Tunable Solid-State Lasers

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Invited Paper

Tunable lasers based on ion-doped solid-state media have emerged from the laboratory into commercial production in the last decade, and have found significant application in photonic-systems research. In the future, they may be a critical component of space-based remote sensing systems, communications systems and laser medicine. In this article we review the basic physics of tunable solid-state lasers and discuss the characteristics of the most significant established and emerging systems.

I. INTRODUCTION

In the last decade tunable solid-state lasers have become an increasingly significant component of quantum electronics. While they were first demonstrated in the 1960's, they have only recently emerged from being a laboratory curiosity to playing key roles in variety of electro-optics systems. Compared to what, until recently, was the most widely used tunable system, the liquid-medium dye laser, solid-state laser media offer unlimited operating and "shelf" lifetimes along with the capability to store energy and thus generate high peak powers via Q-switching. In addition, for some systems one can obtain either a broader tuning range than a given dye or an extension to longer infrared wavelengths. Current applications of tunable solid-state lasers include basic scientific investigations in atomic, molecular and solid-state spectroscopy, laboratory studies of new semiconductor and fiber-optic devices, generation of ultrashort pulses and amplification of such pulses to high peak powers. Future applications now under development include aircraft-and space-based remote-sensing lidars, submarine communication systems and laser medicine. This article will review the field by first providing a brief discussion of the physics of tunable solid-state lasers and then examining a variety of laser systems, as categorized by the laser-active ion. (To be precise, we will cover paramagnetic-ion lasers only; other solid-state laser media employing so-called color centers are considered as an entirely different class of laser systems.)

II. PHYSICAL BACKGROUND

Solid-state lasers operate on stimulated transitions between electronic levels of ions (activators) contained in solid crystalline or glassy media (hosts). To date, activators of practical significance have been positively charged ions from the rare Earth or 3d transition-metal groups of the periodic table. While all solid-state lasers can operate over some range of wavelengths, and thus are tunable, it is common to consider "tunable" lasers as those capable of covering a wavelength range greater than several percent of the laser central wavelength. In the following we discuss some basic laser concepts and consider the mechanisms important for making a laser "tunable."

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A. Linewidth, Cross Section, and Lifetime

The physical characteristic determining tuning range is the linewidth of the laser transition. Two other quantities of general interest to laser design are the gain cross section and the lifetime of the upper laser level.

If we establish, by optical pumping, a population density, N (per cm^3), of ions in upper laser level, and limit our discussion to four-level lasers where the population of ions in the lower level can be neglected, then the optical gain, G, in a laser medium of length l (in cm) is given by the expression

$$G = \exp(\sigma(\lambda)Nl)$$

where $\sigma(\lambda)$ is the gain cross section in cm^2, as a function of wavelength λ. The linewidth of the laser transition is a measure of the range of wavelengths over which the cross section is large, and is usually given as the full-width at half-maximum, i.e., the span between the half-gain points on either side of the peak wavelength. (Unfortunately, in the field of quantum electronics, wavelength and frequency are interchanged all to frequently. We will generally stay with wavelength as the convention for this paper when referring to laser radiation, except for cases where the use of frequency is common.)

The population of the upper laser level, in the absence of laser action, decays at a rate determined by the combined effects of radiative (or, spontaneous) emission and other nonradiative processes considered below. In simple systems the rate is constant, which leads to an exponential decay of the population after the pumping is turned off, and a characteristic lifetime given by the inverse of the decay rate. Lifetimes for levels used in solid-state lasers are in the range

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from 10^{-8} to 10^{-2} s. The upper-level lifetime is important in determining how much pumping power is required to generate the necessary population of ions for laser action. For a desired steady-state population, N, the pump rate, W, in photons/s is simply $N\tau$, where τ is the lifetime.

If the only way the upper laser level decays is by spontaneous emission to the lower laser level, the relation (first developed by Einstein) between stimulated and spontaneous emission can be used to connect the gain cross section, lifetime and linewidth by the following expression:

$$\sigma(\lambda) = F(\lambda)\lambda^5 \cdot [8\pi n^2 c\tau \int F(\lambda)\lambda d\lambda]^{-1}$$

where $F(\lambda)$ is the measured spectral emission curve for the laser transition, in power per unit wavelength interval, n is the refractive index of the laser crystal, and c is the speed of light. If $F(\lambda)$ can be fit to a simple function, such as a Gaussian distribution, the above equation can be simplified, but the basic result of the equation is the product of the gain cross section and τ is inversely proportional to the linewidth.

For tunable systems with large linewidths, large gain cross sections are possible only if the lifetime is short. Conversely, for constant lifetimes, the gain cross section is inversely related to the linewidth. If one assumes a fixed length of laser material and certain level of gain needed for laser operation, one can readily show that the pumping power required to obtain steady-state laser operation is proportional to the linewidth. These kinds of considerations led early investigators to search for solid-state laser materials with narrow linewidths. As techniques for optical pumping improved, including the use of another laser as the pump source, operation on systems with large linewidths became possible.

B. Ions in Host Crystals

A solid-state laser host provides more than just a means for containment of the laser-active ions. In most systems, the environment surrounding the ion serves to modify the ion electronic states in a favorable way, by making possible the interaction between electromagnetic radiation and the electronic states necessary for stimulated emission. In terms of quantum mechanics, the environment can induce between electronic states a dipole moment that would not exist if the ion was in free space. For example, many of the laser transitions we will discuss below involve electrons that stay in the 3d state, and in free space the electric dipole moment for such transitions is parity forbidden, i.e., zero.

Another function of the host crystal is to serve as a sink for excess energy produced by the pumping and lasing cycles of the solid-state system. Typically, the laser ions are excited by optical pumping, and if the pump source is either a gas discharge or tungsten lamp, much of the pump energy is absorbed into ion states higher in energy than that of the upper laser state. Decay of the excited ion from the higher states into the upper state must occur rapidly, accompanied by a loss of ion energy, if the pumping process is to be effective. In a four-level laser system even more energy must be lost after the laser transition takes place, as the ion needs to rapidly decay from the lower laser level into the ground state. The host crystal creates a possibility for rapid loss of energy during the lasing cycle by allowing "nonradiative" transitions to occur between various states of the ion.

In a nonradiative transition, some of the ion energy is converted to mechanical energy in the form of vibrations of the atoms in the host medium, because of vibrational-electronic interaction, or coupling. A quantum-mechanical treatment of vibrations in a solid yields a large set of discrete vibrational states, which are represented by a set of quasiparticles called phonons. Phonon energy is simply the vibrational frequency times $\hbar$, Planck's constant; the properties of the host crystal (number and types of atoms in the unit cell, the overall arrangement of atoms in the crystal structure and the bonding strength between atoms) determine the number of distinct types of phonons and the distribution of phonon energies. All solid media have some characteristic maximum vibration frequency and thus a maximum phonon energy. The total number of phonons present in the host medium is related to the medium temperature. Every time a nonradiative transition occurs phonons are created or, in more classical terms, the medium temperature increases. The rates at which nonradiative transitions take place cover a wide range and depend on a number of variables, including the properties of the active ion, the host crystal, the crystal temperature and the amount of energy given up into phonons. In many systems the rate is greater than 10^9/s, and this is rapid enough for most laser pumping cycles. Even when the energy gap between levels is greater than the maximum phonon energy, there exist processes in which multiple phonons can be created simultaneously, although the rate for such processes decreases rapidly as the number of phonons increase.

While nonradiative transitions are crucial for promoting the decay of levels, they present a problem if they also cause decay of the upper laser level. As we will discuss later, the presence of nonradiative decay of the upper level can have a major effect on the performance of some solid-state tunable lasers. In a host crystal in which all the ions are in the same environment, the fluorescence quantum efficiency for a transition is the spontaneous emission rate divided by the total transition rate. When all the decay is by photon emission the fluorescence quantum efficiency is unity.

C. Broad Linewidths

We first consider the mechanisms giving rise to the finite gain linewidth of a solid-state laser. Unlike gas lasers, the Doppler effect due to the motion of the ions in the host is not of significance. Vibrational frequencies range up to 10^{13} Hz, but the ions are constrained to tiny displacements (10^{-11} m) by the strong forces holding the host medium together, and the resultant maximum velocities and Doppler shifts are relatively small.

The most fundamental cause of a finite linewidth is the finite lifetimes of the upper and lower laser levels, since the uncertainty relation requires that the linewidth of any

level is the inverse of the level lifetime. To invoke quantum mechanics again, we note that the wavefunction for a given ion level has both an amplitude and a phase. In calculating the value to be used for the lifetime of the level we must consider processes which not only cause decay out of the level, but also processes which change the phase. For all but the lowest temperatures the latter occur at a much higher rate than the former, and are due to interactions with the phonons of the host medium. In many common fixed-wavelength solid-state lasers, the linewidth is related to finite level lifetimes; values at room temperature range from 100 GHz and on up. Two well-known examples are ruby, with the transition-metal active ion Cr^{3+} in the host Al_2O_3 (sapphire), and Nd:YAG with the rare-Earth active ion Nd^{3+} in the garnet-structured host $Y_3Al_5O_{12}$.

Lifetime broadening is homogeneous, that is, all the ions in the host are subject to the same broadening mechanism. For disordered host materials, such as glasses, another type of broadening occurs because each ion is in a slightly different environment from another. Since the energy levels of the ion depend to some extent on the surroundings, a distribution of ion energy levels results. The enormous number of ions that interact with a typical optical beam effectively create a smooth distribution of energies, and transitions between levels have a broadened lineshape. This type of broadening is referred to as inhomogeneous, since the properties of ions vary from one to the other. Familiar systems with inhomogeneous broadening include various Nd^{3+}-doped glasses used as active media for laser drivers in inertial-confinement-fusion experiments and another rare Earth ion, Er^{3+}, doped in silica fibers and used as amplifiers for 1550-nm, fiber-transmission systems.

The mechanism giving rise to the large linewidths of nearly all broadly tunable solid-state lasers is different from the two mentioned above, and comes about because of even stronger interaction between ions and the host than previously described. We refer to Fig. 1 to illustrate the mechanism. In the figure we plot energy as a function of the distance between an individual laser-active ion and the surrounding ions of the host crystal, for two different electronic states (1 and 2) of the ion and for two different levels of interaction. Our energy plot is the combined electronic energy of the ion and vibrational (i.e., mechanical) energy of the host crystal, and is referred to as a configuration-coordinate diagram. The energy versus distance relation is in the form of a parabola, a quadratic approximation to the manner in which the atoms of the crystal react to a change in spacing. The host vibrations we referred to earlier occur about the lowest energy point of the parabola, the equilibrium position. For this discussion we consider a simple mode of vibration in which all the atoms surrounding the laser-active ion move in and out at the same time, the "breathing" mode. (In real host crystals there are many different modes of vibration, with much greater complexity of motion than the breathing mode, and each has a particular interaction with the electronic states of the active ion.)

When the active ion changes from state 1 to state 2, the equilibrium position may not be affected at all, as indicated in Fig. 1(a). This is typical for the 4f energy states of rare-Earth ions such as Nd, which have weak interactions with their surroundings. On the other hand, if the surrounding atoms are affected by the active-ion electronic state, they may react by establishing a new equilibrium position, as shown in Fig. 1(b). The latter case leads to the broad linewidths characteristic of most tunable solid-state lasers, as we illustrate in Fig. 2.

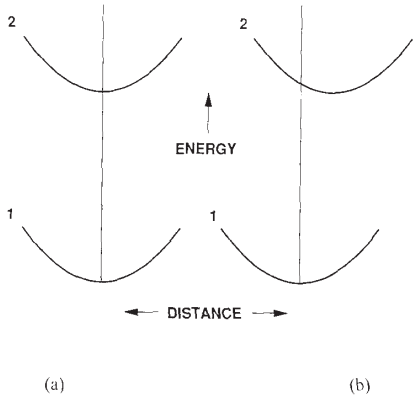

Fig. 1. Energy as a function of distance between active ion and surrounding ions of host crystal, for two different electronic states, 1 and 2. In (a), surrounding ions do not shift equilibrium positions when active ion changes electronic states, while in (b) a shift occurs.

Consider first an electronic transition from state 1 to state 2, which would result from the absorption of energy. An important point about the transition is that the time in which it takes place is much shorter than the inverse of the vibrational frequencies of the atoms in the host medium. As a result, the atoms are essentially "frozen" during the transition. Since the atoms are vibrating, there is in fact a distribution of possible positions for the atomic configuration at the point of an electronic transition, and as Fig. 2 indicates, this leads to a wide distribution of possible transition energies, and thus a broad linewidth. Note that a similar transition for the system represented in Fig. 1(a) would have an energy independent of the atomic position, as the two parabolas have exactly the same spatial function.

Immediately after the upwards transition both the host crystal and the active ion are in higher-energy states. The first component to lose energy is the host crystal, and it does so when the highly localized vibrational energy around the ion is dissipated into the rest of the crystal. If we invoke the concept of phonons again, we would say that phonons local to the ion are "emitted" and travel away from the ion. After this happens the combined ion-crystal system reaches the "relaxed" excited state shown in Fig. 2, in which the crystal is in equilibrium, while the ion is still in an electronic excited state.

In the absence of laser action, the ion makes an electronic transition from state 2 to state 1 due to a combination of spontaneous emission and nonradiative processes. With laser action, stimulated emission provides another channel

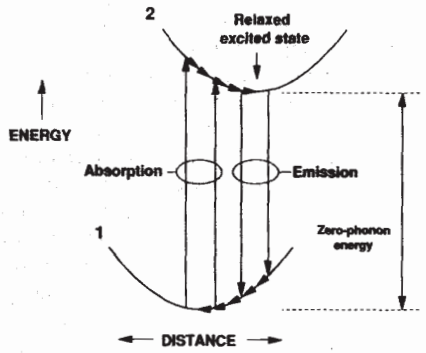

Fig. 2. Energy diagram similar to Fig. 1(b) with details of upward (absorption) and downward (emission) electronic transitions shown. Absorption and emission are spread in energy due to motion about equilibrium position of ions. "Zero-phonon" energy, the purely electronic difference in energy between states 1 and 2 is indicated.

for the transition. As in absorption, the transition linewidth is broad, but the distribution of emission energies peaks at a lower energy than that for absorption. After the downwards transition occurs, the crystal is again left in an excited vibrational state, and relaxation down to the equilibrium condition for state 1 progresses rapidly through phonon emission.

Figure 2 illustrates how the cycle of absorption, phonon emission to the relaxed excited state, stimulated emission into the lower level and phonon emission into the relaxed lower state is equivalent to a four-level system of discrete electronic levels. The difference is that the energy levels are a combination of both electronic and vibrational states, which leads to the use of the term "vibronic" (vibrational-electronic) to describe the types of transitions involved. Other terms applied are "phonon-terminated" and the less accurate "phonon-broadened," the latter of which also applies, as we have noted above, to transitions of the type shown in Fig. 1(a).

In Fig. 2 we show a classical view of how smooth and broad lineshapes for absorption and emission result from the difference in energy between two displaced parabolas. A more rigorous quantum-mechanical analysis of the lineshapes relies on a calculation of overlap integrals between wavefunctions of the phonons in states 1 and 2. This analysis predicts that the energies of vibronic transitions should appear as discrete values, separated by the energy of an individual phonon and clustered on either side of a "zero-phonon" energy. The latter is indicated in Fig. 2 and is the difference between the purely electronic energy of the two states. If there was no displacement of parabolas the zero-phonon line would be the observed transition energy in absorption and emission. As the displacement between the parabolas increases, theory shows that the intensity of the zero-phonon transition decreases while that of vibronic transitions increases. In actuality the discrete vibronic transitions predicted by quantum mechanics are rarely observed, for several reasons. First, each vibronic transition has a large linewidth from lifetime broadening, and the individual vibronic lines often overlap enough to produce a broad, featureless overall lineshape. Second, typically many different phonons, rather than one, interact with the electronic transition, and the effect of the resultant distribution of phonon frequencies obscures individual vibronic lines.

We have assembled, in Fig. 3, a composite of the absorption and emission spectra for many of tunable solid-state laser systems we will discuss in more detail below. In all cases we show the emission band associated with laser operation and the absorption band for the same electronic transition. For some systems we also include higher-lying absorption bands from other electronic transitions. In the systems considered emission bands connected with the more energetic transitions are barely, if at all, observable, as the excitation energy decays rapidly via phonon emission into the lowest-lying electronic excited state.

D. Energy Storage and Extraction

We now consider some of the issues related to Q-switching of and short-pulse amplification by tunable solid-state lasers. One of the favorable attributes of solid-state lasers in general is their ability to generate high peak powers by the Q-switching process. In Q-switching, the laser material is excited by the pump source but laser action is prevented by maintaining the laser cavity in a high-loss (low Q) state. When a high level of excitation in the upper level is attained, the cavity is switched to a high-Q state i.e., the cavity loss is reduced and the laser output rapidly builds up from noise to a point at which a good fraction of the energy stored in the laser medium as excitation is extracted. If the rate of extraction is much faster than the rate at which excitation in the medium can build up again, the laser output falls to zero in a short time. (After the extraction occurs the gain in the medium becomes less than the loss in the laser cavity.) Thus a pulse is generated.

Energy in the laser cavity in the absence of gain in the laser medium decays exponentially with a characteristic time τ_c, related to the round-trip time for the cavity and the loss fraction on each round-trip. Typical values for τ_c are in the ns range and Q-switched output pulsewidths are some multiple of τ_c. A crude estimate of the ratio of the peak power generated by Q-switching to the power under normal conditions is τ/τ_c. The long upper-state lifetimes for most solid-state laser materials lead to high Q-switched powers, in contrast to dye and some gas media, which have ns-range values for τ.

Let us consider how energy storage affects the generation of high Q-switched powers. If the gain cross section for a material is too high, it may be difficult to store much energy in the laser medium before the Q-switch changes to the low-loss state. Even for switches with essentially infinite loss, such as rotating mirrors, stored energy can be lost through parasitic oscillations within the laser medium. Solid-state lasers are particularly prone to parasitics, given that the medium has a high refractive index. High indices make possible unwanted laser cavities formed by reflections off

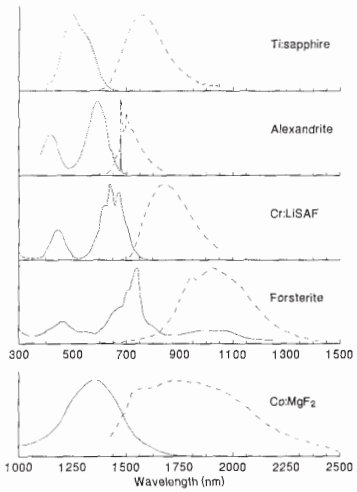

Fig. 3. Absorption (solid line) and emission (dashed line) lineshapes as a function of wavelength for several combinations of laser-active ions and host crystal. The sharp line in the alexandrite emission curve is off-scale. The absorption and emission bands in forsterite result from both Cr^{3+} and Cr4+ ions.

the ends of the media or by internal "bounce" paths based on total internal reflection. Even in systems where parasitic oscillations have been eliminated, ASE (amplification in the medium of spontaneous emission from the upper laser level) serves to deplete upper-level stored energy.

For amplification of short pulses, energy storage is also an issue. In a manner similar to Q-switching, a short-pulse amplifier is pumped up to a high excitation level and then the pulse to be amplified is passed through the laser medium. The amount of amplification is set by limits to the medium stored energy.

In Nd:YAG practical levels of energy storage are reduced by both parasitics and ASE to levels well below the theoretical maximum possible, when all of the Nd ions are pumped to the upper laser level. The relatively (compared to Nd:YAG) low gain cross sections of most tunable solid-state lasers permit high levels of energy storage in both Q-switching and amplification, in some cases close to the theoretical limit.

Unfortunately, a competing issue for Q-switched systems, the ability to extract energy, is a major problem for many tunable systems. An estimate of the energy fluence (J/cm^2) in a Q-switched laser cavity can be obtained by calculating, from the gain cross section, the saturation fluence E_{sat} for the laser transition, given by:

$$E_{sat} = hc/(\lambda\sigma)$$

where h is Planck's constant. Note that the above expression is simply the energy of a laser photon divided by the gain cross section. The actual fluence in the laser cavity is some factor, proportional to the pumping ratio (pump energy/threshold pump energy) multiplied by E_{sat}.

Typical laser-induced damage levels for coated optics and laser media surfaces fall in the range 5–30 J/cm^2, for Q-switched pulses of approximately 10–30 ns in duration. For Nd:YAG lasers, with an E_{sat} of 0.4 J/cm^2, damage and thus energy extraction is not a major concern, but as we point out below, values of E_{sat} for many tunable laser media are 10–100x this level, and energy extraction from Q-switched oscillators is damage-limited.

For short-pulse amplifiers, as long as the output fluence of the amplifier is well below E_{sat} the pulse amplification is linear and energy extraction is not an issue. However, most amplifiers are operated to extract as much possible stored energy as possible from the medium, and simple analysis shows that a high fraction of the stored energy can be obtained when the output fluence is at least twice E_{sat}. As for Q-switched systems, the high values of E_{sat} for many tunable media prevent efficient amplifier operation.

E. Excited-State Absorption

As a final part of our background discussion, we consider the issue of excited-state absorption (ESA), a process that for many tunable solid-state lasers can have a profound effect on system performance. Consider the energy diagram of Fig. 4, in which we include electronic states above the upper energy state for a laser transition. (We have eliminated the atomic displacement to simplify the diagram.) Light can be either amplified by stimulated emission, where the ion drops to a lower-energy state, or can be attenuated through ESA, in which the ion makes a transition from the upper laser level to the higher-lying level. The net optical gain in the system is simply the difference between the cross section for stimulated emission and the cross section for ESA.

If the cross section for ESA is greater than that for stimulated emission laser action is impossible, no matter how high the level of pumping. Even if the net gain is positive, ESA raises the threshold pump power for laser action and reduces the efficiency in converting pump power to laser output power. A variation on ESA is sometimes observed, in which the pump light is absorbed by a transition originating on the upper laser level. The effect of "pump ESA" depends, among other things, on the level of population in the upper level under conditions of laser action. It is particularly detrimental when Q-switched operation is desired, since the upper population can rise to high levels before the Q-switch is changed to the low-loss state.

Prediction of both the magnitude and wavelength dependence of ESA cross sections is difficult for most all tunable solid-state lasers. Our ability to generate the energy curves shown in Fig. 2 from theory is too limited in accuracy for transition-metal ions in crystals. Conventional absorption measurements only access transitions from the ground state of the ion to higher lying states, and are generally not indicative of the nature of transitions from the "relaxed" excited state of the upper laser level. ESA cross sections can be estimated by measuring the optical gain (or loss) in a pumped crystal as a function of wavelength.

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