The Long Journey to the Laser and its Rapid Development After 1960

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Abstract:

The laser, a fascinating new light source with numerous applications in our daily life was first realized some 50 years ago. The principle was initiated in 1916 when Einstein introduced a new concept of radiation-matter-interaction known today as stimulated emission of electromagnetic radiation. It took nearly 40 years before a first practical device based on stimulated emission - the maser - was realized for microwaves in 1954. In 1960, the first laser was operated in the visible. This historical note reports on the way to the laser with its climax in 1960. The subsequent explosive development of new lasers and of fundamental applications is shortly reviewed in the second part of the paper.

1. Introduction

At the beginning of the 19th century two young scientists, Young (1773 – 1829) and Fresnel (1788 – 1827), demonstrated clearly that light is a wave phenomenon, i.e. light waves are oscillations of the ether travelling with a high well defined velocity through space. The wave picture of light allowed to explain numerous optical experiments of the time. For instance, reflection, refraction, interference and diffraction phenomena were quantitatively explained by the wave model of light. But the question remained: what was the light wave really? The breakthrough was made by Maxwell (1831 – 1879) in the second half of the 19th century. He was able to show that light is an electromagnetic wave where the vectors of the electric and magnetic fields are perpendicular to each other. His theory, the famous Maxwell-equations, was able to explain quantitatively most of the existing experiments on light wave phenomena. Even today Maxwell´s equations are of great value in physics.

Three topics should be discussed first which were well investigated experimentally but their interpretation was left to the turn of the century or the following 20th century. More important, these experimental findings and their theoretical explanations were essential for the way to the laser.

(i) The photoelectric effect: After the first discovery by A. E. Becquerel in 1839, it was found later in several laboratories (Hallwachs, Lenard) that metal surfaces – when irradiated in vacuum by ultraviolet light of frequency ν – emitted electrons only when ν was larger than a certain frequency ν_0. Even light of high intensity but with $\nu < \nu_0$, did not release electrons from the metal surface.

(ii) The spectroscopy of gases: The fluorescent emission of atoms in the gaseous state was the subject of extensive investigations in the 19th century. It was found that the atoms of

different chemical elements radiated with a specific set of narrow lines. Of special interest was the spectrum of hydrogen which had several series of lines named after famous spectroscopists, Lyman, Balmer and Paschen. It was quickly noted by spectroscopists that the line spectra were of great value to the analytic chemist. The chemical composition of a certain material could be readily seen in the flame of a Bunsen-burner with the help of a simple spectrometer. Various mathematical formulas gave some order to the numerous emission lines of certain elements, but were not able to explain the emission phenomena. New concepts were necessary to understand the atomic spectra (see below).

(iii) The black body radiation: The light emission of a hot body was investigated prior to 1900. When the emission (more accurately the energy per frequency interval, per area, per unit time) was plotted as a function of frequency one found a curve rising from zero to a maximum and then slowly decaying to high frequencies. Several physicists tried to find an explanation of this radiation curve and derived approximate formulas to the experimental data. W. Wien (1864 - 1928) presented an equation which provided a fairly good approximation for high frequencies. His derivation was based on a statistical picture of the thermodynamics of the days. The Rayleigh-Jeans law gave an approximation of the black-body curve only at very low frequencies, based on classical thermodynamics. The final explanation was given in 1900 by Max Planck.

Planck´s radiation law: With the turn of the 19th century classical physics terminated. The new time of quantum physics started in December of 1900 with a talk of Max Planck (1858 – 1947) at a meeting of the German Physical Society in Berlin. Planck, a professor of theoretical physics, proposed a formula which fitted perfectly the black-body radiation curve over the whole frequency range. But in doing so he introduced something completely new – we may say something revolutionary – for the physicists of his time. He suggested the concept of light quanta where the energy of the light quantum E is proportional to the frequency: $E = h\nu$ (h being a constant, today called after Planck). The proposal of light quanta appeared unacceptable to the contemporary scientific community, which was convinced for more than 100 years that light radiation was a continuous wave phenomenon. Now, light quanta, light packets, should exist; but nobody had ever seen them! In fact, Planck was severely attacked by numerous colleagues for a few years.

Einstein's explanation of the photoelectric effect: The situation changed completely when in 1905 an essential paper appeared in the Annalen der Physik. Einstein (1879 – 1955) a young physicist without prejudices of the older generation explained the external photoeffect (see above) using the quantum picture. To free an electron from the surface of a metal a certain energy E is required which is provided by the light quantum of frequency ν_0 with $E = h\nu_0$. Light with smaller frequency is unable to free electrons and is thus ineffective in the external photo effect. This picture was very convincing and Einstein received the Nobel Prize 1921 for this simple explanation of the external photoeffect. Planck highly respected for the introduction of the quantum-picture received the Nobel Prize 1918.

From the atomic model to quantum mechanics: In 1911 Rutherford (1871 - 1937) postulated a model of the atom where the small positively charged nucleus is surrounded by the negatively charged electrons. Bohr (1885 - 1962) who worked with Rutherford in Manchester developed a theoretical picture of the atom: Electrons exist in stable quantum states around the nucleus and transitions between these states correspond to the observed absorption or emission lines of the specific atoms. Sommerfeld (1868 - 1951) formulated quantization rules which in turn led to the concept of quantum mechanics by Heisenberg (1901 - 1976) and Schrödinger (1887 - 1961) in the years 1925/26.

In the present historical note we want to describe the progress leading to the laser from the viewpoint of a contemporary scientific readership. Therefore for the most exciting period 1950 to 1960 our article is based on the primary literature published at that time in peer reviewed journals. In addition we quote the book of Maiman which we consider as a primary

personal source of information on the course of events in his lab [Maiman 2000], and his speech at the press conference on July 7, 1960 [Maiman 1960a], not accessible from peer reviewed journals.

The expression laser is an acronym introduced in the late 1950s. Laser stands for Light Amplification by Stimulated Emission of Radiation. The word laser nowadays describes an instrument, more precisely a non-classical light source, with new properties:

- emission with high spatial and temporal coherence,
- pencil like emission beam (close to diffraction limit)
- high irradiance or high intensities.

2. Stimulated Emission

In 1916 Einstein published a paper entitled: "Zur Quantentheorie der Strahlung" (on the Quantum Theory of Radiation) where he derived the black body radiation law in a new way different from Planck´s paper [Einstein 1916; Einstein 1917]. Because of the importance of Einstein´s idea for our article, a short remark to the most important points should be given.

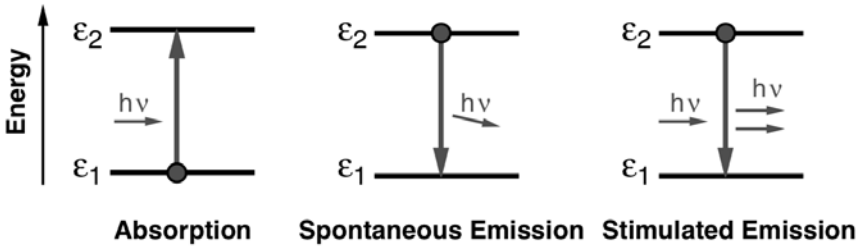


Fig 1: Schematics of the three interaction processes between light and a quantized atomic system represented by two energy levels.

In Fig. 1 two atomic energy states with energy values ε_1 and ε_2 and populations N_1 and N_2 are depicted schematically. In addition to absorption (1) and spontaneous emission (2) Einstein postulated a third interaction process, stimulated emission (3).[1] More detailed:

(1) A system in a state with energy ε_1 absorbs photons from an incident radiation field with spectral density $u(\nu)$ (at a frequency $\nu = (\varepsilon_2 - \varepsilon_1)/h$). This decreases the population N_1 and increases the population N_2 of the excited state ε_2:

$$-\frac{dN_1}{dt} = \frac{dN_2}{dt} = B_{12}u(\nu)N_1 \qquad (1)$$

(2) An excited system reduces its population N_2 by the known spontaneous emission:

$$\frac{dN_2}{dt} = -A_{21}N_2 \qquad (2)$$

(3) The stimulated emission is the essential mechanism for the laser. An excited system in state ε_2 is stimulated by the radiation density u to emit light which decreases the population density N_2.

$$\frac{dN_2}{dt} = -B_{21}u(\nu)N_2 \qquad (3)$$

In thermal equilibrium population changes due to absorption equal those due to emission:

$$B_{12}u(\nu)N_1 = (A_{21} + B_{21}u(\nu))N_2 \qquad (4)$$

With Boltzmann´s distribution of statistical physics we obtain:

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[1] It should be noted that the expression "stimulated emission" was not used in the original publication.

$$\frac{N_2}{N_1} = \exp\left[\frac{\varepsilon_1 - \varepsilon_2}{kT}\right] = \exp\left[-\frac{h\nu}{kT}\right] \tag{5}$$

$$B_{12}u(\nu)\exp\left[\frac{h\nu}{kT}\right] = A_{21} + B_{21}u(\nu) \tag{6}$$

k is known as Boltzmann's constant. For very high temperatures, $T \rightarrow \infty$, the radiation density *u* also diverges, $u \rightarrow \infty$, whereas A_{21} remains constant. Equation (6) leads to $B_{12} = B_{21}$ and one obtains an expression for the radiation density:

$$u(\nu) = \frac{A_{21}}{B_{12}} \frac{1}{\exp\left[\frac{h\nu}{kT}\right] - 1} \tag{7}$$

For small frequencies $h\nu << kT$ comparison with Rayleigh-Jeans law yields $\frac{A_{21}}{B_{12}} = \frac{8\pi h\nu^3}{c^3}$.

Finally one obtains Planck´s radiation law.

$$u(\nu) = \frac{8\pi h\nu^3}{c^3} \frac{1}{\exp\left(h\nu / kT\right) - 1} \tag{8}$$

We wish to repeat: With the introduction of stimulated emission at thermal equilibrium, Einstein succeeded to derive the black body radiation formula. Stimulated emission occurs when an excited system interacts with a well defined radiation field. In other words radiation passing through an excited system is amplified by stimulated emission. It is interesting to note here that the practical use of stimulated emission for a light amplifier requiring non-equilibrium populations was not recognized by the scientific community during the following decades. Terms such as negative absorption or negative dispersion were introduced in the discussion of experiments where stimulated emission played a definitive role (see below).

3. Starting in the 1920s

In his original publication Einstein introduced the three different light-matter interaction processes absorption, spontaneous emission and stimulated emission (Fig. 1) [Einstein 1916; Einstein 1917]. But the effect of the individual interaction processes on the light field itself was not treated in detail. Einstein describes it in a very general way as a process where radiation with energy ε_m - ε_n is released (*"wobei die Strahlungsenergie ε_m - ε_n frei wird"*) and adds that the emission is a directed process: *"..., dass auch der Prozess der Ausstrahlung ein gerichteter Prozess sei"*. In the early 1920s several groups addressed the question of induced light matter interaction:

Einstein and Ehrenfest [Einstein 1923] introduced the expression negative irradiance ("*negative Einstrahlung*") for the emission of a quantum by action of irradiance.

Kramers discussed the light matter interactions in different publications [Kramers 1924a; Kramers 1924b; Kramers 1925]. He used the expression "*negative absorption*" and formulated the term "*negative dispersion*" for the consequences of induced emission on the index of refraction.

In 1924 Van Vleck introduced the expression "induced emission" in [Van Vleck 1924]: *"We shall define as the differential absorption the excess of positive absorption due to the upward transition ..., over the negative absorption (induced emission) for the corresponding downward transition "*.

Bothe [Bothe 1923] used the expression "erzwungene Emission" (forced emission) and repeats that the three radiation processes have to occur -according to Einstein- in a strongly directed way: *"... wenn die drei angegebenen Elementarprozesse streng gerichtet sind"*.

It was Tolman in 1924 [Tolman 1924] who interpreted the expression negative absorption and wrote *"that molecules in the upper state may return to the lower quantum state in such a way as to reinforce the primary beam by "negative absorption""*. He *"... pointed out that for absorption experiments as usually performed the amount of "negative absorption" can be neglected"*.

In a publication on radiation pressure equilibria in the atmosphere of stars [Milne 1924] Milne presented in 1924 an equation describing the change in light intensity during propagation through a medium (absorption cross-section σ) with populations N_2 and N_1 of the upper and lower levels respectively. For a small cone of the radiation his equation is equivalent to

$$\frac{dI}{dx} = \sigma(N_2 - N_1)I \tag{9}$$

which includes the case of amplification ($dI/dx > 0$) for population inversion, $N_2 > N_1$.

In a series of experiments Ladenburg [Kopfermann 1928b; Kopfermann 1928a; Kopfermann 1928c; Kopfermann 1928d; Ladenburg 1928; Ladenburg 1933] focused on "negative dispersion" and succeeded to prove experimentally the existence of negative dispersion in electrically excited neon. This was the first (indirect) experimental evidence for stimulated emission. For excited helium negative dispersion was shown in 1938 [Kruse 1938]. These experiments clearly demonstrated that in an electrical discharge the population densities of high energy states are increased inducing negative dispersion. However, these publications did not yield a direct experimental proof of population inversion and light amplification.

In the 1930s the phenomena negative dispersion (and consequently negative absorption) was generally accepted. However, the intrinsic technical potential of negative absorption – the amplification of light and novel light sources based on stimulated emission – was not considered. In 1939/1940 V.A. Fabrikant in his Doctor of Science thesis addressed light

amplification.[2] Fabrikant proposed to change population densities of excited states (e. g. by impurities) to obtain amplification. In a patent application in 1951 Fabrikant suggested light amplification by stimulated emission from the ultraviolet to the microwaves and proposed to use repetitive propagation through the excited medium.

In 1950/1951 Purcell and Pound found population inversion in a nuclear spin system [Purcell 1951] and used the expression "negative temperature" to describe the artificial population densities. They specified: *"A system in a negative temperature state is not cold, but very hot, giving up energy to any system at positive temperature put into contact with it. It decays to a normal state through infinite temperature."* The expression "negative temperature" was formally used for the two level spin system and describes in this case the population inversion with a higher occupation of the upper state $N_2 > N_1$ using Boltzmann's equation with a negative temperature T.

$$\frac{N_2}{N_1} = \exp\left[-\frac{(\varepsilon_2 - \varepsilon_1)}{kT}\right] \tag{10}$$

However an inverted system is not in thermal equilibrium and therefore Boltzmann's equation is completely inadequate and the expression "negative temperature" is misleading. This fact becomes evident if more than two levels are considered. Even the most simple laser system, a three level system with population inversion produced by optical pumping, can not be described by Boltzmann's equation and a single (negative) temperature T. Nevertheless the expression "negative temperature" was used as a synonym for "population inversion" in the early laser period.

[2] A review of the role of Fabrikant on the way to the laser was given recently by Lukishova [Lukishova 2010].

4. The Maser

Looking back at the previous chapters we clearly see that the stimulated emission introduced by Einstein 1916 was well accepted by numerous physicists and the population of excited states was visualized by reduced absorption and by negative dispersion. Early in the 1950s, work on microwave amplification started at various laboratories [Weber 1953; Basov 1954; Gordon 1954].

Charles Townes, a physics professor at Columbia University, had the idea to use stimulated emission for a microwave oscillator [Townes 1999]. He stated at various occasions, that he had the advantage of extensive experience in microwave technology and at the same time good knowledge in quantum physics. In fact, Townes worked during the war at the Bell Laboratories with engineering groups on Radar-systems and later at the Columbia University he studied microwave spectroscopy of molecules. This combination of knowledge led in 1954 to the first experimental realization of a maser (an acronym of: Microwave Amplification by Stimulated Emission of Radiation) a microwave oscillator using an ammonia transition at 24 GHz (λ = 1.25 cm). A beam of excited molecules passed through a resonant cavity where the amplification by stimulated emission was larger than the losses due to output coupling. The first maser was designed by Townes with his coworkers J. P. Gordon and H. J. Zeiger and went into operation in 1954 [Gordon 1954]. In the same year the possibilities to make a molecular oscillator were discussed by Basov and Prokhorov [Basov 1954]. The ammonia gas maser was a two level system where the resonator had to be refilled continuously by excited molecules. A three level pumping system was introduced in 1955/6 [Basov 1955; Bloembergen 1956] which made continuous operation of the maser possible. In December 1956 an important progress was made through the introduction of solids as maser material by Scovil, Feher and Seidel [Scovil 1957] at the Bell Laboratories. Finally in 1957 ruby was introduced with great success for practical maser amplifiers. In synthetic pink ruby crystals the weak splitting of the electronic ground level by a magnetic field was used for maser operation.

The advantage of the maser was its very low noise level; it was a highly sensitive microwave detector. Two examples: The low noise maser amplifier was applied to radio astronomy by J.A. Giordmaine (one of Townes graduate students) and coworkers [Alsop 1958]. Several years later Penzias and Wilson at Bell Laboratories found the cosmic background radiation originating from the Big Bang with the help of a ruby maser.

5. From the Maser to the Laser

The maser generated microwave radiation. The latter was available since 1940 by devices such as magnetrons or klystrons. But the maser was based on a new concept, the stimulated emission, which initiated the discussion of its possible extension to much higher frequencies. To the people concerned with communication it was a fascinating thought to have coherent and monochromatic electromagnetic radiation in the visible part of the spectrum. This radiation would supply an ideal carrier frequency of several orders of magnitude more information than with existing microwave transmitters. It is not surprising therefore that the first detailed paper which extended the microwave technology of the maser to the infrared and optical region was written 1957 by Schawlow and Townes (as consultant) at the Bell Laboratories; it appeared 1958 in Physical Review [Schawlow 1958]. Of special importance was the introduction of a resonator cavity useful for the optical wavelengths, i.e. for the 500 nm range. The authors proposed an extended Fabry-Pérot system where two plane parallel mirrors are positioned at the ends of the amplifying medium. One partially transparent end-mirror provides the output radiation. A detailed analysis of the mode structure of this resonator suggested the possibility to select one or a few modes in the direction of the system axis. Such a device should emit light with a high degree of spatial coherence, i. e. with a highly collimated beam of low divergence.

The Schawlow-Townes paper of 1958 [Schawlow 1958] was a strong stimulus to start research related to the laser at different industrial laboratories, e.g. Bell Laboratories, IBM, TRG, Hughes, American Optical. Research was focused on materials which appeared promising as laser-active media. The observation of a "negative temperature" in gaseous systems, discussed above suggested gas discharges as possible laser media. As an example the paper by Javan published in 1959 should be quoted [Javan 1959]. He suggested a Helium-Neon gas mixture where helium is electrically excited to a meta-stable state and subsequently transfers its energy to the neon atoms where one of the numerous transitions should lead to laser action (see below).

A paper by Fabrikant's group [Butaeva 1959] published in 1959 in Russian claimed to have observed light amplification of several percent in Mercury and Cesium. However it is not clear that this paper was known to the scientific community in the USA before the realization of the first laser. The experiment was repeated in 1961 [Sanders 1962], when attempts failed to reproduce the optical amplification in mercury.

Interesting laser media were transparent crystals doped with ions fluorescing with high quantum efficiencies and with favorable pumping possibilities by external light sources. As an example we point to tungstates doped with rare earth ions (see below). Of special interest for the following is the ruby crystal consisting of aluminum oxide (Al_2O_3) doped with chromium ions, Cr^{3+}. The color of the crystal depends on the Cr^{3+} concentration. Pink ruby has a doping of only approximately 0.05% chromium oxide while red ruby has a considerably higher Cr^{3+} concentration.

Of importance for the laser action is the energy level diagram of the individual material. In the following pink ruby will be of interest and the corresponding energy states are depicted schematically in Fig. 2. [Sugano 1958; Maiman 1960b]

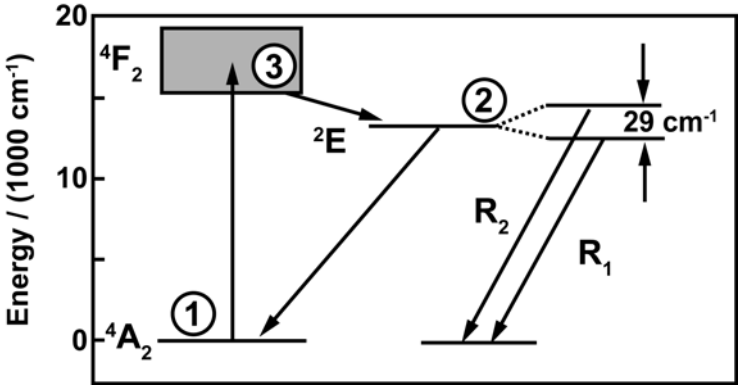


Fig. 2. Level scheme of pink ruby (after [Maiman 1960b]) forming an effective 3-level laser system. (The extremely weak ground-state splitting used for maser operation is omitted.)

Pink ruby has broad and strong absorption bands in the blue and green region of the spectrum due to transitions from the $^4A_2$ ground state to the levels $^4F_1$ and $^4F_2$. From here excitation is rapidly transferred to the lowest excited levels ^2E (split by the crystal field). The ^2E levels were found to have a rather long lifetime (ca. 3 ms at room temperature) [Maiman 1960c]. From here fluorescence emission occurs via the R_1 and the R_2 lines.

As shown in Fig. 2 pink ruby is an effective 3-level system (see Fig. 3) where population inversion between level 1 and 2 is difficult to achieve: More than 50% of the ground state population has to be evacuated. In 1959 Schawlow suggested dark ruby as laser material. In dark ruby (highly doped) strong splitting of the ground state occurs and provides a 4-level system (see Fig. 3). At cryogenic temperatures the population of the terminal state of the fluorescence is strongly reduced and population inversion becomes possible for low pumping power. There is no need to evacuate the ground-state to more than 50% as is the case for a three level system (pink ruby).

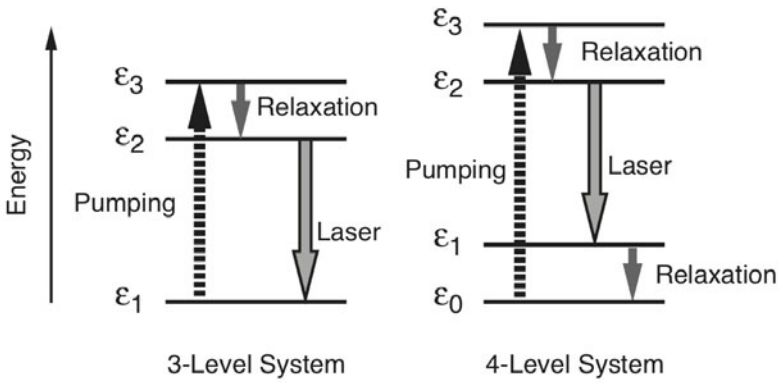


Fig. 3. Schematic of transitions in 3- and 4-level systems. In a 4-level system the lower laser level at ε_1 has - with $\varepsilon_1 - \varepsilon_0 >> kT$ - a low thermal population. Here amplification can be reached at much lower pumping power than in a 3-level system, where the lower laser level is the ground-state.

The Role of Patents: At the end of the 1950s the fundamental concepts and many technical details for the realization of the Laser were established from scientific publications. In the same period a series of patent applications have been submitted. Most prominent are patents by Fabrikant (see above), Dicke (granted in 1958) who proposed the use of a resonator, Schawlow and Townes (granted in 1960) and finally by Gould (filed in 1959 but only granted in 1977). An analysis of the early laser patents can be found in [Myers 2003]. Most of the patents remained in the stage of patent evaluation in 1960, i. e. at the time the laser was realized. Patents did not notably influence the development of the first laser. The essential impact came from scientific publications.

6. The year 1960

In 1960 the first laser was realized and the sequence of events - deduced from published literature - will be discussed below in more detail.

June 1st, 1960: A paper of optical experiments on pink ruby by T. H. Maiman, a physicist at the Hughes Aircraft Corporation was published in Phys. Rev. Lett. [Maiman 1960c]. He had worked for several years on a maser based on pink ruby crystals. With pulsed optical excitation of the ruby Maiman found pronounced depletion of the ground state proven by bleaching of the ground-state absorption and by an excited state absorption. Ground state depletion and excited state absorption decayed with a time constant of ≈ 5 ms. In addition he measured a high quantum efficiency of the fluorescence in contrast to a previous publication [Wieder 1959].

July 7th, 1960: A press conference was held by the Hughes Aircraft Cooperation in New York with the headline of the press release (printed in Maiman's book from 2000 [Maiman 2000] on pages 114 to 117): "*US victor in world quest of coherent light*". In his speech (the document is available from the homepage of HRL-Laboratories (a follow-up of Hughes Aircraft Research Laboratories) [Maiman 1960a]) Maiman made the following statements:

- "*For the first time in history a source of "coherent" light was attained*".
- "*For the first time in scientific history we achieved true amplification of light waves.*"
- "*We will have an important new scientific tool for investigating properties of matter and for performing basic experiments of Physics.*"
- "*Another important property of a laser, indirectly a consequence of its coherence, is that it radiates an almost perfectly parallel beam.*"
- "*A paper describing this laser in detail has been submitted to the U.S. "Journal of Applied Physics" and also to "Nature", a British scientific journal.*"

The next day (July 8th) the New York Times reported under the headline *"Light Amplification Claimed by Scientist"* on the press conference: *"Dr. Maiman reported a power gain of five for the amplified signal over the input. Amplification by a factor of more than 100 is believed to be necessary for applying the laser".*[3]

August 6th, 1960: The first of two scientific publications mentioned in the press conference appeared in "Nature" [Maiman 1960b]. We quote "*... optical pumping technique has been successfully applied to a fluorescence solid, resulting in attainment of negative temperatures and stimulated optical emission at a wavelength of 6943 A; ...*" Only very few experimental details are given in this short paper: "*... a ruby crystal of 1 cm dimensions coated on two parallel surfaces with silver was irradiated by a high power flash lamp*". The important experimental results are depicted in Fig. 4 (originally Fig. 2 of [Maiman 1960b]) where the emission spectra of the ruby obtained under low (a) and high power excitation (b) are shown. The spectral bands from the two transitions R_1 and R_2 were represented in the figure as boxes. Upon increasing excitation energy the R_1 line gained in amplitude (relative to R_2) and was narrowed by a factor of approximately four. The author stated: "*These results can be explained on the basis that negative temperatures were produced and regenerative amplification ensued.*"

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[3] It should be mentioned that according to [Maiman 2000] amplification of light was observed for the first time on May 16, 1960. This date was celebrated in 2010 as the birthday of the laser.

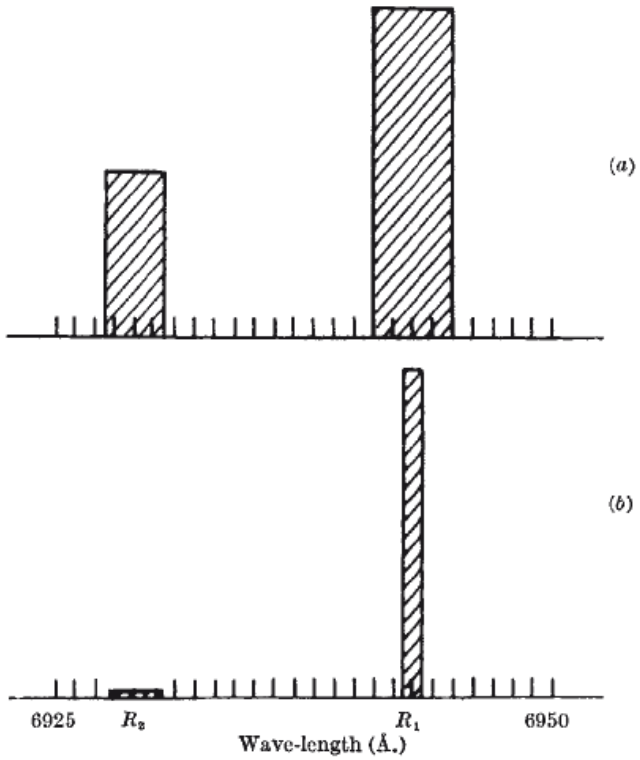


Fig. 4. Emission spectrum of ruby at low (a) and high power excitation (b). (Reproduction of Fig. 2 of [Maiman 1960b]).

The experimental facts in the Nature papers were moderate changes in the relative line intensities of the R_1 and R_2 bands and a weak narrowing of the R_2 band by a factor of four. These features indicate that light amplification in the visible spectral range was realized for the first time. A coherent and nearly parallel beam as claimed in the press conference was not reported. Apparently not all statements given in the press conference were presented in the Nature paper.

September 1960: The second paper was published in "British Communications and Electronics" [Maiman 1960d] containing somewhat more experimental information. The chromium concentration of the ruby crystal was given now, but no additional information on the crystal seize appeared. "*The sample was 0.05 per cent (Cr_2O_3: Al_2O_3) ruby with 1 cm dimensions. Two parallel faces of the crystal were coated with evaporated silver, except for a small hole in one end face through which the fluorescent radiation was observed*". Experimental results: the same emission spectra were given as in the "Nature" paper (see above, Fig. 4). Now numbers for the band narrowing and changes in relative intensities ("*attributed to regenerative effects*") were presented: "*We note that the R_1 line was narrowed from about 4A to less than 1A, R_2 was only slightly narrowed (3A to 2.7A), and finally the peak intensity ratio of R_1 : R_2 increased from 2 : 1 to 50 : 1*". Taking into consideration that the R_1-band was narrowed by a factor of four and that R_2 did not change considerably, the observed intensity ratio corresponds to an increased energy of the R_1 emission by a factor of (50/2)/4 ≈ 6.3. In addition it was shown that the time decay of the emitted intensity became non-exponential for high excitation powers: "*At high pulse power, the decay deviated considerably from the simple exponential with a very fast initial decay constant. With the maximum source intensity available, it was possible to reduce the initial decay constant to 0.6 m sec*" (see Fig. 5).

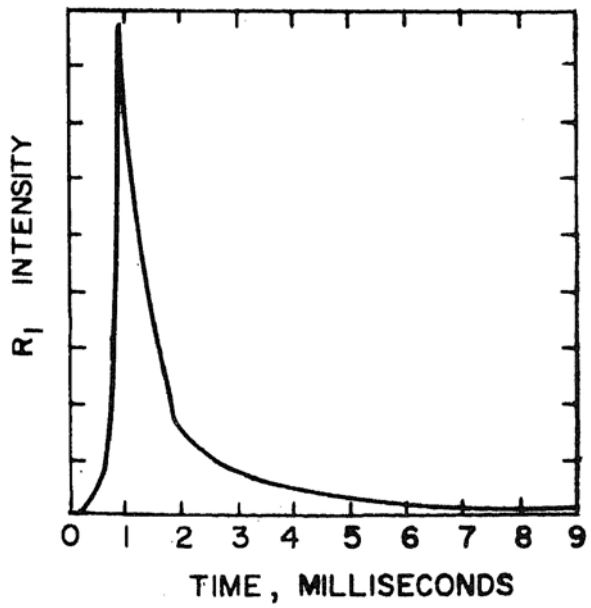


Fig. 5: Oscilloscope trace showing emission decay from ruby after high intensity pulse excitation (Picture reproduced from Ref. [Maiman 1960d], Fig 3).

In the section "Results" of the paper it was stated that "*The optical oscillator described here saturates at low gain and, consequently, the output is not appreciably narrowed. Because of internal reflections in the ruby, the present experimental arrangement shows very little angular discrimination in accepting spontaneous emission noise. A large fraction of the total spontaneous emission power is therefore effective as an input to the amplifier. A reduction of the spectral width as high as* $(D/\lambda)^2$ *(≈* 10^8*) could be obtained in principle ....*" This statement gives an explanation for the deviations of the realized device from the behavior expected for a laser above threshold. In fact [Maiman 1961b] mentions a large divergence of the device of 1 rad.

The two scientific publications describe an optical device operating close to but not necessarily above laser threshold. Light amplification starts to occur but the losses of the laser cavity may not be fully overcome by stimulated emission. It is important to note that at the press conference the impressive properties of a real laser operating well above threshold were presented. The performance of a real working laser was well known at that time from the theory. The essential properties are: the coherence and the intense and narrow beam.

October 1st, 1960: The next step in the development of lasers in 1960 was the publication of a paper by a group of the Bell Laboratories in New Jersey. The title reads "*Coherence, narrowing, directionality, and relaxation oscillations in the light emission form ruby*" by R. J. Collins et al., which appeared in Physical Review Letters [Collins 1960]. Additional results were given at a press conference held in New York on October 5th. The way to this publication has been described recently by some of the previous authors, Nelson, Collins and Kaiser [Nelson 2010]. The ruby work at the Bell Laboratories was triggered by the Hughes/Maiman press conference from July 7th. Collins group used cylindrical ruby crystals "*containing approximately 0.05 % chromium oxide were prepared in the form of rods 0.5 cm in diameter and 4.0 cm in length ... The ends were optically polished and were flat and parallel to 1 minute and were silvered so as to transmit 1 to 5 %*". The flashlight and discharge conditions were given in the paper. In this publication different properties of an operating laser were reported. As an unexpected feature, strong fluctuations of the laser output (spiking) were seen and attributed to relaxation oscillation (see Fig. 6). The various findings are summarized as follows:

- Emitted energy: "*... the ratio (R_1/R_2) for the light seen through the silvered ends, was found to increase as much as three orders of magnitude. The light emerging from the sides of the rod, however, still showed a ratio of the order of unity*". "*The energy emitted through the silvered ends and within the cone during a single pulse, as has been shown, was highly monochromatic and consisted of ~10^{-2} joule.*"
- Beam quality: The beam had very good directionality "*... This shows that the stimulated emission light is confined to an angle of the order of 0.3° to 1°*" (see below).
- Spectral width of the stimulated light: "*From the width of the fringes, the spectral linewidth is estimated at about 0.2 cm^{-1} as compared to the normal R_1 linewidth of 6 cm^{-1}*". I.e. a line narrowing by a factor of 30 was observed.
- The coherence of the emission was checked by Fraunhofer diffraction at a rectangular aperture.
- The imperfection of the ruby crystal was characterized and it was concluded that the oscillation did not occur on a single mode of the crystal etalon but "*Rather it is probable that there is a coherent excitation of a large number of modes at once, although a small fraction of the total modes*".
- Spiking: Instead of one peak with a lifetime of 0.6 ms as published by Maiman (see Fig. 5), Collins at al. observed "*... the excess signal in the R_1 line consists of a series of very intense, very short spikes in emission. Note that this observation differs in an interesting way from the "lifetime decrease" reported by Maiman. In our experience the spiking phenomenon was more pronounced, the heavier the silvering and the more nearly perfect the geometry. The duration of each spike is not greater than the instrumental resolving time (10^{-6} sec).*" The spikes represent relaxation oscillations. An individual spike originates from the rapid rise of intensity due to amplification above threshold and the corresponding depletion of population inversion.

The observations published by Collins et al. give convincing evidence that they had assembled and analysed a working laser.

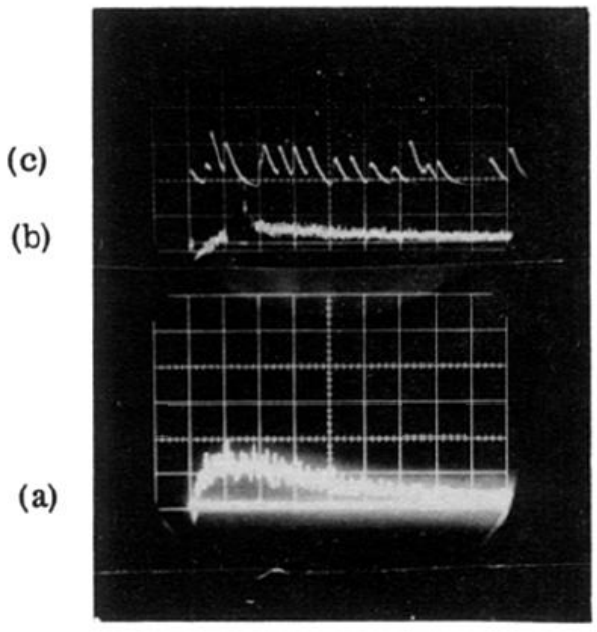


Fig. 6. Oscilloscope traces of the emission from the ruby laser system reproduced from [Collins 1960]. (a) emission recorded at low excitation level (time scale 1 msec/div). (b) Pattern at high excitation level. The break in the trace after 500 μsec results from the rapid increase in signal when stimulated emission begins (time scale 500 μsec/div). (c) Trace of

the stimulated emission region with 1000 fold decrease in ordinate scale compared to (b) and time scale 10 μsec/div showing directly the spiking of the laser emission.

In concluding the topic of the ruby laser we return to the results published by Maiman in two scientific journals and those presented at his press conference on July 7th, 1960.

From the two Maiman papers of 1960 one can directly deduce that light amplification (i. e. amplification of the spontaneous emission generated in the ruby crystal) was established. This fact can be seen from

- the spectral narrowing of the R_1 band by a factor of 4 and
- changes in relative intensities of the R_1 and R_2 bands by a factor of ~25 (when the line narrowing is considered a factor of 6.3 remains for the increase in emitted energy).

The first demonstration of light amplification in the optical range by Maiman was a significant progress. He introduced important achievements: pulsed operation by flash-lamp pumping, the use of a 3-level system (pink ruby) and the inventive lay-out of the ruby laser.

The question whether Maiman had a working laser at the time of his press conference will be discussed now. Of special interest in this context are two papers [Maiman 1961a; Maiman 1961b] which Maiman et al. published subsequently in August 1961. In one of the papers [Maiman 1961b] Maiman states:

"*It was found with high-intensity excitation that the nature of the output radiation from the various ruby samples which were tried could be divided into two categories:*

A. Crystals which exhibited R_1 line narrowing of only 4 or 5 times, a faster but smooth time decay of the output (compared to the fluorescence), an output beam angle of about 1 rad, and no clear-cut evidence of a threshold excitation. This type of behaviour was reported and discussed by Maiman (here [Maiman 1960b] was cited)*.*

B. Crystals which exhibited a pronounced line narrowing of nearly four orders of magnitude, an oscillatory behavior of the output pulse, and a beam angle of about 10^{-2} rad; these crystals were particularly characterized by a very clear-cut threshold input energy where the pronounced line and beam narrowing occured. This second category of behavior was reported by Collins et al. (here Ref. [Collins 1960] was cited)*, and is the subject of further study reported here.*"

Of special interest is Maiman's remark in point A that his early device had an output with an emission angle of about 1 rad, i. e. an emission angle of 55°. For plane parallel mirrors, separated by ca. 2 cm (3/4 in.) as was used in the publication such a large divergence obviously involves spurious internal reflections at the cylindrical surface of the ruby. Apparently the radiation was not efficiently confined in the resonator formed by the mirrors. It is hard to believe that above threshold a laser with the dimensions of the used ruby crystal will emit light of this large divergence.

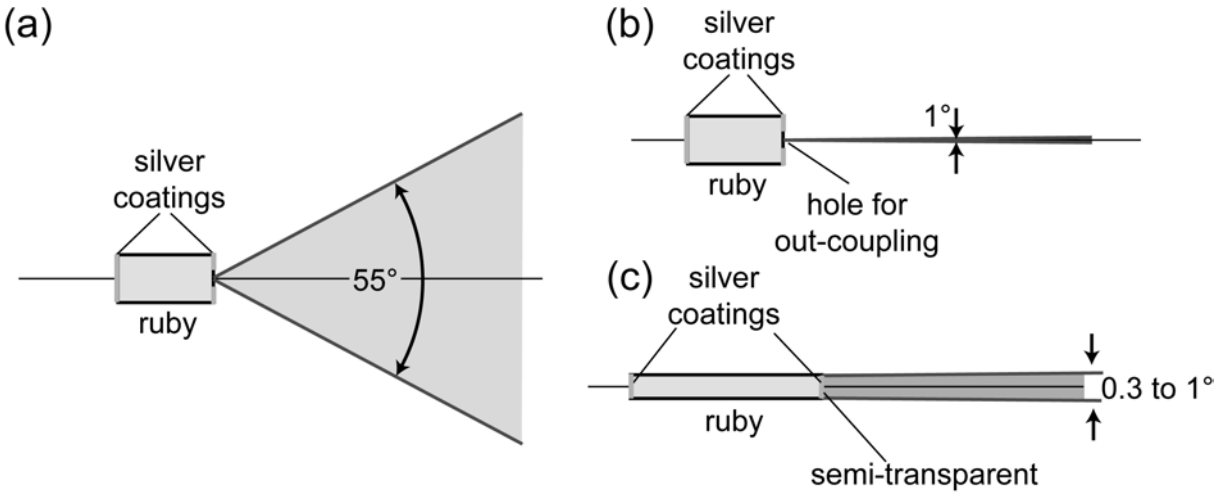


Fig. 7. Schematics of the emission of the devices mentioned in Ref. [Maiman 1961b] (a) refers to Maiman's device used in the 1960 papers. (b) corresponds to the device presented in 1961 [Maiman 1961b]. (c) refers to the laser with semi-transparent coating used by Collins et al. with a beam of divergence of 0.3° to 1°.

The reason for the wide emission angle in Fig. 7a was attributed by Maiman to the poor quality of the ruby crystal used in his initial experiment (see below). We recall the properties of a laser presented at the press conference were claimed to have nearly parallel beams.

The apparent discrepancies can be resolved using Maiman's book, "The Laser Odyssey" published in the year 2000 [Maiman 2000]. Maiman writes on page 125 that he received and tested new ruby crystals form the Linde Co. on July 20. That date is two weeks after his press conference in New York. With these new crystals new results were obtained as he writes: " *When we turned up the power supply this time, we found a very sharply defined threshold and we saw a small brilliant spot on the wall.*" Results on this operation mode were only published in 1961 [Maiman 1961b].

Closing this chapter on the ruby laser we want to present in Fig. 8 the basic set-up used in early ruby lasers. The essential component was the ruby crystal (Al_2O_3 doped at low concentrations with Cr^{3+} ions). A variety of length to diameter ratios of the ruby crystal were used in the early systems to optimize laser action. The polished end faces of the crystal were parallel and silvered in order to serve as the end mirrors for the optical "laser" cavity. Excitation of the ruby was accomplished by a helical flashlamp. The use of a helical lamp was inspired by the photographic equipment of the time using helical flashlamps. In later stages of the laser development the helical lamps were replaced by linear flashlamps specifically designed for high-power laser pumping. External mirrors soon replaced the silvering of the ruby rod to allow more freedom in the design of the optical cavity to improve mode selection. The complete system was operated within a cylindrical "tube" in order to confine the pumping light.

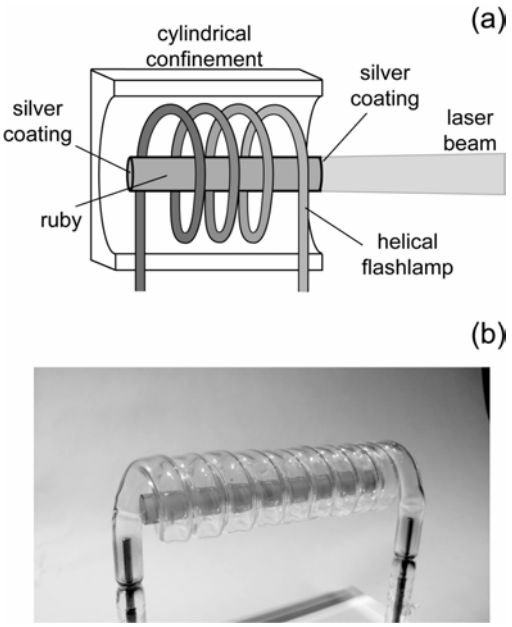


Fig. 8. Components of early ruby lasers. (a) Schematic of the laser with helical flashlamp, ruby crystal and confining "tube". (b) Photograph of helical flashlamp and ruby crystal.

November 1960: The four level laser system: The three energy levels of ruby required extremely high pumping intensities because the most heavily populated ground state had to be evacuated by more than 50 %. Quite different is the situation in a four level system, where the terminal state of the laser emission lies above the ground state by an energy ε and is thus less populated according to the Boltzmann distribution $\exp(-\varepsilon/kT)$. Going to low temperature the terminal state of emission can be strongly depopulated. Excellent candidates for laser materials are transparent crystals such as fluorites, tungstates or sapphire doped with fluorescent ions for example rare earth elements or ions of the 3rd group.

During the second half of the year two potential laser materials appeared on the market. $CaF_2:Sm^{++}$ and $CaF_2:U^{3+}$. At the end of 1960 Sorokin and Stevenson of the IBM research laboratory in Yorktown Heights held a press conference (and had a publication in an IBM journal [Sorokin 1961a; Sorokin 1961b]) reporting laser action at Helium temperature (4.2 K). Both crystals were the first four level systems requiring far less excitation intensities than with ruby. At the same time laser action in $CaF_2:Sm^{++}$ was investigated in great detail by W. Kaiser, C.G.B. Garrett and D.L. Wood [Kaiser 1961b]. During the past 50 years mainly lasers with four level systems were searched for and applied in practice (see below).

December 1960: The first cw-gas-laser: At the end of 1960 the first continuously operating laser of Ali Javan, Bill Bennett and Don Herriot began working at the Bell Laboratories. It was a gas laser. Ali Javan a native of Iran started his PhD thesis in the group of Townes at the Columbia University in New York. In this environment he was an eye witness of the development of the maser and of Townes ideas of the laser. Javan went to the Bell Laboratories with the plan to design a laser system consisting of a He-Ne gas mixture excited in a microwave discharge tube. Electronic excitation should first generate metastable He-atoms which transfer by collision their energy to Ne-atoms generating population inversion between Ne levels (see Fig. 8). The quantitative investigation of the He-Ne system with its numerous parameters turned out to be very time consuming. The gain in a tube of ~ 80 cm was quite small (approximately 1 %) requiring highly reflecting mirrors at the optimum frequency. Javan wanted to be sure that his laser would work when fully assembled. The preparation took more than one year. On Dec 12th 1960 Javan and coworkers started the laser system and – the laser worked exactly as planned [Javan 1961]. One of the present authors (W. K.) remembers a visit in Javan's laboratory the same evening. This laser

operated at the invisible infrared wavelength of 1.15 μm and the narrow beam was made visible by a small image converter showing a bright green spot of approximately 1 mm in diameter at the screen. Later measurements indicated that the beam divergence was smaller than 1 mrad with high monochromaticity of ($\Delta\nu / \nu \approx 10^{-10}$). Indeed, a most impressive new light source. HeNe lasers with other emission frequencies are discussed below.

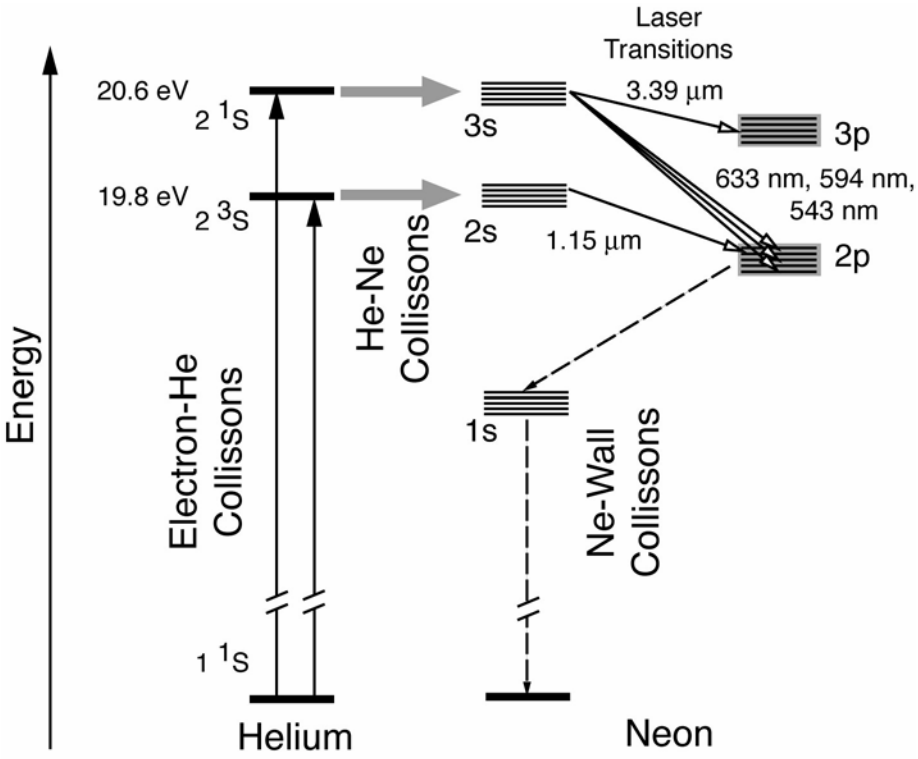


Fig. 9: Diagram of the energy levels important for the operation of a HeNe laser showing the energy uptake from the electric discharge to He levels, the transfer from meta-stable He-levels to Ne-levels and the most prominent laser transitions.

7. The Laser After 1960

The laser was well established at the end of 1960 but there were many open questions on its new properties. The novel light source promised countless applications, and exciting research in many laboratories was started immediately.

The intensity distribution of the laser beam: A more profound understanding of the optical feed-back system was required. What are the spatial properties of the laser output? The problem was studied at Bell Labs by G. Fox and T. Li [Fox 1961]. They calculated the field distribution at one of the plane end mirrors of the Fabry-Perot resonator that reproduces exactly after a series of cavity passes apart from losses due to diffraction. The stationary distributions of amplitude and phase obviously represented stable cavity modes. Shortly later this work was generalized by G. Boyd and J.P. Gordon to resonators with curved mirrors finding minimum diffraction losses for the confocal configuration [Boyd 1961]. Detailed experimental studies of the emission pattern confirmed the theoretical calculations. The discussion of internal cavity modes was extended to the propagation of coherent laser output in optical components by the work of A. Yariv and J.P. Gordon that established the new field of Gaussian Beam Optics [Yariv 1963]. G. Boyd and H. Kogelnik invented more efficient multiple-pass configurations with large mode volume in the laser material that became well-known as unstable resonators [Boyd 1962].

In these early days C.G.B. Garrett, W. Kaiser and W.L. Bond discovered the whispering modes of spheres [Garrett 1961]. Laser light was generated in ring modes, which have low losses due to high internal reflexion. The phenomenon is analoguous to acoustic ring modes, e.g. in the dome of St. Paul's cathedral in London [Rayleigh 1910].

The coherence of the laser beam: The coherence of laser light was a problem to be understood in detail. What are the fundamental differences between laser light and the spontaneous emission of thermal light sources? What kind of theory is required to describe the intensity correlations of light fields in general? Answers to such questions were urgently needed in the early days of the laser. In his work in 1963 R. Glauber gave general expressions for the coherence properties of light in terms of higher order intensity correlations that revealed the differences between laser and classical light [Glauber 1963]. At the same time the behaviour of an atom interacting with a fully quantized radiation field in a resonant cavity was studied by E.T. Jaynes and F.W. Cummings [Jaynes 1963]. A remarkable agreement of the properties of the photon field with semiclassical descriptions was found. A nonlinear theory of laser noise and coherence was published by H. Haken in 1964 [Haken 1964]. In this paper , for the first time the dramatic modification of laser light at laser threshold was analysed. Based on quantum theory it was shown that below threshold the emitted light has Gaussian photon statistics, while above threshold a dramatic change occurs: the laser emission is amplitude stabilized with small fluctuations superimposed and slow phase diffusion. In the transition region critical fluctuations, critical slowing down and symmetry breaking was predicted – typical features of a phase transition. An extension of the theory was given by H. Risken evaluating the photon distribution function [Risken 1965]. Experimental results were delivered in the following year by A.W. Smith and J.A. Armstrong [Smith 1966].]. The theoretical results were confirmed by M.O. Scully and W.E. Lamb Jr [Scully 1966; Scully 1968].

A totally different aspect is the material excitation generated by coherent laser light. The interaction of an intense laser pulse with a resonant ensemble of atoms involves new features of light-matter interaction, since a phase relationship between the induced transition dipoles of the individual ensemble members is built up. Striking effects result in particular if the optical transition is inhomogeneously broadened. Already in 1964 S.R. Hartmann and

coworkers observed a photon echo in ruby that represents the analogue to the Hahn echo in nuclear magnetic resonance [Kurnit 1964]. For the echo effect a pair of excitation pulses with a certain temporal separation is applied to the resonant material transition that is inhomogeneously broadened due to crystal disorder. The first pulse generates a phase relationship that dephases subsequently while the second pulse starts a rephasing of the individual transition dipoles. After a delay time equal to the pulse separation the rephasing gives rise to a macroscopic dipole moment with corresponding coherent emission of a third pulse i.e. the echo radiation. The echo effect allows the measurement of the dephasing time of inhomogeneously broadened optical transitions. The generalization to multiple pulse echoes found important applications in laser spectroscopy up to the present time for the investigation of relaxation phenomena in condensed matter.

A related effect again due to coherent light-matter interaction - not known from an analoguous phenomenon in the NMR of spin systems - was self-induced transparency first studied by S.L. McCall and E.L. Hahn several years later [McCall 1969]. These authors showed that a short laser pulse at resonance with a two-level transition of atoms propagates with anomalously low energy loss. The effect occurs if the pulse width is short compared to dissipative relaxation times of the absorbing material. Ideal transparency persists when the energy loss of the pulse during the rising wing of the pulse is followed by coherent induced emission radiating the same amount of energy back into the beam direction during the second half of the pulse. In other words, a reshaping of the input pulse occurs. Experimental observations of self-induced transparency were made in a ruby sample using ruby laser pulses [McCall 1969].

Novel Laser Systems: The first cw gas laser in the visible was demonstrated in 1962 by A. White and D. Rigden who succeeded to operate the HeNe laser of Javan in the red at 633 nm [White 1962] (see Fig. 8). Using multilayer dielectric mirrors for the laser cavity, the HeNe laser can be operated today at various wavelengths in the visible: in the green (543 nm), yellow (594 nm), orange (612 nm) and the red (633 nm) besides the infrared emission lines at 1153 and 3390 nm. The visible laser emission promoted the understanding of optimum laser structures and visualized the difference between coherent and incoherent light. Now, the demonstration of coherence and optical interference became an easy job. Another example is the speckle pattern of laser light from a rough surface, representing an irregular interference pattern readily seen by visual inspection. The effect was described by J.D. Rigden and E.I. Gordon who showed that the speckle size is determined by the diffraction limit of the viewing optics [Rigden 1962].

An interesting application of a cw laser built with a ring cavity was published by W.M. Macek and D.T.M. Davis [Macek 1963]. These authors demonstrated the first laser gyroscope that measures the rotation rate relative to an inertial reference frame. The counter-propagating modes in the resonator of the rotating laser are slightly frequency-shifted and differ for clockwise and counter-clockwise propagation. The beat frequency of the modes is a direct measure of the angular velocity. Such devices have found wide spread use in inertial navigation systems.

The invention of the ruby laser immediately initiated the search for other solid state laser materials in order to avoid the disadvantage of the 3-level chromium system and to achieve laser emission at new wavelengths. Particularly attractive candidates were rare earth and actinide ions (see above). Their shell electrons responsible for optical transitions are well isolated from the host material by surrounding s-electrons and display narrow emission lines. An important example were the rare earth doped glasses by E. Snitzer at Corning [Snitzer 1961]. Glass can be prepared with high quality at large dimensions and offers the possibility of high power lasers. Particular significance gained the neodymium-doped glass for laser amplification and generation of ultrashort pulses. Another important host material turned out to be yttrium aluminium garnet (YAG), successfully studied at Bell Labs by J.E. Geusic et al.

[Geusic 1964]. By late 1962 continuous laser action had been obtained in several materials, including pink ruby [Nelson 1962]. Today countless laser materials have been investigated.

CO_2 laser: The high power carbon dioxide laser was demonstrated at Bell Labs in 1964 [Patel 1964]. It is still one of the most useful ones because of its large efficiency exceeding 20% and its extremely high continuous output. The laser emits in the mid infrared with two major wavelength bands at 9.4 µm and 10.6 µm. The active laser medium is a gas mixture mainly consisting of similar amounts of carbon dioxide and nitrogen (each 10 to 20%) and a major amount of helium (> 60%). Different from the electronic transitions of earlier laser materials, an electric discharge generates population inversion of the rotational-vibrational levels of CO_2 for laser action, which explains the photon energy in the far infrared.

The CO_2 laser can be constructed for a power level of up to hundreds of kilowatts. It is also easy to Q-switch the laser (see below), yielding pulses with duration in the range 10 – 100 ns and a peak power up to gigawatts. Selecting individual rotational transitions in the rotation-vibration manifold of CO_2 by an intra-cavity grating, narrow-band emission can be achieved that is also tuneable in the range 880 to 1100 cm^{-1}. Such lasers are of interest in research applications.The available high power level and the relatively good efficiency make CO_2 lasers very useful in industrial applications for material processing. The laser has also obtained entry to surgery since the IR emission is strongly absorbed by the human tissue.

Excimer laser: An exciting concept for the generation of laser emission in the ultraviolet are small molecular dimers e.g. noblegas halides, excited by an electric discharge. These dimers only exist in excited electronic states while the ground state is repulsive and therefore not populated. Using a transition to a bound-free ground state for population inversion was suggested already in 1960 [Houtermans 1960]. The first excimer laser was built in 1970 using the xenon dimer Xe_2 and an electron gun for excitation [Basov 1970]. A big improvement was the introduction of the noble gas halides with excitation by an electric discharge [Brau 1975; Ewing 1975; Hoffman 1976]. The emission wavelengths of the various halides are positioned in the UV in the range 193 nm (ArF) to 351 nm (XeF). Pulsed operation with durations of 4 – 40 ns and pulse energies up to the Joule level make such systems well suited for material science and surgery.

Chemical laser: A new pumping method termed dissociative excitation transfer was introduced in a neon:oxygen laser in 1962 [Bennett 1962a]. In this laser, neon atoms are promoted to metastable excited states in a gas discharge. By inelastic collisions energy is transferred from the excited Ne atoms to non-binding excited states of O_2 molecules. The latter dissociate forming atomic oxygen in an excited electronic state suitable for laser action at 840 nm. Subsequently hydrogen fluoride and deuterium fluoride lasers were developed where the chemical interaction of the gas mixture provides the population inversion and laser operation in the infrared. In continuous wave operation - with huge output power levels in the megawatt range - chemical lasers found interest by the military.

Dye laser: In 1966 P.P. Sorokin and J.R. Lankard recognized that population inversion of electronic states of dye molecules could be achieved if pumped by the short and intense pulses of a Q-switched ruby laser [Sorokin 1966]. Placing the phtalocyanine dye solution pumped by a ruby laser between two mirrors they observed an intense laser beam at 755 nm. Independently F.P. Schäfer and coworkers obtained similar results for carbocyanine solutions and demonstrated tunability of a laser in the range 800 - 860 nm [Schäfer 1966]. Shortly afterwards laser action in different dyes was reported in [Stepanov 1967]. Today the dye is usually pumped with an external laser, such as a nitrogen, excimer or frequency-doubled Nd:YAG laser, while flash-lamp pumping is not efficient because of the short lifetime of the excited states of the dye molecules of a few nanoseconds or less. Compared to gases and many solid state laser media with single lines, dye solutions allow broad tuning ranges. With different dyes the complete spectral range from the blue to the near infrared is covered.

For cw operation a special geometry with a free dye jet is required to avoid optical

inhomogeneities due to the heating of the solvent and absorption losses via singlet-triplet crossing in the electronic levels of the dye. The cw laser was developed as a tuneable high-resolution light source making a bandwidth of a few megahertz readily attainable. With sophisticated stabilization schemes even much narrower (by 4 orders of magnitude) spectral emission was achieved.

Interest in dye lasers has decreased during the past two decades because of cw semiconductor lasers and the titanium sapphire laser capable of a larger tuning range and a shorter pulse duration.

Semiconductor laser: The first semiconductor lasers were demonstrated by several groups in North America at GE, IBM and Lincoln Labs. Within a few weeks in 1962 (end of September to November) four papers appeared that reported on the observation of laser emission at 840 nm from GaAs junctions at 77 K [Hall 1962; Holonyak 1962; Nathan 1962; Quist 1962].

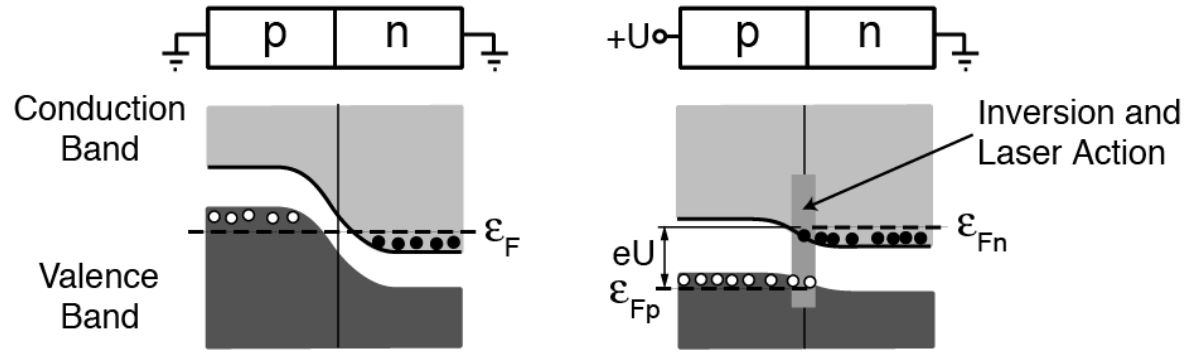


Fig. 10: Semiconductor laser: Schematics of the band structure of highly doped pn-junction. Left: equilibrium situation. Right: Forward biased system (voltage U) where the injection of electrons and holes into the boundary layer leads to stimulated emission. The structure of a modern semiconductor laser is by far more complex.

The semiconductor laser was recognized as a potential breakthrough that could revolutionize telecommunication. Already in the nineteen sixties and seventies articles were dealing with applications for optical data storage and fiber optic communication. In its early form, however, the simple p-n junction was far from any technical application. It took several years to develop double-heterostructures with reduced threshold current for lasing and for continuous operation. Further progress towards high-power lasers required highly sophisticated material processing. Important was the progress of metal-organic chemical vapor deposition (MOCVD) and molecular beam epitaxy (MBE). These two technologies allow the control of the crystal structure with atomic layer accuracy. As a result, uniform materials could be fabricated and quantum well structures with layer dimensions of the order of 10 nm.

Nowadays semiconductor lasers are available with cw operation at room temperature and emission in the wavelength range 375 - 1800 nm, while lasers in the infrared beyond 3 µm are also at hand. Diodes used in laser printers and CD or DVD players only necessitate a moderate power level. But high-power lasers are also available with output of several watts. Large array structures have been designed for high-power systems. They frequently serve for optically pumping of other lasers, e.g. fiber lasers (see below). Industrial lasers have been developed with output as high as 10 kW that are used today for cutting and welding.

Fiber laser: E. Snitzer working at the American Optical Company saw a link between his interests in glass fibers and the Nd:glass laser [Snitzer 1961]. In cooperation with C.J. Koester he achieved amplification by 5 orders of magnitude in a 1m glass fiber with a 10 µm core doped with neodymium [Koester 1964]. More generally the active gain medium of a fiber laser may use doping with rare earth material, e.g. erbium, ytterbium and dysprosium.

Advantages of fiber lasers include long active regions up to several kilometers, compact design and flexible geometry. Pumping is preferentially carried out with efficient semiconductor lasers. The lasers combine long lifetime and simple handling, well suited for numerous applications in telecommunication networks. Recent progress has achieved large mode area fibers that allow a cw power level in the 50 kW range. Applications include material processing and medicine.

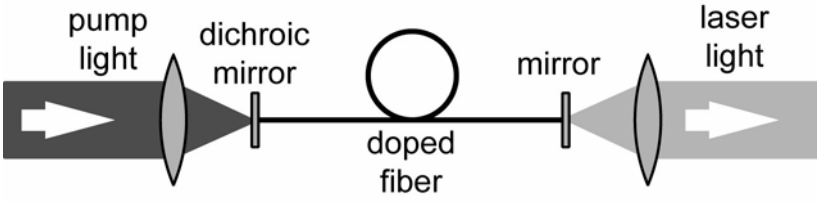


Fig. 11: Schematic view of a fiber laser formed by a doped glass fiber with dielectric mirrors on both ends, pumped by the focussed output of a semiconductor laser.

The Way to Shorter Pulses

The Q-switch: The microsecond spikes emitted by the flashlamp-pumped ruby laser over a period of a millisecond [Collins 1960] stimulated the interest for more defined pulse generation. A first solution of the problem was offered by R. Hellwarth at Hughes Laboratories [Hellwarth 1961]. He proposed Q-switched operation leading to so-called giant laser pulses. In this operation mode the laser cavity is initially blocked so that a large population inversion is generated in the gain medium during pumping. Reinstalling the feedback of the resonator mirrors leads to sudden laser action building up short emission pulses of very high intensity. The first experimental demonstration was made with a nitrobenzene Kerr cell in the cavity for electric control of the resonator feedback [McClung 1962]. Switching the Kerr cell on a sub-microsecond time scale achieved giant output pulses, several orders of magnitude more powerful than in normal operation, while the pulse duration was below 50 nanoseconds. Other switching devices were demonstrated shortly afterwards. Particularly elegant was the invention of passive Q-switching by P.P. Sorokin et al [Sorokin 1964]. In this case the cavity feedback is governed by the nonlinear response of an absorbing dye solution. A simple dye cell with a solution of metallic phtalocyanines in organic solvents performed a photo-physical bleaching at high laser intensities changing the losses of a laser cavity.

The Q-switching techniques achieve high peak power levels of simple laser oscillators in the megawatt range and opened the way for new optical effects known today as nonlinear optics (see below).

Ultrashort laser pulses: An important achievement towards the generation of much shorter - ultrashort - pulses was the locking of cavity modes of a laser. The effect was first shown by L.E. Hargrove, R.L. Fork and M.A. Pollack in 1964 [Hargrove 1964]. These authors introduced a coupling between several frequency modes of a HeNe laser by modulating the internal losses with a frequency close to a multiple of the axial mode spacing. The result was a coherent superposition of axial modes with well-defined amplitude and phase. For their first demonstration of modelocking an acousto-optic modulator was installed in the laser and pulses of 2.5 ns duration were emitted with a repetition period of 17.8 ns.

A big step forward to picosecond pulses was “self modelocking” (also termed passive modelocking) with the help of the nonlinear absorber dye also used for Q-switching. Preliminary results were reported in 1965 [Mocker 1965]. A ruby laser was studied with locking of a few cavity modes and output of a sequence of nanosecond pulses. Several

months later A.J. DeMaria and coworkers achieved better results for Nd:glass lasers [DeMaria 1966]. With the help of a long laser resonator and proper intracavity positioning of the nonlinear dye cell pulses were generated shorter than the time resolution of the detection system of 0.5 ns. Picosecond pulses below 10 ps were reported subsequently [Armstrong 1967]. By selecting a single pulse out of the pulse train of a modelocked glass laser a well-defined pulse of a few picoseconds could be obtained by D. von der Linde, O. Bernecker and W. Kaiser [von der Linde 1970]. For the measurement of these very short pulses novel autocorrelation techniques were developed [Maier 1966; Armstrong 1967; Weber 1967], using second harmonic generation (see below). For the generation of sub-picosecond pulses we mention the pioneering work of E.P. Ippen and C.V. Shank on modelocked cw dye lasers [Shank 1974]. The era of sub-100 fs pulses started several years later [Fork 1981].

Pulse compression: An efficient concept for the generation of very short and intense laser pulses is termed pulse compression. It relies on carefully selected group dispersion properties of a compression device that converts the linear frequency modulation (chirp) of the input pulse to a steeper pulse shape. The reduced pulse width is accompanied by an enlarged peak amplitude (in the ideal case). The phenomenon was invented in the microwave domain for the generation of shorter and more intense radar pulses. In the laser field the technique was proposed by J.A. Giordmaine et al. in 1968 and reported for a HeNe-Laser by M.A. Duguay and J.W. Hansen, achieving a shortening of 0.5 ns pulses by a factor of 1.8 [Duguay 1968; Duguay 1969]. E.B. Treacy discovered that the output of a modelocked Nd:glass laser was frequency modulated and could be compressed by a factor of ten to sub-picosecond pulses [Treacy 1968]. The origin of the frequency chirp was not recognized at that time. As compression apparatus he introduced a pair of identical optical gratings. Nonlinear frequency modulation by the help of the intensity-dependent refractive index of a modulation device with subsequent compression was considered in the computations of R.A. Fisher et al [Fisher 1969] and experimentally demonstrated by A. Laubereau in the same year [Laubereau 1969]. Using a compression setup consisting of an optical grating and a reflexion prism the 20 ps pulses of a Nd:glass laser were shortened by a factor of ten. The method gained great importance in the following decades for the compression of dye laser pulses in the femtosecond time domain and is widely used today in the titanium sapphire laser technology. Instead of gratings, optical prisms and so-called chirped mirrors are used to manipulate the pulse duration [Szipöcs 1994]. Pulses shorter than 4 fs can be generated with the help of the compression technique.

An important property of the compression technique is its connection to group delay, i.e. to the optical dispersion of the laser components. As a consequence it can be inverted to pulse stretching if the sign of the group delay of the optical system is reversed. The pulse lengthening converts bandwidth-limited radiation to a frequency-modulated pulse that in turn can be re-compressed again. In fact, pulses of a few ten femtoseconds duration can be stretched by a factor of up to 10^5 using a grating pair with proper imaging optics. The purpose of the stretching is the corresponding lowering of the peak amplitude. The huge reduction of peak intensity by large stretching factors allows to overcome intensity limitations in laser amplifiers. After amplification of the lengthened pulse, the recompression step is carried out with proper shortening, boosting up the pulse amplitude correspondingly. The concept is termed Chirped Pulse Amplification (CPA), a technique invented in the radar field in 1960. CPA for lasers was first demonstrated by G. Mourou and D. Strickland [Strickland 1985] and is well suited for amplifying a femtosecond pulse up to the petawatt level (10^{15} W).

Nonlinear Optics

The high intensity available in focussed laser beams paved the way for novel optical effects representing light-light interaction in gases, liquids and solids. The advent of the laser

opened a new field termed nonlinear optics.

Second harmonic generation: In 1961 it was realized that high peak amplitudes of the electric field as large as ~10^5 V/cm were accessible with ruby laser pulses, only two orders of magnitude below intra-atomic fields. It was estimated that in the beam focus a nonlinear component of the induced polarization could be generated in solid material leading to higher harmonics. Experiments were started in 1961 to generate the harmonic component at twice the laser frequency [Franken 1961]. Realizing that the harmonic polarization component is proportional to the square of the electric field and can occur only in a crystal without a center of symmetry a quartz crystal was chosen for the first investigation. In fact, P.A. Franken et al observed UV photons at 347 nm, half the ruby laser wavelength, with an efficiency of about 10^{-8}. More efficient second harmonic generation was found for potassium dihydrogen phosphate (KDP) by J.A. Giordmaine. Measuring the angular dependence of the UV emission revealed the strong effect of phasematching between the fundamental and harmonic field components. A dramatic increase of the UV light by several orders of magnitude was found for a phasematched geometry [Giordmaine 1962; Maker 1962].

Two-photon absorption: Also in 1961, W. Kaiser and Garrett at Bell Labs reported the first observation of another nonlinear optical process, two-photon absorption [Kaiser 1961a]. The effect occurs in centrosymmetric media, different from harmonic generation, and was predicted in 1931. At that time it was estimated to be too weak to be observed [Goeppert-Mayer 1931]. Kaiser and Garrett focussed a ruby laser beam (14400 cm^{-1}) into a 1-mm sample of CaF_2 doped with Eu^{2+}. The crystal is transparent at the ruby frequency 14400 cm^{-1}. Excitation of 5d level of europium ions by the two-photon transition was verified by the blue fluorescence observed around 23800 cm^{-1}. A maximum efficiency of ~ 10^{-7} was estimated for the two-photon process in satisfactory agreement with theoretical estimates. The quadratic intensity dependence of the nonlinear effect was demonstrated. Important applications of two-photon absorption were in the spectroscopy of gases and solids. In the former case, Doppler-free absorption lines could be studied by counter-propagating laser beams [Vasilenko 1970], while in the latter cases the different selection rules of the nonlinear process were exploited [Hopfield 1963]. Today two-photon fluorescence is used in microscopy (two-photon-microscopy) and has found wide applications for optical imaging techniques in biochemistry, biophysics and medicine because of the superior penetration depth in biological tissues and improved spatial resolution [Denk 1990].

Spectral hole burning: An important step in high resolution spectroscopy was the spectral hole burning first reported in 1962 [Bennett 1962b]. Normally the long term frequency stability of a gas laser is limited to ~100 MHz because of fluctuations of the laser cavity. The gain profile of the laser medium is determined by the notably wider Doppler broadening, that exceeds the natural linewidth of an individual molecule by two orders of ten. Laser action under normal conditions involves population depletion of a group of molecules which move at a velocity so that the Doppler shift leads to resonant coupling. In the wing of the gain profile, molecular groups with different velocities interact with the two counterprogating components of the laser radiation. Close to the line center, such a Doppler shift is not required and the molecular group interacts with both components of the resonant cavity mode leading to increased depletion and a resulting power dip of that mode. Bennett showed that the effect can be used to stabilize the cavity mode at the center of the Doppler gain profile. The phenomenon was worked out in detail by W. Lamb and became known as the Lamb dip [Lamb 1964]. The width of the dip is of the order of several MHz and determined by the natural linewidth of the gas transition, much narrower than the Doppler width. Correspondingly the width of the laser emission could be stabilized to a few MHz as shown, for example, by K. Shimoda and A. Javan [Shimoda 1965].

Further nonlinear Studies: A theoretical formulation of nonlinear optical processes was given by N. Bloembergen and coworkers in 1962 [Armstrong 1962]. They provided a general quantum-mechanical theory of the nonlinear susceptibility up to terms cubic in the fields. Symmetry relations for the nonlinear polarizability, energy and power relationships were derived by perturbation theory and explicit solutions for coupled amplitude relations. These authors also discussed application of the theory to second harmonic generation. In related work Bloembergen and Pershan [Bloembergen 1962] calculated in detail second harmonic generation and sum-frequency generation at surfaces, allowed by the symmetry breaking at an interface.

The theoretical results promoted further experimental work in 1962 and subsequent years. Examples include sum and difference frequency observations using different lasers, [Bass 1962] phase matching using thin quartz blocks stacked with alternating crystal orientation, and higher order second harmonic generation in centrosymmetric materials. P.D.Maker, R.W. Terhune and coworkers showed that the phase matching condition in second harmonic generation represents a novel interference phenomenon with maxima and minima of the harmonic component [Maker 1962]. D.A. Kleinman introduced valuable symmetry relations based on energy considerations [Kleinman 1962]. In centrosymmetric materials, e.g. liquids, third harmonic generation was achieved in 1965 [Maker 1965]. Continuation along this line over several years led to high harmonic generation (HHG) up to 11^{th} order in 1977 [Burnett 1977]. These authors studied the interaction of intense CO_2 laser pulses with plasmas generated from solid targets. In noble gases HHG was observed in 1988, spanning a spectral range over hundreds of eV [Ferray 1988]. Starting with sub-10 fs laser pulses, the process nowadays gives soft X-ray production and the generation of attosecond pulses in the extreme UV.

Stimulated Raman scattering: In 1962, Woodbury and Ng at Hughes Laboratories accidently observed an intense emission at 767 nm of their ruby laser [Woodbury 1962]. The process was soon identified as stimulated Raman scattering, originating from the nitrobenzene in the intracavity Kerr cell for Q-switching the laser, as recognized by G. Eckhardt , R.W. Hellwarth and collaborators [Eckhardt 1962]. The effect was predicted by M. Goeppert-Mayer much earlier in her theory of 3-photon interaction and estimated to be completely negligible using light sources available at that time [Goeppert-Mayer 1931]. In 1962 with the intensity level of the Q-switched laser, Raman scattering became competitive to the original laser output - an incredible change compared to the weak Raman lines observed in spontaneous scattering. The same group also reported stimulated Raman scattering in a number of other liquids. The field grew rapidly in subsequent years, since an extension of the spectrum of intense coherent light was very desirable. The amplification of the Raman light in the stimulated process was used recently for a new type of Raman microscopy using femtosecond pulses [Ploetz 2007].

Optical parametric oscillation: It was soon realized that according to the theory of sum-frequency generation, the nonlinear process could also occur in reverse direction. Inverting the sum process leads to interesting phenomena termed optical parametric amplification and oscillation. Detailed proposals were made in early 1962 by several authors, a few years in advance of experimental results [Akhmanov 1962; Kingston 1962; Kroll 1962]. Stimulated parametric amplification was first accomplished by C.C. Wang and C.W. Racette [Wang 1965] and independently by S.A. Akhmanov et al. [Akhmanov 1965]. As nonlinear crystals ammonium dihydrogen phosphate (ADP) and potassium dihydrogen phosphate (KDP) were used, achieving parametric gain by factors of approximately 1.2 and 2.5, respectively. Coherent parametric oscillation in a resonant cavity starting from quantum noise was first

observed by J.A. Giordmaine and R.C. Miller in the same year, demonstrating tunability of the nonlinear output in the near infrared in the range 970 – 1150 nm [Giordmaine 1965]. To this end only the temperature of the nonlinear crystal, a 5.4 mm long specimen of lithium niobate, had to be changed from 50 to 62 degrees centigrade. Optical pumping by a frequency doubled Q-switched Nd: $CaWO_4$ laser at 529 nm was used. The large frequency variability of the so-called OPO immediately attracted great interest. Practical use of the device was strongly hindered by crystal quality and optical damage. The problem was solved many years later using picosecond pump pulses in travelling wave geometries. In the femtosecond range non collinear optical parametric amplifiers are used today to cover the spectrum from the near UV to the IR [Riedle 2000].

8. Summary

At the end of this historical paper we wish to repeat some of the steps to the laser. In 1916 Einstein introduced the concept of stimulated emission. The idea to build a light amplifier was published several times during the following decade but it was Townes in 1954 who realized the first working device, an oscillator of electromagnetic radiation. It was for microwaves, the Maser. The extension to shorter wavelengths lasted only several years. In 1960 Maiman invented the flash-lamp pumped ruby laser device and demonstrated for the first time light amplification in the visible spectral range. The observation of laser action with an intense and nearly parallel beam was published first by Collins et al..

In the second part of the present paper we give an introduction of the subsequent rapid development. We have restricted ourselves to major scientific achievements and discussed the dramatic growth of new lasers and scientific fields, not even anticipated in 1960. We did not attempt to write an outline of current numerous laser applications, e. g. in medicine, material processing and information technology. The laser effects our every day life via laser based consumer electronics. It is a major element of the world wide communication network essential for the global economy.

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