

Second law of thermodynamics and thermally excited quantum oscillators

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A review is given of the first investigations formulating the idea of direct conversion of thermal energy into coherent electromagnetic radiation in quantum oscillators. It is shown that the operation of a gasdynamic CO₂ laser can be described, from the energy point of view, by the second law of thermodynamics.

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1. INTRODUCTION

The appearance of a gasdynamic laser, which was the first device to use the principle of quantum generation of electromagnetic oscillations and to convert directly thermal energy into coherent radiation, has stimulated interest in the relationship between the growing subject of quantum electronics and the well-established science of thermodynamics. An examination of the historical background shows that the idea of direct conversion of heat into coherent radiation by means of quantum oscillators was put forward back in 1959 before the appearance of lasers. It has also been found that the second law of thermodynamics applies to heat engines (radiation generators) in which the active material has a discrete energy spectrum with an upper limit.

It is interesting to analyze these undeservedly forgotten investigations. The proof of the second law of thermodynamics based on quantum transitions of particles between discrete energy levels is found to be tutorially simpler than the traditional method employing a cyclic process.

2. THERMAL MASER

It follows from electron spin resonance investigations that the population of spin levels of a paramagnetic particle can be altered by subjecting a sample to an alternating radiofrequency (rf) magnetic field. The rf field should be resonant, i. e., its frequency should be equal to the energy gap between two levels, and it should be sufficiently strong to make the probability of the field-induced quantum transitions comparable with the rate of relaxation processes tending to re-establish the initial distribution of particles between the spin levels. If there is no rf field, a paramagnet is in a thermal equilibrium state and the distribution of the particles between the levels is of the Boltzmann type. In crystals with low concentrations of paramagnetic ions the thermal relaxation process is governed by the interaction between these ions and lattice phonons. Thermal vibrations of the lattice act as a thermostat of the paramagnetic particles. If the number of magnetic levels in the spin system exceeds 2, the simultaneous action of a strong rf field and thermal relaxation may invert

the population of the system that the numbers of ions at some level is higher than the number of ions at a lower level. In the simplest case this population inversion may occur in a three-level system (lower E_1 , intermediate E_2 , and upper E_3 levels) if the rf field induces transitions of the particles between the lower and upper levels (Fig. 1).

The paramagnetic ion and the experimental arrangement are selected so that the times of a relaxational transition from the upper to the intermediate level τ_{32} and from the intermediate to the lower level τ_{21} satisfy the condition $\tau_{32} \ll \tau_{21}$ and both these times are, if possible, shorter than τ_{31} . This produces a population inversion between the intermediate and lower levels. The inversion results because the particles released from the upper level by thermal relaxation are held up by the intermediate level and increase its population. On the other hand, some of the particles are lifted by the rf field from the lower to the upper level and the population of the lower level becomes less than the equilibrium value. Then, a paramagnetic crystal becomes capable of amplifying and generating rf oscillations at a frequency $h\nu_s = E_2 - E_1$.

Although the principle of maser amplification of electromagnetic oscillations has made it possible to generate infrared and optical radiation, at the time of the discovery of the maser action (before the appearance of lasers) there was—in the opinion of physicists—a fundamental difficulty which made it necessary to search for new methods of establishing a population inversion.

The operation of a three-level maser at some frequency ν_s requires the existence of sufficiently powerful and monochromatic radiation of pump frequency $\nu_p > \nu_s$ needed to excite the active medium. In other words, before generating such radiation, one would require a source of the same type of radiation and, on top of that, it should operate at a higher frequency. This conceptual difficulty arose because in the microwave range the ESR lines are relatively narrow and the power of thermal radiation sources is clearly insufficient for a population inversion. Later, investigations in the optical range demonstrated that one could pump wide bands of luminescence centers and to obtain coherent radiation

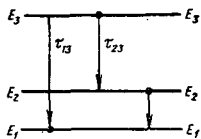


FIG. 1. System of three energy levels. The arrows identify relaxational transitions.

as a result of optical transitions characterized by narrow spectral lines. This seemingly basic difficulty, which hindered the extension of the maser generation principle to the optical range, was later forgotten and an active search for methods of obtaining a population inversion which would not require exciting radiation was stopped.

The discovery of Schulz-DuBois and Scovil,^[1] described below, proposed a new method for creating an amplifying medium in a laser. This method does not require auxiliary radiation because it uses the energy of a thermal source and thus provides a method for direct conversion of thermal energy into a higher form of energy represented by coherent electromagnetic radiation.

Figure 2 shows a system which explains the principle of a thermally excited maser. A cylindrical rod 1 is a single crystal whose central part 2 contains paramagnetic ions with suitable energy levels. For example, the rod may consist of corundum (Al_2O_3) with an admixture of chromium. Investigations carried out by Manenkov and Prokhorov^[2] demonstrated that Cr^{3+} ions in a corundum crystal have a system of magnetic levels which is quite suitable for microwave masers. For example, a sample of rose-colored ruby with 0.05% Cr, kept at a temperature of 1.5°K and subjected to a transverse magnetic field of 2 kOe, can operate as a maser at a frequency of 1420 MHz.^[3] Under these conditions the spin-relaxation times are as follows: $\tau_{12} = 150$ msec, $\tau_{13} = 150$ msec, and $\tau_{23} = 40$ msec.^[1]

A heater 3 is wound on one end of the rod and this heater is connected periodically by a switch to an electrical energy source. The other end of the rod is in good thermal contact with a heat reservoir 4, which should have a sufficiently low temperature T_1 . This reservoir may be, for example, a container with liquid helium. The dimensions of the crystalline rod and its thermal diffusivity should be such that the equalization of the temperature along the whole sample occurs in a time shorter than the spin-lattice relaxation time. In the case of small ruby samples the time needed to establish the lattice temperature can be reduced to ~ 10 μsec .

The central part of the rod is coupled inductively, by

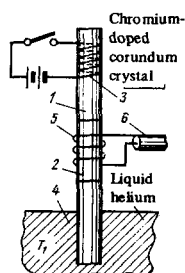


FIG. 2. Schematic diagram of a thermally excited ruby crystal maser.

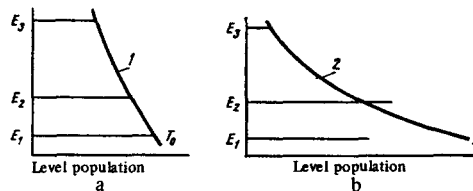


FIG. 3. (a) Distribution of particles between levels at a high initial temperature T_0 (the horizontal lines are proportional to the numbers of particles at the various levels; curve 1 is a Boltzmann distribution with a temperature T_0). b) Distribution of particles between levels at some moment after the crystal lattice is cooled to a lower temperature T_1 (a population inversion is established between levels 2 and 1; curve 2 is a Boltzmann distribution with a temperature T_1).

a loop 5, to a coaxial line 6, which is connected to an external load when a thermal maser is employed as an amplifier or forms an oscillatory circuit when the maser is employed as an oscillator.

The system operates as follows. A current is passed through the heater until the temperature T_0 of the central part of the rod becomes considerably higher than the reservoir temperature T_1 (for example, $T_0 = 20^\circ\text{K}$) and the spin system reaches a thermal equilibrium corresponding to this temperature. The distribution of the particles between the levels during this part of the cycle is shown in Fig. 3a, where the horizontal lines are proportional to the level populations. An equilibrium Boltzmann distribution at $T = T_0$ is represented by curve 1. Next, the heating is stopped so that the rod cools rapidly and the lattice temperature in the central part of the rod becomes close to the thermostat temperature T_1 . Now the particles become redistributed between the levels in the spin system in accordance with the new lattice temperature. The population of the level E_2 increases because of the particles arriving from the level E_3 . The population of the level E_1 also increases due to transitions of particles from E_2 and E_3 but the increase is smaller because the corresponding spin-lattice relaxation times τ_{21} and τ_{31} are longer than τ_{32} . Consequently, for a time the population of the level E_2 may exceed the number of particles at the level E_1 . During this time the crystal rod is capable of amplifying and generating electromagnetic oscillations at the signal transition frequency. This inverted state is shown in Fig. 3b. Subsequently, the populations of the levels tend to a Boltzmann distribution with a temperature T_1 and the inverted state disappears (curve 2). The cycle described above is then repeated again.

Part of the thermal energy transferred from the heater to the crystal is expended in the excitation of the spin system and this energy is then transformed into the energy of electromagnetic oscillations.

It is important to stress that this population inversion in the spin system exists only for a certain part of the cycle and it is due to the fact that the particles at different energy levels have very different thermal relaxation times.

A system of three energy levels with different relaxation times, first proposed by Schulz-DuBois and Sco-

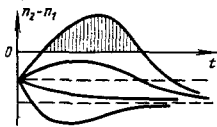


FIG. 4. Time dependences of the difference between the populations of levels 2 and 1. In the time interval when $n_2 - n_1 > 0$ the system can amplify electromagnetic oscillations at the signal transition frequency.

vil,^[1] plays the same fundamental role in thermally excited quantum oscillators as the three-level system proposed by Basov and Prokhorov^[4] in quantum electronics.

A redistribution of particles between the levels, which occurs as a result of cooling or heating, is described by a system of rate equations. The case of instantaneous heating of a three-level system followed by relaxation at a higher temperature is described in^[5]. We shall now give a similar treatment of the cooling case so as to obtain a quantitative description of the process of redistribution of particles between magnetic levels in a thermal maser.

We shall assume that cooling of the crystal lattice from its initial temperature T_0 to the final value T_1 is faster than the spin-lattice relaxation. This means that the spin-lattice relaxation times τ_{ik} are constant and correspond to the temperature T_1 , whereas the populations of the magnetic levels initially ($t=0$) correspond to the temperature T_0 . Since the signal transition couples the first and second levels, the equations for the numbers of particles N_1 and N_2 at these levels are

$$\left. \begin{aligned} \frac{dN_1}{dt} &= -(w_{13} + w_{12})N_1 + w_{21}N_2 + w_{31}N_3, \\ \frac{dN_2}{dt} &= -(w_{21} + w_{23})N_2 + w_{12}N_1 + w_{32}N_3, \\ N_1 + N_2 + N_3 &= N_0, \end{aligned} \right\} \quad (1)$$

where N_0 is the total number of particles localized in all the levels of the spin systems; $w_{ik} = \tau_{ik}^{-1}$ are the probabilities of relaxational transitions. The probability w_{ik} of a transition from an upper E_i to a lower E_k level is always higher than the probability of a transition w_{ki} in the opposite direction:

$$w_{ik} = w_{ki} \exp\left(\frac{h\nu_{ik}}{T}\right), \quad \nu_{ik} = E_i - E_k,$$

where $\nu_{21} = 14.25 \times 10^6 \text{ sec}^{-1}$ and $\nu_{23} = 11.31 \times 10^9 \text{ sec}^{-1}$. The above expression follows from the principle of detailed equilibrium. We shall now introduce relative populations $n_i = N_i/N_0$ and rewrite the inhomogeneous system of equations (1) in the vector form^[6]

$$\left. \begin{aligned} \dot{\mathbf{n}} + A\mathbf{n} &= \mathbf{v}, \\ \mathbf{n} &= (n_1, n_2), \quad A = \begin{vmatrix} w_{13} + w_{12} + w_{31} & w_{31} - w_{21} \\ w_{32} - w_{12} & w_{21} + w_{23} + w_{32} \end{vmatrix}, \\ \mathbf{v} &= (w_{31}, w_{32}), \end{aligned} \right\}$$

The solution of the system (1) is of the form

$$\mathbf{n} = \mathbf{n}_\infty + c_1 e^{-\lambda_1 t} \mathbf{h}_1 + c_2 e^{-\lambda_2 t} \mathbf{h}_2,$$

where $\lambda_1 = 67.86 \text{ sec}^{-1}$ and $\lambda_2 = 21.31 \text{ sec}^{-1}$ are the eigenvalues of the matrix A which correspond to the eigen-

vectors

$$\begin{aligned} \mathbf{h}_i &= (1, b_i), \quad b_i = \frac{\lambda_i - a_{11}}{a_{12}}, \\ c_1 &= \frac{(n_1^{(0)} - n_1^{(1)})b_2 - (n_2^{(0)} - n_2^{(1)})}{b_2 - b_1}, \quad c_2 = \frac{(n_1^{(0)} - n_1^{(1)}) - b_1(n_2^{(0)} - n_2^{(1)})}{b_2 - b_1}; \\ \mathbf{n}_\infty &= (n_1^{(\infty)}; n_2^{(\infty)}) = (0.377; 0.360), \\ (n_1^{(0)}; n_2^{(0)}) &= (0.337; 0.336), \end{aligned}$$

$n_{1,2}^{(0)}$ and $n_{1,2}^{(1)}$ are the relative populations of the levels 1 and 2 when the initial temperature is T_0 and the final temperature is T_1 . The difference between the populations of these two levels is

$$n_2 - n_1 = c_1(b_1 - 1)e^{-\lambda_1 t} + c_2(b_2 - 1)e^{-\lambda_2 t} + n_2^{(\infty)} - n_1^{(\infty)}.$$

The time dependence of the difference $n_2 - n_1$ is plotted in Fig. 4. During the initial stage the population of the upper level is less than that of the lower level and then the function $n_2 - n_1$ changes its sign, so that the population inversion appears and lasts up to $t = 4.3$ msec; this is followed by a slow fall of $n_2 - n_1$ to the value $n_2^{(\infty)} - n_1^{(\infty)}$, which corresponds to the equilibrium populations at the final temperature T_1 . It should be noted that the additional population of the level 2 appears because of the particles arriving from the higher level 3 whose population changes most during the relaxation time, the change being from $n_3^{(0)} = 0.328$ to $n_3^{(\infty)} = 0.262$.

This calculation shows that, from the point of view of relaxation processes, a thermal laser can be made of a ruby crystal. The authors of the present review are not aware of whether the system has been made to work but its value is that it demonstrates for the first time the principle of thermal excitation of quantum systems in generation of electromagnetic radiation. The idea was discussed at the First International Conference on Quantum Electronics. At this conference Schulz-DuBois^[7] presented a paper showing that thermal generation of coherent radiation could be realized experimentally and that this could be practical interest. The first heat engine in which the principle of thermal excitation was put into practice has been the gasdynamic laser.

3. GASDYNAMIC LASER AS A HEAT ENGINE

A thermally excited three-level quantum-mechanical system can be regarded as a heat engine and its thermal efficiency can be calculated by applying the thermodynamic approach.

Following the paper of Scovil and Schulz-DuBois,^[8] we shall consider the specific case of a three-level maser. As is usual in thermodynamics, we shall introduce some idealized properties of the active substance and system. A system of this kind may be very difficult to realize in practice. However, this does not affect the correctness of the final conclusion if none of the properties of the active substance or system are in conflict with the first and second laws of thermodynamics.^[9]

We shall assume that no relaxation processes take place in our three-level system but that radiative transitions are allowed between any pair of levels. Applying the laser terminology, we shall call the $1 \rightarrow 3$ transition the excitation or pump transition at a frequency ν_p , the $1 \rightarrow 2$ transition the signal of frequency ν_s , and the tran-

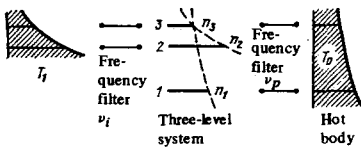


FIG. 5. System of three energy levels in thermal contact with two thermostats.

sition $2 \rightarrow 3$ the idler of frequency ν_i . We shall assume the existence of idealized frequency filters which do not emit any thermal radiation and pass without attenuation the radiation near a certain frequency ν but absorb completely the thermal radiation of all other frequencies.

We shall assume that the three-level system is brought into simultaneous thermal contact with two thermostats (Fig. 5), one of which is kept at a high temperature T_0 and the other at a much lower temperature T_1 . Contact is made by radiation, which is transmitted by a filter passing at the frequency ν_p in the case of the thermostat T_0 and is transmitted by a filter passing a frequency ν_i in the case of the thermostat T_1 . In this case the relative population of the levels 1 and 3 corresponds to the higher temperature T_0 and the relative population of the levels 2 and 3 to the lower temperature T_1 :

$$\frac{n_2}{n_1} = \frac{n_2}{n_3} \frac{n_3}{n_1} = \exp\left(\frac{h\nu_i}{T_1}\right) \exp\left(-\frac{h\nu_p}{T_0}\right), \quad (2)$$

where n_i is the population of the level in question.

Amplification and generation of an rf field at a frequency ν_s are possible only if the population relating to the signal transition is inverted $n_2/n_1 \geq 1$. Each thermal radiation quantum $h\nu_p$ arriving from this thermostat T_0 and absorbed by the system creates one particle at the upper level 3. This particle drops successively to the levels 2 and 1 emitting quanta $h\nu_i$ and $h\nu_s$. The $3 \rightarrow 2$ transition is induced by the thermal radiation from the thermostat T_1 , which absorbs the photon energy $h\nu_i$ and converts it into heat. The quantum energy $h\nu_s$ can be extracted from the system in the form of a useful signal. In our idealized lossless maser system each quantum $h\nu_p$ produces one quantum $h\nu_s$ at the signal transition frequency so that the quantum efficiency of the system is $\eta_q = \nu_s/\nu_p$. The quantum efficiency is the energy characteristic of the maser system acting as a converter of the thermal energy taken from the reservoir T_0 into the energy of electromagnetic oscillations at the signal transition frequency.

Transforming Eq. (2) subject to the condition $\nu_p = \nu_s + \nu_i$, we find that

$$\frac{n_2}{n_1} = \exp\left[\frac{h\nu_s}{T_1} \left(\frac{\nu_p}{\nu_s} \frac{T_0 - T_1}{T_0} - 1\right)\right].$$

The expression $\eta_c = (T_0 - T_1)/T_0$ is the efficiency of a heat engine operating in the Carnot cycle when the thermostat T_0 is used as a heater and the thermostat T_1 is used as a refrigerator. Allowing for the population inversion condition $n_2/n_1 \geq 1$ and the physical meaning of the frequency ratio ν_s/ν_p , we find that

$$\eta_q \leq \eta_c.$$

This means that it is impossible to construct a system based on the principles of quantum generation of electromagnetic radiation which would take energy from a thermal reservoir, and transform it into the energy of coherent radiation without any changes in the state of the surrounding bodies. In other words, thermally excited quantum oscillators obey the second law of thermodynamics and their operation requires not only a heater but also a refrigerator. The energy efficiency of such oscillators cannot be higher than the efficiency of a cyclic heat engine employing the same heater and the same refrigerator.

We shall now consider the actual physical systems which can be used as masers, thermostats, and filters. One such system is considered by Scovil and Schulz-DuBois,^[8] in which the system of energy levels is provided by gadolinium ions and the high-temperature thermostat is a gas-discharge flashlamp whose radiation is passed through a cutoff waveguide filter. The low-temperature thermostat is represented by the vibrations of a lattice kept at a low temperature. A crystal is assumed to contain not only gadolinium ions, but also cerium ions which—on the one hand—are coupled by the spin-spin interaction to the idler transition at a frequency ν_i in gadolinium ions and—on the other—have a short spin-spin lattice relaxation time and are therefore in good thermal contact with the lattice vibrations. Clearly, in actual realization of this system there is no need for thermal radiation and frequency filters considered in our analysis (in the above example the energy transfer at one frequency is performed by cerium ions).

Another example is a gasdynamic CO_2 laser, first discussed by Konyukhov and Prokhorov,^[10,11] Gasdynamic lasers are a variety of molecular gas lasers in which energy is provided by vibrationally excited molecules present in a gas heated to a high temperature and an amplifying medium is formed by thermal relaxation of these molecules in the course of their flow through a supersonic nozzle. A gas medium in such a laser consists of three components. The first component, whose concentration is highest (molecular nitrogen, 80–90 vol. %) is a molecular gas with a long vibrational relaxation time. Its molecules can remain for a relatively long time in a vibrationally excited state which they acquire at the high temperature of the gas mixture. Consequently, the losses of these vibrationally excited molecules are slight during the motion of the gas mixture through the supersonic nozzle. The vibrational energy of the nitrogen molecules is thus the thermostat T_0 from which energy is acquired in generation of laser radiation. The vibrational temperature T_0 of molecular nitrogen is of the order of 1000°K.

The second component (carbon dioxide gas, 5–10 vol. %) is a molecular gas with a short vibrational relaxation time. The CO_2 molecule has allowed dipole transitions in the infrared part of the spectrum and, in particular, a vibrational-rotational band at 10.6 μ , so that it can interact with an electromagnetic field. The CO_2 molecule has a vibrational level 00^0_1 ($E = 2349 \text{ cm}^{-1}$), which is almost identical in energy (energy defect $E = 18 \text{ cm}^{-1}$) with the first vibrational level of the N_2 mole-

cule so that a resonant exchange of the vibrational excitation takes place between these levels. The lower active level of carbon dioxide, 10^0 ($E = 1388 \text{ cm}^{-1}$), is located high above the ground vibrational state of this molecule. The vibrational levels of the CO_2 molecule, comprising the upper laser level (00^01), the lower level (10^00), and the ground state (00^00), forms a three-level system with an allowed $3 \rightarrow 2$ radiative transition, which corresponds to the signal transition in the maser scheme. The transfer of energy from the thermostat T_0 to the three-level system occurs without participation of thermal radiation because energy is transferred in molecular collisions in the gas. The selectivity of the interaction between the thermostat T_0 and the system is ensured by the resonant nature of the transfer of excitation from the N_2 to the CO_2 molecules.

The third component (H_2O vapor, 1–2 vol. %) is a molecular gas with an even shorter vibrational relaxation time. The purpose of this component is to reduce the lifetime of the CO_2 molecule at the lower laser level and to accelerate the transfer of the CO_2 molecules to the ground state. In emitting radiation the CO_2 molecule passes through a cycle which consists of the transition from the ground state to the (00^01) level as a result of collisions with the excited nitrogen molecules, the radiative transition between the active levels, and the relaxational transition to the ground state. If the third component is absent or its concentration is slight, the bottleneck in this chain is the transition from the lower laser level to the ground state. The H_2O molecules provide the coupling between the three-level system and the thermostat T_1 by the transition $2 \rightarrow 1$, which corresponds to the idler transition in the case of a maser. The thermostat T_1 represents the translational and rotational degrees of freedom of all the molecules in the mixture. On the one hand, the H_2O molecules have a short vibrational relaxation time, so that the populations of their vibrational levels is in equilibrium characterized by the temperature T_1 and, on the other, they ensure a high probability of the exchange of the vibrational quanta between H_2O and the CO_2 molecules at the lower laser level. The selectivity of this interaction is less than that of the CO_2 – N_2 vibrational exchange in the upper level so that a high concentration of water vapor (greater than 5–8 vol. %) in the mixture accelerates the relaxation of the CO_2 molecules from the upper active level as well and, consequently, it results in a significant deactivation of the nitrogen molecule.

The low-temperature thermostat T_1 does not exist initially in the hot gas before its expansion because all the degrees of freedom of the molecules are in thermal equilibrium: this thermostat appears only during the motion of the gas through the nozzle. The gas expansion is accompanied by the conversion of the energy of random motion of the molecules into the energy of the directional mass motion of the gas. Since the flow is adiabatic, without supply of heat, the energy represented by the random thermal motion of the molecules decreases and a low translational and rotational gas temperature $T_1 \sim 300 \text{ }^\circ\text{K}$ is established in the supersonic stream.

We shall now see what physical assumptions have to be made to apply the model of a three-level system, which is in thermal equilibrium with two thermostats, to the operation of a gasdynamic laser. The three levels mentioned above belong to a polyatomic molecule which is rich in vibrational levels. However, these levels are located fairly low above the ground state so that we can introduce the concept of a normal oscillation (mode) and they belong to different modes. This means that the thermostats interact with different vibrational modes of the molecule. The exchange of quanta within a mode is a rapid process compared with the exchange between the modes so that the modes can be regarded as independent and we can apply to them the concept of a mode temperature T_i . The values of T_i should be equated to the temperatures of the relevant thermostats. Symmetric ($v_a, 0, 0$) and deformation ($0, v_d, 0$) modes are in equilibrium with the thermostat T_1 , whereas an asymmetric mode ($0, 0, v_s$) is in equilibrium with the thermostat T_0 . The partition function of this state is

$$\frac{1}{Q(T_0, T_1)} = \left[1 - \exp\left(-\frac{\theta_a}{T_0}\right) \right] \left[1 - \exp\left(-\frac{\theta_d}{T_1}\right) \right]^2 \left[1 - \exp\left(-\frac{\theta_s}{T_1}\right) \right],$$

where θ is the characteristic vibration frequency. The number of molecules at a level i is

$$n_i = \frac{1}{Q} \exp\left(-\frac{E_i}{T}\right),$$

where instead of T we substitute T_1 on T_0 , depending on which mode this level belongs.

The next assumption in dealing with an idealized laser system has been the exclusion of all the relaxation transitions. This has meant that there are no other physical factors, apart from the interaction with the thermostats, which would influence the distribution of particles between the energy levels. In the case of the CO_2 molecule this assumption means that the intermode exchange processes and the V – T relaxation have a negligible influence on the population of the (00^01) and (10^00) levels, compared with the V – V exchange with the N_2 molecules at the (00^01) level and with the H_2O molecules at the (10^00) level. This assumption is satisfied in the operation of a gasdynamic laser.

The relationship (2) for a gasdynamic laser reads as follows:

$$\frac{n_3}{n_2} = \exp\left(-\frac{E_3}{T_0}\right) \exp\left(\frac{E_2}{T_1}\right) = \exp\left[\frac{h\nu}{T_1} \left(\frac{E_3}{h\nu} \frac{T_0 - T_1}{T_0} - 1\right)\right];$$

$$E_2 = E(10^00) - E(00^00),$$

$$E_3 = E(00^01) - E(00^00), \quad h\nu = E_3 - E_2.$$

In addition to the conclusions relating to the validity of the second law of thermodynamics in the case of thermally excited quantum oscillators, which have been deduced earlier, the above relationship has a consequence which applies to the gasdynamic CO_2 laser.

There is a lower limit of the temperature drop (T_0/T_1) between the entry and exit of a supersonic nozzle above which a population inversion may be established in the stream of gas flowing through such a nozzle. This drop is governed solely by the position of those molecular vibrational levels which act as the upper

and lower laser levels. In the case of the carbon dioxide molecule, we find that $(T_0/T_1)_{\min} = E_3/E_2 = 1.69$.^[12] This temperature drop corresponds to the minimum expansion of the gas flowing through the nozzle. If this is represented by the Mach number at the nozzle exit, we find that

$$\frac{T_0}{T_1} = 1 + \frac{\gamma-1}{2} M^2, \quad M_{\min} = \sqrt{\frac{2}{\gamma-1} \frac{h\nu}{E_2}},$$

where γ is the adiabatic exponent (specific-heat ratio).

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