A Reinterpretation of Stimulated Emission as Spontaneous Emission Under Non Thermodynamic Equilibrium Conditions

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In 1916, Einstein postulated the existence of induced emission (einstrahlung), where a quantum of radiation from a light beam induced a molecule in an excited state to emit a quantum of radiation of the same frequency. The stimulating photon was unchanged by the ghostly interaction and the net effect of induced emission, which is now commonly known as stimulated emission, was indistinguishable from spontaneous emission (ausstrahlung). Here I show that the existence of stimulated emission, postulated by Einstein, is an artifact of his use of the Boltzmann distribution for continuous energy states. Here I show that with the use of Planck’s distribution for discontinuous energy states, stimulated emission becomes superfluous. By using the quantum-theoretical distribution, it becomes clear that at thermodynamic equilibrium, the emission of all quanta of radiation are indistinguishable from the spontaneous emission of quanta.

When molecules where the lower energy state and excited states are in thermodynamic equilibrium are subject to an intense light composed of radiation quanta of the appropriate frequency, the molecules enter a non-equilibrium state. In the non-equilibrium state, the population of energy states becomes inverted such that there are more excited states than lower energy states. Such a distribution is thermodynamically equivalent to a negative temperature state. In response to the incident light, the molecules spontaneously emit radiation quanta with the same frequency as the incident quanta. The transformation between incident radiant energy and emitted radiant energy is a consequence of the first and second laws of thermodynamics. There is no need to postulate a photon that induces the response, unchanged by the interaction.

1. Introduction

In 1916, in order to give Planck’s blackbody radiation law a more quantum-theoretical foundation than an electromagnetic foundation, Einstein [1] derived Planck’s blackbody radiation law using Bohr’s model of the atom applied to molecules, instead of using an electromagnetic-mechanical analysis. Einstein considered the molecule, not as an electromagnetic-mechanical resonator as Planck did, but as a quantized system that had two possible quantum states $Z_n$ and $Z_m$ that were characterized by energies $\varepsilon_n$ and $\varepsilon_m$, ($\varepsilon_n < \varepsilon_m$). Einstein related the mean energy of the molecule, a function of temperature, to the energy density of the radiation and the frequency of radiation. The transition from $Z_n$ to $Z_m$ resulted from the absorption of a quantum of energy ($h\nu_{nm}$) and the transition from $Z_m$ to $Z_n$ resulted from the emission of the same quantum of energy.

Using Boltzmann’s principle, Einstein determined that the relative number ($N_n$) of molecules that were in lower energy state $Z_n$ is given by:

$$N_n = p_n e^{\varepsilon_n / kT}$$  \hspace{1cm} (1)

Where, $k$ is Boltzmann’s constant, $T$ is the absolute temperature and $p_n$ is the statistical weight of state $Z_n$. Similarly, the relative number ($N_m$) of molecules in the excited state $Z_m$ is given by:

$$N_m = p_m e^{\varepsilon_m / kT}$$  \hspace{1cm} (2)

Where, $p_m$ is the statistical weight of state $Z_m$. At thermodynamic equilibrium, the ratio of $N_n$ to $N_m$ is given by:

$$\frac{N_n}{N_m} = \frac{p_n}{p_m} e^{\varepsilon_m - \varepsilon_n / kT}$$  \hspace{1cm} (3)

At thermodynamic equilibrium, the number of transitions from $Z_n$ to $Z_m$ that involve the absorption of a quantum of radiation is equal to the number of transitions from $Z_m$ to $Z_n$ that involve an emission of a quantum of radiation. The number of transitions per unit time that involve an emission of a quantum of radiation is given by $A^n_m N_m$, where $N_m$ is the number of molecules in state $Z_m$ and $A^n_m$ is a constant that characterizes the spontaneous transition from $Z_m$ to $Z_n$. Such emission is
independent of the incident radiation and takes place spontaneously.

Einstein considered the incident radiation to affect both the induced emission of a quantum of energy as a molecule goes from \( Z_m \) to \( Z_n \) and the induced absorption of a quantum of energy as a molecule goes from \( Z_n \) to \( Z_m \). The number of transitions per unit time that involve an induced emission is given by \( B^m_n N_m \rho_v \), where \( B^m_n \) is a constant that characterizes the induced transition from \( Z_m \) to \( Z_n \), \( N_m \) is the number of molecules in state \( Z_m \), and \( \rho_v \) is the energy density of the effective frequency (\( \nu_{nm} \)) of the resultant radiation. The number of transitions per unit time that involved an induced absorption is given by \( B^m_n N_n \rho_v \), where \( \rho_v \) is the energy density of the effective frequency (\( \nu_{nm} \)), \( N_n \) is the number of molecules in state \( Z_n \), and \( B^m_n \) is a constant that characterizes the induced transition form \( Z_n \) to \( Z_m \).

At thermodynamic equilibrium:

\[
A^m_n N_m + B^m_n N_m \rho_v = B^m_n N_n \rho_v
\]

Combine Eqn. (3) with Eqn. (4) to get:

\[
A^m_n N_m + B^m_n N_m \rho_v = B^m_n N_m \frac{p_n}{p_m} e^{\frac{\epsilon_m - \epsilon_n}{kT}} \rho_v
\]

Cancel \( N_m \), rearrange, and factor out \( \rho_v \) to get:

\[
A^m_n p_m = \rho_v (B^m_n p_m e^{\frac{\epsilon_m - \epsilon_n}{kT}} - B^m_n p_m)
\]

The L.H.S. is a finite expression. \( \rho_v \) and the R.H.S. would go to infinity if the temperature approached infinity unless the expression in the parentheses went to zero. In Einstein’s development, he postulated that the spectral energy density must approach infinity with increasing temperature (\( \rho_v \rightarrow \infty \), as \( T \rightarrow \infty \)), and thus

\[
B^m_n p_n = B^m_n p_m
\]

Of course the accuracy of Eqn. (7) depends on how the terms that contain \( \rho_v \) and \( T \) behave as they approach infinity. Substitute Eqn. (7) into Eqn. (6) to get:

\[
A^m_n p_m = \rho_v (B^m_n p_m e^{\frac{\epsilon_m - \epsilon_n}{kT}} - B^m_n p_m)
\]

Factor out \( B^m_n p_m \) to get:

\[
A^m_n p_m = \rho_v B^m_n p_m (e^{\frac{\epsilon_m - \epsilon_n}{kT}} - 1)
\]

and

\[
\rho_v = \frac{\frac{A^m_n}{B^m_n}}{(e^{\frac{\epsilon_m - \epsilon_n}{kT}} - 1)}
\]

which gives Planck’s blackbody radiation law when

\[
\frac{A^m_n}{B^m_n} = \frac{8 n m v^3}{c^2}
\]

That is, the ratio of the Einstein’s A and B constants of spontaneous and stimulated emission is equal to the temperature-independent prefactor in Planck’s blackbody radiation law.

Einstein [2,3] provided additional support for Eqn. (11) by considering conservation of linear momentum. In order for the analysis to be free of contradictions, at thermodynamic equilibrium, the linear momentum transferred to the molecules by the collision of photons must result in a stable Maxwellian distribution of velocities. The mean kinetic energy \( \frac{p^2}{2m} = \frac{1}{2} kT \) per degree of freedom in a blackbody radiation field of temperature \( T \) must be equal to \( \frac{1}{2} kT \) and the distribution must be independent of both the absorbed or emitted frequencies and the chemical nature of the molecule.

Einstein showed that the distribution would remain constant at thermodynamic equilibrium if the induced absorption resulted in a transfer of linear momentum to the molecule in the direction of the incident light, the stimulated emission resulted in a transfer of linear momentum antiparallel to the direction of the incident light, and the spontaneous emission of energy quanta were equally probable for all directions. Based on these assumptions, Einstein [2] obtained Eqn. (11). Einstein’s A and B coefficients, along with stimulated emission became a part of quantum theory [4-9], and textbooks of modern physics [10-14] and quantum electronics [15]; yet the question remains: how can a photon that induces stimulated emission remain unchanged as a result of the interaction? I now show that it is unnecessary to postulate such a ghostly interaction. The concept of stimulated emission is superfluous.

2. Results

Einstein [2] was led to the idea of stimulated emission “from a desire to postulate in the simplest manner the quantum-theoretical behavior of molecules in a manner analogous to the classical theory of Planck’s resonator”. Notice, however, that Einstein’s [1] derivation was not free from
classical foundations. He used the Boltzmann principle for continuous distributions of energy. Here I derive the temperature-independent prefactor for Planck’s blackbody radiation law using Planck’s distribution formula for quantized distributions. By building on a completely quantum-theoretical foundation, we discover that the concept of induced emission is superfluous. If we postulate that induced emission does not occur, at thermodynamic equilibrium we get:

$$A_m^N N_m = B_m^N N_m \rho \nu$$  \hspace{1cm} (12)

And if we postulate further that the ratio of the quantized states is not given by the Boltzmann distribution, which yields Eqn. (3), but by the Planck distribution, we get the following equation:

$$\frac{N_n}{N_m} = \frac{P_n}{P_m} \left( e^{ \frac{E_n - E_m}{kT} } - 1 \right)$$  \hspace{1cm} (13)

Substituting Eqn. (13) into Eqn. (12), we get:

$$A_m^N N_m = B_m^N N_m \frac{P_n}{P_m} \left( e^{ \frac{E_n - E_m}{kT} } - 1 \right) \rho \nu$$  \hspace{1cm} (14)

After cancelling $N_m$, we get:

$$A_m^N = B_m^N \frac{P_n}{P_m} \left( e^{ \frac{E_n - E_m}{kT} } - 1 \right) \rho \nu$$  \hspace{1cm} (15)

Solving for the spectral energy density ($\rho \nu$), we get:

$$\rho \nu = \frac{A_m^N P_m}{B_m^N P_n} \frac{P_n}{P_m} \left( e^{ \frac{E_n - E_m}{kT} } - 1 \right)$$  \hspace{1cm} (16)

which gives Planck’s blackbody radiation law if

$$\frac{A_m^N P_m}{B_m^N P_n} = \frac{8 \pi \nu^3}{c^2}$$  \hspace{1cm} (17)

At thermodynamic equilibrium, the statistical weight of states $Z_n$ and $Z_m$ are equal, $\frac{P_m}{P_n} = 1$, and the temperature-independent prefactor of Planck’s blackbody radiation law is given by the ratio of the spontaneous emission constant $A_m^N$ to the induced absorption constant $B_m^N$.

$$\frac{A_m^N}{B_m^N} = \frac{8 \pi \nu^3}{c^2}$$  \hspace{1cm} (18)

We arrive at Planck’s blackbody radiation law from Bohr’s model of the atom without the need to postulate the existence of spontaneous emission. This is an extraordinary result given that Hawkes and Latimer [16] wrote in Lasers: Theory and Practice, “The development of the laser has been one of the great triumphs of science in the twentieth century. The foundations were laid by Einstein in 1917, who pointed out that the equation proposed by Planck to describe the spectral distribution of light emitted from a black body could be derived quite simply by assuming the existence of a hitherto-unknown type of light-emission process which has since become known as stimulated emission”.

3. Discussion

LASER is an acronym that stands for Light Amplification by Stimulated Emission of Radiation [17-19]. Laser, which is based on visible waves of light, was used in analogy to maser, which stood for Microwave Amplification by Stimulated Emission of Radiation. Arthur Schawlow considered the acronym loser, to emphasize oscillation over amplification [19]. Acronyms considered for instruments based on waves outside the visible spectrum included, iraser, for infrared, gaser for gamma rays and raser for radio waves [20]. According to Basov [21], in semiconductor lasers, “under the influence of a quantum, an electron may be transferred from the conduction band to a vacant place (hole) on the valent band. Such a transfer will be accompanied by the emission of a light quantum identical in frequency, direction of propagation and polarization to the quantum which produced the emission. This process is connected with an increase of the field energy and is called stimulated emission. We recall that stimulated emission was discovered by A. Einstein in 1917 during an investigation of thermodynamical equilibrium between radiation fields and atoms”.

As a biophysical plant cell biologist [22,23], well versed in Einstein’s Photochemical Law of Equivalence and the first and second laws of thermodynamics, I know that any photon that initiates a biological process ranging from photosynthesis to vision, must be affected by the interaction. In order to remove the inconsistency between the biological observations and the concept of stimulated emission, I went back to the original papers by Einstein [1-3], and found the point at which the concept of stimulated emission was introduced. By using Planck’s distribution function for discontinuous states instead of Boltzmann’s distribution function for continuous states, I have been able to show that the concept of
If spontaneous emission does not happen and there is no amplification of the energy output relative to the energy input, how does the laser work? Laser action can be interpreted to result from the intense excitation of molecules from thermodynamic equilibrium to a non thermodynamic equilibrium state. The input of energy by optical pumping or an electrical potential causes the electrons to transition from the lower energy state to the excited state according to the First Law of Thermodynamics sans amplification. In fact, consistent with the Second Law of Thermodynamics, in a laser, much of the energy is lost in the transformation of the input energy to the useful output energy. The intense excitation results in a population inversion elsewhere characterized by a negative absolute temperature [21,26-30]. Since the inverted population of molecules in the laser are in an environment characterized by a positive temperature, they simply are not in thermodynamic equilibrium with that environment. Consequently, the molecules return to thermodynamic equilibrium by emitting light with a frequency dependent on the transition from the excited state to the lower energy state. The radiant energy out is related to the input energy through the first and second laws of thermodynamics. There is no amplification resulting from a ghostly interaction between an input photon and the photons emitted by stimulated emission. Again, this is an extraordinary result since Millonni and Eberly [31] wrote in Lasers, “The word laser is an acronym for the most significant feature of laser action: light amplification by stimulated emission of radiation. There are many different kinds of laser, but they all share a crucial element: each contains material capable of amplifying radiation. This material is called the gain medium, because radiation gains energy passing through it. The physical principle responsible for this amplification is called stimulated emission, and was discovered by Albert Einstein in 1916.”

The spontaneously emitted radiation quanta are coherent, monochromatic and collimated. The coherence, monochromaticity, and collimation results in part from the choice of a lasing material with the correct chemico-spectral properties, and from optical engineering design principles that include using highly-reflecting end walls separated by a prescribed distance [30], and a small angular aperture [32]. While the concept of stimulated emission gave impetus for the invention of lasers [21,24,33] and laser spectroscopy [34,35], it is not necessary to explain the action of lasers. The thermodynamic explanation given here has the advantage of not requiring a ghostly interaction of photons that can cause a transformation without being changed themselves.

I have previously interpreted the temperature-independent prefactor of Planck’s blackbody radiation law in terms of the volume of the photon [36] and interpreted the sign of the prefactor in terms of matter and antimatter [37]. Here I also show that the prefactor can also be interpreted as proportional to the ratio of spontaneous emission to induced absorption.

Laser light is important in almost every facet of human endeavor [20,38], and unsurprisingly, laser light is instrumental in basic research done by biophysical cell biologists and others to visualize cells and tissues with confocal [39,40] and multiphoton microscopy [41]; to determine the mechanical properties of cytoplasm [42]; to measure the force of organelles [43,44], and the force of adhesion between them [45]; to measure the force of cellular motors [46,47]; to perform microsurgery [48,49] and microdissection [50]; to isolate organelles [51]; to measure the speed of intracellular flow [52]; to localize and activate cellular photoreceptors [53,54]; to determine the mechanism of energy transfer in photosynthesis [55]; to determine the DNA content of nuclei [56]; to sequence DNA [57]; to measure the diffusion of molecules [58,59]; and to visualize molecules with super-resolution [60-62]. Here I show that laser light is understandable, not in terms of stimulated emission, but in terms of the first and second laws of thermodynamics and the spontaneous emission of photons from an inverted population. According to the results presented here, in laser light, there is no stimulated emission and no amplification, which makes the acronym LASER, a misacronym. The fundamental principle of laser action is not stimulated emission but population inversion due to optical pumping or an electrical potential.
Population inversion [20,63,64] was the key to inventing the laser.

In their Nobel lectures, Basov [21] Prochorov [24] and Townes [33] considered light sources that produce light by stimulated emission to be fundamentally different from light sources such as sunlight, filament lamps, luminescent lamps, whose light is emitted by spontaneous emission. Here I have shown that light emitted by lasers is readily viewed as a result of spontaneous emission, which puts laser light in the same category as sunlight, candlelight, and light produced by bioluminescent organisms such as fireflies and glowworms.

References


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