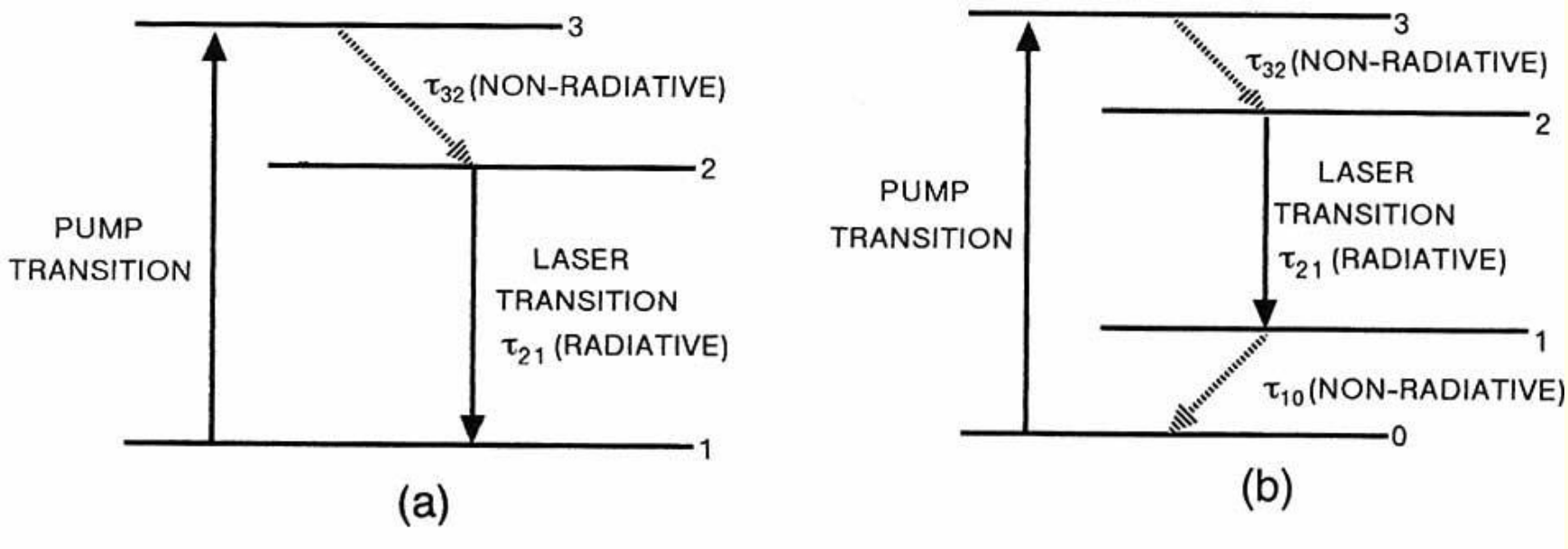


Population of energy levels (a) at equilibrium and (b) far from equilibrium after pumping.

(Adapted from: *The essence of optoelectronics*, K. Booth and S. Hill, Prentice Hall, 1998)

Three- and four-level lasing systems

(e.g. Er^{3+}) **(e.g. Nd^{3+})**



Simplified energy diagrams used to explain the operation of optically pumped (a) three-level and (b) four-level laser systems.

(Adapted from: *Glasses for photonics*, M. Yamane and Y. Asahara, Cambridge Univ. Press, 2000)

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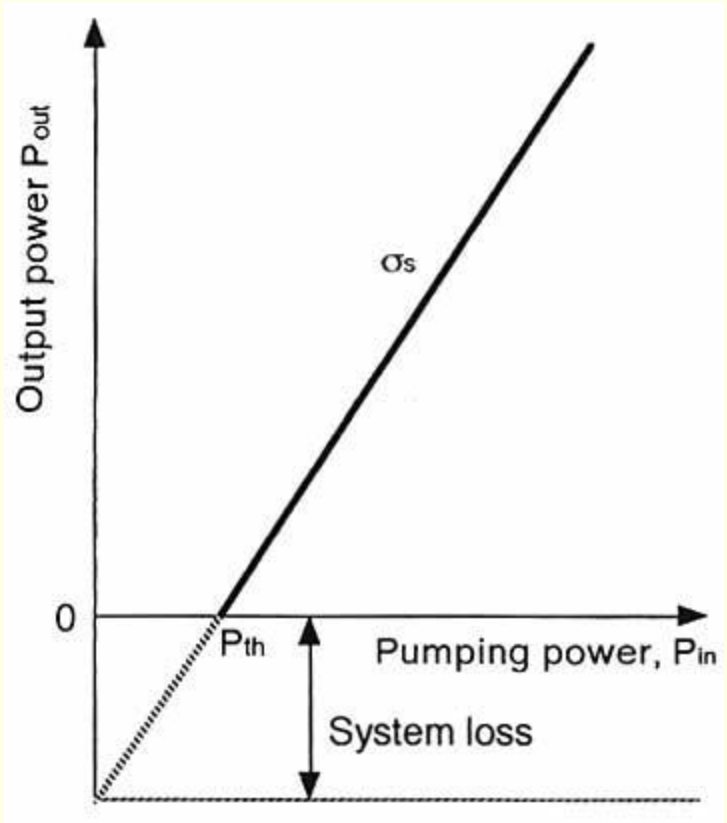
In a laser, assuming that the pump rate is a linear function of the input pump power, P_{in}, the population inversion is a function of the pump intensity and the output power, P_{out}, is a function of P_{in}:

$$P_{out} = \sigma_s (P_{in} - P_{th})$$

where **P_{th} is the minimum required, or *threshold*, input power**; **σ_s is the *slope efficiency*** of the laser curve. P_{th} is higher for a 3-level laser than for a 4-level one (shown below).



Output power versus pump input for a four-level system.

(Adapted from: *Glasses for photonics*, M. Yamane and Y. Asahara, Cambridge Univ. Press, 2000)

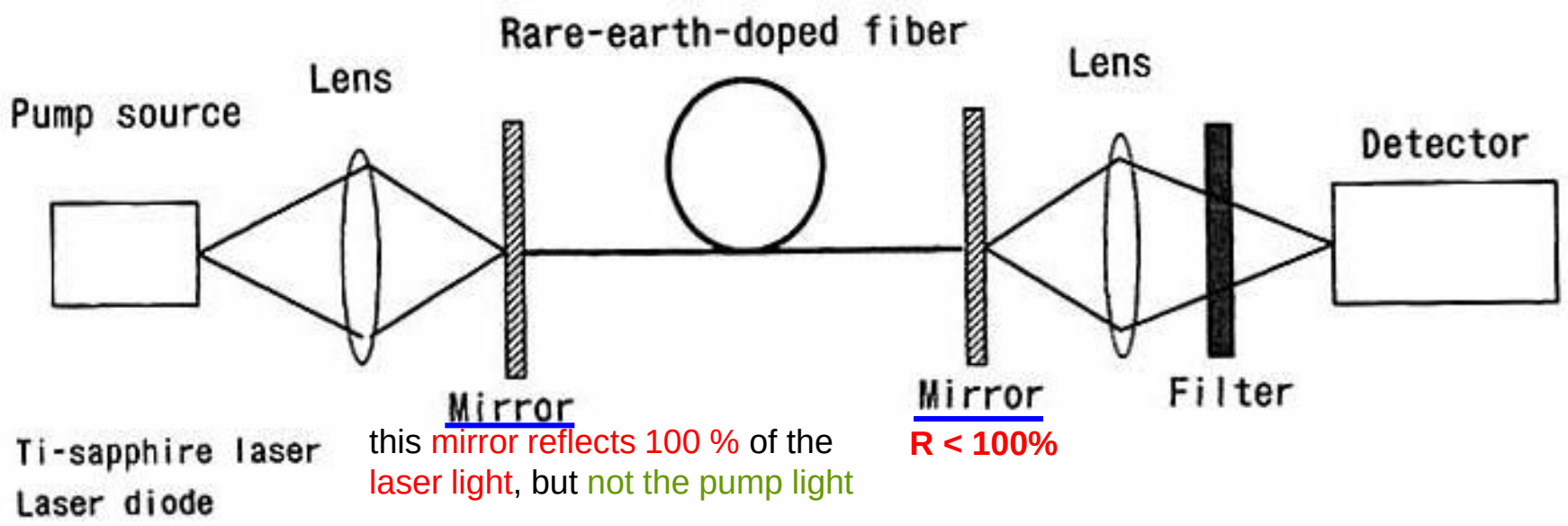
The laser output power is proportional to the excess pump power above threshold.

Fiber laser oscillators

Both bulk and fiber glass **lasers** and **amplifiers** are used.

E.g. RE-doped **fiber lasers** can be used in both long-haul (1.5 μm) and local area network (1.3 μm) communications, as light sources. Such systems typically **use *single mode* fibers**.

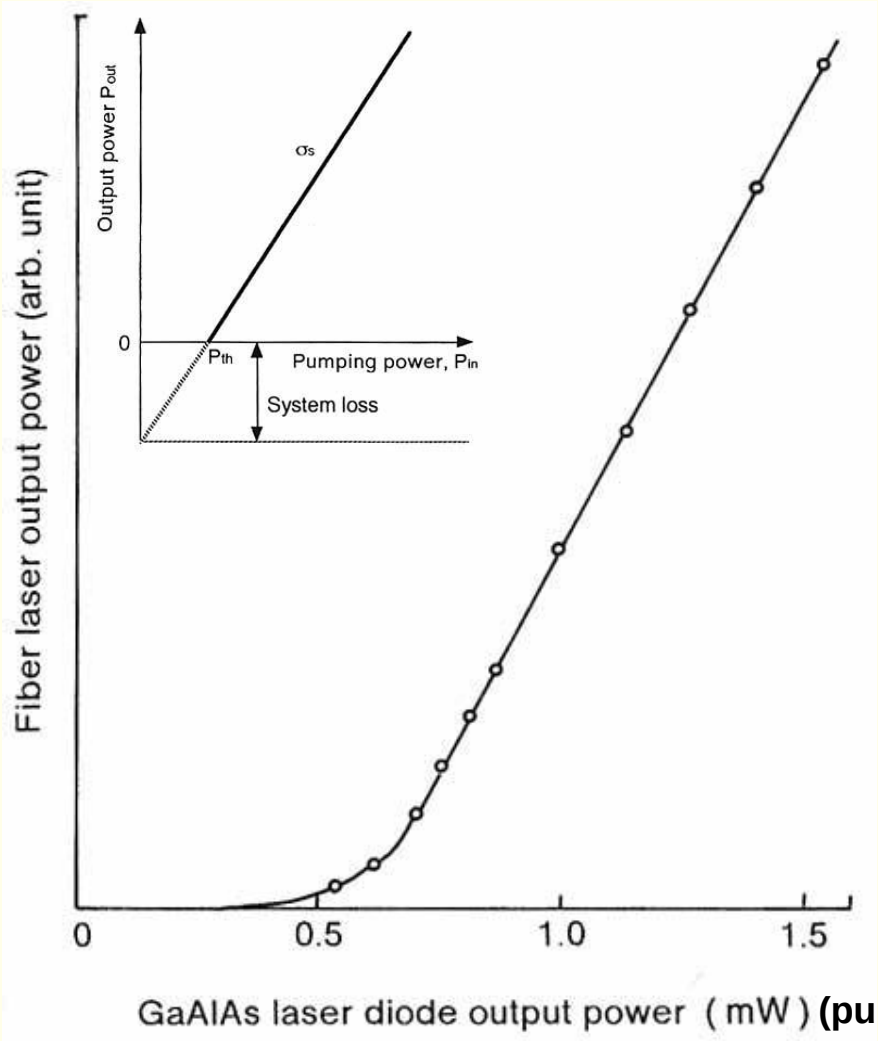
The tight confinement of light in a fiber helps the achievement of a large population inversion density and glass fiber lasers readily operate in CW mode, at room temperature, without the need for cooling.



Typical configuration of fiber laser oscillator.

(Adapted from: *Glasses for photonics*, M. Yamane and Y. Asahara, Cambridge Univ. Press, 2000)

Typical output power vs. pump power curve for a fiber laser oscillator

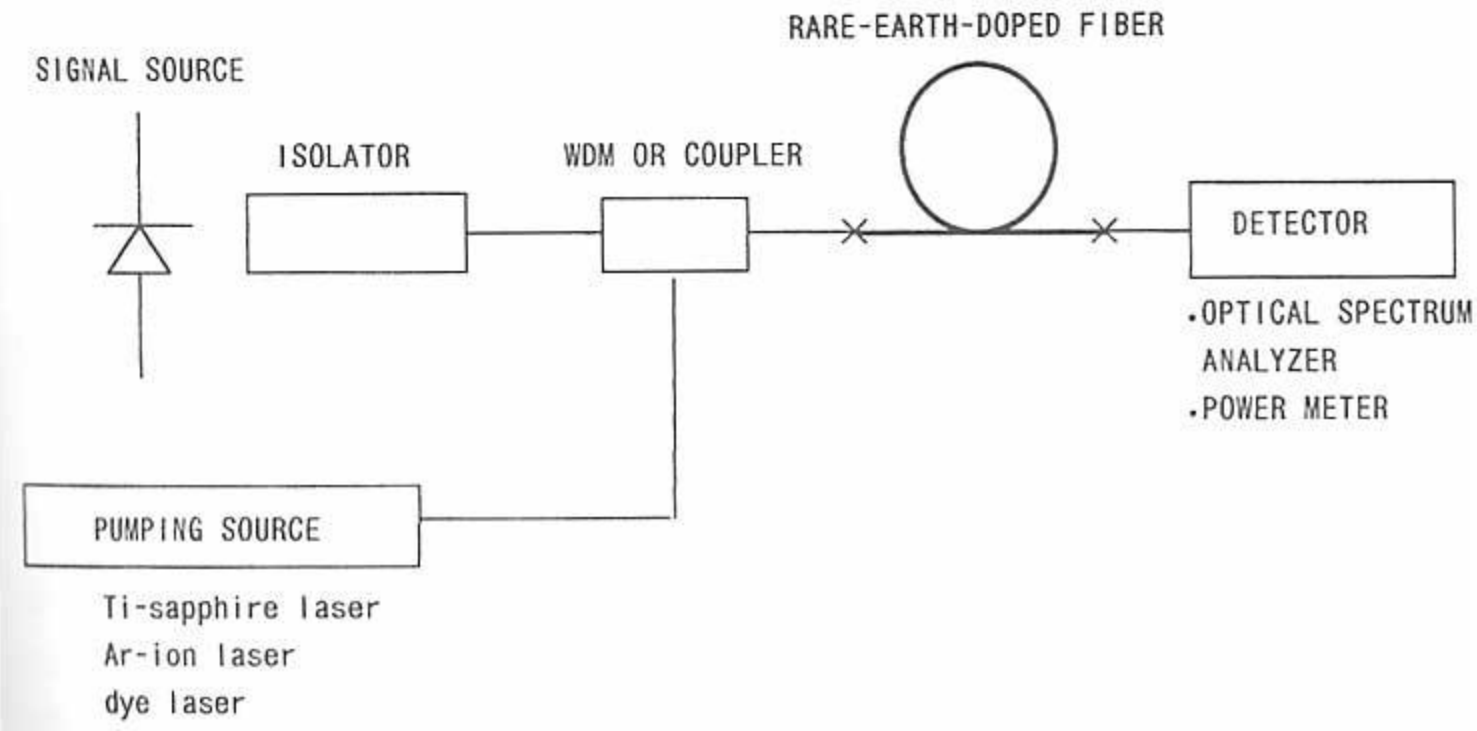


Typical example of the output power as a function of pump power. [Reprinted from R. J. Mears, L. Reekie, S. B. Poole and D. N. Payne, *Electron. Lett.* **21** (1985) 738, copyright (1985) with permission from the Institute of Electrical Engineers.]

(Adapted from: *Glasses for photonics*, M. Yamane and Y. Asahara, Cambridge Univ. Press, 2000)

Fiber amplifiers

The driving force for the development of RE-doped fiber amplifiers was their application in fiberoptics operating at 1.5 μm, or for the installed systems at 1.3 μm. It suffices **to splice a section of RE-doped fiber into the transmission fiber and injecting into it pump light** through a fiber coupler, as shown below. The incoming signal will stimulate emission of the excited RE ions, which amplifies the signal.



Typical experimental configuration of fiber optical amplifier.

(Adapted from: *Glasses for photonics*, M. Yamane and Y. Asahara, Cambridge Univ. Press, 2000)

A fiber laser differs from a fiber amplifier in that : (1) each fiber laser end must be coupled to a mirror which provides reflection at the laser signal wavelength; (2) no signal is injected into the laser cavity.

Schematic comparison between *fiber laser* **oscillators** and *fiber* ***amplifiers***

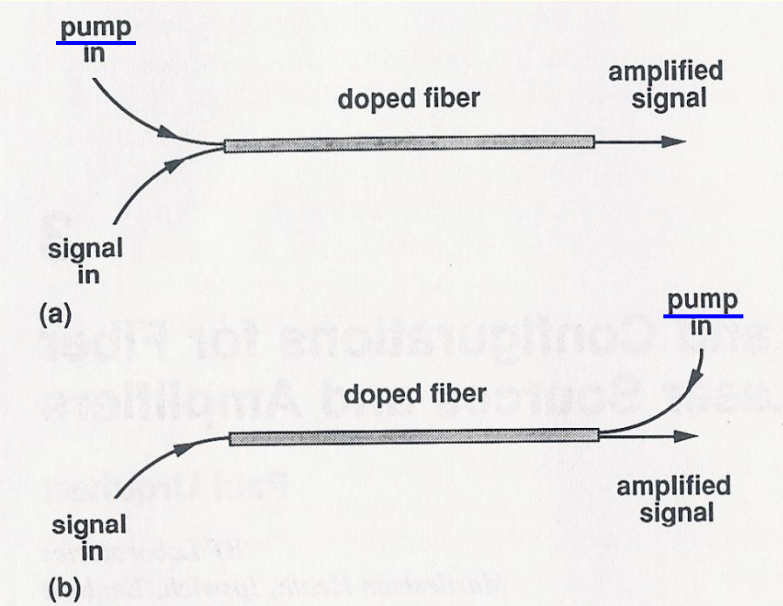
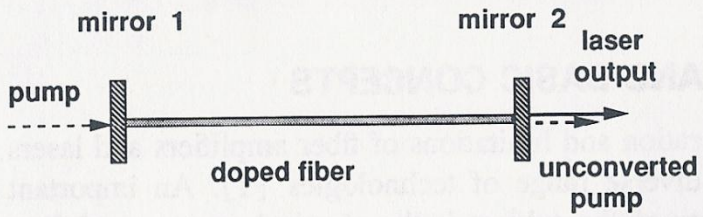


Figure 1 Schematic of a fiber amplifier.



Schematic of a fiber laser.

(Adapted from: *Rare earth doped fiber lasers and amplifiers*, M.J.F. Digonnet, Marcel Dekker, 1993)

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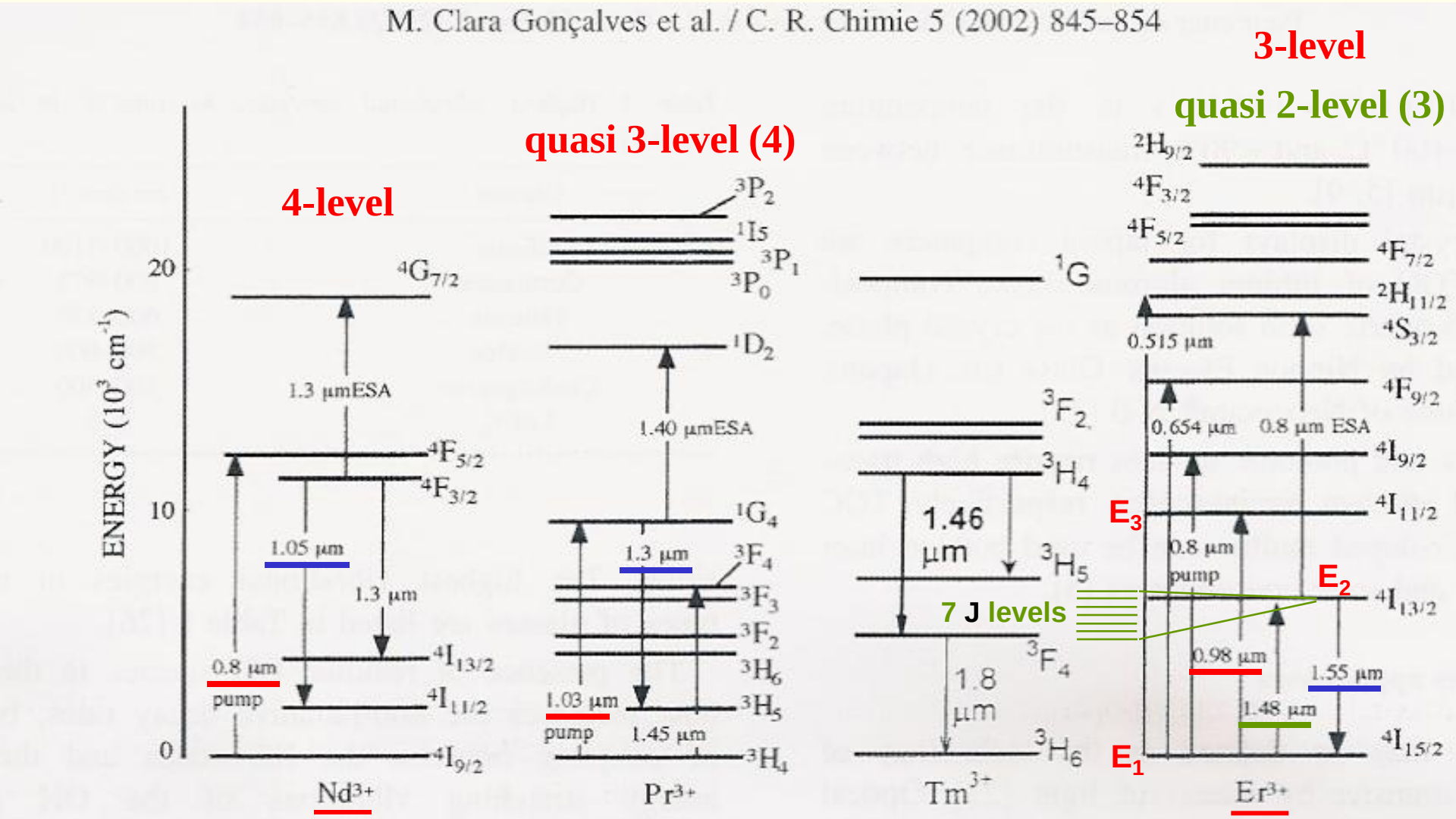
The example of Er^{3+}

Er-doped fiber amplifiers

(EDFAs)

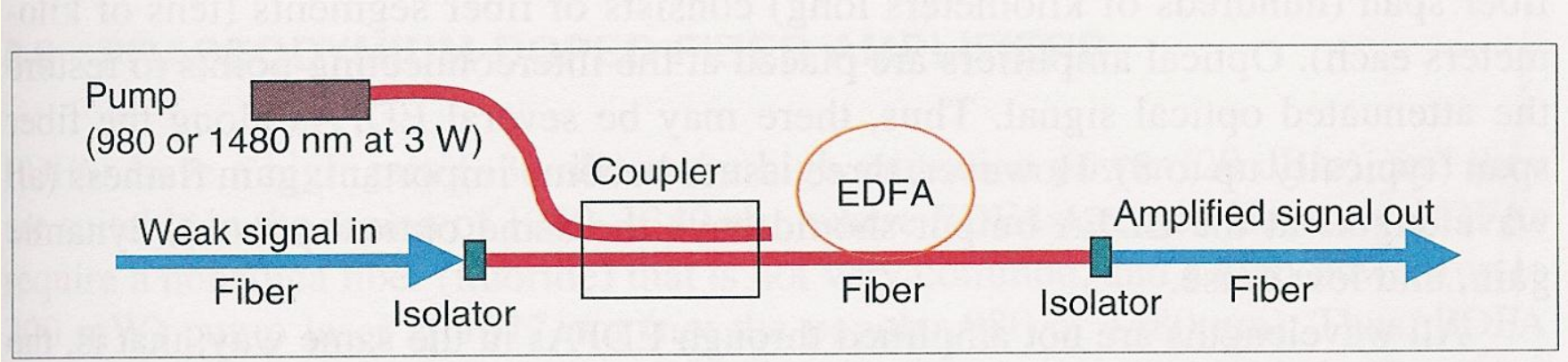
Possible RE ion (active) dopants for glassy hosts



Simplified electronic energy level diagrams of three different RE ions and relevant transitions for pumping and emission.

A fiber amplifier is a section of doped fiber; pump and signal waves are launched into this fiber either at the same end, or at opposite ends and the amplified signal emerges at the opposite end from which the signal was launched.

Example: Erbium Doped Fiber Amplifier

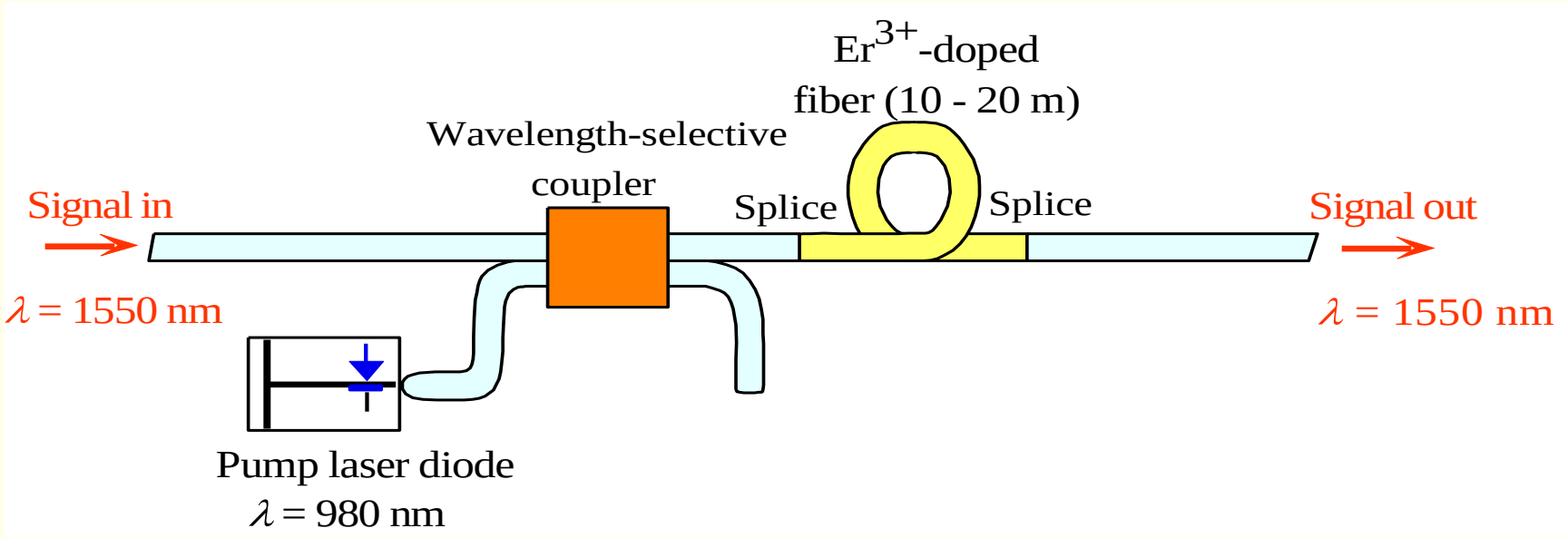


An EDFA amplifier consists of an erbium-doped silica fiber, an optical pump, a coupler, and isolators at both ends.

(Adapted from: *Introduction to DWDM technology*, S.V. Kartalopoulos, IEEE Press, 2000)

Er-doped fiber amplifiers

It is necessary to regenerate a light signal after it has travelled several thousand kms. The SiO_2-GeO_2-Al_2O_3 core region of the active optical fiber is doped with Er^{3+} ions and spliced by fusion to a long distance passive optical fiber. **Upon pumping @ 980 nm, the metastable (E_2) level is reached with a long lifetime ~ 10 ms.**



A simplified schematic illustration of an EDFA (optical amplifier). The erbium-ion doped fiber is pumped by feeding the light from a laser pump diode, through a coupler, into the erbium ion doped fiber.

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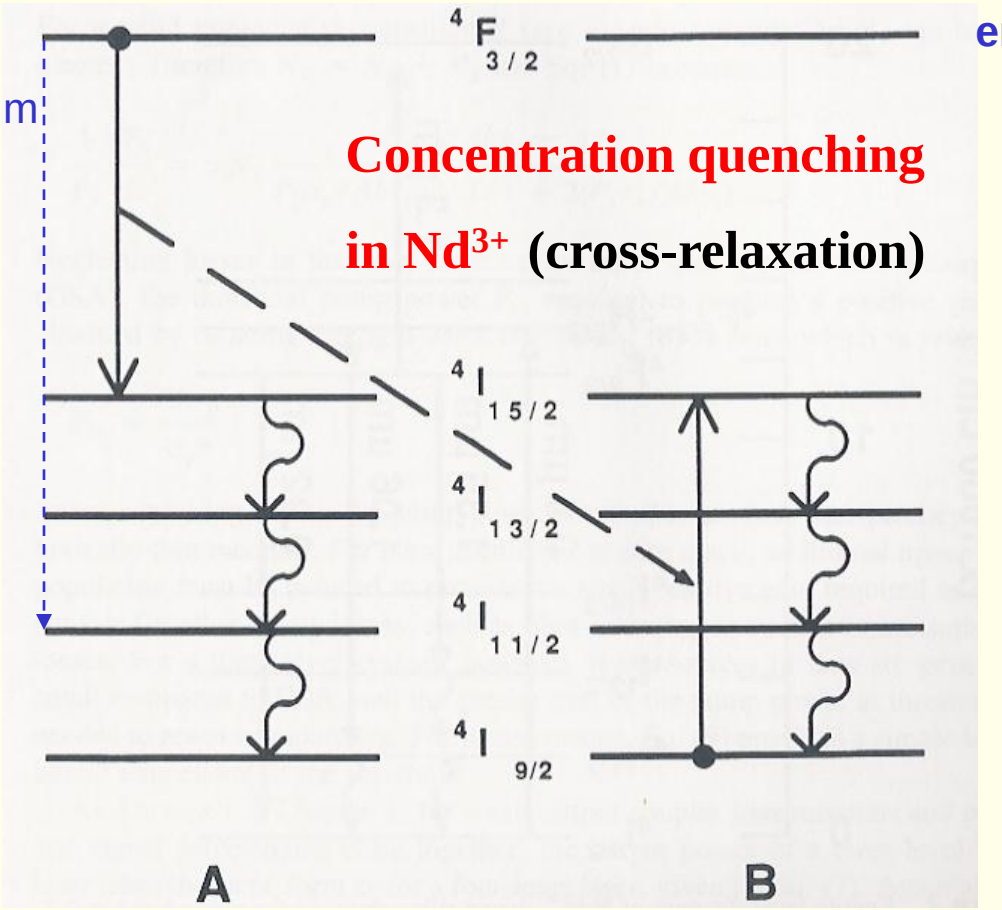
From: *Principles of Electronic Materials and Devices*, *Third Edition*, S.O. Kasap (© McGraw-Hill, 2005)

Nd^{3+}-doped bulk glass lasers and amplifiers

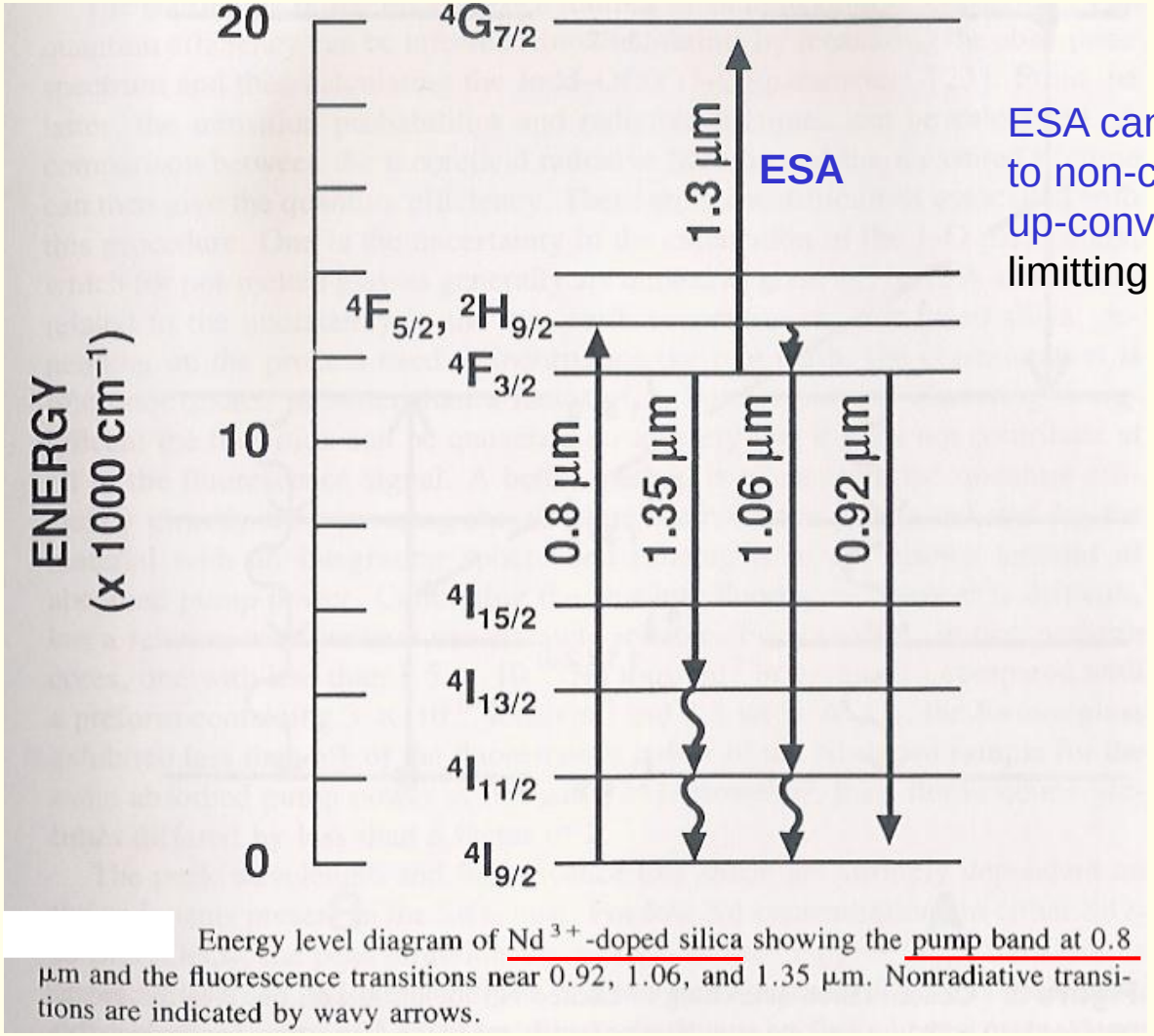
The *Quantum efficiency* of fluorescence (η_Q), for a given RE-glass system, sets the upper limit of the RE concentration, limited by *concentration quenching*. The figure shows *cross-relaxation* for Nd^{3+}, between an excited ion A and a neighboring ion B in the ground state, with both ions making non-radiative transitions to the intermediate $^4I_{15/2}$ state, from which they decay non-radiatively to the ground state. ***Cooperative up-conversion* is another possible mechanism for concentration quenching**.

$$\eta_Q = \frac{\text{\# emitted photons}}{\text{\# pump photons}}$$



(Adapted from: *Rare earth doped fiber lasers and amplifiers*, ed. M.J.F. Digonnet, Marcel Dekker, 1993)

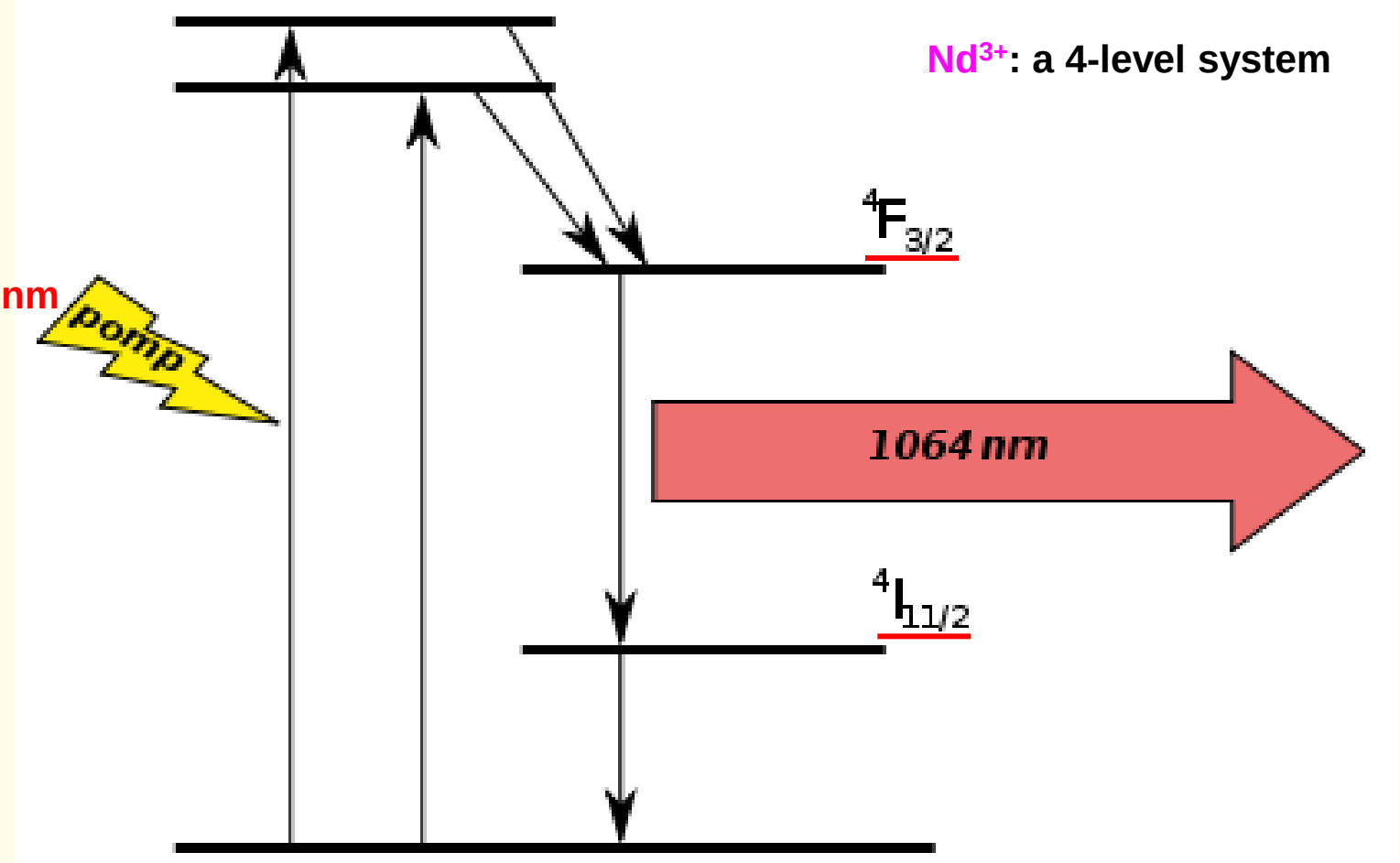
Relaxation of the $^4F_{3/2}$ level of Nd^{3+} is primarily radiative for all the more common glasses, with the exception of the high vibrational energy **borates**, which **have high non-radiative relaxation rates** leading to $^4F_{3/2}$ excited state lifetimes as short as 45 μs (η_Q ~ 10-15 %) . The presence of impurity OH groups (ν (O-H) ~ 2.9 μm) can also be a problem, for any glass host.



Energy level diagram of Nd^{3+}-doped silica showing the pump band at 0.8 μm and the fluorescence transitions near 0.92, 1.06, and 1.35 μm. Nonradiative transitions are indicated by wavy arrows.

ESA can also lead to non-cooperative up-conversion (thus limitting η_Q).

(Adapted from: *Rare earth doped fiber lasers and amplifiers*, ed. M.J.F. Digonnet, Marcel-Dekker, 1993)



Neodymium ions in various types of ionic crystals and in **glasses** act as a **laser** gain medium, typically **emitting 1064 nm** light from a particular atomic transition in the Nd^{3+} ion, after being "pumped" into excitation from an external source.

Adapted from: http://en.wikipedia.org/wiki/File:Laser_glass_slabs.jpg (7 Jan 2012)

1.06 µm. *Range of spectroscopic properties for the $^4F_{3/2}$–$^4I_{11/2}$ transition of Nd^{3+} observed in different glasses at 295 K [Reprinted from M. J. Weber, J. Non-Cryst. Solids **123** (1990) 208, copyright (1990) with permission from Elsevier Science]*

Nd^{3+} Host glass	Refractive index n_d	Cross section σ_p (pm^2)	Peak wavelength λ_p (µm)	Effective linewidth $\Delta\lambda_{eff}$ (nm)	Radiative lifetime τ_R (µs)
Oxide					
silicate	1.46–1.75	0.9–3.6	1.057–1.088	35–55	170–1090
germanate	1.61–1.71	1.7–2.5	1.060–1.063	36–43	300–460
tellurite	2.0–2.1	3.0–5.1	1.056–1.063	26–31	140–240
phosphate	1.49–1.63	2.0–4.8	1.052–1.057	22–35	280–530
borate	1.51–1.69	2.1–3.2	1.054–1.062	34–38	270–450
Halides					
beryllium fluoride	1.28–1.38	1.6–4.0	1.046–1.050	19–29	460–1030
aluminium fluoride	1.39–1.49	2.2–2.9	1.049–1.050	28–32	540–650
heavy metal fluoride	1.50–1.56	2.5–3.4	1.048–1.051	25–29	360–500
chloride	1.67–2.06	6.0–6.3	1.062–1.064	19–20	180–220
Oxyhalides					
fluorophosphate	1.41–1.56	2.2–4.3	1.049–1.056	27–34	310–570
chlorophosphate	1.51–1.55	5.2–5.4	1.055	22–33	290–300
Chalcogenides					
Sulfide	2.1–2.5	6.9–8.2	1.075–1.077	21	64–100
oxysulfide	2.4	4.2	1.075	27	92

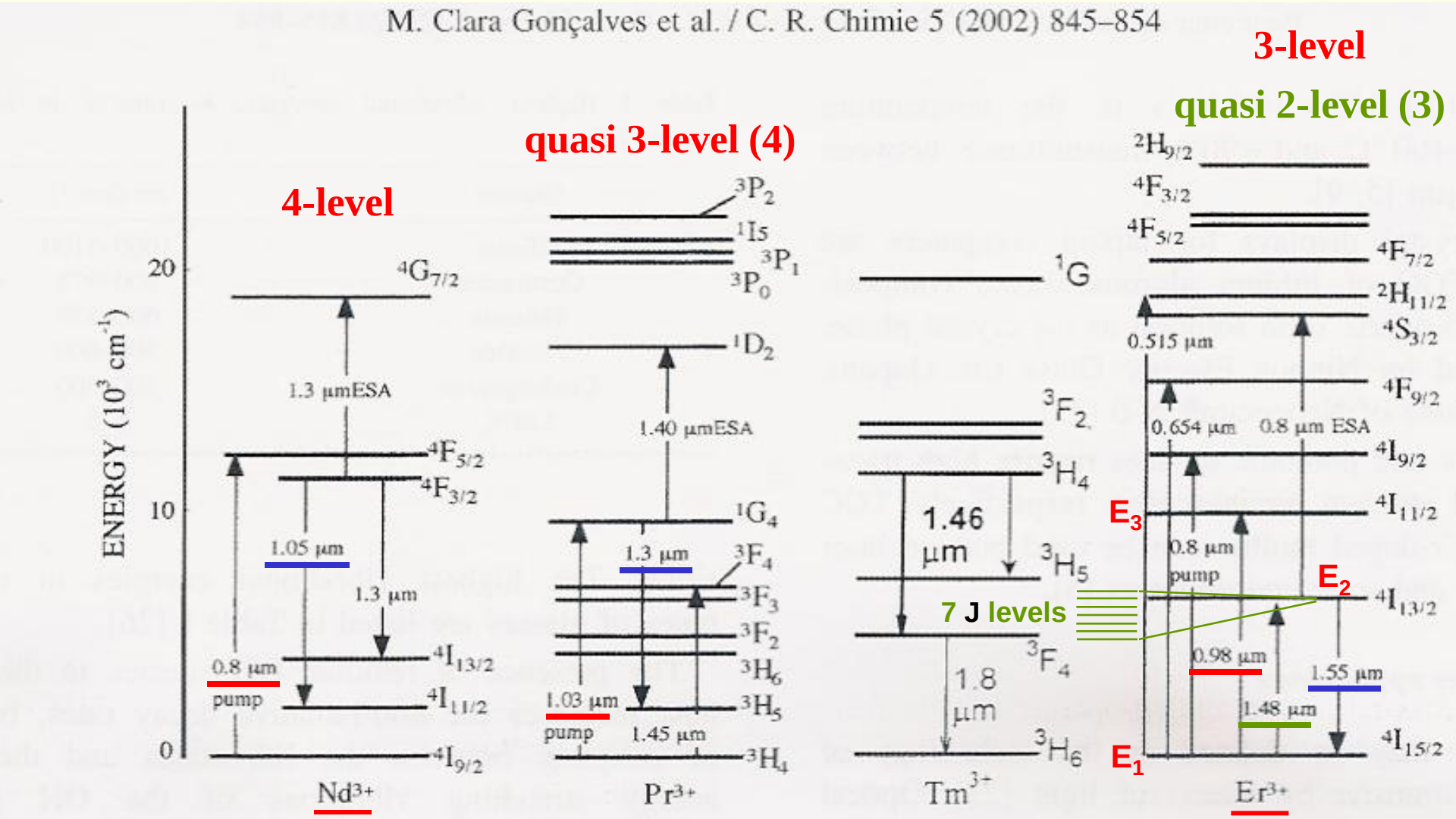
(Adapted from: *Glasses for photonics*, M. Yamane and Y. Asahara, Cambridge Univ. Press, 2000)

Peak broadening

The ***homogeneous broadening*** of the transitions between the Stark components of different J multiplets is caused by lifetime broadening ($\Delta E.\Delta\tau \sim \hbar$) in both crystals and glasses, dominated by rapid phonon-induced transitions between the individual Stark components within a given multiplet (thermalization). Such transitions occur on a ~ ps time scale at low temperature, but become much faster at room temperature, causing **homogeneous broadening** to **increase significantly with temperature**.

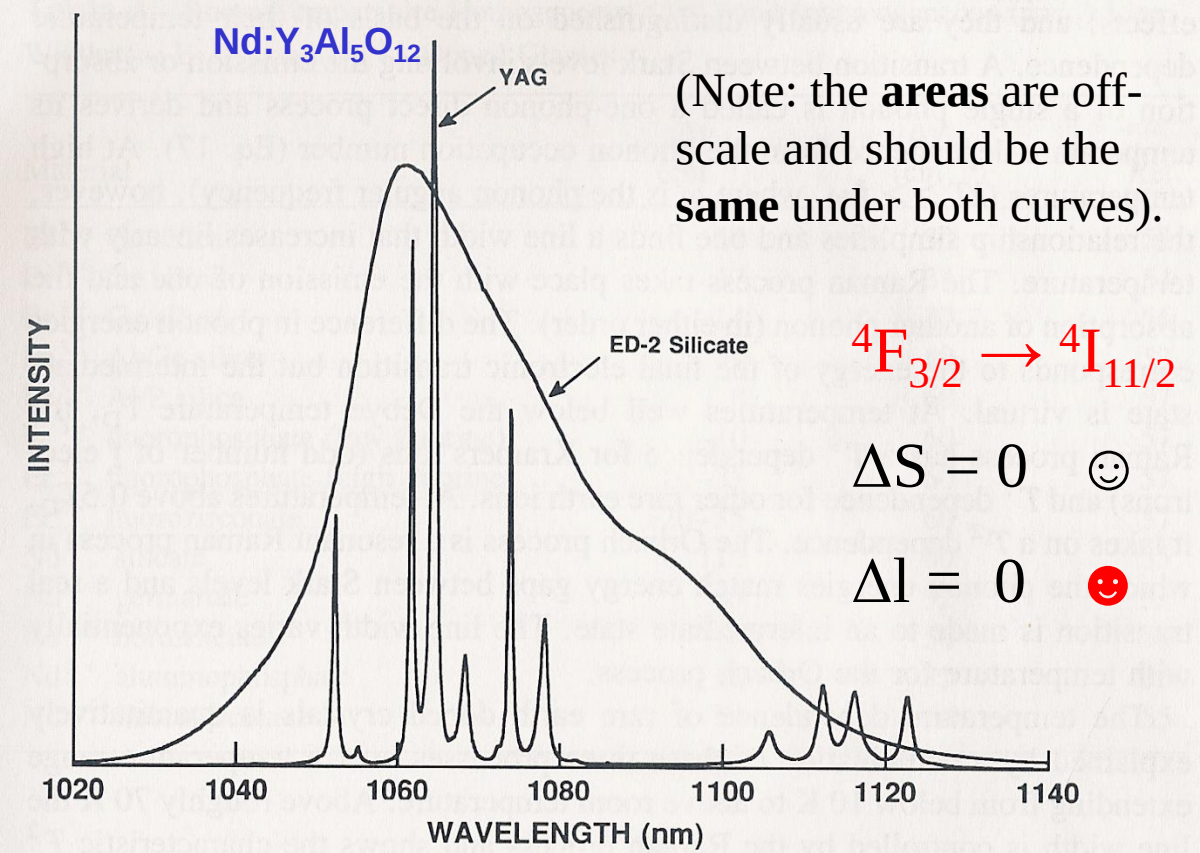
In addition to homogeneous line broadening, **a *glass* host will also cause** the so-called ***inhomogeneous broadening***, due to site-to-site variations in the local atomic structure around the different RE ions present.

Possible RE ion (active) dopants for glassy hosts



Simplified electronic energy level diagrams of three different RE ions and relevant transitions for pumping and emission.

The absorption and emission transitions between the individual Stark components of different J multiplets can usually be observed at room temperature as discrete lines in RE-doped **crystals**, but not in **glasses**. While **crystalline hosts provide high cross sections at nearly discrete wavelengths**, **glass hosts have lower cross sections over a broad range of wavelengths**.



Comparison of the emission spectra for Nd^{3+}-doped YAG and Nd^{3+}-doped ED^{-2}silicate glass.

(Adapted from: *Rare earth doped fiber lasers and amplifiers*, ed. M.J.F. Digonnet, Marcell-Dekker, 1993)

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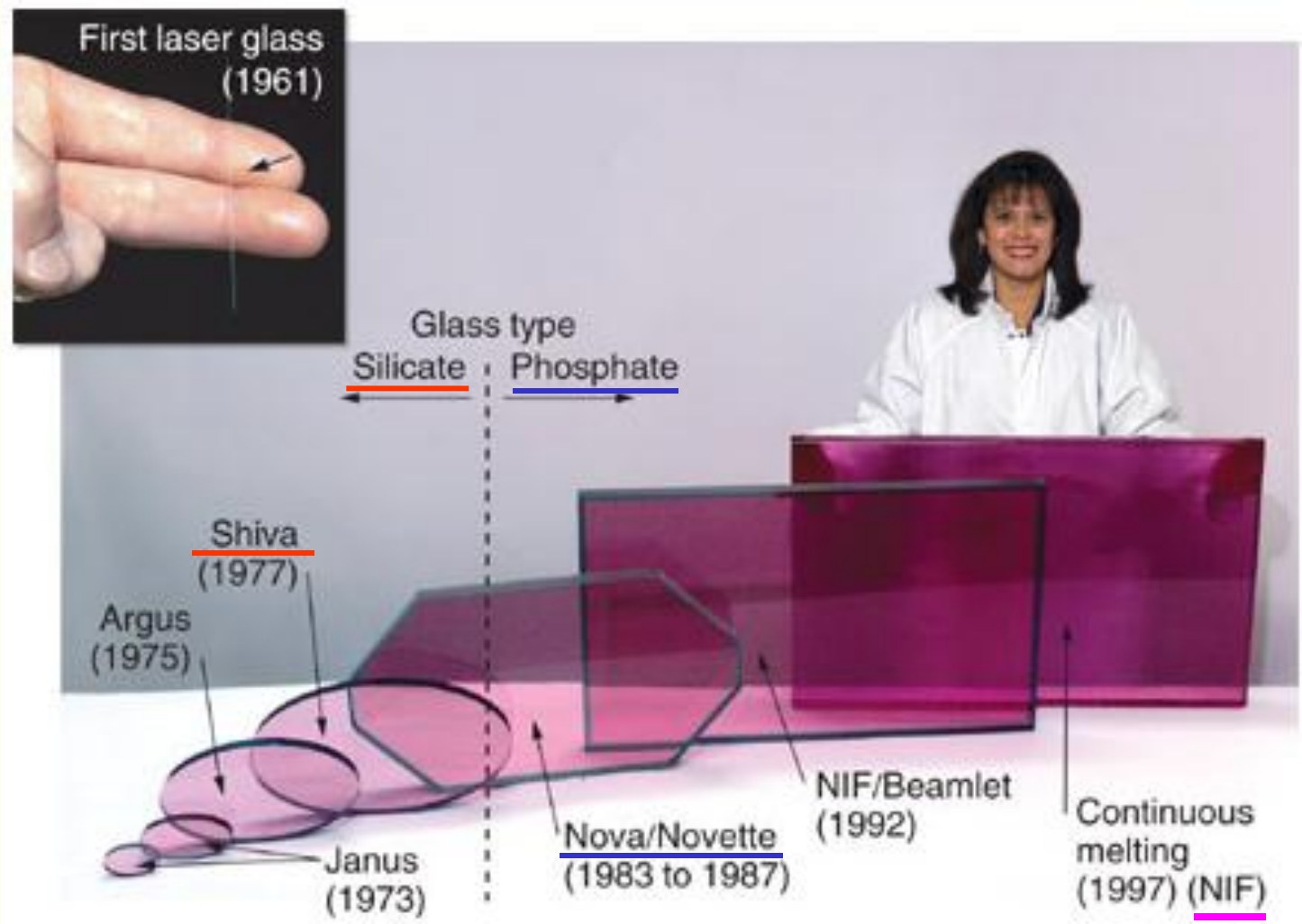
Glass lasers and amplifiers for inertial confinement nuclear fusion

One of the important applications of **Nd:**glass lasers and amplifiers is in the area of inertia confined laser nuclear fusion research. The plasma generated during the fusion process is so hot (**> 10^7 K**) that it cannot come into contact with the surrounding materials. Here, as opposed to the magnetic confinement used in Tokamak test reactors, the confinement is achieved by the target fuel's own inertia.

Directed onto a tiny deuterium-tritium pellet (the source of such fuel is essentially endless), the enormous energy influx evaporates the outer layer of the pellet, producing energetic collisions which drive part of the pellet inward. The inner core is increased a thousandfold in density and its temperature is driven upward to the ignition point for fusion. Accomplishing this in a time interval of 10 ps to 1 ns does not allow the ions to move appreciably because of their own inertia; hence the name inertial confinement.

In the US, these projects have been developed at the Lawrence Livermore National Laboratory (LLNL) in California.

Nd-doped lasers: Nd-doped glass slabs used in extremely powerful lasers for inertial confinement fusion. Pumping can be done with a broadband Xe flashlamp or with a diode laser.



Adapted from: http://en.wikipedia.org/wiki/File:Laser_glass_slabs.jpg (7 Jan 2012)

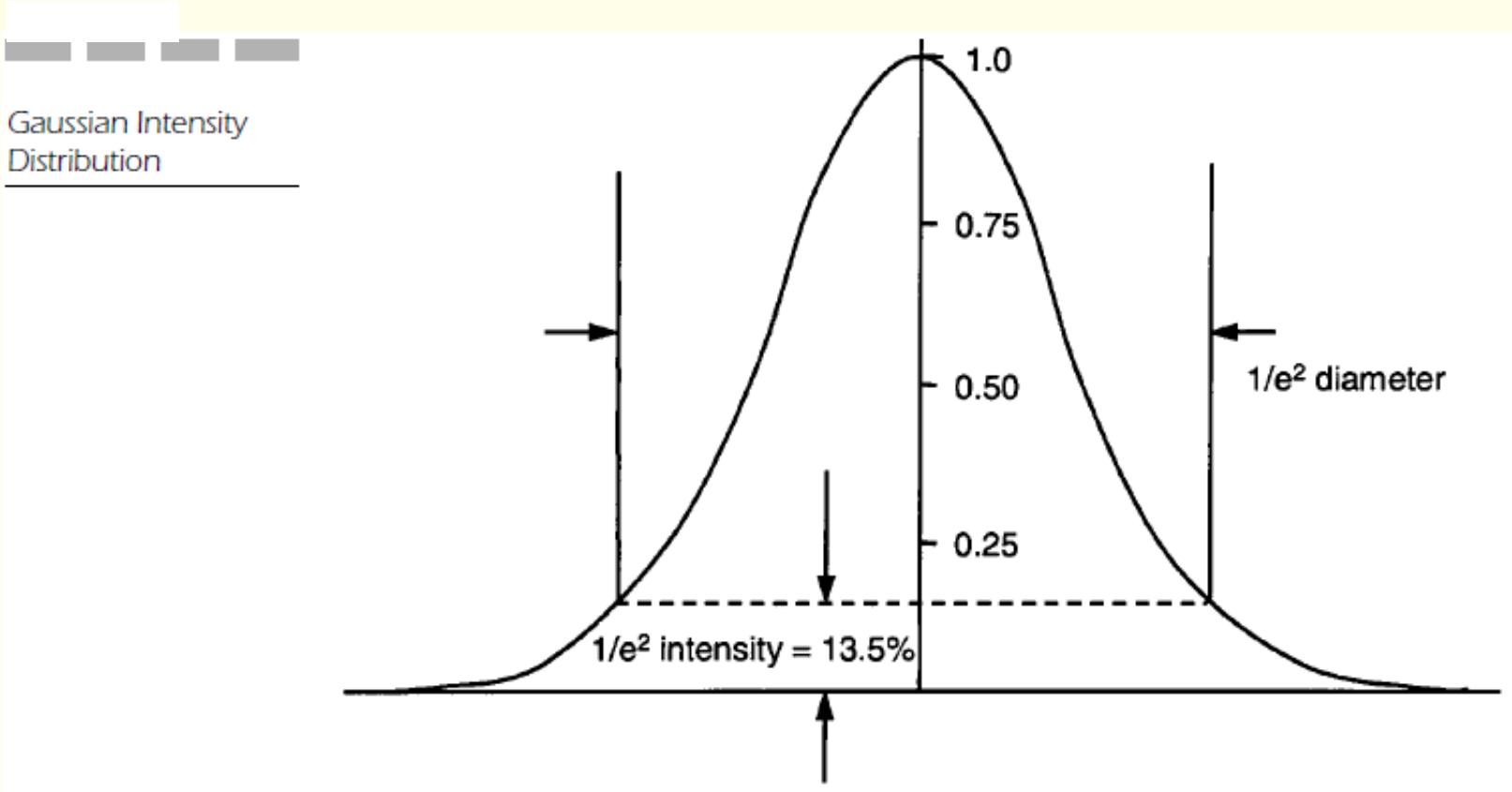
In the first such project, called Shiva and put in operation in 1977, a collection of 20 **Nd:silicate glass** laser beams at 1054 nm were focused to a precise position in a target chamber. The multi-laser device, called Shiva after the multi-armed Hindu god, sought to initiate laser fusion in small microballoons of a deuterium-tritium gas mixture. Each 100 μm pellet supposedly contained the energy equivalent of a barrel of oil. The first generation Shiva system was operated until 1981.

A second, more powerful machine called Nova was built by 1983, offering the possibility of reaching the fusion breakeven point, where usable energy gotten from the nuclear fusion process exceeds the input energy.

Nova is the name given to the second generation laser fusion device at LLNL. It employed lasers ten times more powerful than the Shiva system and it attempted to reach the breakeven point for fusion. Nova made use of 10 **Nd:phosphate glass** lasers at 351 nm (i.e. frequency tripled), focused on a 1 mm diameter target area, dumping 100,000 joules of energy (0.1 MJ) into the target in a nanosecond. Fused silica glass lenses were also part of this laser system.

The switch from silicate (Shiva) to phosphate glasses (Nova) was due mostly to the need to reduce non-linear optic (NLO) effects. The main problem was attributed to the non-linear refractive index, the source of self-focusing and (gaussian) beam distortion.

TEM_{00} mode has Gaussian intensity distribution



Adapted from: http://www.mhprofessional.com/downloads/products/0071472487/0071472487_chap11.pdf

NLO effects

The **intensity dependent refractive index** is generally given as:

$$n = n_o + n_1 \mathbf{E} + n_2 \mathbf{E}^2$$

where n_o is the linear refractive index, n_1 is the **Pockels coefficient** (assumed negligible for glass, which is isotropic) and n_2 is the so-called ***non-linear refractive index*** or Kerr coefficient (from the **optical Kerr effect**). Since the intensity of the electric field of the laser light is equal to the square of its amplitude, for **glass** one can also write:

$$n = n_o + n_2'' \mathbf{I} \qquad (n_2'' \text{ in } cm^2/W)$$

n_o **and** n_2 are usually **directly correlated**, such that low index (n_o) materials like fluoride and phosphate glasses have also low n_2 and are important in high power applications to minimize self-focusing, leading to beam distortion and local damage to the optical glass.

Nd-doped silicate glasses had $n_o \sim 1.56$; Nd-doped phosphate glasses have $n_o \sim 1.50 – 1.53$ for the sodium D-line ($\lambda = 589.3$ nm). Even lower indices are possible with fluoro-phosphate (~ 1.49) and, especially, fluoride glasses like those based on BeF_2 (~ 1.28 – 1.38).

Some of the problems associated with the new, higher power Nova system still included NLO effects, which led to **self-focusing** and **pulse distortion**, but also optical damage of the glass due to heating of very small Pt particles present in the glassy material, which generated a **yellow tint** due to blue absorption near 400 nm, until by 1987 techniques were developed to fabricate Pt-free phosphate laser glass.

Techniques to fabricate Pt-free phosphate laser glass included melting under inert (O_2) or reactive (Cl_2) gases, thus controlling the redox state of the melt in order to dissolve the Pt particles. OH reduction was also achieved, by melting under Cl_2 atmosphere.

To ensure a high degree of glass homogeneity, the refractive index was controlled within $+/- 2 \times 10^{-6}$ (e.g. 1.506700 +/- 0.000002) throughout the large area laser and amplifier elements, by annealing each glass plate for several weeks near T_g (~ 460 – 480 ^oC).

Composition and properties of Nd-doped phosphate laser glasses LHG-8 and LG-770 [14,16,17]

Composition[a] or property (mol %)	LHG-8	LG-770
P_2O_5	55–60	58–62
Al_2O_3	8–12	6–10
K_2O	13–17	20–25
BaO	10–15	–
MgO	–	5–10
Nd_2O_3[b]	0–2	0–2
Other	<2	<2
O/P (± 0.1)	3	3
Optical		
Refractive index[c]		
n_d (587.3 nm)	1.5296	1.5067
n_l (1053 nm)	1.5201	1.4991
Non-linear refractive index		
n_2 (10^{-13} esu)	1.12	1.01
γ (10^{-20} m^2/W)	3.08	2.78
Abbe number (±0.05)	66.5	68.4
Laser		
Emission cross-section (10^{-20} cm^2) (±0.2)	3.6	3.9
Radiative lifetime (zero-Nd) (μs) (±3)	365	372
Judd–Ofelt radiative lifetime (μs) (±10%)	351	349
Emission band width (nm) (±0.1)	26.5	25.4
Thermal		
Thermal conductivity, 90°C (W/mK) (±0.03)	0.58	0.57
Thermal diffusivity (10^{-7} m^2/s)	2.7	2.9
Specific heat, Cp (J/gK) (±0.02)	0.75	0.77
Coeff. thermal expansion, 20–300°C (10^{-7}/K) (±3)	127	134
Glass transition temperature[c], T_g (°C) (±5)	485	460
Mechanical		
Density (g/cm^3)[c] (±0.01)	2.83	2.59
Poisson's ratio (±0.01)	0.26	0.25
Fracture toughness (MPa $m^{1/2}$) (±0.02)	0.51	0.43
Hardness (GPa) (±0.10)	3.43	3.58
Young's modulus (GPa) (±1.0)	50.1	47.3

$[Nd^{3+}]$ ~ 1 at%

(~ 5 x 10^{20} ions/cm^3)

[a] Range in composition values reflect variations due to Nd-doping concentration, melt volatility, and batching variations.
[b] Nd-doping levels typically <2 mol% Nd_2O_3 (<5 × 10^{20} Nd ions/cm^3); the NIF and LMJ use a doping of 4.2 (± 0.1) × 10^{20} ions/cm^3.
[c] Values may vary slightly with Nd-doping level.

Adapted from: J.H. Campbell et al., J. Non-Cryst. Solids 263&264 (2000) 342.

Nd phosphate laser glasses have large stimulated emission cross section, low non-linear refractive index and good physical, chemical and mechanical properties ([Nd] ~ 1 at%).

High power lasers require that the glass active medium have large dimensions, high optical homogeneity and high quantum efficiency. The refractive index difference within a glass blank should be $< 2 \times 10^{-6}$ and the absorption coefficient due to OH groups at ~ 3.5 μm should be < 10 cm^{-1} ([OH] < ~ 100 ppm weight).

When Nova was dismantled to make way for its final upgraded version called National Ignition Facility (NIF), the target chamber was lent to France for temporary use during the development of Laser MegaJoule (LMJ) facility, a system similar to NIF. Work on NIF was not formally completed until 2009. The NIF employs 192 Nd:doped phosphate glass laser beam lines capable of reaching > **1 MJ** of laser energy at 351 nm (equivalent to the energy consumed by 10,000 100-Watt light bulbs in 1 s). Each beamline contains 16 Nd-doped laser glass plates.

National ignition facility (NIF) (Laser Bay 2)

Adapted from: http://www.boston.com/bigpicture/2010/10/the_national_ignition_facility.html

High-power Nd:glass laser facilities for nuclear fusion research

	Starting date	# laser beams	lasing λ	Glass	Energy
Shiva	1977	20	1054 nm	Nd:silicate	10 kJ
Nova	1983	10	351 nm	Nd:phosphate	100 kJ
NIF	2009	192	351 nm	Nd:phosphate	1.8 MJ

Q.

References:

http://hyperphysics.phy-astr.gsu.edu/hbase/nucene/finert.html (6 Jan 2012)

C.M. Baldwin, R.M. Almeida and J.D. Mackenzie, J. Non-Cryst. Solids 43 (1981) 309.

M. J. Weber and R. M. Almeida, J. Non-Cryst. Solids 43 (1981) 99.

Jiang Yasi et al., J. Non-Cryst. Solids 80 (1986) 623.

M.J. Weber, J. Non-Cryst. Solids 123 (1990) 208.

J.H. Campbell et al., J. Non-Cryst. Solids 263&264 (2000) 342.