

Chapter 5

Fluoride Laser Crystals: YLiF₄ (YLF)

Fluoride crystals are among the most important hosts for laser materials because of their special optical properties. Of these, LiYF₄ (YLF) is one of the most common rare-earth-doped laser materials, with a variety of efficient mid-IR laser lines from the UV (Ce³⁺:YLF)¹ to mid-IR range. Generally, YLF has good optical properties with high transparency throughout the emission spectrum of the conventional sources used for pumping solid state lasers. YLF does not show UV damage, and it has lower nonradiative decay rates for processes occurring between electronic levels participating in the pumping and lasing process. YLF also has a low, two-photon absorption coefficient. Because of its low nonradiative rates, the material can be used for cascade emission² between intermediate levels as well as an up-converter, as will be discussed later.

YLF is also a good medium for mode locking at 1047 or 1053 nm and 1.313 μm as a result of its natural birefringence and low thermal lensing. Mode-locked pulses from YLF are shorter thanks to its broader linewidth, both for the 1047/1053-nm and 1.313- μm emission peaks. The crystallographic structure of LiYF₄ (or YLF) is the same as CaWO₄, which was developed years ago as a potential laser material.³ However, when a trivalent rare-earth material substitutes for the Ca²⁺ ion, charge compensation is necessary. However, the process of charge compensation may result in inhomogeneities in the crystal and is a source of disordered crystal structure. No charge compensation is necessary with YLF throughout the doping process, since the trivalent rare-earth-ion substitutes for the Y³⁺ ion. As a result, a single undisturbed site exists. The crystal has tetragonal symmetry; the important optical and physical properties are shown in Tables 4.1 and 4.2. Figure 5.1 shows a schematic energy-level diagram of those levels participating in the lasing process in Nd:YLF.

5.1 Thermal and Mechanical Properties of YLF

Thermal and mechanical properties of $\alpha\beta\text{Ho:YLF}$ were measured by Chicklis et al.⁴ See also Tables 4.1 and 4.2. The authors described and analyzed the esti-

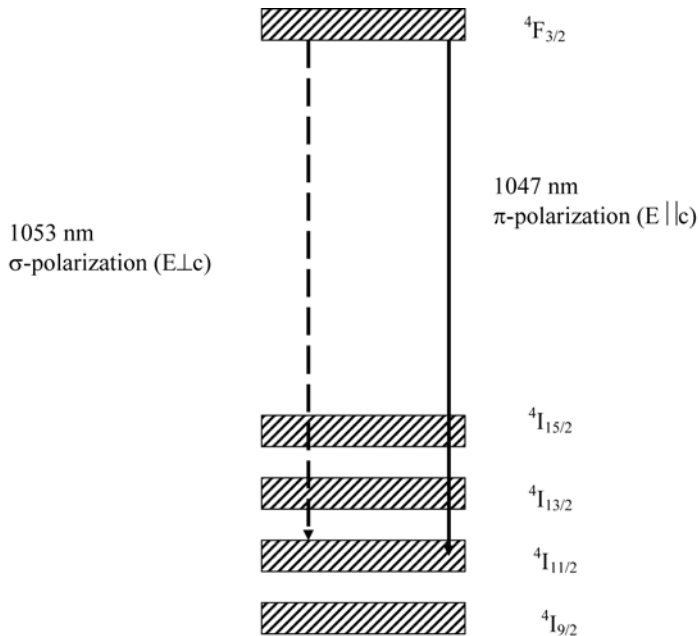


Figure 5.1 Schematic energy-level diagram of electronic levels participating in the lasing process in Nd:YLF. Broken line: 1053 nm (σ polarization, $E \perp c$); full line: 1047 nm (π polarization, $E \parallel c$).

mated power loading at fracture. The following sections explain some of the concepts used in thermal-load analysis.

5.1.1 Estimate of thermal load at fracture

Unused energy deposited in a laser crystal is converted into heat. Two main reasons account for heat accumulation:

1. The quantum gap between the absorbed pump light and the lasing energies, e.g., the energy difference between absorbed pump light and fluorescence energies.
2. An inefficient pumping source. The spectral distribution of the pump light is broad relative to the narrow absorption lines of the lasing ion. The undesired pumping energy is absorbed by the host and is transformed into heat.

The heat generated owing to the above mechanisms and the radial heat flow resulting from the cooling process of the laser rod surface together cause the thermal effects in a laser rod.

In order to calculate the temperature distribution in a laser rod, these assumptions are made:

- The heat generates uniformly in the laser rod. The cooling process is uniform along the laser rod surface.
- The laser rod is an infinitely long cylindrical rod of radius r_0 .
- The heat flow is radial.
- Small end effects occur.

The cross-sectional geometries generally used for lasers are cylindrical and heat removal is carried out through the circumferential surface of the cylinder. Therefore, radial temperature distribution has a parabolic profile, which is given by

$$T(r) = T(r_0) + \frac{1}{4K} Q(r_0^2 - r^2), \quad (5.1)$$

where $T(r)$ is the temperature at a distance r from the rod axis, $T(r_0)$ is the temperature at the rod surface, r_0 is the rod radius, K is the thermal conductivity, and Q is the heat per unit volume dissipated in the rod. As seen from Eq. (5.1), the radial temperature distribution inside a laser rod has a parabolic profile. Therefore, temperature gradients are formed inside the cylindrical laser rod, and these gradients lead to the following effects:

- Mechanical stresses inside the laser rod.
- Photoelastic effects and a change in the refraction index.
- Thermal lensing owing to changes in the refraction index.
- End-face curvature resulting from mechanical stresses relating to temperature gradients.
- Thermal-induced birefringence.
- Depolarization of polarized light.

The thermal load and the mechanical stresses formed inside the laser rod can lead to a rod fracture. The value of the thermal load at the fracture of a uniformly heated laser rod, cooled at the surface, is an important parameter in estimating the average output power available from a given host.

A stress distribution gradient is accompanied by a temperature gradient,⁴

$$\sigma_r(r) = \frac{2\alpha E}{(1 - \mu)16k} Q(r^2 - r_0^2), \quad (5.2)$$

$$\sigma_\theta(r) = \frac{2\alpha E}{(1 - \mu)16k} Q(3r^2 - r_0^2), \quad (5.3)$$

$$\sigma_z(r) = \frac{2\alpha E}{(1 - \mu)16k} Q(4r^2 - 2r_0^2), \quad (5.4)$$

where the parameters appearing in Eqs. (5.2) to (5.4) are defined as $\sigma_r(r)$, the radial stress at distance r ; $\sigma_\theta(r)$, the tangential stress at distance r ; $\sigma_z(r)$, the axial

stress at distance r ; μ is Poisson's ratio; E is Young's modulus; and α is the thermal expansion coefficient. See Fig. 5.2 for a demonstration of these parameters. From these expressions, it is seen that the radial component of the stress disappears at the rod's surface while the tangential and axial components do not vanish. Therefore, the rod is under tension, which may cause it to crack. The value of the power loading per unit length for YLF is 11 W/cm, while for YAG it is 60 W/cm.

Perhaps one of the most important factors affecting the laser performance of YLF crystal is its refractive index. YLF has a negative change of refractive index with temperature: $dn/dT < 0$, where n is the refraction index and T is the crystal temperature. This minimizes the thermal lensing effects in the crystal and improves the fraction of the available power with the TEM₀₀ mode, improving the beam quality. Assume an absorbing medium heated by radiation. If its temperature is increased by ΔT at a certain point owing to heat formation, the refractive index upon irradiation is given

$$n(\Delta T) = n(0) + \left(\frac{dn}{dT} \right) \cdot \Delta T, \quad (5.5)$$

where $n(0)$ is the refractive index at any point without pumping the absorbing medium and dn/dT is the dependence of the refraction index on temperature.

Assuming also a cylindrical lasing medium cooled through its surface, the temperature will have maximum value along the axis and minimum value at the surface, and it will drop gradually from the center to the peripheral region. If the condition $dn/dT > 0$ is fulfilled, the axis region will be optically denser than the surface [according to Eq. (5.5)], and the radiation along the rod axis will be focused since rays will be deflected into the region containing a higher value of n . In the case of $dn/dT < 0$, the periphery will be denser than the axis, and rays propagating along the rod axis will be defocused. In the case of $dn/dT > 0$, the active element is identical to a convergent lens, and in the case of $dn/dT < 0$, it is identical to a divergent lens. The phenomenon in which the laser element acts as a lens is called *thermal lensing*. The radial temperature gradient causes a refractive index gradient along the radius of the crystal, giving the laser rod the characteristics of a graded index (GRIN) lens. Another contribution to thermal lensing is

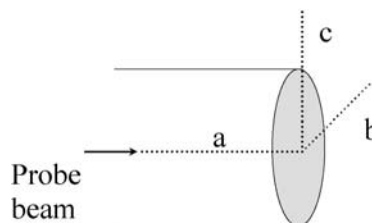


Figure 5.2 Crystallographic directions of the laser rod in the thermal lensing experiment performed by H. Vanherzeele. The σ polarization at 1053 nm is the ordinary polarization ($E \perp c$); π polarization at 1047 nm is the extraordinary polarization ($E \parallel c$).

the effect of the crystal faces bending under strong thermomechanical stresses. Koechner⁵ analyzed the thermal lensing effects in a Nd:YAG laser rod theoretically and experimentally under flashlamp pumping and external probe laser. The expression for the total focal length obtained by Koechner contains the GRIN lens contribution as well as elasto-optical terms, which contribute to the end-face curvature,

$$f = \frac{KA}{P_{\text{in}}\eta} \left[\frac{1}{2} \frac{dn}{dT} + \alpha C_{r,\phi} n_0^3 + \frac{\alpha l_0 (n_0 - 1)}{L} \right], \quad (5.6)$$

where A is the rod cross section, K is the thermal conductivity of the laser rod, P_{in} is the input incident pump power, η is the heat dissipation factor ($Q = \eta P_{\text{in}}$), n_0 is the refraction index at the center of the rod, α is the thermal expansion coefficient, l_0 is the depth of the end effect (the length up to the point where no significant contribution to surface bending occurs), L is the length of the laser rod, and $C_{r,\phi}$ is a functional representation of electro-optical coefficients with the radial and tangential components of the orthogonal polarized light.

The combination of the two effects (radial temperature gradients and crystal-face bending) can be approximated by a thin lens located at the end of the laser rod, with dioptric power of⁶

$$D_{\text{R}} = D_{\text{E}} + D_{\text{T}}, \quad (5.7)$$

where D_{R} is the dioptric power of the overall thermal lensing, D_{T} is the GRIN lens effect, and D_{E} is the bending end effect of the laser rod. The depth of the end effect and the radius of the rod can be assumed to be roughly equal, e.g., $r_0 \approx l_0$. Therefore, the two contributions to thermal lensing, the GRIN lens, D_{T} , and the end effect D_{E} , are given respectively as

$$D_{\text{T}} = \frac{QL}{K} \left(\frac{1}{2} \frac{dn}{dT} + \alpha C_{r,\phi} n^3 \right) \quad (5.8)$$

and

$$D_{\text{E}} = \frac{1}{K} [\alpha Q r_0 (n - 1)], \quad (5.9)$$

where Q is the heat generated in the laser rod per unit volume, K is the thermal conductivity, L is the length of the laser rod, T is the rod temperature, α is the thermal expansion coefficient, n is the refraction index, and r_0 is the radius of the rod. The end effect contribution to thermal lensing for homogeneous pumping was estimated to be about 20%; it is independent of the absorption coefficient. In the case of inhomogeneous pumping, such as longitudinal diode pumping, the actual magnitude of the absorption coefficient (or doping level), the pump spot

Chapter 6

Photophysics of Solid State Laser Materials

6.1 Properties of the Lasing Ion

Light emission occurs as a result of interaction between light and matter. Let us assume a two-level atom with levels 1 (ground state) and 2 (excited state). The energies of the ground and excited states are E_1 and E_2 , respectively, and the energy difference is therefore given by the difference $\Delta E_{12} = E_2 - E_1$. When light with photons of energy equal to this difference is absorbed by the atomic or molecular system, an electron will be excited from level 1 to level 2. The energy of the photons is given by $\Delta E_{12} = E_{12} = h\nu_{12}$ (h is Planck's constant and ν_{12} is the frequency of light resulting from the level 1 \rightarrow 2 transition). This energy is absorbed by an atom or a molecule that has energy levels, separated by ΔE , where these energy levels are the ground and the excited states. This kind of "quantum jump" of an electron between two states occurs in atomic systems between electronic levels; it can be extended to molecular systems, where vibrational and translational energy levels participate in the quantum jump. The interaction between light and matter involves the transition of electrons between different states. This interaction results in the absorption of photons (stimulated absorption) as well as spontaneous and stimulated emission. These processes can be described using Einstein's A and B coefficients, as will be described in the next paragraphs.

6.1.1 Absorption

The absorption of light by an object is a fundamental phenomenon in nature. Visible objects scatter the light that falls on them. However, colored objects absorb light at certain wavelengths (or frequencies), while scattering or transmitting other frequencies. For example, an object that will absorb all the frequencies in the visible range will appear black. A green object will absorb light throughout the visible spectral regime except that wavelength which defines the green color.

Let us assume that we have a nondegenerate two-level atomic system, with ground and excited states levels 1 and 2. Initially, the atom is in the ground state 1.

When an external electromagnetic field with a frequency of $h\nu_{12} = E_2 - E_1$ is applied to the atomic system, it is probable that the atom will undergo a transition from level 1 to level 2. This process is termed absorption. If N_1 is the volume density of the atoms in the ground state, the temporal rate of change of the density is given by the equation

$$\frac{dN_1}{dt} = W_{12}N_1, \quad (6.1)$$

where W_{12} is the absorption rate, which is related to the photon flux, I , by

$$W_{12} = \sigma_{12}I, \quad (6.2)$$

where σ_{12} is the absorption cross section. The intensity of a monochromatic light, I_λ , which propagates a distance Δz in an absorbing medium, is given by

$$I_\lambda(z) = I_\lambda(0) \exp[-\alpha(\lambda) \cdot \Delta z]. \quad (6.3)$$

$I_\lambda(0)$ is the initial light intensity at the entrance of the absorbing medium, $z = 0$, while $I_\lambda(z)$ is the final intensity after the light has propagated a distance Δz inside the absorbing material. The quantity α introduced in Eq. (6.3) is called the *absorption coefficient*. This equation is valid under thermal equilibrium conditions, $N_1g_1 > N_2g_2$, where g_1 and g_2 are the degeneracies of levels 1 and 2, respectively. The inverse magnitude, α^{-1} , measures the optical path for which the light intensity is decreased by a factor of e^{-1} as a result of absorption only by the medium. If it is assumed that the density of the absorbing medium is N atoms per unit volume, the absorption coefficient is related to the absorption cross section as

$$\alpha(\lambda) = N\sigma_{12}. \quad (6.4)$$

It should be noted that the absorption coefficient in this model can be obtained by two methods: The first is based on the imaginary part of the refraction index, the second on the rate at which an atom absorbs energy from an external field. In both cases, the energy absorption is described by a simple classical electron oscillator model, or Lorentz model, of the atom. The Lorentz model was developed before atomic structure was known. The model assumes that in the absence of external forces, the electron in the atom is in an equilibrium position. When an external electromagnetic field with a driving frequency ω is applied to the atomic system, the electron will be displaced from the equilibrium position and will oscillate back and forth, owing to elastic forces, at a natural frequency of ω_0 .

The electron motion relative to the nucleus can be described by the rate equation

$$m \frac{d^2x}{dt^2} = e \cdot E(R, t) - C \cdot x, \quad (6.5)$$

where m is the reduced mass of the electron-nucleus system, R is the center-of-mass coordinate, E is the electromagnetic field, and $C \cdot x$ describes the oscillatory motion of the electron (C is the spring constant). This equation can also be written in terms of natural frequency, ω_0 , as

$$\left(\frac{d^2}{dt^2} + \omega_0^2 \right) x = \frac{e}{m} E(R, t), \quad (6.6)$$

where the electron natural frequency ω_0 is defined as $\omega_0 = \sqrt{C/m}$.

When a periodic external field is applied, the oscillatory motion of the bound electron is described in terms of a driven harmonic oscillator. If the electron is displaced by Δx from its equilibrium state, the dipole moment of the system is $P = e \cdot \Delta x$ (where e is the electron charge). The external field provides energy that maintains the oscillation at a frequency of the applied field, ω .

In the case of a damped oscillator, driven by external electric field, the equation of motion of the oscillating electron is

$$\frac{d^2 x}{dt^2} + 2\gamma \frac{dx}{dt} + \omega_0^2 x = \vec{\epsilon} \frac{e}{m} E_0 \cos(\omega t - kz). \quad (6.7)$$

The term γ , introduced in Eq. (6.7) pertaining to the electron oscillator model, is the damping parameter of the average harmonic displacement of the electron, x is the average electron displacement, ω_0 is the natural oscillation frequency, e is the electron charge, m is the reduced mass of the electron-nucleus system, E_0 is the wave amplitude of the induced external field, and $\vec{\epsilon}$ is a unit vector. It is assumed that the electric field in Eq. (6.7) is a plane wave that propagates along the z axis, with a wave vector k , which relates to the wavelength λ by $k = 2\pi/\lambda$. A damping parameter is also defined as $\Gamma = 2\gamma$.

The solution to Eq. (6.7) can be described by two components, one in phase and a second that is out of phase with the driving force,

$$x(t) = A \sin \omega t + B \cos \omega t, \quad (6.8)$$

where the coefficients A and B are given by

$$A = \frac{e \cdot E_0}{m} \frac{\Gamma \omega}{[(\omega_0^2 - \omega^2)^2 + (\Gamma \omega)^2]} = A_{\text{ab}} \quad (6.9)$$

and

$$B = \frac{e \cdot E_0}{m} \frac{(\omega_0^2 - \omega^2)}{[(\omega_0^2 - \omega^2)^2 + (\Gamma \omega)^2]} = A_{\text{disp}}. \quad (6.10)$$

The term A_{ab} is called the absorptive amplitude, while A_{disp} is called the dispersive amplitude. The averaged input absorption power is a consequence of the term

$A_{ab} \sin \omega t$. The term A_{disp} is averaged out to 0 over one oscillation cycle. It was found that the time-averaged, absorbed power over the oscillation period is given after some mathematical manipulation by

$$P_{av} = \frac{1}{2} (e \cdot E_0 \omega A_{ab}) = \frac{1}{2} m \alpha_0^2 \cdot \frac{\omega^2 \Gamma}{(\omega_0^2 - \omega^2)^2 + (\omega^2 \Gamma^2)} \quad (6.11)$$

and

$$\alpha_0 = e \cdot \frac{E_0}{m}. \quad (6.12)$$

The energy transfer from the driving field to the oscillatory system will be maximized at resonance, when $\omega = \omega_0$. The energy absorption by the system will be at maximum when the frequency of the driving force coincides with the natural frequency of the oscillating system. At resonance,

$$P_0 = \frac{1}{2} \frac{m \alpha_0^2}{\Gamma} \quad (6.13)$$

is obtained. Combining Eqs. (6.11) and (6.13) produces

$$P_{av} = P_0 \frac{\Gamma^2 \omega^2}{(\omega_0^2 - \omega^2)^2 + \Gamma^2 \omega^2}. \quad (6.14)$$

The absorptive amplitude and power [Eqs. (6.9) and (6.14), respectively] depend on the quantity C that is defined as

$$C = (\omega_0^2 - \omega^2)^2 + \Gamma^2 \omega^2 = (\omega_0 - \omega)^2 (\omega_0 + \omega)^2 + \Gamma^2 \omega^2. \quad (6.15)$$

This quantity (C) is changed rapidly at resonance, when $\omega_0 = \omega$, or near resonance, where ω is within the range $\omega - 10\Gamma < \omega < \omega_0 + 10\Gamma$. On the other hand, the term with ω alone contributes a much slower variation in A . In near-resonance and weak damping conditions such as $\Gamma \ll \omega_0$, it is assumed that ω is equal to ω_0 in the expression C except in the factor $(\omega_0 - \omega)^2$ of the first term. Then C may be approximated as

$$C = (\omega_0 - \omega)^2 (\omega_0 + \omega_0)^2 + \Gamma^2 \omega_0^2 = 4\omega_0^2 \left[(\omega_0 - \omega)^2 + \left(\frac{1}{2} \Gamma \right)^2 \right]. \quad (6.16)$$

By inserting the last approximation for C into Eqs. (6.9) and (6.14), it can be observed that both the *absorptive amplitude* and the *averaged absorbed power* are proportional to the factor

$$\frac{[(1/2)\Gamma]^2}{(\omega_0 - \omega)^2 + [(1/2)\Gamma]^2}. \quad (6.17)$$

This frequency-dependent factor determines the lineshape of the absorption amplitude or the absorbed power. The origin of this lineshape, called a Lorentzian lineshape, can be understood if the statistical nature of the atomic system is considered. Therefore, the average time between collisions affects the temporal behavior of the driving EM field and should be included. These effects are included in the lineshape function g , which has the form

$$g(\omega - \omega_0) = \frac{T_2}{\pi} \frac{1}{[1 + (\omega_0 - \omega)^2 T_2^2]}, \quad (6.18)$$

where T_2 is the average time between two collisions. The lineshape $g(\omega - \omega_0)$ defines a Lorentzian lineshape. If one plots $g(\Delta\omega)$ vs. $\Delta\omega$, where $\Delta\omega = \omega - \omega_0$, one obtains a lineshape with a maximum at T_2/π and a FWHM intensity ($\Delta\omega_0$) with a value of $2/T_2$. If the Lorentz statistical model for atomic collisions is introduced, the result is a relation of the average time between collisions (T_2) to the damping parameter γ by

$$T_2 = \frac{1}{\gamma}. \quad (6.19)$$

The Lorentzian lineshape is therefore a result of collision broadening between atoms. Collision effects are statistically averaged, so physical quantities associated with a Lorentzian lineshape are also averaged. The collision broadening dephases, i.e., the phase of the electron's oscillation before collision is uncorrelated to the phase after the collision. As a result of the collisions, the average displacement of the bound electron decays at a rate equal to the collision rate. The dephasing effect associated with atomic collisions appears as a damping parameter in Eqs. (6.7) and (6.20).

Equation (6.7) can be rewritten in terms of average electron displacement,

$$\frac{d^2}{dt^2}\langle x \rangle + 2\gamma \frac{d}{dt}\langle x \rangle + \omega_0^2 \langle x \rangle = \bar{\mathbf{e}} \frac{e}{m} E_0 \cos(\omega t - kz), \quad (6.20)$$

where the parameters appearing in Eq. (6.20) have already been defined in Eq. (6.7).

The effect of the damping parameter γ , which includes the meaning of collision rate, should be emphasized here. The friction parameter or the collision rate dephases the electron oscillations so that the phase of the oscillatory motion of the electrons after the collision is completely uncorrelated to the precollision phase. This dephasing results from collisions between electrons, which leads to a decay of the average electron displacement. When no inelastic collisions occur, the oscillatory motion satisfies the Newton Equation [Eq. (6.6)],

$$\frac{d^2 x}{dt^2} + \omega_0^2 x = \varepsilon \cdot \frac{e}{m} E_0 \cos(\omega t - kz), \quad (6.21)$$