RELAXATION OF THE VELOCITY AND POLARIZATION DISTRIBUTIONS OF EXCITED ATOMS DURING COMPLETE TRAPPING OF RESONANCE RADIATION

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An exact solution is obtained of the equation describing the trapping of resonance radiation in an infinite medium. The results are used to solve problems on the competition between modes with different polarizations in a gas laser.

1. In the course of trapping of resonance radiation a photon emitted by an excited atom is absorbed by another (unexcited) atom and the process occurs several times before the photon leaves the container filled with the gas. If the linear dimensions of the container are much greater than the photon mean free path the spontaneous emission of resonance photons has practically no effect on the number of excited atoms in the gas. Under these conditions one could speak of complete trapping of resonance radiation. An excited atom produced as a result of photon absorption has a velocity which, in general, is different from that of the atom which has radiated the photon. It follows that photon reabsorption gives rise to a change in the velocity distribution of excited atoms. There is an analogous redistribution over the Zeeman sublevels of the excited state. These processes are important in effects connected with interference between atomic states[1,2] and have an important influence on the parameters of gas lasers.[3,4]

Reabsorption of resonance photons is similar to atomic collisions in that it leads to a Maxwellization of the velocity distribution of excited atoms, to an equalization of Zeeman sublevel populations, and to destruction of the coherence of states at low pressures at which true atom collisions are still unimportant.

In the present paper we shall consider the change in an arbitrary velocity distribution of excited atoms and the distribution over the Zeeman sublevels in the case of complete trapping of resonance radiation. An exact solution of the integral equation describing the relaxation of the density matrix for the excited atoms will be obtained and the corresponding relaxation times will be found. Moreover, we shall discuss the possibility of applying these results to the study of polarization phenomena in luminescence and the competition between polarizations in the gas laser.

2. The equations for the density matrix $\hat{f}(\mathbf{v}, t)$ for excited atoms during complete trapping is of the form[2]

$$\frac{\partial \hat{f}(\mathbf{v}, t)}{\partial t} = -\gamma \hat{f}(\mathbf{v}, t) + \gamma(\hat{\mathscr{P}}\hat{f}), \tag{1}$$

where γ is the reciprocal of the excited-state lifetime and

$$(\hat{\mathscr{P}}\hat{f})_{mm'} = \int d^3\mathbf{v}' \sum_{m_1 m_1'} K_{m\,m'}^{m_1 m_1'}(\mathbf{v}, \mathbf{v}') f_{m_1 m_1'}(\mathbf{v}', t). \tag{2}$$

In this expression m and m′ identify the Zeeman sublevels of the excited states. The second term on the right-hand side of Eq. (1) describes the trapping of resonance radiation. The kernel in Eq. (2) is evaluated in[2] and is given by

$$K_{m\,m'}^{m_1 m_1'}(\mathbf{v}, \mathbf{v}') = F(v) v_0 \int d\Omega_n \exp(\mathbf{nv}/v_0)^2 S_{m\,m'}^{m_1 m_1'}(\mathbf{n}) \delta[\mathbf{n}(\mathbf{v} - \mathbf{v}')]. \tag{3}$$

In this expression

$$F(v) = \exp(-v/v_0)^2 / \pi^{3/2} v_0^3 \tag{4}$$

is the Maxwellian velocity distribution of normal atoms, $v_0 = (2kT/M)^{1/2}$,

$$S_{m\,m'}^{m_1 m_1'}(\mathbf{n}) = \frac{9}{8} \frac{1}{\pi^{3/2}} \frac{2j_1 + 1}{d^4} \sum_{\mu\mu'\sigma\sigma'} (\mathbf{e}_{n\sigma} \mathbf{d}_{m_1\mu'})^* (\mathbf{e}_{n\sigma} \mathbf{d}_{m\mu}) (\mathbf{e}_{n\sigma'} \mathbf{d}_{m_1'\mu'}) (\mathbf{e}_{n\sigma'} \mathbf{d}_{m'\mu})^*,$$

μ and μ' identify the Zeeman sublevels of the ground state, $\mathbf{d}$ is the dipole moment of a resonance transition,

$$d = (j_1 \| \mathbf{d} \| j_0) \tag{6}$$

is the reduced matrix element, j_1 and j_0 are, respectively, the total angular momenta of the upper and lower states, $\mathbf{n}$ is a unit vector, $\mathbf{e}_{n\sigma}$ is the polarization vector of the photon propagating in the direction of the unit vector $\mathbf{n}$, and the integration in Eq. (3) is carried out with respect to the angles of this unit vector.

Henceforth we shall be concerned with finding the "eigenoperators" $\hat{\varphi}(\mathbf{v})$ and the eigenvalue of the operator $\hat{\mathscr{P}}$, i.e., the solution of the equation

$$(\hat{\mathscr{P}}\hat{\varphi}) = (1 - \lambda)\hat{\varphi}. \tag{7}$$

The eigenvalues are denoted by $1 - \lambda$ so that $\hat{\varphi}(\mathbf{v}) e^{-\gamma\lambda t}$ is the solution of Eq. (1). It is clear that the general solution of Eq. (1) is a linear combination of such exponential relaxing solutions.

The operator $\hat{\mathscr{P}}$ is spherically symmetric, i.e., it commutes with the simultaneous rotation of the internal coordinates of the atom and of its velocity, so that the solution of Eq. (7) can be classified in accordance with the irreducible representations of the rotation group. An arbitrary density matrix for an atomic state with angular momentum j_1 can be expanded in terms of the irreducible tensor operators $\hat{T}_q^\kappa$, where κ lies between 0 and $2j_1$ and q between $-\kappa$ and κ. The operators $\hat{T}_q^\kappa$ transform as the eigenvectors of the angular momentum κ during the rotation of the internal coordinates of the atom. Since, in our case, the density matrix depends

on the atomic velocity v, the coefficients of its expansion in terms of $\hat{T}_q^K$ are functions of the spherical coordinates ϑ and φ of the velocity v so that they, in turn, can be expanded in terms of the spherical harmonics $Y_{LM}(\vartheta, \varphi)$.

It follows that, in its general form, the density matrix is the sum of the products of the operators $\hat{T}_q^k$ and the functions $Y_{LM}(\vartheta, \varphi)$ in which the expansion coefficients depend on the modulus of the velocity. To obtain the tensor operators which depend on the angles of the velocity, and which transform in accordance with the irreducible representations of the rotational group, it is clearly sufficient to combine κ and L. Consequently,

$$\hat{\Psi}_{JS}^{\kappa L}(\vartheta, \varphi) = \sum_{qM} (\kappa LJS | \kappa LqM) \hat{T}_q^{\kappa} Y_{LM}(\vartheta, \varphi), \tag{8}$$

will be operators of this kind, where $(\kappa LJS | \kappa LqM)$ are the Clebsch-Gordan coefficients. We shall take the normalization to be such that

$$(\hat{T}_q^{\kappa})_{m'm} = (-1)^{j_1 - m'} \sqrt{2\kappa + 1} \begin{pmatrix} j_1 & \kappa & j_1 \\ -m' & q & m \end{pmatrix}. \tag{9}$$

We then have the orthogonality relation

$$\sum_{mm'} (\hat{T}_q^{\kappa +})_{mm'} (\hat{T}_{q_1}^{\kappa_1})_{m'm} = \delta_{\kappa\kappa_1} \delta_{qq_1}, \tag{10}$$

from which it follows that the operators $\Psi_{JS}^{\kappa L}$ are orthogonal, i.e.,

$$\sum_{mm'} \int d\Omega [\hat{\Psi}_{JS}^{\kappa L +}(\vartheta, \varphi)]_{mm'} [\hat{\Psi}_{J_1 S_1}^{\kappa_1 L_1}(\vartheta, \varphi)]_{m'm} = \delta_{\kappa\kappa_1} \delta_{LL_1} \delta_{JJ_1} \delta_{SS_2} \tag{11}$$

The solution of Eq. (7) will be sought in the form

$$\hat{\varphi}(v) = \sum_{\kappa L} \varphi_{\kappa L}(v) \hat{\Psi}_{JS}^{\kappa L}(\vartheta, \varphi). \tag{12}$$

Since the operator $\hat{\mathscr{L}}$ commutes with inversion in velocity space, the sum in Eq. (12) should contain either only even or only odd momenta L. The equations for the coefficients $\varphi_{\kappa L}(v)$ can be obtained by substituting Eq. (12) in Eq. (7), multiplying both sides of the equation from the right by $\hat{\Psi}_{JS}^{\kappa L +}(\vartheta, \varphi)$, integrating with respect to the angles of the velocity and, finally, taking the trace over the magnetic quantum numbers m. After simple but laborious transformations we finally obtain

$$\sum_{\kappa' L'} A_{\kappa L}^{\kappa' L'}(J) \hat{K}_{LL'} \varphi_{\kappa' L'} = (1 - \lambda) \varphi_{\kappa L}. \tag{13}$$

In this expression $\hat{K}_{LL'}$, is an integral operator such that

$$\hat{K}_{LL'} \varphi_{\kappa' L'} = \frac{e^{-v^2}}{u} \int_{-u}^{u} dx\, P_L\left(\frac{x}{u}\right) e^{x^2} \int_{|x|}^{\infty} du'\, u' P_{L'}\left(\frac{x}{u'}\right) \varphi_{\kappa' L'}(u'), \tag{14}$$

where $u = v/v_0$, $u' = v'/v_0$, and $P_L(x/u)$ is the Legendre polynomial. The coefficients $A_{\kappa L}^{\kappa' L'}$ depend on the resultant angular momentum J but are independent of its projection S. They are given by

$$A_{\kappa L}^{\kappa' L'}(J) = \frac{1 + (-1)^{\kappa + \kappa'}}{2} \sum_{q=0,2} B_{\kappa L}^{q}(J) B_{\kappa' L'}^{q}(J), \tag{15}$$

where

$$B_{\kappa L}^{q}(J) = 3[(2j_1 + 1)(2\kappa + 1)(2L + 1)]^{1/2} \times \begin{Bmatrix} \kappa & 1 & 1 \\ j_0 & j_1 & j_1 \end{Bmatrix} \begin{pmatrix} \kappa & 1 & 1 \\ -q & 1 & q-1 \end{pmatrix} \begin{pmatrix} \kappa & L & J \\ -q & 0 & q \end{pmatrix}. \tag{16}$$

Equation (13) is still an integral equation in the modulus of the velocity. It can therefore be reduced to an algebraic equation as follows. Let

$$\Phi_n^L(u) = e^{-u^2} u^L L_{(n-L)/2}^{L+1/2}(u^2) \left[2\left(\frac{n-L}{2}\right)! \Big/ \Gamma\left(\frac{n+L+3}{2}\right)\right]^{1/2}, \tag{17}$$

where $L_n^{\alpha}(u^2)$ is the Laguerre polynomial. The kernel $\hat{K}_{LL'}$, has the following property:

$$\hat{K}_{LL'} \Phi_n^{L'} = b_n^L b_n^{L'} \Phi_n^L, \tag{18}$$

where

$$b_n^L = \pi^{1/4} \left[n!/2^{n+1} \left(\frac{n-L}{2}\right)! \Gamma\left(\frac{n+L+3}{2}\right)\right]^{1/2}. \tag{19}$$

This can be proved by using the formulas

$$\int_{|x|}^{\infty} dy\, e^{-y^2} y^{L+1} L_n^{L+1/2}(y^2) P_L\left(\frac{x}{y}\right) = \frac{(-1)^n e^{-x^2}}{2^{2n+L+1} n!} H_{2n+L}(x), \tag{20}$$

$$\int_{-u}^{u} dx\, P_L\left(\frac{x}{u}\right) H_n(x) = (-1)^{(n-L)/2} \pi^{1/2} u^{L+1} L_{(n-L)/2}^{L+1/2}(u^2)\, n! \Gamma^{-1}\left(\frac{n+L+3}{2}\right) \tag{21}$$

where $H_n(x)$ is the Hermite polynomial. The formula given by Eq. (21) is valid for $n \geq L$, where n and L have the same parity since, otherwise, the integration on the left-hand side of Eq. (21) is zero. Equations (20) and (21) can be proved with the aid of the Rodriguez representation of the Legendre polynomials and integration by parts.

The functions $\Phi_n^L(u)$ for given L and n = L, L + 2, L + 4, ... form a complete set of orthonormal functions. Therefore, by substituting

$$\varphi_{\kappa L}(v) = c_{\kappa L} \Phi_n^L(u) \tag{22}$$

where $c_{\kappa L}$ are numerical coefficients, and giving n the above values, we obtain all the solutions of Eq. (13). The coefficients $c_{\kappa L}$ must satisfy the following set of linear algebraic equations:

$$\sum_{\kappa' L'} A_{\kappa L}^{\kappa' L'}(J) b_n^L b_n^{L'} c_{\kappa' L'} = (1 - \lambda) c_{\kappa L}. \tag{23}$$

Therefore, the complete orthogonal set of solutions of Eq. (7) is made up of the operator functions

$$\hat{\varphi}_N(v) = \sum_{\kappa L} c_{\kappa L} \Phi_n^L(v/v_0) \hat{\Psi}_{JS}^{\kappa L}(\vartheta, \varphi), \tag{24}$$

where $\Psi_{JS}^{\kappa L}(\vartheta, \varphi)$ is an operator which depends on the angles of the velocity and is given by Eq. (8), while $\Phi_n^L(v/v_0)$ is given by Eq. (17). The eigenvalues λ are characterized by the quantum numbers n and J and the parity of the number κ. There is one further quantum number which corresponds to the number of the solution of Eq. (23). Each eigenvalue has a degeneracy of 2J + 1 with respect to the quantum number S. The index N denotes the set of all such quantum numbers which characterizes a given eigenfunction. The eigenfunction $\hat{\varphi}_N(v)$ corresponding to the quantum numbers n and J contains in Eq. (24) the values L of the same parity as n, and $L \leq n$. The possible values of κ and L in this sum are, in addition, restricted by the vector sum $\kappa + L = J$ for fixed parity of κ.

Let us now list the first few eigenfunctions and eigenvalues for the most interesting case, namely, $j_1 = 1$, $f_0 = 0$ (κ can then assume the values 0, 1, 2).

A. For n = 0 the number L can assume the single value L = 0; we then have $\kappa = J$ and the sum in Eq. (24) contains only one term for each J. The eigenvalues λ are 0, 1/2, 3/10 for j = 0, 1, 2, respectively, which is in agreement with the values obtained in[2] for a Maxwel-

lian velocity distribution of excited atoms. The three values of λ, in this case, characterize the population, orientation, and alignment decay of the excited state.

B. When n = 1 the number L can again assume only one value, namely, L = 1. The eigenfunctions with odd κ contain only $\kappa = 1$. For each J there is then only one eigenvalue λ ($\lambda = 1/2, 1, 4/5$ for J = 0, 1, 2). The eigenfunctions with the even κ include $\kappa = 0$ and $\kappa = 2$. The value J = 0, in that case, is not possible. When J = 1 there are two eigenvalues: $\lambda = 1$ and $\lambda = 3/5$. The corresponding eigenfunctions contain two terms each in Eq. (24) with the coefficients $c_{01} = 1/\sqrt{6}$, $c_{21} = -\sqrt{5/6}$, and $c_{01} = \sqrt{5/6}$, $c_{21} = 1/\sqrt{6}$. For J = 2 and 3 we have single eigenvalues, namely, $\lambda = 3/5$ and $\lambda = 57/70$, respectively. The sum in Eq. (24) then includes the single term with $\kappa = 2$, L = 1.

3. The above results enable us to consider the relaxation of the density matrix for excited atoms for any initial velocity and polarization distribution. In the case of a Maxwellian velocity distribution and complete trapping there are only two relaxation times, namely, for orientation and alignment. If the distribution of the excited atoms is not Maxwellian then, as shown in the present paper, the density matrix can be written in the form of a series, each term of which decays with its own relaxation time. The non-Maxwellian distribution in the case of optical excitation will appear if the spectral width of the exciting radiation does not exceed the Doppler line width. This situation is encountered quite frequently in luminescence experiments and is typical for laser problems.

In the presence of excitation, Eq. (1) must be augmented by the term F(v, t) which describes the entry of atoms into the state under consideration. We shall also take into account the possible decay of this state due to nonresonant transitions (for which there is no trapping) and will denote the corresponding contribution to the level width by γ'. Instead of Eq. (1) we then have

$$\frac{\partial \hat{f}(\mathbf{v},t)}{\partial t} = -(\gamma+\gamma')\hat{f}(\mathbf{v},t)+\gamma(\hat{\mathscr{D}}\hat{f})+\hat{F}(\mathbf{v},t). \tag{25}$$

$\hat{F}(v, t) = \hat{F}(v)$ in steady-state excitation. Again, in the steady state, the derivative on the left-hand side of Eq. (25) is zero. The solution of Eq. (25) can readily be found in the form of an expansion in terms of the orthogonal set of functions $\hat{\varphi}_N(v)$ defined above:

$$\hat{f}(\mathbf{v}) = \sum_N f_N \hat{\varphi}_N(\mathbf{v}). \tag{26}$$

To achieve this, let us expand $\hat{F}(v)$ in terms of the same functions, i.e.,

$$\hat{F}(\mathbf{v}) = \sum_N F_N \hat{\varphi}_N(\mathbf{v}) \tag{27}$$

and use Eq. (7). We then find that

$$f_N = F_N/(\gamma' + \lambda_N\gamma). \tag{28}$$

The coefficients F_N which characterize excitation can be found from Eq. (27) in the following form:

$$F_N = \mathrm{Sp}\int d^3\mathbf{v}\,\hat{\varphi}_N^{+}(\mathbf{v})\hat{F}(\mathbf{v})\exp(\mathbf{v}/\mathbf{v}_0)^2. \tag{29}$$

Let us begin by considering luminescence. The luminescence intensity integrated over the Doppler spectrum can be expressed in terms of the density matrix $\hat{f}(v)$ integrated over the velocities. In this integration the sum in Eq. (26) contains only terms with n = 0, and these correspond to the contribution of the Maxwell velocity distribution $\hat{f}(v)$. Thus, the polarization and angular distribution of luminescence integrated over the spectrum for any mode of excitation are such as if the excited-atom velocity distribution were Maxwellian. This is not, however, correct for each spectral interval individually.

4. Let us now consider the effect of trapping of resonance radiation on the polarization parameters of laser generation. The nature of the generation near the threshold is determined by the relation between the dipole moment and the laser radiation field:

$$p_q = \chi_q E_q, \tag{30}$$

where q = ±1 identifies the right- and left-handed circular components of the dipole moment p and field **E**, and the nonlinear polarizability χ_q is of the form

$$\chi_1 = a_1 - b_{1,1}|E_1|^2 - b_{1,-1}|E_{-1}|^2 \tag{31}$$

and similarly for χ_{-1}. The type of polarization in a laser without anisotropic elements is determined by the ratio of the coefficients $b_{qq'}$. When the cavity is accurately tuned to the center of the atomic transition the stability conditions for plane-polarized generation yield[5] $b''_{1,1} > b''_{1-1}$ ($b''_{qq'}$ is the imaginary part of $b_{qq'}$). In the opposite case, the circular polarization of the radiation is stable.

The coefficients $b_{qq'}$ depend on the angular momenta of the working levels. When $j_1 = 1$ and $j_0 = 0$ for the upper and lower state, respectively, and trapping and collisions are neglected, $b''_{1,1} = b''_{1,-1}$ so that the type of polarization remains undetermined. Nevertheless, Fork et al.[6] and de Lang[7] have reported stable circularly polarized generation due to the $2s_2 \rightarrow 2p_1$ transition in the He-Ne laser. It has been suggested[6,8,9] that this is connected with the effect of depolarizing collisions which are more effective in changing the orientation than alignment. Wang et al.[10] placed a laser in a weak magnetic field and observed the transition from circular to plane polarization when this field reached a critical value H_c. The magnitude of the critical field was related by these workers to the difference between collision cross sections representing the decay of orientation and alignment in the $2s_2$ state.

Under the conditions reported in[10], the trapping of resonance radiation from the $2s_2$ level was practically complete (the mean free path of a resonant photon at the line center was 0.03 mm at a partial pressure of 0.3 torr). It is therefore necessary to elucidate the influence of trapping of resonance radiation on the polarization parameters of a gas laser. We have therefore calculated the increments $\Delta b_{qq'}$ of the coefficients $b_{qq'}$ due to trapping. The calculations were performed as in[5], including in the equation for the density matrix of the upper working level a term corresponding to resonance radiation trapping.[4] It was found that we could confine our attention to terms with n = 0, J = 0, 1, 2 in the expansion in Eq. (26). As a result, we obtained the following expressions for the coefficients

$b_{qq'}$ (for accurate cavity tuning and the Doppler limit $kv_0 \gg \gamma$):

$$\Delta b_{1,1} = ig\frac{4\sqrt{\pi}}{9\gamma_1}\frac{\gamma_{10}}{kv_0}\left[\frac{1}{3}S_0 + \frac{1}{2}S_1 + \frac{1}{6}S_2\right], \quad (32)$$

$$\Delta b_{1,-1} = ig\frac{4\sqrt{\pi}}{9\gamma_1}\frac{\gamma_{10}}{kv_0}\left[\frac{1}{3}S_0 - \frac{1}{2}S_1 + \frac{1}{6}S_2 + \frac{\tilde{\gamma}_1\gamma_1(1-\lambda_2)}{(\gamma_1' + \lambda_2\tilde{\gamma}_1 - 2i\Omega)(\gamma_1 - 2i\Omega)}\right] \quad (33)$$

where g is the constant defined in[5], $\gamma_1 = \tilde{\gamma}_1 + \gamma_1'$ is the width of the upper working level (reciprocal of lifetime), $\tilde{\gamma}_1$ is the component of the width connected with transition to the ground state, k is the wave number of the working level, $\gamma_{10} = (\gamma_1 + \gamma_0)/2$, γ_0 is the width of the lower working level, and

$$S_J = \tilde{\gamma}_1(1-\lambda_J) / (\gamma_1' + \lambda_J\tilde{\gamma}_1), \quad (34)$$

where $\lambda_0 = 0$, $\lambda_1 = 1/2$, $\lambda_2 = 3/10$, Ω is the frequency separation between the Zeeman sublevels of the upper working state due to the magnetic field.

In zero magnetic field ($\Omega = 0$) we have $\Delta b''_{1,1} < \Delta b''_{1,-1}$ since $\lambda_1 > \lambda_2$. Hence, the trapping of resonance radiation itself ensures stable circular polarization of the laser radiation. The difference $b''_{1,1} - b''_{1,-1}$ increases with increasing magnetic field and, beginning with the critical field H_c, it becomes positive so that for $H > H_c$ plane polarization becomes stable. The expressions for $b_{qq'}$ were obtained in[5] without taking trapping into account (we shall denote them by $b^0_{1,1}$ and $b^0_{1,-1}$):

$$b^0_{1,1} = \frac{ig}{9}\left(\frac{1}{\gamma_1} + \frac{1}{\gamma_0}\right)\left(1 + \frac{\gamma_{10}}{\gamma_{10} - i\Omega}\right), \quad (35)$$

$$b^0_{1,-1} = \frac{ig}{9}\left[\frac{1}{\gamma_0}\left(1 + \frac{\gamma_{10}}{\gamma_{10} - i\Omega}\right) + 2\frac{\gamma_{10}}{\gamma_{10} - i\Omega}\frac{1}{\gamma_1 - 2i\Omega}\right]. \quad (36)$$

These formulas correspond to precise cavity tuning to the atomic transition frequency. Using Eqs. (32)–(36), we can find the critical field from the equation

$$\mathrm{Im}(b_{1,1} + \Delta b_{1,1}) = \mathrm{Im}(b_{1,-1} + \Delta b_{1,-1}). \quad (37)$$

If we assume that $\gamma_1 = 12 \times 10^7$ sec^{-1},[11] $\gamma_1' = 1.04 \times 10^7$ sec^{-1},[12] $\gamma_0 = 7 \times 10^7$ sec^{-1},[13] and that the g factor for the $2s_2$ level is 1.22,[14] we find from Eq. (37) that $H_c = 1$ G. The experimental value is about 2 G[10] and varies by roughly 20% in the pressure range 2–3 torr. It is clear that resonance radiation trapping has an important effect on the polarization parameters of a laser. When collision cross sections are calculated without taking this into account the result turns out to be too high. We note that, when Wang et al.[10] calculated the cross sections from the dependence of the critical field on pressure, they used the ratio of the relaxation times for orientation and alignment (5/3) reported in[15] which is too high (numerical calculations[16] show that this ratio is 1.1 in the case of the van der Waals interaction). This, in turn, led to underestimated cross sections.

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