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Tunable lasers

Three ways have already been found of tuning lasers to emit light of particular wavelengths chosen from a wide range. This ability is rapidly extending the uses to which lasers can be put

THEODORE H. MAIMAN, of Hughes Aircraft Company in California, announced to the press on July 7, 1960, that he had successfully operated the first laser. Thus ended a race between a number of laboratories which had started in 1958 following two independent publications of the theoretical idea of a laser. These proposals were made by the Americans A. L. Schawlow and C. H. Townes and the Russians N. G. Basov and A. M. Prokhorov. Before the end of 1960 Maiman's laser, and those built by the close runners-up in this race, had caught the imagination of hundreds of scientists and engineers and research in this field began to expand very rapidly.

The results of the past eight years of intensive research have been quite dramatic. The different materials in which laser operation has been obtained now number in the hundreds. Lasers have been operated which produce visible radiation, ultraviolet, infrared and even submillimetre radiation. Laser powers have been increased by many orders of magnitude and it is now possible to obtain continuous powers of more than ten kilowatts or powers of tens of thousands of megawatts in pulses as short as one millionth of a microsecond.

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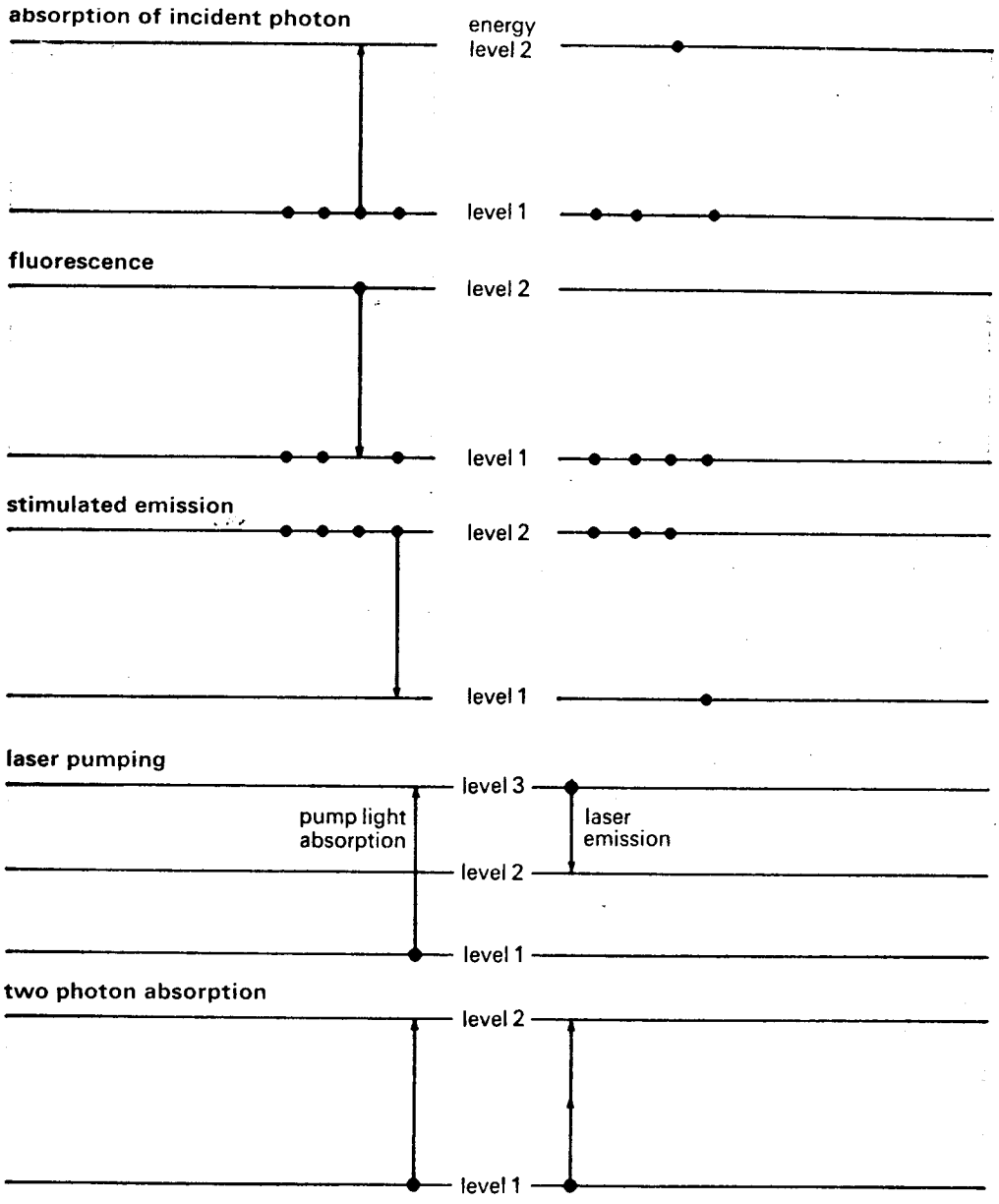
However, despite all these dramatic improvements, there has been a long felt need amongst laser scientists and potential laser users for a laser whose wavelength could be controllably varied. Many early lasers did in fact have small tuning ranges, but what was lacking were lasers with really wide tuning ranges. Such lasers have recently become a reality.

TUNABLE LASERS have been made in three quite different ways. The impetus for the two most important methods came from the use of the extremely high power lasers, of fixed wavelength, which were already in existence.

In 1966 Peter Sorokin, working at the IBM Laboratories at Yorktown Heights near New York, obtained laser action from a solution of an organic dye This new laser showed a wide range of tuning. The secret to obtaining laser action from such materials is that they must be excited by an extremely short and powerful burst of light and this was available only from another laser. By using different dyes and different lasers to excite them it has been possible to produce lasers which work over a range which spreads without break from the ultraviolet to the near infrared. The idea of using a laser to drive another laser may sound extravagant but it is becoming increasingly used and can be a very efficient process. As much as 25 per cent of the radiation from a ruby laser has been converted into the tunable radiation of a dye laser. A systematic search amongst strongly fluorescent organic materials has now revealed that some materials, including the well known laboratory indicator fluorescein, can be excited by a fairly conventional flash lamp. This means that lasers capable of producing megawatts of power and tunable right through the visible spectrum have been brought well within the financial and technical resources of even a small laboratory.

The second technique for producing tunable coherent radiation to some extent complements and to some extent rivals these organic lasers. This technique uses a device known as a parametric oscillator which, although not properly described as a laser, is capable when excited by a laser of producing a beam of light with all the properties of coherence and directionality which are the hallmarks of laser radiation. Parametric oscillators have produced powerful coherent radiation in the visible and near infrared parts of the spectrum, and will be capable of producing radiation into the far infrared region. Perhaps the most important aspect of recent parametric oscillators is that they have been operated continuously as well as pulsed. The dye lasers are of course limited to pulsed operation and then only in the visible and near-visible regions of the spectrum.

The idea of the parametric oscillator arose through the study of the entirely new field of physics known as non-linear optics. This is



ENERGY LEVEL CHANGES are the mechanism by which atoms or molecules absorb and emit light. If a photon of frequency ν strikes and is absorbed by an atom or molecule (one of the black dots) that atom is immediately raised into an upper energy level by the addition of energy equal to a constant (Planck's constant, h) multiplied by the photon's frequency. This packet of energy is lost when the atom later spontaneously emits a similar photon by fluorescing. Laser action is based on the fact that when photons encounter a population of atoms already in the excited upper energy level they cause stimulated emission of further photons exactly in step with those of the incident light. Laser pumping is shown with three energy levels arranged to allow 'population inversion', prerequisite for laser action. The incident photons from the flash lamp used for pumping the laser raise the atoms from the ground level 1 to excited energy level 3. Intermediate energy level 2 is practically empty, so population inversion is produced between levels 3 and 2. Atoms return from level 3 to level 2, emitting laser radiation at a wavelength longer than that of the pumping radiation. Two photon absorption takes place when quantum theory forbids the raising of an atom's energy by absorption of a single photon of energy $h\nu$. An intense source of light provides so many incident photons that the atom can instead absorb two simultaneously, each having half the frequency and thus energy of $\frac{1}{2}h\nu$

the study of the behaviour of materials subjected to light of such intensity as to change some of the parameters of the material. These extremely high optical fields can be obtained only from lasers and therefore research into lasers and non-linear optics go hand in hand. The study of non-linear optics started in 1961 when Peter Franken and co-workers at the University of Michigan demonstrated a phenomenon known as 'second harmonic generation'. They showed that, when a powerful beam of red light from a ruby laser was passed through certain crystals, some of the red light was converted into a beam of ultraviolet light with a wavelength exactly half that of the red light. This is the optical analogue of harmonic distortion which has plagued many a hi-fi enthusiast.

With more powerful lasers and better crystals this conversion process becomes very efficient and is now commonly used as a way of generating powerful short wavelength radiation from existing lasers. Since Franken's discovery the study of non-linear optics has expanded rapidly. Many interesting phenomena have been discovered, of which one is parametric oscillation. A parametric oscillator consists of a crystal which converts a powerful laser beam at one wavelength into another coherent beam of light having different wavelengths. Unlike second harmonic generation these new wavelengths are longer than the laser wavelength; but most important is the fact that they can be varied even though the laser wavelength is fixed. This variation or tuning is achieved either by changing the crystal temperature or by altering its orientation.

Although the first parametric oscillators required very powerful lasers to pump them, new and exotic crystals have been made to oscillate by a laser having rather less than one watt of power. Conventional light sources still cannot pump parametric oscillators, since the essence of the pump light source is its extreme spectral purity and beam parallelism. In these terms even a laser with a power of only one milliwatt far exceeds the capabilities of any conventional light source.

The third approach to tunability is to use a special kind of semiconductor laser. The narrower the band gap of a semiconductor—the difference in energy between its valence and conduction bands—the longer the laser wavelength. By applying mechanical pressure to the material the band gap can be changed and thus also the laser wavelength. The tunable wavelengths are in the infrared and the powers obtainable are low compared to dye lasers and parametric oscillators.

DYE LASERS, the first of the three tunable types described, are like any other laser in that their essential component is a substance which can amplify a light wave which passes through it. This amplifying medium is

placed between two parallel mirrors so that light travelling perpendicular to the mirrors is reflected back and forth and so made to pass through the medium repeatedly. The light wave thus grows to a high intensity and a small transmission through one of the mirrors allows the light to escape in the form of a powerful, collimated beam.

The process of amplification in a laser is the exact opposite of the process by which light is absorbed. Under normal conditions a collection of atoms absorbs rather than amplifies light because the higher the atomic energy the smaller the number of atoms possessing this energy. As each atom absorbs light it is raised from a low energy level to a level whose energy is higher by an amount equal to the energy of one photon—a quantum of this light. This energy is equal to Planck's constant, h, multiplied by the frequency, ν, of the light. These atoms can return spontaneously to the low energy level by remission (fluorescence) of photons of the same frequency. In a laser the atoms are so arranged that the atoms having the higher energy level can outnumber the rest, a condition known as population inversion. Under this condition a photon of energy $h\nu$, instead of being absorbed, will stimulate an atom in the high energy level to drop to the lower level. As it does so the atom emits a fresh photon of energy $h\nu$ which adds itself coherently to the beam of photons (light) passing through the medium.

Absorption does not take place at one precise frequency and there is always some narrow band of frequencies over which absorption can take place. Since laser amplification is the opposite of absorption there is a corresponding spread of frequency over which amplification can take place. This opens the way to a tunable laser, since a filter can be placed between the laser mirrors which stops all the light except for a very narrow frequency range, much narrower than the range for amplification. The laser can then operate only at the frequency passed by the filter; and, by tuning the filter, the laser frequency can be tuned.

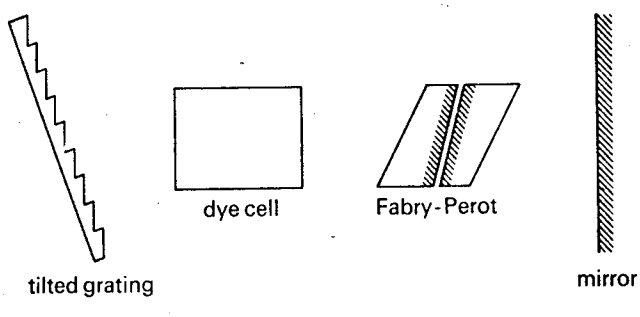
For a wide tuning range clearly a wide amplification range is needed. A considerable number of organic molecules have broad and intense fluorescence bands which suggest that they might be used to produce tunable laser oscillation. An obstacle to their use was the fact that the fluorescence is extremely short lived, lasting only for a few thousandths of a microsecond. Sufficient population inversion can be obtained only with a correspondingly short and intense source of light to pump the molecules into higher energy levels. The beam from a ruby laser can provide just such a pump source and in this way Sorokin obtained laser oscillation from a solution of the blue dye chloroaluminium phthalocyanine. The blue colour of this dye is due to its strong absorption of red light and it is therefore well suited to being pumped by the red light at 694·3 nm (1 nanometre is 10^{-9} metre) from a ruby laser. This absorption corresponds to molecules being raised from the base level 1 to excited level 3. Laser oscillation from the dye, level 3 to level 2, has a wavelength of 750 nm which looks a very deep red colour since it is on the fringe of the infrared.

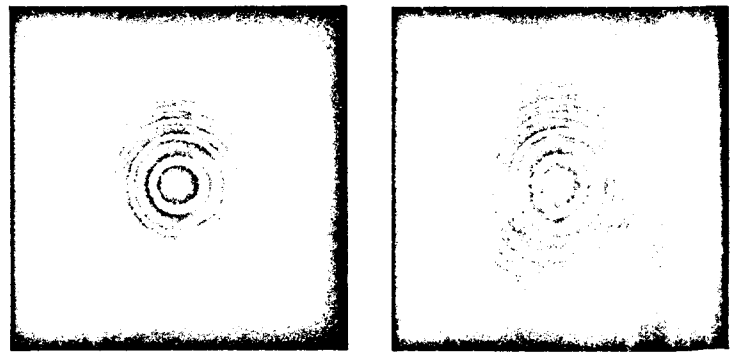
Since Sorokin's discovery many more organic molecules have shown laser action pumped in the same way by a ruby laser. Not all of these molecules can be correctly described as dyes but the term 'dye laser' is generally used. Different dyes lase at different wavelengths and, since typically each dye has a tuning range of a few tens of nanometres, it has been possible to span without a break the range 700-1000 nm by using some 20 different dyes.

One method of tuning involves replacing a mirror of the dye laser by a diffraction grating. When used as a reflector a grating has the interesting property that it can reflect light back along the same path even though the light wave is not travelling perpendicular to the grating. In this situation only a narrow wavelength range is reflected and the particular wavelength that is reflected depends on the angle of incidence of the light on the grating. Thus, by tilting the grating, the reflected wavelength can be changed. An even more precise tuning effect can be achieved with a Fabry-Perot interference filter. Dan Bradley's group at Queen's University, Belfast, have used both types of filter together, the grating for coarse tuning and the Fabry-Perot for fine tuning. Their tuning range for one dye was more than 10 nm and when tuned to any particular wavelength within this range the wavelength spread of the laser was less than 10^{-3} nm. Despite the tremendous spectral narrowing, a factor of 10,000, the laser power was essentially unchanged.

The wavelength given by a dye when pumped by a ruby laser is necessarily longer than the ruby laser wavelength, since fluorescence is always at a longer wavelength than the absorbed pump light. However the technique of second harmonic generation allows the ruby laser beam to be converted to ultraviolet light, and the powerful infrared beam from a neodymium laser to green light. These second harmonic light sources have been used to pump yet more dyes which absorb in the ultraviolet or green and these lasers have extended the tuning range to cover 400-1000 nm.

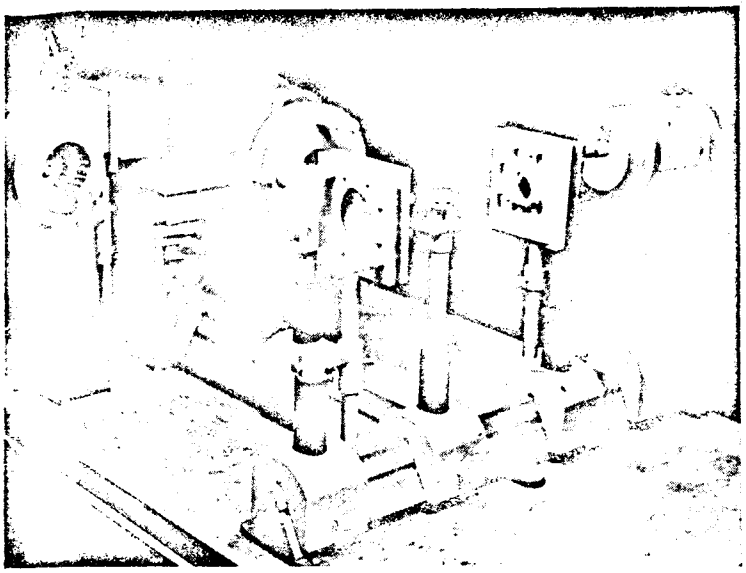
AN IMPORTANT ADVANCE was then made by Sorokin when he showed that many of these dyes could be pumped directly by a flash lamp, provided the flash was of sufficiently short duration. Flash lamps produce copious amounts of green, blue and ultraviolet light. They are used to pump both ruby and neodymium lasers, with a flash duration of about one thousandth of a second, but for dye lasers the flash must be shorter than one microsecond. Longer times than this result in the excited molecules finding their way

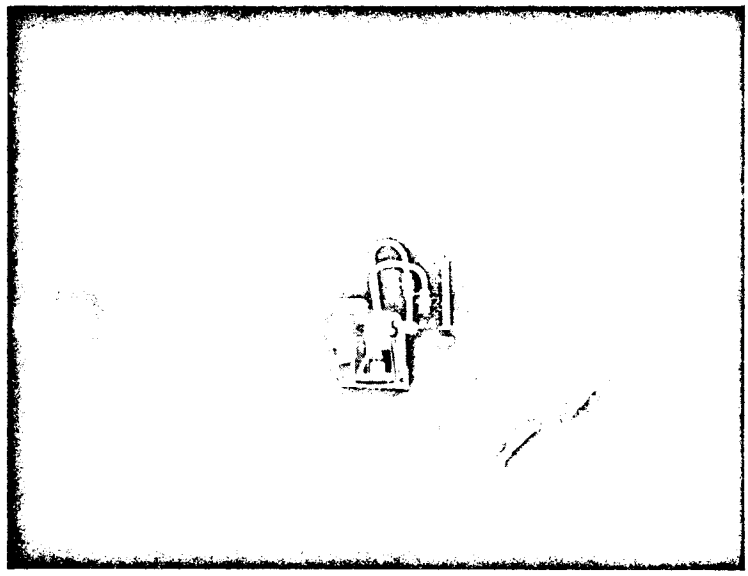




DYE LASER, by a group at Queens University, Belfast, having extremely narrow spectral emission. The filtering action, which eliminates almost all wavelengths, is obtained by the combined effects of the diffraction grating, set at the desired angle, and the Fabry-Perot interferometer with two parallel reflecting faces

INTERFEROGRAMS reveal the extraordinary spectral purity of light from the Belfast dye laser (right). The photograph compares the dye emission with the light from a helium-neon laser (left). The extreme sharpness of the rings from the dye laser indicates a spectral width of less than 0·001 nm—one millionth of a micron





TUNABLE DYE LASER in the illustration is the small rectangular glass cell in the centre foreground placed between two exactly parallel mirrors. The cell contains the dye which is pumped to the necessary excited condition by the air cooled ruby laser visible in the background. In the right hand picture Dr Peter Sorokin is seen using his dye laser pumped by a flash lamp. The intense beam produces the bright spot on the screen. This laser can be tuned to emit light having a colour ranging from red through yellow to green

into other energy levels, notably the so-called triplet level, in which molecules give rise to absorption instead of amplification.

A short flash can easily be obtained by eliminating electrical inductance from the flash lamp circuit. Unfortunately the glass envelope of a conventional lamp is then rapidly destroyed; in fact, it is usual to add inductance deliberately to avoid this. The solution is to use a robust metal envelope and, as this is optically opaque, the dye solution must also be contained within the flash lamp glass, inside a tube which is not destroyed since it can easily withstand compressive forces. The use of a flash lamp to pump the dye results in much greater heating of the dye than is the case when using a laser or second harmonic beam. The dye solution must be cooled by pumping through a heat exchanger if it is to be used in a system with the high flash rate that is desirable for many applications.

The power from a flash lamp pumped dye laser can be of the order of a megawatt. This means that efficient second harmonic operation is possible giving useful powers which can be tuned down to 200 nm, well into the ultraviolet. This should be of particular interest to chemists. We should emphasize that this kind of dye laser is very cheap and simple to build. In fact J. R. Lankard, a colleague of Sorokin, is designing a dye laser for the 'home hobbyist' costing only $50.

PARAMETRIC OSCILLATORS follow a completely different approach to amplification which involves changing periodically some parameter affecting the motion of a wave. Sound waves, radio waves and any other kind of waves can, in principle, be amplified by a 'parametric process'.

An everyday example of this parametric amplification is a boy working a swing by standing upon the seat. He lowers his body on the down-swing and raises it on the up-swing, driving the swing higher and higher. What he is doing, of course, is producing greater change of his potential energy with displacement than when swinging freely, which results in an extra force on the swing. An alternative view is that a parameter is being modulated which governs the motion of the swing. This parameter is the distance between the swing hinge and the boy's centre of gravity, which determines the natural frequency of oscillation. To increase the amplitude of the swing the modulation must be at twice the natural frequency and with the correct phase to keep in step with the swing. In the language of parametric amplifiers, energy is then transferred from the pump frequency of the boy to the signal frequency (the motion of the swing).

Non-linear optics, which gives rise to the phenomenon of harmonic generation, also provides the time varying parameter required to amplify optical waves. With suitable crystals a large optical field will modulate the refractive index at the optical frequency, thus providing the desired time varying parameter. Another wave at exactly half this pump frequency will then be amplified, just as in the example of the swing, since the optical energy in the crystal increases with refractive index. Again the phase of this signal wave has to be correct, and the two waves must travel through the crystal with the same phase velocity. This condition poses a problem since the velocity of light waves in crystals varies with frequency. The solution is to use an anisotropic crystal in which the velocity depends upon the direction of the light beam and how the light is polarized. By using different polarizations for pump and signal, and the correct beam direction, it is possible to match the velocities. As crystal refractive indices depend on temperature, this phase match can be finely tuned by altering the crystal temperature.

The parametric amplifiers discussed so far have been rather special in that the signal frequency is exactly half the pump frequency. In fact a pair of waves can be amplified. The first of these is the signal wave and the second an 'idler wave' whose frequency is the difference between the pump frequency and the signal frequency. For a given set of frequencies there will be a particular crystal orientation and temperature for which the waves are phase-matched and therefore amplified. By changing the angle or temperature, for a fixed pump frequency, the signal and idler frequencies can be tuned.

A parametric amplifier can be turned into a tunable oscillator by the addition of two parallel mirrors. These mirrors are normally chosen to reflect both signal and idler although, provided the pump is sufficiently strong, it is possible to operate without the idler being reflected, and this offers practical advantages. The oscillator is tuned by varying the angle or temperature of the crystal. Tuning is continuous, with the shortest wavelength approaching the pump wavelength and the longest being limited by the onset of infrared absorption in the crystal. More than half the pump power can be converted into the two tunable wavelengths.

THE NON-LINEAR CRYSTAL should by now have emerged as the most important part of a parametric oscillator. For a crystal to be suitable it must be transparent for all the wavelengths of interest, it must be highly non-linear—giving a large change in refractive index for a reasonable strength of the optical field—and it must have the right properties to permit phase matching. The whole progress of parametric oscillators has therefore been determined by crystal development. To date four crystals have been used successfully and two more look very promising. Considerable effort is being expended in the search for new crystals.

Groups at the Bell Telephone Laboratories in New Jersey and at Moscow University were the first to demonstrate parametric oscillation at optical frequencies. The Bell Labs scientists used a crystal of lithium niobate for their experiments and the Russians used KDP (potassium dihydrogen phosphate) and ADP (ammonium dihydrogen phosphate). Both groups pumped their oscillators with megawatt powers of green light obtained by second harmonic generation of the light from neodymium lasers. Much progress has been made since these early experiments and, with more refined techniques, lithium niobate oscillators have been tuned over two octaves of frequency, have given 40 per cent conversion from pump to signal and idler, and have been operated without the idler reflected: KDP has given tunable wavelengths right down to the green.

The oscillators described above operate in short pulses. A breakthrough was made last year when two laboratories ran oscillators continuously for the first time. One of these was at the Bell Labs and used a new crystal developed there: barium sodium niobate ($Ba_2NaNb_5O_{15}$, hence its nickname of bananas). This crystal is an improvement over lithium niobate which is damaged by green laser radiation. The second continuous oscillator was built by Stephen Harris and his students at Stanford University, California. In this case lithium niobate was used but, by keeping the crystal at 300°C, the damage was annealed out as fast as it was produced. Both oscillators gave several milliwatts of tunable output. This is the order of power available from many commercial gas lasers at a fixed wavelength.

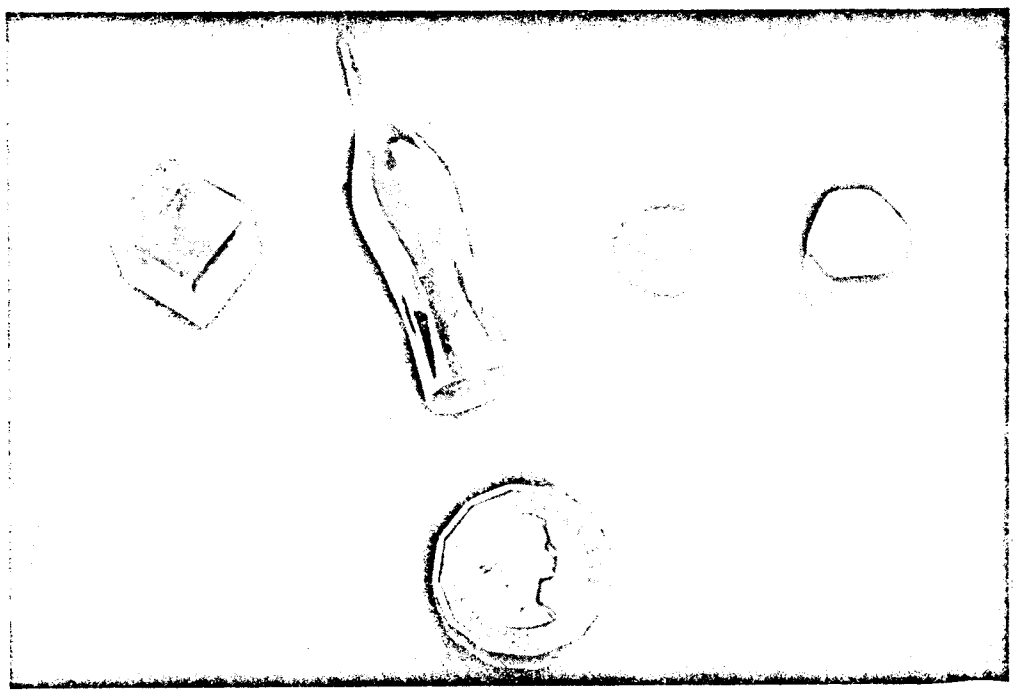
The shortest wavelength produced so far by a parametric oscillator is 530 nm and the longest is 2·5 microns. Looking to the future, a continuous oscillator using ADP and going down to 300 nm seems feasible, since high power ultraviolet lasers are becoming available for use as pumps. There are also crystals which should work far into the infrared. Proustite, an exciting new crystal developed at the Royal Radar Establishment, Malvern, opens the prospect of oscillators operating over the range 2-12 microns. Tellurium, an elemental crystal, has already been shown to amplify a far infrared signal and should oscillate at 10-25 microns. In the near future we expect to see parametric oscillators covering the whole spectrum between the ultraviolet and far infrared. Research is urgently needed into techniques for obtaining a narrower spectral output and a more reproducible wavelength for these oscillators.

TUNABLE SEMICONDUCTOR LASERS are based on the fact that the energy of each photon from a laser equals the difference in energy between two atomic levels in the lasing material. Thus the frequency is also tied to the energy difference: if this can be altered then the laser can be tuned. The small tuning range of the ruby laser is obtained in this way; its energy difference decreases with rising temperature, so changing the temperature of the ruby tunes the laser frequency. Lasers using gallium arsenide semiconductor diodes can also be temperature tuned over a small range.

Normally the change in energy difference is small and the tuning range limited. But J. M. Besson and W. Paul at Harvard University with A. R. Calawa at MIT have recently obtained very considerable tuning of lead selenide diode lasers in the far infrared. Hydrostatic pressure, rather than temperature, was used to alter the band gap. By increasing the pressure up to 14,000 atmospheres the laser was tuned from 7·5 to 22 microns. Despite the difficulty of these first experiments—for instance the diode was held in a bath of liquid helium—it can be expected that practical tunable lasers will eventually be the result.

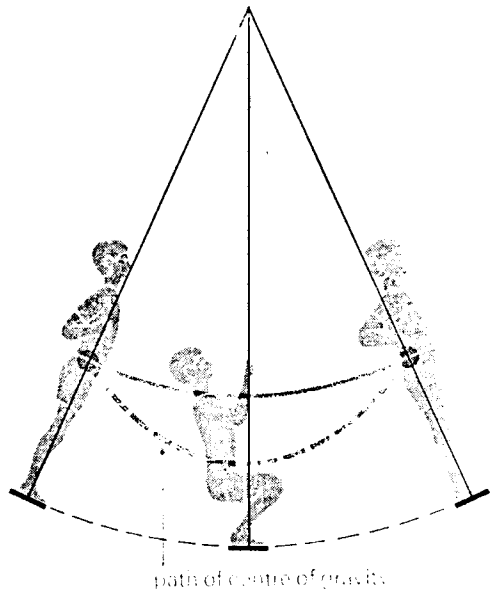
APPLICATIONS of lasers have since 1960 been the subject of ceaseless speculation. Early lasers suffered from many faults which limited their usefulness and as a result much of the laser research effort was concentrated on understanding and improving the laser rather than upon its uses. Applications have therefore been fairly slow in coming although now an impressive list of successes can be made (see "Practical uses of lasers", SCIENCE JOURNAL, June 1966). Many of these applications show the laser in what may be one of its most important roles for some time to come, as an extremely versatile research tool not only in physics and engineering but also in chemistry, medicine and the biological sciences. The tunability of the laser now greatly extends this versatility and brings many proposed applications closer to realization.

Spectroscopy is one of the most obvious fields for exploitation of laser tunability. Conventional absorption spectroscopy needs a light source of very narrow spectral width which can be tuned over a wide range of wavelengths. This is achieved by using a light source with a broad spectral spread, such as a tungsten filament lamp, and following it with a prism or grating monochro-

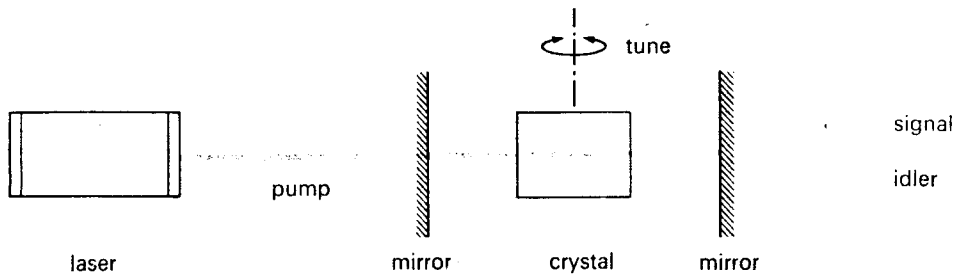


CRYSTALS suitable for use in parametric oscillators must be non-linear, transmit over a useful range of wavelengths and be phase matchable. From the left, these are ADP for visible oscillation, lithium niobate for the near infrared, proustite for infrared and tellurium for emission in the far infrared. The threepenny piece (2 cm) gives an index of size

SWINGING CHILD illustrates the concept of a parametric amplifier. The parameter that the child varies is the radius of swing of his centre of gravity; by raising and lowering his body at twice the swing's natural frequency he increases the amplitude of the swing. An optical parametric amplifier is pictured in the diagram below



PARAMETRIC OSCILLATOR (below) uses a single frequency laser to pump a suitable crystal which effectively becomes a tunable laser. Parametric oscillation normally calls for the crystal to be placed between mirrors which reflect at two frequencies known as the signal and the idler. The device can be tuned by altering the angle at which the pump beam strikes the crystal (as indicated) or, alternatively, by changing the crystal's temperature



mator to act as a filter which can be tuned to pass only the required portion of the spectrum. A tunable laser obviates the need for a monochromator and in addition allows much greater resolution. Spectroscopic applications of this sort were demonstrated with some of the very first ruby lasers. Since it has only a small degree of tunability, the ruby laser is restricted to the study of atomic or molecular systems which have absorption lines that happen to fall within this small tuning range. Despite this restriction, not only were the absorption spectra of certain lines in bromine vapour measured using a tuned ruby laser, but certain chemical reactions involving bromine vapour were selectively photocatalysed by tuning the laser to be absorbed by particular bromine lines. With the more recent tunable lasers effects such as this can now be studied throughout the visible and near visible spectrum instead of only on fortuitously placed absorption lines.

By using a pulsed laser, with pulses much shorter than one microsecond, it becomes possible to examine spectra of very short lived species such as excited molecules and the intermediate products in fast chemical reactions. For example the flash photolysis measurements pioneered by British Nobel prizewinners R. G. W. Norrish and G. Porter can now be performed using dye lasers instead of flash tubes, with great improvements in both time and spectral resolution.

High power tunable lasers now permit a new type of absorption spectroscopy. This involves a phenomenon coming from the area of non-linear optics known as two photon absorption. With the low light intensities used in conventional spectroscopy the two photon absorption is very feeble and cannot readily be observed. The likelihood of two photon absorption taking place increases very greatly if the light intensity is increased, and with available laser powers the effect becomes quite pronounced. The data obtained from the two photon absorption spectrum complement those obtained from the conventional absorption spectrum.

One of the most firmly established applications for powerful laser pulses of short duration is in optical radar (see "Lidar", SCIENCE JOURNAL, February 1968). Optical radar measurements have been widely used to investigate the atmosphere where the targets may be clouds, dust layers or even the atomic and molecular constituents of the atmosphere. For many of these measurements the wavelength used is not important, but there are resonant scattering processes which are strongest at a particular wavelength and a tunable laser is required to match this wavelength. At the Radio and Space Research Station, Slough, light scattered from sodium atoms in the atmosphere at a height of about 90 kilometres was recently observed by tuning the wavelength of a rhodamine-6G dye laser to coincide with the well known yellow D lines of sodium. When this was done the return signal was greatly enhanced by resonant absorption and re-emission of the laser radiation. In this way the abundance of sodium atoms was measured.

Other potential applications for tunable lasers are in the fields of medicine and communications. Surgery has already been performed with lasers but it may be an advantage to use a particular wavelength in order to destroy a piece of tissue selectively. In communications the generation of a number of frequencies permits multi-channel operation. For high security communications a variable frequency should defeat the efforts of any eavesdropper.

Summarizing, then, dye lasers provide high power pulses with wavelengths tunable across the visible and near visible. As a result of their tunability and high power, dye lasers will certainly be used in specialized spectroscopy. Since they are so cheap and easy to construct it is likely that they will replace ruby and neodymium lasers in many applications. Parametric oscillators are more complex but can be tuned over a much wider range. They have been operated both pulsed and continuously. Probable applications are in optical communications and as an alternative to conventional spectrometers. Tunable semiconductor lasers may also find spectroscopic application. Since tunable lasers are now in an advanced state of development the time has surely come for scientists and engineers to consider seriously the potentialities of such lasers in their own fields of activity.

FURTHER READING

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