

Masers and Lasers

MOLECULAR AMPLIFICATION

AND OSCILLATION

BY STIMULATED EMISSION



Gordon Troup

*Lecturer in Physics, Monash University,
Victoria, Australia*

LONDON METHUEN & CO LTD
NEW YORK JOHN WILEY & SONS INC.

First published October 22nd, 1959

Second edition 1963

Reprinted 1963

© 1959 and 1963 Gordon John Fordyce Troup

Printed in Great Britain

by Butler & Tanner Ltd Frome and London

2.2

Cat. No. (Methuen) 2/4072/11

Contents

Preface to 1st Edition	<i>page</i>	vii
Preface to 2nd Edition		viii
1 Introduction		1
2 Stimulated and Spontaneous Emission of Radiation		9
2.1 <i>The Approach from Thermodynamics</i>		9
2.2 <i>The Approach from Quantum Mechanics</i>		16
3 Amplification, and Factors Affecting the Process		34
3.1 <i>The Possibility of Amplification</i>		34
3.2 <i>Factors Affecting the Amplification Process</i>		36
3.3 <i>A Specimen Problem: Paramagnetic Molecules</i>		45
4 Excitation Methods		51
5 Microwave Amplifier Systems		64
5.1 <i>The Line Amplifier – General Considerations</i>		64
5.2 <i>Slow-Wave Structures</i>		71
5.3 <i>Noise Temperature of the Maser Line Amplifier</i>		75
5.4 <i>Resonant Cavity Maser Amplifier</i>		80
5.5 <i>Noise Temperature of the Resonant Cavity Maser</i>		87
6 The Microwave Maser Oscillator		94
6.1 <i>Introduction</i>		94
6.2 <i>Conditions for Oscillation: Emitted Power</i>		97
6.3 <i>The Frequency of Oscillation</i>		100

Contents

7	Infra-Red and Optical Masers	<i>page</i> 110
7.1	<i>General Considerations</i>	110
7.2	<i>The Parallel Plate Resonator – Elementary Treatment</i>	113
7.3	<i>Optical Waveguide</i>	117
8	Experimental Work	120
8.1	<i>Early Gaseous Devices: Amplification and Oscillation</i>	120
8.2	<i>Gaseous Devices: Noise Temperature</i>	127
8.3	<i>Early Solid-State Devices: Amplification and Oscillation</i>	132
8.4	<i>Solid-State Devices: Noise Temperature</i>	138
8.5	<i>A Practical Microwave Line Amplifier</i>	142
8.6	<i>A Practical Microwave Cavity Amplifier</i>	145
8.7	<i>Infra-Red and Optical Masers</i>	146
9	Applications and Future Work	154
9.1	<i>Applications</i>	154
9.2	<i>Future Work: Materials</i>	159
9.3	<i>Techniques</i>	161
9.4	<i>Conclusion</i>	162
	Acknowledgements	164
	Bibliography	165
	References	167
	Appendix I – Quantum Mechanical Treatment of the Harmonic Oscillator	174
	Appendix II – Introduction to the Theory of Paramagnetic Ions in a Crystal Field and a Steady Magnetic Field	181
	Index	189

CHAPTER 1

Introduction

The word MASER as originally coined stood for 'Microwave amplification by stimulated emission of radiation'; however, devices based on the stimulated emission principle have been developed in the radiofrequency, ultra-high-frequency, and optical regions of the electromagnetic spectrum. Since these devices use molecular processes to generate or amplify electromagnetic radiation, the initial 'm' in maser has come to stand for 'molecular' so that one can talk about 'UHF' or 'optical' masers. (The obvious abbreviations 'Laser', 'Iraser', and 'Raser' are also current.) The historical order of development will be followed here. Initially, the simpler microwave approach will be emphasized; subsequently, the discussion will be widened to include the optical region.

In conventional microwave amplifiers and oscillators such as the Klystron, an alternating electromagnetic field interacts with elementary particles, electrons, by virtue of their charge. The electrons are given a steady (d.c.) kinetic energy, part of which is ultimately converted into electromagnetic field energy. For this to occur, the electrons must interact with the alternating electric field in the proper phase: that is, the electrons must be retarded by this field.

It is known that electromagnetic waves can interact with other elementary particles by virtue of changes in the

internal energy of the particles. These particles may be uncharged – atoms and molecules – or charged – ions. In the following text, ‘molecule’ will be used to include the cases of atom and ion when statements made may apply to all these types of particle. A molecule consists of an assembly of electrons and atomic nuclei. These can assume only the motions and orientations which yield a discrete set of energies: that is, energy states or levels as opposed to an energy continuum. The internal energy of the system is quantized. A molecule can interact with electromagnetic radiation by making a transition from one energy level to another. The change will be from one value of internal energy, corresponding to particular motions and orientations of the electrons and nuclei, to another value of internal energy, involving a difference in either motions or orientations, or in both. If the final internal energy of the molecule exceeds the initial energy, the energy difference must be supplied from the electromagnetic radiation field, and the process is one of absorption of radiation. Conversely, when energy is given to the radiation field, the process is one of emission of radiation, and the final energy state of the molecule must have less energy than the initial state.

The radiation field is also quantized in energy, so that it can exchange only discrete amounts of energy with interacting molecules. If the energy of a molecule in an upper energy state, m , is W_m and in a lower energy state, n , is W_n ($W_m > W_n$), then the frequency f_{mn} of the radiation involved in a transition upward ($n \rightarrow m$) or downward ($m \rightarrow n$) of the molecule between states m and n is given by the Bohr frequency condition,

$$W_m - W_n = hf_{mn} \quad (1.1)$$

Where h is Planck’s constant (6.6×10^{-34} joule sec.). The

Introduction

radiation of frequency f_{mn} is quantized into 'packets' or photons of energy hf_{mn} , and the energy of radiation of frequency f_{mn} can change only by this amount when interaction with a single molecule occurs. The molecule absorbs a photon and increases its internal energy, or decreases its internal energy by emitting a photon. However, the treatment of electromagnetic radiation in terms of continuously varying electric and magnetic fields is justified at microwave frequencies, because of the minute size of an energy quantum (6.6×10^{-24} joule for $f = 10,000$ Mc/s), and hence the enormous number of quanta present in practical cases (1.5×10^{17} photons per sec. in radiation at 10,000 Mc/s carrying 10^{-6} watts of power). Provided that the wavelength of the radiation is very much greater than the size of the molecule, and there are relatively many quanta present, the field treatment also holds good for the optical region.

Not all transitions between energy states of a molecule are possible. Theory predicts and experiment confirms that only certain transitions can occur, and 'selection rules' can be formulated which enable permitted transitions between various energy levels to be determined.

It is found that molecular transitions occur by interaction with either the electric or magnetic component of the radiation field, depending upon whether the change of molecular internal energy is primarily electric or magnetic in character. Thus an effective oscillating electric or magnetic moment is associated with each radiative transition; this moment is usually dipolar, but may be of higher order.

In a typical optical transition, the energy difference $W_m - W_n$ between the levels is of the order of 1 electron-volt (1.6×10^{-19} joule); such energy differences exist between the successive electron shells open to the outermost

Masers

electrons of atoms or ions. However, $W_m - W_n$ must be very much smaller for the frequency to lie in the microwave range. In atoms and ions, this small energy difference can exist in the fine structure of electronic energy levels (due to electron spin and relativistic effects) and in the hyperfine structure of these levels (due to nuclear spin) (1). In molecules, energy differences between vibrational energy levels or rotational levels are often of the right magnitude. Atomic, ionic and molecular energy levels split into more numerous components under the influence of an applied steady (d.c.) electric or magnetic field (Stark effect and Zeeman effect (1)). Energy differences between such Stark or Zeeman components of energy levels may also be sufficiently small for frequencies associated with permitted transitions to lie in the microwave range. Finally, paramagnetic ions also present the possibility of suitably spaced energy levels. These particles possess permanent steady magnetic moments, which will attempt to align themselves in the direction of an applied d.c. magnetic field. The energy of these elementary magnets in the applied field depends upon the magnitude of the field, and upon the angle between the vector of the field and the vector of the magnetic moment. Once again, only certain energy values are allowed, corresponding to certain angles between the vectors. Transitions between particular energy levels (orientations of the magnetic moment relative to the applied field) are permitted, and by choosing a suitable paramagnetic substance and value of d.c. magnetic field, transition frequencies in the microwave region can be obtained. (See Appendix II.)

For simplicity, but without loss of generality, we now consider a molecule having an upper energy state and a ground state between which transitions are permitted. It is found that, when the molecule is in the upper state, there

is a definite probability that after a period of time the molecule will revert to the ground state with the emission of radiation. This probability has two components, one of which is constant and the other variable. The constant component may be likened to the probability for the decay of a radioactive substance: it depends on the molecule and the particular transition involved, and is independent of outside circumstances (with an exception to be discussed later). The variable component of the probability for the downward (emissive) transition to occur is found to depend linearly on the energy density (say joule metre⁻³ or photons metre⁻³) of radiation at the transition frequency incident on the molecule. The presence of radiation at the transition frequency increases the probability for the emission of radiation by the molecule in the upper state.

The process whereby excited molecules may revert to the ground state by emitting radiation even when no radiation is present, and which gives rise to the constant component of the transition probability, is known as the spontaneous emission of radiation. In the presence of radiation at the transition frequency, the molecules may still revert to the ground state spontaneously: the emitted radiation then bears no definite phase relationship to the incident radiation. However, incident radiation at the transition frequency increases the probability of transition as described above, and induces the molecules to emit radiation which will bear a definite phase relationship to the incident radiation. This process is known as the induced or stimulated emission of radiation. In the microwave region, this process greatly predominates over spontaneous emission at practical field strengths (energy densities) of radiation at the transition frequency.

Thus an assembly of molecules in the upper of the two

energy states is a potential source of electromagnetic energy gain. The matter is complicated by the fact that molecules in the lower of the two energy states are found to absorb radiation at the transition frequency with a probability equal to that for stimulated emission by molecules in the upper energy state. Hence, in order to obtain a gain of energy from an assembly of molecules, we must ensure that there is a large excess of molecules in the upper energy state, and maintain this excess by some means. It is unnecessary that the molecules should first absorb radiation at the transition frequency in order to reach the upper state: the requisite energy may be obtained from electron bombardment, or optical illumination if more than two states are involved; and other excitation methods are possible.

We see that, in order to make use of the coupling of a radiation field to the internal energy of molecules to obtain amplification, we must satisfy conditions somewhat analogous to those fulfilled in a conventional electronic amplifier, and mentioned at the very beginning. We must have a source of energy for the molecules, to put them in the excited state (analogous to the supply of d.c. kinetic energy to the electrons); and the molecules must be in the excited state before energy can be delivered to the radiation field (corresponding to the electrons being in a retarding electric field before energy can be surrendered to it). We shall see that, as well as amplification, oscillations sustained by radiation from emitting molecules ('self-induced' emission, so to speak) are also possible.

A short historical review of the literature and reported developments is in order. Einstein (2) postulated the existence of stimulated emission in 1917 from considerations of thermal equilibrium. The maser was proposed by Townes (3) as a source of millimetre waves in 1951; the first

Introduction

paper in the open literature on the subject was by Weber (4) in 1953. By 1954, a molecular amplifier and oscillator had been constructed, using a transition frequency from Ammonia, by Gordon, Zeiger and Townes (5, 6, 7). In 1956, Bloembergen (9) indicated that paramagnetic ions with three or more energy levels could be used to make a continuously operating solid-state maser; the excitation method (suggested independently by Basov and Prokhorov (10)) involved using three levels, and was capable of ensuring a continuous excess of upper-state molecules. This was followed up experimentally by the work of Scovil, Feher and Seidel (11) who first made and operated an amplifier based on Bloembergen's proposals. Subsequently, many masers of the Bloembergen type were reported. Much theoretical work on microwave masers was published in 1957-8, and measurements reported in late 1957 and early 1958 confirmed the theoretical predictions as to bandwidth and low-noise performance of maser amplifiers. From then on, masers could be considered established, and were operated in radars (12) and radiotelescopes (13, 14). Late 1958 saw Townes' and Schawlow's paper on maser techniques in the optical region (15); in 1960 the first demonstration of optical stimulated emission in a solid was reported by Maiman (16), and the first coherent optical oscillator was described by Collins *et al.* (17).

In this dissertation, we shall first consider the theory of stimulated and spontaneous emission in some detail (spontaneous emission is important because it is a source of noise in the maser). After a discussion of conditions under which amplification can be achieved, and of natural processes affecting amplification, we shall deal with the methods of obtaining sufficient molecules in the excited state. The theory of microwave and optical stimulated emission amplifiers and oscillators will be presented,

Masers

followed by a description of some experimental devices reported in the literature. An assessment of future developments in the field, and of possible and actual applications of masers, will conclude the survey.

CHAPTER 2

Stimulated and Spontaneous Emission of Radiation

2.1 The Approach from Thermodynamics

The first treatment of stimulated and spontaneous emission of radiation by molecules was given by Einstein (2), who used thermodynamical arguments. His treatment is well worth considering quite fully, both because of its fundamental nature and because it conveniently introduces many topics necessary to the discussion of amplification by stimulated emission.

Einstein considers an assembly of molecules in thermal equilibrium with a surrounding enclosure at absolute temperature T . The molecules have energy levels between which radiative transitions occur. Since the whole system is in thermal equilibrium, the radiation present in the enclosure must be 'complete' or 'black body' radiation, and each part of the system must absorb as much radiation as it emits.

Black body radiation is characterized by the fact that its energy density (say u joule metre⁻³) depends only on the absolute temperature of the associated system. The total energy of radiation is distributed over a wide frequency range and the distribution of the energy with frequency is well established theoretically and experimentally. If u_ν is the energy density of radiation per unit frequency range at

the frequency f (say u_f joule metre⁻³ cycle⁻¹ sec.), such that

$$\int_0^{\infty} u_f df = u$$

then the value of u_f for any frequency f and temperature T is given by the Planck radiation formula (18)

$$u_f df = \frac{8\pi f^2}{c^3} \left\{ \frac{hf}{\exp(hf/kT) - 1} + \frac{hf}{2} \right\} df \quad (2.1)$$

where h is Planck's constant as previously

k is Boltzmann's constant (1.37×10^{-23} joule deg.⁻¹ mol.⁻¹)

and c is the velocity of light (2.997×10^8 metre sec.⁻¹).

The quantity $(8\pi f^2/c^3) df$ is the number of wave types, or modes of oscillation, per unit volume, of the radiation in the enclosure (large compared with the wavelength at frequency f) in the frequency range f to $(f + df)$. The quantity $\{(hf/2) + hf[\exp(hf/kT) - 1]^{-1}\}$ is the average energy in an oscillation mode at frequency f : $(hf/2)$ is the zero-point energy, usually omitted, since the inclusion makes the total integrated energy-density infinite, because the number of modes is infinite. Empirically though, we only look at a restricted frequency range, so that the total energy-density in the range will be finite. At microwave frequencies, where $hf \ll kT$ even for quite low values of T , the average energy per mode reduces to the familiar kT ; at optical frequencies, where $hf \gg kT$ for ordinary values of T , the average energy per mode becomes $(hf/2)$. Since we cannot detect half-quanta, and the thermal radiation 'noise' in an optical mode does exist, we conclude that the 'thermal noise' in an optical mode must be hf . (A rigorous demonstration would use fluctuation theory.) Had we omitted the zero-point energy, the radiation noise in an optical mode would have tended to zero for high quantum

Stimulated and Spontaneous Emission of Radiation

energies; however, at microwave frequencies where $hf \ll kT$, the zero-point energy can be omitted quite safely.

Einstein postulates that the radiation emitted and absorbed by the molecules in making transitions between energy states must also obey the Planck radiation law, equation (2.1), since the molecules are in equilibrium with their surroundings. Suppose the molecules have upper states m , energies W_m , and lower states n , energies W_n . Then for equilibrium the total probability for transitions $m \rightarrow n$ with the emission of a quantum of radiation $hf_{mn} = W_m - W_n$ must equal the total probability for a transition $n \rightarrow m$ with the absorption of a quantum of radiation. The overall probabilities of the transitions depend upon the probabilities of the molecules being in the states m or n . These probabilities, P_m for molecules in state m , P_n for molecules in state n , are given by

$$\begin{aligned}P_m &= C \exp(-W_m/kT) \\P_n &= C \exp(-W_n/kT)\end{aligned}\tag{2.2}$$

where C depends on the temperature. We have neglected statistical weighting factors for the different states for simplicity. These relations, which apply in thermal equilibrium at temperature T , are well established in thermodynamics, and we will have occasion to use them often.

Einstein then proceeds to put forward forms for the probabilities of downward and upward transitions and to show that these forms allow the Planck radiation formula to be satisfied if certain relationships exist between the probabilities. He first considers a molecule in an upper energy state m in the absence of radiation. It is assumed that in an interval of time dt , the probability of the downward transition $m \rightarrow n$ is

$$dP_{mn} = A_{mn} dt\tag{2.3}$$

where A_{mn} is a constant, the spontaneous emission coefficient. This spontaneous emission of radiation is likened to the process of radioactive decay, where the probability of a nuclear transition is independent of physical conditions.

Einstein next considers molecules in the presence of radiation of energy density per unit frequency range u_f at the transition frequencies. He assumes that the probabilities of upward and downward transitions depend on u_f , and writes these probabilities as

$$\begin{aligned} dP_{nm} &= u_f B_{nm} dt & \text{for transitions } n \rightarrow m \\ dP_{mn} &= u_f B_{mn} dt & \text{for transitions } m \rightarrow n \end{aligned} \quad (2.4)$$

where B_{nm} , B_{mn} are constants, the coefficients of absorption and of induced emission.

The condition for equilibrium can now be written. Since each part of the system in thermal equilibrium must absorb as much radiation as it emits, we must have the total probability for upward transitions equal to the total probability for downward transitions. Hence from (2.2), (2.3), (2.4),

$$u_f B_{nm} \cdot \exp(-W_n/kT) = (u_f B_{mn} + A_{mn}) \exp(-W_m/kT) \quad (2.5)$$

Now as the temperature T becomes indefinitely high, $T \rightarrow \infty$, the exponential terms tend to equality: that is, the upper energy states become more densely populated at the expense of the population of the lower energy states. Also, u_f must be of black body form because the system is in thermal equilibrium: and hence as $T \rightarrow \infty$, u_f also becomes very large (equation 2.1).

Consequently we must have

$$B_{nm} = B_{mn}$$

That is, the coefficients of absorption and of induced emission, and hence the respective elementary probabilities

Stimulated and Spontaneous Emission of Radiation

P_{nm} , dP_{mn} , are equal. Using this fact, and equation (2.5d), we may find an expression for u_f ,

$$u_f = \frac{A_{mn}/B_{mn}}{\exp\left(\frac{W_m - W_n}{kT}\right) - 1}$$

Comparing this with the Planck formula for u_f , we find that it will be satisfied if

$$W_m - W_n = hf$$

which is the Bohr frequency condition, and

$$A_{mn} = \frac{8\pi f^2}{c^3} hf \cdot B_{mn} \quad (2.6)$$

Thus Einstein's assumptions regarding the existence of spontaneous and induced emission, and the forms for their respective probabilities, are shown to be compatible with both the Planck radiation formula and the Bohr frequency condition. The probabilities for absorption and for induced emission must be equal, and an important relation between the induced emission coefficient B_{mn} and the spontaneous emission coefficient A_{mn} exists. We see from equation (2.6) that the relationship is

$$A_{mn} = (\text{number of radiation modes per unit volume per unit frequency range}) \times (\text{quantum energy at the transition frequency}) \times B_{mn} \quad (2.6')$$

This enables us to calculate the spontaneous emission coefficient A_{mn} from the absorption coefficient B_{nm} which is equal to the emission coefficient B_{mn} , and which is a readily measurable quantity. We have said that $8\pi f^2/c^3$ is the number of radiation modes per unit volume per unit frequency range in an enclosure very large compared with the wavelength at frequency f . In a microwave cavity enclosing molecules and resonant at a molecular transition