Review

Extension of frequencies from maser to laser

How the laser evolved and was extended to terahertz during my research life

A personal review

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(Contributed by Jun-ichi NISHIZAWA, M.I.A.)

Abstract: The present review describes the author’s involvement and contributions to the development of the semiconductor laser and terahertz oscillators at his laboratory during the period between 1957 and now. The author cites records to show that the idea of a semiconductor laser was documented as a Japanese patent in April 1957 prior to those of G. Gould in 1957 and C.H. Townes in 1958. Terahertz oscillators of high Q values with the use of GaP were developed and applied to areas like investigations of molecular dynamics, cancer diagnosis, etc., thus extending the frontiers of science.

Keywords: laser, terahertz oscillator, crystal and structural molecular vibration, detection of structural defects in organic compounds, mapping of molecular structure by reflection and by penetration, non-destructive diagnosis by terahertz oscillator

1. Introduction

In the present article, the author intends to introduce the results of efforts of his research group, in work related to an extension of the usable frequencies to ranges for which appropriate Electronic Oscillators for the analysis and for the communication, etc. were not available at that time. This creative work had been carried out in the fourth period (after about 1955) of the author’s research life, mainly at Tohoku University and at the Semiconductor Research Institute, which belongs to Semiconductor Research Foundation.

Those efforts came from motivation given to the present author by Prof. Hidetsugu Yagi, the inventor of the Yagi antenna. He used to tell younger generation researchers, “The final goal of the communication technology should be to open a communication channel between any pair of people at any time and any place. In order to realize such a requirement, communication engineers must work hard to develop communication technology towards higher frequencies through terahertz, visible, ultraviolet, and even further toward the X-ray frequencies.”

Fortunately, the development of optical communication technology provided an enormous number of parallel communication channels, and we could eliminate such a difficult task as working toward the X-ray communication technology, but we certainly needed lasers as optical frequency generators.

It was the influence of such strong words, which the present author heard when he was a second-year student at a middle school, that drove him to work for the development of high-frequency electromagnetic radiation sources, and to be led to inventing the laser using a semiconductor as the working material.

Our research efforts began with our proposal of an “optical maser” using a semiconductor as the active material. Such a device is essentially the same in principle as devices presently called semiconductor lasers. Even though semiconductor lasers are presently available for use in a broad range of wavelengths from infrared to the visible-light range, it was impossible at that time for the industrial community to understand its feasibility and industrial importance.
The author intended to realize such a device. However, a broad range of research on semiconductor materials and on the characteristics of the semiconductor diode was needed. Such work was far beyond the capability of a university laboratory at that time, in terms of the available budget and human resources. Our efforts to persuade the industrial community to initiate this work to realize such a device turned out to be impossible at that time. Under such conditions, the only option left for us was to abandon the idea of realizing this device, and to record the concept as a patent, as described in the following section, before seeking research funds from outside.

At a much later date, the R&D capacity of my laboratory was boosted by Government support, mainly through Japan Science and Technology Agency (JST), and I continued my efforts on the same research project to realize newer frequencies by applying the same principle of the laser and to extend the frequency range covered by the electronic oscillator, towards the THz range.

2. History of proposing the semiconductor laser (Table 1)

It is generally recognized that the laser is one of the greatest inventions of the twentieth century, along with the electronic computer and semiconductor electronics, but there seems to have been some complicated struggles in the patent rights, concerning the invention of the laser, as detailed in a book by C.H. Townes, entitled “How the Laser Happened”. Recently, credit for the invention of Laser was transferred from C.H. Townes in 1958 to G. Gould in 1957. A new patent was granted to G. Gould in 1987, based on a memo dated November 1957 in a notebook used for an experiment on stimulated emission.

However, our patent (Japanese patent number 273217, registered as JAP No. 762975, dated 22nd April, 1957) seems to have predated Gould’s invention by 7 months. The application of my patent on the maser using a semiconductor as the working material is essentially the same as the semiconductor laser of today. The claim said:

“A semiconductor maser, including as a part: a semiconductor body, at least a part of the semiconductor is configured to be applied with an electromagnetic field, the semiconductor body comprising: a means for injecting free carriers at a high-energy level into a portion of the semiconductor body where the electromagnetic field is applied.”

Table 1. Chronology of laser

| Year | Month | Event |
|------|-------|-------|
| 1957 | April | J. Nishizawa applied for the patent for semiconductor optical maser (Japanese patent 273217) |
| 1957 | Nov.  | G. Gould made a document to express idea of equipment for stimulated emission with simple formula and named it as LASER (Light Amplification by Stimulated Emission of Radiation) |
| 1958 | July  | A. L. Schawlow & C. H. Townes applied for the patent for LASER |
| 1959 | April | G. Gould applied for the patent for LASER |
| 1960 | March | A. L. Schawlow & C. H. Townes got the patent right for LASER (USP2,929,922) but disappeared |
| 1987 | Nov.  | G. Gould got the patent right for LASER (USP4,704,583) |

Similar situation
J. P. Eckert & J. W. Mauchly were the first to patent a digital computing device (ENIAC). But, J. V. Atanasoff and C. Berry are now recognized as the legal inventors of the electronic digital computer (Atanasoff-Berry Computer (ABC)).
A semiconductor laser that appeared in the application form, where 1 stands for the semiconductor, 2 the container, the walls of which work as one of the reflectors composing an optical resonator; 5 and 6 are contacts for injecting electrons into the semiconductor. The temperature of the semiconductor may be controlled, for example, by circulating a coolant through a couple of funnels, 7 and 8. The region shown as 9 is the conduction band and 10 is the valence band; 12 and 13 are respectively for the electrons in the conduction band and the impurity level, and 14 for a hole in the valence band.

It may be said that the schematic diagram shown in Fig. 1 contains all of the essential components needed for a typical junction-type semiconductor laser that is used today. We believe that this is the first recorded schematic of a laser at that time. It had been filed about one year earlier than the paper by A. Schawlow and H. Townes, published in the Physical Review, which is widely understood to be the first publication of the optical maser, presently called the laser.

We believe it should be interesting to mention the motivation that gave the present author an idea to extend the maser frequency from microwaves towards the infrared and visible ranges.

3. From where and when the idea of lasers came to the author

My teacher, who had already retired, was giving a special lecture, on Prof. K. Honda’s recommendation, to a select group of about 20 students from a class of 200 of the 2nd year class in middle school in Sendai. He said that, "Josef Fraunhofer, a German scientist, was observing a star. He found that the star became very brilliant when it was observed through the corona of the Sun."

If two particles that have a recombination energy are induced to recombine by an electromagnetic wave of the same energy, the energy from the recombination is released in the form of electromagnetic waves, which not only have the same energy, but also the same direction as the triggering electromagnetic wave. As a result, the electromagnetic waves are intensified after traveling through the corona.

Therefore, a certain star that is just about visible from Earth suddenly brightens when the star is at a position from where its radiation reaches the observer's eye through the corona around the Sun. The Sun’s plasma abounds in electrons and ions, which can emit radiation of exactly the same energy as the star’s radiation. This causes an amplification of the star's image. This amplification occurs in one direction, i.e. the direction of the star's radiation. At the same time, in other directions, natural radiation is decreased because of the amplification in the direction of stimulated recombination and it causes a dark line in the spectrum," he explained. Afterwards, I continued the consideration. This phenomenon has come to be known as stimulated emission of radiation.
The resultant radiation makes the image of the star brighter in the direction of the star, and causes a black line in the solar spectrum when viewed from other directions. In 1914 this phenomenon was explained by A. Einstein.\(^6\)

Unfortunately, it was just after the First World War, and conditions were not quite conducive to academics/research. However, I was fortunate enough to be able to study the related theory from a book.\(^6\)

Just after this, Dr. Isidor Rabi who had received the Nobel Prize in 1944 visited our Tohoku University and gave an inspiring lecture about molecular beams and molecular masers. In 1957, J. F. Wittke\(^7\) introduced the solid state ruby maser, which was a very interesting maser. In this maser the amplification ceased after some time, depending on the number of electrons in the trapped level. This phenomenon itself is useful for pulse amplifiers. However, I felt that some new method is necessary to excite electrons into a higher energy level for amplification, and it was then that the idea to use a semiconductor flashed into my mind. In a semiconductor, carriers for the masers can easily be fed by electron or hole flows (Table 2) and make possible to irradiate this very high frequency region toward optical wavelength and this irradiate frequency limit toward optical frequencies and then two frequencies to use audio communication of \(2 \times 10^3 \times 3\) cycles for two communication channels for the every body of about \(60 \times 10^8\) peoples and, at the same time, many applications for spectrum analysis, also.

At the same time, an optical transmission guide was also proposed for communication, which the author also extended toward the use of total reflection\(^8,9\) at the reflecting point near the surface to improve the transmission characteristics of the glass fiber. This was the world’s first low loss transmission line which used to be called multimode and was followed by C. K. Kao\(^10\) and by others.\(^11-13\) Kao’s transmission line used to be called single mode, which is narrow band, and has been so much afraid for limited transmission of information.

That was the trigger that helped the author to envision the concept of amplification by the stimulated emission of radiation. Since it had been shown to work in the microwave frequency range, it should also work at optical frequencies. That means that masers working in the optical frequency range should be possible.

As a working material for masers at optical frequency, the author chose semiconductors.\(^14-19\) Even though there were no prototypes of a maser in which a semiconductor was used as the active medium, we considered that a conduction electron injected into a semiconductor can have sufficiently high energy to emit infrared or visible photons by recombination with a positive hole having sufficient mobility to fill the vacancies of electrons, and that it should be possible to develop optical masers (later became to be called as laser) based on such a principle.\(^14-19\)

The author would like to add here that the purpose of reviewing this history is not just to advertise our achievements, but also to discuss the circumstances in which research and development activities can be more creative and competitive, thereby contributing to future progress, rather than trying to compete just by moving into an already existing field of international competition, or merely learning the present status of the field from papers already published in overseas technical journals.

The advancements of laser technology should have been triggered much earlier during the initial period of laser technology, itself, if such an idea as the one we proposed had been understood and supported by those laboratories, whose activities were triggered by a demonstration of the double hetero-junction semiconductor laser at room temperature, more than ten years after our patent application. Unfortunately, the author could not be supported financially in the year of 1960 in Japan. However, after the fevered development of semiconductor lasers, the author fortunately found the chance to challenge the realization of terahertz oscillation in a semiconductor based on the same principle.

4. Realization of terahertz oscillation

It may be appropriate to introduce here the results of my efforts to increase the available frequencies of electromagnetic oscillators. This was done by the use of stimulated Raman scattering. In the early stage of an investigation of nonlinear optics by using lasers, it was found that stimulated Raman radiation can be built up in an optical resonator. Not only the Raman Stokes lines, but also anti-Stokes radiation was found to be present. Of course, there are a large number of reports on how to obtain such frequencies using the spectrometry of a flame or gas discharge and the corresponding absorption spectra. These results expanded the field of molecular research, pioneered by Prof. S. Mizushima,\(^20\) followed by Profs. H.
Yoshinaga, Akiyoshi Mitsuishi and Eiji Hirota et al. in Japan.

Two approaches were followed in my laboratory. In one approach we used a tunnett diode, which is a tunnel injection transit-time-negative resistance diode invented by myself in 1958, and attained an oscillation frequency very close to 1 THz. This was very simple and useful for experiments that did not demand high accuracy. In another approach, we applied the same mechanism of optical maser principle, in other words, frequency conversion. First, laser light was generated by the Raman effect or by other means. Then, a higher frequency \( \omega_0 + n\omega_r \) was generated by an anti-Stokes Raman Effect. Based on recently obtained results from research on the Raman effect in intermetallic compounds, my staff chose GaP. After maser resonance at \( \omega_0 + \omega_r \), the oscillating frequency was led out of the resonator, and then led into another resonator of frequency \( \omega_0 \). Due to the Stokes-Raman effect, the laser output had a frequency of \( \omega_0 \), which could be controlled over some frequency range. Under some conditions, if factors such as the

| Year | Event |
|------|-------|
| 1957 | J. Nishizawa Proposal of semiconductor optical maser (now laser) (Japanese Patent) |
| 1957 | G. Gould First naming as LASER (in notebook) |
| 1958 | J. Nishizawa Proposal of TUNNETT diode |
| 1958 | A. Schawlow & C. Townes Proposal of gas laser |
| 1960 | T. Maiman Realization of ruby laser |
| 1961 | A. Javan Realization of gas laser |
| 1962 | M. Naithan, R. Hall, T. Quist & J. Pankove Realization of semiconductor laser |
| 1963 April | J. Nishizawa Proposal of THz-wave generation via molecular and lattice vibrations |
| May | R. Loudon |
| 1964 | J. Nishizawa & I. Sasaki Proposal of optical fiber communication and focusing optical fiber (Graded Index) |
| 1965 | J. Nishizawa Proposal of terahertz wave generation via molecular and lattice vibrations together with tunneling |
| 1966 | K. Kuo Estimation of low absorption loss optical fiber |
| 1968 | J. Nishizawa Realization of TUNNETT diode |
| 1969 | R. Pantell Observation of frequency shift via lattice vibrations |
| 1973 | P. Solokin, J. Wynne & J. Lankard Four wave parametric effect in alkaline metals |
| 1979 | J. Nishizawa Proposal of Ideal Static Induction Transistor (Ballistic SIT) |
| 1979 | J. Nishizawa & K. Suto Realization of semiconductor Raman laser with lattice vibration |
| 1980 | J. Nishizawa Realization of TUNNETT diode oscillating at 0.34 THz |
| 1983 | J. Nishizawa & K. Suto Difference frequency wave generation via semiconductor Raman laser (12.1 THz) |
| 1996 | K. Kawase, M. Sato, T. Taniuchi & H. Ito THz wave generation via LiNbO₃ by Nishizawa-Pantell method |
| 1999 | J. Nishizawa, P. Plotka Realization of Ballistic SIT (scattering free SIT) |
| 2000 | K. Kawase, J. Shikata, K. Imai & H. Ito THz wave parametric generation by injection seeding |
| 2000 | J. Nishizawa Proposal of application of THz-wave to diagnosis and medical treatment of cancer |
| 2006 | J. Nishizawa & T. Tanabe CW THz-wave generation from GaP with using semiconductor laser diodes |
field intensity, angle, distance etc. are changed, the wavelength of the emergent beam can be changed. By using this property, the change in the characteristic spectrum of organic compounds can be measured. The characteristics of more than 200 compounds were measured and published in database. A large molecule will effectively have a larger mass. Component atoms in a molecule are held together by binding forces that can be represented by springs. The spring effectively becomes stronger when the binding force gets stronger, which means that lighter molecules will resonate at a relatively higher frequency.

Electromagnetic waves have been widely applied, as shown in Fig. 2, including the special range for communication, which was prepared in the author’s laboratory. As a result of our work, if we are asked to, we can realize any frequencies up to 100 THz. Finally, we can say that we have succeeded in bridging the terahertz gap effectively from both ends; radio waves and optical waves. Thus, we can feed any frequency as we like, realizing the thesis by Prof. Yagi; some monopolizing peculiar frequency to any of six billion people on the earth. Now, it became possible to connect one wherever and whenever he is.

5. Type of oscillators

While we worked on realizing oscillators, and succeeded in the laboratory, many researchers in this field, most of them from Japan, had been using some form of infrared radiation source. Prof. Hirota used higher harmonics. In contrast, only our group...
started to realize oscillators. The general principle involved frequency conversion using lattice vibrations, like Raman. Following the former result, we chose GaP, and using the anti-Stokes Raman effect we succeeded to generate oscillations of 12.1 THz and higher. Subsequently, we tried to induce a reverse effect, namely the Stokes Raman effect, and obtained 12.1 THz in 1983, following up-conversion based on an anti-Stokes Raman effect in 1979.

On the whole, today, simple oscillators can be realized using GaP and other materials as Raman-effect media, and also by using LiNbO3. The latter is an example of applying of microscopic dielectrics. In my laboratory, continuously oscillating semiconductor lasers were already showing exciting results, even at room temperature. A Q value as high as $10^6$ at about 3 THz without cooling was attained already. If we introduce cooling, we are sure to obtain a higher value for Q. (Fig. 2)

As a result of this development, the GaP Raman laser was shown to have very high Q values for spectrum analyses. To demonstrate the potential impact of the very high precision, molecular structural defects were measured. Figures 3 and 4 show absorption spectra of different crystalline polymorphism of sulfanilamide (alpha-, beta- and gamma-forms). There is nearly no difference in the μm wavelength range of IR, whereas in the THz range the difference becomes significantly large. Figure 4 shows the characteristics of 2 types of thalidomides, which are optical polymorphs. With amino acids these show characteristics of serine, depending on the type of the partner thalidomide. The glucose content was measured before and after γ-ray irradiation. The THz transmittance is shown in Fig. 5. The effect is visible in the shift of the black curve relative to the red curve. The peak shift is towards lower frequency, which corresponds to the effectively lighter weight atoms.

This is the first report about a successful diagnosis of defects in organic molecules. We are trying to improve the accuracy of the measurement, and are trying to obtain more precise information about crystalline defects inside an organic compound. This shows that our equipment can produce the best results in the world. The excitement has not ended there. Academician Kanno forecasted that "Cancer seems to be induced by molecular structural defects in DNA" components (Fig. 6). And in future, these are expected to be measurable, even in a solution. I believe that our equipment will be able to contribute to cancer research. We have already published measurement results in many science journals, which can be looked up on the Internet.

The impact of applications of such equipment...
for checking organic molecules is profound. In the case of cancer, it is possible to detect changes in a molecule, which will provide a means of obtaining concrete evidence of cancer. The equipment can also give us additional information about the cancer-type, structure of concerned molecules, etc., which can be very important information about the cancer. In a similar manner, we can estimate the type of defect in a specimen or some drug, which has molecular defects, perhaps due to manufacturing processes. Such information will be sufficient to identify the country and the manufacturer. Molecular vibration shows sharp absorption, and heats up only the molecules that resonate. This may open up the possibility of applying this heating up technique to cancer treatment; the resonance heating will heat up only the cancer cells, and will not affect other cells.

A major problem in the measurement is the fact that too many peaks are observed, and therefore some mathematical method is necessary to analyze the spectral pattern. It is like locating a needle in a haystack. At this time, a meaningful measurement of reflection spectra is very important. Moreover, at such frequencies, electromagnetic waves penetrate
into the substance and some of the electromagnetic waves are reflected by the internal structure of the substance. These waves also have important information about the optical properties of the substances just under the surface, and therefore it can be a very effective non-invasive method for medical examination. THz spectroscopy is basically a low-energy physical test.

6. Detection of some bacterium and virus

A bacterium and a virus are both made of interconnected molecules. The set of connected atoms in the molecules has a resonance frequency. By measuring this resonance frequency, we can obtain insight about the structure of the molecule. Conversely, one can infer the type of molecules or chained atoms that is expected in the specimen, as has been shown for a slice of cancerous liver in Fig. 7 (left). Figure 7 (right) shows a map of absorption at two frequencies: 1.56 and 3.70 THz, respectively. These seem to be mostly due to resonance absorption caused by the principal molecular structures. Usually, in solids molecules are densely packed. This makes the identification of defects very difficult, which means that dilution technology is very important both in the physical and computational context. At any rate, the map shown is sufficient to diagnose the presence (or absence) of cancer in a very short time; the corresponding diagnosis by microscope usually takes four days.

THz reflection spectroscopy and mapping have wide applicability. The attenuated total reflection (ATR) spectroscopy can also be used for measurements of a small amount of species with large absorption for the surface analysis of thin films. We are working on developing diagnosis procedures for diabetes that can be used by common people within legal means.

7. Applications

A molecule is composed of two or more atoms. The charged atoms form a dipole at the junction of two atoms. The degree of charging an atom in a molecule mainly depends on the nature of the chemical combination, and is usually unique for the pair of combining atoms. The same degree of charging is scarcely seen for 2 different types of pairs of atoms, and rarely for 3 different types of pairs of atoms. This relationship can be represented as a matrix. The charging is also influenced by the energy levels of the second and third nearest neighbors. The effect of γ-ray irradiation for α-D glucose has already been
reported,\textsuperscript{37,38} and accounted for by the deformation of an atom after radiation. These situational charges occur even in organic compounds. Theoretical analysis is beginning to show interesting effects in the combination of atoms in a molecule. In practical situations, an actual measurement can be conveniently preceded by measurements of transparency and/or reflection using terahertz waves.

The possibility of measuring the concentration of a specified molecular part in a material by examining the spectrum of the reflected and/or transmitted terahertz wave has far-reaching consequences.

One of the already successfully demonstrated examples in my laboratory is the detection of dangerous chemical compounds for medical purposes. Prof. T. Sasaki and I have already succeeded to estimate the relative concentrations based on the idea of polymorphism of thalidomide from spectral measurements.\textsuperscript{39} For Racemic compounds, the difference of the spectrum in the terahertz range is significant, though there is no noticeable difference in the infrared range. Checking all tablets manufactured by the medical industry presently seems to be a practical proposition.

In the same manner, the author is planning to apply the same measurement technique for some bacterium and virus, because bacterium and virus are also organic compounds. The binding of atoms in a compound is easily understood in terms of a spring, like connection between pairs of atoms. This concept gives the compound a characteristic resonant frequency that can be measured with the same technique. Already, the author’s group has published a part of the results, which will be useful to ascertain the existence of cancer in a suspected part before an operation, in less than ten minutes by looking for the specified characteristic frequency in the measured absorption or reflected spectrum. The author’s group has also already published a part of a result about the process of hydration on a surface, as an example of atomic reactions in layered structures using specified electrode structures, using a specified structured electrode.

Anyhow, this is a proposal of new method based on the great precision of the specified frequencies in the terahertz range, which reaches a Q-value as high as 10\textsuperscript{6}, the best that has ever been achieved.

Another very interesting result is the transition between a deep level and a band. It was already forecasted in the original patent that deep levels are caused by doped impurities. Recently, in Germany and Russia, the same situations were realized even in Si or Ge, at liquid-helium temperatures. Transitions between an edge of the band and such deep levels were reported to produce a terahertz oscillation. Prof. M. Kimura reproduced the same phenom-
enon. We found the same effect, not only at very low temperatures, but also at room temperatures. It will be reported very soon.

Acknowledgments

Frequency generation is of course of prime importance in the field of communications. The author succeeded to extend the range of generated frequencies from a low frequency of about a few hundred cycles to a high frequency of several tens of terahertz using semiconductors. The most significant idea in the generation of waves was that of light and microwave amplification by the stimulated emission of radiation, which was reported by Josef Fraunhofer as a result of his lonely observation of stars in 1814, and was explained theoretically by Albert Einstein.6)

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Profile

Jun-ichi Nishizawa, a member of the Japan Academy, was born on 12 September, 1926. He entered Tohoku University, Faculty of Engineering, Electrical Engineering Department as a student just after the departure of Prof. H. Yagi to Osaka University. After experiencing some difficulties, he was selected as a special student to conduct research; his theme was the application of electron physics to engineering. Since he was poor, he had to start his semiconductor experiments by first melting semiconductor powders using a vacuum-tube oscillator constructed by himself. After many experiments he ascertained the now accepted theory, and developed junction theory. He introduced the structures of a pin diode and a npn transistor, achieved an innovative inhomogeneous mesh structure and succeeded to fabricate several very nice devices like diodes, triodes and thyristors. Subsequently, he jumped into optical communications after inventing the semiconductor optical maser in 1957. However, because of a lack of financial support he restarted his research on devices based on the same principle from 1962. This can be called the terahertz laser, which he successfully developed in 1983 with Assistant Professor K. Suto. For these achievements he received the Academy Prize from Japan Academy in 1974, the Jack A. Morton Award in 1983 and Edison Medal from IEEE (2000). The IEEE also created the Nishizawa Medal in 2002, and Humboldt University presented an Honorary Doctor degree to him. He is a member of the Academies of Russia, Poland and Japan. He also successively served as the Presidents of Tohoku University, Iwate Prefectural University and Tokyo Metropolitan University, and still remains Emeritus President of Iwate Prefectural University and Tokyo Metropolitan University. He is currently Adviser to the Sophia School Corporation and Professor (Special Appointment) of Sophia University. In 1989 he was awarded the Order of Cultural Merit by the Emperor.