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# Mirrorless lasers

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**Abstract.** Experimental realization of mirrorless lasers in the last decade have resulted in hectic activity in this field, due to their novelty, simplicity and ruggedness and their great potential for application. In this article, I will review the various developments in this field in roughly chronological order, and discuss some possible applications of this exciting phenomenon, also termed as 'random lasing'.

**Keywords.** Random amplifying media; multiple scattering; random lasers.

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A typical laser consists of an amplifying medium, enclosed within a resonant cavity formed by two mirrors (figure 1). The amplifying medium is pumped, and a spontaneously emitted photon is amplified as it travels through the gain medium. The cavity mirrors cause the light to traverse back and forth repeatedly. One of the mirrors is made partially transmitting, to provide an output port for the laser. The build-up of intensity within the amplifying medium is due to stimulated emission – the emitted photon being in phase with, and travelling in the same direction as the stimulating photon. Thus the laser light is well-collimated, and is highly coherent – and these two features have become the hallmark of lasers, distinguishing them from ordinary light sources. It is also obvious that the mirrors and the amplifying medium must be free from defects – scattering is detrimental to laser action, as it increases losses. The net amplification is reduced as the photon may get scattered out of the gain medium, and the coherence is also lost due to random multiple scattering off the defects.

While the active medium may have a broad range of emission, as in the case of a dye, the conventional laser can support only select wavelengths – those that form stable modes inside the cavity formed by the two mirrors. Only those wavelengths $\lambda$, that can have an integral number of half wavelengths in the distance $L$ between the two mirrors, i.e., $\lambda = 2L/n$, where $n$ is a whole number, can be supported, as only such waves can have nodes at the reflectors. Thus, by altering the resonator configuration, one can change the wavelength of operation thereby tuning the laser. Another way of expressing this is by saying that only those modes are possible for which the mirrors provide a coherent and resonant feedback, i.e., it satisfies two conditions – it returns some electromagnetic energy back into the active medium, and forms a stable electromagnetic configuration in the cavity because of constructive interference between the incident and reflected waves.


**Figure 1.** Schematic diagram of a conventional laser. An active medium is enclosed in a resonant cavity formed by two mirrors, one of which is partially transmitted to allow light to exit the system.

Thus, in a conventional laser, the cavity mirrors perform two tasks – reflecting the light back and forth into the gain medium so as to build up intensity and selecting the wavelength of operation. Considering the crucial role of mirrors in a laser, the phrase ‘mirrorless lasers’ seems to be a paradoxical one. However, in what follows, we will see how one can indeed have mirrorless lasers, and how these differ from conventional lasers.

To begin with, the first lasers [1] were observed nearly a century before the first optical laser was made on earth [2]. These were the stellar lasers discovered by Wolf and Rayet in 1867. Over the years, astronomers have found some extremely intense astronomical sources of light and radio waves. For example, in 1965, astronomers reported [3] radio frequency emission in the direction of certain regions of interstellar ionized hydrogen (HII regions). Subsequently, a number of HII regions with high intensity emissions at 1.35 cm wavelength were discovered [4]. These sources exhibit some peculiar features like anomalous line intensity ratio, i.e., the ratio of intensities of different spectral lines are different from those observed in laboratory sources. The lines are strongly polarized, and have a very small spectral width. Based on the emission profile and brightness, one can estimate the temperature of an object. The brightness temperature of some of the spectral lines of these sources is as high as $10^{13}$–$10^{17}$ K, whereas the Doppler width of these lines corresponds to kinetic temperatures of 10–100 K that are usual for neutral clouds of interstellar gas. It was then proposed that these anomalies were the result of coherent amplification of the radio emission of the star by the surrounding stellar medium, that was in a population inverted state [5–7]. This mechanism could yield amplifications as large as $10^7$. This is the ‘cosmic laser amplifier’, where light from the star is amplified in a single pass, by the surrounding medium. This mechanism, which does not require any feedback mechanism, is also known as amplified spontaneous emission (ASE).

While the cosmic laser amplifier seemed to account for the anomalies in these sources, soon, other more powerful laser outbursts were observed, the characteristics of which could not be explained within the framework of travelling wave amplifier as this cannot provide the requisite gain. A generator mechanism with non-resonant feedback was proposed by Weaver *et al* [8] and Letokhov [9]. It was suggested that the emission originates in the amplifying stellar medium, that is made of electron gas or microscopic dust, and is multiple scattered by these particles. The photons bounce back and forth due to random multiple scattering. This increases the path length in the gain medium, which in turn increases amplification. As the random multiple scattering causes the light to travel in much larger paths in the amplifying medium than it would have done otherwise, extremely high intensities can result. This is the ‘cosmic laser’, in which the scattering by the free electrons or by interstellar dust provides feedback. Unlike conventional lasers, where the feedback is resonant and coherent. It is, in general, not so in the cosmic laser.

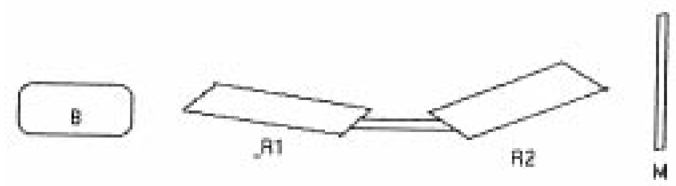


**Figure 2.** Layout of the first non-resonant feedback laser. Two ruby rods R1 and R2 provide amplification. The mirror, M, on the right is partially transmitting. The block, B, is either a volume or surface scatterer.

Such amplifier systems are now referred to as 'random laser' or 'mirrorless lasers'. Though initially hypothesised to explain superluminous stellar sources, lasers using non-resonant feedback were first demonstrated in the laboratory by Letokhov and coworkers [9,10]. In a conventional laser, if one of the cavity mirrors is made rough, the reflection will not be specular, but diffuse, and this was used in the first implementation of a random laser. The feedback into the gain medium is non-resonant and incoherent. In the absence of a well-defined cavity there is no resonant selection of wavelength. Figure 2 shows the schematic of such a laser. The active medium consists of two ruby crystals whose ends were cut at Brewster angle to prevent self-excitation. Instead of the two end mirrors of a conventional laser, the random laser had one partially reflecting mirror for providing an output port, and a surface or volume scatterer at the other end to provide non-resonant feedback. The volume scatterer was a suspension of chalk particles (about 20 microns diameter), in water and surface scatterer was a plate with a layer of spluttered MgO. In some cases particles of sulfur were suspended in water. This system was found to lase beyond a threshold.

The emission characteristics of these random lasers is quite different from those of conventional lasers. Due to the absence of a definite reflecting boundary, there are no discrete equi-spaced resonant modes. The interaction of various modes, and the large losses suffered by each mode resulted in a continuous emission spectrum for these devices. Consequently, the output frequency of the laser with scattering feedback is determined by the resonance frequency of the active medium rather than by the resonator as in the case of conventional lasers. Line narrowing is determined by the resonant properties of the active substance due to the fact that photons at the frequency that has maximum gain are amplified more than photons of other frequencies. It was found that the process of spectral narrowing was much slower in random lasers than in an ordinary laser with resonant feedback, and this limits the width of the spectrum in the pulsed mode operation. In cw operation, the width is limited by fluctuations the main sources of which are spontaneous emission and Brownian motion of the scatterers. The statistical properties of the laser with non-resonant feedback have been shown to be that of an extremely bright black body in a narrow range of spectrum [11]. The emission of such a laser has no spatial coherence, and no stable phase.

So far, mirrorless lasers in the laboratory had an amplifying medium, and external scatterers in the form of rough mirrors or colloidal suspensions. However, as in the stellar lasers, the scatterers can be in the amplifying medium itself. These are the 'powder lasers' [12–16], where amplifying microcrystals, often laser crystals that have been crushed and ground, constituted the random laser. These microcrystals, typically those with rare-earth inclusions, provided amplification, and also non-resonant feedback due to the random scatterings off the microcrystallite faces. This form of random lasers too were found to give rise to stimulated emission beyond a clear threshold. They too display the effects of strong narrowing of the emission spectra, increase in luminiscence intensity, showing that it is possible to construct quasimonochromatic bright sources of radiation using fine grained powder.

There has, however, been a spurt of activity in this field over the last few years, after Lawandy and coworkers found mirrorless lasing in colloidal suspensions in liquid dye [17]. The system, which consists of $TiO_2$ microspheres suspended in a solution of rhodamine 640 perchlorate, is extremely easy to prepare. The dye has a very broad emission spectrum (about 40 nm wide) in the orange-red and gives rise to ASE at high pump powers. On addition of scatterers to the dye, the emission is found to have spectral and temporal characteristics of a multimode laser oscillator. There appears a significant increase in the emission intensity, and emission intensity vs pump power exhibits an S-shape, typical of lasers. A sudden line-width collapse occurs at the threshold, and the emission narrows down to a few nanometers width, as is shown in figure 3. The threshold excitation for laser action is found to be surprisingly low. The pulsed emission have a duration of 4 ns below threshold, and it reduces to about 400 ps beyond threshold.

During the last decade, a large number of studies have been conducted on various dye–scatterer systems. It was found that the threshold for stimulated emission is dependent on concentration of both laser dye and scattering particles [18]. The lasing threshold could be reduced by more than two orders of magnitude as compared to pure dye, when the density of scatterers ranged from $5 \times 10^9$/cc to $2.5 \times 10^{12}$/cc. In one particular study, a thousand-fold increase in the peak emission intensity was observed upon addition of scatterers [19]. For a range of dye and scatterer concentrations, bi-modal emission, with competition between the modes was seen.

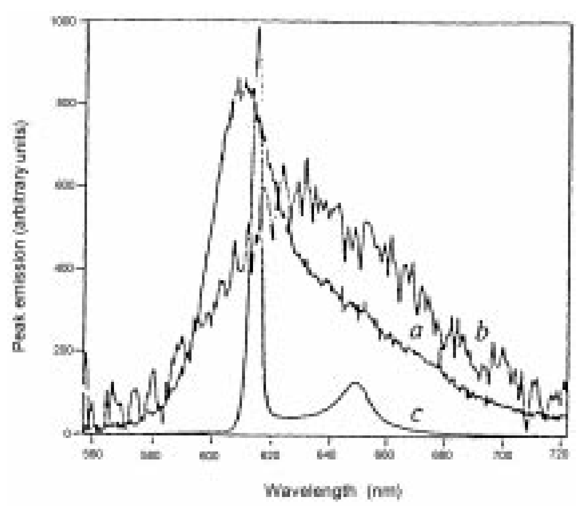


**Figure 3.** The emission spectrum of a $TiO_2$–rhodamine system at three different pump powers. Curve *b* has been scaled up by a factor 10, while curve *c* has been scaled down by a factor of 20. Pump power for b < pump power for a < pump power for c (figure from ref. [17]).

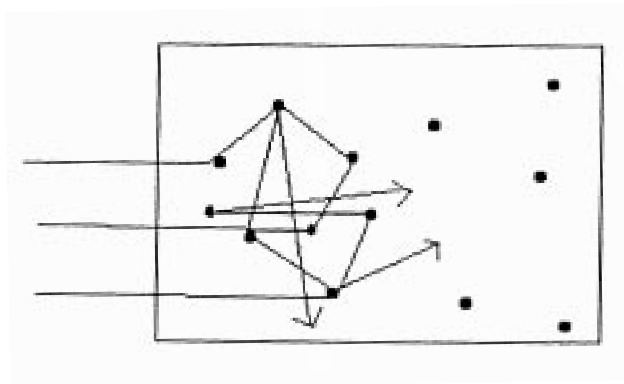

**Figure 4.** Diffusion propagation of light in a medium with random scatterers.

How does the lasing action occur? It is obvious that multiple scattering enhances the path length of a photon within the amplifying medium, which results in increased gain. The scattering of light by a particle is described by the Mie theory, where three regimes are considered – the weak scattering regime, the diffusion regime, and the localization regime. The three regimes may be quantified in terms of the transport mean-free path, $l^*$, and the system size, $L$. The transport mean-free path, $l^*$, defined as the average distance the photons travel before their direction of propagation is randomized, depends on the size and the number density of scatterers. As the name suggests, the weak scatterer regime occurs when there are very few scatterers and so $l^* \gg L$. The diffusion regime corresponds to the intermediate case where $l^* \sim L$, and the strong scattering regime, $l^* \ll L$. The lasing picture above represents the diffusion regime, where a photon now appears to diffuse through the gain medium. Though the path of the photon is rectilinear, due to the random change in direction off each scatterer, the net effect is one of diffusion-like propagation (figure 4). In the diffusion regime, the path length of photons in the gain medium is vastly increased and hence large amplifications are seen.

The above picture seemed quite satisfactory, till some researchers found lasing in very small samples, for example, in samples of about 30 microns in length, which was much smaller than the transport mean-free path of 200 microns [17]. In another experiment, Prasad and coworkers [19] made use of polystyrene microspheres which were smaller and had a lower refractive index in contrast with the surrounding medium as compared to other studies. Both factors served to increase $l^*$ up to 3.5 cm in some cases. In addition, an aqueous solution of dye with much poorer efficiency was used. Yet, lasing action was observed even for samples of size 1 cm $\times$ 1 cm $\times$ 1 cm, where clearly $L < l^*$. In this low scattering limit, the effect of the scatterers is minimal, and no new effect is expected to arise. To explain the observed lasing, a simple probabilistic model was proposed according to which the very rare sub-mean free path scatterings are rendered important due to the gain of the system. Based on this hypothesis, an expression was derived [19] for lasing threshold in random lasers with distributed non-resonant feedback, and it was shown that for typical values of the parameters, threshold condition can indeed be met for $L \ll l^*$.

The other extreme case of $l^* \ll L$, i.e., the strong scattering regime too is very interesting. The photons are expected to be localized and to follow closed loop paths, due to recurrent multiple scattering, much like the electronic analog of Anderson localization. There is, however, one significant difference. Due to the bosonic nature of light, it can be coherently amplified. Kumar and Pradhan [20] showed that in the presence of gain in a disordered optical medium the Anderson localization enhances coherent amplification, i.e., the localization not only enhances amplification, it also maintains coherence due to the resonant closed loop paths. This should lead to a temporally coherent emission, albeit in a random direction. Examining a 1-d system, they conclude that the enhancement of emission does not arise merely due to an increase in sojourn time of the photon inside the gain medium, but there exists synergy between Anderson localization and coherent amplification.

After the first experimental report on the localization of light [21], a group from Illinois reported experimental observation [22] of lasing action from a scattering amplifying medium that was capable of localizing light. Not only is the emitted light of narrow spectral width (0.09 nm), it is also shown to have a large temporal coherence, which other random lasers do not have. The system consisted of micron-sized particles of disordered semiconductors like ZnO, and so has earned the name ‘random microlaser’. A typical cluster of 1.7 microns contains roughly 20 000 ZnO nanocrystallites. Usual microlasers require expensive state-of-the-art crystal growth and microfabrication facilities. The random microlaser, in contrast, is extremely simple to prepare. Figure 5 shows the emission from random microlasers at different pump powers. Because of the strong scattering, it is suggested that the physical mechanism of optical confinement is based on Anderson localization.

Recently Joshi and Jayannavar [23] studied the statistics of transmission and reflection from a random medium with stochastic amplification as opposed to coherent amplification. Their model for amplification, in the framework of time-independent wave equations gives qualitatively different results for statistics of super reflection in the strong amplification regime. A closer examination of the experimental data would help distinguish between coherent and stochastic amplification.

There has also been some effort in trying to explain the characteristics of emission and to predict *a priori* the emission profile of random lasers. Balachandran and coworkers [24] have developed a model based on transient two-level laser equations and includes detailed spectral properties of dye gain system. With this they could simulate threshold behavior for lasing action, when feedback was provided by scatterers. They have shown from their simulation that ASE is inadequate to describe the observed threshold behavior in these systems as they do not provide sufficient gain-length products.

Recently Monte-Carlo simulations have been used to derive the emission profile of random lasers. The authors show that the peak emission wavelength is determined by the absorption and emission profiles of the dye. Given the dye-scatterer system, they predict the spectral characteristics of random lasers, including bimodal emission and the competition between modes [25].

Now we turn to the possibility of applications of mirrorless lasing. Due to the simplicity and ruggedness of these systems, they are likely to replace conventional lasers where coherence and linewidth are not important, i.e., where it is merely necessary to have a high intensity source. The disadvantage in these systems at present is that they need a pump laser to excite the emission. With the development of mirrorless lasers with flashlamp or other simpler form of pumping, the use of random lasers will become more prevalent.

One of the earliest applications envisaged is ‘photonic textiles’. For several years, polymer materials have been used as hosts for dye lasers [26,27]. This provides the tunability of dye lasers with the practical convenience of solid-state lasers. Balachandran and coworkers

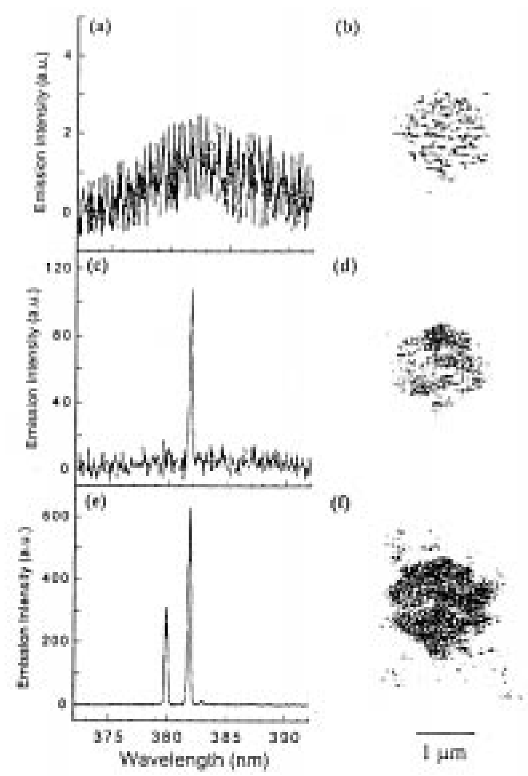


**Figure 5.** (**a**), (**c**) and (**e**) are the spectra of emission from a ZnO cluster about 1.7 micron across at pump pulse energies of 0.26 nJ, 0.35 nJ and 0.50 nJ respectively. (**b**), (**d**) and (**f**) are the corresponding spatial distributions of emission intensity from the cluster (figure from ref. [22]).

[28] have shown an interesting application of mirrorless lasing. They have tumble-coated laser dyes like pyromethene 567, rhodamine 590 chloride, rhodamine 640 perchlorate and scatterers like $TiO_2$ onto textile resins like nylon-6, polyester, PET, Kevlar, Nomex and drawn them into fibres. Fibres which had no scatterers on them showed morphological resonances in their emission due to their cylindrical shape (figure 6). On the addition of scatterers, the morphological dependent resonance disappeared, and the emission spectrum consisted of a single narrow peak. A pair of twisted fibres containing different dyes gave an emission of two distinct peaks, separated in wavelength (figure 7). This then gives the possibility of incorporating codes into the weave of textiles, by using fibres with different dyes and scatterers. These codes may be remotely read out by exciting emission in these coded random lasers. A possible military application is to have a small area in a person's dress with a coded textile that may be excited with a weak laser or flashlamp from a distance, and read out the code and identify the person as a friend or a foe. A somewhat similar, but civilian application is remote bar-code reading. It is estimated that 100 wavelengths can be coded into the usable range of a typical silicon detector array. A code with

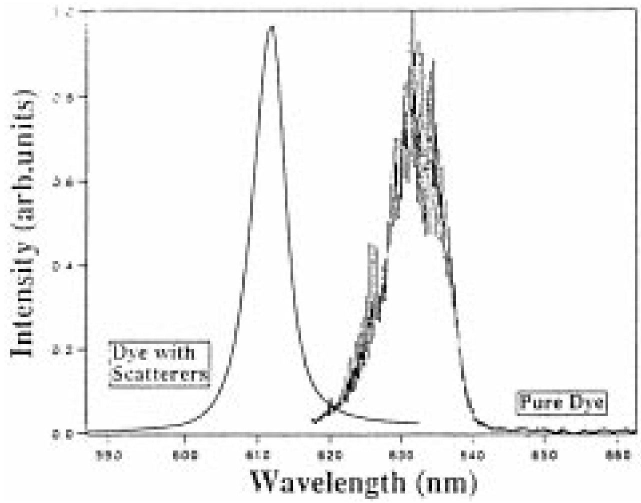


**Figure 6.** Emission from nylon-6 fibers containing rhodamine 640 perchlorate dye, with and without scatterers. The spikes seen in the pure dye case are modes due to morphological resonance (from ref. [28]).

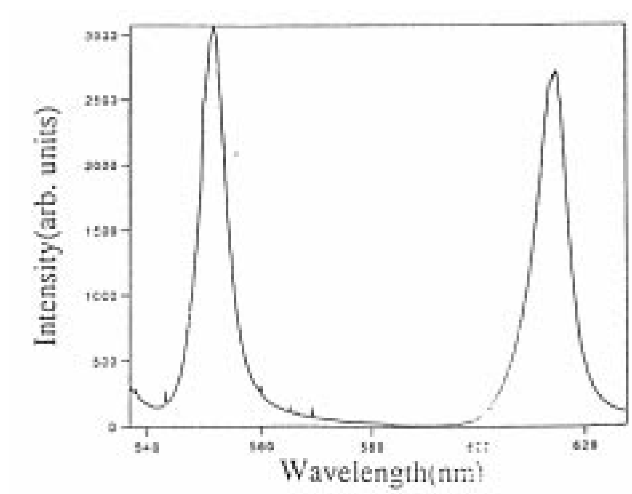


**Figure 7.** Emission from a braided pair of nylon fibers, one with pyromethene dye and the other rhodamine. Both contain $TiO_2$ scatterers (from ref. [28]).

$N$ possible entries can produce $2^N - 1$ unique signatures, which for $N = 100$, is about $10^{30}$. Proof-of-principle experiments demonstrating validity of concept, and ranging field tests were carried out at Goddard Space Flight Centre, USA.

Another suggestion is that dresses of persons undertaking perilious missions like deep sea diving, may be made of photonic textile, so that in an emergency, they may be easily located by virtue of the strong emission from their clothing. Yet another suggestion is to have flat panel displays in television and computer monitors, made up of random lasers.

It has also been shown [29] that photonic layers of polymers can be made, and that successive layers be radiatively coupled. Figure 8 shows the schematic view, where layers A and B (typically monomethylmethacrylate sheet, 1 mm in thickness), have different dyes and scatterers dispersed in them and polymerized. The emission of the top layer pumps the lower layer. Layer A is transparent to the emission of layer B. In such a system, if viewed from the top, the emission spectrum will be a combination of emissions of each individual layer, as is seen in figure 9. Thus, by multiple stacking of layers, it is possible to incorporate wavelength domain codes. These codes can be implemented

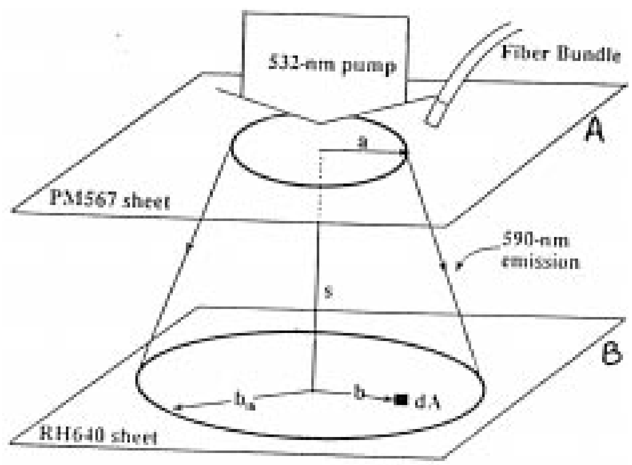


**Figure 8.** The polymer sheets A and B both contain $TiO_2$ scatterers, but different dyes. Sheet A is pumped with an external laser at 532 nm, while its emission pumps sheet B. The emission of both sheets can be viewed using the fiber bundle at the top (from ref. [29]).

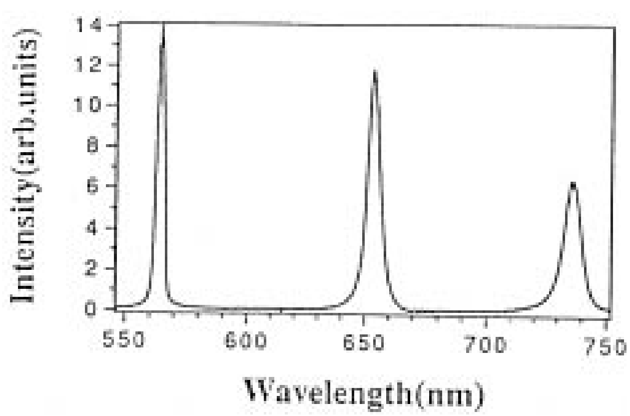


**Figure 9.** Laser emission from a three-layer stack (from ref. [29]).

by the use of polymeric sheets of liquid-base paint coatings to yield multilayer stacks on a rigid structure. The lower layers are invisible, but can be read out by a single excitation laser. Using a two-layer structure with 16 stripes, one can generate roughly $4 \times 10^{9}$ photonic codes that permit unique signatures. Such codes find applications in a variety of fields like anti-counterfeiting, long range identification of military friend or foe, hazardous materials marking, search and rescue applications, etc.

Another application of mirrorless lasers is in making ‘optical diodes’, an optical analog of the ubiquitous diode in electronics. A diode permits the flow of current in one direction, and prohibits it in the other. It has recently been shown [30] that a capillary with dye with graded gain permits the transmission of light of certain wavelengths in one direction, and not in the other. The addition of scatterers into the dye helps tailor the pass band of wavelengths. Thus, graded gain mirrorless lasers can provide directional transmission of light.

To summarize, mirrorless lasers that were proposed to explain some astrophysical phenomena, have now been realized in the laboratory, and even on micrometer scale. These devices, based on weak or strong localization of light, provide very simple means of obtaining high intensity light sources. Numerous applications have been proposed and its practical use appears only to be limited by our imagination.

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