THE EFFECT OF STIMULATED EMISSION ON THE DIFFUSION OF RESONANCE EXCITATION THROUGH A GAS

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ABSTRACT: A method of treatment of the diffusion of resonance excitation in a gas without regard to the effect of stimulated emission was reported in the Astrophysical Journal (May 1961) by McIntyre and Fowler. Also it was considered that the number of unexcited states far exceeded the number of excited states. Both of these restrictions are relaxed. It is seen that the probability of a stimulated emission can be treated as a nega-

tive probability of absorption and therefore a direct relationship may be derived between the total radiation flux from emitters in the gas to simply the rate of spontaneous emission throughout the volume.

INTRODUCTION

The purpose of this work is to extend the kinetic-theoretical description[1] of the diffusion of resonance excitation through a gas to include the effect of stimulated emission and to greatly expand the range of excitation over which the previous work is appiicable.

The term "resonance excitation" is applied to radiation emmited by an atom in a transition from one excited state to another. Since it is highly absorbable by other atoms identical to the emitter atom, the process of diffusion is quite complex. In early treatments of the subject[2-7] attempts were made to link the physical phenomena to a diffusion-type equation for the density of excited atoms. In addition, all but R.G. Fowler[3] neglected the displacement of an excited atom during the period between absorption and reemission. R.G. McIntyre and R.G. Fowler[1] further relaxed the former restriction by approaching the problem from a purely kinetic-theoretical basis and refraining from taking averages over velocity space too quickly. However, the assumption was made that the relative density of excited states was sufficiently low that stimulated emission was negligible. It is intended in this report to generalize the physical situation.

DESCRIPTION OF THE PROBLEM AND DERIVATION OF THE EQUATIONS OF TRANSPORT

Let a volume contain a monatomic gas. Since the dynamics of the gas itself are so much more rapid than the radiation diffusion process, one may assume an initial and uniform steady-state velocity-distribution $f(\mathbf{c})$ applicable over a wide range of probiems. We consider that only two states of the gas exist and that there is a known initial distribution of each state. Furthermore, it is assumed that the initial radiation field and any impinging radiation fields are known. Let us define $f_1(\mathbf{r}, \mathbf{c}, t)$ as the velocity-distribution function of the excited states. Their sum must equal $f(\mathbf{c})$. The symbols $\mathbf{r}$ and $\mathbf{c}$ represent position and velocity respectively.

We further assume that the free path distance of atoms between collisions with other atoms, is very large compared to (a) the mean displacement of an excited atom during the period between an absorption and re-emission and (b) the expected distance, herein referred to as the ***skip distance***, that a photon might travel during an emission-reabsorption process. The above assumptions do not preclude the creation of excited states by collision. It does imply, however, that either an excited atom emits before collision or else its loss to a particular cell in phase space is exactly compensated for by collisions of the second kind.

As was pointed out earlier, one source of radiation is that radiation field is impinging on the vessel. A second source is any axcited atom. In dealing with this source it is desirable to look back to its source of excitation. If the atom became excited by any source other than the radiation field itself, we have an internal driving term. The two most common internal driving mechanisms are atom-atom collisions and electron-atom collisions. Other sources may be treated exactly as the above two, i.e. some mechanism is producing excited atoms using as raw material the unexcited atoms. This may or may not be a process dependent upon the velocity of the unexcited atom. One must determine rate coefficients, each of which is the probability that an unexcited atom with a given velocity will be given excitation energy per unit time. These coefficients will be discussed more fully later on. Once an atom becomes excited, its loss of excitation energy will be considered to be either spontaneous emission or stimulated emission. The sinks of radiation are absorption by unexcited atoms and loss through the walls of the vessel.

Now, as in reference (1) we let A equal the probability per unit time that an excited atom will radiate and let B equal an excitation cross-section defined as that area which, divided by unit area, gives the probability of absorption per unit length of path of a photon in a region wherein eligible absorbers have a density of one per unit volume. We let D equal the stimulated emission cross-section defined as that area which, divided by unit area, gives the probability of an induced emission per unit length of path of a photon in a region wherein excited atoms, which could if unexcited be eligible absorbers, have a density of one per unit volume. The emitted photon will, of course, move in the same direction as the photon inducing emission and will, therefore, have essentially the same Doppler shift away from the natural line shape. A is seen to be simply the Einstein coefficient of spontaneous emission. B and D are, however, not the transition probabilities in the standard textbook treatment, being a probability per unit length of path, rather than per unit length of time.

The term "eligible absorber" is used as in reference (1). A photon emitted from an atom with a velocity component c_ρ in the direction of motion, will require an absorber to have a velocity component along its line of sight near this value to make the photon appear to be within the limits of the natural line shape. If c'_ρ is the velocity component of the absorber it is assumed that $|c_\rho - c'_\rho| \lessgtr \Delta c/2$ and that the constant Δc is just the speed of an atom necessary to give a Doppler shift roughly equal to the width of the natural line shape.

In this paper a relationship will be established between the absorption of a photon either to the spontaneous emission of a photon somewhere else in the gas or to a radiation source such as incident radiation or reflection at the walls. For the time being let us consider only emission-absorption processes in the interior of the gas.

Suppose that we have at a point r in the gas an unexcited atom with velocity $\mathbf{c}$ at time t. As we have pointed out, all radiation reaching the neighborhood of r may be considered to have been spontaneously emitted at some source or to have been produced by stimulated emission which has been induced by a photon emitted farther away from the absorber and along the line of sight from the absorber to the emitter. Therefore, we look for a way to relate the intensity of radiation in the neighborhood of r to the distribution of excited states throughout the vessel. Because of the direct relationship of the rate of production of excited states to the intensity of absorbable radiation, we may determine directly a relationship between the rate of production of excited states about a point $\mathbf{r}$ to the density of excited states throughout the volume of gas. Therefore, at this point, we do not need to calculate an expression for the radiation field. Therefore, we may reduce the core of the problem to the derivation and solution of an equation for the velocity distribution function of excited states. Implicit in this remark is the fact that the time of flight of a photon is so short that we can consider the emission-absorption process instantaneous and, therefore, one may associate each photon with some atom in the gas. Let us use the symbol $p(r, \mathbf{c}, t)$ for the velocity-distribution function of excited states, previously referred to as $f_2(r, \mathbf{c}, t)$.

In reference (1) occulting was assumed negligible and we now continue under that assumption. Therefore, the probability that a photon emitted along ρ from an atom with velocity $\mathbf{c}$ will be absorbed in a length $d\rho$ will be B times the number of eligible absorbers in a volume element of unit cross-section and thickness $d\rho$. If we take the x and y directions as two directions mutually orthogonal with ρ the above probability is given by B times the integral

$$\int_{x}^{x+1}\int_{y}^{y+1}\int_{\rho}^{\rho+d\rho}\left\{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{c_\rho-(\Delta c/2)}^{c_\rho+(\Delta c/2)} f_1(\mathbf{r},\mathbf{c},t)\,dW\,dV\,dU\right\} d\rho\,dy\,dx\,. \tag{1}$$

The integration over the space variables is equivalent to multiplying by $d\rho$. Since Δc is very small the integration over W is equivalent to multiplying by Δc. Hence the integral reduces to

$$d\rho\Delta c\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} f_1(x,y,z,U,V,c_\rho,t)\,dV\,dU \tag{2}$$

and the probability per unit length of path may be expressed as

$$B\Delta c\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} f_1(x,y,z,U,V,c_\rho,t)\,dV\,dU\,. \tag{3}$$

Now let us think of stimulated emission as negative absorption since an induced emission yields a photon traveling in the same direction as the photon inducing the emission. First, let us consider the probability per unit length of path that a photon emitted along ρ by an atom with velocity $\mathbf{c}$ will induce an excited atom to emit. The derivation of this term is exactly the same as for absorption given by

$$D\Delta c\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} p(x,y,z,U,V,c_\rho,t)\,dV\,dU\,. \tag{4}$$

Thus, one may, considering the above as a negative probability of absorption, arrive at $Q(c_\rho)$ the total probability of absorption per unit length of path for a photon emitted from an atom with velocity component c_ρ in the direction of emission to be at a point r

$$Q(c_\rho) = \Delta c\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\left[Bf_1(r,U,V,c_\rho,t) - Dp(r,U,V,c_\rho,t)\right] dV\,dU\,, \tag{5}$$

which reduces to

$$Q(c_\rho) = \Delta c \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} [Bf(r, U, V, c_\rho) - (B + D)p(r, U, V, c_\rho, t)]\, dV\, dU \ . \tag{6}$$

Using the assumption that the gas is dynamically at a uniform steady state, the first of the integrals above may be evaluated directly, and the expression reduces to

$$Q(c_\rho) = nB\Delta c\, [m/(2\pi kT)]^{\frac{1}{2}} \exp[-mc_\rho^2/(2kT)] - (B + D)\Delta c \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} p(r, U, V, c_\rho, t)\, dV\, dU \ . \tag{7}$$

It may well be that the above *probability* of absorption is negative. Such a case would imply a cascading effect. If we define $P(\rho, c_\rho)$ to be the probability that a photon emitted from an atom with a velocity component c_ρ along the line of sight of the photon travels at least a distance ρ before re-capture we have[9]

$$\frac{1}{P}\frac{dP}{d\rho} = -Q(c_\rho) \tag{8}$$

or, on integrating

$$P(\rho, c_\rho) = \exp[-\int Q(c_\rho)\, d\rho] \ , \tag{9}$$

since $P(0, c_\rho)$ equals unity. The above may be written

$$P(\rho, c_\rho) = \exp\{-nB\Delta c\, [m/2\pi kT)]^{\frac{1}{2}} \rho \exp - [mc_\rho^2/(2kT)] + (B + D)\Delta c \int_0^\rho \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} p(r'', U, V, c_\rho, t)\, dV\, dU\, d\rho\} \ , \tag{10}$$

where $\mathbf{r}''$ is denoted with double primes to indicate that it is a variable point along the line of sight between some emitter at the point $\mathbf{r}'$ and an absorber at the point $\mathbf{r}$. Again, $P(\rho, c_\rho)$ may be greater than unity. Also, though it is not indicated in the argument of $P(\rho, c_\rho)$, the term is a function of both $\mathbf{r}$ and $\mathbf{r}'$ since $p(\mathbf{r}, \mathbf{c}, t)$ is a function of position.

The explicit time dependence of the distribution function $p(\mathbf{r}, \mathbf{c}, t)$ depends upon five processes:

(1) Spontaneous emission, expressed by $-Ap(\mathbf{r}, \mathbf{c}, t)$.

(2) Loss and gain of members caused by movement into and out of the space co-ordinate ranges by atoms with the required velocity co-ordinates, expressed by the usual second term in the Boltzmann equation, i.e., $-\mathbf{c} \cdot \nabla p(\mathbf{r}, \mathbf{c}, t)$.

(3) Absorption by unexcited atoms of photons emitted, either spontaneously or by induced emission, in other regions of the gas.

(4) Rate of increase of the excited states by impinging radiation.

(5) Rate of increase of the excited states due to the cumulative effect of driving mechanisms.

The number of excited atoms in a small neighborhood about $\mathbf{r}'$ capable of emitting photons which could, if emitted in the proper direction, be absorbed in the neighborhood of $\mathbf{r}$ by atoms whose velocity component along the vector $\rho = \mathbf{r} - \mathbf{r}'$ is in range $c_\rho \pm (\Delta c/2)$ is simply

$$d\tau_{R'} \oint p(\mathbf{r}', \mathbf{c}, t)\, d\tau_{v'} \qquad (11)$$

where the symbol $d\tau_{R'}$ means the volume element about $\mathbf{r}'$ and the integration in velocity space is between the limits $-\infty$ to ∞ in two mutually orthogonal directions, both to $\mathbf{r} - \mathbf{r}'$, and over $c_\rho \pm (\Delta c/2)$ in the direction of $\mathbf{r} - \mathbf{r}'$. The last integration is equivalent to multiplying by Δc. The above number of photons multiplied by A and divided by $4\pi\rho^2$ where $\rho = |\mathbf{r} - \mathbf{r}'|$ gives the rate of spontaneous emission of photons from the volume element which are in the proper direction to pass through a unit area about $\mathbf{r}$ and normal to ρ. This number multiplied by $P(\rho, c_\rho)$ gives the actual number of photons that do cross such a unit area. Next, the fraction of these photons that are absorbed per unit volume about $(\mathbf{r}, \mathbf{c})$ in a velocity range $d\mathbf{c}$ such that $\mathbf{c}$ has a component $c_\rho \pm (\Delta c/2)$ along ρ is $(Bf_1 - Dp)\, d\mathbf{c}$.

Therefore the total contribution to the explicit time dependence of $p(\mathbf{r}, \mathbf{c}, t)$ is

$$\frac{A\Delta c}{4\pi\rho^2} \oint_{R'} [Bf(\mathbf{c}) - (B+D)p(\mathbf{r}, \mathbf{c}, t)]\, P(\rho, c_\rho) \oint_{S_{v'}} p(\mathbf{r}', \mathbf{c}', t)\, dS_{v'}\, d\tau_{R'} \tag{12}$$

where $\mathbf{c}'$ is subject to the constraint that c'_ρ equals c_ρ and the velocity integration is over an infinite plane in velocity space, $S_{v'}$ normal to ρ.

The fourth process, the rate of change of the excited state distribution function, is arrived at in a manner similar to the one used in deriving (12) above. In most cases the impinging radiation will probably be taken to be uni-directional at some flat wall of the vessel or normal to some cylindrical or spherical shape. In any event the precise expression for the contribution to the excited states can not be written down, except simbolically, for the general case. However, considering the impinging radiation in the same way as radiation spontaneously emitted in the gas, and taking into account of $P(\rho, c_\rho)$ and the distribution functions in the neighborhood of $\mathbf{r}$ one can arrive at an expression similar to (12). We shall denote this term only symbolically as $F_4(\mathbf{r}, \mathbf{c}, t)$.

Similarly, the fifth process, the cumulative effect of driving terms can be represented only symbolically as $G(\mathbf{r}, \mathbf{c}, t)$ though for most physical problems G might take on a very simple form or perhaps equal zero.

Considering all five processes above our equation for the velocity distribution of excited states becomes

$$\frac{\partial p(\mathbf{r}, \mathbf{c}, t)}{\partial t} + \mathbf{c} \cdot \nabla p(\mathbf{r}, \mathbf{c}, t) + Ap(\mathbf{r}, \mathbf{c}, t) - F_4(\mathbf{r}, \mathbf{c}, t) - F_5(\mathbf{r}, \mathbf{c} t) =$$

$$= \frac{A\Delta cB}{4\pi\rho^2} \oint_{R'} [Bf(\mathbf{c}) - (B+D)p(\mathbf{r}, \mathbf{c}, t)]\, P(\rho, c_\rho) \oint_{S_{v'}} p(\mathbf{r}', \mathbf{c}', t)\, dS_{V'}\, d\tau_{R'}$$

where $\mathbf{c}'$ is subject to the restriction that $c'_\rho = c_\rho$ and $P(\rho, c_\rho)$ is given by (10). One is reminded that the terms F_4 and F_5 are only symbolic and must be arrived at independently for each new set of conditions. Also reflection, or partial reflection at the boundary is not considered in the above. However, the method for treatment of reflection should be clearly indicated by the above treatment.

It is obvious that the only feasible approach to solutions of the above is by numerical calculations on a high speed computer. Symmetry of a vessel is of no help since at every point off an axis or point of symmetry an integration over R' is not symmetric. Since the use of high-speed computers require funds not available to the authors, no illustrations of the method are given.

However, one and two dimensional analogies have been drawn and calculations are now being made with students having access to large-scale machines. Results of these calculations will be reported separately.

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RESUMEN

McIntyre y Fowler reportaron en Astrophysical Journal un método para estudiar la difusión de excitación por resonancia en un gas, sin tomar en cuenta los efectos de la emisión estimulada. Se consideró, además, que el número de estados sin excitación excedía grandemente el número de estados con excitación. En este trabajo, ambas restricciones se han suprimido. Se ve entonces como la probabilidad de emisión estimulada puede ser tratada como la probabilidad negativa de absorción y, por lo tanto, una relación directa se puede obtener entre el flujo de emisión espontánea en el gas y el flujo total de radiación.