Tunable Solid State Lasers

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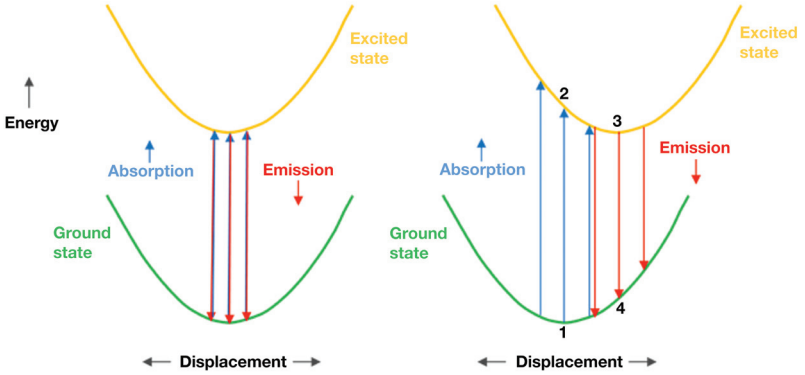
While the wavelength of any laser can be varied, lasers get classified as tunable when their tuning range becomes a substantial fraction of their center wavelength. Despite having lower optical gain than narrow-line rare-earth doped crystal lasers such as Nd^{3+}-doped YAG, tunable lasers are desirable for a number of reasons. In laser-based spectroscopy, laser tuning allows one to access spectral features of interest, while in laser propagation through the atmosphere, tuning can be used to avoid atmospheric absorption lines. A large tuning range implies the ability to generate and amplify short pulses of light. The development of practical and efficient tunable solid state lasers has led to a scientific revolution and an emerging industrial revolution in laser processing of materials, based on the generation of electromagnetic pulses with femtosecond and recently attosecond duration.

Most broadly tunable lasers employ ions from the "3d" portion of the periodic table. Figure 1 presents so-called configuration-coordinate diagrams that help explain the broad tunability of 3d ions. The diagrams are a greatly simplified schematic representation of the combined energy of the laser-active ion and its environment as a function of the positions of the atoms surrounding the ion. In equilibrium, the overall energy is minimized, and the system energy increases as the coordinate deviates from the equilibrium position. Deviation occurs as a result of the always present vibrations of the atoms, which appear even at the lowest temperatures from the uncertainty principle of quantum mechanics. The left-hand diagram shows the case where, when the ion energy level changes from a "ground" state to an "excited" state, the equilibrium position for the configuration coordinate is unchanged. The right-hand side shows the case where the equilibrium position does change.

An important concept regarding the linewidth of the transitions between the ion ground and excited states is the Franck–Condon principle. Stated in classical terms, when an active ion undergoes a transition, it occurs so quickly that the atomic surroundings do not move, as shown by the vertical arrows in the diagrams. The left-hand diagram is representative of the type of narrow-linewidth transitions among levels of the rare-earth ions, since changing the electronic state of the spatially compact wavefunctions of the rare earths has negligible effect on the surrounding atoms.

The electronic wavefunctions of 3d ions have a larger spatial extent than those of rare-earths and have a stronger interaction with their environment. The case illustrated in the right-hand diagram shows what happens with a strong interaction, exciting the electronic level leads to a new equilibrium position. As is evident from the arrows, the energy associated with ground-to-excited-state transitions does vary with the displacement, leading to a large spread in energies and hence a large linewidth. The energies for the absorption (ground-to-excited) transitions are generally distinct and higher than those for emission and possible laser operation (excited-to-ground transitions). As a result, even with only two electronic transitions, one can observe four-level laser operation (as shown by the numbers in Fig. 1) as the peak absorption and emission wavelengths do not overlap. These types of transitions are often referred to as "vibronic," a concatenation of vibrational and electronic.

After the demonstration of the ruby laser, and around the same time as the development of rare-earth-doped lasers, there were demonstrations of the first broadly tunable solid state lasers, based on 3d-ion transitions. In particular, in 1963 L. F. Johnson and co-workers at Bell Labs

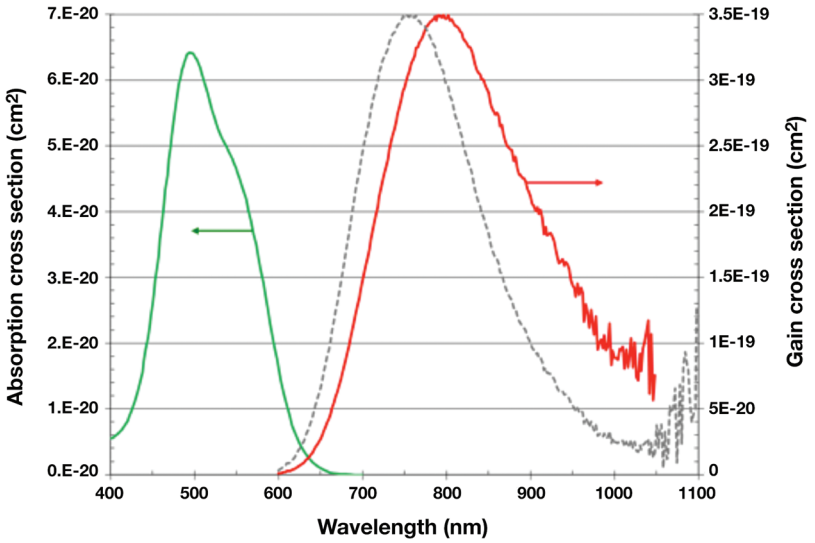


▶ **Fig. 1.** Configuration-coordinate diagrams for two cases of paramagnetic-ion transitions.

reported "optical maser oscillation from Ni^{2+} in MgF_2 involving simultaneous emission of phonons," which, translated to now-accepted terminology, would be "laser operation on vibronic transitions." Subsequent work by the same group showed operation on vibronic transitions from Co^{2+} ion in MgF_2 and ZnF_2 around 1750–2150 nm, prism-based tuning, albeit in discontinuous segments from Ni:MgF_2, and operation from V^{2+}-doped MgF_2 around 1100 nm. The major drawback to these first vibronic lasers was that, because of thermally induced non-radiative processes, relatively low-threshold operation with lamp pumping required cooling of the laser crystals to cryogenic temperatures. The author, working at MIT Lincoln Laboratory in the 1970s, became aware of the early Bell Labs work and realized that the use of lasers, rather than lamps, as pump sources could greatly reduce the engineering complexity of the systems. In particular, Nd-doped solid state lasers operating around 1300 nm proved effective in pumping both Ni^{2+} and Co:MgF_2 lasers. In the subsequent work, he had some success with the Co:MgF_2 laser, which proved capable of tuning from 1630–2080 nm at LN_2 temperatures and 1750–2500 nm at room temperature. Other 3d systems he studied showed clear evidence of a problem that has plagued many tunable solid state lasers: excited-state absorption (ESA). For most ions there are a number of 3d levels above the first excited state, i.e., the upper laser level. Depending on the positions of the levels in the configuration-coordinate diagram, it is possible that, for the desired laser wavelength, induced transitions to one or several of these levels may be possible. The net cross section that determines the laser gain is the cross section for transitions to the lower laser level minus the cross section for transitions to the higher-lying states, and this reduces laser efficiency and can even prevent laser operation.

The announcement of room-temperature, 750-nm-wavelength-region, tunable laser operation from Cr^{3+}-doped $BeAl_2O_4$ (alexandrite) in 1979 re-ignited interest in Cr^{3+}-doped lasers beyond ruby. At first, laser operation was thought to be, like ruby, on a narrow-line transition but spectroscopic investigation showed that it was in fact a vibronic. However, the gain in alexandrite lasers is relatively low, limiting applications, and today the most widespread use of alexandrite is in lamp-pumped, long-pulse lasers used for a variety of medical applications. The majority of other Cr^{3+}-doped tunable materials studied showed low conversion of pump to laser power, generally attributed to ESA. One class (colquirite structure) of materials, first developed at Lawrence Livermore National Laboratory, includes the crystals $LiCaAlF_6$ (LiCAF) and $LiSrAlF_6$ (LiSAF) and was shown to have relatively weak ESA and thus high efficiency. However, the thermo-mechanical properties of the colquirite host crystals (with thermal conductivities 10%–20% of the sapphire and alexandrite host crystals) significantly limit their ability to generate high average powers free of significant thermo-optic distortion of the output beam and, ultimately, free of fracture to the laser material.

While listening to a presentation on a particular type of color-center laser the author noted the simplicity of that system: there were no excited states above the upper laser level that could cause ESA. A subsequent review of the periodic table showed that one 3d ion, Ti^{3+}, has only a single 3d electron. The five-fold degenerate free-space state for that electron placed in a typical crystal, to first order, splits into a three-fold degenerate ground state, $^2T_2$, and a doubly degenerate upper state, ^2E. Any higher-lying states result from transitions that take the single electron out of the 3d shell and



◀ **Fig. 2.** Absorption (green) and emission (red) cross sections for Ti:sapphire and a relative plot (dashed gray curve) of the measured fluorescence spectrum. The noise in the long-wavelength region is from the detection system.

could be so high in energy as to not create ESA. There were reports on the basic spectroscopy of Ti^{3+}-doped Al_2O_3 (Ti:sapphire) with data on absorption and fluorescence. Given the superior thermo-mechanical properties of sapphire, proven with the ruby laser, it looked to be a good choice for a Ti^{3+} host.

The author obtained crystal samples from Robert Coble's group at MIT, where they had been studying the diffusion of oxygen in sapphire by using the oxidation state of Ti as a tracer. (Coble was the developer of the first transparent ceramics, paving the way for sodium arc lamps and, later, laser-quality ceramics.) The author's measurements of the absorption cross section and fluorescence spectra, shown in Fig. 2, showed much broader emission than earlier reports. When one converts the emission data to gain cross section (also plotted in Fig. 2), multiplying by the necessary $(\text{wavelength})^5$ correction, the tuning range is unusually broad. The spectral breadth of the emission results, in part, from Jahn–Teller splitting of both the ground and upper levels of the ion, leading to a more complicated configuration coordinate than shown in Fig. 1, where both the ground and excited states have multiple-energy versus displacement curves. The author also determined the fluorescence decay time and found a room-temperature value of 3.15 μs. The short lifetime seemed to indicate low quantum efficiency, but if one estimates the radiative value based on the strength of the measured absorption in the material, as well as on optical gain measurements, one finds high quantum efficiency, on the order of 80% at room temperature. The short lifetime and associated high gain cross section (in the range $3–4 \times 10^{-19}$ cm^2) result from the trigonal symmetry of the Ti^{3+} site in sapphire, which acts to strongly activate the dipole-forbidden $^2E \rightarrow {}^2T_2$ transitions.

The author first obtained laser operation from the material in May of 1982 and reported the results in June at the Twelfth International Quantum Electronics Conference in Munich. There was a delay in publication in a fully refereed journal until 1985 while the author worked, unsuccessfully, to patent the system, became engaged in other technical work, and left MIT Lincoln Laboratory to help start a company. The results published in 1985 included demonstrations of pulsed laser operation with lamp-pumped, dye-laser pumps, frequency-doubled, Q-switched, Nd:YAG laser pumps, and continuous-wave (CW) operation with argon-ion-laser pumps, with cryogenic cooling used to obtain true-CW operation. In laser operation, tuning experiments showed that the observed tuning range and that predicted by fluorescence measurements were in good agreement, confirming that ESA was not a factor in laser operation.

The first commercial Ti:sapphire laser product, an argon-ion-laser pumped CW device, was introduced by Spectra-Physics in 1988 and was followed shortly after by one from the author's company, Schwartz Electro-Optics, that included an option for a single-frequency, ring-laser configuration. Early applications of the products included use as a diode-laser substitute in the development of

other solid state lasers, notably Er-doped fiber amplifiers pumped at 980 nm for telecom applications and later Yb-doped, high-efficiency crystal lasers. With the discovery that nonlinear effects in the CW Ti:sapphire laser crystal, namely Kerr-effect lensing, could lead to generation of 60-fs-duration pulses, the utility of Ti:sapphire lasers greatly expanded. The irony of this is that the nonlinearity in the solid state laser medium might have been expected to be a limit to the mode-locking properties of the system, but it in fact provided a path to generation of femtosecond pulses. Subsequent technology improvements, including dispersion-compensating intracavity elements, broadband mirrors with appropriate optical dispersion and phase characteristics, and sophisticated pulse diagnostics, led to direct generation of 3.6-fs-duration pulses at 800 nm, slightly more than one optical cycle. These are claimed to be the shortest pulses directly generated by any laser system and close to the limit expected from the 100-THz gain bandwidth of Ti:sapphire. Commercial mode-locked Ti:sapphire lasers emerged in 1991 with picosecond-duration pulses, followed shortly by Kerr-lens-based systems providing 100-fs-duration pulses, and brought reliable ultrafast-laser technology to a broader base of users, replacing dye-laser-based sources that required long setup times with "turn-key" sources that allowed users to devote more time to science and much less to laser maintenance. The high Ti:sapphire laser gain cross section yields a pulse saturation fluence on the order of 0.8 J/cm^2, comparable with 0.7 J/cm^2 of Nd:YAG-generated high-energy pulses. If one uses a Q-switched, frequency-doubled, Nd:YAG solid state pump laser, the Ti:sapphire medium will be able to integrate and store the pump energy, which must then be extracted within a microsecond or so of the pump pulse.

The combination of femtosecond-duration pulses produced by CW Ti:sapphire lasers and high-gain, high-energy amplifiers pumped by pulsed, Q-switched lasers has led to widely used systems for high-intensity pulse generation. A key technology for this combination is the chirped-pulse amplification (CPA) technique of Strickland and Mourou, first reported in 1985 and nicely matched to the properties of Ti:sapphire. With the availability of large-aperture Ti:sapphire crystals, the ultimate limit on energy is set by the pump laser, and the limit on pulse rate is set by a combination of the pump laser and thermal effects in the Ti:sapphire material. At present, regenerative systems are widely available on a commercial basis, with pulse energies of tens of millijoules and pulsewidths <40 fs, with cryogenic cooling used for systems producing 20–30 W average power. In sum, commercial sales of Ti:sapphire lasers to date, including associated green-wavelength pump lasers, are on the order of $1 billion, not counting very-high-power systems installed or being built at major research laboratories.

At this writing, there is active development to scale up the peak power/energy of Ti:sapphire CPA systems. The highest reported power is 2×10^{15} W (2 PW), from a system in Shanghai, with a final stage pumped by a Nd:glass laser providing 140 J of 527 nm pump energy. The APPOLON Ti:sapphire laser system, under construction in France, has a goal of 10 PW in a pulse of 150 J in 15 ps. Figure 3 shows the pumped final stage of the one the Gemini amplifiers at the Central Laser Facility (CLF), Rutherford Appleton Laboratory, Oxford, UK, generating 25 J of pulse energy in a 30-fs pulse.

Key new advances in tunable solid state lasers are now almost entirely driven by their application to ultrafast pulse generation and include diode-pumped, rare-earth, Yb-doped crystals that can generate pulses on the order of 100 fs, and Cr^{2+}-doped ZnSe and similar II-VI

▲ **Fig. 3.** One of the two Gemini amplifiers at Rutherford Appleton Laboratory, showing, in the center, a green-laser-pumped Ti:sapphire crystal 90 mm in diameter and 25 mm thick. With 60 J of pump energy the system has generated 25 J of output energy in a 30 fs pulse. (STFC Gemini Laser Facility/Chris Hooker).

semiconductor hosts, providing high-gain operation similar to that of Ti:sapphire lasers but centered at 2500 nm. The longer wavelength is of great interest for attosecond pulse generation in the x-ray wavelength region through high-harmonic generation. The limited number of pages for this article requires that we leave out further discussion of these developments in tunable solid state lasers.

Other articles in this book provide details of the exciting science and Nobel-prize-winning work that has been enabled by the development of tunable solid state lasers.