Forward scattering (polarisation spectroscopy) of resonance radiation in partially coherent fields

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Abstract. Previous theoretical treatments of polarisation spectroscopy (forward scattering) are generalised so as to include laser temporal coherence effects. It is shown that for the general case it is impossible to describe the signal obtained in terms of birefringence and dichroism. The generalisation of the concept of the polarisability tensor is given together with the approximate expression for the signal in terms of it. The simple example of the $J_g = 0 \rightarrow J_e = 1$ transition in the forward scattering experiment is discussed in detail. Comparison is made between the signal lineshapes obtained for the phase diffusion model and the chaotic field model. The necessity of taking into account the statistical properties of the laser light in the forward scattering (polarisation spectroscopy) experiment is pointed out.

1. Introduction

Polarisation spectroscopy (PS), a very efficient method of high-resolution laser spectroscopy, has been developed from saturation spectroscopy (Wieman and Hänsch 1976). It was shown, however, (Gawlik and Series 1979, Nakayama et al 1980) that optical pumping, not saturation, is essentially responsible for the signals obtained. Gawlik and Series (1979) pointed out a strong physical resemblance between PS and forward scattering (FS) spectroscopy, developed many years earlier (Corney et al 1966, Durrant 1972, Laloe 1971). Recently it has been shown experimentally by Gawlik et al (1982) that with increased power of the probe beam in PS one can obtain resonances narrower than the natural lifetime, with a width partially limited by the laser bandwidth. Therefore, it is interesting to investigate the influence of the laser temporal coherence on signals obtained in PS and FS.

Usually the signal in such an experiment has been described by the polarisability tensor or what is equivalent by the refractive indices. The expression for the signal could then be decomposed into two parts, one corresponding to dichroism and the second to birefringence induced in the sample. It turns out that such a description is incorrect in the general case for partially coherent light; it may serve sometimes as a good approximation only.

The theory which takes into account the laser temporal coherence effects is given in § 2. Section 3 deals with the generalised (in order to cover the partially coherent case) polarisability tensor. In § 4 the simplest example of the $J_g = 0 \rightarrow J_e = 1$ transition in the FS case is discussed. Particular attention is paid to the difference in the theoretical

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lineshapes due to the different models of the laser light fluctuations. The phase
diffusion model (PD) as well as the chaotic field model (CF) is used. The validity of
the 'decorrelation approximation' (introduced in § 3) is investigated.

2. Notation, expression of the signal

The scheme of the experiment is presented in figure 1. A partially coherent, polarised
laser beam L (directed along OZ) interacts with atoms in the cell (or as in figure 1 in
the perpendicular atomic beam). Atoms are perturbed either by the magnetic field
B (FS) or by the counter-propagating laser beam (PS, shown by a broken arrow in
figure 1) or both. This perturbation introduces an asymmetry into the scattering
process and it is possible to observe a non-zero signal after a crossed analyser is put
directly in the way of the beam L. In PS the intensity of the beam is usually small.
We shall not restrict ourselves to such a case, as recent experiments show that when
L is strong enough to influence the dynamics of the atoms (L ceases to be a probe
beam in the normal sense) one gets very interesting resonances (Gawlik et al 1982).
In FS the most interesting effects occur for a strong laser beam.

We will use the following notation (shown in figure 1). P is the polariser unit
vector, A is the analyser unit vector, $E^{(+)}$ is the positive frequency part of the electric

![Figure 1.](image-url)

Figure 1. The set-up for forward scattering and polarisation spectroscopy experiments.
The atoms in the beam are irradiated by the laser light L (full arrow). They are influenced
by the longitudinal magnetic field B (FS) or by the counter-propagating laser beam (broken
arrow). P, polariser; A, analyser.)
field $E^{(+)} = |E^{(+)}|$ and $(\frac{1}{2}\pi - \varphi)$ is the angle between $P$ and $A$. The part of $E^{(+)}$ which enters the photomultiplier (PM) consists of $E^{(+)R}$, which is scattered by atoms and $E^{(+)0}$, which is the part of the incident light which passes without interaction with the atoms. The intensity measured at point $z_0$ (figure 1) is:

$$L_F(z_0, t) = \langle(E^{(-)}_0(z_0, t) + E^{(-)}_R(z_0, t)) \cdot A(E^{(+)}_0(z_0, t) + E^{(+)}_R(z_0, t)) \cdot A \rangle_{av} \tag{1}$$

which for small $\varphi$ becomes

$$L_F = (\varphi + \theta)^2(E^{(-)}_0 E^{(+)}_0)_{av} + (\varphi + \theta)(E^{(-)}_0 E^{(+)}_R)_{av} + (E^{(-)}_R E^{(+)}_0)_{av} + (E^{(-)}_R E^{(+)}_R)_{av} \tag{1a}$$

where $\theta$ is the imperfection of the polarisers. In FS one measures $L_F(z_0, t)$ as a function of the magnetic field $B$, whereas in PS it is measured as a function of the laser frequency $\omega_L$. We restrict ourselves to the one-photon transition, we assume that the laser is tuned near the resonance with the $|J_g\mu \rangle \rightarrow |J_e m \rangle$ transition, where $|m\rangle \langle \mu \rangle$ are magnetic sublevels of the upper (lower) level. $E^{(+)}_R$ scattered from $N$ atoms is a sum of contributions $E^{(+)}_R(i\mu)$, which are proportional to the electric dipole operator $D^{(+)}(i\mu)$ of the $i$th atom (Mollow 1969, Ackerhalt and Eberly 1974); all calculations are carried out in the Heisenberg picture. In terms of atomic operators $\sigma^{(l)}_{\mu \mu}$ is given by

$$d^{(l)}_{\mu \mu} = \langle iJ_{\mu \mu} | D^{(+)}(i\mu) | -\infty \rangle | J_e m \rangle \tag{3}$$

We assume that the sample of atoms under consideration is optically thin. That means that we neglect the influence of the light scattered by one atom on the dynamics of the other atoms. This assumption which has been frequently made (Laloe 1971, Kaftandjian et al. 1979) together with the known property of the light scattered in the forward direction—equal optical paths for scattering from atoms in different positions allows us to neglect the position dependence in (2) (Durrant 1972). Thus, we obtain for the measured intensity:

$$L_F(z_0, t + z_0/c) = (\varphi + \theta)^2(E^{(-)}_0 E^{(+)}_0)_{av} - iC(\varphi + \theta)N \sum_{m\mu} \langle(E^{(-)}_0(t) \sigma^{(1)}_{\mu \mu}(t))_{av} d^{(l)}_{\mu \mu} \cdot A - HC \rangle$$

$$+ C^2 N(N - 1) \sum_{m\mu, m' \mu'} A \cdot d^{(l)}_{m\mu} d^{(l)}_{m' \mu'} \cdot A \langle \sigma^{(1)}_{m\mu}(t) \sigma^{(2)}_{m' \mu'}(t) \rangle_{av}$$

$$+ C^2 N \sum_{m\mu, m' \mu'} |d^{(l)}_{m\mu}|^2 \langle \sigma^{(1)}_{m \mu}(t) \rangle_{av} \tag{4}$$

where we have used an equality $\sigma^{(1)}_{m \mu} \sigma^{(1)}_{m' \mu'} = \delta_{m \mu} \sigma^{(1)}_{m \mu}$. (Milonni 1976) and have assumed that all the atoms are of the same kind. The last term in (4), which is responsible for the Hanle signal is negligible in comparison with the other terms as $E^{(+)}_0 \gg E^{(+)}_R$, and for large $N$. For the lateral scattering only this term exists, as there is no $E^{(+)}_0$ field passing to the detector and the third term would be smeared out due to the negative interference. The first term in (4) is not interesting as it gives the constant background, the second term, which will later be called the 'linear' term, as it is proportional to $N$, is due to the positive interference between the incident and the scattered light.
The third term, proportional to $N^2$, the 'square' term, results from the light scattered by different atoms. By choosing $\varphi = -\theta$ we can have nothing but the square component; if $\varphi \neq -\theta$ then, as $E_0^{(r)} \gg E_R^{(r)}$, only the linear term is important.

Let us now concentrate on the meaning of the $\langle \ldots \rangle_{av}$ symbol. It is an average over the Maxwell distributed atomic velocities, over statistical properties of the light and over the density matrix of the system:

$$\rho(t) = \rho(0) = \rho^{(1)}(0) \otimes \rho^{(2)}(0) \otimes \ldots \otimes \rho^{N}(0)$$

where $\rho^{(i)}(0)$ is the density matrix of the $i$th atom. We denote the three successive averaging processes as $\langle \ldots \rangle_D$, $\langle \ldots \rangle_L$ and $\langle \ldots \rangle_p$ so that

$$\langle \ldots \rangle_{av} = \langle \langle \ldots \rangle_D \rangle_L \rangle_p$$

and the order of the averaging is arbitrary. Denoting for the sake of brevity $\langle \langle A \rangle_D \rangle_p = \bar{A}$ from (4) we get:

$$L_F(z_0, t + z_0/c) = iC(\varphi + \theta)N \sum_{m\mu} \langle \langle \bar{\sigma}_{m\mu}^{(1)}(t)E_0^{(+)}(t) \rangle_L d_{m\mu} \cdot A - \langle \langle E_0^{(-)}(t)\bar{\sigma}_{m\mu}^{(1)}(t) \rangle_L d_{m\mu} \cdot A \rangle + C^2N^2 \sum_{m\mu} \sum_{m'\mu'} A \cdot d_{m\mu} A \cdot d_{\mu'm'} \langle \langle \bar{\sigma}_{m\mu}^{(1)}(t)\bar{\sigma}_{\mu'm'}^{(2)}(t) \rangle_L$$

where we have used the fact that due to the form of $\rho(0)$:

$$\langle \langle \sigma_{ab}^{(1)}\sigma_{cd}^{(2)} \rangle_p \rangle = \langle \langle \sigma_{ab}^{(1)} \rangle_p \langle \sigma_{cd}^{(2)} \rangle_p$$

and the same obviously holds for $\langle \ldots \rangle_D$. We have also omitted the Hanle and the background terms. We point out that, similar to (7), decorrelation for $\langle \ldots \rangle_L$ is not possible. This important point will be discussed in the following sections.

According to (6) we need to know $\langle \ldots \rangle_L$ over atomic operators in order to calculate the signal. The interaction of atoms with the occupied modes of the radiation field is governed by the Hamiltonian (in the rotating-wave approximation)

$$\mathcal{H} = \sum_{i=1}^{N} H_i = \sum_{i=1}^{N} \left( H_{at}^{(i)} - \sum_{m\mu} (\sigma_{m\mu}^{(i)}d_{m\mu} \cdot E^{(+)}(t) + HC) \right)$$

through the set of Heisenberg equations of motion:

$$\dot{\sigma}_{ab}^{(i)} = -i\hbar [\mathcal{H}, \sigma_{ab}^{(i)}] + \hat{\Gamma}(\sigma_{ab}^{(i)}).$$

In (8) $E^{(+)}$ is the total electric field acting on the atoms, $E^{(+)} = E_0^{(+)} + E_C^{(+)}$, $E_C^{(+)}$ is the electric field of the counter-propagating laser beam (see figure 1). $\hat{\Gamma}(\sigma_{ab}^{(i)})$ is a linear superoperator added to include the spontaneous emission (Letokhov and Chebotayev 1977); its form is determined by the configuration of the investigated levels (Cohen Tannoudji 1977). An influence of the magnetic field is included in $H_{at}^{(i)}$. Such a Hamiltonian leads us to the generalised Bloch equations for $\sigma_{ab}^{(i)}$ (see an example in § 4).

3. The polarisability tensor

Many authors have expressed the FS (PS) signal in terms of the polarisability tensor (Gawlik and Series 1979, Durrant 1972, Laloe 1971, Kaftandjian et al 1979) defined
as the 'constant' (depending on $E$ for non-linear processes) of proportionality between the electric dipole moment $\mathcal{P} = \langle D \rangle_D$ induced in the atom, and the electric field which induces that moment:

$$\langle \mathcal{P}^{(+)}(t) \rangle_i = \sum_k \alpha_{ik} \langle E^{(+)}(t) \rangle_k$$  \hspace{1cm} (10)

where the subscripts $i, k$ denote, for example, spherical basis components of the $\mathcal{P}$, $E$ vectors. That description is convenient because one can associate the classical optical phenomena, such as dichroism and birefringence, induced in the sample with various components of the signal. In this section we want to answer two questions:

(i) Is (10) valid for fluctuating laser fields? It is possible to generalise (10) so that it can describe fields which fluctuate?

(ii) Can one express the signal in terms of the $\alpha_{ik}$ components thus making possible the classical interpretation?

To answer (i) let us refer back to (10). If $E^{(+)}$ is a fluctuating quantity then $\mathcal{P}^{(+)}$ also fluctuates. However, in order to express the signal with $\alpha_{ik}$ components, $\alpha_{ik}$ should not fluctuate and, on the other hand, should contain information about the stochastic behaviour of the process. Therefore (10) cannot be used directly for calculating $\alpha_{ik}$ because in (10) the polarisability tensor is defined in terms of fluctuating instantaneous fields instead of the averaged fields. These requirements can be fulfilled by using

$$\langle E_{0i}^{(-)}(t) \mathcal{P}_i^{(+)}(t) \rangle_L = \sum_k \alpha_{ik} \langle E_{0i}^{(-)}(t) E_k^{(+)}(t) \rangle_L$$  \hspace{1cm} (10a)

as a definition of $\alpha_{ik}$ instead of (10). The left-hand side can again be expressed in terms of $\langle \sigma_{ab} \rangle_D$:

$$\langle E_{0i}^{(-)}(t) \mathcal{P}_i^{(+)}(t) \rangle_L = -iCN \sum_k \alpha_{ik} \sum_{\mu\nu} (d_{\mu\nu})_k \langle E_{0i}^{(-)}(t) \sigma_{\mu\nu}^{(+)}(t) \rangle_L.$$  

If we assume some known stochastic process for the laser field we can obtain $\alpha_{ik}$ exactly in the same way as for the coherent field providing we know the $\langle E^{(-)} \sigma_{\mu\nu} \rangle$ averages. (Then we have to average $\alpha_{ik}$ over Maxwell-distributed velocities, but this can be done at the very end of the calculation as usual.)

Looking at the signal (6) we can immediately answer the second question. The definition of the polarisability involves only one-atom averages over light fluctuations. Therefore it is clear that the square term containing two-atom averages can by no means be expressed exactly by the polarisability. It would be possible if we could decorrelate these averages, for example:

$$\langle \tilde{\sigma}_{\mu\nu}^{(1)}(t) \tilde{\sigma}_{\mu\nu}^{(2)}(t') \rangle_L = \langle \langle E_0^{(-)}(t) E_0^{(+)}(t) \rangle \rangle^{-1} \langle \tilde{\sigma}_{\mu\nu}(t) E_0^{(+)}(t) \rangle_L \langle E_0^{(-)}(t) \tilde{\sigma}_{\mu\nu}(t) \rangle_L.$$  \hspace{1cm} (11)

Such a form of the decorrelation has some advantages:

(a) it allows for the signal interpretation in terms of dichroism and birefringence;

(b) it is obviously correct for the perfectly coherent case;

(c) it is also correct in first-order perturbation theory.

The major drawback of the decorrelation is that it is unjustified in general and its range of validity must be restricted. We will refer to (11) as the decorrelation approximation (DA). An example below shows what difference it produces for the phase/amplitude fluctuations of the laser field.
4. Forward scattering on the $J_e = 0 \rightarrow J_e = 1$ transition

In this section we discuss the theoretical lineshapes in the experiment (figure 1) with the atomic beam (to avoid the Doppler effect) and with a longitudinal magnetic field. The mean frequency of the $\sigma$-polarised laser beam $\omega_L$ is tuned near the resonance with the $J_e = 0 \rightarrow J_e = 1$ transition. The laser couples the ground state $|J_e, 0\rangle$ (denoted by $|0\rangle$) with the excited sublevels $|J_e, \pm 1\rangle$ (denoted by $|+\rangle$ and $|-\rangle$). We have therefore an effective three-level system (figure 2). In our case the atomic part of the Hamiltonian (8) takes the form of

$$H_{at}^{(i)} = \sum_{m=-1,1} \sigma_{mm}^{(i)}(\omega_0 + m\omega_B)$$

(12)

where $\omega_B$ is the Larmor precession frequency (Zeeman splitting) and $\omega_0$ is the frequency of the transition for $B = 0$. The relative phases of the excited sublevels with respect to ground state $|0\rangle$ will be chosen in such a way that

$$d_{+0} \cdot P = d_{-0} \cdot P = +id.$$

Then

$$d_{+0} \cdot A = -d(1 + i\varphi)$$
$$d_{-0} \cdot A = d(1 - i\varphi).$$

From (8), (9) and (12) we obtain generalised Bloch equations for our system (in the rotating frame)

$$\sigma_{++}^{(1)} = -\Gamma \sigma_{++}^{(1)} + dE(t)\sigma_{++}^{(1)} + dE^*(t)\sigma_{0+}^{(1)}$$
$$\sigma_{+-}^{(1)} = -\Gamma \sigma_{+-}^{(1)} + dE(t)\sigma_{+-}^{(1)} + dE^*(t)\sigma_{0-}^{(1)}$$
$$\sigma_{+0}^{(1)} = (2i\omega_B - \Gamma)\sigma_{+0}^{(1)} + dE(t)\sigma_{+0}^{(1)} + dE^*(t)\sigma_{0+}^{(1)}$$
$$\sigma_{-+}^{(1)} = -(2i\omega_B + \Gamma)\sigma_{-+}^{(1)} + dE(t)\sigma_{-+}^{(1)} + dE^*(t)\sigma_{0-}^{(1)}$$
$$\sigma_{00}^{(1)} = [(\omega_0 - \omega_L + \omega_B)i - \gamma]\sigma_{00}^{(1)} - dE^*(t)(\sigma_{++}^{(1)} + \sigma_{+-}^{(1)}) - \sigma_{00}^{(1)}$$

(13)

$$\sigma_{0-}^{(1)} = [(\omega_0 - \omega_L - \omega_B)i - \gamma]\sigma_{0-}^{(1)} - dE^*(t)(\sigma_{++}^{(1)} + \sigma_{+-}^{(1)}) - \sigma_{0-}^{(1)}$$
$$\sigma_{0+}^{(1)} = [-(\omega_0 - \omega_L + \omega_B)i - \gamma]\sigma_{0+}^{(1)} - dE(t)(\sigma_{++}^{(1)} + \sigma_{+-}^{(1)}) - \sigma_{0+}^{(1)}$$
$$\sigma_{00}^{(1)} = \Gamma(\sigma_{++}^{(1)} + \sigma_{++}^{(1)}) - dE(t)(\sigma_{++}^{(1)} + \sigma_{++}^{(1)}) - dE^*(t)(\sigma_{00}^{(1)} + \sigma_{00}^{(1)})$$

where $\Gamma$ and $\gamma$ are longitudinal and transversal relaxation terms respectively, $E(t)$ is the amplitude of the positive frequency part of the electric field:

$$E^{(+)}(t) = E(t) \exp(-i\omega_L t).$$

4.1. The linear signal case

We have to calculate:

$$-i \sum_m \langle E^*(t)\sigma_{mm}^{(1)}(t) \rangle L d_{0m} \cdot A - \langle \sigma_{mm}^{(1)}(t)E(t) \rangle L d_{m0} \cdot A.$$

(14)

As models for the laser light fluctuations we take either the phase diffusion model (PD) or the chaotic field model (CF).
4.1.1. For the PD model where \( E(t) = E_0 \exp(-i\phi(t)) \) and a Gaussian phase fulfils \( \langle \phi(t)\phi(t') \rangle_L = 2\Gamma_\Delta \delta(t-t') \) it is well known (Wódkiewicz 1979) that the exact equations for the averages can be obtained from (13) by making the substitutions

\[
\gamma \rightarrow \tilde{\gamma} = \gamma + \Gamma_L \quad \sigma^{(1)}_{\mu m}(t) \rightarrow \langle \exp(i\phi(t))\sigma^{(1)}_{\mu m}(t) \rangle_L \quad \sigma^{(1)}_{mm'}(t) \rightarrow \langle \sigma_{mm'}(t) \rangle_L.
\]

For the resonant case \( \omega_L = \omega_0 \) in the stationary limit we obtain, after some tedious algebra, a rather complicated formula for the signal:

\[
L^\text{PD}_F(\omega_B) = 2CN(\varphi + \theta)[Y(\omega_B, \Omega^2) + (\varphi + \theta)X(\omega_B, \Omega^2)]
\]  

(15)

where

\[
X(\omega_B, \Omega^2) = \Omega^2 \frac{2\Gamma\Omega^2 + \tilde{\gamma}(\Gamma^2 + 4\omega_B^2)}{(\tilde{\gamma}^2 + \omega_B^2)(\Gamma^2 + 4\omega_B^2) + \Omega^2[10\tilde{\gamma}\Gamma^2 + 8\omega_B^2(3\tilde{\gamma} - \Gamma)]\Gamma^{-1} + 16\Omega^4} 
\]

(15a)

\[
Y(\omega_B, \Omega^2) = \omega_B\Omega^2 \frac{\Gamma^2 + 4\omega_B^2 - 4\Omega^2}{(\tilde{\gamma}^2 + \omega_B^2)(\Gamma^2 + 4\omega_B^2) + (\Omega^2/\Gamma)[10\tilde{\gamma}\Gamma^2 + 8\omega_B^2(3\tilde{\gamma} - \Gamma)] + 16\Omega^4} 
\]

(15b)

and

\[
\Omega^2 = |E_0|^2 d^2.
\]

4.1.2. For the CF model we assume that the amplitude \( E(t) \) obeys a one-dimensional complex Markov process described by the Langevin equations with \( \delta \)-correlated random Gaussian forces (Louisell 1973, Zoller 1979). To calculate the signal (14) we follow the 'marginal characteristic function approach' proposed by Zoller. For details see Zoller (1979), Zoller et al (1981) and the appendix.

After some algebra we obtain the solution for the signal in terms of the continued fraction. An explicit expression for the signal is given in the appendix because of its complicated form.
4.1.3. We present the linear signal lineshapes \( (L_F) \) for various laser strengths and spectral bandwidths in figures 3-6. The continued fraction (A.2) had to be truncated for some \( M \) in the numerical calculations. The accuracy was assessed for \( \Gamma_L = 0 \), for which the convergence should be worse and the exact result can be easily evaluated (Zoller 1979). For our purposes \( M = 15 \) was enough to obtain the assumed accuracy.

The lineshapes in figures 3–6 are composed of two dispersion-shaped resonance curves. The wider resonance curve takes its origin in optical coherences and for small...
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Figure 5. Same as figures 3 and 4, $\Omega = 1$ and $\Gamma_L = 5$. The laser linewidth broadens the optical and narrows the Zeeman resonance curves, compare with figure 4.

Figure 6. Same as figures 3–5, $\Omega = \sqrt{10}$ and $\Gamma_L = 1$. In the same range of $\omega_B$ only the inner resonance is left due to the power broadening. For such a laser strength the power broadening is much bigger for PD (broken curve) than for CF.

intensities it is the only one which appears since it results from the first-order perturbation theory. With increasing intensity a second resonance curve, inverted in comparison with the ‘optical’ resonance curve, appears in the centre. At the same time the ‘optical’ resonance curve becomes power broadened (figure 3). The second resonance is due to the Zeeman coherence $\sigma_{-+}^{(1)}$ induced in the upper level and it also broadens with increasing intensity (figure 4). Power broadening for the PD model is more effective than for the CF model. The laser linewidth broadens the ‘optical’ and narrows the ‘Zeeman’ resonance curves (figures 4 and 5). It is caused by the fact that
with a larger laser spectral width the spectral density is distributed over a wider spectral range, so that the ‘Zeeman’ resonance curve is smaller in amplitude and less power-broadened.

4.2. The square component case

Let us assume now that \( \varphi = -\theta \) so we can measure the square component only. For our case (figure 2) the signal (6) reduces to:

\[
L_E(\omega_B) = C^2 N^2 \sum_m \sum_{m'} (d_{m0} \cdot A)(d_{0m} \cdot A)(\hat{\sigma}^{(1)}_{m0}(t)\sigma^{(2)}_{0m}(t))_L.
\]

(16)

The main problem in obtaining solutions for \( \langle \hat{\sigma}^{(1)}_{m0}(t)\hat{\sigma}^{(2)}_{0m'}(t) \rangle_L \) averages is the number of equations involved. For the most simple \( J_e = 0 \rightarrow J_e = 1 \) transition we have eight equations for one-atom averages \( \langle \sigma^{(1)}_{ab} \rangle_L \). If we want to write equations for two-atom averages \( \langle \sigma^{(1)}_{ab} \sigma^{(2)}_{cd} \rangle_L \) the number of equations grows to 72. (This number can be reduced a little due to the symmetry properties of our problem but it still remains very large.) Thus, it would be very convenient to apply the ‘decorrelation approximation’ (DA), §3; its range of validity is however uncertain, except for the very low intensities. We will show how DA works for both the phase and the amplitude fluctuations.

4.2.1. For the PD model we write equations for \( \langle \sigma^{(1)}_{ab} \sigma^{(2)}_{cd} \rangle_L \) (denoted for the sake of brevity as \( \langle 1; 2 \rangle \)) and for the corresponding \( \langle 1 \rangle \langle 2 \rangle \) averages and compare coefficients in these equations. Without the DA we have to write equations for \( \sigma^{(1)}_{ab} \sigma^{(2)}_{cd} \) and average them according to general rules (Wódkiewicz 1979). In the DA we obtain the required equations directly from the averaged set of equations which are the equivalent to (14). The only difference between the two sets of equations obtained lies in the damping coefficients for the averages involving two optical coherences coupled together. \( \langle \sigma_{m0} \sigma_{0m'} \rangle_L \) is damped with a rate \(-2\gamma\) whereas the corresponding element in DA, \( \langle \sigma_{m0} \exp(-i\varphi) \sigma_{0m'} \exp(i\varphi) \rangle_L \), is damped with a rate \(-2(\gamma + \Gamma_L)\). \( \langle \sigma_{m0} \exp(-i\varphi) \rangle_L \langle \sigma_{m0} \exp(-i\varphi) \rangle_L \) is damped with a rate \(-2(\gamma + \Gamma_L)\) in DA, whereas \( \langle \sigma_{m0} \sigma_{0m} \exp(-2i\varphi) \rangle_L \) and \( \langle \sigma_{0m} \sigma_{0m} \exp(2i\varphi) \rangle_L \) have a damping coefficient \(-2(\gamma + 2\Gamma_0)\). As other coefficients in the differential equations are the same it is clear that the DA works for \( \Gamma_L \ll \gamma \). For the radiation damping only, \( \gamma = \frac{1}{2} \Gamma \), so the laser linewidth must be much smaller than the natural decay constant of the upper level. When \( \gamma \) is large (e.g. when collisions are taken in to account in the phase-interrupting model (Wódkiewicz 1979) the laser bandwidth can exceed \( \Gamma \) without violating the DA.

Both sets of equations have been solved numerically (in the stationary limit) and the resulting lineshapes are shown in figures 7 and 8. It turns out that for \( \Gamma_L > \gamma \) the differences between the exact and DA solutions are significant. The amplitudes of the ‘Zeeman’ resonance curves are different for both methods but the positions and the widths are practically the same. On the other hand the widths of the ‘optical’ resonance curves are quite different for both methods (see figure 7). This difference will be smaller for atoms in the cell if the Doppler effect is taken into account.

4.2.2. For the CF model the situation is more difficult to handle for arbitrary laser bandwidth. However, for \( \Gamma_L = 0 \) the exact solution can be obtained easily by averaging the perfectly coherent solution over the Glauber P distribution of the intensities,
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Figure 7. The square component of the signal in the PD model. The broken curve corresponds to the exact solution, the full curve to the DA result. The resonance curves are symmetrical around \( \omega_B = 0 \); only the part corresponding to \( \omega_B > 0 \) is plotted. The right-hand side (b) shows the whole signal, the left-hand side (a) the (enlargened) region in the vicinity of \( \omega_B = 0 \) only. The values of the parameters are \( \Omega = \sqrt{10} \) and \( \Gamma_L = 1 \).

Figure 8. The square signal in the PD model. The curves are symmetrical around \( \omega_B = 0 \). (a), the signal for \( \Omega = 10 \) and \( \Gamma_L = 1 \), whereas (b) corresponds to \( \Omega = 10 \) and \( \Gamma_L = 5 \). Both (a) and (b) have independent intensity units. The narrowing of the Zeeman resonance curve can be seen as in the linear case.

\begin{align*}
\langle 1 \rangle_p (R^2) d R^2 &= \frac{1}{\Omega^2_L} \exp(-\Omega^2/(\Omega^2_L)) d \Omega^2.
\end{align*}

Thus

\begin{align*}
L_p^\Delta(\omega_B) &= \frac{N^2 C^2}{\Omega^2_L} \int d \Omega^2 P(\Omega^2) Y(\omega_B, \Omega^2).
\end{align*}

(Glauber 1965):
and
\[ L^{CF}_F(\omega_B) = N^2 C^2 \int d\Omega^2 P(\Omega^2) \frac{Y^2(\omega_B, \Omega^2)}{\Omega^2} \] (18)

where \( L^{DA}_F \) and \( L^{CF}_F \) denote the signal obtained with and without the DA. \( Y(\omega_B, \Omega^2) \) is given by (15b). Both the integrals in (17) and (18) can be expressed in terms of the exponential integrals \( Ei(x) \) or calculated numerically. The results are presented in figures 9–11. For small intensities all three resonance curves are similar to the squared dispersive curves. The slight difference between the coherent and the chaotic cases is well approximated by the DA. As for the linear case it is the ‘optical’ resonance which is responsible for the shape in figure 9. For larger intensities the second, ‘Zeeman’ resonance curve arises in the centre of the picture. It is clearly distinguishable for the coherent case as well as for the DA case giving rise to the three-minima structure of the lineshape. Only slight differences can be seen between the coherent and the DA cases in the widths and amplitudes of the curves. The exact result, without the DA solution, is entirely different. For pure radiation damping the power broadening of the outer optical resonance curve is so small that the three-minima structure vanishes, leaving one wide resonance only. The only trace of the ‘Zeeman’ resonance can be seen on the slopes of the wide resonance curve figure 10. For larger \( \gamma \) the optical resonance broadens and the three-minima structure appears again in figure 11; we believe that the same happens when the Doppler effect is taken into account.

5. Conclusions

We have formulated the theory of forward scattering (polarisation spectroscopy) valid for partially coherent, arbitrarily strong laser fields. It turns out that in general the signal cannot be described in terms of dichroism and birefringence as in the perfectly coherent case. In other words one cannot speak about the global properties of the sample and use concepts inherited from classical optics. The square component of the signal can be calculated using the decorrelation approximation only for lasers
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Figure 10. The same as figure 9. The values of parameters are $\Omega = \sqrt{10}$, $\Gamma_L = 0$ and $\gamma = 0.5$. The one-minimum curve corresponds to the exact solution, without the DA.

Figure 11. The square signal for $\gamma = 2.5$, $\Omega = \sqrt{10}$ and $\Gamma_L = 0$. The full curve and the broken curve correspond to the DA solution and to the exact solution respectively.

well described by the PD model and if the laser bandwidth is smaller than the transverse damping constant of the atoms investigated. The amplitude fluctuations make the DA inapplicable when we are interested not only in positions of the resonances but also in their shapes. Although the results presented above were checked for the particular, simple, case we believe that they are quite general, e.g. if the Maxwell distribution of the atomic velocities is taken into consideration. We discussed in detail only the forward scattering (there was no counter-propagating pump beam) but similar results should be obtained in PS as the pump beam plays a similar role in PS as the magnetic field does in the FS.

It turns out that the signal in the FS (and, we think, in PS) is very sensitive to the statistical properties of the laser light. The proper interpretation of the experimentally obtained curves should therefore contain information about the laser light statistics. On the other hand, the FS experiment can serve as a useful tool for obtaining information about the laser light statistics. Because of its relative experimental simplicity it may be in this sense competitive with the resonance fluorescence or multiphoton ionisation experiments.
In calculations concerning the chaotic field model we directly follow the method developed by Zoller (Zoller 1979, Zoller et al 1981). The system of equations (13) to be solved belongs to the class described by equation (37) of Zoller et al (1981). The averages \( \langle E^*(t)\sigma_{\mu n}(t)\rangle_L, \langle \sigma_{\mu n}(t)E(t)\rangle_L \) can be expressed as

\[
d\langle E^*(t)\sigma_{\mu n}(t)\rangle_L = (d\langle \sigma_{\mu n}(t)E(t)\rangle_L)^* \\
= \langle (\Omega^2_L)^{1/2}(\exp(i\varphi(t))(\Omega^2(t))^{1/2}L_0^{\dagger}(\Omega^2(t))/(\Omega^2_L)^{1/2}L_0\rangle_L \sigma_{\mu n}(t) \rangle_L
\]  

(A.1)

where \( L_n^\alpha(x) \) is a Laguerre polynomial. We follow the notation of Zoller (1979). The average on the right-hand side of (A.1) belongs to the infinite set of equations obtained by Zoller. The signal may be expressed as:

\[
L_F(\omega_B) = \frac{4CN(\varphi + \theta)}{\omega_B^3 + (\gamma + \Gamma_L)^2}(\Omega^2_L)[[\omega_B(\varphi + \theta) - (\gamma + \Gamma_L)](f + f^0) \\
+ [\omega_B + (\varphi + \theta)(\gamma + \Gamma_L)](w - w^0 + c + c^0)]
\]  

(A.2)

where \( w^n, f^n \) and \( c^n \) are given by the matrix continued fraction resulting from the recursion formulae:

\[
(\Gamma + \Lambda_{nn})w^n - 3(\Gamma + \Lambda_{nn})c^n + 6 \omega_B f^n = -\Gamma \delta_{n0}
\]

\[
(L_n + L_{n-1})w^n + (\Gamma + \Lambda_{nn} + L_n + L_{n-1})c^n + (D_n + D_{n-1} - 2\omega_B)f^n \\
= L_n(w^{n+1} + c^{n+1}) + D_{nf} f^{n+1} + L_{n-1}(w^{n-1} + c^{n-1}) + D_{n-1} f^{n-1} - L_{nf} f^{n-1}
\]

\[
(D_n + D_{n-1})w^n + (D_n + D_{n-1} - 2\omega_B)c^n - (\Gamma + \Lambda_{nn} + L_n + L_{n-1})f^n \\
= D_n(w^{n+1} + c^{n+1}) + D_{n-1}(w^{n-1} + c^{n-1}) - L_{nf} f^{n-1} - L_{nf} f^{n-1}
\]

(A.3)

In these expressions \( w^n, c^n \) and \( f^n \) are given by

\[
w^n = \langle (\sigma_+^{(1)} - \sigma_{00}^{(1)})L_n^0(\Omega^2(t)/(\Omega^2_L)^L) \rangle_L
\]

\[
c^n + if^n = \langle \sigma_+^{(1)}L_n^0(\Omega^2(t)/(\Omega^2_L)^L) \rangle_L
\]

and

\[
\Lambda_{nn} = \Gamma_L(2n + |\alpha|)
\]

\[
L_n = 2(\Omega^2)^L_{\omega_B^2 + (\gamma + \Lambda_{1n})^2}
\]

\[
D_n = 2(\Omega^2)^L_{f^2 + (\gamma + \Lambda_{1n})^2}
\]

where \( w^0, c^0 \) and \( f^0 \) represent the averages of the atomic operators:

\[
w^0 = \langle \sigma_+^{(1)} - \sigma_{00}^{(1)} \rangle_L \quad \quad c^0 + if^0 = \langle \sigma_{+^+}^{(1)} \rangle_L.
\]

One notices that at the same time we obtain the solution for the Hanle part of the signal (as it is dependent on \( \sigma_{\mu n}^{(1)} \)) in the chaotic field case.
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