EEE 6503

A Report on Fast-Pulse Production & Nonlinear Optics

Prepared by: A F. M. Saniul Haq Student ID: 0412062205

Submitted To: Dr. Md. Nasim Ahmed Dewan Associate Professor, Department of EEE, BUET, Dhaka-1000.

Chapter 1

Fast-Pulse Production

Pulsed solid-state lasers are among the most important and widely deployed commercial laser for applications such as marking, cutting, drilling, range finders, and retinal surgery. For many of these applications, the shorter the pulse, the better. Fast, powerful pulses tend to ablate material quickly without heating surrounding material or tissue. A CW laser beam or one with a long pulse would tend to heat the material surrounding the target area and either damage the substrate or alter the resistor in an uncontrolled way by changing the properties of the resistor film. For this reason a technique called Q-switching is used to produce very short pulses for these types of applications.

1.1 CONCEPT OF Q-SWITCHING

The most simplest method to produce a pulse laser is to to switch the gain of the medium on and off by switching pumping energy on and off simultaneously. When pump energy is sufficient to allow laser gain to exceed the threshold, an output beam appears. The problem with this scheme is that the output pulses will be quite rounded, since there is a delay as population inversion and hence gain builds in the laser; this also sets limits on the pulse length and repetition rate for the laser.

In a Q-switching technique, the laser output is switched by controlling loss within the laser cavity as outlined in Figure 1.1. Q-switching is loss switching in which a loss is inserted into the cavity, thus spoiling it for laser action. In the simplest manner, a Q-switch can be though of as an optical gate blocking the optical path to one cavity mirror and hence causing laser action to cease. In reality, it is not necessary to block the optical path completely. Simply inserting a loss high enough to raise the lasing threshold beyond the maximum gain of the laser is sufficient. When the Q-switch is on, it blocks the intracavity beam. This state is called a low-Q state, meaning that the quality factor, or Q, of the cavity (which measures the ability of a laser cavity to act as a resonator) is ruined. With the switch off, losses in the cavity are reduced, and the cavity is resonanta high-Q state. This technique allows energy to be stored in the laser medium during the low-Q state and released in a single massive pulse. The peak power of the pulse is much larger than is possible with gain switching, which for a CW laser, yields a peak power equal to the CW output power of the laser.



Figure 1.1: Q-switching a LASER.

1.2 INTRACAVITY SWITCHES

A Q-switch consists of a mechanism that spoils the resonance of the laser cavity. There are a number of ways to accomplish this, from altering the alignment of a cavity mirror mechanically (e.g., by a rotating mirror), insertion of an optical switch within the cavity of the laser itself (e.g., an EO or AO modulator), or a saturable dye switch within the cavity. These methods are outlined in Figure 1.2, in which each type of Q-switch is shown within in a solid-state laser cavity.



Figure 1.2: Q-switching methods

A Rotating Mirror

A rotating mirror, the simplest method, is rarely used except in a few older military rangefinders that use ruby rods. A high-speed motor drives the mirror, which, once per revolution, aligns with the other cavity mirror. Just prior to this alignment, the optical pump (usually, a flashlamp) is fired to ensure that a massive population inversion has built up before the cavity is aligned.

EO and AO Modulator

Electrooptic and acoustooptic modulators work quite literally as optical switches, allowing intracavity light to pass only when the switch is open. These are controllable and allow the laser to be fired at the operators command, as required by many experiments.

Saturable Dye

Saturable dye switches are simply a cell filled with organic dye (similar to the dye used in a dye laser) placed inside the laser cavity. These work by absorbing intracavity radiation, inducing a large loss in the cavity, until the dye becomes saturated or bleached, at which point it cannot absorb more laser energy and the remaining light passes through the cell to be amplified in the laser.

1.3 ENERGY STORAGE IN LASER MEDIA

The entire concept of Q-switching relies on the fact that energy can be stored in the lasing medium itself in the form of an excited atomic population at the ULL. The very definition of cavity quality factor (or Q factor) basically defines the situation:

$$Q = 2\pi \times \frac{\text{energy stored in the cavity}}{\text{energy lost per cycle}}$$
(1.1)

A large Q factor represents a low-loss resonator that can store a large amount of energy. In Q-switching the Q of the cavity is spoiled (the Q factor is purposely made low), so that it is not resonant and hence lasing is not possible. Energy storage takes place, but rather than within the cavity as for optical energy, energy is stored in the atomic population.

In a Q-switched laser the laser medium itself is used as a sort of capacitor, storing energy gradually and releasing it in a single burst. Not surprisingly, the capacity of the medium to store energy depends on the lifetime of the upper lasing level (ULL). A long lifetime implies that the lasing species can absorb energy over a long period without losing it to spontaneous emission; hence it has a large storage capacity and is a good candidate for Q-switching.

The primary limit on this inversion is the lifetime of the upper lasing level, which serves to deplete the upper level through spontaneous emission. With continuous pumping the inversion builds until reaching a level equal to the rate of pumping times the lifetime of the upper level. The population rate equation during this interval when the cavity is blocked becomes

$$r_{\rm inversion} = r_{\rm pumping} - \frac{\Delta N(t)}{\tau_{\rm ULL}}$$
 (1.2)

where $r_{inversion}$ is the rate at which the inversion builds, $r_{pumping}$ the rate at which the laser is pumped, and $\Delta N(t)$ the population difference at any time t. The solution to this equation at any time t becomes

$$\Delta N(t) = (r_{\text{inversion}} \tau_{\text{ULL}}) \left[1 - exp\left(-\frac{t}{\tau_{\text{ULL}}} \right) \right]$$
(1.3)

The solution to this equation is plotted in Figure 7.3.1, which depicts how the population builds in time. It is clear that in such a Q-switched laser it is not productive to pump the laser medium

with energy beyond two or three times the lifetime of the upper lasing level. After this period the population inversion does not really grow much but rather, slowly approaches (and never reaches) the maximum level. Implications are that any laser medium may be Q-switched, but



Figure 1.3: Energy storage in Q-switched media.

it should be evident that the lifetime of the upper lasing level (ULL) must be much longer than the opening time for the Q-switch. For most gas lasers with ULL lifetimes of under a 1 ns, Qswitching simply will not work, since no practical switch exists that can open in considerably less than this time. One of the few exceptions is the carbon dioxide gas laser, which has an exceptionally long ULL for a gas laser.

Solid-state lasers, on the other hand, invariably feature long ULL lifetimes. The ruby laser, for example, has a lifetime of 3 ms, and the YAG laser has a lifetime of 1.2 ms. These are ideal candidates for Q-switching, which is a standard option on most solid-state lasers.

1.4 PULSE POWER AND ENERGY

We begin a mathematical analysis of a Q-switched laser by considering the number of coherent photons inside the cavity (per unit volume) at any given time, denoted n. More specifically, we

consider the rate at which the number of photons increases or decreases in the cavity as

$$\frac{dn}{dt} = \frac{n}{\tau_c} + \Delta N W_{\text{pump}} \tag{1.4}$$

where τ_c is the lifetime of a photon in the cavity, ΔN is the population inversion at any given time *t*, and W_{pump} is the pumping probability.

The equation can be simplified by using

$$W_{\text{pump}} = \frac{n}{\Delta N_{th} \tau_c} \tag{1.5}$$

So, Equation 1.4 bocomes,

$$\frac{dn}{dt} = \frac{n}{\tau_c} + \frac{\Delta Nn}{\Delta N_{th}\tau_c}$$
(1.6)

$$=\frac{n}{\tau_c}\left(1+\frac{\Delta N}{\Delta N_{th}}\right) \tag{1.7}$$

In a three-level laser, for example, the rate equation for the ULL becomes

$$\frac{dN_2}{dt} = \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{21}} - W\Delta N \tag{1.8}$$

where the first term represents input from the pump level; the second term, spontaneous decay. We may develop an exceedingly simply expression by making a few assumptions: namely, that the Q-switch pulse occurs so quickly that the effects of decay from the pump level (the first term) as well as the effects of spontaneous emission (the second term) are quite negligible during the time the actual pulse is generated. We also note that for a threelevel laser a loss of one atom from the ULL results in a gain of one atom in the LLL, so the total change in the difference (ΔN) is 2. The answer is then simply

$$\frac{d\Delta N}{dt} = -2W\Delta N \tag{1.9}$$

Further substitution of W yields,

$$\frac{d\Delta N}{dt} = -2\frac{n}{\tau_c}\frac{\Delta N}{\Delta N_{th}}$$
(1.10)

The expression for the change in the number of coherent photons with respect to the change in population inversion:

$$\frac{dn}{d\Delta N} = \frac{\frac{\Delta N}{\Delta N_{th}} - 1}{-2\frac{\Delta N}{\Delta N_{th}}}$$
(1.11)

$$=\frac{1}{2}\Big(-1+\frac{\Delta N_{th}}{\Delta N}\Big) \tag{1.12}$$

This differential equation may be mathematically integrated to yield an answer in terms of the number of coherent photons:

$$n = \frac{1}{2}\Delta N_{th} \ln \Delta N - \frac{1}{2}\Delta N + k \tag{1.13}$$

where k is a constant, which is calculated using initial condition. Replacing k, the final form of the equation,

$$n = \frac{1}{2} \Delta N_{th} \ln \frac{\Delta N}{\Delta N_{initial}} - \frac{1}{2} \left(\Delta N - \Delta N_{initial} \right)$$
(1.14)

We know the volume of the cavity and the energy of each photon ($h\nu$), so that the output power for the Q-switched pulse can be computed as

 $P_{output} = \text{OC Transmission} \times \text{energy per photon} \times \text{number of photon} \times \text{cavity loss per line}$ (1.15)

By replacing all the values, we get,

$$P_{output} = \left(1 - R_{OC}\right)h\nu nV\frac{1}{\tau_c}$$
(1.16)

This expression, then, gives the instantaneous power of the pulse (in watts) at any time t. The total energy of the pulse can be determined by integrating the power over the entire time frame of the pulse:

$$E_{pulse} = \int \left((1 - R_{OC}) h v n V \frac{1}{\tau_c} \right) dt$$
(1.17)

The integration may be made easier by realizing that we may integrate over ΔN since we know that it starts at value $\Delta N_{initial}$ and ends at some terminal value ΔN_{final} which we may, for many practical purposes, substitute $\Delta N_{thresold}$. Then the integral becomes,

$$E_{pulse} = 2 \int_{\Delta N_{initial}}^{\Delta N_{threshold}} \left[\left\{ (1 - R_{OC}) h \nu n V \frac{1}{\tau_c} \right\} \frac{dt}{d\Delta N} \right] d\Delta N$$
(1.18)

By replacing the value of $\frac{dt}{d\Delta N}$ from Eqn. 1.10 and conducting the integration yields,

$$E_{pulse} = (1 - R_{OC}) Vhv \Delta N_{th} \ln \frac{\Delta_{initial}}{\Delta_{threshold}}$$
(1.19)

At peak power, the rate of change of the number of photons will be zero. Setting dn/dt = 0in equation 1.12, we find that the peak power occurs when $\Delta N = \Delta N_{th}$. By substituting this value into equation 1.14, we can obtain an exact expression for the number of coherent photons per unit volume at the peak of the pulse. Further simplification is possible if we assume that $\Delta N_{initial}$ is much, much larger than ΔN_{th} , so we neglect many terms in the result to find,

$$n_{peak} = \frac{1}{2} \Delta N_{initial} \tag{1.20}$$

Substituting this value in equation 1.15 yields an straightforward equation,

$$P_{peak} = \frac{1}{2} \left(1 - R_{OC} \right) h \nu \Delta N_{initial} V \frac{1}{\tau_c}$$
(1.21)

And the width of the pulse is given by,

$$t_{pulse} = \frac{E_{pulse}}{P_{peak}} \tag{1.22}$$

1.5 ELECTROOPTIC MODULATORS

Electrooptic modulators work on the principle of *birefringence*, which is induced in a crystal by the application of an external electric field. Birefringence is the effect of effect of separating a incident ray into two different ray that travels in a different direction, direction depending on ray's polarization. For each of the two rectangular states of polarization (perpendicular and parallel), the light will travel in a different direction as it exits the crystal. This effect is also

called *double refraction*. Calcite is a natural crystal that exhibits birefringence.

Figure 1.4 shows the effect of birefringence on a beam of unpolarized light that is incident on the surface of a birefringent crystal at an angle u. The two different components of polarization (perpendicular and parallel) take two separate paths through the crystal. This effect is caused by the index of refraction of the crystal, which has the property of being dependent on the polarization of the incident beam. For such a crystal, two indexes of refraction exist, one for each polarization of the incident light.



Figure 1.4: Birefringence.

There are also a number of crystals that exhibit birefringence only when an external electric field is applied. This phenomenon, called the electrooptic effect, can be used in the case of a Q-switch. It may also be used to deflect beams. An electrooptic modulator is shown schematically in Figure 1.5. Incident light is polarized and then passed through the electrooptic material (this polarization is not needed if the incident light is already polarized). Light exiting the crystal is passed through a second polarizing filter, called an *analyzer*. Since the crystal causes no change to the light passing through it when not energized, the analyzer, oriented at 90° to the polarizer, prevents any light from being transmitted through the device. In essence, when the crystal is not energized, the device acts as a shutter. When an external electric field is applied to the crystal, the direction of the polarization of light passing through it is rotated by 90°. Then the light will pass through the analyzer; effectively the shutter is open. Two electrooptic effects are possible,



Figure 1.5: Electrooptic modulator.

classified according to how the refractive index changes relative to the applied electric field: the *Pockels effect* and the *Kerr effect*. Which effect occurs depends on the material involved. In the *Pockels effect* the change in refractive index is a linear function of an applied electric field E:

$$\Delta n = a_1 E \tag{1.23}$$

where all is the linear electrooptic coefficient. The coefficient a_1 is a property of a material and is zero in any noncrystalline substance.

The other electrooptic effect, in which the change in refractive index is a function of the square of the applied electric field, is the *Kerr effect*, in which the change of refractive index is described by

$$\Delta n = a_1 E^2 \tag{1.24}$$

where a2 is a second-order electrooptic coefficient, or, more generally as

$$\Delta n = K\lambda E^2 \tag{1.25}$$

where K is the Kerr coefficient, a property of a particular material. All materials exhibit the Kerr effect, although the effect is very small in many materials.

Common materials used for Kerr-type EO modulators include the liquid nitrobenzene (which is toxic) and glass. Kerr-type modulators are uncommon in laser use, and essentially all modern modulators use the Pockels effect. These are often called Pockels cell modulators. The phase change for light passing through a modulator is proportional to the thickness of the EO material as well as the change in refractive index generated by the electric field according to

$$\Delta \phi = \frac{2\pi \Delta nL}{\lambda} \tag{1.26}$$

which allows us to express the transmission of the modulator (T) according to the change in refractive index according to

$$T = T_0 \sin^2\left(\frac{\pi \Delta nL}{\lambda}\right) \tag{1.27}$$

where *T* is the transmission, T_0 the maximum transmission of the assembly when open, Δn the difference in refractive index for the two polarizations, *L* the length of the crystal, and λ the wavelength of the light. The maximum transmission occurs when the term $\frac{\pi \Delta nL}{\lambda} = \frac{\pi}{2}$ or when

$$\Delta n = \frac{\lambda}{2L} \tag{1.28}$$

This phenomena occurs at a voltage called *half-wave voltage* $V_{1/2}$. The half-wave voltage is a property of the material itself, but this voltage increases with the wavelength according to

$$\frac{V_{1/2}^{\lambda_1}}{V_{1/2}^{\lambda_2}} = \frac{\lambda_1}{\lambda_2}$$
(1.29)

EO modulators operating in the infrared thus require much higher voltages than those operating in the visible region (which often operate at voltages of 5 to 10 kV).

1.6 ACOUSTOOPTIC MODULATORS

The simplest and most common modulator is the acoustooptic modulator. An acoustic wave, at radio frequencies, originates from a piezoelectric crystal which generates a surface acoustic wave in a crystal as outlined in Figure 1.6. The waves generated by the piezoelectric crystal form almost entirely flat plane wavefronts in the crystal. This wave is transmitted through a quartz crystal and induces a strain (rarefication and compression) in the crystal which changes its refractive index on a localized scale.



Figure 1.6: Acoustooptic modulator.

The acoustic wave, although at radio frequencies, propagates through the crystal at the speed of sound, so the wavelength of the acoustic wave inside the crystal is

$$\Lambda = \frac{v_{acoustic}}{f} \tag{1.30}$$

where Λ is the wavelength inside the crystal. Thus, we have a relatively long wavelength in the crystal. Regions of high and low index of refraction will similarly exist in the crystal at these points, as outlined on Figure 1.7. In the case of an AO modulator, the acoustic wave inside a



Figure 1.7: Acoustic waves in an AO modulator (Bragg diffraction condition).

crystal sets up parallel planes of varying indexes of refraction, which like the parallel planes of atoms in the crystal, generate Bragg diffraction of an incident light beam. The wavelength of the acoustic wave in the crystal corresponds to the wavelength of the incident light beam just as the spacing of the atoms in the crystal corresponds to x-ray wavelengths. An incident light beam on an AO modulator crystal is deflected only when an acoustic wave is present in the crystal (i.e., it is driven by a radio-frequency (RF) source). Mathematically, this deflection can be expressed in a manner similar to that for an optical diffraction grating:

$$2\Lambda\sin\theta_B = \frac{\lambda}{n} \tag{1.31}$$

where θ_B is the Bragg angle and λ is the wavelength in air. The exit beam is deflected out of the crystal at the same angle as it enters. This type of diffraction may be used to deflect a beam and hence may be used as a beam scanner.

It must be noted that the angle of diffraction does not depend on the intensity of the acoustic wave: Increasing this parameter (by increasing the RF drive signal) simply changes the intensity of the diffracted light beam. Of course, there are physical limits on how much energy a given switch will tolerate.

The second type of diffraction, RamanNath diffraction, occurs when the incoming beam is aligned perpendicular to the alternating layers, which now act like parallel slits in a transmission diffraction grating. In this regime the diffraction is modeled to occur from a thin acoustic wave beam inside the crystal. The angle of diffraction is quite different from the Bragg angle and the incoming light beam is deflected in both directions (both upward and downward).



Figure 1.8: RamanNath diffraction from an AO modulator.

AO modulators are by far the most common for laser Q-switches. They are inexpensive (the crystal for most modulators is made of quartz), and the drive circuitry is a simple RF oscillator and power amplifier. With a suitable antireflective coating on the faces of the crystal (which is wavelength specific), the insertion loss is almost negligible, allowing it to be used with low-gain lasers.

1.7 CAVITY DUMPING

If a laser is constructed with no output coupler, but two high reflectors, then there would be no output beam but the intracavity power would be very high. If suddenly one of the mirror is removed, massive output beam would occur. All of the energy circulating in the cavity would exit in a single pulse with a pulse length equal to the round-trip time for radiation in the cavity. This is the premise for cavity dumping.



Figure 1.9: Cavity dumping using an EO cell.

Theoretically, a mechanism to remove a mirror instantly could be used as a cavity dumper, but such an arrangement would not be practical given the large time it would take to move the mirror physically. Practically, an EO modulator may be used as a cavity dumper by changing the polarization of intracavity radiation, which is then deflected from an intracavity polarizing beamsplitter as shown in Figure 1.9.

Cavity dumping techniques will work with any laser, including solid-state and gas lasers, to

produce fast pulses, up to about 20 times more powerful than CW laser power. Such a technique can be done repetitively to produce a train of pulses.

1.8 MODELOCKING

The technique used to generate the shortest pulses of light is known as *modelocking*. In a modelocked laser, a single packet of light that traverses the laser, reflecting from cavity mirrors and through the gain medium. The pulse inside the cavity is much shorter than the cavity itself: If the cavity was 1 m long, the pulse might typically be 10 cm within this cavity. If the cavity contains a partially reflecting mirror as an output coupler, a short output pulse is transmitted each time the pulse is reflected from that mirror.

The modelocked laser consists of a normal Q-switched laser in which the Q-switch is opened at regular intervals corresponding to the transit time of the pulse within the cavity (c/2L). Once per round trip through the laser cavity the Q-switch is opened (Figure 1.10c) to allow the pulse to pass; at all other times the switch is closed to prevent any other light from oscillating in the cavity except for this modelocked pulse. If it is placed near one mirror in the cavity (as in the figure), only one opening of the switch is required per round trip.



Figure 1.10: Modelock pulse development in the time domain.

The output of a modelocked laser with the configuration described is a continuous series of

short pulses. In the case of a laser with mirrors 1 m apart, the pulses will appear at a frequency of c/2L or 150 MHz. Pulse duration depends, among other factors, on the time for which the Q-switch is open as well as the gain bandwidth of the lasing species. Q-switches for a modelocked laser must open and close in a very short time period. Regular Q-switches, such as the AO modulators(100 ns) used for a Q-switched laser, are generally not fast enough for these purposes. Standing wave scheme is used to use AO modulator as the Q-switch. Compared to AO modulators, EO modulators have much faster opening times of 1 to 2 ns and so can be used directly as modelockers. A further advantage is that EO modulators can be inserted directly into the cavity without using a polarizer/analyzer filter combination as usually required when used as an optical switch.

The final method to modelock a laser is to use a saturable dye absorber (also used as a Q-switch). These absorbers also serve to Q-switch the laser at the same time; the output is a pulse train with a Q-switched amplitude envelope. As with a simple Q-switch, an incident pulse inside the laser cavity bleaches (saturates) the dye, which opens the switch, allowing the pulse through it.

1.9 MODELOCKING IN THE FREQUENCY DOMAIN

When considering a practical laser it becomes apparent that many longitudinal modes (with slightly different frequencies) can oscillate simultaneously in a cavity. In a modelocked laser, all of these components are locked together in phase with each other. Where these components interfere constructively, this is the center of the modelocked pulse with a high peak power.

This occurs at all because all modes in the laser are given equal amplification (at the same time) when the switch is open, so that energy is distributed among these modes equally. Where the modes interfere constructively (i.e., they are all in phase), the modelocked pulse peaks; where they interfere destructively, the pulse is extinguished. In most practical lasers the bandwidth of the gain is wide enough to allow a number of longitudinal modes, each of slightly different frequency,

$$E(t) = \dots + E_{n-2}\sin\omega_{n-2}t + E_{n-1}\sin\omega_{n-1}t + E_n\sin\omega_n t + E_{n+1}\sin\omega_{n+1}t + \dots$$
(1.32)

The amplitudes of each of these component waves is summed (as happens where multiple waves are present) by interference. When all component waves are in phase, constructive interference occurs, producing a large amplitude. At some point these amplitudes will sum to zero, and the resulting summed amplitude will also be zero. This situation is outlined in Figure 1.11, where three individual modes, each with a slightly different frequency, are plotted in time.



Figure 1.11: Modelocked pulse synthesis.



Figure 1.12: Effect of number of modes on pulse shape.

Considering that numerous longitudinal modes are required in the laser for this technique to work, the more modes that oscillate, the shorter the resulting pulse will be. A laser species with a wide spectral range will allow more modes to oscillate, and hence the pulse will be shorter. This situation is demonstrated in Figure 7.9.2, where the pulse shape is plotted when three and $\frac{17}{17}$

15 component modes are summed. With three modes active, the envelope is obvious, but as more modes are summed, the envelope becomes quite narrow. As more modes are allowed to exist, the pulse shape becomes shorter, which explains why the shortest pulses ever produced have been from modelocked lasers with extraordinarily wide spectral width.

Chapter 2

Non-Linear Optics

Certain material can double the lasing frequency when the laser light is passed through them, but to a very small proportion. much study has been made of nonlinear materials and techniques have been devised to exploit this behavior in a efficient way. Here, the physics behind these nonlinear optical effects and applications have been discussed.

2.1 LINEAR AND NONLINEAR PHENOMENA

In most of the physical phenomena, when the quantity is small it shows linear relationship (like relation between the bending of a rular to the applied force), but when the quantity is increased above a certain value, non-linear behavior exhibits. Many optical material behaves in a similar manner. Consider the structure of a quartz crystal: a lattice with atoms placed at regular patterns. Electrons in the crystal are held to the nucleus of each atom by a force similar to a spring. When low and moderate light intensities are incident on such a crystal, electrons behave in a linear manner. Bonds stretch in response to an electric field in a linear manner.

Using Bohr analogy, the atom consisting of a positive nucleus and negative electrons in different shells orbiting them. Now when light impinges on atom, it can dislocate the valence electron, as it has a very weak binding with the nucleus. When an electric field is applied, it only affects the electrons, as the nucleus is much heavier. It makes the valence electrons to move to an end of the atom which polarizes the whole atom. A positive and a negative end develop on the atom, and an electric dipole with a dipole moment develops in the direction of

the electric field applied.

In the presence of an applied electric field, atoms in the crystal become polarized in this manner, and the entire material becomes polarized on a localized level (around the intense beam). This is a *macroscopic charge polarization* of the material, and the amount of charge polarization depends on the intensity of the applied electric field according to

$$P = aE \tag{2.1}$$

where a is the coefficient of polarizability for the material and E is the applied electric field in V/cm. This depicts a linear relation but it hold only for small electric field applied producing a polarization in step with the electric field. This polarization and reradiation of photons results in a slowing of the velocity of light through the crystal, but no change occurs in the nature of the light passing through the crystal.

Now, when the applied force is very large, nonlinear effects are exhibited for the same reason that the ruler exhibits nonlinear behavior. The charge polarization becomes nonlinear according to a geometric series:

$$P = a_1 E + a_2 E^2 + a_3 E^3 + \cdots$$
 (2.2)

The nonlinearity of the charge polarization becomes apparent, and high-order terms such as a_2 and a_3 contribute to the polarization.

To accomplish this nonlinear polarization, an intense electric field is required. The electron is held to the positive nucleus by an internal force of about 10^9 V/cm. Normal light sources with electric fields below 100 V/cm exhibit only linear optical effects. Few sources exist with a high enough electric field (comparable to the internal force) to generate nonlinear effects. A focused laser beam, for example, can impress an electric field of 10^6 to 10^7 V/cm on a crystal. Even higher intensities, with even higher electric fields, can be found inside the cavities of most lasers. To demonstrate mathematically, we replace $E = E_0 \cos \omega t$ in 2.2,

$$P = a_1 E_0 \cos \omega t + a_2 E_0^2 \cos^2 \omega t + \cdots$$
(2.3)

Trigonometric substitution can be made for the cos^2 term as

$$\cos^2 \omega t = \frac{1}{2} + \frac{1}{2}\cos 2\omega t$$
 (2.4)

So, the Eqn. 2.3 becomes,

$$P = a_1 E_0 \cos \omega t + \frac{1}{2} a_2 E_0^2 + \frac{1}{2} a_2 E_0^2 \cos 2\omega t + \cdots$$
 (2.5)

This leads to an interesting conclusion since the third term in equation contains a term with order 2ω ; one component of the charge polarization equation is at twice the fundamental frequency of the original laser beam. As the incident beam falls on the crystal, oscillating electric dipoles absorb light at a frequency ω and reradiate it at the same frequency ω and at twice the original frequency 2ω . The total energy of the beam is not altered much, but is now split between two components. This is the second harmonic of the original beam and the effect is called *second-harmonic generation* (SHG).



Figure 2.1: Generation of second harmonic light.

2.2 PHASE MATCHING

The three terms found from the right hand side of the Eqn. 2.5 represents three different polarization terms. The total is given by the summation of all three terms and that is shown in the Fig. 2.2. The peaks and valleys of the summed wave still coincide with those of the 21 original incident wave (i.e., it is in phase with the original v term), but the wave is shifted in one direction this case, upward. This is possible only if the nonlinear crystal has no internal symmetry. About 10% of all crystals have such a lack of symmetry and hence can be used as nonlinear media. In most crystals the index of refraction will be higher for the higher-frequency



Figure 2.2: Output terms from equation 2.5.

component (2ω) than for the lower-frequency component. A higher index of refraction means that light travels more slowly. Figure 2.3 illustrates the situation in which the second harmonic light becomes out of phase with the fundamental component as it passes through the crystal, causing destructive interference, which leads to extinction of the second harmonic component. The distance over which the waves travel before becoming 1808 out of phase is called the *coherence length*. If, the crystal is thicker and happens to have a thickness that is an odd multiple of the coherence length, the generated second harmonic light is entirely extinguished, due to destructive interference.

The problem of phase mismatch can be solved exploiting the effect of birefringence. this is an effect in which the index of refraction depends on the polarization of light passing through the crystal, or in this case, the individual light component passing through the crystal. By tilting the crystal such that the index of refraction for both the fundamental (with one specific polarization) and the harmonic component (with a different polarization) are the same, phase matching can be accomplished. When the direction of propagation of light in the crystal is chosen so that the indexes of refraction for both the fundamental and second harmonic are exactly the same, the crystal is said to be phase matched. The second harmonic component



Figure 2.3: Output terms from equation 2.5.

produced in the crystal now exits in phase with the fundamental component, so that constructive interference occurs.

Types of Phase-matched Crystal

Nonlinear crystals are classified by the type of phase matching required, each type requiring a different polarization of each input component. Type I phase-matched crystals utilize input beams with polarizations parallel to each other, while type II phase-matched crystals utilize input beams with polarizations perpendicular to each other. Both types of crystals are outlined in Figure 2.4, in which input components ω_1 and ω_2 (identical for a second-harmonic generator) are mixed to produce an output at frequency ω_3 .



Figure 2.4: Types of phase matching.

Process of Doubling Frequency

a small green DPSS laser can be considered, in which vanadate (a common solid-state laser crystal normally with an output at 1064 nm in the infrared) is frequency-doubled to produce green light at 532 nm using a type II phase-matched crystal of KTP (a common nonlinear crystal). Vanadate produces a polarized output at 1064 nm that is oriented at 45 degrees to the optical axis of the KTP crystal. This output is hence split into two polarization components (the o and e components) perpendicular to each other, as required for type II phase matching. A second-harmonic beam is generated with a rotation of 45 degrees relative to the 1064-nm output from the vanadate, which exits through the output coupler. The optical train of such a laser is described in Figure 8.2.4 along with relative polarizations of each component.



Figure 2.5: Type II phase matching in a DPSS laser.

For a crystal with unknown axes, phase matching can be accomplished by mounting the crystal in a mount allowing angular adjustment, and passing the beam from a Q-switched laser through the crystal (a Q-switched laser is used since the peak intensities are extremely high, as required for high second-harmonic generation efficiency). By rotating the crystal while the $\frac{24}{24}$

beam from the laser passes through it, second-harmonic light will be generated when the phasematching angle is encountered. The angle may then be optimized for peak second-harmonic output, at which point the phase-matching direction has been found.

Other approaches include the optical pumping of a semiconductor laser (as opposed to a normal solid-state laser crystal) with a nonlinear doubling crystal inside the cavity. Solid-state lasers such as the YAG, on the other hand, feature much narrower gain bandwidths, so are already ideally suited for doubling. In addition to angular adjustment, phase matching of some nonlinear crystals may be achieved by changing the temperature of the crystal itself. Such crystals exhibit indexes of refraction that change with temperature. Niobates such as lithium niobate (LiNbO₃) exhibit such behavior.

An alternative method to ensure phase matching is quasi-phase matching using a periodically poled nonlinear material. In such a material, thin layers of nonlinear material are stacked with polarizations in opposite directions. As the fundamental and harmonic waves both propagate as they pass through the crystal, phase shifts occur due to the differing refractive indexes at each wavelength. By reversing the phase mismatch periodically, phase matching can be accomplished without the use of angular rotation.

2.3 NONLINEAR MATERIALS

It is important to choose a good candidate for the material for harmonic light generator. These nonlinear coefficients (a_2 and a_3), as well as the coefficient for the linear term (a_1), are properties of the medium itself but can be determined and depend on the applied electric field. At small electric field values, the linear term dominates, but as this value increases, the nonlinear coefficients increase as well, so the material behaves in a nonlinear manner.

The linear term is related to the index of refraction by

$$a_1 = \varepsilon_0 (n^2 - 1) \tag{2.6}$$

where ε_0 is the permittivity of free space (8.85 × 10¹² F/m). This makes this term on the numerical order of 10¹² whereas the second-order coefficient a_2 for many nonlinear crystals is

on the order of 10^{24} . Although this coefficient seems extremely small, it is multiplied by E_0^2 in the equation, which gives it a reasonable value.

Common crystals for use as frequency doublers have large nonlinear coefficients, but not all crystals in nature have this feature. Only crystals that have no symmetry have a nonzero a_2 coefficient. These crystals are similar to those used in Pockels cell EO modulators and are piezoelectric as well. Quartz and other glasses have relatively small coefficients and hence make poor generators. Other crystals commonly used for harmonic generators that have large nonlinear coefficients include phosphates such as ADP (ammonium dihydrogen phosphate), KDP (potassium dihydrogen phosphate), KTP (potassium titanyl phosphate), and niobates such as lithium niobate (LiNbO₃).

| Material | Nonlinear Coefficient |
|--|--|
| Quartz | 2.6×10^{26} to 3.0×10^{24} |
| $ADP\left(NH_4H_2PO_4\right)$ | 6.8×10^{24} |
| KDP (KH ₂ PO ₄) | 3.8×10^{24} to 4.1×10^{24} |
| KTP (KTiOPO ₄) | 4.4×10^{23} to 1.2×10^{22} |
| Lithium niobate (LiNbO ₃) | 2.3×10^{23} to 3.9×10^{22} |

Commercially available nonlinear crystals are often specified by the type of phase-matching required, phase-match angle, and coatings on the crystal faces. In general, crystal faces must be coated with antireflective coatings suitable for both the fundamental and harmonic wavelength involved. An uncoated crystal would lead to a large intracavity loss, which may well prevent oscillation of the laser.

Finally, a good nonlinear crystal must be able to take the enormous power densities incident on the crystal without permanent damage. A nonlinear crystal may be damaged when the incident power reaches a high enough level. In this particular case, heat generated by the incident laser beam caused the crystal to fracture. Each type of crystal then has a characteristic damage threshold, which varies from 20MW/cm² for a lithium niobate crystal to 500MW/cm² for an ADP crystal.

2.4 SHG EFFICIENCY

The second-harmonic conversion efficiency, defined as the ratio of the power of the secondharmonic output component to the power of the fundamental component, is directly proportional to the intensity of the electric field, $I = P_{\text{fundamental}}/A$. This explains why many harmonic generator crystals are placed inside the cavity of the laser where the intensity is highest. Incident beams may also be focused inside the crystal to increase the localized intensity in the crystal as the beam passes through it. To prevent damage in this case, one must ensure that the intensity does not exceed the damage threshold of the crystal.

Efficiency is also proportional to the square of the length of the second harmonic crystal so that doubling the length of the crystal quadruples the second-harmonic output. We find that the relationship between incident power and second-harmonic output power is,

$$P_{\text{second harmonic}} = \frac{Kl^2 P_{\text{incident}}^2}{A}$$
(2.7)

where K is a proportionality constant, I the length of the crystal, and A the beam area in the crystal.

2.5 SUM AND DIFFERENCE OPTICAL MIXING

So far we have predicted and anticipated the production of second harmonic generation only, but in general, non-linear process extend much further, to the production of both summed and difference frequency components of the source frequencies. In the world of electronics, this process of mixing is used in all radio receivers to generate intermediate frequencies, which are then detected. This process is followed in both AM and FM radio receivers. This scheme, called superheterodyning, allows the tuning of the receiver to any frequency desired by tuning an oscillator. The same mixing effect can be accomplished with light by mixing it in a nonlinear crystal; in fact, most nonlinear optical effects have similar analogies in the world of electronics. In the effect described here, when two beams with frequencies ω_1 and ω_2 interact in a nonlinear crystal, they induce a polarization oscillating at $\omega_1 + \omega_2$ and $\omega_1 - \omega_2$: in other words, a sum and a difference signal. This interaction is depicted in Figure 2.6. Where λ_1 and λ_2 interact



Figure 2.6: Model for mixing in a nonlinear crystal.

they can sum together to produce λ_4 or a difference in λ_3 . Remember that energies of these waves add and subtract so that the shorter wavelength sum (λ_4) contains the sum of energies in photons λ_1 and λ_2 . Although laser beams are generally used for nonlinear mixing, intense incoherent sources can be used as well: Lasers simply offer a convenient source of intense light.

2.6 HIGHER-ORDER NONLINEAR EFFECTS

As mentioned earlier, not all crystal material have a_2 co-efficient of non-linearity, only 10% material have them. But all material, including glass, have third order harmonic component, say a_3 . However, as this is much smaller compared to second order harmonic, the efficiency of third order harmonic is expected to be much smaller. Calcite, for example, can be used as a third-harmonic generator with efficiencies of less than 1025. Alternatively, series nonlinear crystals can be used to generate third-harmonic light. Both process is shown in Fig. 2.7.



Figure 2.7: Generation of high-order harmonics.

2.7 **OPTICAL PARAMETRIC OSCILLATORS**

In the process of harmonic generation two photons combine their energy into a single photon. We have also seen that the reverse process, in which a single photon splits its energy into two photons which will have lower energy than the incident photon, is also possible. By conservation of energy, the energy of the resulting photons must sum to the energy of the incident photon. In an optical parametric oscillator (OPO) configuration a pump beam incident on a nonlinear crystal produces two resultant photons at different wavelengths. One wavelength, called the signal, exits the device as the output beam; the second wavelength, essentially useless, called the idler beam, stays within the cavity of the device, as depicted in Fig. 2.8. The frequencies of the idler and output beam sum to the frequency of the pump beam. An OPO is not a laser since it does not amplify; rather, it is an oscillator only. It is, however, a coherent oscillator producing laser light.



Figure 2.8: Simple OPO device.

A simple OPO produces two output beams, but only one of them is used as output, because only one of the outputs is phase-matched at a time. This is also how the OPO is tuned: by using the same methods as those used to phase-match a second-harmonic generator crystal. The temperature of the crystal or the angle of the crystal within the cavity can be changed to tune the laser. In the case of angular tuning, often two crystals are used that rotate in opposite directions. This scheme eliminates displacement of the beam in the cavity, which leads to walk-off loss.