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New quantum representation for treating recoil effects in non-linear spectroscopy

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Abstract. The problem of saturated absorption by a gas of two-level molecules, including recoil effects, is formulated in a new quantum representation for the molecular motion. The relationship to other representations is clarified in a simple chart. The new formulation requires the solution of simultaneous linear differential equations with mixed boundary conditions, instead of linear difference equations in two variables followed by an integration over velocity. Analytic solutions are presented for one running wave with Doppler line broadening, and for a weak probe beam in the Doppler limit. The latter lineshape is analysed to show when power broadening obscures the recoil splitting.

1. Introduction

Consider a molecule or atom with two long-lived internal states whose energies differ by $\hbar\omega_0$. Radiative transitions between these states are resonant when the angular frequency $\omega = cq$ of the external field is exactly equal to ω_0—provided the molecule is held fixed. But if the molecule has a finite mass M and is free to recoil, it will acquire a recoil velocity

$$V_r = \hbar q/M \tag{1.1}$$

to conserve momentum with the absorbed or emitted photon. To conserve energy also the absorption resonance must be shifted to $\omega = \omega_0 + \delta_r$, and the emission resonance to $\omega = \omega_0 - \delta_r$, where the recoil shift is

$$\delta_r = \hbar q^2/2M. \tag{1.2}$$

In atomic and molecular physics the recoil shift is very small, usually much smaller than spectral linewidths. However, the techniques of non-linear spectroscopy with laser sources have now permitted the observation of the recoil splitting in the 3·39 μm line of methane (Hall *et al* 1976), even though the relative splitting $2\delta_r/\omega_0$ is only 2×10^{-11}.

Even without recoil the theoretical analysis of non-linear interactions leading to very narrow Doppler-free lines becomes quite complicated (Haroche and Hartmann 1972, Shirley 1973). A pioneering attempt to include recoil effects was made by Kol'chenko *et al* (1969) using the Wigner representation. They obtained lineshapes for one travelling wave and for a standing wave in third order in the field strength.

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Stenholm (1974) used a momentum representation to formulate the problem with recoil as coupled recursion relations in two variables. There the absorption of a weak probe beam was treated and a discussion of the physics of its dependence on the velocity of the absorber was presented. Aminoff and Stenholm (1976) have solved the recursion relations numerically in the 'rate equation' approximation. The results show how power broadening can obliterate the recoil splitting, but can leave an asymmetry in the lineshape when the lifetimes of the two levels are unequal.

In all of this previous work the absorption is found first as a function of the velocity of the molecule and then integrated over a velocity distribution. The integration is always a problem, even when done numerically, since the integrand contains much more structure than the final result. In an effort to avoid this velocity integration, yet retain recoil effects, we have developed a new quantum representation for particle motion. In § 2 we formulate the saturated absorption problem in this representation.

Other representations are discussed in § 3 and their interrelationships clarified. The mathematical features encountered in solving the equations of our new formulation are discussed in § 4. The solution for one travelling wave is presented as an example of the method in § 5. Finally in § 6 we derive an analytic expression for the lineshape observed with a weak probe beam. We also discuss its interpretation in terms of overlapping, power-broadened resonances. Numerical solutions based on our new representation will be described in a following paper.

2. Development of equations of motion

For the Hamiltonian of a molecule we write three terms: the kinetic energy, the energy of the two internal states of interest, and the interaction energy with the electromagnetic field:

$$H = \hat{p}^2/2M + \tfrac{1}{2}\hbar\omega_0\sigma_3 - \mu E(\hat{z}, t)\sigma_1. \tag{2.1}$$

The internal structure of a two-level molecule is described by 2×2 σ matrices which obey the commutation rules of Pauli spin matrices. To treat the motion of the molecule quantum mechanically we also regard the position $\hat{z}$ and momentum $\hat{p}$ appearing in H as being operators obeying the commutation rule $[\hat{z}, \hat{p}] = \mathrm{i}\hbar$. For a plane-wave field only molecular motion in the z direction is important in the interaction. We shall not explicitly treat the other two dimensions.

Assume the externally applied electromagnetic field to consist of two monochromatic plane waves with amplitudes E_+ and λE_+ running in the positive and negative z directions respectively:

$$E(\hat{z}, t) = E_+ \cos(q\hat{z} - \omega t) - \lambda E_+ \cos(q\hat{z} + \omega t). \tag{2.2}$$

Let us rewrite the interaction energy μE_+ as $2\hbar bA$ where A is a dimensionless field amplitude and b is a coupling constant in frequency units (compare Shirley 1968, 1973). Then in the rotating wave approximation the interaction term becomes

$$-\hbar bA(\mathrm{e}^{\mathrm{i}q\hat{z}} - \lambda\ \mathrm{e}^{-\mathrm{i}q\hat{z}})\,\mathrm{e}^{-\mathrm{i}\omega t}\sigma^+ + \mathrm{HC}$$

where HC stands for Hermitian conjugate and $\sigma^\pm = \frac{1}{2}(\sigma_1 \pm \mathrm{i}\sigma_2)$, as usual.

We assume that the average behaviour of the assembly of molecules making up our absorbing medium is described by an ensemble-averaged density matrix ρ obeying the

equation of motion

$$\frac{\partial\rho}{\partial t}=\frac{1}{\mathrm{i}\hbar}[H,\rho]+\left(\frac{\partial\rho}{\partial t}\right)_{\text{relax}} \tag{2.3}$$

The commutator term fully describes the interaction of a freely moving molecule with the field. The additional term incorporates the average effect of relaxation processes. We shall use for it just simple decay terms with different decay rates for the different matrix components, plus a diagonal source term with a thermal velocity distribution:

$$\left(\frac{\partial\rho}{\partial t}\right)_{\text{relax}}=\frac{1}{\mathcal{N}}\begin{pmatrix}\Lambda_a & 0\\ 0 & \Lambda_b\end{pmatrix}\exp(-\hat{p}^2/2MkT)-\begin{pmatrix}\gamma_a\rho_{aa} & \gamma_2\rho_{ab}\\ \gamma_2\rho_{ba} & \gamma_b\rho_{bb}\end{pmatrix}, \tag{2.4}$$

where $\mathcal{N}=\mathrm{Tr}[\exp(-\hat{p}^2/2MkT)]$. As relaxations remove molecules from interaction with the field, the source term makes them available again with thermally redistributed velocities. In the absence of the external field the medium relaxes to a steady-state condition

$$\rho=\frac{1}{\mathcal{N}}\begin{pmatrix}\Lambda_a/\gamma_a & 0\\ 0 & \Lambda_b/\gamma_b\end{pmatrix}\exp(-\hat{p}^2/2MkT). \tag{2.5}$$

To proceed we adopt a configuration space representation for the molecular motion; that is, we take matrix elements of the equation of motion (2.3) between position eigenstates $\langle z_1|$ and $|z_2\rangle$ and write the result as a partial differential equation for the 2×2 matrix function $\rho(z_1,z_2)\equiv\langle z_1|\rho|z_2\rangle$:

$$\begin{aligned}\frac{\partial}{\partial t}\rho(z_1,z_2)=&\frac{\mathrm{i}\hbar}{2M}\left(\frac{\partial^2}{\partial z_1^2}-\frac{\partial^2}{\partial z_2^2}\right)\rho(z_1,z_2)-\frac{\mathrm{i}\omega_0}{2}[\sigma_3,\rho(z_1,z_2)]\\&+\mathrm{i}bA[(\mathrm{e}^{\mathrm{i}qz_1}-\lambda\,\mathrm{e}^{-\mathrm{i}qz_1})\sigma^+\rho(z_1,z_2)-\rho(z_1,z_2)(\mathrm{e}^{\mathrm{i}qz_2}-\lambda\,\mathrm{e}^{-\mathrm{i}qz_2})\sigma^+]\,\mathrm{e}^{-\mathrm{i}\omega t}\\&+\mathrm{i}bA[(\mathrm{e}^{-\mathrm{i}qz_1}-\lambda\,\mathrm{e}^{\mathrm{i}qz_1})\sigma^-\rho(z_1,z_2)-\rho(z_1,z_2)(\mathrm{e}^{-\mathrm{i}qz_2}-\lambda\,\mathrm{e}^{\mathrm{i}qz_2})\sigma^-]\,\mathrm{e}^{\mathrm{i}\omega t}\\&+\left[\frac{\partial}{\partial t}\rho(z_1,z_2)\right]_{\text{relax}}.\end{aligned} \tag{2.6}$$

The function $\rho(z_1,z_2)$ has the same transformation properties as the product of two one-particle wavefunctions $\psi(z_1)\,\psi^*(z_2)$. In two-particle problems it is often convenient not to work with z_1 and z_2, but with centre of mass and relative coordinates. In the same way, we find it more convenient to work with the sum and difference variables

$$Z=\tfrac{1}{2}(z_1+z_2)\qquad\text{and}\qquad z=z_1-z_2. \tag{2.7}$$

The sum variable corresponds to the average position of the molecule, while the difference variable describes the spread of the quantum mechanical wave packet associated with the molecule. In terms of these variables the kinetic energy term in (2.6) becomes the mixed partial derivative $(\mathrm{i}\hbar/M)\partial^2\rho(Z,z)/\partial Z\,\partial z$; while the source term contains

$$\begin{aligned}\mathcal{N}\mathcal{F}(z)&\equiv\langle Z+\tfrac{1}{2}z|\exp(-\hat{p}^2/2MkT)|Z-\tfrac{1}{2}z\rangle\\&=(4\pi)^{-1/2}(qu/V_{\mathrm{r}})\exp[-\tfrac{1}{4}(quz/V_{\mathrm{r}})^2],\end{aligned} \tag{2.8}$$

where $u=(2kT/M)^{1/2}$ is the most probable thermal speed and V_{r} is the recoil velocity defined in (1.1).

The standing wave part of the external field induces a spatially periodic structure in the polarization of the medium. This structure appears in the variation of ρ with the sum variable Z. We, therefore, expand this dependence in a Fourier series:

$$\rho(Z, z)=\sum_{\nu}\rho(\nu, z)\,\mathrm{e}^{\mathrm{i}\nu qZ}. \tag{2.9}$$

Writing out the matrix components explicitly we obtain the following infinite set of coupled linear first-order differential equations:

$$\begin{aligned}
&\left(\frac{\partial}{\partial t}+\gamma_a+\nu V_{\mathrm{r}}\frac{\partial}{\partial z}\right)\rho_{aa}(\nu, z)\\
&\quad=\Lambda_a\mathscr{F}(z)\delta_{\nu,0}+\mathrm{i}bA\left(\rho_{ba}(\nu-1, z)\,\mathrm{e}^{-\mathrm{i}\omega t}-\rho_{ab}(\nu+1, z)\,\mathrm{e}^{\mathrm{i}\omega t}\right)\mathrm{e}^{\mathrm{i}qz/2}\\
&\qquad-\mathrm{i}b\lambda A\left(\rho_{ba}(\nu+1, z)\,\mathrm{e}^{-\mathrm{i}\omega t}-\rho_{ab}(\nu-1, z)\,\mathrm{e}^{\mathrm{i}\omega t}\right)\mathrm{e}^{-\mathrm{i}qz/2}\\
&\left(\frac{\partial}{\partial t}+\gamma_b+\nu V_{\mathrm{r}}\frac{\partial}{\partial z}\right)\rho_{bb}(\nu, z)\\
&\quad=\Lambda_b\mathscr{F}(z)\,\delta_{\nu,0}-\mathrm{i}bA\left(\rho_{ba}(\nu-1, z)\,\mathrm{e}^{-\mathrm{i}\omega t}-\rho_{ab}(\nu+1, z)\,\mathrm{e}^{\mathrm{i}\omega t}\right)\mathrm{e}^{-\mathrm{i}qz/2}\\
&\qquad+\mathrm{i}b\lambda A\left(\rho_{ba}(\nu+1, z)\,\mathrm{e}^{-\mathrm{i}\omega t}-\rho_{ab}(\nu-1, z)\,\mathrm{e}^{\mathrm{i}\omega t}\right)\mathrm{e}^{\mathrm{i}qz/2}\\
&\left(\frac{\partial}{\partial t}+\gamma_2+\mathrm{i}\omega_0+\nu V_{\mathrm{r}}\frac{\partial}{\partial z}\right)\rho_{ab}(\nu, z)\\
&\quad=\mathrm{i}bA\left(\rho_{ab}(\nu-1, z)\,\mathrm{e}^{\mathrm{i}qz/2}-\rho_{aa}(\nu-1, z)\,\mathrm{e}^{-\mathrm{i}qz/2}\right)\mathrm{e}^{-\mathrm{i}\omega t}\\
&\qquad-\mathrm{i}b\lambda A\left[\rho_{bb}(\nu+1, z)\,\mathrm{e}^{-\mathrm{i}qz/2}-\rho_{aa}(\nu+1, z)\,\mathrm{e}^{\mathrm{i}qz/2}\right]\mathrm{e}^{-\mathrm{i}\omega t}\\
&\left(\frac{\partial}{\partial t}+\gamma_2-\mathrm{i}\omega_0+\nu V_{\mathrm{r}}\frac{\partial}{\partial z}\right)\rho_{ba}(\nu, z)\\
&\quad=-\mathrm{i}bA\left[\rho_{bb}(\nu+1, z)\,\mathrm{e}^{\mathrm{i}qz/2}-\rho_{aa}(\nu+1, z)\,\mathrm{e}^{-\mathrm{i}qz/2}\right]\mathrm{e}^{\mathrm{i}\omega t}.\\
&\qquad+\mathrm{i}b\lambda A\left[\rho_{bb}(\nu-1, z)\,\mathrm{e}^{-\mathrm{i}qz/2}-\rho_{aa}(\nu-1, z)\,\mathrm{e}^{\mathrm{i}qz/2}\right]\mathrm{e}^{\mathrm{i}\omega t}.
\end{aligned} \tag{2.10}$$

Note that since $\mathscr{F}$ is independent of the sum variable Z (the source is uniformly distributed in the medium), the source appears only in the $\nu=0$ equations.

For boundary conditions we shall require that $\rho(\nu, z)$ vanish as z goes to plus or minus infinity. That is, we assume the molecular motion is described by wave packets of finite spatial extent so that $\langle z_1|\rho|z_2\rangle$ goes to zero as $|z_1-z_2|$ goes to infinity. A momentum eigenstate does not obey this boundary condition, but our source term (2.8) does for any finite temperature (non-zero u). In equations (2.6) the inhomogenous source terms generate values for $\rho_{aa}(0, z)$ and $\rho_{bb}(0, z)$ of finite extent. Interaction with the field spreads these values among the other components of ρ with non-zero Fourier indices ν, but does not spread them to larger z values.

To find the influence of the medium on the applied field, we calculate the induced macroscopic polarization, which we use as a source in Maxwell's equations. The complex amplitude of the running wave polarization will be proportional to the expectation value

$$\langle\sigma^-\,\mathrm{e}^{\pm\mathrm{i}qz}\rangle=\mathrm{Tr}(\rho\sigma^-\,\mathrm{e}^{\pm\mathrm{i}qz})=\int\langle z_1|\rho_{ab}|z_1\rangle\,\mathrm{e}^{\pm\mathrm{i}qz_1}\,\mathrm{d}z_1=L\rho_{ab}(\nu=\mp1, z=0), \tag{2.11}$$

where L is the length of the medium traversed by the field†. Thus we do not need a complete solution of equations (2.10) for all ν and z. The attenuation coefficients or susceptibilities are obtained from just the two specific values in (2.11). This is the feature which makes our new representation attractive.

3. Relationship to other formulations

In the development of our equations of motion we could equally well have chosen to use a momentum representation for the molecular motion, writing

$$\rho(p_1, p_2) = \langle p_1|\rho|p_2\rangle.$$

For this representation the kinetic energy term in the equation of motion is just $-\mathrm{i}(2M\hbar)^{-1}(p_1^2 - p_2^2)\rho(p_1, p_2)$ and contains no derivatives. On the other hand, $\mathrm{e}^{\mathrm{i}q\hat{z}}$ is a translation operator in momentum space, so the interaction couples in $\rho(p_1 \pm \hbar q, p_2)$ and $\rho(p_1, p_2 \pm \hbar q)$. We thus obtain a set of difference equations in two variables, rather than differential equations.

Just as we did in coordinate space let us introduce sum and difference momentum variables:

$$P = \tfrac{1}{2}(p_1 + p_2) \qquad \text{and} \qquad p = p_1 - p_2, \tag{3.1}$$

where P describes the average momentum and p the spread in momentum for a molecule. Then the kinetic energy term becomes just $-\mathrm{i}Pp/M\hbar$. The source terms contain

$$\langle P + \tfrac{1}{2}p|\exp(-\hat{p}^2/2MkT)|P - \tfrac{1}{2}p\rangle = \exp(-P^2/M^2u^2)\delta(p). \tag{3.2}$$

Thus the source term generates a continuous range of values for P, but only the value zero for p. The interaction couples $\rho(P, p)$ to other ρ components whose arguments differ by integral multiples of $\hbar q$, hence only a discrete set of values for p will occur. We make this explicit by writing

$$P = Mv + \tfrac{1}{2}\mu\hbar q = M(v + \tfrac{1}{2}\mu V_{\mathrm{r}}) \qquad \text{and} \qquad p = \nu\hbar q, \tag{3.3}$$

with μ and ν integers and v a continuous velocity parameter varying only over a range V_{r}. Then the equations of motion for

$$\rho(\nu, \mu, v) \equiv \langle Mv + \tfrac{1}{2}(\mu + \nu)\hbar q|\rho|Mv + \tfrac{1}{2}(\mu - \nu)\hbar q\rangle \tag{3.4}$$

become coupled difference equations in μ and ν identical to equations (2.19)–(2.21) in Stenholm (1974). Our derivation is equivalent to the earlier one except we have not used second quantization for the molecules.

To calculate the polarization in the momentum representation we need to evaluate

$$\mathrm{Tr}(\rho\sigma^- \mathrm{e}^{\pm \mathrm{i}q\hat{z}}) = \int \langle p_1|\rho_{ab}|p_1 \pm \hbar q\rangle \, \mathrm{d}p_1 = \sum_{\mu} \int \rho_{ab}(\nu = \mp 1, \mu, v) M \, \mathrm{d}v. \tag{3.5}$$

That is, after solving the coupled difference equations one must perform an integral over the velocity parameter and a sum over the μ index. The total mathematical

† The normalization factor $\mathcal{N}$ is also proportional to L, hence our results will be independent of it.

problem becomes rather cumbersome with recoil included. However, it has been carried out (Aminoff and Stenholm 1976) when all components with $|\nu| > 1$ have been neglected (rate equation approximation).

The momentum representation has the advantage of automatically providing the Fourier expansion of the spatial dependence (ν index). It also provides the dynamics explicitly for each momentum class of molecules. In this form the physics of the interactions (Doppler and recoil shifts, multiphoton processes, etc) can best be understood (Haroche and Hartmann 1972, Stenholm 1974).

In quantum theory the position and momentum representations are related by Fourier transformations such that (z_1, p_1) and (z_2, p_2) are Fourier transform variable pairs. In the same way our sum and difference variables form the transform pairs (Z, p) and (z, P). Thus the momentum representation equations of Stenholm (1974) can be obtained from the position representation equations (2.10) by Fourier transforming both Z and z. If we transform just one of these variables we obtain a mixed representation in terms of the two sum variables or the two difference variables. What are these representations like?

The choice of the sum variables Z and P for a representation is tempting, since we expect them to become the classical position and momentum variables in the correspondence limit. In fact, the representation in terms of Z and P was proposed long ago by Wigner (1932) and an extensive literature has been built up on the methods of doing quantum mechanics in this representation (Moyal 1949). Kol'chenko *et al* (1969) used this representation in their early work on the recoil problem. However, the quantum mechanical features of the motion are not essential in their calculation. Their equations can be interpreted as those for a classical distribution in position and momenta with the added requirement that a molecule's velocity changes by V_r when its internal state changes. The Wigner formulation does assure one, however, that the off-diagonal components ρ_{ab} and ρ_{ba} should be assigned a classical velocity half way between that of the components ρ_{aa} and ρ_{bb} to which they are coupled by the interaction with the field.

Despite the appeal of classical understanding, the Wigner representation becomes cumbersome to solve with recoil included. Without recoil it reduces to the conventional formulation used in laser theory (Lamb 1964). The latter has been best solved by Fourier expanding the spatial dependence (Stenholm and Lamb 1969, Feldman and Feld 1970), which we now see corresponds to using the momentum variables P and p.

The representation used in this work is based on the difference variables† z and p. It is proportional to the characteristic function $\langle \exp[\mathrm{i}(z\hat{p} - p\hat{z})/\hbar] \rangle$ discussed by Moyal (1949). To our knowledge no one has previously used this representation. We have derived it from the position representation by Fourier expansion of the Z dependence. We could also have obtained it from the momentum representation by Fourier transformation of the P dependence. Since $P = Mv$ classically, our new formulation is mathematically just a Fourier transformation of the velocity dependence in conventional formulations. The detailed information found in the velocity dependence, such as hole burning, now appears in the z dependence of the solutions of (2.10). Our present derivation provides a physical interpretation of the new variable z.

The momentum representation can be related to our new representation in another way which reproduces the index μ in (3.3). Mathematically the differential equations

† Because we have assumed our source to be thermal and our interaction to be with a monochromatic external field, we have been able to restrict the values of p to discrete ones (index ν), using a Fourier series rather than integral in (2.9).

(2.10) are linear with periodic coefficients $e^{\pm iqz/2}$. Applying Floquet's theorem (Moulton 1958) the homogeneous solution has the form

$$\rho(v, z) = e^{ikz} \sum_{\mu} \rho(v, \mu, k)\, e^{i\mu qz/2}, \tag{3.6}$$

where k is a Floquet exponent. The new coefficients $\rho(v, \mu, k)$ then obey the same difference equations as $\rho(v, \mu, v)$ defined in (3.4) with $k = Mv/\hbar$. The exponent k is not fixed by the boundary conditions, but by the source term. When the inhomogeneous solution is constructed from (3.6) and the source function (2.8), an integral over real k values results, corresponding to the velocity integration in the momentum representation. The index μ then corresponds to a Fourier index in the expansion of the periodic coefficients in the Floquet solution of (2.10).

The four representations we have discussed are summarized in the chart below:

Fourier transform

Variables $z \leftrightarrow P$

Fourier transform $Z \updownarrow p$

Position representation	Wigner representation
Representation used in this paper	Momentum representation

Each representation depends on two variables, one chosen from each of the Fourier transform pairs (Z, p) and (z, P). The formulation of our problem can be converted from any one of the four representations to any other one by an appropriate Fourier transformation. Hence all are equally valid. The choice of representation for solving a problem can be based solely on mathematical convenience.

In the above scheme, the number of basic variables does not change when recoil effects are retained in the formulation, yet the problem becomes much more difficult mathematically. In the Wigner and momentum representations this occurs because the recoil associated with the interaction couples together the behaviour of molecules of different momentum classes, whereas, without recoil the equations for each momentum class are independent. In our present formulation the effect of recoil is to insert periodic coefficients into differential equations. This greatly complicates the analytic solution of the equations, but makes little difference when the equations are solved numerically. That is why we have chosen the present formulation for the numerical solutions to be reported in a following paper.

4. General features of solving the equations

4.1. Dimensionless variables

For algebraic and numerical convenience we shall rewrite our basic equations in terms of dimensionless variables and parameters. First we rewrite the density matrix as its

field-free steady state plus a correction to describe the interaction with the field:

$$\rho(\nu, z) = \begin{pmatrix} \Lambda_a/\gamma_a & 0 \\ 0 & \Lambda_b/\gamma_b \end{pmatrix} \mathcal{F}\delta_{\nu,0} - \frac{\bar{W}}{2V_r\mathcal{N}} \begin{pmatrix} \gamma_2 a_\nu & ibAy_\nu \, e^{-i\omega t} \\ -ibAz_\nu \, e^{i\omega t} & \gamma_2 b_\nu \end{pmatrix} \tag{4.1}$$

where $\bar{W} = \Lambda_a/\gamma_a - \Lambda_b/\gamma_b$ is the field-free steady-state population inversion. In the off-diagonal elements we have also factored out the amplitude and time dependence of the field. This permits our new variables to be time independent in steady state and to depend on the field amplitude only through the intensity parameter†

$$\alpha = 2b^2A^2/\gamma_1\gamma_2, \tag{4.2}$$

where

$$\gamma_1 = \gamma_a\gamma_b/\gamma_{ab} \qquad \text{and} \qquad \gamma_{ab} = \tfrac{1}{2}(\gamma_a + \gamma_b). \tag{4.3}$$

The factor $\bar{W}/2V_r\mathcal{N}$ in (4.1) has been so chosen that $y_{\pm 1}(0)$ give directly the ratios of the susceptibility χ or attenuation coefficient β to its unsaturated value in the Doppler limit (compare the notation of Shirley 1973)

$$\beta^+ = \beta_0 y_1(0), \qquad \beta^- = -\lambda^{-1}\beta_0 y_{-1}(0)$$

or (4.4)

$$\chi_1 = i\chi_0'' y_1(0), \qquad \chi_{-1} = -i\lambda^{-1}\chi_0'' y_{-1}(0).$$

We scale our system to the relaxation rate γ_2 (unsaturated homogeneous linewidth) by defining the relaxation rate ratios

$$r_i = \gamma_i/\gamma_2 \qquad (i = a, b, ab, 1), \tag{4.5}$$

the detuning parameter

$$\tan\psi = (\omega - \omega_0)/\gamma_2, \tag{4.6}$$

the recoil shift parameter

$$\epsilon = \delta_r/\gamma_2, \tag{4.7}$$

and the Doppler parameter

$$\eta = \gamma_2/qu. \tag{4.8}$$

Finally we replace the position variable z by

$$s = \gamma_2 z/V_r. \tag{4.9}$$

† Our intensity parameter α corresponds to the following parameters in other papers:

Holt (1970) and Shirley (1973)	α
Kol'chenko *et al* (1969)	$\frac{1}{2}\kappa$
Feldman and Feld (1970)	$\frac{1}{4}I$
Baklanov and Chebotayev (1971)	$\frac{1}{2}\chi$
Haroche and Hartmann (1972)	$\frac{1}{2}I^2$
Stenholm (1974)	$\frac{1}{2}\eta I_+$

The equations of motion (2.10) then become

$$\left(\nu\frac{\mathrm{d}}{\mathrm{d}s}+r_a\right)a_\nu = \tfrac{1}{2}r_1\alpha(y_{\nu+1}+z_{\nu-1})\,\mathrm{e}^{\mathrm{i}\epsilon s}-\tfrac{1}{2}r_1\lambda\alpha(y_{\nu-1}+z_{\nu+1})\,\mathrm{e}^{-\mathrm{i}\epsilon s}$$

$$\left(\nu\frac{\mathrm{d}}{\mathrm{d}s}+r_b\right)b_\nu = -\tfrac{1}{2}r_1\alpha(y_{\nu+1}+z_{\nu-1})\,\mathrm{e}^{-\mathrm{i}\epsilon s}+\tfrac{1}{2}r_1\lambda\alpha(y_{\nu-1}+z_{\nu+1})\,\mathrm{e}^{\mathrm{i}\epsilon s}$$

$$\left(\nu\frac{\mathrm{d}}{\mathrm{d}s}+1-\mathrm{i}\tan\psi\right)y_\nu \tag{4.10}$$

$$= 2\mathscr{G}\delta_{\nu,1}-2\lambda\mathscr{G}^*\delta_{\nu,-1}-a_{\nu-1}\,\mathrm{e}^{-\mathrm{i}\epsilon s}+b_{\nu-1}\,\mathrm{e}^{\mathrm{i}\epsilon s}+\lambda a_{\nu+1}\,\mathrm{e}^{\mathrm{i}\epsilon s}-\lambda b_{\nu+1}\,\mathrm{e}^{-\mathrm{i}\epsilon s}$$

$$\left(\nu\frac{\mathrm{d}}{\mathrm{d}s}+1+\mathrm{i}\tan\psi\right)z_\nu = 2\mathscr{G}\delta_{\nu,-1}-2\lambda\mathscr{G}^*\delta_{\nu,1}-a_{\nu+1}\,\mathrm{e}^{-\mathrm{i}\epsilon s}+b_{\nu+1}\,\mathrm{e}^{\mathrm{i}\epsilon s}+\lambda a_{\nu-1}\,\mathrm{e}^{\mathrm{i}\epsilon s}-\lambda b_{\nu-1}\,\mathrm{e}^{-\mathrm{i}\epsilon s}.$$

We have dropped the time derivative since we shall be concerned only with the steady-state solution in what follows. Our definition of a_0 and b_0 has transferred the source terms to the $\nu = 1$ equations in the form

$$\mathscr{G}(s) = (2\eta\sqrt{\pi})^{-1}(n_+\,\mathrm{e}^{-\mathrm{i}\epsilon s}-n_-\,\mathrm{e}^{\mathrm{i}\epsilon s})\exp(-s^2/4\eta^2) \tag{4.11}$$

$$= (2\eta\sqrt{\pi})^{-1}[\cos\epsilon s-\mathrm{i}(n_++n_-)\sin\epsilon s]\exp(-s^2/4\eta^2), \tag{4.12}$$

where

$$n_+ = \Lambda_a/\gamma_a\bar{W} \qquad \text{and} \qquad n_- = \Lambda_b/\gamma_b\bar{W}.$$

For $\nu = 0$ there is no derivative term, so a_0 and b_0 can be eliminated from (4.10) algebraically.

In any solution of equations (4.10) we shall always truncate the Fourier expansion (2.9) so that only a finite number of equations will be considered. We know from previous work without recoil (Shirley 1973, Holt 1970) that truncation gives a very good approximation for β or χ whenever the Doppler width well exceeds the saturation width. We expect this to remain true with recoil present.

4.2. The homogeneous solution

Mathematically equations (4.10) are linear differential equations with periodic coefficients (period $2\pi/\epsilon$). By Floquet's theorem (Moulton 1958) we know their homogeneous solution is of the form $\Sigma_i C_i(s)\exp(\xi_i s)$ where the ξ_i are constants and the C_i are periodic functions of s. But such a solution cannot simultaneously vanish at both plus and minus infinity no matter what the real part of ξ_i is. Hence the boundary conditions rule out the homogeneous solution, admitting only the solution generated by the inhomogeneous source term. (Compare the discussion in Aminoff and Stenholm (1976, appendix 3).) In seeking approximate solutions one should, therefore, always start with the source terms and then work away from them.

4.3. Symmetry conditions

According to quantum mechanics, the density operator is Hermitian:

$$\langle z_1|\rho_{\alpha\beta}|z_2\rangle^* = \langle z_2|\rho_{\beta\alpha}|z_1\rangle.$$

Since this property is preserved both by the interaction with the field and by $(\partial\rho/\partial t)_{\text{relax}}$,

we can apply it to derive the symmetry condition

$$\rho^*_{\alpha\beta}(\nu, z) = \rho_{\beta\alpha}(-\nu, -z). \tag{4.13}$$

For our dimensionless variables this becomes

$$\begin{aligned} a^*_\nu(s) &= a_{-\nu}(-s) \qquad & y^*_\nu(s) &= z_{-\nu}(-s) \\ b^*_\nu(s) &= b_{-\nu}(-s) \qquad & z^*_\nu(s) &= y_{-\nu}(-s). \end{aligned} \tag{4.14}$$

By utilizing this symmetry one need solve equations (4.10) only for the variables with positive ν index, or only for positive s values. In the latter case we evaluate the symmetry conditions at $s = 0$. We then use them as boundary conditions at $s = 0$ to replace those at minus infinity while still defining a unique solution for positive s. But since we are seeking $y_{\pm 1}$ at $s = 0$, the problem becomes one of finding those initial conditions at $s = 0$ which give a solution vanishing at plus infinity. The complete solution for all s is not needed. Numerically, the initial conditions can be found with reasonable accuracy, even though the numerically generated solution of the differential equations based on these initial conditions may be inaccurate.

4.4. Doppler limit

In calculations neglecting recoil much attention has been given to the Doppler limit case in which the Doppler width qu is much greater than the homogeneous linewidth γ_2. In the present formulation this limit alters the problem slightly. As η approaches zero, the source function $\mathcal{G}(s)$ approaches a delta function in s, independent of recoil. This source can be easily handled, however, by integrating the equations over an infinitesimal distance across the delta function. The functions $y_{\pm 1}$ and $z_{\pm 1}$ thereby experience discontinuities, while all other functions remain continuous. By incorporating half the discontinuity into the boundary conditions at $s = 0$, the boundary value problem for positive s alone remains well defined (has continuous solutions), but becomes one for homogeneous equations instead of inhomogeneous ones.

4.5. Recoilless limit

The recoil shift can be neglected in the present formulation by simply letting ϵ go to zero. The differential equations then have constant coefficients and can be solved by conventional techniques. By so doing we have reproduced several of the special solutions given by Shirley (1973). The amount of algebraic effort required in the two methods is comparable. Our new formulation offers no particular advantage in analytic calculation.

In the general case of a strongly saturated system the observables have to be evaluated numerically. Then our new formulation seems to offer advantages, because it can incorporate additional features without requiring any change in the method of solution. As we have seen, recoil is included merely through the introduction of periodic coefficients into the system of differential equations. Furthermore, the numerical accuracy is concentrated where we want it, in $y_{\pm 1}(0)$, and not in the values at large s.

5. Solution for one running wave

To illustrate a solution of the inhomogeneous equations (4.10) we consider only one

running wave to be present ($\lambda = 0$). In this case, examination of the equations (4.10) shows that they separate into independent sets of four equations each for the variables a_ν, b_ν, $y_{\nu+1}$, and $z_{\nu-1}$. The inhomogeneous terms appear only in the $\nu = 0$ set of equations, so only this set has a non-zero solution. Eliminating a_0 and b_0 algebraically the exponential factors cancel leaving the following two equations:

$$\begin{aligned}\left(\frac{\mathrm{d}}{\mathrm{d}s}+1-\mathrm{i}\tan\psi\right)y_1+\alpha(y_1+z_{-1})&=2\mathscr{G}\\ \left(-\frac{\mathrm{d}}{\mathrm{d}s}+1+\mathrm{i}\tan\psi\right)z_{-1}+\alpha(y_1+z_{-1})&=2\mathscr{G}.\end{aligned}\tag{5.1}$$

Equations (5.1) can be solved by the method of variation of parameters in the homogeneous solution. The result is

$$y_1=[(Q^{-1}-1)f_-(s)\,\mathrm{e}^{Qs}+(Q^{-1}+1)f_+(s)\,\mathrm{e}^{-Qs}]\,\mathrm{e}^{\mathrm{i}s\tan\psi}$$

and (5.2)

$$z_{-1}=[(Q^{-1}+1)f_-(s)\,\mathrm{e}^{Qs}+(Q^{-1}-1)f_+(s)\,\mathrm{e}^{-Qs}]\,\mathrm{e}^{\mathrm{i}s\tan\psi},$$

where $Q=(1+2\alpha)^{1/2}$. The new functions $f_\pm(s)$ are defined by the integrals

$$f_+(s)=\int_{-\infty}^{s}\mathscr{G}(t)\,\mathrm{e}^{(Q-\mathrm{i}\tan\psi)t}\,\mathrm{d}t$$

and (5.3)

$$f_-(s)=\int_{s}^{\infty}\mathscr{G}(t)\,\mathrm{e}^{-(Q+\mathrm{i}\tan\psi)t}\,\mathrm{d}t.$$

In this form we can see explicitly how the solution (5.2) goes to zero at $\pm\infty$ to meet the boundary conditions. We can also express $f_\pm(s)$ in terms of the error function of a complex argument:

$$\begin{aligned}f_+(s)&=n_+\exp(-\zeta_+^2)\left[1+\mathrm{erf}\left(\frac{s}{2\eta}+\mathrm{i}\zeta_+\right)\right]-n_-\exp(-\zeta_-^2)\left[1+\mathrm{erf}\left(\frac{s}{2\eta}+\mathrm{i}\zeta_-\right)\right]\\ f_-(s)&=n_+\exp(-\zeta_+^{*2})\left[1-\mathrm{erf}\left(\frac{s}{2\eta}+\mathrm{i}\zeta_+^*\right)\right]-n_-\exp(-\zeta_-^{*2})\left[1-\mathrm{erf}\left(\frac{s}{2\eta}+\mathrm{i}\zeta_-^*\right)\right].\end{aligned}\tag{5.4}$$

We have used the abbreviation $\zeta_\pm=(\tan\psi\pm\epsilon+\mathrm{i}Q)\eta$. Note that $f_+^*(s)=f_-(-s)$ in agreement with the symmetry condition (4.14). Using the symmetry at $s=0$ we can write the attenuation coefficient as

$$\beta=\beta_0[(2/Q)\,\mathrm{Re}\,f_+(0)+2\mathrm{i}\,\mathrm{Im}\,f_+(0)].\tag{5.5}$$

The dominant effect of recoil is the shifting of the absorption and emission Doppler lines relative to each other as shown by the exponentials in (5.4). This feature was mentioned in the work of Kol'chenko *et al* (1969). The separation of the lines is blurred when the homogeneous width Q is comparable to or larger than ϵ.

If we introduce the plasma dispersion function (Fried and Conte 1961) we can write

$$f_+(0)=(-\mathrm{i}/2\sqrt{\pi})(n_+Z(\zeta_+)-n_-Z(\zeta_-)).\tag{5.6}$$

Letting ϵ go to zero then recovers the solution without recoil given by Shirley (1973).

When η becomes small, $\mathrm{erf}(s/2\eta)$ approaches a step function, while $\zeta_\pm$ approach zero. In (5.4) the exponentials flatten while the two error functions merge. In the Doppler limit $f_\pm(0) = \frac{1}{2}$. The recoil effects disappear. The solution (5.4) shows in detail how the Doppler limit is reached.

6. Lineshape observed with a weak probe beam

6.1. Derivation of solution

When the oppositely running wave is weak we find an approximate solution by a perturbation series in λ about the one running wave solution. From the basic equations (4.10) we see that each set of variables a_ν, b_ν, $y_{\nu+1}$, $z_{\nu-1}$ is coupled in order λ to an adjacent set with ν index differing by two. Starting from the inhomogeneous terms the variables of the different sets are thus of order $\lambda^{|\nu|/2}$. To get the lowest-order value for the attenuation coefficient β^- of the weaker wave we solve the set of equations including y_{-1} to order λ:

$$\begin{aligned}
&\left(1 - i\tan\psi - \frac{d}{ds}\right) y_{-1} + a_{-2}\, e^{-i\epsilon s} - b_{-2}\, e^{i\epsilon s} = -2\lambda\mathcal{G}^* + \lambda(a_0\, e^{i\epsilon s} - b_0\, e^{-i\epsilon s}) \\
&\left(r_a - 2\frac{d}{ds}\right) a_{-2} - \tfrac{1}{2} r_1 \alpha (y_{-1} + z_{-3})\, e^{i\epsilon s} = -\tfrac{1}{2} r_1 \alpha \lambda z_{-1}\, e^{-i\epsilon s} \\
&\left(r_b - 2\frac{d}{ds}\right) b_{-2} + \tfrac{1}{2} r_1 \alpha (y_{-1} + z_{-3})\, e^{-i\epsilon s} = \tfrac{1}{2} r_1 \alpha \lambda z_{-1}\, e^{i\epsilon s} \\
&\left(1 + i\tan\psi - 3\frac{d}{ds}\right) z_{-3} + a_{-2}\, e^{-i\epsilon s} - b_{-2}\, e^{i\epsilon s} = 0.
\end{aligned} \tag{6.1}$$

We have omitted terms in y_{-3} since they are of order λ^3. If we insert the one running wave solution for a_0, b_0 and z_{-1} then the terms on the right-hand sides are all inhomogeneous terms of order λ.

We shall carry out the solution of (6.1) for positive s in the Doppler limit. From (5.2) we obtain

$$z_{-1} = (Q^{-1} - 1)g$$

and

$$a_0\, e^{i\epsilon s} - b_0\, e^{-i\epsilon s} = \left(\frac{\alpha}{r_{ab} Q}\right) g (r_b\, e^{2i\epsilon s} + r_a\, e^{-2i\epsilon s}) \tag{6.2}$$

where $g = \exp[(-Q + i\tan\psi)s]$. The inhomogeneous terms thus introduce two different oscillation frequencies into equations (6.1). The solutions contain both frequencies (exponentials) in the form

$$\begin{aligned}
y_{-1} &= \lambda(C\, e^{2i\epsilon s} + C'\, e^{-2i\epsilon s})g \\
a_{-2} &= \lambda(A\, e^{3i\epsilon s} + A'\, e^{-i\epsilon s})g \\
b_{-2} &= \lambda(B\, e^{i\epsilon s} + B'\, e^{-3i\epsilon s})g \\
z_{-3} &= \lambda(D\, e^{2i\epsilon s} + D'\, e^{-2i\epsilon s})g,
\end{aligned} \tag{6.3}$$

where A, B, C, D and their primed counterparts are constant coefficients found by substituting (6.3) and (6.2) into (6.1) and equating coefficients of like terms.

At $s=0$ we have $y_{-1}=\lambda(C+C'-1)$ where the last term arises from the Doppler limit discontinuity generated by the $\mathscr{G}^*$ term in (6.1). We then obtain the attenuation coefficient for the weaker wave from (4.4) as

$$\beta^- = \beta_0(1-C-C'). \tag{6.4}$$

In the absence of the stronger wave the inhomogeneous terms (6.2) vanish, hence C and C' vanish, and β^- reduces to the unsaturated value β_0. The non-linear, Doppler-free lineshape (Lamb-dip) of interest to spectroscopists is the change in attenuation due to the presence of the oppositely running wave. In the present case it is just proportional to $\mathrm{Re}(C+C')$.

The variables a_{-2} and b_{-2} are associated with spatial modulation of the level populations induced by the standing-wave part of the field. If we neglect these variables in (6.1) (rate equation approximation) we obtain directly

$$C = C_a \equiv (\alpha/r_{ab}Q)r_b/h_1$$

and (6.5)

$$C' = C'_a \equiv (\alpha/r_{ab}Q)r_a/h'_1$$

where

$$h_1 = 1+Q-2\mathrm{i}(\tan\psi+\epsilon) \qquad \text{and} \qquad h'_1 = 1+Q-2\mathrm{i}(\tan\psi-\epsilon). \tag{6.6}$$

The lineshape is then a superposition of two Lorentzian curves centred at $\omega=\omega_0\pm\delta_0$ with heights in the ratio γ_a/γ_b and power-broadened widths $\gamma_2(1+Q)/2$. This is the simplest case to solve which gives a power-broadened lineshape. The perturbation result of Kol'chenko *et al* is obtained from (6.5) and (6.6) by replacing Q with unity.

Unfortunately, when the relaxation rates γ_a, γ_b and γ_2 are of comparable size and power broadening is significant, the rate-equation approximation is not very good. Retaining all four equations (6.1) we find $C=C_a+C_r$ and $C'=C'_a+C'_r$ with

$$C_r = \tfrac{1}{2}r_1\alpha(Q^{-1}-1)\frac{h_3[h_a+(r_b/2r_{ab})(1+Q)(h_a+h_b)/h_1]}{h_1h_ah_bh_3+\frac{1}{2}r_1\alpha(h_a+h_b)(h_1+h_3)}. \tag{6.7}$$

The new h's are

$$h_a = r_a+2Q-2\mathrm{i}(\tan\psi+3\epsilon)$$

$$h_b = r_b+2Q-2\mathrm{i}(\tan\psi+\epsilon) \tag{6.8}$$

and

$$h_3 = 1+3Q-2\mathrm{i}(\tan\psi+3\epsilon).$$

C'_r is obtained from C_r by exchanging the subscripts a and b and reversing the sign of ϵ. When ϵ vanishes we recover equation (120) in the work of Shirley (1973). The effect of C_r is to reduce the peak of the Lorentzian in C_a, fatten its tail, and introduce a slight asymmetry shifting the maximum of C inward (see figure 1). The total lineshape from (6.5) and (6.7) agrees with the recent analytic result of Bordé (1976).

6.2. Discussion of the lineshape

In order to obtain a preliminary view of the results obtainable by the present method of

calculation, we have investigated the accuracy of the rate-equation approximation (REA) in the limit of a weak probe.

Due to recoil the lineshape acquires two separated peaks. For equal relaxation rates the curve remains symmetric, but from the earlier work (Aminoff and Stenholm 1976) we expect the REA result to overemphasize the doublet structure. This is, indeed, borne out by the results of figure 1 where REA is compared with the exact calculation. The same trend is observed in figure 2 where we have asymmetric peaks due to unequal relaxation rates $(\gamma_a/\gamma_b) = 3$. We observe, however, that the position of the strong peak remains the same also in the REA. When power is increased the two peaks tend to merge, but the maximum is only displaced slightly from the position of the stronger peak. This effect of power is seen in figure 3. This set of curves should be compared with figure 3 of Aminoff and Stenholm (1976).

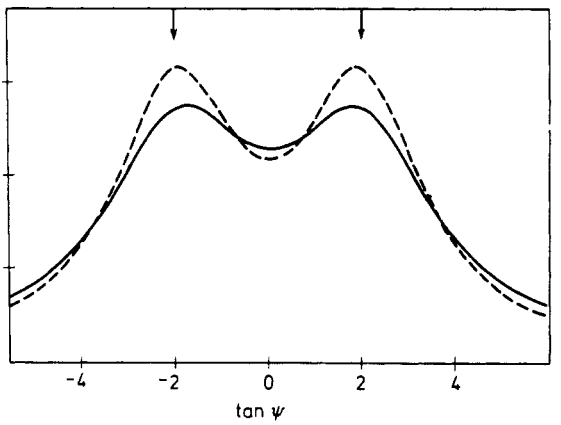

Figure 1. The lineshape of the response to a weak probe beam, as a function of the detuning parameter $(\omega - \omega_0)/\gamma_2 = \tan\psi$. The parameters are $\alpha = 2$, $\epsilon = 2$ and $(\gamma_a/\gamma_b) = 1$. The broken curve is the rate-equation result, the full curve is the complete solution. The arrows show the positions of the peaks in the low-intensity limit.

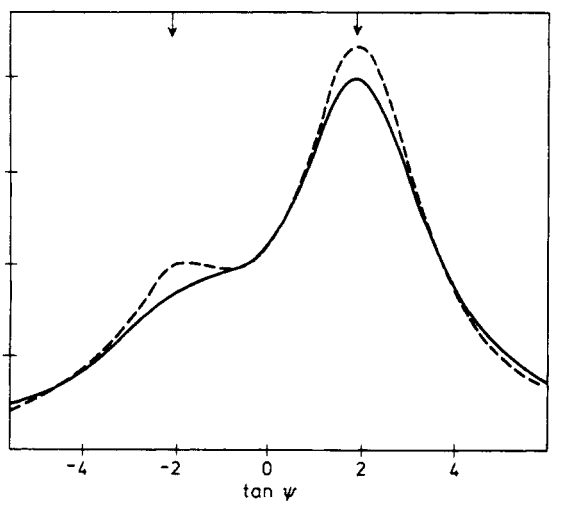

Figure 2. The same as figure 1, but with unequal relaxation rates, $(\gamma_a/\gamma_b) = 3$.

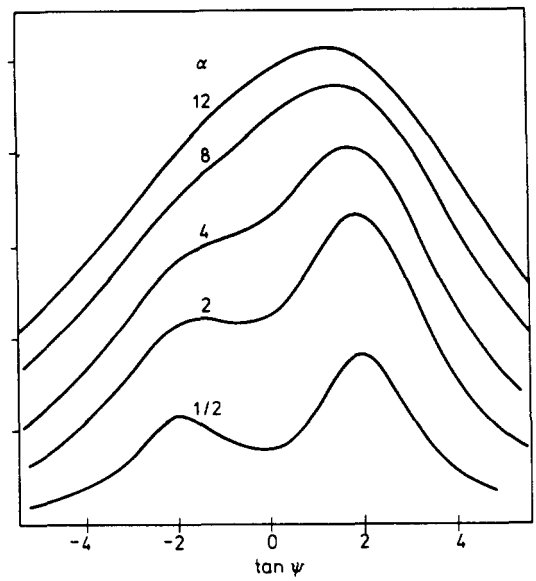

Figure 3. Lineshapes for unequal relaxation rates: $\gamma_a/\gamma_b = \frac{5}{3}$, $\gamma_{ab} = \gamma_2$, at various power levels, showing how power broadening obliterates the recoil splitting.

The earlier theory has been used to evaluate the resolvability of the recoil splitting. Here we carry out a similar comparison to see the difference between the REA and the exact theory. The criterion on resolvability we choose here is based on the fact that the curves in figure 1 can be reasonably well approximated by two Lorentzians of equal strengths and widths. If their centres are separated by 2ϵ the mathematical condition for a minimum to exist between the two Lorentzians is that the half-width w satisfies the condition

$$w < 3^{1/2}\epsilon \approx 1{\cdot}73\epsilon. \tag{6.9}$$

In a previous publication (Aminoff and Stenholm 1976) the criterion of a 1% dip was used. This corresponds to a condition $w < 1{\cdot}52\epsilon$. For convenience in handling numerical data, we adopt the criterion that the height of the line at the minimum is less than the height at a detuning equal to ϵ. This gives instead of (6.9) the condition

$$w < 2^{1/2}\epsilon \approx 1{\cdot}41\epsilon. \tag{6.10}$$

For the symmetric case, $w = 2^{1/2}\epsilon$ leaves a dip of 2·4% and the splitting between the two peaks is decreased by 32% due to overlap.

In figure 4 we show the power levels α which just obliterate the recoil doublet as a function of the recoil splitting. For the REA the width $w = \frac{1}{2}(1+Q) = 2^{1/2}\epsilon$ gives the upper curve

$$\alpha = 2(2)^{1/2}\epsilon(2^{1/2}\epsilon - 1) \tag{6.11}$$

(see equations (6.5)–(6.6)). The lower curve shows the effect of C_r and C_r' which increases the power broadening and thus reduces the power range over which the splitting is resolved. For unequal decay rates the curve lies between the two shown.

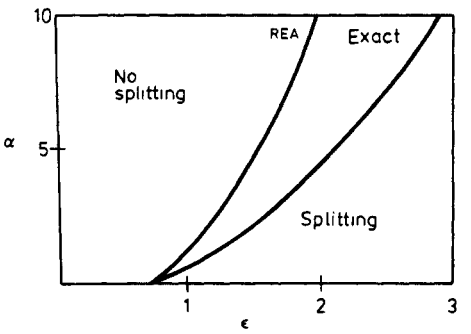

Figure 4. Strong beam intensities which just obliterate the recoil splitting in the probe beam lineshape as a function of recoil shift. The upper curve is the rate-equation result, valid for $\gamma_a = \gamma_b \ll \gamma_2$. The lower curve, valid for all relaxation rates equal, shows how much the region of resolved lines is reduced when population modulation processes are important.

Figure 4 should be compared with figure 6(a) of Aminoff and Stenholm (1976). Except for the exchange of axes they are seen to agree. This shows that the REA is not very useful in deciding resolvability questions, because, even if its average error is small, it gives an incorrect lineshape already for moderate power levels. This limits the usefulness of the calculation published earlier. We note, however, that the perturbation result by Kol'chenko *et al* gives only the point at $\alpha = 0$, where the two calculations coincide.

7. Conclusions

We have developed a new method to calculate the results of non-linear spectroscopy in Doppler-broadened media. This provides an alternative to the conventional method which just evaluates the response for a given velocity and only at the final stage averages out the velocity dependence. In the new method the velocity average is computed directly and the unobservable details of the velocity dependence never enter the calculations. Another advantage is that new features of the problem are easily introduced. This point is stressed in the present paper, where photon-induced recoil is taken into account. The method incorporates this feature easily, and some simple, analytically solvable cases are treated. A comparison between the present results and earlier ones is included.

The real advantage of the present scheme emerges when strong-signal theory with recoil is attempted. No other method has been able to give results for that case. Due to the complexity of the ensuing equations, the calculations have to be wholly numerical. The approach has, however, been made to work, and the details will be reported in a forthcoming publication.

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