# Lasers: Fundamentals, Types, and Operations

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The acronym LASER, constructed from *L*ight Amplification by Stimulated Emission of Radiation, has become so common and popular in every day life that it is now referred to as *laser*. Fundamental theories of lasers, their historical development from milliwatts to petawatts in terms of power, operation principles, beam characteristics, and applications of laser have been the subject of several books [1–5]. Introduction of lasers, types of laser systems and their operating principles, methods of generating extreme ultraviolet/vacuum ultraviolet (EUV/VUV) laser lights, properties of laser radiation, and modification in basic structure of lasers are the main sections of this chapter.

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### 1.1 Introduction of Lasers

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### 1.1.1 Historical Development

The first theoretical foundation of LASER and MASER was given by Einstein in 1917 using Plank's law of radiation that was based on probability coefficients (Einstein coefficients) for absorption and spontaneous and stimulated emission of electromagnetic radiation. *Theodore Maiman* was the first to demonstrate the earliest practical laser in 1960 after the reports by several scientists, including the first theoretical description of *R.W. Ladenburg* on stimulated emission and negative absorption in 1928 and its experimental demonstration by *W.C. Lamb* and *R.C. Rutherford* in 1947 and the proposal of *Alfred Kastler* on optical pumping in 1950 and its demonstration by *Brossel, Kastler*, and *Winter* two years later. *Maiman's* first laser was based on optical pumping of synthetic ruby crystal using a flash lamp that generated pulsed red laser radiation at 694 nm. Iranian scientists *Javan* and *Bennett* made the first gas laser using a mixture of He and Ne gases in the ratio of 1 : 10 in the 1960. *R. N. Hall* demonstrated the first diode laser made of gallium arsenide (GaAs) in 1962, which emitted radiation at 850 nm, and later in the same year *Nick Holonyak* developed the first semiconductor visible-light-emitting laser.

#### 1.1.2

#### **Basic Construction and Principle of Lasing**

Basically, every laser system essentially has an active/gain medium, placed between a pair of optically parallel and highly reflecting mirrors with one of them partially transmitting, and an energy source to pump active medium. The gain media may be solid, liquid, or gas and have the property to amplify the amplitude of the light wave passing through it by stimulated emission, while pumping may be electrical or optical. The gain medium used to place between pair of mirrors in such a way that light oscillating between mirrors passes every time through the gain medium and after attaining considerable amplification emits through the transmitting mirror.

Let us consider an active medium of atoms having only two energy levels: excited level  $E_2$  and ground level  $E_1$ . If atoms in the ground state,  $E_1$ , are excited to the upper state,  $E_2$ , by means of any pumping mechanism (optical, electrical discharge, passing current, or electron bombardment), then just after few nanoseconds of their excitation, atoms return to the ground state emitting photons of energy  $hv = E_2 - E_1$ . According to *Einstein's* 1917 theory, emission process may occur in two different ways, either it may induced by photon or it may occur spontaneously. The former case is termed as *stimulated emission*, while the latter is known as *spontaneous emission*. Photons emitted by stimulated emission have the same frequency, phase, and state of polarization as the stimulating photon; therefore they add to the wave of stimulating photon on a constructive basis, thereby increasing its amplitude to make lasing. At thermal equilibrium, the probability of stimulated emission is much lower than that of spontaneous emission (1 : 10<sup>33</sup>), therefore most of the conventional light sources are incoherent, and only lasing is possible in the conditions other than the thermal equilibrium.

#### 1.1.3

#### **Einstein Relations and Gain Coefficient**

Consider an assembly of  $N_1$  and  $N_2$  atoms per unit volume with energies  $E_1$ and  $E_2(E_2 > E_1)$  is irradiated with photons of density  $\rho_{\nu} = N h\nu$ , where [N] is the number of photons of frequency  $\nu$  per unit volume. Then the stimulated absorption and stimulated emission rates may be written as  $N_1\rho_{\nu}B_{12}$  and  $N_2\rho_{\nu}B_{21}$  respectively, where  $B_{12}$  and  $B_{21}$  are constants for up and downward transitions, respectively, between a given pair of energy levels. Rate of spontaneous transition depends on the average lifetime,  $\tau_{21}$ , of atoms in the excited state and is given by  $N_2A_{21}$ , where  $A_{21}$  is a constant. Constants  $B_{12}$ ,  $B_{21}$ , and  $A_{21}$  are known as *Einstein coefficients*. Employing the condition of thermal equilibrium in the ensemble, Boltzmann statistics of atomic distribution, and Planck's law of blackbody radiation, it is easy to find out  $B_{12} = B_{21}$ ,  $A_{21} = B_{21}(8\pi h\nu^3/c^3)$ , known as *Einstein relations*, and ratio,  $R = \exp(h\nu/kT) - 1$ , of spontaneous and stimulated emissions rates. For example, if we have to generate light of 632.8 nm ( $\nu = 4.74 \times 10^{14}$  Hz) wavelength at room temperature from the system of He–Ne, the ratio of spontaneous and stimulated emission will be almost  $5 \times 10^{26}$ , which shows that for getting strong lasing one has to think apart from the thermal equilibrium. For shorter wavelength, laser, ratio of spontaneous to stimulated emission is larger, ensuring that it is more difficult to produce UV light using the principle of stimulated emission compared to the IR. Producing intense laser beam or amplification of light through stimulated emission requires higher rate of stimulated emission than spontaneous emission and self-absorption, which is only possible for  $N_2 > N_1$  (as  $B_{12} = B_{21}$ ) even though  $E_2 > E_1$  (opposite to the Boltzmann statistics). It means that one will have to create the condition of *population inversion* by going beyond the thermal equilibrium to increase the process of stimulated emission for getting intense laser light.

If a collimated beam of monochromatic light having initial intensity  $I_0$  passes through the mentioned active medium, after traveling length x, intensity of the beam is given by  $I(x) = I_0 e^{-\alpha x}$ , where  $\alpha$  is the absorption coefficient of the medium, which is proportional to the difference of  $N_1$  and  $N_2$ . In the case of thermal equilibrium  $N_1 \gg N_2$  the irradiance of the beam will decrease with the length of propagation through the medium. However, in the case of population inversion,  $(N_2 > N_1) - \alpha$ , will be positive and the irradiance of the beam will increase exponentially as  $I(x) = I_0 e^{kx}$ , where k is the gain coefficient of the medium and may be given by  $k = (nN_d hv_{21}B_{21})/c$ , where  $N_d$  is  $N_2 - N_1$ , c is speed of light, and n is refractive index of the medium.

#### 1.1.4

#### Multilevel Systems for Attaining Condition of Population Inversion

Considering the case of two energy level system under optical pumping, we have already discussed that  $B_{12} = B_{21}$ , which means that even with very strong pumping, population distribution in upper and lower levels can only be made equal. Therefore, optical as well as any other pumping method needs either three or four level systems to attain population inversion. A three level system (Figure 1.1a) irradiated by intense light of frequency  $v_{02}$  causes pumping of large number of atoms from lowest energy level  $E_0$  to the upper energy level  $E_2$ . Nonradiative decay of atoms from  $E_2$  to  $E_1$  establishes population inversion between  $E_1$  and  $E_0$  (i.e.,  $N_1 > N_0$ ), which is practically possible if and only if atoms stay for longer time in the state  $E_1$  (metastable state, i.e., have a long lifetime) and the transition from  $E_2$  to  $E_1$  is rapid. If these conditions are satisfied, population inversion will be achieved between  $E_0$  and  $E_1$ , which makes amplification of photons of energy  $E_1 - E_0$  by stimulated emission. Larger width of the  $E_2$  energy level could make possible absorption of a wider range of wavelengths to make pumping more effective, which causes increase in the rate of stimulated emission. The three level system needs very high pumping power because lower level involved in the lasing is the ground state of atom; therefore more than half of the total number of atoms have to be pumped to the state  $E_1$  before achieving population inversion and in each of the cycle, energy used to do this is wasted. The pumping power can be greatly reduced if the lower level involved in the lasing is not ground state, which requires at least a four level system (Figure 1.1b). Pumping transfers atoms from ground state to  $E_3$ , from where they decay rapidly into the metastable state  $E_2$  to make  $N_2$  larger than

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Figure 1.1 Energy level diagram for (a) three- and (b) four level laser systems.

 $N_1$  to achieve the condition of population inversion between  $E_2$  and  $E_1$  at moderate pumping.

### 1.1.5 Threshold Gain Coefficient for Lasing

Laser beam undergoes multiple oscillations (through active medium) between pair of mirrors to achieve considerable gain before it leaves the cavity through partially reflecting mirror. Laser oscillation can only sustain in the active medium if it attains at least unit gain after a round-trip between mirrors and maintains it overcoming various losses inside the cavity. If we incorporate these losses, the effective gain coefficient reduces to  $k - \Upsilon$ , where  $\Upsilon$  is the loss coefficient of the medium. If round-trip gain G were less than unity, the laser oscillation would die out, while it would grow if the G value were larger than unity. Let us consider that the laser beam of intensity I<sub>0</sub> passes through the active medium, homogeneously filled in the length L between the space of two mirrors  $M_1$  and  $M_2$  with reflectivities  $R_1$  and  $R_2$ , respectively. The beam of intensity  $I_0$  initiates from the surface of  $M_1$  and attains intensity  $I_1(I_1 = I_0 \exp(k - \Upsilon)L)$  after traveling a length L to reach at the surface of  $M_2$ . After reflection from  $M_2$  and traveling back to  $M_1$ , the light intensity becomes  $I_2$  $(I'_1 = I_1 R_2$  due to reflection and  $I_2 = I'_1 \exp(k - \Upsilon)L$ ), which finally becomes  $I'_2$  after reflection from M<sub>1</sub> to complete a round-trip  $(I'_2 = I_2 R_1 = I_0 R_1 R_2 \exp(k - \Upsilon)L)$ . Waves starting from the surface of mirror M<sub>1</sub> and those that have completed one or more round trips are in the same phase. Now, the gain  $G(I'_2/I_0)$  attained in a round-trip should be at least unity to sustain the laser oscillation inside the cavity, therefore  $R_1 R_2 \exp(k - \Upsilon)L = 1$  is the threshold condition, which gives a value of  $\Upsilon + (2L)^{-1} \ln(R_1 R_2)^{-1}$  for threshold gain (*k*th) coefficient.

### 1.1.6 Optical Resonator

An optical resonator is an arrangement of optical components, which allows a beam of light to circulate in a closed path so that it retraces its own path multiple times, in order to increase the effective length of the media with the aim of large light amplification analogous to the positive feedback in electronic amplifiers. Combination of optical resonator with active medium is known as optical oscillator. A set of two parallel and optically flat mirrors, with one highly reflecting  $M_1(R \approx 100\%)$ and another partially transmitting  $M_2(R > 95\%)$ , makes a simple optical oscillator as shown in Figure 1.2. Some of the pumped atoms in the excited states undergo spontaneous emission generating seed photons, which pass through the active medium and get amplified through stimulated emission. Most of the energy gets reflected from both the mirrors, passes through the active medium, and continues to get amplified until steady state level of oscillation is reached. After attaining this stage, amplification of wave amplitude within the cavity dies away and extra energy produced by stimulated emission exits as laser output from the window M2. The gain coefficient inside the cavity should be greater than the threshold gain coefficient (kth) in order to start and maintain laser oscillation inside the cavity. Owing to the diffraction effects, it is practically difficult to maintain a perfectly collimated beam with the combination of two parallel plane mirrors, which causes significant amount of diffraction losses. Such losses could be reduced by using a combination of concave mirrors and other optics in different optical arrangements. The optical configurations, which are able to retain the light wave inside the cavity after several transversals, are known as stable resonators. Some of the stable resonators are shown in the Figure 1.3. Laser oscillators with different geometries have their own benefits and losses. For example, in an oscillator having assembly of two parallel mirrors, it is difficult to align them in a strictly parallel manner. A slight deviation from the parallel geometry of the laser beam causes its walk away from the cavity axis after few reflections. However, it is beneficial in the sense that a large fraction of the active medium (mode volume) is pumped in this geometry. Confocal resonators are very simple to align, although lesser fraction of the active medium is being pumped.

Every laser resonator is characterized by a quantity *Q* termed as *quality factor*, which is defined by  $Q = (2\pi \times \text{energy stored})/(\text{energy dissipated per cycle})$ . The *Q* value of laser cavities lies in the range of  $\sim 10^5 - 10^6$ . Significance of higher *Q* value lies in the sense of capacity to store larger energy. In terms of line width  $\Delta v$ ,



Figure 1.2 Basic geometry of laser cavity: (1) 100% and (2) 95-98% reflecting mirrors, (3) active medium, (4) pumping source, and (5) laser output.

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Figure 1.3 Different geometries of stable optical resonators.

and frequency  $\nu$ , the quality factor can be defined as  $Q = \nu/\Delta\nu$ . A higher Q value associates with lower relative line width and vice versa.

A resonator that cannot maintain laser beam parallel to its axis is termed as *unstable resonator*. Such resonators suffer from high losses, but can make efficient use of the mode volume and have easy way of adjustment for the output coupling of the laser. Figure 1.4 illustrates an unstable resonator having active medium between the mirrors. Output power of the laser and inner diameter of the annular-shaped beam can be easily adjusted by varying the distance between the two reflecting



Figure 1.4 A sketch of unstable optical resonator with annular beam shape.

mirrors. Resonators having low irradiation volume or unstable cavities require active media with a large gain coefficients, such as CO<sub>2</sub> gas.

### 1.1.7 Laser Modes

The output of laser beam actually consists of a number of closely spaced spectral lines of different frequencies in a broad frequency range. The discrete spectral components are termed as *laser modes*, and coverage range is the line width of the atomic transition responsible for the laser output. Laser modes are categorized into axial and transverse modes.

- 1) Axial modes: Let  $d\phi = 2\pi/\lambda^*(2L)$  be the phase change in the laser wave after a round-trip in the cavity. In order to sustain laser oscillation inside the cavity, the phase change should be an integral multiple of  $2\pi$ , that is,  $2\pi/\lambda^*(2L) = 2p\pi$ . In terms of frequency, this expression transforms to v = pc/2L; therefore separation between two adjacent *p* and *p* + 1 modes is given by  $\Delta v = c/2L$ (Figure 1.5). In the particular case of Nd: YAG (neodymium-ion-doped yttrium aluminum garnet) laser,  $\lambda = 1064$  nm and L = 25 cm,  $p = 2L/\lambda \approx 47 \times 10^4$ axial mode exists inside the laser cavity. If line width of the laser at 1064 nm is about  $\Delta w = 1$  GHz, then only  $\Delta w / \Delta v \approx 1$  axial mode oscillates in the cavity, while others die out. The axial modes are constructed by the light waves moving exactly parallel to the cavity axis. Light incident on a mirror and that reflected from that mirror construct a standing wave similar to a string bounded at both the ends. All the axial modes are due to the propagation of plane waves along the line joining centers of two reflecting mirrors.
- Transverse modes: Unlike the plane waves propagating along the axis of the 2) cavity in axial modes, there are some other waves traveling out of the axis that are not able to repeat their own path termed as transverse electromagnetic



Figure 1.5 Axial laser modes (a) a simple illustration and (b) inside the laser line width, which shows that the mode at the center of the line has maximum intensity.



Figure 1.6 Various TEM modes of the laser.

(TEM) modes. These modes can be practically seen in the form of pattern when the laser beam falls on any surface. These modes are assigned by two integers p and q in the form of TEM<sub>pq</sub>, where p and q are the number of minima in the horizontal and vertical directions, respectively, in the pattern of the laser beam. TEM<sub>00</sub> means that there is no minima in the beam spot, and this is known as *uniphase mode*. On the contrary, TEM<sub>01</sub> shows that there is no minima in the horizontal scanning and one minima in vertical. Laser beam spots on the screen with several TEM modes are displayed in Figure 1.6.

### 1.2

### Types of Laser and Their Operations

Depending on the nature of the active media, lasers are classified into three main categories, namely, solid, liquid, and gas. Scientists and researchers have investigated a wide variety of laser materials as active media in each category since 1958, when lasing action was observed in ruby crystal. It is inconvenient to discuss all lasers having these materials as active media. Here, representative active medium for each of the categories and their operating principle with energy level diagram is discussed.

### 1.2.1 Solid Laser

### 1.2.1.1 Doped Insulator Laser

Solid state lasers have active media obtained by embedding transition metals (Ti<sup>+3</sup>, Cr<sup>+3</sup>, V<sup>+2</sup>, Co<sup>+2</sup>, Ni<sup>+2</sup>, Fe<sup>+2</sup>, etc.), rare earth ions (Ce<sup>+3</sup>, Pr<sup>+3</sup>, Nd<sup>+3</sup>, Pm<sup>+3</sup>, Sm<sup>+2</sup>, Eu<sup>+2,+3</sup>, Tb<sup>+3</sup>, Dy<sup>+3</sup>, Ho<sup>+3</sup>, Er<sup>+3</sup>, Yb<sup>+3</sup>, etc.), and actinides

such as U<sup>+3</sup> into insulating host lattices. Energy levels of active ions are only responsible for lasing actions, while physical properties such as thermal conductivity and thermal expansivity of the host material are important in determining the efficiency of the laser operation. Arrangement of host atoms around the doped ion modifies its energy levels. Different lasing wavelength in the active media is obtained by doping of different host materials with same active ion. Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>, YAlO<sub>3</sub>, Y<sub>3</sub>Ga<sub>5</sub>O<sub>12</sub>, Y<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub>, YLiF<sub>4</sub>, Y<sub>2</sub>SiO<sub>5</sub>, Y<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub>, Y<sub>3</sub>Sc<sub>2</sub>Ga<sub>3</sub>O<sub>12</sub>, Ti:Al<sub>2</sub>O<sub>3</sub>, MgAl<sub>2</sub>O<sub>4</sub> (spinel), CaY<sub>4</sub>[SiO<sub>4</sub>]<sub>3</sub>O, CaWO<sub>4</sub> (Scheelite), Cr:Al<sub>2</sub>O<sub>3</sub>, NdP<sub>5</sub>O<sub>4</sub>, NdAl<sub>3</sub>[BO<sub>3</sub>]<sub>4</sub>, LiNdP<sub>4</sub>O<sub>12</sub>, Nd:LaMgAl<sub>11</sub>O<sub>19</sub>, LaMgAl<sub>11</sub>O<sub>19</sub>, LiCaAlF<sub>6</sub>, La<sub>3</sub>Ga<sub>5</sub>SiO<sub>4</sub>, Gd<sub>3</sub>Sc<sub>2</sub>Al<sub>3</sub>O<sub>12</sub>, Gd<sub>3</sub>Ga<sub>5</sub>O<sub>12</sub>, Na<sub>3</sub>Ga<sub>2</sub>Li<sub>3</sub>F<sub>12</sub>, Mg<sub>2</sub>SiO<sub>4</sub> (Forsterite), CaF<sub>2</sub>, Al<sub>2</sub>BeO<sub>4</sub> (Alexandrite), and so on, are some of the important hosts. Active atom replaces an atom in the host crystal lattice. Nd:YAG is one of the best lasing material and is representative of solid state lasing materials.

### 1.2.1.1.1 Dopant Energy Levels in the Host Matrices

Transition metal and rare earth ions have partially filled and unfilled 3d and 4f subshells, respectively. For example, the electronic configurations of trivalent Cr and Nd ions are as follows:

$$Cr^{+3}: 1s^2 2s^2 2p^6 3s^2 3p^6 3d^3$$
 
$$Nd^{+3}: 1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 4f^3 5s^2 5p^6$$

There are unshielded partially filled d electrons in the transition metal ions, while partially filled 4f electrons of the rare earth ions are shielded by 5p and 5s sub shells. Owing to the electronic shielding of inner subshells in rare earth ions, crystal field effect on the energy levels of transition metal ions are pronounced as compared to that on energy levels of rare earth ions. When one of these ions is doped into a host lattice, three main types of interactions occur: (i) columbic interaction between electrons in the unfilled shell, (ii) the crystal field, and (iii) spin–orbit interactions. The columbic interaction between electrons causes splitting of energy levels of a single electron configuration into several levels denoted by pair of values of *L* and *S* (*L* and *S* are vector sum of angular, *l*, and spin, *s*, momenta, respectively, of electrons). Crystal field splitting dominates for transition metal, while spin–orbit interaction is the major contributor for rare earth ions in the modification of energy level of isolated host atom. The energy level diagram for  $Cr^{+3}$ :Al<sub>2</sub>O<sub>3</sub> (ruby) and Nd<sup>+3</sup>:YAG are displayed in Figure 1.7.

### 1.2.1.1.2 Pumping Techniques in Solid State Lasers

Pumping of electrons from the ground state to the excited state to achieve population inversion condition is an essential requirement for lasing. Optical pumping is the best and most efficient pumping method for solid state active media due to their broad optical absorption bands. A significant fraction of incident optical energy can be easily used for the pumping of ground state electrons using pulsed as well as continuous light sources. Excess light energy raises temperature of the laser materials; therefore pulsed light sources are more suitable for dissipation of heat



Figure 1.7 Energy level diagrams for doped insulator lasers: (a) ruby and (b) Nd:YAG lasers.

through circulating water jackets. Low-pressure quartz/glass-sealed krypton/xenon lamps are mostly used for pulsed pumping light sources, while tungsten halogen lamps and high-pressure mercury discharge lamps are utilized for continuous optical pumping. An inductive, capacitive, and resistive (LCR) circuit and trigger unit as shown in Figure 1.8 is basically used for operating the flashtube. The detail circuit diagram of the power supply is presented in Ref. [3]. High-voltage pulse of the trigger coil ionizes some gas in the tube and makes it conductive. This causes rapid discharge of the capacitor through the tube and generation of intense optical radiation for almost few milliseconds. A small inductor in the series protects damage of the tube due to high capacitor discharge current. Light source and active medium should be arranged in such a way that maximum pumping radiation falls on the active medium. Active media in solid state lasers are cylindrical and rod shaped with few millimeters diameter and few centimeter lengths. Several arrangements of cylindrical flash lamp and rod-shaped active media are used for optical pumping to get laser radiation. The flash lamp and active medium assembly are placed inside gold-plated reflectors of circular or elliptical cross section. In the first practical operating laser, ruby rod was pumped by helical flash lamp inside the cylindrical reflecting cavity. Such arrangement has significant uniformity of irradiation inside the rod but exhibits poor optical coupling. Side-by-side arrangement of flash lamp and laser rod inside the cylindrical gold-plated reflector or wrapping both together with a metal foil are simpler approaches having good optical coupling



Figure 1.8 Trigger unit and LCR circuit diagram for solid state lasers.

but poor uniformity of irradiation. An elliptical reflector having flash lamp at one focus and laser rod at the other focus is the most popular and best way of optical pumping in the solid state lasers. Light radiation leaving from the first focus gets focused close to the axis of laser rod placed at the second focus to make uniform energy distribution. Combination of a number of elliptical reflectors having laser rod at the common foci and several flash lamps at the other foci is used for better optical pumping with more uniform energy distribution. Various geometries for the arrangement of laser rod and flash lamps are illustrated in Figure 1.9. Nd:YAG laser is widely used in the processing of materials and various characterizations. Here we discuss energy level diagram and operating principles of Nd:YAG lasers.

### 1.2.1.1.3 Nd:YAG Laser Construction and Operation

The schematic diagram of Nd:YAG laser head as shown in Figure 1.10, consists of oscillator section, rear mirror, quarter-wave plate, Pockel cells, polarizer, pump chambers, injection seeder, output coupler, D-Lok monitor, fold mirrors, amplifier section, harmonic generator (HG), temperature controller, dichroic mirrors, and Beam Lock pointing sensor. It may have single or multipump chambers, and each chamber consists of single or multiple flash lamps depending on the power of laser. The laser head end panel contains coolant, output connector, coolant input connector, neutral/ground connector, control cable connector, high-voltage connector, Q-switch input connector, and nitrogen purge input connector. The HGs have potassium di-hydrogen phosphate (KDP) and beta barium borate (BBO) crystals for frequency doubling and tripling, respectively. It can be operated in long pulse and Q-switch modes. Long pulse mode has light pulses of almost 200 µs duration and separated from each other by  $2-4\,\mu$ s. The total energy of the pulse

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Figure 1.9 Different geometries for the arrangement of flash lamp and laser rod in solid states lasers.

train is similar to that of a single Q-switched pulse. During Q-switched operation, the pulse width is less than 10 ns and the peak optical power is tens of megawatts.

The properties of Nd:YAG are the most widely studied and best understood of all solid state laser media. Its energy level diagram, optical arrangements for Q-switching and stable and unstable resonators are depicted in Figure 1.11. The active medium is triply ionized neodymium, which is optically pumped by a flash lamp whose output matches principle absorption bands in the red and near infrared (NIR). Excited electrons quickly drop to the  $F_{3/2}$  level, the upper level of the lasing transition, where they remain for a relatively longer time ( $\sim$ 230 µs). The strongest transition is  $F_{3/2} \rightarrow I_{11/2}$ , emitting a photon in NIR region (1064 nm). Electrons in the I11/2 state quickly relax to the ground state, which makes its population low. Therefore, it is easy to build up a population inversion for this pair of states with high emission cross section and low lasing threshold at room temperature. There are also some other competing transitions at 1319, 1338, and 946 nm from the same upper state, but having lower gain and a higher threshold than the 1064 nm wavelength. In normal operation, these factors and wavelength-selective optics limit oscillation to 1064 nm. A laser comprising just an active medium and resonator will emit a pulse of laser light each time the flash lamp fires. However, the pulse duration will be long, about the same as the flash lamp and its peak power will be low. When a Q-switch is added to the resonator to shorten the pulse, output peak power is raised dramatically. Owing to the long lifetime of  $F_{3/2}$ , a large



Figure 1.10 Assembly of various components in the head of an Nd:YAG laser system with four pump chambers.

population of excited neodymium ions can build up in the YAG rod in a way similar to which a capacitor stores electrical energy. When oscillation is prevented for some time to build up high level of population inversion by electro-optical Q-switching and after that if the stored energy gets quickly released, the laser will emit a short pulse of high-intensity radiation.

### 1.2.1.2 Semiconductor Laser

Semiconductor lasers also known as *quantum well lasers* are smallest, cheapest, can be produced in mass, and are easily scalable. They are basically p-n junction diode, which produces light of certain wavelength by recombination of charge carrier when forward biased, very similar to the light-emitting diodes (LEDs). LEDs possess spontaneous emission, while laser diodes emit radiation by stimulated emission. Operational current should be higher than the threshold value in order to attain the condition of population inversion. The active medium in a semiconductor diode laser is in the form of junction region of 2 two-dimensional layers. No external mirror is required for optical feedback in order to sustain laser oscillation. The reflectivity due to the refractive index differences between two layers or total internal reflection to the active media is sufficient for this purpose. The diodes end faces are cleaved, and parallelism of reflecting surfaces is assured. Junction



**Figure 1.11** (a) Energy level diagram for the transition of Nd:YAG laser (b) The Q-switch comprises a polarizer, a quarter-wave plate, high quality reflector, and pockels cell, and (c) stable and unstable resonator configurations.

made from a single type of semiconductor material is known as *homojunction*, while that obtained from two different semiconductors is termed as *heterojunction*. Semiconductors of p and n type with high carrier density are brought together for constructing p-n junction with very thin ( $\approx 1 \mu m$ ) depletion layer. Figure 1.12 illustrates GaAs homojunction semiconductor diode laser. Lasing occurs in the confined narrow region, and optical feedback is done by reflections between cleaved end faces. For GaAs n = 3.6, therefore reflectivity *R* from the material–air interface is  $R = (n - 1)^2/(n + 1)^2 = 0.32$ , which is small but sufficient for lasing.

When the operating current is small, the population inversion built compensates losses in the system and no lasing action is done. Increase of the current above a critical value named as *threshold current* commences lasing action, and the intensity of laser radiation increases rapidly with further increase in the operating current. Semiconductor lasers have large divergence compared to any other laser systems, which is due to their small cross section of active region. Actual dimension (*d*) of the active medium is of the order of light wavelength ( $\lambda$ ), which causes diffraction and hence divergence by an angle of  $\theta \approx \lambda/d$ . Homojunction semiconductor lasers have some disadvantages over heterojunction lasers. Both of the laser systems should have confinement of injected electrons and emitted light in the junction region in order to initiate efficient stimulated emission process. In



Figure 1.12 Basic geometry of semiconductor laser system.

the homojunction laser, confinement of light is the consequence of the presence of hole and electrons close to the junction. Homojunction lasers operate under such confinement mechanism but have high threshold current density and low efficiency. Electrons have to travel different distances before they recombine with the holes. In contrast, heterojunction lasers exhibit much higher lasing efficiency and low threshold current density compared to their homojunction counterparts. Another difficulty with homojunction laser is to prevent the radiation from spreading out sideways from the gain region, which causes loss instead of gain. Therefore they can only be used in the pulsed mode.

Heterojunction lasers are constructed by sandwiching a thin layer of GaAs between two layers of ternary semiconductor compound  $Ga_{1-x}Al_xAs$  with comparatively lower refractive indices and higher band gap energy. Lower refractive indices of surrounding layers causes confinement of laser radiation inside the active medium by the mechanism of total internal reflection, which makes laser oscillation to sustain in the medium. Higher band gap energy of the surrounding media creates potential barrier to prevent charge carriers to diffuse from the junction region, that is, provides a way for the confinement of charge carriers in the junction region, which enhances the condition of population inversion and hence stimulated emission. The electrical circuit for pumping the semiconductor diode lasers is similar to the doped insulator lasers.

#### 1.2.2 Gas Laser

Gas lasers are widely available in almost all power (milliwatts to megawatts) and wavelengths (UV-IR) and can be operated in pulsed and continuous modes. Based on the nature of active media, there are three types of gas lasers viz atomic, ionic, and molecular. Most of the gas lasers are pumped by electrical discharge. Electrons in the discharge tube are accelerated by electric field between the electrodes. These accelerated electrons collide with atoms, ions, or molecules in the active media and



Figure 1.13 Construction of gas laser system (argon ion laser with prism-based wavelength tuning).

induce transition to higher energy levels to achieve the condition of population inversion and stimulated emission. An example of gas laser system is shown in Figure 1.12.

### 1.2.2.1 Atomic Gas Laser; He:Ne Laser

He-Ne laser is the simplest and representative of atomic gas lasers. The active medium is a 10 : 1 mixture of He and Ne gases filled in a narrow tube of few millimeter diameters and 0.1-1 m long at a pressure of about 10 Torr. Discharge tube and circuit are very similar as shown in Figure 1.13. A resistant box is used in series with power supply in order to limit the discharge current because tube resistance falls too low once discharge is initiated. Energy levels of Ne atom are directly involved in the laser transitions, and He atoms provide an efficient excitation mechanism to the Ne atoms. Helium atoms from their ground state 1<sup>1</sup>S, are pumped to the excited atomic states 2<sup>1</sup>S and 2<sup>3</sup>S by impact with accelerated electrons in the discharge tube. Neon atoms have 3s and 2s atomic states, which are closer to the 21S and 23S states of the helium atoms, respectively. Collision between excited helium atoms in the 21S and 23S states and neon atoms in the ground states reinforce transfer of energy from helium to neon atoms. Helium atoms in the 21S and 23S states excite neon atoms from ground state to the 3s and 2s states, respectively, and return to the ground state. Excited states 3s and 2s of Ne atom have longer life times as compared to its lower (3p and 2p states), therefore they serve as metastable states and are used in achieving the condition of population inversion between s and p states. Transitions  $3s \rightarrow 3p$ ,  $3s \rightarrow 2p$ , and  $2 \text{ s} \rightarrow 2 \text{p}$  of neon atoms are consequences of lasing at 3.39  $\mu$ m, 632.8 nm, and 1.15 µm wavelengths, respectively. Lifetimes of 3p and 2p atomic states are shorter; therefore Ne atoms from these states rapidly decay to the 1s state by nonradiative transitions. Neon atoms in the 1s state go to the ground state after losing energy through collision with the wall of the tube. The energy level diagram of the He-Ne laser is displayed in Figure 1.14. Another important atomic laser is copper vapor laser, but it is beyond the scope of this book.



Figure 1.14 Energy level diagram for He-Ne laser system.

#### 1.2.2.2 Ion Laser: Argon Ion Laser

#### 1.2.2.2.1 Physical Construction

Argon ion laser is one of the widely used ion gas lasers, which typically generates several watts power of a green or blue output beam with high beam quality. The core component of an argon ion laser is an argon-filled tube made of ceramics, for example, beryllium oxide, in which an intense electrical discharge between two hollow electrodes generates a plasma with a high density of argon ( $Ar^+$ ) ions. A solenoid around the tube (not shown in Figure 1.13) is used for generating a magnetic field, which increases output power of the beam by magnetic confinement of the plasma near the tube axis.

A typical device, containing a tube with a length of the order of 1 m, can generate 2.5–5 W of output power of laser beam in the green spectral region at 514.5 nm, using several tens of kilowatts of electric power. The dissipated heat is removed with a chilled water flow around the tube. The laser can be switched to other wavelengths such as 457.9 nm (blue), 488.0 nm (blue–green), or 351 nm (ultraviolet) by rotating the intracavity prism. The highest output power is achieved on the standard 514.5 nm line. Without an intracavity prism, argon ion lasers have a tendency for multiline operation with simultaneous output at various wavelengths.

### 1.2.2.2.2 Working of Ar Ion Laser

The argon ion laser is a four level laser, which facilitates to achieve population inversion and low threshold for lasing. The neutral argon atoms filled between two hollow electrodes inside the plasma tube (Figure 1.13) are pumped to the 4p energy level by two steps of collisions with electrons in the plasma. The first step ionizes atoms to make ions in the 3p ( $E_1$ ) state, and the second one excites these ions from the ground state  $E_1$  either directly to the 4p<sup>4</sup> levels ( $E_3$ ) or to the 4p<sup>2</sup> levels ( $E_4$ ), from which it cascades almost immediately to the 4p<sup>2</sup> ( $E_3$ ). The 4p ions

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Figure 1.15 Energy level diagram for argon ion laser system.

eventually decay to 4s levels ( $E_2$ ), either spontaneously or when stimulated to do so by a photon of appropriate energy. The wavelength of the photon depends on the specific energy levels involved and lies in between 400 and 600 nm. The ion decays spontaneously from 4s to the ground state, emitting a deep ultraviolet photon of about 72 nm. There are many competing emission bands as shown in Figure 1.15. These can be preferentially selected using a prism in front of one of the end mirrors. This prism selects a specific wavelength to send it back through the cavity to stimulate identical emissions, which stimulates more and more emissions and make regenerative process. This facilitates laser to operate at a single wavelength. Removal of the prism allows for broadband operation, that is, several wavelengths are kept rather than keeping only a particular wavelength. The mirrors reflect a number of lines within a maximum range of about 70 nm. Energy level diagram showing various transitions of Ar ion laser is illustrated in Figure 1.15.

#### 1.2.2.3 Molecular Laser

Unlike isolated atoms and ions in atomic and ionic lasers, molecules have wide energy bands instead of discrete energy levels. They have electronic, vibrational, and rotational energy levels. Each electronic energy level has a large number of vibrational levels assigned as *V*, and each vibrational level has a number of rotational levels assigned as *J*. Energy separation between electronic energy levels lies in the UV and visible spectral ranges, while those of vibrational–rotational (separations between two rotational levels of the same vibrational level or a rotational level of one vibrational level to a rotational level from other lower vibrational level) levels, in the NIR and far-IR regions. Therefore, most of the molecular lasers operate in the NIR or far-IR regions.

### 1.2.2.3.1 Carbon Dioxide (CO<sub>2</sub>) Laser

Carbon dioxide is the most efficient molecular gas laser material that exhibits for a high power and high efficiency gas laser at infrared wavelength. It offers maximum industrial applications including cutting, drilling, welding, and so on. It is widely used in the laser pyrolysis method of nanomaterials processing. Carbon dioxide is a symmetric molecule (O=C=O) having three (i) symmetric stretching [i00], (ii) bending [0j0], and (iii) antisymmetric stretching [00k] modes of vibrations (inset of Figure 1.16), where *i*, *j*, and *k* are integers. For example, energy level [002] of molecules represents that it is in the pure asymmetric stretching mode with 2 units of energy. Very similar to the role of He in He–Ne laser, N<sub>2</sub> is used as intermediately in CO<sub>2</sub> lasers. The first, V = 1, vibrational level of N<sub>2</sub> molecule lies close to the (001) vibrational level of CO<sub>2</sub> molecules. The energy difference between vibrational levels of N<sub>2</sub> and CO<sub>2</sub> in CO<sub>2</sub> laser is much smaller (0.3 eV) as compared to the difference between the energy levels of He and Ne (20 eV) in He–Ne laser; therefore comparatively larger number of electrons in the discharge tube of CO<sub>2</sub> laser having energies higher than 0.3 eV are present. In addition to



Figure 1.16 (a) Absorption, emission curves, and (b) energy level diagram of dye laser system.

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this, V = 1 state of N<sub>2</sub> is metastable, which provides longer time for the collision between excited N<sub>2</sub> molecules and the ground state CO<sub>2</sub> molecules to excite them to (001) state. These two favorable conditions make it easy to attain high level of population inversion between 001 and 100, and 020 vibrational states of CO<sub>2</sub>.

Transitions between 001 initial level to 100 and 020 final vibrational states make stimulated emissions of several IR radiations between 9.2 and 10.6  $\mu$ m wavelengths. Helium gas is also mixed in the gas mixture in order to increase efficiency of lasing. Helium helps in transporting waste heats to the tube wall and de-exciting (100) and (020) energy levels by collision process. The amounts of N<sub>2</sub>, CO<sub>2</sub>, and He in CO<sub>2</sub> laser depends on the type and application of system, but usually, the amount of nitrogen and CO<sub>2</sub> molecules are comparable, while helium concentration is higher than either. Low pressure ( $\sim$ 10 Torr) is generally used for CW lasers, while quite higher pressure is used for high-energy and pulsed laser applications. Depending on power level and beam quality of CO<sub>2</sub> lasers, various internal structures are being used, and they are called *sealed tube laser*, *gas flow laser*, transversally excited atmospheric (TEA) laser, and *gas dynamic laser*. Detailed discussions of these are beyond the scope of the book, although interested readers may consult Ref. [4].

In the far-IR region of  $10\,\mu$ m wavelength, the usual optical material has large absorbance and therefore cannot be used as windows and reflecting mirrors in the cavity. Materials such as Ge, GaAs, ZnS, ZnSe, and some alkali halides having transparency in the IR region are used.

#### 1.2.2.3.2 Nitrogen Laser

Lasing transition in  $N_2$  laser takes place between two electronic energy levels, therefore this laser operates in the ultraviolet region at 337 nm wavelength. Here, upper electronic level has a shorter lifetime compared to the lower one, hence CW operation cannot be achieved, but pulsed operation with narrow pulse width is possible. The pulse width is narrow because as soon as lasing starts, population of the lower state increases, while that at upper state decreases and rapidly a state at which no lasing is possible is rapidly achieved. Such a laser system is known as *self-terminating*.

#### 1.2.2.3.3 Excimer Lasers

Excimers are molecules such as ArF, KrF, XeCl, and so on, that have repulsive or dissociating ground states and are stable in their first excited state. Usually, there are less number of molecules in the ground state; therefore direct pumping from ground state is not possible. Molecules directly form in the first excited electronic state by the combination of energetic halide and rare gas ions. Condition of population inversion can be easily achieved because the number of molecules in the ground state is too low as compared to that in the excited state. Lasing action is done by transition from bound excited electronic state to the dissociative ground state. Population in the ground state always remains low because molecules dissociate into atoms at this point. Usually a mixture of halide such as F<sub>2</sub> and rare gas such as Ar is filled into the discharge tube. Electrons in the discharge tube dissociate and ionize halide molecules and create negative halide ions. Positive Ar<sup>+</sup> and negative F<sup>-</sup> ions react to produce ArF<sup>\*</sup> molecules in the first excited bound state, followed by their transition to the repulsive ground state to commence lasing action. Various excimer lasers are developed in the wavelength range of 120–500 nm with 20–15% efficiency and up to 1 J peak and 200 W average powers. These lasers are widely used in materials processing and characterizations as well as for the pumping of dye lasers.

### 1.2.3 Liquid Laser

Liquids are more homogeneous as compared to solids and have larger density of active atoms as compared to the gasses. In addition to these, they do not offer any fabrication difficulties, offer simple circulation ways for transportation of heat from cavity, and can be easily replaced. Organic dyes such DCM (4-dicyanomethylene-2-methyl-6-p-dimethylaminostyryl-4H-pyran), rhodamine, styryl, LDS, coumarin, stilbene, and so on, dissolved in appropriate solvents act as gain media. When the solution of dye molecules is optically excited by a wavelength of radiation with good absorption coefficient, it emits radiation of longer wavelength, known as fluorescence. The energy difference between absorbed and emitted photons is mostly used by nonradiative transitions and creates heat in the system. The broader fluorescence band in dye/liquid lasers facinates them with the unique feature of wavelength tuning. Organic dye lasers, as tunable and coherent light sources, are becoming increasingly important in spectroscopy, holography, and in biomedical applications. A recent important application of dye lasers involves isotope separation. Here, the laser is used to selectively excite one of several isotopes, thereby inducing the desired isotope to undergo a chemical reaction more readily. The dye molecules have singlet ( $S_0$ ,  $S_1$ , and  $S_2$ ) and triplet ( $T_1$  and  $T_2$ ) group of states with fine energy levels in each of them (Figure 1.17). Singlet



Figure 1.17 Energy level diagram for carbon dioxide (molecular) laser system. Inset shows three mode of vibration of  $CO_2$  molecule.

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and triplet states correspond to the zero and unit values of total spin momentum of electrons, respectively. According to selection rules for transitions in quantum mechanics, singlet-triplet and triplet-singlet transitions are quite less probable as compared to the transitions between two singlet or two triplet states. Optical pumping of dye molecules initially at the bottom of S<sub>0</sub> state transfers them to the top of S1 state. Collisional relaxation of these molecules takes them to the bottom of  $S_1$  state, from where they transit to the top of  $S_0$  state with stimulated emission of radiation. Most of the states in the complex systems are usually neither pure singlet nor pure triplet. Singlet states have small contribution of triplet and vice versa. In the case of most of the dye molecules, unfortunately, T1 state lies just below the  $S_1$ , therefore few molecules transit from  $S_1$  to  $T_1$  by losing some energy through nonradiative transitions. Difference between T<sub>1</sub> and T<sub>2</sub> states is almost same as the wavelength of lasing transition, therefore emitted lasing radiation gets absorbed, which reduces laser gain and may cease the laser action. Therefore, some of the dye lasers operate in the pulsed mode with the pulse duration shorter than the time required to attain a significant population in the state T1. Some of the dyes also absorb laser radiation corresponding to the transition from S1 to upper singlet transitions. Therefore, one should select the dye molecule so that energy differences between these states do not lie between the ranges of laser radiation.

### 1.3 Methods of Producing EUV/VUV, X-Ray Laser Beams

EUV and VUV coherent light sources, that is, EUV/VUV lasers, are in high demand in order to continue the validity of Moore's law in future, in the high-density data writing on disks, materials synthesis and characterizations, and spectroscopic investigations. For example, in photolithography for making the micro/nanopatterns on microelectronic chips, the width of the pattern is proportional to the wavelength of laser light used. Therefore, we would reach a limit for further miniaturization of electronic devices, which causes failure of the well-known Moore's law if shorter wavelength lasers would not be developed. Similarly, for data writing on the optical discs, if we have shorter wavelength lasers, larger amount of data can be written on the same size of disc. In addition to these, such sources are the future of 3D high-density data writing, microscopy at the atomic scale, crystallography, and medical sciences. Following are the methods for developing short wavelength lasers.

### 1.3.1

### Free Electron Lasers (FEL)

In contrast to the other laser sources, free electron lasers (FELs) have an active medium that consists of a beam of free electrons, propagating at relativistic velocities in a spatially periodic magnet (undulator). Here, electrons experience the Lorentz force, execute transverse oscillations, and emit synchrotron radiation in the forward direction (Figure 1.18). We know that an accelerated charged particle



Figure 1.18 Construction geometry of free electron laser system.

moving with relativistic velocity emits radiation, which may be considered as spontaneous. The emitted spontaneous radiation interacts with the electron beam in order to stimulate laser radiation. Since electric field of light is perpendicular to the direction of its propagation, electrons cannot interact with photons unless they also have a velocity component parallel to the light electric field. Hence, electron beam is allowed to travel through wiggler magnet, which generates a spatially periodic magnetic field along the direction of propagation of electron beam. After passing through such field, electrons undergo transverse oscillations as shown in Figure 1.18. In the present case, wavelength of emitted radiation is given by the expression  $\lambda = \lambda_u/2\gamma^2$  where  $\lambda_u$  is the undulator length and  $\gamma$  is the ratio of total electron energy to its rest energy. Similar to other lasers, emitted radiation oscillates between two mirrors of the laser cavity and interacts with the electron beam to enhance power of the laser radiation before leaving the cavity as laser output. Since mirrors have very low reflectivity in the X-ray region, therefore there is severe problem in achieving intense X-ray laser beam. In another mechanism known as self-amplified stimulated emission (SASE), radiation of the spontaneous emission from relativistic electron interacts with the same electron to stimulate it for stimulated emission. Following are the salient features of FELs.

 Tunability: FELs generate coherent, high-power radiation that is widely tunable, currently ranging in wavelength from microwaves, through terahertz radiation and infrared, to the visible spectrum, to soft X-rays. They have the widest frequency range of any laser type. Wavelength of FELs can be easily tuned by varying electron energy and period and amplitude of magnetic field. More shorter wavelength can be produced by harmonic generation.

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  - 2) **Pulse duration:** Based on the electron beam time structure pulse duration ranges from CW to ultrashort pulsed regime (fraction of picoseconds)
  - 3) **Coherence:** It is transverse and longitudinal for oscillators and coherent seed amplifiers, while only transverse for SASE
  - 4) Brilliance: Depending on the status of the art of the electron beam technology, the FEL brilliance can be larger, in some spectral regions (in particular in VUV-X), by many orders of magnitude than the brilliance of the existing sources (lasers and synchrotron radiation).

We do not go into much details, but interested readers are referred to Refs. [6-11].

### 1.3.2 X-Ray Lasers

X-ray lasers provide coherent beam of electromagnetic radiation with the wavelength range from  $\sim$ 30 nm down to  $\sim$ 0.01 nm. The first X-ray laser beam was initiated in 1980 by underground nuclear explosion at Nevada test site, while the first laboratory demonstration of X-ray laser was made in 1984 in the form of Nova laser. Most of the experimental demonstration of light amplification in this spectral range has come from the high-density plasma produced by interaction of high-energy laser with the solid target. Like the above case of FEL, unavailability of good-quality of cavity mirrors has utilized the mechanism of SASE in X-ray lasers. In such case intensity of the laser beam depends on the amplifier length. Gain of the active medium depends on the mass and temperature of the ions and multiplicity and population of the upper/lower levels. Most of the X-ray lasers utilize transitions among L, M, N, and so on, shells of highly ionized atoms. The short life times of the excited states (picoseconds) require very large pumping rates in order to achieve and maintain the condition of population inversion. Laser produced plasmas (LPPs) have high electron temperatures ( $\sim 0.1-1.0$  keV) and densities ( $\sim 10^{18-21} \, \mathrm{cm}^{-3}$ ), which is required for higher degree of ionization and excitation in X-ray lasers. Therefore LPPs are used as primary medium for X-ray lasers. In addition to these, LPPs have uniform density and temperature and thus can provide good media for amplification and propagation of X-rays. Of the various processes suggested, two main (i) collisional excitation and (ii) electron recombination are responsible for attaining the population inversion condition. In case of collisional excitation upper state of the lasing has lower probability of decay with dipole radiation, compared to the lower state, which creates the condition of population inversion between them. In contrast, the rate of population at any state by three-body recombination process is proportional to the forth power of the principle quantum number. The combination of preferential population of upper levels and fast radiative decay of lower states leads to an inversion amongt the  $N = 2, 3, 4, \dots$  states [12].

### 1.3.3 EUV/VUV Lasers through Higher Harmonic Generation

Higher harmonic generation (HHG) is a nonlinear optical process used for the generation of shorter wavelength laser light from the interaction of high-intensity longer wavelength lasers source with nonlinear optical medium. The obtained new frequencies are integral multiples ( $n\omega$ ) of the fundamental ( $\omega$ ) frequency of original laser light. This phenomenon was first observed in 1961 by Franken et al. [13] with ruby laser and quartz as nonlinear medium. The first HHG result was found in 1988, which shows that intensity of the spectra decreases with the increase of order, reaches a plateau, where the intensity remains constant for several orders, and finally ends abruptly at a position called *high harmonic cutoff*. They are portable sources of EUV/soft X-rays, synchronized with the fundamental laser and operated at the same frequencies with much shorter pulse width. These are more spatially coherent compared to X-ray lasers and cheaper than FELs. The harmonic cutoff increases linearly with increasing laser intensity up to the saturation intensity I<sub>sat</sub> where harmonic generation stops. The saturation intensity can be increased by changing the atomic intensities of lighter noble gases, but these have lower conversion efficiency. HHG strongly depends on the driving laser field; therefore the produced harmonics have similar spatial and temporal coherence. Owing to the phase matching and ionization conditions required for HHG, the produced new pulses are with shorter pulse duration compared to the driving laser. Mostly, harmonics are produced in very short time frame, when phase matching condition is satisfied. Instead of shorter temporal window, they emit colinearly with the driving laser pulse and have very tight angular confinement. Gaseous media and LPP on the solid surfaces are two sources used as nonlinear active media for the generation of harmonics.

The shortest wavelength producible with the harmonic generation is given by cutoff of the plateau, which is given by the maximum energy gained by ionized electron from the light electric field. The energy of cutoff is given by  $E_{\text{max}} = I_P + 3.17 \text{U}_P$ , where  $\text{U}_P$  is the pondermotive potential from the laser field and  $I_P$  is the ionization potential. It is assumed that electron is initially produced by ionization into the continuum with zero initial velocity and is accelerated by the laser electric field. After half period of the laser electric field, direction of motion of electron is reversed and it is accelerated back to the parent nuclei. After attaining high kinetic energy of the order of hundreds of electron volts (depending on the intensity of laser field), when electron enters into the parent nuclei, it emits Bremsstrahlung-like radiation during a recombination process with the atoms as it returns back to the ground state. The three-step model of ionization, acceleration of electron, and its recombination with parent nuclei by the emission of EUV photons is shown in Figure 1.19.

Wide wavelength ranges of laser systems starting from hundreds of micrometers (molecular liquid lasers) to X-ray regions (FELs, X-ray lasers, and HHG lasers) with CW and pulsed lasers having various pulse width from milliseconds to attoseconds and repletion rates from single pulse to the megahertz are available nowadays.



**Figure 1.19** Three-step model of ionization and acceleration of electron and its recombination with parent nuclei by the emission of EUV photons.

Figure 1.20 represents the spectrum of available laser system with wavelength range and active media.

## 1.4

### **Properties of Laser Radiation**

Light produced from the lasers have several valuable characteristics not shown by light obtained from other conventional light sources, which make them suitable for a variety of scientific and technological applications. Their monochromaticity, directionality, laser line width, brightness, and coherence of laser light make them highly important for various materials processing and characterization applications. These properties are discussed separately in the following subsections.

### 1.4.1

### Monochromaticity

Theoretically, waves of light with single frequency  $\nu$  of vibration or single wavelength  $\lambda$  is termed as *single color* or *monochromatic* light source. Practically, no source of light including laser is ideally monochromatic. *Monochromaticity* is a relative term. One source of light may be more monochromatic than others. Quantitatively, degree of monochromaticity is characterized by the spread in frequency of a line by  $\Delta \nu$ , line width of the light source, or corresponding spread in wavelength  $\Delta \lambda$ . For small value of  $\Delta \lambda$ , frequency spreading,  $\Delta \nu$ , is given as  $\Delta \nu = -(c/\lambda^2)\Delta \lambda$  and  $\Delta \lambda = (c/\nu^2)$ . The most important property of laser is its spectacular monochromaticity. Based on the type of laser media, solid, liquid, or gas and molecular, atomic, or ions, and the type of excitation, produced laser line consists of color bands that range from broad (as dye laser  $\Delta \lambda \sim 200$ ) to narrow (for gas discharge lines,  $\Delta \lambda \sim 0.01$  nm). Utilizing suitable filters one can get the monochromaticity as good as a single line of lasing transition. But such a single line also contains a set of closely spaced lines of discrete frequencies, known as *laser* 



Figure 1.20 Spectrum of available laser systems, their wavelength range and active media.

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modes (Figure 1.5). Suppressing all other modes excluding the central intense mode using mode locking one can increase monochromaticity of laser line. Suppressing of modes is possible by increasing the separation ( $\Delta v_{sep} = c/2L$ ) between two modes, which can be done by reducing the cavity length. When the axial mode separation approaches the line width,  $\Delta \nu$ , of the lasing transition it is possible that only single mode oscillates. Now, the line width of the laser is equal to that of the single longitudinal mode, which is too narrow. The width of the laser line is directly related to the quality factor Q of the cavity and is given by  $Q = \nu/\Delta\nu$ . The quality factor *Q* actually defines ( $Q = 2\pi \times \text{energy}$  stored in the resonator at resonance/energy) dissipated per cycle) the ratio of energy stored in the cavity at resonance condition and energy dissipated per cycle. High degree of monochromaticity of laser line is required in the diagnosis of closely spaced rotational levels of molecules using selective excitation of that level. The laser line would be absolutely monochromatic if it is oscillating with single frequency, that is, width of the laser line is zero  $(\Delta v = 0)$ . The single-mode laser has the highest degree of monochromaticity, but it has also not achieved the ideal monochromaticity condition.

### 1.4.2 Directionality

One of the most striking properties of laser is its directionality, that is, its output is in the form of an almost parallel beam. Owing to its directional nature it can carry energy and data to very long distances for remote diagnosis and communication purposes. In contrast, conventional light sources emit radiation isotropically; therefore, very small amount of energy can be collected using lens. Beam of an ideal laser is perfectly parallel, and its diameter at the exit window should be same to that after traveling very long distances, although in reality, it is impossible to achieve. Deviation in the parallelism of practical laser beam from the ideal is not due to any fault in the laser design, but due to diffraction from the edges of mirrors and windows. From the theory of diffraction, we know that circular aperture has angle of diffraction given by  $\theta = \sin^{-1}(1.22\lambda/D)$ . The spreading of the beam does depend on the physical nature of aperture and on the type of transverse mode oscillating inside the cavity. In the particular case of TEM<sub>00</sub> mode oscillating inside the nearly confocal cavity, the value of minimum diameter at the center of cavity is given by  $w_0 = (\lambda r/2\pi)^{1/2}$ , where r is the radius of curvature of cavity mirrors (Figure 1.3). The beam diameter, w, varies with distance, z, from the point of minimum diameter and is given by  $w = w_0(2z/r)$ . The beam radius at any point inside and outside the cavity is determined by the distance from the cavity axis where intensity reduces 1/e times of its maximum value.

### 1.4.3 Coherence

Coherence is one of the striking properties of the lasers, over other conventional sources, which makes them useful for several scientific and technological applications. The basic meaning of coherence is that all the waves in the laser beam remain spatially and temporarily in the same phase. Photons generated through stimulated emission are in phase with the stimulating photons. For an ideal laser system, electric field of light waves at every point in the cross section of beam follows the same trend with time. Such a beam is called *spatially coherent*. The length of the beam up to which this statement is true is called *coherence length*  $(L_{\rm C})$ of the beam. Another type of coherence of the laser beam is temporal coherence, which defines uniformity in the rate of change in the phase of laser light wave at any point on the beam. The length of time frame up to which rate of phase change at any point on the laser beam remains constant is known as *coherence time* (*t*<sub>C</sub>). Let P<sub>1</sub> and P<sub>2</sub> be two points on the same laser beam at time *t* and  $t + \tau$ . The correlation function between phases at these two points is termed as mutual coherence function, which is a complex number with magnitude between 0 and 1 (0 for completely incoherent beam and 1 for ideally coherent). Coherence time  $(t_c)$  is also defined as the time taken by the atoms/molecules in active medium to emit a light wave train of length  $L_{\rm C}$ . These two coherences are thus related by  $t_{\rm C} = L_{\rm C}/c$ . The coherence time of the laser beam is almost inverse ( $t_{\rm C} \simeq 1/\Delta \nu$ ) of the width,  $\Delta v$ , of the laser transition. The lasers operating in the single mode (well-stabilized lasers) have narrow line width, therefore they exhibit higher coherence time and coherence length compared to those operating in multimode. Spatial and temporal coherences of continuous laser beams are much higher compared to those of pulsed laser systems because temporal coherence in the pulse lasers are limited by the presence of spikes within the pulse or fluctuation in the frequency of emission.

### 1.4.4 Brightness

Lasers are more intense and brighter sources compared to other conventional sources such as the sun. A 1 mW He–Ne laser, which is a highly directional low divergence laser source, is brighter than the sun, which is emitting radiation isotropically. Brightness is defined as power emitted per unit area per unit solid angle. In the particular case of 1 mW He–Ne laser with  $3.2 \times 10^{-5}$  rad beam divergence and 0.2 mm spot diameter at exit window the solid angle ( $\pi$  ( $3.2 \times 10^{-5}$ )<sup>2</sup>) is  $3.2 \times 10^{-9}$  sr and spot area ( $\pi$  ( $2 \times 10^{-4}$ )<sup>2</sup>) at the exit window is  $1.3 \times 10^{-7}$ m<sup>2</sup>. Thus the brightness of the beam is given by ( $(1 \times 10^{-3})/(1.3 \times 10^{-7} \text{ m}^2 \times 3.2 \times 10^{-9} \text{ sr})$ ) =  $2.4 \times 10^{12} \text{ W/m}^{-2} \text{ sr}^{-1}$ , which is almost  $10^6$  times brighter than the sun ( $1.3 \times 10^6 \text{ W/m}^{-2} \text{ sr}^{-1}$ ).

### 1.4.5 Focusing of Laser Beam

In practice, every laser system has some angle of divergence, which increases the spot size of laser beam and reduces its brightness. If a convergent lens of suitable focal length is inserted in the path of the laser beam, it focuses laser energy into

small spot area at focal point. If  $w_L$  is the radius of the beam and f is the focal length of convergent lens, then radius of the spot at focal point is given as  $r_s = \lambda f / \pi w_L$ , where  $\lambda$  wavelength of laser radiation. If D is the lens diameter and the whole aperture is illuminated by laser radiation (i.e.,  $w_L = D/2$ ) then  $r_s = 2\lambda f / \pi D$  or  $r_s = 2\lambda F / \pi$ , where F = f / D is f number of the lens.

In the case of a particular Nd:YAG laser operating at 1064 nm wavelength and 35 mJ/pulse energy, and 10 ns pulse width. It is focused by a convex lens of F = 5, and whole of the lens area is illuminated by laser. The spot diameter at the focal point is given by  $r_{\rm s} = 2 \times 1.064 \times 10^{-6} \times 5/\pi = 3.4 \,\mu\text{m}$ . The irradiance of the laser beam is given by I = E(J)/pulse width (s)  $\times (\pi r_{\rm s}^2) = 35 \times 10^{-3}/(10 \times 10^{-9} \,\text{s} \times \pi (3.4 \times 10^{-6})^2) \approx 10^{16} \,\text{W m}^{-2}$ .

### 1.5

### **Modification in Basic Laser Structure**

Addition of some electronic, optical, or electro-optical systems between the active media and mirror to modify the pulse width, pulse shape, and energy/pulse and generation of integral multiple of laser frequency is important for advanced technological applications. Mode locking or phase locking, Q-switching, pulse shaping, pulse compression and expansion, frequency multiplication, and so on, are some commonly used methods in advanced laser technology.

### 1.5.1

#### Mode Locking

#### 1.5.1.1 Basic Principle of Mode Locking

Mode locking is a technique in optics by which a laser can be made to produce light pulses of extremely short duration of the order of picoseconds ( $10^{-12}$  s) or femtoseconds  $(10^{-15} \text{ s})$ . The basis of this technique is to induce constant phase relationship between the modes of laser cavity. Simply, same phase of  $\delta$  can be chosen for all laser modes. Such a laser is called mode-locked or phase-locked laser, which produces a train of extremely narrow laser pulses separated by equal time intervals. Let N modes are oscillating simultaneously in the laser cavity with  $(A_0)_n$ ,  $\omega_n$ , and  $\delta_n$  being the amplitude, angular frequency, and phase of the nth mode. All these parameters vary with time, therefore modes are incoherent. The output of such laser is a linear combination of *n* different modes and is given by  $A(t) = \sum_{n=0}^{N} (A_0)_n e^{i(\varpi_n t + \delta_n)}$  expression. For simplicity, frequency of the *n*th mode can be written as  $\omega_n = \omega - n\Delta\omega$ , where  $\omega_n$  is the mode with highest frequency and  $\Delta \omega = c\pi/L$  is the angular frequency separation between two modes. If all the modes have same amplitude and we force the various modes to maintain same relative phase  $\delta$  to one another, that is, we mode lock the laser such that  $\delta_n = \delta$ , then the expression for resultant amplitude will be  $A(t) = A_0 e^{i(\varpi t+\delta)} \sum_{n=0}^{N} e^{-i\pi nct/L} = A_0 e^{i(\varpi t+\delta)} \sin(N\phi/2) / \sin(\phi/2),$ where  $\phi = \pi ct/L$ . The irradiance of the laser output is given by  $I(t) = A(t)A(t)^* =$  $A_0^2 \sin^2(N\phi/2)/\sin^2(\phi/2)$ , which is the periodic function of the period  $\Delta \phi = 2\pi$  in the time interval t = 2L/c (time of round-trip inside the cavity). The maximum value of irradiance is  $N^2A_0^2$  at  $\phi = 0$  or  $2p\pi$  (*p* is integer). Irradiance has zero value for  $N\phi/2 = p\pi$ , where *p* is an integer with values neither zero nor a multiple of *N*. This makes  $\phi = 2p\pi/N = \pi ct/L$  or t = (1/N)(2L/c)p. Therefore, separation between two consecutive minima, that is, pulse width of a single laser pulse is  $\Delta t = (1/N)(2L/c)$ . Hence, the output of a mode-locked laser has sequence of short pulses of pulse duration (1/N)(2L/c) separated in time by 2L/c. The ratio of pulse separation to the pulse width is equal to the number of modes *N*, which shows that there should be a large number of modes in the cavity in order to get high-power short duration (picoseconds and femtoseconds) laser pulses.

### 1.5.1.2 Mode Locking Techniques

#### 1.5.1.2.1 Active Mode Locking

We have discussed that mode locking is achieved by inducing the longitudinal mode to attain the fixed phase relationship, which may be exploited by varying the loss of the laser cavity at a frequency equal to the intermode separation c/2L. Let us consider a shutter between the active medium and output mirror, which is closed for most of the time and is opened after every 2L/c seconds (corresponding to the time of round-trip) and remains open for short duration of (1/N)(2L/c) seconds. If the laser pulse train is as long as the shutter remains opened and arrives at the shutter exactly at the time of its opening, the pulse train is unaffected by the presence of shutter. The segment of the pulse, which arrives before opening and after closing of the shutter, will be clipped. Thus phase relationship of the modes is maintained by periodic oscillation of the shutter. An electro-optical or acousto-optical crystal operating on the principle of Pockels or Kerr effect respectively, may be used as a periodic shutter.

In the former case of electro-optical switching, a polarizer and an electro-optical crystal are arranged in between the active medium and laser exit mirror so that laser beam passes through the polarizer before entering into the crystal. Laser light from the active medium passes through the polarizer and gets plane polarized. When this polarized beam passes twice through the crystal (with appropriate electric field along the direction of light propagation) before returning to the polarizer, the plane of polarization is rotated by an angle of  $90^{\circ}$ , which does not allow the light beam to enter into the active media through the polarizer. In other words, the shutter is effectively closed. If there is no field along the crystal in the direction of propagation of light, there will be no rotation of the plane of polarization of light and it can pass though the polarizer and enter into the active medium (shutter is open). Similarly in acousto-optical switching, a 3D grating pattern is established in a medium (water or glass, not active medium of lasing) by the incident and reflected sound waves created by piezoelectric transducer attached at one end of the medium. This grating diffracts a part of the laser beam and creates a high loss.

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#### 1.5.1.2.2 Passive Mode Locking

Passive mode locking method consists of placing a saturable absorber inside the cavity. Saturable absorbers are molecules having a nonlinear decrease in absorption coefficients with the increase in the irradiance of light. The saturable absorber is placed between active laser medium and mirror. If an intense pulse of light passes through the saturable absorber placed inside the laser cavity, the low-power tails (weaker modes) of the pulse are absorbed because of the absorption of dye molecules. The high-power peak of the pulse is, however, transmitted because the dye is bleached. Owing to this nonlinear absorption, the shortest and most intense fluctuation grows, while the weaker dies out.

1.5.2

### Q-Switching

*Q-switching*, sometimes known as *giant pulse formation*, is a technique by which a laser can be made to produce a pulsed output beam. The technique allows the production of light pulses with extremely high (of the order of approximately gigawatts) peak power, much higher than would be produced by the same laser if it were operating in a CW mode. Compared to mode locking, another technique for pulse generation with lasers, Q-switching leads to much lower pulse repetition rates, much higher pulse energies, and much longer pulse durations. Both techniques are sometimes applied at once. In contrast to mode locking, where we achieve a train of pulses, Q-switching provides a single strong and short pulse of laser radiation.

Q-switching is achieved by putting a variable attenuator inside the laser's optical resonator. When the attenuator is functioning, light that leaves the gain medium does not return and lasing cannot begin. This attenuation inside the cavity corresponds to a decrease in the *Q* factor or quality factor of the optical resonator. A high Q factor corresponds to low resonator losses per round-trip, and vice versa. The variable attenuator is commonly called a "Q-switch," when used for this purpose. Initially the laser medium is pumped while the Q-switch is set to prevent feedback of light into the gain medium (producing an optical resonator with low Q). This produces a population inversion, but laser operation cannot yet occur since there is no feedback from the resonator. Since the rate of stimulated emission is dependent on the amount of light entering the medium, the amount of energy stored in the gain medium increases as the medium is pumped. Owing to losses from spontaneous emission and other processes, after a certain time, the stored energy will reach some maximum level; the medium is said to be gain saturated. At this point, the Q-switch device is quickly changed from low to high Q, allowing feedback and the process of optical amplification by stimulated emission to begin. Because of the large amount of energy already stored in the gain medium, the intensity of light in the laser resonator builds up very quickly; this also causes the energy stored in the medium to be depleted almost as quickly. The net result is a short pulse of light output from

the laser, known as a *giant pulse*, which may have very high peak intensity. Similar to mode locking, active and passive techniques are used for Q-switching. Electro-optical and opto-acoustic (active Q-switching) switches, and saturable absorbers (passive Q-switching) are used to prevent feedback signal into the active media.

### 1.5.3 Pulse Shaping

Pulse shaping is a technique of optics, which modifies temporal profile of a pulse from laser. It may lengthen or shorten pulse duration, can generate complex pulses, or generate multiple pulses with femtosecond/picosecond separation from a single laser pulse. A pulse shaper may act as a modulator. Modulating function is applied on the input pulse to get the desired pulse. The modulating function in pulse shapers may be in time or a frequency domain (obtained by Fourier transform of time profile of pulse). In the pulse shaping methods optical signal is converted into electronic signal, where the presence and absence of pulses are designated by 1 and 0, respectively. There are two well-known pulse shaping techniques (i) direct space to time pulse shaper (DST-PS) and (ii) Fourier transform pulse shaper (FT-PS). In DST-PS, electrical analog of the desired output signal is used as a modulator with the electrical analog of input pulses. In contrast to DST-PS, the FT-PS uses a modulating function, which is a Fourier transform of the required sequence. In other words, a specific temporal sequence of pulses, the modulating function, is its Fourier transform and acts in frequency domain.

Optical design is a common experimental arrangement consisting of a combination of two sets of grating, a pair of mirrors, a pair of convex lenses, and a pulse shaper (see Figure 1.21a). By placing different types of optical components we can design various types of laser pulses. For example, employing suitable filter at focal plane can remove optical radiation of undesired frequency. A slab of transparent material with varying thickness can offer different path lengths for different frequency components and can decide which component of light will come out first. A concave lens at the focal plane provides longer path length for higher frequency component, and vice versa (Figure 1.21b). This causes the lower frequency component to come first and the higher frequency one comparatively later, resulting in positively chirped<sup>1)</sup> output pulse (frequency increases with time). In contrast to this, a planoconvex or biconvex lens makes longer path length for lower frequency, and vice versa, which makes higher frequency to come out first and shorter frequency after that, that is, negative chirped output. Both positive and negative chirping lengthens the pulse duration, that is, stretching of the laser pulse. Removing or reducing the chirp causes compression (reduction of pulse duration) of the pulse.

 Chirping is a mechanism by which different components of light frequency of a pulse from laser comes out at different time. When frequency of light components of pulse increases with time it is called *positive chirping*, decreases with time negative chirping and if constant with time no chirping (un-chirped).

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Figure 1.21 (a) Basic optical geometries for laser pulse shaping, (b) positive chirped, and (c) negative chirped pulse shaping.

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