Addendum

i) The third paragraph on page 2 should read:

When $\Delta \neq 0$ the spectrum remains symmetric, the central peak shifts to follow the laser and both width and shift are functions of $\Delta$ (Mollow, 1969). If collisions are included (see below), the spectrum becomes asymmetric, with the side peak furthest away from $\omega_{21}$ becoming insignificant. In either case, in the extreme limits of laser decoupling (low field strength and severe detuning) the spectrum must return to the ordinary spontaneous emission profile. If the external field is not monochromatic ...

ii) On the Contents page, the following should be added:

5. CONCLUDING REMARKS

and the following text inserted between pages 101 and 102:

5. CONCLUDING REMARKS

The many-body theory developed in this thesis presents a powerful technique for calculating susceptibility profiles in complex radiation-matter systems. Because of the simple correspondence between sets of Feynman diagrams and related physical processes, it is very easy to see how to build into a calculation, in a straightforward and self-consistent fashion, a variety of approximations, such as the impact approximation, forward scattering, T-matrix "coherence"
between initial and final states, resonant radiation interactions and so on.

Nevertheless, the theory is severely limited due to the need to begin from a system capable of being described by a thermal grand canonical statistical operator.

An alternative, and widely used approach, begins from a master equation of motion for the reduced density matrix of the atom which has been averaged over the ensemble of perturbers and radiation field modes (see for example Carmichael and Walls, 1976, and references therein). Whether or not a dressed-atom approach is followed, typical calculations involve Liouville or similar techniques along with the quantum fluctuation-regression theorem of Lax (1967) to relate solutions of the density matrix equation to the two-time dipole-matrix correlation functions which represent absorption and emission profiles (but see Cooper, 1980, who has avoided making the implicit Markovian collision approximation by retaining information contained in the t=0 density matrix).

While this approach overcomes the limitation of the many-body technique, it lacks the simple interpretation in terms of basic physical processes provided by the Feynman diagrams. In addition, it is not clear how Doppler-collision coherences and higher order perturber-density effects might be accommodated.
Both theories begin from essentially the same point. Because Green's functions are ensemble averages of particle-field states (Fetter and Walecka, 1971) the equation of motion for the Green's function is closely related to that of the density matrix. Both follow from the equation for the time evolution operator. This close similarity suggests that research towards removing the many-body thermal limitation would be worthwhile.

The failure of the experiment to measure the strongly driven collision-broadened resonance fluorescence raises questions about alternative methods. In the present case, dust or other macroscopic particles swept from the shock tube side and/or rear walls gave rise to a swamping spurious scattering signal. This is possibly a result of the particular split-bar hollow-collar design adopted for the shock tube, and also of the use of reflected shock mode. Because of the advantages of shock tubes - test gas uniformity, wide range of operating conditions, ability to achieve simultaneously high temperatures and densities - it is worthwhile attempting to overcome these problems. One likely approach would be to construct, using spark erosion, an un-split square cross-section shock tube and to conduct experiments at the exit of the tube in the straight-through mode. For this, the tube would need to exhaust into a dump tank, with observation windows far removed from the hot gas.

Other possible sources are piston compressors (Eckart, 1975) and arc-discharges (Griem, 1964). Piston compressors
are likely to suffer from much the same problems as shock tubes, due to the length of time available for contaminant mixing (a compression takes several tens of milliseconds), flush mounted windows (to reduce turbulence) and blow-past at the piston edges. Arc discharges suffer from arc inhomogeneities, difficulties in arranging diagnostics, possibly excessive electron number densities, problems in attaining high perturber number densities and difficulties in controlling the concentration of emitters (Na atoms). Nevertheless, the experiments of Wiese et al (1965) in measuring electron broadening indicate that carefully controlled arc experiments are at least a possibility.

The experiments reported in this thesis, measuring the He-broadened Na profiles, did demonstrate the utility of shock tubes run without a cookie-cutter test section. The results resolve a previous inconsistency and allow the $\frac{1}{2} - \frac{1}{2}$ width of the broadened Na 5890 Å line to be used with considerable confidence as a diagnostic for He number density (ignoring the Na 5896 Å line) for densities up to $2 \times 10^{20}$ cm$^{-3}$ and temperatures up to 5000 °K. The shock tube should provide a useful means for establishing other similar diagnostic oriented measurements.

iii) The following references on experimental measurements of the fluorescent spectrum in the absence of collisions should be added to the bibliography in the interests of completeness:


iv) The following references to the new section 5. CONCLUDING REMARKS should be added to the bibliography:

Cooper, J., Lectures at the 1980 New Zealand Laser Physics Summer School, University of Waikato, Hamilton, New Zealand.


STRONGLY DRIVEN PRESSURE BROADENED

SODIUM ATOMS

by

Charles Leedman

A thesis submitted for the degree of
Doctor of Philosophy
Australian National University

May 1980
A many-body Feynman diagram solution is developed for the susceptibility of a two state atom simultaneously perturbed by atomic collisions and a laser tuned to resonance. It is shown how the specific details of the collisions enter naturally into the emission spectrum and how this is related to the work of Baranger and Mollow, and also to the behaviour of real atoms. An experiment to verify this theory for Na atoms in a hot He bath is discussed. Construction and testing of a suitable shock tube source is described, along with some experimental limitations. Results for Na collision broadening by He are reported which resolve a previously published inconsistency (Eckart, 1975; Baird et al., 1979), and show that under the given conditions the He collision potential does not give rise to any asymmetric contributions to the spectral profile, which can thus be viewed as that from two isolated lines.
Statement of Authorship

Except where indicated the contents of this thesis are entirely my own work.

May 1980
I wish to thank all the technical staff for their patience and skill in producing excellent equipment to difficult specifications, and in particular Roland French, Don Hughes, Ken Marshall and Graham Davis. To Dr. R.J. Sandeman and Dr. H. Hornung I express my gratitude for many helpful discussions. For his guidance, mathematical and practical advice and most of all for his enthusiasm, I am grateful to my supervisor, Dr. Mark Eckart.
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1. INTRODUCTION

The development of tunable dye lasers has led to considerable interest in the behaviour of atomic radiators in the presence of strong electromagnetic fields. The solution for the emission from an isolated two level non-degenerate atom in the absence of collisions is now well established, both theoretically (Mollow 1969, 1975; Carmichael and Walls, 1976; Cohen-Tannoudji and Reynaud, 1977) and experimentally (Wu et al., 1975; Grove et al., 1977). The spectrum consists of three components, which originate from the splitting into doublets of the unperturbed atomic energy levels under the influence of the strong electric field associated with intense radiation. This is the Rabi, or A.C. Stark, effect and is represented schematically in Fig. 1.1.

If the external field is monochromatic at frequency \( \omega_k \) near the atomic resonance frequency \( \omega_{21} \) then Mollow (1969) gives the positions of the three components as \( \omega_k, \omega_k \pm \Omega' \), where \( \Omega' \) is the Rabi splitting frequency (\( \Omega' = \Omega \sqrt{2} = \sqrt{2} \left| \vec{d}_o \cdot \vec{E} \right| \)),

\[
\Omega' = \sqrt{\omega^2 + \Delta^2},
\]

(1.1)

\( \Delta = \omega_k - \omega_{21} \) is the detuning of the external field from the atomic resonance and \( \Omega = \sqrt{2} \left| \vec{d}_o \cdot \vec{E} \right| \) is the magnitude of the interaction of the atomic dipole vector \( \vec{d}_o \) with the field \( 2^{-\frac{1}{2}} \vec{E} \left[ e^{-i\omega_k t} + e^{i\omega_k t} \right] \). This assumes \( \Omega >> \gamma \), the natural linewidth, and that natural broadening is the dominant mechanism in the absence of the field. When \( \Delta = 0 \) the ratios of the peak height and
The halfwidth of the central component compared to those of the side peaks are respectively 3:1 and 2:3.

These predictions are in agreement with the results of the experiment of Wu et al. (1975) in which a 30 mW single frequency CW dye laser of 250 kHz spectral width was focused orthogonally onto an atomic beam of Na atoms producing up to 920 mW cm\(^{-2}\) in the interaction volume. The laser was tuned to the Na hyper-fine \(3^2P_{3/2}(F=3) - \mathbf{3}^2S_{1/2}(F=2)\) resonance transition and laser induced fluorescence in the absence of collisions was observed. Due to experimental uncertainties only qualitative agreement was obtained for the ratio 2:3. In a similar experiment, Grove et al. (1977) confirmed this ratio.

When \(\Delta \neq 0\) the spectrum becomes asymmetric, the central peak shifting to follow the laser and the side peak furthest away from \(\omega_{21}\) becoming insignificant. As \(\Delta/\gamma \to \pm \infty\) the ordinary spontaneous emission profile is regained. If the external field is not monochromatic, such that its bandwidth is more than approximately 20\% of the unperturbed linewidth, the three components broaden and merge appreciably (Kimble and Mandel, 1977).

The spectral behaviour when perturbing collisions are introduced in addition to the strong external field is not as well established. Mollow (1970, 1977) has introduced phenomenological damping terms into his previous density matrix treatment of the collisionless case, but does not describe in any detail how these are to be evaluated. The correspondence between the features of his spectral profile and specific collision processes is not clear, especially when coherent processes are important, for example when the initial and final states of a photon-absorbing emitter interact through collisions with the same perturber. Moreover, it is difficult to see how the kinematics of the interacting atoms, including Doppler broadening and terms of second order in the perturber density, may be fully incorporated.
This suggests that a many-body approach may be useful. Ross (1966) has considered pressure broadening as a many-body problem in the absence of an external field and has used a T-matrix expansion to describe the collisions in order to rederive the well known impact approximation for the line width $W$ and shift $D$ first obtained by Baranger (1958)

$$iW + D = V = -\left(2\pi N_p/m_p\right) \langle f_2(0) - f_1^*(0)\rangle_{av}$$

$$+ iN_p \langle v_p \int d\Omega f_2(\Omega) f_1^*(\Omega)\rangle_{av}$$ (1.2)

The perturbers have number density $N_p$, mass $m_p$ and velocity $v_p$. $\langle \rangle_{av}$ represents a Maxwellian average over $v_p$ and the elastic scattering amplitudes $f_2(\Omega)$ and $f_1(\Omega)$ describe collisions involving excited and ground state emitters respectively. In terms of $V$ and the emitter number density $N$ the susceptibility is

$$\chi(\omega) = N |\bar{d}_0|^2 (\omega - \omega_{21} + V)^{-1}$$ (1.3)

The absorption profile, proportional to $\text{Im} \chi$, is thus Lorentzian.

In the first part of this thesis a many-body, quantum electrodynamic solution is developed for $\chi$ when both collisions and a strong external field are important. It is explicitly demonstrated how the precise nature of the atomic interactions may be consistently built into the predicted spectrum and how this is related to the work of Mollow. Extensions to more complex systems are discussed.

Several experiments have been conducted on the collisional redistribution and saturation of near-resonant scattered light at low perturber densities ($N_p \leq 10^{16}$ cm$^{-3}$) and/or low gas temperatures ($\mathcal{T} \leq 2 \times 10^3$ $^\circ$K) and/or low laser powers (causing insignificant Rabi shifts compared to the collisional width) (Carlsten, Szöke and Raymer, 1977). Of greater interest is the behaviour at stellar-like conditions, $\mathcal{T} \geq 4 \times 10^3$ $^\circ$K and $N_p \geq 10^{18}$ cm$^{-3}$. 
To investigate the predictions of the theory outlined above for sodium atoms at such conditions, an experiment was designed to measure emission and absorption profiles. A shock-tube source was designed and constructed to produce a uniform He perturber bath at $N_p \sim 10^{13} - 10^{20}$ cm$^{-3}$ and $T \sim (4-6) \times 10^3$ °K with Na as a controlled impurity at $N \sim 10^{13}$ cm$^{-3}$. A tunable flashlamp-pumped dye laser was used to provide the external strong field. Experimental details are described in the second part of this thesis, including the reporting of some results for the collision broadening of Na in He which resolve a previously published inconsistency (Eckart, 1975; Baird et al., 1979) and show that under the given conditions the He collision potential does not give rise to any asymmetric contributions to the spectral profile, which can thus be viewed as that from two isolated lines.

The theoretical and experimental results presented in this thesis are being submitted as papers to the Journals of Physics A and B.
2. THEORY

2.1 Hamiltonian and Eigenstates

The system we consider is that of a gas of interacting particles, consisting of two species; two-level atoms (emitters), of number density \( N \) and mass \( m \), and single-level atoms (perturbers), of number density \( N_p \gg N \) and mass \( m_p \). The gas is irradiated by a monochromatic field described by a frequency \( \omega_k \) and wave vector \( \vec{k} \) (the laser field) which interacts with the emitters only. Emitters are mutually non-interacting. We assume that \( \Delta/\omega_{21} \ll 1 \) where \( \Delta = \omega_k - \omega_{21} \) and \( \omega_{21} \) is the resonance frequency of the transition between the unperturbed emitter upper and lower states, \( |2> \) and \( |1> \) respectively. We wish to consider the susceptibility of such a system to a weakly perturbing additional, not necessarily monochromatic, electromagnetic field (the probe field).

We proceed by first finding the eigenstates for the second-quantized dressed-emitter Hamiltonian \( H_D \) which describes the sub-system of emitter plus laser field.

\[
H_D = \omega_k a^+ a + \frac{\omega_{21} \sigma_z}{2} + \kappa a^+ \sigma - m + \kappa^* \sigma_+ m^+ + \frac{P \cdot P}{2m} \tag{2.1}
\]

where \( a^+, a \) are the laser photon creation and destruction operators and \( a^+ a \) is the laser photon number operator;

\( \sigma_+ , \sigma_\) are the usual emitter raising and lowering (pseudo-spin) operators with \( \sigma_z = \sigma_+ \sigma_- - \sigma_- \sigma_+ \);

\( m^+_z , m^-_z \) are atomic momentum operators defined by \( m^{z}_\pm |n, \vec{p}, \alpha> = |n, \vec{p} \pm \vec{k}, \alpha> \) where \( \alpha = 1,2 \) and \( |n, \vec{p}, \alpha> \) describes a state characterised by an emitter in state \( |\alpha> \), with 3-momentum \( \vec{p} \), plus a laser field of \( n \) photons;

\( \kappa \) is a coupling constant between the laser field and the emitter \( (\kappa^* \) is the complex conjugate). We choose \( n \) such that \( 4|\kappa|^2 n = \Omega^2 \) and we
will always require $\Omega << \omega_{21}$; and

$P$ is a momentum operator for the atom defined by $P|n, \vec{p}, \alpha> = \vec{p}|n, \vec{p}, \alpha>$. The eigenstates of $H$ are

$$|n, +, \vec{p}> = u(n,\vec{p}) |n, \vec{p}, 1> + v(n,\vec{p}) |n-1, \vec{p} + \vec{k}, 2>$$

(2.2)

$$|n, -, \vec{p}> = -v(n,\vec{p}) |n, \vec{p}, 1> + u(n,\vec{p}) |n-1, \vec{p} + \vec{k}, 2>$$

and the corresponding eigenvalues are

$$E(n,+,\vec{p}) = \omega_{k}(n-\frac{1}{2}) + \frac{p^2}{2m} + \phi(n,\vec{p})$$

(2.3)

$$E(n,-,\vec{p}) = E(n,+,\vec{p}) - \phi(n,\vec{p})$$

where $\phi(\vec{p}) = (\vec{p} \cdot \vec{k}) / m + \frac{\vec{k}^2}{2m}$

$$\phi(n,\vec{p}) = ([\Delta - \phi(\vec{p})]^2 + 4|\kappa|^2 n)^{1/2} > 0$$

$$u(n,\vec{p}) = (1+y/z)^{1/2} v(n,\vec{p}) = u(n,\vec{p})|_{y \rightarrow z}$$

$$y = \phi(n,\vec{p}) - \Delta + \phi(\vec{p}) > 0$$

$$z = \phi(n,\vec{p}) + \Delta - \phi(\vec{p}) > 0$$

(2.4)

The dressed-emitter eigenvalues form an infinite sequence as shown in Fig. 2.1.

Fig. 2.1: Dressed atom energy level scheme
We assume that radiative transitions take place only between states of different $n$, while collisions and collision induced transitions involve states of the same $n$ (in most transitions the quantum number $\tilde{p}$ will alter).

From the definition of $\phi(n,\tilde{p})$ it follows that for a given $n$ and $\tilde{p}$,

$$E(n,+,\tilde{p}) > E(n,-,\tilde{p}).$$

We consider interaction of the $H_D$ system with both the perturber bath (infinite order perturbation) and the weak probe field (linear response). The Hamiltonian describing this interaction is $H_I = H_{DR} + H_{DP}$ where, as in Eq. (2.1),

$$H_{DR} = \sum_{q,\omega} \left\{ \chi_{q,\omega}^* b_{q,\omega}^+ \hat{\sigma}_- + \chi_{q,\omega} \hat{\sigma}_+ b_{q,\omega} \right\}$$

describes the probe-photon dressed-emitter part, with $b^+$, $b$ and $\hat{\sigma}_+$, $\hat{\sigma}_-$ the probe-photon creation/destruction and dressed-emitter raising/lowering operators respectively (with momentum included). $\chi_{q,\omega} = \omega \hat{A}_q$ is a coupling constant. A similar expression may be written for the perturber dressed-emitter part $H_{DP}$ involving operators which change the 3-momentum quantum numbers and a coupling constant characteristic of the perturber-emitter interatomic potential.

The problem thus defined differs from that considered by Carmichael and Walls (1976), in their treatment of the A.C. Stark effect using a quantum mechanical master equation approach, by the inclusion of collisions and other atomic kinematic effects, and in the provision for $\Delta \neq 0$. 
2.2 Formalism

We consider linear response of the colliding dressed-emitter to the weak probe field and find (Ross, 1966) that the time-averaged power per unit volume absorbed from the probe beam at frequency $\omega_q$ is

$$P(\mathbf{q}, \omega_q) = \frac{3}{2} \omega_q^3 \left| A(\mathbf{q}, \omega_q) \right|^2 \operatorname{Im} \{ \hat{e} \cdot \overline{\chi}(\mathbf{q}, \omega_q) \cdot \hat{e} \}$$  \hspace{1cm} (2.5)

where $A$ is the amplitude of the vector potential, $\hat{e}$ is the probe beam polarization unit-vector and $\overline{\chi}$ is the retarded susceptibility. Following Ross we introduce the so-called imaginary time (finite temperature) Green's function propagators for each type of particle.

This is straightforward for perturbers and probe photons but not for dressed-emitters. An essential (periodic) boundary condition for the defining differential equation for the Green's function for a given particle is that the corresponding grand canonical statistical operator be of the form $e^{\beta(\hat{N} - H + \mu \hat{N})}$, where $\beta^{-1} = k_B T$ refers to some well defined temperature $T$, $\hat{N}$ is the thermodynamic potential, $\mu$ is the chemical potential and $\hat{N}$ is the number operator (see Fetter and Walecka, 1971, and Kadanoff and Baym, 1962). It is clear that this is the case for perturbers, whose momentum distribution reflects a well defined kinetic temperature. However, this is not true for the laser field whose sharp peaking about some mean photon number $n_0$ means that the field $|n\rangle$ and the levels of Fig. 2.1 cannot be thermal.

Just before the laser-emitter interaction is turned on, the emitter momentum and internal states are described by a thermal distribution at the same temperature as the perturbers (assuming collisions are sufficiently rapid for LTE) and we will assume that the laser field has exactly $n$ photons.
The initial system then consists of the states $|n, \bar{p}, 1\rangle$ and $|n, \bar{r}, 2\rangle$ occupied in the ratio $1:e^{-\beta_0 2\hbar}$ (for average $\bar{p}$ and $\bar{r}$ not necessarily different). Immediately after the interaction is turned on, the states $|n, \pm, \bar{p}\rangle$ and $|n+1, \pm, \bar{r}\rangle$ will be occupied in the same ratio, while all other states will be empty. Inelastic collisions and spontaneous/stimulated emission and absorption processes populate these initially empty states, a typical process (in the undressed picture) being an inelastic collision inducing the transition $|2\rangle \rightarrow |1\rangle$ followed by absorption $|1\rangle \rightarrow |2\rangle$ removing a photon from the laser field. This process represents the heating up of the gas by the laser to an eventual temperature equal to the black body temperature corresponding to a radiation density (in the laser mode) equivalent to the laser field.

However, because $N << N_p$ and the emitter transition is rapidly saturated by the external field, negligible energy is absorbed from the laser in typical experimental times of $\mu$s and this heating effect can be ignored.

The dressed-emitter level populations may nevertheless not be in LTE. For each dressed-emitter doublet labelled by $n$ we define spontaneous emission, upward and downward inelastic collision rates $S_n$, $U_n$, and $D_n$ respectively. We might expect these to be similar to those for the emitter, i.e.

$D_n \gg S_n, U_n$, and to change very little with $n$ for large $n >> 1$. Then each level population $P_n$ satisfies

$$\frac{dP_n}{dt} = P_{n+1} \left( D_{n+1} + S_{n+1} \right) + P_{n-1} U_{n-1} - P_n \left( D_n + S_n + U_n \right)$$

and the original ratio $1:e^{-\beta_0 2\hbar}$ may be expected to change significantly in a time $\gg D_n^{-1}$.

For times shorter than this the dressed-emitter states clearly satisfy the periodic boundary condition, and we will solve for the susceptibility accordingly. The restrictions this places on the applicability of the solution will be discussed in a later section.
After introducing the Green's function propagators, we calculate $\chi(q,\omega_q)$ as the analytic continuation $i\omega_q + \omega_q + i\epsilon$, $\epsilon > 0$ (from the imaginary frequency $\omega_q$ to the real frequency $\omega_q$) of the causal susceptibility $S(q,\omega_q)$. In the dipole approximation the latter is then the two-particle finite-temperature propagator with free-free propagation removed and external vertices added (see for example Fetter and Walecka, 1971; Abrikosov et al. 1963; Ross, 1966). In terms of Feynman diagrams we write (4-momentum is conserved - see below)

$$S(q,\omega_q) = \sum_{\alpha,\beta,\gamma,\delta} q, p+q, p+k$$

which represents the absorption of a probe photon of 4-momentum $q = (q,\omega_q)$, the propagation of both an excited dressed-emitter and a hole in the sea of unexcited dressed-emitters and their coalescence with the re-emission of the photon $q$. For convenience we suppress the explicit dependence of $S$ on $k$ and $n$.

Greek indices label the first two quantum numbers for the dressed-emitter and are subject to the selection rules regarding perturber collisions and radiative transitions mentioned earlier. Consistent with the above discussion on level populations, we consider only diagrams in which the excited dressed-emitter state is $|n+1, \pm, \bar{p}+\bar{q}\rangle$ and the unexcited state is $|n, \pm, p+k\rangle$. We assume that the propagators are diagonal (discussed below) so that $\beta = \gamma = (n+1,\pm)$ and $\alpha = \delta = (n,\pm)$. Note that $\bar{p} + \bar{q}$ and $\bar{p} + \bar{k}$ refer to the momentum attributable to the emitter only (in the ground state), while the corresponding dressed-emitter momenta are $p + q + (n+1)k$ and $p + k + nk$, which include the laser field momentum. Thus 4-momentum is conserved at each vertex.
The shaded region represents the vertex function which includes the so-called coherent processes, in which (in this case) the incoming hole and outgoing excited dressed-emitter react via collisions with the same perturber. It is this type of process which is responsible for the third term in Baranger's line shape, Eq. (1.2). Explicitly, we write:

\[-S(q, w) = \sum_{a, \beta} \int \frac{d^4 p}{(2\pi)^4} \Lambda(a, \beta; p, p+q) \Lambda(\beta, a; p+q, p+k) \times G(a, p+k) G(\beta, p+q)\]

(2.7)

where \(\Lambda\) is the external (unshaded) vertex at \(\gamma, \delta\) and the integration represents an integral over the possible values of the 3-momentum \(p\), and a summation over the imaginary frequency \(w\). We solve for the Green's function propagators by inverting Dyson's equation (see Fetter and Walecka, 1971)

\[G(\delta, a; p) = \left\{G(\delta, a; p) \right\}^{-1} \Sigma(\delta, a; p)^{-1}\]

(2.8)

where \(G\) is the propagator for the dressed-emitter in the absence of collisions, \(\Sigma\) is the proper self energy (also called the mass operator) and we have relaxed, for the moment, the diagonal condition \(a = \delta\). Note that Dyson's equation involves the inversion of a matrix in the indices \(\delta, a\).

Any approximation for \(\Sigma\) will yield, by the method of functional differentiation described by Kadanoff and Baym (1962), an infinite-order perturbative, conserving approximation for the susceptibility. We choose a \(\Sigma\) which includes two-body, dressed-emitter perturber, impact regime collisions only. This is an entirely reasonable assumption for typical laboratory conditions in which perturber pressure-broadened line widths far exceed those of natural broadening while being much less than the inverse of the collision duration, \(N_p\) is small enough that three-body collisions may be ignored and \(N\) is small enough that emitter-emitter
collisions may be ignored along with all Dicke-type co-operative effects (Dicke, 1954). This is the case for most relevant compressor and shock-tube experiments (Eckart, 1975; Baird et al., 1979) and forms the design criterion for the shock-tube described in the second part of this thesis.

The appropriate self energy is then given by the lowest order T-matrix approximation:

\[
\Sigma(\delta, \alpha; p) = \int \frac{d^4 r}{(2\pi)^4} T(\delta, \alpha; p, r; 0) G^{oo}(r) \quad (2.9)
\]

which represents an ensemble average, over perturber momentum \( r \), of a collision involving an incoming dressed-emitter of state \( \delta \) and momentum \( p \). \( G^{oo}(r) \) is the perturber propagator which, due to the absence of internal perturber states, is a free-particle propagator. The T-matrix is a sum of ladder diagrams in the two-body perturber-emitter interatomic potential expectation value \( V(\delta, \alpha; p, t) \):

\[
T(\delta, \alpha; p, r; t) = \sum_{t} \frac{d^4 r}{(2\pi)^4} V(\delta, \alpha; p, r; t) G^{oo}(r) G(s, \alpha; p, t) T(s, \alpha; p, t) T(s, \alpha; p, t)
\]

\[
= V(\delta, \alpha; p, t) + \sum_{s, t} \frac{d^4 r}{(2\pi)^4} V(\delta, \alpha; p, r; t) G^{oo}(r) G(s, \alpha; p, t) \times T(s, \alpha; p, t)
\]

\( (2.10) \)
A similar equation may be written for the vertex function:

\[ \Lambda(\alpha, \beta; p+k, p+q) = \]

\[ = \]

\[ = \]

\[ = \]

\[ \Gamma(\alpha, \beta; \varepsilon; p+k, r+q; r-p) \]

\[ \times G(\gamma, \varepsilon; r+k) G(\delta, \eta; r+q) \Lambda(\gamma, \delta; r+k, r+q) \] \hspace{1cm} (2.11)

where \( \Gamma \) is the irreducible four-point function which contains all possible two-particle interactions not divisible by means of particle-hole cuts, and is obtained by functional differentiation of the self energy with respect to particle propagators (Kadanoff and Baym, 1962) \( \Gamma = \frac{\delta \Sigma}{\delta G} \).

Various combinations of indices in \( \Sigma \) and \( G \) yield corresponding terms in \( \Gamma \) but as perturbers cannot become emitters, and vice-versa, the only surviving term (Ross, 1966) is

\[ \Gamma(\alpha, \eta; \beta, \varepsilon; p+k, r+q; r-p) = p+s-r \]

\[ = - \int \frac{d^4 s}{(2\pi)^4} G^{00}(s) G^{00}(p+s-r) T(\alpha, \varepsilon; p+k, s; r-p) T(\eta, \beta; r+q, p+s-r; p-r) \] \hspace{1cm} (2.12)

We now have all the necessary elements for a solution to the susceptibility of Eq. (2.7). After choosing a particular inter-atomic potential \( V \), the properties of the T-matrices are determined by making some suitable approximations for the propagator \( G \) in Eq. (2.10).
Corresponding self energies are then calculated and substituted into Dyson's equation, yielding an approximation for the dressed-emitter propagator that can be used in a solution of the vertex equation and finally in the susceptibility. Such an approximation is fully conserving in that the conservation laws, namely energy, momentum and particle number conservation, are automatically satisfied. On a microscopic level this implies that there are no spurious or omitted Feynman diagrams in our approximation: probability is conserved.

2.3 T-Matrices

Let \( V_1(\overline{\epsilon}) \) and \( V_2(\overline{\epsilon}) \) be the interatomic virtual potentials for collisions involving a momentum transfer \( \overline{\epsilon} \) between a perturber and respectively an unexcited and excited emitter. Then substitution of the eigenstates of Eq. (2.2) into \( V(\delta,\alpha;p,t) \) yields

\[
\begin{align*}
V(l',1';p,t) &= a_1 V_1(\overline{\epsilon}) + a_2 V_2(\overline{\epsilon}) \quad a_1 = u(n,\overline{p}) u(n,\overline{p}+\overline{\epsilon}) \\
V(2',2';p,t) &= a_2 V_1(\overline{\epsilon}) + a_1 V_2(\overline{\epsilon}) \quad a_2 = v(n,\overline{p}) v(n,\overline{p}+\overline{\epsilon}) \\
V(l',2';p,t) &= -a_3 V_1(\overline{\epsilon}) + a_4 V_2(\overline{\epsilon}) \quad a_3 = u(n,\overline{p}) v(n,\overline{p}+\overline{\epsilon}) \\
V(2',1';p,t) &= -a_4 V_1(\overline{\epsilon}) + a_3 V_2(\overline{\epsilon}) \quad a_4 = u(n,\overline{p}+\overline{\epsilon}) v(n,\overline{p})
\end{align*}
\]

where \( l' = (n,+) \) and \( 2' = (n,-) \). Note that as \( \Delta/4\kappa^2 \eta \rightarrow 0 \) and Doppler shift and atomic recoil effects become small so that \( \Theta(\overline{p})/4\kappa^2 \eta \rightarrow 0 \) then both \( u, v \rightarrow 1/\sqrt{2} \) independent of \( \overline{p} \) and \( \overline{\epsilon} \), and \( V(1',1') \rightarrow V(2',2') \) while \( V(1',2') \rightarrow V(2',1') \). If in addition there is no broadening of the spectral line due to collisions involving the unexcited emitter state, so that \( V_1(\overline{\epsilon}) = 0 \) (the no lower-broadening limit), then all the \( V(\delta,\alpha) \) are equal to \( V_2(\overline{\epsilon})/2 \).
To be consistent with our choice of inclusion of two-body collisions only, we assume that the propagator $G(\epsilon, \eta)$ in Eq. (2.10) itself contains no collisions and is therefore the unperturbed, diagonal, dressed-emitter propagator $\tilde{G}(\epsilon) = G(\epsilon, \eta)_{\epsilon=\eta}$. The T-matrix equations then decouple into two sets of two integral equations:

$$T(1',1';p,r;t) = V(1',1';p,t) + \int \frac{d^4s}{(2\pi)^4} V(1',1';p,s)A + \int \frac{d^4s}{(2\pi)^4} V(1',2';p,s)B$$

$$T(2',1';p,r;t) = V(2',1';p,t) + \int \frac{d^4s}{(2\pi)^4} V(2',1';p,s)A + \int \frac{d^4s}{(2\pi)^4} V(2',2';p,s)B$$

where $A = \tilde{G}^{oo}(r-s) \tilde{G}(1',p+s) T(1',1';p+s,r-s;t-s)$

and where identical equations hold with $1' \rightarrow 2'$, $2' \rightarrow 1'$. By considering an iteration of Eq. (2.14) it is clear that in each case the T-matrices are independent of $\omega^\tau$ and depend only on the combination $\omega^p + \omega^r$ due to the fact that the potentials $V$ are functions of 3-momenta only (make the substitution $s+r-s$). Then the T-matrices in A and B can be taken outside the imaginary frequency sums of Eq. (2.14). The propagators are (see Fetter and Walecka, 1971; and recall that there are $n$ laser photons)

$$[\tilde{G}^{oo}(r-s)]^{-1} = iw_r - iw_s - E_p(r-s)$$

$$[\tilde{G}(1',p+s)]^{-1} = iw_p + iw_s + iw_k - \omega_k(n^{-\frac{1}{2}}) - E(p+s) - \frac{1}{2} \phi(n,\bar{p}+\bar{s})$$

$$[\tilde{G}(2',p+s)]^{-1} = [\tilde{G}(1',p+s)]^{-1} + \phi(n,\bar{p}+\bar{s})$$

where $E_p(r-s) = (r-s)^2/2m - \mu_p$, $m$ is the perturber mass, $\mu_p$ is the perturber chemical potential and similarly $E(p+s) = (p+s)^2/2m - \mu$ for the dressed-emitter. Strictly, Eq. (2.15) is incorrect as it gives each particle an
infinite lifetime. To each inverse propagator there should be added a term $i\Gamma \text{sgn}(F)$, where $\Gamma > 0$ represents some finite lifetime and where $F = \omega_r - \omega_s$ for $G^{00-1}$ and $F = \omega_p + \omega_s + \eta \omega_k$ for both $\overline{G}^{-1}$. We omit such terms here as they are unimportant; in later sections however they form a necessary part of essential analytic properties.

To keep account of statistics, we choose our atomic particles to be fermions (this choice is arbitrary, as our observables are calculated in the classical limit, but avoids problems with Bose condensation) while photons and virtual interactions such as $V$ in the T-matrix equations are described by bosons. We perform the frequency sum in the usual way by interpreting it as a Cauchy integral along a contour $C$ which encloses the imaginary axis in the anticlockwise direction and is then pushed to infinity:

$$\int_C \frac{dz}{e^{\beta z} + 1} e^{\eta z} f(z)$$

where $\eta$ is a convergence factor ensuring that the contribution at infinity goes to zero and $\frac{1}{\beta} = kT$, $k$ = Boltzman's constant and $T$ = absolute temperature. In the classical limit $e^{\beta \mu_i} \ll 1$ for all $i$ so that $(e^{\beta (\mu_i - \mu_j)} + 1)^{-1} \rightarrow e^{-\beta (\mu_i - \mu_j)}$ for both statistics. Note that $e^{\beta \mu w} = + 1$ for $w$ describing fermions or bosons respectively. Explicit dependence on the chemical potential vanishes with the normalisations:

$$e^{\beta (\mu - \omega_k (n - \frac{1}{2}))} = \left( \frac{2\pi \beta}{m} \right)^{3/2} N$$

$$e^{\beta \mu_p} = \left( \frac{2\pi \beta}{m_p} \right)^{3/2} N_p .$$

Consider the first integral in Eq. (2.14). Both $G^{00}$ and $\overline{G}$ have poles in the complex $\omega_s$-plane that are now picked up by the contour $C$. That due to $\overline{G}$ will introduce a factor $N$ while the $G^{00}$ pole contributes no factors of $N$ or $N_p$. By hypothesis $N$ is vanishingly small (dilute system) so we ignore.
the G pole and find that the residue of the $G^{00}$ pole yields

$$\int \frac{d^3 \vec{s}}{(2\pi)^3} \frac{V(l',l';p,s) T(l',l';p+s,r-s;t-s)}{[i\omega_p + i\omega_r + i\omega_k - \omega_k(n-\frac{1}{2}) - E_p(r-s) - E(p+s) - \frac{1}{2}\phi(n,p+s)]}$$

(2.17)

In the complex $w_p$-plane $T(l',l';p,r;t)$ thus has a semi-infinite branch line along $\text{Im} z = -\omega_r - n\omega_k$ to the right of the point

$$[\omega_k(n-\frac{1}{2}) + \{E_p(r-s) + E(p+s) + \frac{1}{2}\phi(n,p+s)\}] \text{minimum w.r.t. } s > |\mu + \mu'|,$$

so that in any $w_p$-sum it will introduce a factor $N_N/n_p$. The other integrals in Eq. (2.14) yield similar results.

To demonstrate the form of our T-matrices we make the hard-sphere approximation which sets an upper limit $s'$ on the momentum transferred by a T-matrix collision: $s' = \hbar$ where $r$ is the sum of the perturber and emitter hard-sphere radii and $\hbar$ is Planck's constant. The hard sphere potentials $V_1(s)$ and $V_2(s)$ are constant for $|s| < s_1$, $s_2'$ respectively and are assumed large but finite. For typical experimental conditions involving dilute Na emitters in a bath of He perturbers at 5-6000°K and $N \sim 10^{18} - 10^{21}$ cm$^{-3}$, results show that this is a reasonable approximation (Eckart, 1974).

In addition, the forward scattering approximation is then valid;

$$|s| \ll |\vec{r} + \vec{p}|_{\text{average}}$$

so that scattering is highly peaked in the forward direction (in the spirit of the undeviated classical path approximation).

Then in Eq. (2.14) $T(l',l';p+s,r-s;t-s) \sim T(l',l';p,r;t)$ and may be taken outside the 3-momentum integration. This reduces Eq. (2.14) to a set of algebraic equations for the T-matrices. In obvious matrix notation $T = V + KT$, where $K$ is a $2 \times 2$ matrix whose elements are all of the same form as

$$K_{11} = \int_0^\infty \frac{ds}{(2\pi)^2} s^2 V(l',l';p,s) \int_{-1}^1 d(\cos \theta) \left[i\omega_p + i\omega_r + i\omega_k - \omega_k(n-\frac{1}{2}) - E_p(r-s) - E(p+s) - \frac{1}{2}\phi(n,p+s)\right]^{-1}$$

(2.18)
where $\tilde{V}^0 = \tilde{p}/m - \tilde{r}/m$, $s.\tilde{V}^0 = sV^0 \cos \theta$ and the hard sphere potential is defined using the Heaviside step function $\mathbb{H}_e(x) = \begin{cases} 0 & x < 0 \\ 1 & x \geq 0 \end{cases}$ as

$$V(l', l''; p, s) = a_1V_1[1 - \mathbb{H}_e(s - s') + a_2V_2[1 - \mathbb{H}_e(s - s'_2)] .$$

This is to be compared to Eq. (2.13). Note that the collision process has been assumed axisymmetric about $\tilde{V}^0$.

In terms of $K$ we have:

$$T(l' , l''; p; r; t) = \frac{V(l', l''; p; t) (1 - K_{22}) + V(2', l''; p; t)K_{12}}{[(1 - K_{11})(1 - K_{22}) - K_{12} K_{21}]}$$

$$T(2', l''; p; r; t) = \frac{V(l', l''; p; t) K_{21} + V(2', l''; p; t)(1 - K_{11})}{[(1 - K_{11})(1 - K_{22}) - K_{12} K_{21}]}$$

with identical equations $l' \leftrightarrow 2'$ and corresponding changes in $K_{11}, K_{22}$ etc.

The impact approximation for collisions limits the off-energy-shell behaviour of the T-matrices, so that for the denominator of Eq. (2.18) we can write

$$\sigma + i\omega + i\mathcal{V}_k - \omega_k(n - \frac{1}{2}) - E_{p}(r - s) - E_{p}(p + s) - \frac{i\lambda}{2}(n, p + s) \rightarrow \alpha + i\beta$$

where $\alpha, \beta$ are real numbers at most of the order of the collision broadened shift or $\frac{1}{2} - \frac{1}{2}$ width. Letting $\alpha' = \alpha/V^0s', \beta' = \beta/V^0s'$ and $x = s/s'$ yields

$$K_{11} = \frac{(a_1V_1s_1^2 + a_2V_2s_2^2)}{4\pi V^0} \int_0^1 dx x^2 \int_0^{+1} d\cos \theta \frac{[\alpha' = x \cos \theta - \beta']}{[(\alpha' - x \cos \theta)^2 + \beta'^2]}$$

where the appropriate $s'_1, s'_2$ is understood in $\alpha', \beta'$. The major contribution to the susceptibility will occur for on-energy-shell T-matrices (Ross, 1966) that is, in the limit $\alpha', \beta' \rightarrow 0$. Then

$$K_{11} = -\frac{i}{8\pi V^0} (a_1V_1s_1^2 + a_2V_2s_2^2)$$
with similar terms for $K_{22}$ etc. The T-matrices then follow from Eq. (2.19) and in the limit $V_1, V_2 \to \infty$ we obtain after some algebra:

$$T(1',l';p,r;|s, s') = -i8\pi V^0 \left\{ \frac{a_1^2 + a_4^2}{s_1^2} + \frac{a_2^2 + a_3^2}{s_2^2} \right\}$$

$$T(2',l';p,r;|s, s') = -i8\pi V^0 \left\{ \frac{a_2 a_4 + a_3 a_1}{s_1^2} - \frac{a_2 a_4 + a_1 a_3}{s_2^2} \right\}$$

(2.20)

with identical equations $1' \leftrightarrow 2'$ and the interchange $1 \leftrightarrow 2, 3 \leftrightarrow 4$ on the $a$ subscripts. Note that in this approximation $T(2',l') = T(1',2')$. As $t$ increases past $s'_1, s'_2$ first one term, then both terms, within each of the curly brackets of Eq. (2.20) go to zero discontinuously. A similar analysis in the absence of the laser and beginning from the unmixed states $|1>, |2> yields $-i8\pi V^0/s'_1^2$ and $-i8\pi V^0/s'_2^2$ for the lower and upper state T-matrices respectively.

Hence, as might have been expected, the T-matrices of Eq. (2.20) are simple combinations of the no-laser T-matrices, the combination in any particular case depending on laser parameters through the dependence of $a_1, a_2, a_3, a_4$ on $\Delta$ and $4\kappa^2 n$.

Some special cases are worth mentioning:

- $\Delta = 0$ and $V_1 = 0$ (no lower state broadening). Then all the

$$K_{ij} = \frac{-i V_2 s_2^2}{16\pi V^0}$$

and all the T-matrices are equal to $-i4\pi V^0/s_2^2$ which is one half of the result obtained in the absence of the laser for the upper level T-matrix. This is in fact a general result following from Eq. (2.14) and is independent of the hard sphere approximation.
The diagonal T-matrices are both equal to one half of the sum of the no-laser upper and lower T-matrices, but the non-diagonal T-matrices now depend on the relative magnitude of $s_1$ and $s_2$. When these are equal (the broadening is the same for upper and lower states) the non-diagonal T-matrices are zero.

### 2.4 Propagators

We return to Dyson's equation, Eq. (2.8), into which we substitute the self energy Eq. (2.9) to obtain:

$$\Sigma(\delta, \alpha, p) = \int \frac{d^3r}{(2\pi)^3} \frac{T(\delta, \alpha; p, r; 0)}{[i\omega_r - E_p(r)]}$$

(2.21)

$$= N_p \left( \frac{2\pi \hbar}{m_p} \right)^{3/2} \int \frac{d^3r}{(2\pi)^3} e^{-\beta r^2/2m_p} T(\delta, \alpha; p, r; 0) i\omega_p + E_p(r)$$

where the frequency dependence of the forward scattering T-matrix has been written explicitly as a superscript and contributions due to the T-matrix branch line in the complex $w_r$-plane have been ignored due to their being of order $N_p$. The perturber average is over a Maxwellian velocity distribution. Inverting the matrix of Eq. (2.8) and dropping the argument $p$ we have:

$$G(\delta, \alpha) =$$

$$\left\{ \begin{array}{c} \bar{G}(1', 1')^{-1} - \Sigma(1', 1') - \frac{\Sigma(1', 2') \Sigma(2', 1')}{\bar{G}(2', 2')^{-1} - \Sigma(2', 2')} \end{array} \right\}^{-1}$$

$$\left\{ \begin{array}{c} \bar{G}(2', 2')^{-1} - \Sigma(2', 2') - \frac{\Sigma(1', 2') \Sigma(2', 1')}{\bar{G}(1', 1')^{-1} - \Sigma(1', 1')} \end{array} \right\}^{-1}$$

(2.22)
The off-diagonal parts of $G(\delta, \alpha)$ are thus proportional to $N_p$, and may be ignored under the dilute perturber approximation (they will introduce factors of at least $N_p^2$ in the $\nu_p$-sum of Eq. (2.7)). Similarly the last term in each curly bracket is proportional to $N_p^2$. $G(\delta, \alpha)$ may therefore be assumed diagonal, which justifies the use of Eq. (2.7) in the form given.

2.5 Vertex Functions and Susceptibility

Labelling $(n+1, +)$ and $(n+1, -)$ by $3'$ and $4'$ respectively, there are now only four non-vanishing diagrams in Eq. (2.7):

$$S(\bar{q}, \omega_q) \omega_q^2 = \begin{array}{c}
\begin{array}{c}
3' \quad p+q \\
1' \quad p+k
\end{array}
\end{array} + \begin{array}{c}
\begin{array}{c}
4' \quad p+q \\
2' \quad p+k
\end{array}
\end{array} + \begin{array}{c}
\begin{array}{c}
4' \quad p+q \\
1' \quad p+k
\end{array}
\end{array} + \begin{array}{c}
\begin{array}{c}
4' \quad p+q \\
2' \quad p+k
\end{array}
\end{array}
$$

(2.23)

Examination of Eq. (2.11) indicates that the vertex equation then becomes an inhomogeneous system of four simultaneous integral equations in which all the terms are non-zero. Application of the forward scattering approximation yields an inhomogeneous system of four simultaneous algebraic equations, whose solution demands the inversion of a $4 \times 4$ matrix. Rather than attempt this we write:

$$\Lambda(\alpha, \beta; p+k, p+q) = \frac{\Lambda(\alpha, \beta; p+k, p+q)}{1-F(\alpha, \beta; p+k, p+q)}$$
whence

\[ F(\alpha, \beta; p+k, p+q) = \Lambda^{-1}(\alpha, \beta; p+k, p+q) \sum_{\gamma, \delta} \int \frac{d^4r}{(2\pi)^4} G(\gamma, r+k) G(\delta, r+q) \]

\[ \times \Gamma(\alpha, \delta; \beta, \gamma; p+k, r+q; r-p) \Lambda(\gamma, \delta; r+k, r+q) \left\{ \frac{1 - F(\alpha, \beta; p+k, p+q)}{1 - F(\gamma, \delta; r+k, r+q)} \right\} \]  

(2.24)

If our expression for the susceptibility is to be at all correct, we must have \(|F| < 1\) (otherwise we have chosen a poor starting basis). Thus, as a first approximation to a solution of Eq. (2.24) we set the term in curly brackets equal to unity, and iterate to obtain better solutions if necessary. Note that \(\alpha, \gamma\) are restricted to \(1'\) or \(2'\) while \(\beta, \delta\) can take on only the values \(3'\) or \(4'\). This procedure eliminates the need to make the forward scattering approximation in order to invert the vertex equation.

Substitution of the eigenstates of Eq. (2.2) into \(\Lambda(\alpha, \beta; p+k, p+q) = \langle \beta, 
\begin{align*}
\Lambda(1', 3'; p+k, p+q) &= \omega_2 d_0 u(n, \overline{p}) (n+1, \overline{p}+q) \\
\Lambda(1', 4'; p+k, p+q) &= \omega_2 d_0 u(n, \overline{p}) u(n+1, \overline{p}+q) \\
\Lambda(2', 3'; p+k, p+q) &= -\omega_2 d_0 v(n, \overline{p}) v(n+1, \overline{p}+q) \\
\Lambda(2', 4'; p+k, p+q) &= -\omega_2 d_0 v(n, \overline{p}) u(n+1, \overline{p}+q)
\end{align*}

(2.25)

where \(d_0 = \langle 2| \overline{d}| 1\rangle\) is the emitter dipole matrix element. Note that \(\Lambda(\alpha, \beta) = \Lambda^*(\beta, \alpha)\).

The four-point function \(\Gamma\) is given by Eq. (2.12). In the \(w_s\)-sum the contributions from each of the T-matrix branch lines are of order \(N_pN\) and may be neglected. For reasons to be discussed below, the perturber can no longer be assumed to have no self energy. Off-energy-shell behaviour is accommodated by writing \(G^{oo}(r)^{-1} = i\omega_r E_p(r) + ie \text{sgn}(\omega_r)\) where \(e > 0\).
This transforms the perturber pole into a branch along $\text{Im} z = 0$ in the complex $w_r$-plane. The sign of $e$ is determined by the Lehmann representation of diagonal propagators, which requires that the imaginary part of the self energy be less than zero when $\omega_r > \mu_p$ (Abrikosov et al., 1963; we have set energy levels such that $\mu_p = 0$).

Ignoring for the moment the analytical structure of the $T$-matrices in the complex $w_s$-plane, in Eq. (2.12) the contour pushed to infinity now looks like

![Diagram](image)

and instead of considering residues at perturber poles we must evaluate branch line integrals. Although we have chosen $w_r - w_p < 0$, the result is independent of this choice. Just above and below each branch line we have

$$G^{00}(s)^{-1} G^{00}(p+s-r)^{-1} = [iw - E_p(s) + i\epsilon n_1] [iw + iw - E_p(s+p-r) + i\epsilon n_2]$$

with respectively $n_1 = n_2 = 1$; $n_1 = -1 = -n_2$; $n_1 = -1 = -n_2$ and $n_1 = n_2 = -1$. This function has poles in the second or non-physical sheet. We will evaluate the branch line integrals in an approximate fashion by taking residues at these poles. This involves small errors in detailed balancing of the order $(\omega_q - \omega_{21})/\omega_{21}$, which in this case may be neglected in calculating the susceptibility. Only the terms with $n_1 = -1 = -n_2$ contribute (other terms have both poles in the same non-physical half-plane) and recalling that $e^{\beta (w_r - w_p)} = 1$ we obtain for the four point function:
\[ \Gamma (\alpha, \delta; \beta, \gamma; r+k, r+q; r-p) \]
\[
= \int \frac{d^3s}{(2\pi)^3} \left[ i(w_r - w_p) + E_p(s) - E_p(p+s-r) + 2ie \right]^{-1}
\]
\[
\times \frac{T(\alpha, \gamma; p+k, s; r-p) i\omega_p + i\omega_k + E_p(s) + ie}{\left( e^{\beta s^2/2m_p} - \beta i p + 1 \right)} \frac{i\omega_p + i\omega_k + E_p(s) + ie}{T(\delta, \beta; r+q, p+s-r; p-r) i\omega_q + i\omega_k + E_p(s) + ie}
\]
\[
= \frac{T(\alpha, \beta; p+k, s; r-p) i\omega_r + i\omega_k + E_p(p+s-r) - ie}{\left( e^{\beta (p+s-r)^2/2m_p} - \beta i p + 1 \right)} \frac{T(\delta, \beta; r-q, p+s-r; p-r) i\omega_r + i\omega_k + E_p(p+s-r) - ie}{e^{\beta (p+s-r)^2/2m_p} - \beta i p + 1}
\]

(2.26)

Note that only the second pair of T-matrices depends on \( w_r \).

Consider now the \( w_r \)-sum of Eq. (2.24). In the complex \( w_r \)-plane both the G's have branch lines by virtue of the T-matrices contained in their self energies. Recalling the selection rules for \( \gamma \) and \( \delta \), \( G(\delta, r+k) \) and \( G(\delta, r+q) \) have branch lines along \( \text{Im} z = -(n+l)\omega_k \) and \( \text{Im} z = -\omega_q - (n+l)\omega_k \) respectively in the complex \( w_r \)-plane. In addition, from Eq. (2.26) for \( \Gamma \) (before carrying out the \( s \) integral), the term in square brackets contributes a pole and the second pair of T-matrices have semi-\( \omega \) branch lines coincident with the full branch lines of the G's. Only the pole does not contribute factors of \( N \) or \( N^N \), and making the classical approximation after collecting terms (necessary for correct statistics) we obtain:

\[ F(\alpha, \beta; p+k, p+q) = \hat{\sum}^{-1} (\alpha, \beta; p+k, p+q) \sum_{\gamma \delta} \int \frac{d^3r}{(2\pi)^3} \int \frac{d^3s}{(2\pi)^3} e^{-\beta (s^2/2m_p - u_p)} \]
\[
\times G(\gamma, r+k) G(\delta, r+q) T(\alpha, \gamma; p+k, s; r-p) i\omega_p + i\omega_k + E_p(s) + ie
\]
\[
T(\delta, \beta; r+q, p+s-r; p-r) i\omega_q + i\omega_k + E_p(s) + ie \hat{\sum} (\gamma, \delta; r+k, r+q)
\]

(2.27)

where, at the pole, \( i\omega_r = R = i\omega_p + E_p(s) - E_p(p+s-r) + 2ie \).
We can now calculate the susceptibility as

\[-S(q, w)\omega^2 = \sum_{\alpha, \beta} \int \frac{d^4p}{(2\pi)^4} G(\alpha, p+k) G(\beta, p+q) \frac{\mathcal{X}(\alpha, \beta; p, q) \mathcal{X}(\alpha, \beta; p+k, q)}{[1 - F(\alpha, \beta; p+k, p+q)]}\]

\[(2.28)\]

In the \(w\)-sum, \(G(\alpha, p+k)\) and \(G(\beta, p+q)\) contribute branch lines in the complex \(w\)-plane along \(\text{Im}z = -(n+1)\omega_k\) and \(\text{Im}z = -(n+1)\omega_q\) respectively. The propagators and T-matrices of \(F\) contribute branch lines parallel to these but displaced slightly due to the \(ie\) terms. The integration contour now looks very much like that for \(\Gamma\) (we assume \(w > 0\) as we shall later want to analytically continue \(iw_q \to w_q + ie\) at a positive \(\omega_q\)).

We proceed as before, picking up poles in the non-physical sheet. Just above and below the upper set of branch lines we have, for example,

\[G(1', p+k, +ie)^{-1} = [iw_p + i(n+1)\omega_k - (n-\frac{1}{2})\omega_k - E(p+k) - \frac{1}{2}\phi(n, p+k) - N_p \langle T(1', p+k, 0) \rangle]|_{iw_p \to \text{Re}z - i(n+1)\omega_k + ie}\]

where \(\langle T(1', p+k, 0) \rangle\) represents in obvious notation the perturber average of Eq. (2.21). Arguments identical to those leading to the determination of the sign of \(e\) yield \(-\text{Im}T(1', p+k, 0)^+ie > 0\) (it is also proportional to a cross section by the optical theorem) and \(T(1', p+k, 0)^{-ie} = T^*(1', p+k, 0)^{+ie}\).
Hence \( G(l', p+k, \pm i\varepsilon) \) has a pole in the \( \{ \text{lower} \} \) non-physical half-plane.

We find as for the integration for \( \Gamma \) that only the inner two branch line integrals need be considered, and that in this case the contribution from the lower is \( -e^{-\beta \omega q} \) times that for the upper. This represents stimulated emission, and we will henceforth not include this term in our calculations.

We close the upper branch line in the upper half-plane, picking up the \( G(\alpha, p+k, -i\varepsilon) \) pole (label it \( J_\alpha \)), in the neighbourhood of which both \( G(\beta, p+q) \) and \( G(\delta, r+q) R+i\omega_q \) are slowly varying and nearly equal. In Eq. (2.27) we therefore cancel these in the term \( \Gamma \) and write the contribution to \( -S(q, \omega_q) \omega_q^2 \) as

\[
\text{Sum} \int \frac{d^3\mathbf{p}}{(2\pi)^3} \frac{\pi \alpha(\alpha; p+q, p+k) \pi \alpha(\beta; p+k, p+q)}{[G(\beta, p+q)]^{-1} J_\alpha - \Gamma'(\alpha, \beta; p+k, p+q) | J_\alpha]} \tag{2.29}
\]

where \( \Gamma'(\alpha, \beta; p+k, p+q) | J_\alpha \) is \( \{ \Gamma(\alpha, \beta; p+k, p+q) \) with the term \( G(\delta, r+q) R+i\omega_q \) eliminated \( | J_\alpha \).

Consider a typical case; \( \alpha = 1', \beta = 3'. \) The contribution is

\[
C_{1', 3'} = \omega_2 \int_0^2 d\omega \frac{v(n, p)}{v(n+1, p+q)^2} \frac{d^3\mathbf{p}}{(2\pi)^3} \frac{\pi \alpha(\alpha; p+q, p+k)}{N \frac{2\pi \beta}{m}} e^{-\beta p^2/2m} \frac{2}{\omega - \omega_k} \left[ \omega_k + \frac{\tilde{p} \cdot (\tilde{q} - \tilde{k})}{m} \right] N_p \{ \langle T^\ast(1', p+k, 0) \rangle - \langle T(3', p+q, 0) \rangle \}
\]

\[
- \Gamma'(1', 3'; p+k, p+q) | J_1' \] \(-1 \]

where atomic recoil effects have been ignored. In evaluating the \( \Gamma'(1', 3'; p+k, p+q) | J_1' \) term, note that

\[
G(\gamma, r+k) R+i\omega_k | J_\alpha = [\Delta E_{\alpha \gamma} + 2\varepsilon]^{-1}
\]
where
\[ \Delta E_{\alpha\gamma} = \frac{(p+k)^2}{2m} - \frac{(r+k)^2}{2m} + \frac{s^2}{2m} - \frac{(p+s-r)^2}{2m} + \frac{1}{2} \frac{\phi(n,p+k)}{m} \]
\[ + \frac{1}{2} \frac{\phi(n,r+k)}{m} + \sum_{p} \left\{ \langle T^*(\alpha,p+k,0) \rangle - \langle T^*(\gamma,r+k,0) \rangle \right\} \]

and \( I_\eta = \pm 1 \) for \( \eta = 1', 2' \) respectively. This gives rise (ignoring the small principal part) to an energy conserving factor \( \delta(\Delta E_{\alpha\gamma}) \), and this delta function may be used to remove part of the \( \int d^3 r \) integral. This is the reason for retaining \( \pm ie \) in the perturber propagators. A change of variables to a perturber momentum is convenient; \( \bar{r} + \bar{r}' = p+s-r \). Then

\[ \Delta E_{\alpha\gamma} = \frac{(p+k)^2}{2m} - \frac{(p+s-r'+k)^2}{2m} + \frac{s^2}{2m} - \frac{r'^2}{2m} \cdot \]

If \( \frac{m}{p} \ll m \) then \( s, r' \ll p \) so that the delta function leads to \( r' \sim s > 0 \) which is a necessary condition given that the range of possible \( r' \) is 0 to \( \infty \). Note that if \( \frac{m}{p} \) is not \( \ll m \) more complicated substitutions are required (Ross, 1966). Writing

\[ \int d^3 r' = \frac{m}{p} \int d\left(\frac{r'^2}{2m} \right) r' \int d\Omega_{r'}, \]

ignoring the momentum arguments of \( \Xi \) (valid when the Rabi-shift is greater than the Doppler width) and noting that the T-matrices are now on the energy shell we obtain:

\[ F'(\alpha,\beta;p+k,p+q) = i \sum_{\gamma,\delta} \left( \frac{d^3 s}{(2\pi)^3} \right) e^{-\beta(s^2/2mp-\mu p)} \frac{r'mp}{(2\pi)^2} \]
\[ \int d\Omega_{r'}, T^*(\alpha,\gamma;p+k,s;\bar{s}-r') T(\delta,\beta;p+s-r'+q,\bar{r'};\bar{r}'-\bar{s}) / r' = s \]
Thus, with $v_p = r'/m_p$ and using Eq. (2.24),

$$F'(1',3';p+k,p+q)|_{r'} = iN_p v_p \int d\Omega r \left\{ T^*(1',1') T(3',3') \right\}_{r'=s}$$

$$+ \frac{u}{v} T^*(1',1') T(4',3') - \frac{v}{u} T^*(1',2') T(3',3') - T^*(1',2') T(4',3') \right\}_{r'=s}$$

where the momentum arguments of Eq. (2.31) are understood and where

$$< > = \frac{m_p^2}{(2\pi)^2} \int \frac{d^3s}{(2\pi)^3} e^{-\beta s^2/2m} \left( \frac{2\pi\beta}{m_p} \right)^{3/2}.$$

Therefore Eq. (2.30) is almost the convolution of a Doppler profile with a Lorentzian centred on $\omega_k + D$ with (see Eq. (1.2))

$$i\omega + D = -N_p \left\{ \langle T(3',p+q;0) \rangle - \langle T^*(1',p+k;0) \rangle + i \langle v_p \int d\Omega r \{ \cdots \} \rangle \right\}_{r'=s}$$

where $\{ \cdots \}$ is the curly bracket in (2.32). It differs in that $(q-k)$ appears in the Doppler term rather than $q$, and $i\omega + D$ is a (weak) function of momenta. This form is identical to that of Ross (1966), Eckart (1974) and Baranger (1958) with T-matrices now requiring different calculation (Eq. (2.14) and Eq. (2.19)) and $F'$ containing additional terms (Eq. (2.31)).

As the field strength $4\kappa$ → 0 (and ignoring Doppler effects), $u, v \to \frac{1}{2}$ and in Eq. (2.20) all the a-coefficients → $\frac{1}{2}$. The T-matrices are then averages of the upper and lower non-laser T-matrices (T(2) and T(1) respectively) so that in $\{ \cdots \}$ of Eq. (2.32) the middle two terms cancel and the remaining terms yield $T^*(1) T(2)$. The term $C_{1,3}'$ of Eq. (2.30) is then exactly $\frac{1}{4}$ of the susceptibility given by Ross (1966) and Baranger (1958). In an identical fashion (see below) the other three terms in the susceptibility of Eq. (2.23) also give $\frac{1}{4}$ of Ross's result,
so that the non-laser result is regained correctly. In addition, in the no-detuning, no lower state broadening case the coherence term $F'$ goes to zero, as it must (the contribution of coherence terms cannot depend on the choice of the basis set of eigenstates). These results are a powerful check on the preceding analysis.

After some algebra, the remaining terms in the susceptibility are:

$$C_{2,3}' = \omega_{21} \frac{d^2}{d_0} v(n,p) v(n+1,p+q) \frac{d^3p}{(2\pi)^3 N} \frac{1}{m^{3/2}} e^{-\beta_p^2/2m}$$

$$\times \left[ \omega_q - \omega_k + \phi - \frac{p^*(q-k)}{m} \right] + N_p \left\{ <T^*(2',p+k;0)> - <T(3',p+q;0)> \right\}$$

$$- F'(2',3';p+k,p+q) \bigg|_{J_2^1}^{-1}$$ (2.33)

where the difference between $\phi(n,p+k)$ and $\phi(n+1,p+q)$ has been ignored, and

$$F'(2',3';p+k,p+q) \bigg|_{J_2^1} = i N_p \left\{ - \frac{u}{v} T^*(2',1') T(3',3')$$

$$- \frac{u^2}{v^2} T^*(2',1') T(4',3') + T^*(2',2') T(3',3') + \frac{u}{v} T^*(2',2') T(4',3') \right\} \bigg|_{r'=s}$$ (2.34)

Note that the peak is shifted by the Rabi shift and has a different width and height to the $1',3'$ peak. The $C_{2,4}'$ term yields a result similar to
the \( c_{1',3'} \) term i.e. a peak centred on the laser frequency \( \omega_k \) (assuming the shift \( D \) is small), while the \( c_{1',4'} \) term gives a peak shifted by the Rabi shift on the opposite side of the laser to the \( c_{2',3'} \) peak. These terms are

\[
C_{2',4'} = \omega_{21} \int_{0}^{2} d_0 \left( \frac{1}{2} u(n,p)^2 u(n+1,p+q)^2 \right) \int \frac{d^3 p}{(2\pi)^3} N \left( \frac{2\pi R}{m} \right)^{3/2} e^{-\beta p^2/2m} \\
\times \left[ \omega_q - \omega_k - \frac{p^*(q-k)}{m} + N_p \{ \langle T^*(2',p+k;0) \rangle - \langle T(4',p+q;0) \rangle \} \\
- F'(2',4';p+k,p+q) \right]_{J_2}^{-1}
\]

(2.35)

with \( F'(2',4';p+k,p+q) \mid_{J_2} = i N_p \left\{ \int d\Omega_r \left( -T^*(2',1') T(3',4') - \frac{u}{v} T^*(2',1') T(3',4') + \frac{v}{u} T^*(2',4') T(3',4') \right) \right\}_{r'=s}^r \)

(2.36)

And \( c_{1',4'} = \omega_{21} \int_{0}^{2} d_0 \left( \frac{1}{2} u(n,p)^2 u(n+1,p+q)^2 \right) \int \frac{d^3 p}{(2\pi)^3} N \left( \frac{2\pi R}{m} \right)^{3/2} e^{-\beta p^2/2m} \\
\times \left[ \omega_q - \omega_k - \phi - \frac{p^*(q-k)}{m} + N_p \{ \langle T^*(1',p+k;0) \rangle - \langle T(4',p+q;0) \rangle \} \\
- F'(1',4';p+k,p+q) \right]_{J_1}^{-1}
\]

(2.37)

with \( F'(1',4';p+q,p+q) \mid_{J_1} = i N_p \left\{ \int d\Omega_r \left( -T^*(1',1') T(3',4') - \frac{u}{v} T^*(1',2') T(3',4') + \frac{v}{u} T^*(1',2') T(4',4') \right) \right\}_{r'=s}^r \)

(2.38)
Two further points require discussion. **Assuming no detuning**, ignoring Doppler corrections and supposing $n \gg 1$, it is clear that (bearing in mind the form of our T-matrices) **it is the $F'$ terms which are responsible for any differences in the widths of the side peaks compared to the central peak.** This is a result which is easy to understand. The $F'$ terms represent coherent interactions between initial and final dressed-emitter states. In the absence of such coherence, the four states represented in Fig. 1.1 act independently and there is no mechanism capable of enhancing one transition rate over another. With coherence, a more complicated set of decay paths is available and different widths become a possibility. When there is laser detuning, the solution is more complex and is discussed in section 2.6.

Finally, we must consider the implications of our thermal Green's function assumption. Provided $n \gg 1$ (i.e. the laser is above saturation) then the dependence of the susceptibility on $n$ is extremely weak. As inelastic collisions populate states $n \pm 1, n \pm 2, \ldots n \pm j$ so the original ratio $l; e^{-\omega_2 I}$ changes and our ensemble is no longer a thermal one. Transitions $n \pm j \leftrightarrow n \pm j + 1$ occur, but because of the weak $n$-dependence we hypothesise that the line profile will remain unchanged. This hypothesis enables us to greatly extend our validity criterion to include experiments of duration less than the time required for significant population of states of such different $n$ that differences in the profile are also significant. This time is much longer than our original limit.
2.6 Susceptibility Profiles and Conclusions

Rather than evaluate the T-matrix integrals explicitly, in itself a difficult process and in the present case involving integrals of a form not considered in previous calculations, we will assume that our T-matrices behave very much like the hard-sphere T-matrices of section 2.3. To this level of approximation, all the T-matrices are pure imaginary (a good approximation for Na \((3^2S)\) and \((3^2P)\) in He at \(5 \times 10^3 \, ^0\text{K}\) at which conditions the D-lines show almost no shift (Eckart, 1975)). Our T-matrix integrals then become simple sums of the well-known (non-dressed) emitter upper and lower state T-matrix integrals, the coefficients being combinations of those in Eq. (2.20).

We introduce the parameters \(W^0\), the non-dressed emitter collisional \(\frac{1}{2}-\frac{1}{2}\) width;

\[ Q = \frac{\langle T(1) \rangle}{\langle T(2) \rangle}, \]

the ratio of lower state to upper state broadening; and

\[ R = i \langle \nu_p \int d\Omega_r T^*(1) \, T(2) \rangle / \langle T(2) \rangle, \]

the ratio of the coherent broadening to the upper state broadening. The result of ordinary collision broadening theory is that (assuming zero shift)

\[ iW^0 = - \frac{N}{p} \left\{ \langle T(2) \rangle - \langle T^*(1) \rangle + i \langle \nu_p \int d\Omega_r T^*(1) \, T(2) \rangle \right\} \quad (2.39) \]

from which it follows that

\[ - \frac{N}{p} \langle T(2) \rangle = iW^0/(1+Q+R) \quad (2.40) \]
Ignoring Doppler broadening, the imaginary part of the susceptibility profile (un-normalized) is then the sum of four terms

\[ C_{1',3'} = -u^2 v^2 W_{1',3'} \left[ (\omega_2 \omega_2 - \Delta)^2 + W_{1',3'} \right]^{-1} \]

\[ C_{2',3'} = -u^2 v^2 W_{2',3'} \left[ (\omega_2 \omega_2 - \Delta + \Phi)^2 + W_{2',3'} \right]^{-1} \]

\[ C_{2',4'} = -u^2 v^2 W_{2',4'} \left[ (\omega_2 \omega_2 - \Delta)^2 + W_{2',4'} \right]^{-1} \]

\[ C_{1',4'} = -u^4 W_{1',4'} \left[ (\omega_2 \omega_2 - \Delta - \Phi)^2 + W_{1',4'} \right]^{-1} \] (2.41)

where

\[ W_{1',3'} = \frac{W^0}{(1+Q+R)} \left\{ 2M_1 + \frac{R}{Q} \left[ M_1^2 + M_1 M_2 \left[ \frac{u}{v} - \frac{v}{u} \right] - \frac{M_2^2}{u^2} \right] \right\} \]

\[ W_{2',3'} = \frac{W^0}{(1+Q+R)} \left\{ 1 + Q + \frac{R}{Q} \left[ M_1 M_3 + \frac{u}{v} M_2 \left[ M_3 - M_1 \right] - \frac{M_2^2}{u^2} \right] \right\} \]

\[ W_{2',4'} = \frac{W^0}{(1+Q+R)} \left\{ 2M_3 + \frac{R}{Q} \left[ M_3^2 + M_2 M_3 \left[ \frac{v}{u} - \frac{u}{v} \right] - \frac{M_2^2}{v^2} \right] \right\} \]

\[ W_{1',4'} = \frac{W^0}{(1+Q+R)} \left\{ 1 + Q + \frac{R}{Q} \left[ M_1 M_3 + \frac{v}{u} M_2 \left[ M_1 - M_3 \right] - \frac{M_2^2}{u^2} \right] \right\} \] (2.42)

and where

\[ M_1 = (a^2 + a^2) Q + a^2 + a^2 \]

\[ M_2 = (a^2 a^2 + a^1 a^3) (1-Q) \]

\[ M_3 = (a^2 + a^2) Q + a^2 + a^2 \] (2.43)
The susceptibility is thus the sum of four Lorentzians (to this order of approximation) whose relative weights, positions and \( \frac{1}{2} - \frac{1}{2} \) widths depend on laser and broadening parameters.

Measured values of \( W^0 \) are available (Eckart, 1975 and Baird et al., 1979; and see section 4.4 below) while estimates of \( Q \) and \( R \) may be made from the interatomic potentials discussed by Wilson and Shimoni (1975(a) and (b)). Fourier transforming the momentum space potential

\[
V(\vec{k}) = \begin{cases} 
V & k < k' \\
0 & k \geq k'
\end{cases}
\]

we obtain

\[
V(\vec{r}) = \frac{2V}{(2\pi)^2} \frac{[\sin(k'r) - k'r \cos(k'r)]}{r^3}
\]

which, as a function of \( k'r \), represents a decaying step potential with oscillations superimposed, looking qualitatively like the potentials of Wilson and Shimoni. On this basis, we estimate \( Q \) to be \( 0.4 \pm 0.15 \). \( R \) may be evaluated by approximating the terms \( \langle \mathbf{v} \int d\Omega T^*(1) T(2) \rangle \) and \( \langle T(2) \rangle \) by substituting the hard sphere \( T \)-matrices and calculating the integrals at the peak of the emitter and perturber Maxwellian momentum distributions. Determining an absolute value for \( k' \) from \( V(\vec{r}) \), this yields \( R \approx -0.5 \) for a temperature of 4,500 °K, but this is expected to be a poor approximation. For this reason, \( R \) was left as a parameter in the susceptibility curves of Figs. 2.2 to 2.5, all of which have the same vertical scale and integrated area. \( R \) is clearly bounded by \(-1 < R < 0\).
Fig. 2.2: $\Delta = 0$, $R = 0$, $Q = 0.4$. Curves are:

- $\Omega = 0$ Unpumped Lorentzian,
- $\Omega = 4\omega^0$ Three-peaked profile.
Fig. 2.3: $\Delta = 1, \ R = -0.5, \ Q = 0.4$

$\Omega = 0$ Shifted Lorentzian

$\Omega = 4W^0$ Three-peaked profile
Fig. 2.4: $\Delta = 1$, $\Omega = 4\omega^0$, $Q = 0.4$ Curves are:
R = 0  Lower central peak, higher wings
R = -0.5 Higher central peak, lower wings
Fig. 2.5: $\Delta = 2$, $\Omega = 4W^0$, $Q = 0.4$. Curves are:

- $R = -0.1$ Lower central peak, higher wings
- $R = -0.4$ Higher central peak, lower wings
Several interesting points emerge from examination of Eq. (2.42). Because \( R/Q \) goes to a constant as \( Q \) (and thus \( R \)) goes to zero, it is clear that the widths of the four Lorentzians are not necessarily equal even in the absence of lower state broadening. They are the same if and only if the detuning is zero, in which case \( u = v \), all the \( a_i = \frac{1}{2} \), \( M_1 = M_3 = (1+Q)/2 \) and \( M_2 = (1-Q)/2 \) and the profile consists of three peaks all with \( \frac{1}{4} \) width \( \omega^0 \) and the central peak twice the height of the side peaks.

When the laser is detuned, coherence terms survive even when \( Q = 0 \) because mixing of the original eigentstates by the laser field ensures that the four-point function \( \Gamma \) still contains \( \int d\Omega_T \ T^4(2) T(2) \) terms.

Mollow (1977) has introduced phenomenological relaxation coefficients into a density matrix equation of motion for a two level collisionally damped atom in the presence of a strong field and has obtained the emission spectrum (his Eq. (4))

\[
f(\omega) = \frac{S_o A_o}{[(\omega - \omega_2)^2 + \sigma_o^2]} + \frac{\sigma A_+}{[(\omega - \omega_2 - \Delta)^2 + \sigma^2]} + \frac{\sigma A_-}{[(\omega - \omega_2 - \Delta + \phi)^2 + \sigma^2]}
\]

where Rayleigh scattering has been ignored, and where

\[
S_o = \frac{(r_1 \Delta^2 + r_2 \Omega^2)}{\phi^2}
\]

\[
\sigma = \frac{2 r_1 \Omega^2 + r_2 (\Omega^2 + 2 \Delta^2)}{2 \phi^2}
\]

\[
A_o = \frac{\eta^2 \Omega^2 + (2\eta - 1) \Delta^2}{\phi^2 (\eta \Omega^2 + \Delta^2)^2}
\]

\[
\phi^2 = \Omega^2 + \Delta^2
\]

\[
A_+ = \frac{\frac{1}{\phi^2} \eta^2 (\phi + \Delta) [\eta (\phi + \Delta) + \Delta]}{(\phi^2 (\eta \Omega^2 + \Delta^2))}
\]

\[
(2.44)
\]

\[
A_- = \frac{\frac{1}{\phi^2} \eta^2 (\phi + \Delta) [\eta (\phi + \Delta) - \Delta]}{(\phi^2 (\eta \Omega^2 + \Delta^2))}
\]

\[
(2.45)
\]
with \( r_1 = R_1 \), \( r_2 = \frac{1}{2}(R_1 + R_E) \) and \( \eta = \frac{R_E}{R_1} \). \( R_1 \) and \( R_E \) are respectively the inelastic and elastic mean collision rates by which the two level system is de-excited and dephased. The above profile is valid for \( \phi \gg r_1, r_2 \), and depends strongly on the ratio \( \eta \). When \( \eta = 1 \) (the strong collision case) the profile is three peaked with all peaks having the same \( \frac{1}{2}r_2 \) widths \( r_2 \) and the central peak twice the height of the side peaks. The evaluation of \( r_2 \) (at least when Doppler-pressure broadening correlations are unimportant) is a familiar problem in line broadening theory (Baranger, 1958; Omont, Smith and Cooper, 1972) and leads to \( r_2 = W^0 \). Thus, when the laser is not detuned, the theoretical spectrum developed in this thesis agrees exactly with Mollow's strong collision result.

The reasons for this agreement are two-fold. In making the hard-sphere and forward scattering approximations we assume that the probability of transferring momentum \( \vec{p} \) during a collision is a constant for all \( p \) smaller than some cutoff momentum, and that this is independent of the relative kinematics of the scattering particles (we could, for example, imagine the scattering Na atom to be replaced by a scattering centre of force). Thus the collisions are of the 'black sphere' absorbing type described by Mott and Massey (1965, p.326) in which incoming particles described by a certain energy are 'destroyed' and new particles described by a new energy are 'created'. For these collisions, \( R_E = R_1 \) from which follows the observed agreement. In addition, the approximations made in deriving the final transformed vertex function \( F \) demand that changes in \( \frac{1}{2}r_2 \) widths due to coherence terms must be small compared to \( W^0 \) if our approximation is to be a good one. This limitation of smallness may be removed by iterating the solution for \( F \) given by Eq. (2.24). Calculations based on the use of accurately determined T-matrices corresponding to realistic interatomic potentials will similarly provide solutions for general values of \( R_E, R_1 \).
It is worth noting that both theories yield profiles satisfying the general condition for no detuning that the integrated central peak area is twice that of each of the side peaks. This follows from the fact that for no-detuning the mixing coefficients for the levels of Fig. 1.1 described by Eq. (2.4) are all equal, and thus effective oscillator strengths for each of the possible four transitions are equal. This condition is also satisfied by the no-collision profile (Mollow, 1969).

When the laser is detuned the two theories no longer give exactly the same profile. Mollow predicts that the side peak and central peak widths will remain unchanged, while the present work predicts small changes resulting from the fact that the mixing coefficients are no longer all equal and thus each eigenstate contains different proportions of $|1\rangle$ and $|2\rangle$. Such a result is intuitively plausible and would serve as the basis for an experiment to discriminate between the two theories.

In summary, we have demonstrated that a many-body quantum electrodynamic approach may be used to derive a profile that agrees, in the case of no-detuning, with that of Mollow. All the details of the collision mechanism appear in a natural way, the modifications to the coherent processes caused by the strong external field are clearly elucidated and the kinematics of the colliding atoms (Doppler broadening) are consistently taken into account in a straightforward way. In addition, at each step the approximations related to the dilute perturber assumption are clearly set out, so that the generalisations required for the inclusion of three-body collisions and other dense perturber effects are evident.

The present theory can be extended to include radiative transitions to a third level (not coupled to the laser) from the set of dressed-emitter levels. It is easy to show that a two-peaked Autler-Townes like profile results.
Finally, we note the limitations of the above Feynman diagram approach. It is difficult to see how it might be used to describe non-steady state situations, and is severely limited by the short time criterion described in section 2.5. The latter would seem to be an area of possibly fruitful future work.

The mathematical theory developed above is being submitted as a paper for publication in the Journal of Physics A.
3. EXPERIMENTAL DESIGN

3.1 Experiment

In order to investigate the theoretical model developed in section 2 an experiment was designed to examine the behaviour of Na atoms interacting simultaneously with a bath of He perturbers and the field from a high-power, pulsed dye laser tuned to one of the Na D-lines. Layout of the experiment is shown in Fig. 3.1.

After reflection at the back wall of the shock tube the shocked gas, which is stationary in the laboratory frame, was pumped by the pulsed laser orthogonally to the shock tube and emission or absorption was observed simultaneously along an axis normal to both these directions. A large collision-broadened Na D-line $\frac{\lambda}{2}$ width lowers demands on the narrowness of the pulsed laser bandwidth (Kimble and Mandel, 1977) and on the resolution of the detection optics, but increases the laser power density, required to fully resolve the Rabi shifted side peaks from the central peak in the emission profile. A set of compromise conditions was determined to be:

- Na D-line collision broadened $\frac{\lambda}{2}$ width of approximately $1^\circ$

Corresponding to a He number density of $\sim 10^{20}$ cm$^{-3}$ and kinetic temperature of $4 - 6 \times 10^3$ °K. Such conditions are easily obtainable in shock tubes, are of astrophysical interest, offer the possibility of highly resolved profiles (detection optics), mean that Doppler broadening can be neglected as insignificant, and correspond to shock tube operating conditions which provide reasonable pressure differentials across incident and reflected shocks enabling the use of the pressure transducers described in section 4.1 as shock timing stations.
Fig. 3.1: Experimental layout
the above conditions require a laser with bandwidth no larger than 0.4 - 0.6 Å, for well defined Rabi-shifted peaks, and a power density in the interaction volume of some 4 MW cm\(^{-2}\) to yield a Rabi shift equal to approximately twice the \(\frac{1}{2}-\frac{1}{4}\) width of the collision-broadened line in the absence of the laser (Wu et al., 1975). With the measured detection system resolution of 0.2 Å, power densities of 1 MW cm\(^{-2}\) would still provide an acceptably large change in the emission profile to be accurately measurable. These power levels are easily obtained using Xe-flashlamp pumped dye lasers.

At such shock tube conditions, Na, which is typically a residual and not always easily controllable impurity, exists at number densities in the range \(10^{12} - 10^{16}\) cm\(^{-3}\). To ensure observable optical depths the interaction volume optical axis must be 0.1 - 1 cm in extent. Taking into account possible shock tube boundary layer effects and the desirability of pumping all of the shocked gas uniformly (to avoid observation through unsaturated atoms, fringe effects if non-linear processes are shown to be important and Abel inversions) a square shock tube of 1 cm\(^2\) section is indicated.

Several potentially significant non-linear processes require consideration. Coherent interaction of Na atoms through intercoupling via the laser field (see for example Arecchi and Courtens, 1970) is possible in principle. Typically, the laser irradiates a slug of gas 1 cm × 0.3 cm in section by 1 cm deep in which Na number density is of the order of \(3 \times 10^{13}\) cm\(^{-3}\). The corresponding maximum co-operation number \(N_c\), the co-operation time \(\tau_c\) and the maximum co-operation length are \(2.3 \times 10^{10}\), \(2.2 \times 10^{-13}\) s and \(6.6 \times 10^{-3}\) cm respectively. Thus, were they to be initially in a superradiant Dicke state, coherent emission from a group of some \(2.3 \times 10^{10}\) Na atoms would occur. Throughout the gas slug there would be many such groups, radiating coherently within themselves but
incoherently with respect to other groups. Averaged over an observation time of many $\tau_c$ these rapid, enhanced emissions would not be detectable and, because of superradiant narrowing into a small forward cone, the nett effect would be preferential forward scattering of the laser and almost no signal at other angles. Mitigating against this are collisions, occurring on average every $6 \times 10^{-12}$ s and which tend to destroy coherence, and the fact that the system is never actually in a Dicke superradiant state, being stimulated towards states of lower total (pseudo) angular momentum long before the maximum total angular momentum of $4N_c$ can be attained. This latter constraint ensures that orthogonal emission experiments are still possible.

A second process, restricted to absorption experiments, is amplification rather than attenuation of a weak probe beam in certain parts of the absorption profile (Mollow, 1972(b) and Wu et al., 1977). The difference between the emission and absorption profiles is that in the latter the probe beam can interact with the laser beam and atom in a non-linear multiphoton process (see Fig. 4(c) of Wu et al.) which under certain circumstances (of very intense laser fields) effectively removes laser photons and creates additional probe photons. Such processes are explicitly excluded from the formalism of section 2, whose linear response, thermodynamic basis carries the inherent assumption that emission and absorption profiles are the same for optically thin sources. Observation of emission, rather than absorption, is then a test for the results of section 2.

A further non-linear process is laser induced ionization of Na to Na$^+$ (Lucatarto and McIlrath, 1976; McIlrath and Lucatarto, 1977; Bearman and Leventhal, 1978; Skinner, 1980) via collisions between excited Na atoms and super- elastically heated electrons $e^-$.
The super-elastically heated electrons arise from collisions between seed electrons (supplied by residual impurities in the case of shock tubes) and the excited atom population maintained by the laser:

\[ \text{Na}(3^2\text{P}) + e \underset{\text{se}}{\rightarrow} \text{Na}(\text{autoionizing}) + e \rightarrow \text{Na}^+ + 2e \]

Such processes also occur in the absence of the laser, due to electrons in the far Maxwellian wing. By suddenly increasing the excited atom population the laser causes a (non-equilibrium) enhancement of these highly energetic electrons. As part of the relaxation process towards the new thermal equilibrium represented by the energy source of the laser, the super-elastically heated electrons transfer energy from the laser to the Na atoms, causing ionization.

Such processes can be remarkably efficient. Lucatarto and McIlrath (1976) observed approximately 90% ionization in an oven containing Na at \(10^{16} \text{ cm}^{-3}\) by pumping the 5896 Å line with a 500 ns, 1MW cm\(^{-2}\) pulsed dye-laser with a bandwidth of \(\sim 0.5 \text{ Å}\). Similar results have been reported by other authors (see Skinner, 1980). On the other hand, Bearman and Leventhal (1978) observed much lower levels of ionization in an oven containing Na at \(10^{13} \text{ cm}^{-3}\) pumped by a cw dye laser producing \(10^2 \text{ W cm}^{-2}\) in a bandwidth of 40 GHz.

Efficient ionization relies on the laser maintaining saturation in the pumped transition and on the mean free path of the electrons being such as to enable sufficient collisions to occur to super-elastically heat the electrons and then to ionize the Na atoms by electron impact. In Bearman and Leventhal's experiment neither of these conditions was satisfied, the
estimated electron mean free path through Na(3P) of 25 cm being some 40 times the cell diameter of 6 mm. Since only two Na(3P) + e → Na(3S) + e collisions are required before the electrons have sufficient energy to impact ionize the excited atoms, significant ionization in a 1 cm cube interaction volume might be expected when the Na(3P) density > 5 × 10^{14} cm^{-3}. This is supported by the results of Skinner, who observed a rapid decrease in the percentage ionization for Ca, Ba and Sr below approximately 2 × 10^{14} cm^{-3}. A theoretical model for the ionization process has been reported by Measures (1977 and 1979) who solves numerically a set of coupled rate equations describing radiative and collisional processes into and out of the resonance states, in the absence of foreign perturbers (other species of atoms). The addition of collisions with He alters this model in several ways, the most important being that there is now a sink for the energy of the super-elastically heated electrons. Ionization criteria depend on relative Na and He densities and electron impact cross sections. Blaauw et al. (1980) give a total e-He cross section of σ_{e-He} = 5.9 × 10^{-16} cm^2 for electrons with kinetic energy of a few eV; Bearman and Leventhal (1978) quote σ_{se} = 80 × 10^{-16} cm^2 for the e + Na(3P) → e + Na(3S) process (for a single collision) and an estimate of σ_+ = 2 × 10^{-15} cm^2 may be made for the electron impact ionization cross section from Na(3P), based on the work of Kunc (1980). Because of the large mass ratio, the He atoms can absorb very little energy per collision from the fast electrons. Let a typical dimension of the interaction volume be l_0. Because 3σ_+ << 2σ_{se}, of the three mean free paths

\[ l_{se} = 3(n_{Na} \sigma_{se})^{-1} \]  (two collisions required)

\[ l_{p} = 2(n_{He} \sigma_{e-He})^{-1} \]

\[ l_{+} = 2(n_{Na} \sigma_{+})^{-1} \]
the ionization criterion will depend primarily on $l_r/l_p$. Let $l_o = x(l_r + l_{se})$

Then significant ionization can be expected if

$$\frac{n_{He} \sigma_{e-He}}{n_{Na} \sigma_{+}} \leq M_r \left(1 - \frac{1}{x}\right) x \geq 1$$

where $M_r = 7.34 \times 10^3$ is the mass ratio of He to e. For $l_o \sim 1$ cm and $n_{Na} \sim 3 \times 10^{13}$ cm$^{-3}$, $l_{se} \sim 12.5$ cm and ionization is clearly not expected. If $n_{Na} \sim 3 \times 10^{15}$ cm$^{-3}$, then $x \sim 2$ and ionization is expected if $n_{He} \leq 3 \times 10^{19}$ cm$^{-3}$. Note that in these equations, $n_{Na}$ is the number density of Na($^2P$).

In an actual experiment, the changing intensity of the laser with time and the non two-level nature of the Na atom produce difficulties for the interpretation of measured profiles in terms of the expressions developed in section 2. The former can be eliminated, at least in part, by shuttering the detection optics so as to switch out all but the central, flat part of the laser pulse. Polarization of the laser output can be used to eliminate effects arising from the degenerate magnetic sublevels of the Na($^2S$) and Na($^2P$) states (see for example Shore, 1978). If the laser output is linearly polarized and tuned to the 5890 Å line, only the $M_J = \frac{1}{2} \rightarrow M_J = \frac{1}{2}$ and the $M_J = -\frac{1}{2} \rightarrow M_J = -\frac{1}{2}$ transitions will be pumped. The Rabi-shifts for these are the same. Collisional relaxation amongst the sublevels to $M_J = \pm\frac{3}{2}$ is so fast that equilibration can be assumed over any experimental observation time. The 5890 Å line profile in emission will thus be the sum of a three-peaked profile for transitions originating from $M_J = \pm\frac{3}{2}$ and a two-peaked profile for transitions originating from $M_J = \pm\frac{1}{2}$, with the widths, shifts and peak heights given by the theory of section 2. It is easy to see from Fig. 3.2 that these contributions may be partly separated by a polarization analysis of the emission signal and,
in particular, that if a linear analyzer is used the observed profile is that of a two-level system such as considered in section 2.

![Diagram of energy levels and transitions](image)

**Fig. 3.2** Full lines represent the pumping laser, wavy lines linearly polarized emission and broken lines circularly polarized emission.

The above assumes that the sublevel collisions do not mix the $M_J$ quantum states sufficiently to invalidate the polarization selection rules, a situation of greatly increased complexity. Similarly, we have ignored the presence of the Na($3^2P_{\frac{3}{2}}$) states, so that the 5896 Å line profile in this approximation would appear unaffected by the laser, but enhanced in intensity over that observed without the laser (sensitized resonance fluorescence). Both of these assumptions may be unrealistic but are in any case experimentally verifiable. Helium collisions which give rise to diagrams of the form

![Detailed diagram of helium collision](image)
are those that are responsible for the non-Lorentzian terms that appear in the spectral profile of overlapping lines. This is further discussed in section 4.4, in which the results of an experiment to measure this non-Lorentzian behaviour are presented and it is found that to a very good approximation the contribution of such diagrams is zero and the two Na D-lines can be considered isolated lines (in the sense of Baranger, 1958(b)). This could be due to either of two causes. The potential represented by the diagram

\[
\begin{align*}
\alpha & \rightarrow \beta \\
| r | \\
| | \\
| | \\
\end{align*}
\]

\[\alpha, \beta = \begin{cases} 
3^2 \text{P}_{3/2} \\
3^2 \text{P}_{3/2}
\end{cases} \quad \alpha \neq \beta\]

could be identically zero, or the collisions represented by the T-matrices might not be capable of changing the spin the lower propagator, a necessary condition if momentum is to be conserved. The first is unlikely in practice (it also describes inelastic collisions). Therefore observation of a non-Lorentzian component in the 5896 Å line while the 5890 Å line is being pumped tests for the presence of any spin-flip behaviour induced by the laser.

3.2 Shock Tube Design

Experiments were conducted on a free-piston shock tube (Stalker, 1967) shown schematically in Fig. 3.3. Upon firing, compressed air in the reservoir accelerates a mild steel piston into the compression tube helium such that the unscored aluminium diaphragm of 2,3000 psi burst pressure is just ruptured at peak compression. The resulting shock wave propagates into
Fig. 3.3: Shock tube schematic.
Fig. 3.4: Shock tube construction.
the helium in the shock tube and after reflection at the end wall typical steady conditions of helium temperature 5000 °K and number density $10^{20}$ cm$^{-3}$ last for approximately 30 μs. As described above, a shock tube of ~1 cm square cross section is desirable. Although knife-edged square tubes have been used to slice out suitable sections at the exits of circular shock tubes (Baird, 1977), there are uncertainties in gas conditions due to the complex shock behaviour at the leading edges. Bow shocks form at the entry, attached or unattached depending on the composition of the gas and the wedge angles of the knife edges. These then interact via a complicated series of reflections.

The design of the square shock tube had to take into account non-uniform stress and vacuum sealing problems and reflected shock pressures of up to 20,000 psi (allowing a design factor of four). Eventual construction was the annular-collar diagonally-split design shown in Fig. 3.4. An 18 inch section of $\frac{3}{4}$" diameter tube immediately follows the diaphragm to maintain even, symmetric rupture. A smooth transition of constant cross sectional area over a distance of 1" precedes the 36" long .44" square section. This length is sufficient to reduce rupture and transition caused shock-front disturbances to insignificance at the test section. Four flush-mounted windows are orthogonally positioned .64" from the end-wall, as indicated in Fig. 3.4. The whole structure is mounted on a wooden frame set in a steel base, which provides complete vibration isolation from other laboratory equipment. Recoil, photographed during test times, is 3.75 ± .25 mm and is reproducible over many shots. Provided the shock tube is evacuated to approximately .03 mm Hg prior to firing, shot to shot variation in shock speed of less than 2% can be obtained. Performance decreases slowly, due to wear on piston compression rings, which are replaced after approximately 100 shots.
A satisfactory description of the gas behaviour is provided by the perfect gas relations for normal shocks at constant cross-section (see for example Liepmann and Roshko, 1957), there being negligible ionization of helium at 5000°K. Across the incident shock, the equations of continuity and momentum and energy conservation yield

\[
\frac{\rho_2}{\rho_1} = \frac{u_1}{u_2} = \frac{(\gamma+1)M_s^2}{[(\gamma-1)M_s^2 + 2]} \quad (3.1)
\]

\[
\frac{P_2}{P_1} = 1 + \frac{2\gamma}{(\gamma+1)} (M_s^2 - 1) \quad (3.2)
\]

\[
\frac{T_2}{T_1} = \frac{\rho_1}{\rho_2} \cdot \frac{P_2}{P_1} \quad (3.3)
\]

where \( M_s = \frac{u_1}{a_1} \) is the incident shock Mach no., \( a_1 \) is the pre-shock speed of sound, \( \gamma \) is the ratio of specific heats, equal to \( \frac{5}{3} \) for He, and other variables are defined in shock stationary co-ordinates in Fig. 3.5.

Reiteration for the jump across the reflected shock requires only the reflected shock Mach no. \( M_R \):

\[
M_R = K + \left[1 + K^2\right]^{\frac{1}{2}}
\]

\[
K = M_s\left(1 - \rho_1/\rho_2\right) \sqrt{\frac{T_1}{T_2}} \frac{\gamma + 1}{4} \quad (3.4)
\]
Incident Shock

\[ u_2, \rho_2, T_2, P_2 \] \hspace{1cm} \[ u_1, \rho_1, T_1, P_1 \]

Reflected Shock

\[ u_1 - u_2 + U_R \] \hspace{1cm} \[ U_R, \rho_5, T_5, P_5 \]

Fig. 3.5: Definitions of shock - stationary variables.
Determination of the conditions of the stationary gas (in the laboratory frame) between the reflected shock and the end wall thus requires a knowledge of $P_1$, $T_1$ and $u_1$. Typical experimental values are $P_1 = 380$ mm Hg, $T_1 = 294^\circ$K, $u_1 = 4.64 \times 10^3$ m s$^{-1}$ which produces $M_s = 4.60$, $M_R = 2.08$, $u_R = 2.43$ km s$^{-1}$, $T_5 = 4.80 \times 10^3$ °K, $\rho_5 = 1.03 \times 10^{20}$ cm$^{-3}$ and $P_5 = 9.95 \times 10^3$ psi.

3.3 Optics

The dye-laser was based on a Phase-R type DL-15B co-axial, annular-quartz, Xe-flashlamp pumped laser head having a cylindrical active dye volume of $\sim 28$ cm in length and $\sim 1.3$ cm in diameter. Rhodamine 6G dye of $5 \times 10^{-5}$ molar in absolute ethanol was filtered to 2 $\mu$m and settled to remove bubble and contaminant scattering centres, cooled to below $\sim 22^\circ$C to reduce laser-inhibiting thermal gradients and pumped at from 0 to $\sim 5$ litres/minute via a magnetically coupled dye pump. Two high voltage fast discharge capacitors were dumped in series at 30 kV through the laser head via dry air pressurized triggerable spark gaps. In an untuned cavity with a 40% output mirror approximately 3J in 500 ns was produced, with the pulse shape shown in Fig. 3.6. Tuning was achieved with two high dispersion prisms, whose flat faces at approximately the Brewster angle produced nearly complete linear polarization. An air-space calcite polarizer placed within the cavity ensured a completely linearly polarized output beam, whose plane of polarization could be rotated using a suitably biased Inrad 250 series Pockels cell. With the output mirror stopped down to 1 cm diameter typical peak power densities of approximately 0.6 MW cm$^{-2}$ were measured, using a 163 $\mu$VJ$^{-1}$ calibrated thermopile and a $\mu$V-meter. Far-field patterns and burn spots on emulsion targets indicated
Fig. 3.6: Typical laser pulse

Fig. 3.8: OMA gating unit output
Overlay of 3 shots
1000X attn.
Fig. 3.7: OMA gating unit
that the power density was extremely uniform, and that the half-angle beam divergence was approximately 0.1 mrad. At the expense of a slight increase in divergence, removal of the 1 cm output stop produced a beam of similar power density over a 1.4 cm² spot. Pulse shape remained as for Fig. 3.6. Careful focussing of these beams using a combination of one or both of a plano-cylindrical lens and a plano-convex lens enabled an interaction volume to be defined which in section ranged from a 1 cm diameter circle (at 1 MW cm⁻², used for absorption experiments in which it was necessary to pump all of a 3 mm vertical column defined by the smaller shock tube windows), through a 1 cm x 3 mm vertical rectangle (at approximately 2 MW cm⁻³, for absorption experiments but with a slight decrease in power density uniformity) to a spot of approximately 3.5 mm in diameter (at 2-4 MW cm⁻³, for emission experiments in which it is not necessary to pump all of the vertical column). After traversing the shock tube, the laser was beam-dumped using a combination of angled, matt black neoprene sheets.

The flashlamp and detection optics were assembled to observe the interaction volume through the vertical, small, shock tube windows. Several combinations of lamps and spectrometers were used and details of these are given in the relevant experimental sections. In most cases, spectra were recorded using a Princeton Applied Research Corporation optical multichannel analyser (OMA) equipped with a 1025D detector head and a 102A console. Only the modifications to these along with minimal operating information will be described here.

The detector head consists of a silicon intensified target (SIT) RCA 4804 camera tube, with associated voltage supplies and an extremely low-noise high-gain signal preamplifier. Photons strike a photo-cathode and emitted electrons are accelerated to approximately 10 keV before being
imaged onto a 0.4" by 0.5" silicon chip, where they cause secondary electron cascades. The consequent charge distribution is read by an electron beam generated in the rear of the SIT tube which sweeps the back of the silicon chip. Channel size is determined by electron beam diameter and current integration times. In normal operation the chip is 'divided' into two 500-channel horizontal arrays, a signal array and a dark noise array respectively.

The instrument, including the analog to digital converter housed in the console, is specified as linear to 2% with respect to counts versus incoming intensity at the photocathode, for all 500 channels simultaneously. A dynamic range of $10^3$ per reading scan is possible within this linearity.

Gating is effected by reverse biasing by 1-2 kV the photocathode to chip potential thus holding off the primary electrons except for a time determined by a negative-going gating pulse. Because this pulse also affects the focussing of the primary electrons, it needs to be as flat topped and as square edged as possible. A further specification relating to coincidence triggering of other apparatus required that the delay from trigger input to leading edge output be less than 400 ns.

Experimentation led to the design shown in Fig. 3.7. Typical pulse output into 10 MΩ is shown in Fig. 3.8. Repeatability was found to be excellent; less than 3% in amplitude with jitter $\leq 20$ ns in the leading edge and $\leq 40$ ns in the trailing edge. Pulse length range may be altered by changing the values of $C_1$ and $C_2$. To prevent the RF noise accompanying firing of the laser from triggering the gating curcuit, all of the laser except for the flashlamp head was housed in an aluminium box. Experiments were performed with pulse lengths of 400 ns and 800 ns. In these times relevant conditions in the shock tube are essentially unchanging. In both the non-gated and 800 ns gated modes, OMA linearity was confirmed by a
number of tests including comparison of signals from a stable calibrated source attenuated by a variety of accurately calibrated neutral density filters. The 400 ns case is examined in section 4.5.

In the non-gated mode, comparison of the tabulated spectral output from a standard lamp with that recorded by the OMA indicated that relative OMA channel gain fell off by approximately 10% for the forty or so channels at each end of the silicon array. In all cases, digital ASCII output from the OMA was fed to a teletype paper tape punch and thence to a computer.

Coincidence timing of the OMA gate pulse with the 400 ns flat central part of the laser pulse demands that a zero time indicator be available from the laser itself, because jitter in the operation of the shock tube and laser spark gaps was observed to be several milliseconds and microseconds respectively. The rising of the ground plane of the laser by several hundred volts simultaneously with discharge of the flashlamp head provided a signal enabling coincidence jitter to be reduced to 40 ns. Both laser spectral jitter and bandwidth were sensitive functions of dye flow and laser operating voltage, increasing rapidly with increasing flow and higher voltages. This was due, in spite of water cooling of the laser head, to thermal gradients and fluctuations in the dye causing small angular deviations in optical path for the lasing modes, with respect to which the double prism arrangement is very sensitive. Optimised jitter and bandwidth were measured to be respectively $\pm 0.3 \, \text{Å}$ and an approximate triangle with full-width half-maximum of $1.0 \pm 0.1 \, \text{Å}$ with the laser tuned near the Na D-lines. These results are barely sufficient for the Rabi shifted peaks to be more than qualitatively identified (Kimble and Mandel, 1977).
4. EXPERIMENT

4.1 Transducer Design and Shock Timing

Along with measurement of the reflected shock pressure, determination of the incident shock speed is a major shock tube diagnostic. The short distance between timing stations (typically 20 cm) in combination with shock speeds of \( \approx 4.5 \text{ km s}^{-1} \) necessitated the use of timing transducers and amplifiers with jitter and response times \( \leq 0.2 \mu s \) if uncertainties in shock speeds and calculated temperatures were not to exceed 1.5% and 3% respectively. The latter was an important criterion for evaluation of the electron number density via Saha's equation (see section 4.3). For such an extremely fast risetime, given that typical incident shock pressure rises were \( \approx 600-700 \text{ psi} \) and may well be less at the transducer face (see Fig. 3.4), commercial units of sufficient sensitivity were not available.

Eventual transducer construction and output are shown in Fig. 4.1. Final design was a compromise between the requirements of front diaphragm flexibility (the pressure change should generate as large a force as possible onto the face of the crystal) and rigidity (good electrical and mechanical contact is essential over the entire front and rear crystal faces respectively). The front diaphragm must not bulge forwards at all when the shock tube is evacuated prior to a shot. Construction details are important, particularly the soldering and then paring back of a stainless steel cylinder to form the front diaphragm. The active transducer component was a 3 mm \( \times \) 1.5 mm PZT5A disc piezoelectric crystal which, when mounted and subjected to typical incident shock pressure rises, produced into 1 k\( \Omega \) a negative going pulse with leading edge slew rate \( \geq 150 \text{ mV/\mu s} \). This fast pulse was fed into the preamplifier of Fig. 4.2. The gain of the two very fast 733 op-amps was adjusted until the \( \approx 200 \text{ mV} \) above just-turn-on voltage required to ensure full-turn-on of the 319N voltage
Mylar insulation washer

Signal wire

Crystal

Rear contact piece

Transducer body

Insulation

0.010"

front diaphragm

Tension locking screw

Transducer design, excluding brass shock-tube-port mount

Fig. 4.1: Transducer design and specifications

Typical transducer output
Fig. 4.2: Transducer preamplifier schematic
Fig. 4.3: Main shock timing board schematic

All resistors in kΩ
comparator appeared in less than the desired jitter time of 0.2 μs. One-shot lockout and TTL pulse-shaping was performed by a modified version, shown schematically in Fig. 4.3, of a circuit originally developed by S. Ngan (1977). Overall electronic jitter between final main board outputs for identical square wave preamplifier inputs was ≤ 0.1 μs.

Jitter between mainboard outputs due to errors and drifts in setting preamplifier trigger levels, shot to shot variation in transducer output, and uncertainties in transducer separation accounted for a further 0.4 μs at typical shock speeds. For timing, an external 10 MHz source was used with Advance TC11 counters having resolution of 0.1 μs. Shock speed and calculated temperature uncertainties were estimated to be 1.2 - 1.5%, and 2.4 - 3.0% respectively. This system proved to be reliable, insensitive to both mechanical and electronic noise, capable of withstanding shock flash temperatures and easily maintainable, allowing rapid replacement of faulty crystals.

4.2 Viscous Effects and Shock Tube Performance

Viscosity alters the perfect-gas ideal shock tube behaviour, decreasing the incident shock speed and accelerating the contact surface, as mass is removed from the interlying region by means of the wall boundary layer. In addition, this boundary layer affects the propagation of the reflected shock, which encounters higher densities and lower velocities and temperatures near the wall. The result is a reduction of ~ 50% in test time from the ideal (due to early arrival of contact surface contamination), slight alteration of the reflected shock conditions from the calculated perfect gas quantities and different mechanisms for impurity mixing in the reflected shock region.
Helium number density uniformity and test times were measured interferometrically, using a 120 mW cw He-Cd laser, operating at 4678 Å, to illuminate the test section of the shock tube which was centred in one arm of a small Mach-Zehnder interferometer, producing horizontal fringes in undisturbed gas. The image of a narrow (100 μm) vertical slit was then swept by a Space Technology Laboratories ID/4B image converter camera, see Fig. 4.4. Results from typical shots are shown in Fig. 4.5. Planar, sharp incident and reflected shocks are clearly visible. He number density uniformity was found to be excellent (±5%) during test times of 30 μs to 55 μs, and wall boundary layers were too thin to be resolved (≤ 0.2 mm).

Simultaneous measurements of wall pressure and overall luminosity were made through the small top and bottom window apertures in the shock tube test section (see Fig. 3.4) using a Kistler type 2601 pressure transducer, and a fast PIN diode. These measurements indicated stable test times which were in agreement with the interferograms and which had reasonably constant pressure (approximately 15% fall overall) and luminosity (20% rise), with both increasing on the arrival of contact surface disturbances transmitted by a weak additional shock due to the further reflection of the reflected shock off the contact surface ( overtailing).

An x-t diagram, Fig. 4.6, illustrates the shock behaviour. Using the turbulent boundary layer theory of Mirels (1963, see below) and measured pressure rises to estimate the behaviour of the contact surface and re-reflected shock, calculated test times of ~40 μs were found to be in good agreement with the measured values.

However, measured fringe shifts were in poor agreement with shifts predicted from recorded shock speeds and perfect gas relations, even after allowing for possible further adiabatic expansion or compression to reach
Fig. 4.4: Interferometry schematic
Fig. 4.5: Typical image converter camera interferograms
Fig. 4.6: X-T diagram for typical 380 torr He shot

- Test point (centre of window)
- Re-reflected shock
- Sonic line
- Predicted test time
- Contact surface (from Mirels (1963))
- Reflected shock
- Incident shock

Distance from shock tube end wall (cm)

Time (μs)
the measured pressure (see below). Fringe shifts were measured by traversing a diode sensitive to reflected light intensity across a print, such as shown in Fig. 4.5, along a line perpendicular to the undisturbed fringes. A light source and appropriate focusing lenses and alignment pinholes were incorporated into the diode assembly. Digitized results for 256 sample points were then Fourier analyzed for undisturbed and disturbed fringe traverses. Helium densities up to 40% higher than predicted were observed. These were due to viscous effects, as described below.

An appropriate Reynolds number for the shock tube flow is (Mirels, 1963)

$$R_e = \frac{u_2 (u_1/u_2 - 1)^2 (u_1 t)}{\nu \omega}$$  \hspace{1cm} (4.1)

where $t$ is the elapsed time at the window between incident and reflected shocks, and $\nu \omega$ is the kinematic viscosity at the wall. All experimental conditions fall into the range $0.5 \leq R_e \times 10^6 \leq 4$ which Mirels quotes as the laminar to turbulent transitional regime. In addition, he gives estimates of turbulent boundary layer growth in terms of the displacement thickness, $\delta$:

$$\delta = \beta(u,t) \left[ \frac{\nu \omega}{u_1(1-u_2/u_1)} \right]^{1/5}$$  \hspace{1cm} (4.2)

where $\beta$ is a function of incident shock jump values. For the typical conditions described in 3.2 this yields $\delta \sim 0.2 \text{mm}$, corresponding to a sufficient increase in He number density at the wall to account for an overall increase in optical path length of about 30%.
A consistent explanation is that there exists a jet of cool, dense gas passing through the reflected shock near to the wall (Fig. 4.7) which accounts for the increased optical path length. Upon arrival and mixing at the end wall this results in an initial increase in pressure, due to interaction of the colliding wall jets, followed by a decrease as mixing and cooling proceed. The transitional Reynolds number implies haphazard jet repeatability, due to shot to shot changes in the point along the shock tube walls at which the laminar to turbulent transition occurs. Such a jet is also necessary to explain the observations, described below, that Na impurities enter the reflected shock region almost entirely from the back wall, that the Na Boltzmann temperature is uniform to ±7% throughout the tube and is the reflected shock temperature, that the number density of He responsible for the observed Na absorption profiles is in excellent agreement with perfect gas calculations, and that the measured stagnation pressures show the qualitative behaviour to be expected due to such a wall jet.

A possible mechanism for production of a wall jet is shock bifurcation (Fig. 4.8) in which part of the boundary layer cannot negotiate the pressure rise across the reflected shock and is carried along in a separation bubble, and part forms a relatively cool jet with associated vortex sheet at the bifurcation apex. Davies (1969) gives criteria for bifurcation to occur in terms of the maximum stagnation pressure $P_{st}$ in the boundary layer. The position of the corresponding steamline determines how much of the boundary layer is trapped in the separation bubble. Measurement of boundary layer pressure profiles is extremely difficult. The assumption of linear profiles gives $P_{st}/P_5 \sim 1.1$ for the ratio of maximum stagnation pressure to reflected shock pressure, whereas Davies' criterion is $P_{st}/P_5 \leq 0.9$. In view of the approximation made, the model of a wall jet is thus at least plausible.
Fig. 4.7: Behaviour of the wall jet

Fig. 4.8: Schematic of bifurcated foot
Overtailoring rise

Test time at rear wall

Fig. 4.9: PCB Output, 380 torr He

Fig. 4.14: Typical Optical Multichannel Analyser Output

Sodium D-lines (see text, p. 88 and Fig. 4.15)

\[ N_{\text{He}} = 1.7 \times 10^{20} \text{ cm}^{-3} \]
\[ T_{\text{He}} = 3.8 \times 10^3 \text{ oK} \]
In addition to the measured incident shock speed, a major shock tube diagnostic was the reflected shock pressure, which was measured with a subminiature transducer, PCB type 105A33, mounted in the rear shock tube wall. The latter incorporated its own charge to voltage amplifier and was supplied with a specific calibration curve, with a measured sensitivity 0.412 mV/psi.

Typical output from the PCB transducer is shown in Fig. 4.9. Note that the rise time is transducer limited to \( \approx 2 \mu s \). Ringing is due partly to an inherent resonance at 250 kHz, and partly to a double layer of PVC insulation tape (found to have no effect on averaged output) mounted to protect the transducer face from the high reflected shock temperatures and from small aluminium diaphragm fragments. The slight fall in pressure during the first 30 \( \mu s \) is consistent with cooling due to a dense wall jet, and the later rise with overtailing.

Initial shock reflected pressures, measured with an uncertainty of \( \pm 5\% \) using Tektronix 7000 series oscilloscopes, were typically up to 20\% higher than those calculated from the measured shock speeds. This jump was thought to be due to the effect of the axisymmetric wall jets colliding at the centrally positioned PCB transducer. Pressure changes measured at the back wall were assumed to effect conditions at the test point (Fig. 3.4) after a time determined by the sonic speed in the reflected shock region (some 4-5 \( \mu s \)) and to cause adiabatic compression (or, rarely, expansion) of the test gas, from the pressure calculated from perfect gas assumptions and measured shock speeds to the pressure estimated from transducer readings. This assumption was justified experimentally by agreement between predicted and obtained Na collision-broadened half-half widths (see section 4.4).
Na atom number density in the reflected shock region was controlled by periodically moistening the shock tube rear wall with saliva. This would provide some five shots with gradually decreasing optical depth. If not remoistened, after approximately eight shots the maximum optical depth would fall to $\tau \leq 0.1$. Applying Na to the side walls had very little effect in the succeeding shot. Observations made after the incident shock but before the return of the reflected shock showed optical depths smaller than those measured during the reflected shock test time by a factor of $\sim 6$. Only shots with $2.2 \geq \tau \geq 0.6$ in the line core were recorded, values outside this range leading to uncertainties in the accuracy of the computer data reduction of spectral line profiles.

To estimate the uniformity of Na atom distribution along the optical path, a measurement was made of the source function $S(\lambda, T)$ which appears as an emission correction in the expression for the observed transmitted intensity in an absorption experiment

$$I(\lambda) = I_o(\lambda) e^{-\tau(\lambda)} + S(\lambda, T) [1-e^{-\tau(\lambda)}].$$

Before the absorption run, an emission shot is used to establish that

$$S(\lambda, T) [1-e^{-\tau(\lambda)}]_{\text{max}} \ll [I_o(\lambda) e^{-\tau(\lambda)}]_{\text{min}}.$$

Assuming LTE, $S(\lambda, T)$ is the black body function, so that in the absorption shot a calculation of $S(\lambda, T)$ for a number of points within the spectral line profile yields corresponding temperatures $T$ for each point. Points of large optical depth reflect the behaviour of atoms near the outer surface of the shocked gas. Spectroscopic details are described in a later section.
Typical results indicated a source function consistent with a temperature uniform throughout the profile to $\pm 7\%$, which in turn implied that an insignificant number of atoms were located in the cool wall boundary layer and that the temperature distribution was fairly uniform throughout the non-boundary layer gas.

A further powerful diagnostic enabling a determination of the number density of He perturbers is the measurement of the collision broadened Na $5890 \, \text{Å}$ and $5896 \, \text{Å}$ D-line half-half widths (Eckart, 1975). Experimental details and data are contained in section 4.4. Assuming an adiabatic compression, from the reflected shock pressure calculated from measured shock speeds to the stagnation pressure measured at the rear wall, excellent agreement was obtained between the measured $5890 \, \text{Å}$ widths (based on some 1500 data points) and the mean of the data of Eckart. Because the width depends on different powers of $n_{\text{He}}$ and $T$ (an absolute temperature characteristic of collision kinetic energies) this agreement is a powerful verification of the viscous perfect gas operation of the shock tube outlined above.

4.3 Impurity Concentrations

Because of the low ionization of He at $5,000^\circ\text{K}$ and the small thermal transfer per He-Na collision (Eckart, 1974) the presence of electrons from donating impurities dominates the approach to LTE of Na swept from the shock tube walls. The electron number density, $n_e$, was measured directly using Thompson scattering and indirectly, applying Saha's equation to relative intensities of CaI and CaII resonance lines.

For the former method, a Union Carbide "Korad" K-1 ruby laser was used, in the Pockels-cell Q-spoiled mode, producing a triangular pulse, with a FWHM of 40 ns, of $\sim 60 \, \text{MW cm}^{-2}$ peak intensity over a 1 cm diameter
Scattering at right angles was observed using an RCA Quantacon C31034 photomultiplier, mounted on a SPEX one-meter Czerny-Turner monochromator equipped with an 1800 line/mm holographic grating peaked at \( \approx 500 \text{ nm} \) and giving \( 5\frac{5}{3} \text{ Å/mm} \) in the exit plane. With a 100 \( \mu \) input slit, the ratio of electron scattered signal to spurious scattering from shock tube windows was maximized by selecting an exit window to collect only the outer half of one wing of the \( \sim 9 \text{ Å}_{1/2} \) width electron Doppler-profile, giving a \( 10^3 \) improvement factor compared to collection of the entire profile. System calibration was provided by Rayleigh scattering using both \( \text{N}_2 \) and \( \text{He} \) at 1 atm. Shot to shot variations in laser intensity were monitored using a fast Hewlett-Packard PIN photodiode.

In spite of optimised anti-reflection measures, the use of BK7-glass windows, beam focussing and OBl0-glass beam dumping, the small size of \( 7.9 \times 10^{-26} \text{ cm}^{-2} \text{ sr}^{-1} \) for the differential Thompson cross-section at \( \pi/2 \) and the inescapable proximity of the shock tube windows to the scattering volume combined to yield a spurious-scattering limited best value of \( n_e = (1.0 \pm 1.3) \times 10^{16} \text{ cm}^{-3} \), for reflected shock conditions of \( n_{\text{He}} \sim 5 \times 10^{19} \text{ cm}^{-3} \), \( T_{\text{He}} \sim 5,500^{0}K \). At these conditions, the ratio of collisional de-excitation to spontaneous decay for the Na \( 3^2P_{3/2} \) level is \( R \sim 25 \times 10^{-16} n_e \) (Van Regemorter, 1962) so that the ratio of actual population to LTE population is \( R' = (1+R^{-1})^{-1} \sim 0.96 \), and the Na Boltzmann temperature differs from \( T_{\text{He}} \) by less than 1%.

The major expected impurities were Fe, Cr, Ca, C, Al, K and Na. The conveniently close CaI 4227 Å and CaII 3968 Å, 3934 Å resonance lines were observed simultaneously in emission, through the small shock tube windows, using a Russian constant deviation prism spectrometer having a dispersion of \( \sim 47 \text{ Å/mm} \) in the exit plane. Spectra were recorded digitally, using the
OMA which was gated on for ~ 9 μs during the first half of the steady reflected shock test time. The central part of a tungsten-filament standard lamp, positioned at point P of Fig. 3.3, provided a relative intensity calibration for the system of collection optics, spectrometer and OMA over the $6 \times 10^2 \text{Å}$ recorded exit window.

The shock tube test section was imaged onto an 80 μ input slit, and the corresponding instrument function of ~ $1.3 \text{Å}$ FWHM and typical sections of the 500-channel spectrum are shown in Fig. 4.10. Shot to shot reproducibility of relative (absolute) line positions was better than one (two) channels, corresponding to approximately one (two) angstroms.

Variations in absolute intensity were generally ≤ 25%. Absolute wavelength versus channel assignments were obtained using Ca and Cr hollow cathode lamps for the unionized resonance lines and narrow band filters for the CaII resonance lines. Other lines were inferred from spectroscopic tables (Meggers et al., 1961 and Wiese et al., 1969).

To determine collision broadened line shifts (if any) to assist in wavelength and atomic line assignation, and also to obtain estimates of the optical depth in the core of each line, absorption experiments (non-simultaneous) were carried out for the lines in the spectrum of Fig. 4.10. With a 20 μ input slit and the OMA at the exit plane of the SPEX 1-metre spectrometer, set up as for the Thompson scattering experiment, the system instrument function was a three channel triangle, the two side channel magnitudes being approximately $\frac{1}{3}$ of the central channel. This was the standard high resolution arrangement, giving $0.136 \text{Å}$/channel at 5890 Å. A typical absorption spectrum is shown in Fig. 4.11. In this, and all other, absorption runs background illumination was provided by an ILC type 4LC quartz-Xenon linear-arc flashlamp used to dump 40 μF charged to ~ 2.1 kV. Output, measured with a broadband, fast Hewlett-Packard PIN diode and shown
Fig. 4.10

Emission spectrum

Instrument function

Channel number
Fig. 4.11: Absorption spectrum (high resolution)

Overlay of three shots

Fig. 4.12: Flashlamp characteristics

High resolution spectrum near NaD lines
in Fig. 4.12(a), was close to being critically damped and was found to be very reproducible (peak height better than 5%). At these conditions the lamp was spectrally quite flat; Fig. 4.12(b) includes the effect of non-uniform OMA sensitivity, which is responsible for most of the curvature. The lamp was imaged through the small shock tube windows onto the test section and again onto the entrance slit of the SPEX so as to closely match the fl0 optics of the spectrometer. Immediately prior to each absorption run a 500 channel spectrum of the flashlamp was recorded, and the position of at least one line determined using hollow cathode or arc-discharge lamps. For the high resolution arrangement, shot to shot reproducibility in relative and absolute line positions was ±1 and ±3 channels respectively, and was better than 25% for absolute intensities, this last being limited by repeatability in shock tube recoil and associated vignetting and movement of the image on the sensitive part of the OMA.

In contrast to the situation for Na atoms, both the Cr and Ca impurity concentrations, as evidenced by optical depth measurements, showed excellent shot to shot repeatability (better than 10%) indicating as probable source the mild steel shock tube rear wall.

With the temperature $T$ in °K, the ionization potential $\chi$ and the lowering of the potential $\Delta \chi$ in cm$^{-1}$ and the species number densities $n$ in cm$^{-3}$ Saha's equation becomes (see for example Drawin and Felenbok, 1965)

$$\frac{n_{\text{Ca II}}}{n_{\text{Ca I}}} = 4.825 \times 10^{15} T^{3/2} \exp \left(-1.4388 \frac{\chi - \Delta \chi}{T} \right) \frac{U_{\text{Ca II}}(T)}{U_{\text{Ca I}}(T)}$$

(4.3)

where $U_{\text{Ca}}$ are the partition functions as functions of temperature. For Ca transitions from atomic states characterised by upper level energies $E$, degeneracies $g$, line strengths $S$ and emission intensities $I$, the relation
for optically thin lines allows \( n_e \) to be obtained without the need to calculate \( U_{Ca} \). Optical depth corrections to \( I_I \) and \( I_{II} \) were performed using a numerically computed curve of growth, assuming a purely Lorentzian pressure broadened profile for both lines (Doppler broadening is insignificant). Values of \( \chi, E, g \) and \( S \) were obtained from Wiese et al. (1969).

Using Debye's theory for the lowering of the ionization potential (since the number of electrons in a Debye sphere is \( >>1 \)), \( \Delta\chi \) was found to be negligible (Drawin and Felenbok, 1965). A well-defined temperature was assumed to exist (valid given determined values for \( n_e \)), equal to the kinetic helium temperature obtained from measured shock speeds and pressures.

Shots with \( P_1 = 380 \) torr, giving \( T_5 \approx 5 \times 10^3 \) °K and \( n_{He} \approx 1 \times 10^{20} \) cm\(^{-3}\), and shots with \( P_1 = 190 \) torr, giving \( T_5 \approx 6 \times 10^3 \) °K and \( n_{He} \approx 6 \times 10^{19} \) cm\(^{-3}\) were analysed for the 4227 Å/3934 Å ratio yielding \( n_e = (3.6 \pm 0.9) \times 10^{15} \) cm\(^{-3}\) and \( n_e = (1.7 \pm 0.8) \times 10^{15} \) cm\(^{-3}\) for the former, and \( n_e = (2.5 \pm 2.0) \times 10^{16} \) cm\(^{-3}\) and \( n_e = (1.8 \pm 0.8) \times 10^{16} \) cm\(^{-3}\) for the latter, in fair agreement with the Thompson scattering result. The large uncertainties are due to poor signal to noise ratio for the comparatively less intense 3934 °A line, and uncertainties in reflected shock temperatures, with respect to which this procedure is extremely sensitive. Differences between results for the same shock filling pressures are indicative of typical shot to shot repeatability in \( T_5 \), in the concentration of electron donating contaminants and of the effects of adiabatic compression due to the wall jet. No corrections have been made for ionization and radiation energy losses, respectively estimated to be of the order of 0.03% and 0.2% of the kinetic He energy. A more precise
estimate of the latter is impossible due to the difficulty of modelling all important radiation mechanisms. In any event, such uncertainties may be ignored in view of the shot to shot variations. The significance of these results for $n_e$ is that electron broadening is of the same order as Doppler broadening at these conditions (Benett and Griem, 1971) and may likewise be ignored and the Na-D lines assumed purely collision broadened.

Estimates of atomic number number densities $n_i$ may be obtained from the expression for the optical depth $\tau_i$ of an homogeneous layer of gas of depth $x_o$ for a transition between lower state $i$ and upper state $k$

$$\tau_i(\omega) = 2\pi^2 \Gamma_0 c n_i x_o f_{i,k} \left( 1 - \frac{g_i n_k P^\text{SE}(\omega)}{g_k n_i P^\text{A}(\omega)} \right) P^\text{A}(\omega)$$

(4.5)

where $\Gamma_0$ is the classical electron radius, $c = \text{speed of light}$, $f_{i,k}$ is the (absorption) transition f-value, $g$ denotes degeneracy and $P^\text{A}$, $P^\text{SE}$ are absorption and stimulated emission profiles normalised to unity.

$$\int_{\text{line}} P(\omega) \, d\omega = 1.$$  Under present circumstances (in the absence of the laser), stimulated emission is negligible. Hence, for an isolated Lorentzian line of $\frac{1}{2}$-width $W_i$, we find

$$n_i = \frac{W_i \tau_0}{2\pi r_0 c x_o f_{i,k}}$$

where $\tau_0$ is the optical depth in the centre of the line profile.

Ignoring the contribution of the 5896 Å line, which is a small correction to the 5890 Å Lorentzian profile, substitution of typical measured values for $W_i$ and $\tau_0$ yields Na ground state number densities of $\sim 3 \times 10^{13}$ cm$^{-3}$.
In order to estimate impurity concentrations, absorption profiles were obtained for the CaI 4227 Å and CrI 4254 Å, 4275 Å lines using the standard high resolution arrangement described earlier. Data reduction procedures are dealt with in section 4.4. Because of high optical depths (approximately 3.2 for the CaI line) and observed profile widths that approached the instrument width (thus making deconvolution inaccurate) only approximate reduced \( \frac{1}{2} - \frac{1}{2} \) widths were obtained for \( n_{\text{He}} \sim 1.2 \times 10^{20} \text{ cm}^{-3} \) and \( T \sim 4.7 \times 10^3 \text{ K} \):

<table>
<thead>
<tr>
<th>Shot no.</th>
<th>Observed ( \frac{1}{2} - \frac{1}{2} ) width (channels)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>4227</td>
</tr>
<tr>
<td>207</td>
<td>4.6</td>
</tr>
<tr>
<td>208</td>
<td>4.9</td>
</tr>
</tbody>
</table>

\[
\begin{align*}
\text{HHW}_{4227} &= (0.5 \pm 0.3) \times 10^{-20} \text{ Å cm}^3 \\
\text{HHW}_{4254} &= (0.3 \pm 0.2) \times 10^{-20} \text{ Å cm}^3 \\
\text{HHW}_{4275} &= (0.2 \pm 0.1) \times 10^{-20} \text{ Å cm}^3
\end{align*}
\]

These CaI results are to be compared with those reported by Malvern (1977) and Driver and Snider (1976). The latter measured \( \text{HHW}_{4227} = (0.25 \pm 0.03) \times 10^{-20} \text{ Å cm}^3 \) at \( 4.7 \times 10^3 \text{ K} \) in a piston compressor with \( n_{\text{He}} = (2-4) \times 10^{20} \text{ cm}^{-3} \), which is in fair agreement with the figure \( 0.3 \times 10^{-20} \text{ Å cm}^3 \) obtained by extrapolating Malvern's theoretical data. The present higher result is most likely due to poor peak height data resulting from high optical depths. Values of absorption oscillator strengths for these three ground state transitions are \( f_{4227} = 1.75 \pm 0.08 \) (Wiese et al., 1969) and \( f_{4254} = 0.259 \pm 0.12, f_{4275} = 0.196 \pm 0.010 \) (Huber and Sandeman, 1977) and typical
number densities were \( n_{\text{CaI (gnd)}} \sim 2 \times 10^{13} \text{ cm}^{-3} \) and \( n_{\text{Cr (gnd)}} \sim 4 \times 10^{13} \text{ cm}^{-3} \).

The absence in emission or absorption of FeI lines leads to the conclusion that FeI(gnd) number densities are lower than those for CaI(gnd). Because of the high ionization energy, negligible FeII are expected to be present.

The question of a source for the electrons now arises, since observed atomic impurity densities are far too low to be the major contributors. He may be rejected, as less than \( 10^{-5} \) of those present are ionized. The most likely sources are \( \text{Al}_2\text{O}_3 \) and molecular compounds formed by combustion of driver gas residues (coming from delrin piston rings and neoprene piston buffers).

4.4 Sodium Absorption Line Profiles

The absorption profiles of Na D-lines broadened by collisions with He have been measured by Eckart (1975) and Baird et al. (1979) who obtained significantly different results for the reduced \( \frac{1}{2} - \frac{3}{2} \) width of the 5890 Å line in the region of temperatures common to both experimental techniques. In respective experiments, hot dense He was produced at \( 3-6 \times 10^3 \) °K and \( 2-5 \times 10^{20} \text{ cm}^{-3} \) in a piston compressor, and \( 5-10 \times 10^3 \) °K and \( 4-12 \times 10^{19} \text{ cm}^{-3} \) in a free-piston shock tube. Results for these two experiments (as well as for the present work) are plotted in Fig. 4.13. Note that not only do the magnitudes of the reduced \( \frac{1}{2} - \frac{3}{2} \) widths differ (at 5,000 °K Eckart gives \( (0.98 \pm 0.08) \times 10^{-20} \) Å cm\(^3\) while Baird et al. give \( (0.81 \pm 0.04) \times 10^{-20} \) Å cm\(^3\)) but the exponent \( \delta \) in the temperature dependence \( T^\delta \) of the reduced \( \frac{1}{2} - \frac{3}{2} \) width is given respectively as 0.41 and 1.1.

Several possible explanations were put forward by Baird et al. for the discrepancies with the work of Eckart. The conclusion arrived at was that the most likely of these was that mixing of the upper states \( 3^2 P_{3/2} \)
and \(^3\!^2\!P_{3/2}\) by the perturber ensemble produced a non-Lorentzian anti-symmetric contribution to each lineshape and this affected mostly the results of Eckart because of his higher densities. The origin of such non-Lorentzian behaviour for overlapping lines was pointed out by Baranger (1958(b)) and in the present case for S-state He scatterers was discussed in section 3.1. Experimental limitations of the optical sampling apparatus prevented Baird et al. from measuring the non-Lorentzian component and thus testing their hypothesis.

Using the standard high resolution set up described in section 3.3 absorption profiles for both D-lines were measured simultaneously, for two shock tube conditions - \(n_{\text{He}} \sim 1.7 \times 10^{20} \text{ cm}^3\) and \(T \sim 4 \times 10^3 \text{ °K}\); \(n_{\text{He}} \sim 0.7 \times 10^{20} \text{ cm}^3\) and \(T \sim 6 \times 10^3 \text{ °K}\). A total of 4 shots representing some 1500 usable data points were recorded. At these conditions emission corrections were found to be insignificant (<1%). Typical 500 channel output from the OMA for a data acquisition gating time of \(\sim 800\) ns is shown in Fig. 4.14 for \(n_{\text{He}} = (1.7 \pm .1) \times 10^{20} \text{ cm}^{-2}\), \(T_{\text{He}} = (3.8 \pm .2) \times 10^3 \text{ °K}\). The corresponding optical depth is shown in Fig. 4.15 and has a maximum value in the core of the 5890 Å line of \(\sim 1.4\). To determine the general form of the D-line absorption profile we write the susceptibility as:
where $|L\rangle$, $|1\rangle$ and $|2\rangle$ are the lower state and the two upper states respectively and $\Gamma$ is the corresponding four-point function (note the similarity to the laser field case of section 2). It is the last two diagrams which represent those interactions between the upper two states which could give rise to antisymmetric terms in the absorption profile. When the isolated line approximation is valid, the last two diagrams can be ignored, the vertex equation matrix is diagonal in the indices 1,2 and may be trivially inverted using the formalism developed in section 2, and we then obtain a susceptibility of the form (which gives rise to two Lorentzians)

$$S \propto 2(\Delta_1 + i\gamma_1)^{-1} + (\Delta_2 + i\gamma_2)^{-1}$$

where $\gamma_1, \gamma_2$ and $\Delta_1, \Delta_2$ are $\frac{1}{2}$-$\frac{1}{2}$ widths and detunings of the probe frequency from the resonance wavelengths for the $|1\rangle \leftrightarrow |L\rangle$ and $|2\rangle \leftrightarrow |L\rangle$ transitions respectively. When the last two diagrams are included, the vertex equation involves the inversion of a $4 \times 4$ matrix all of whose terms are non zero. Considering only these two last terms, and following the formalism of section 2 in which the imaginary time susceptibility is evaluated at the pole of the lower state propagator, the additional contribution to the susceptibility is of the form

$$C' (\bar{\Delta}_1 + i\bar{\gamma}_1)^{-1} (\bar{\Delta}_2 + i\bar{\gamma}_2)^{-1}$$

where $C'$ is some complex quantity independent of $\omega_q$ and the $\bar{\Delta}$'s and $\bar{\gamma}$'s may be slightly different from the $\Delta$'s and $\gamma$'s (M. Eckart (private communication) has actually evaluated $C'$ in terms of the four point function and reduced dipole matrix elements for the case of the Na D-lines). These differences are, however, very small as the propagators are almost identical to those appearing in the isolated line case. Hence, separating the above into partial fractions, we obtain
\[ P \left\{ \left( \Delta_1 + iY_1 \right)^{-1} - \left( \Delta_2 + iY_2 \right)^{-1} \right\} \]

where

\[ C' = iP \left( \gamma_2 - \gamma_1 \right), \]

and thus the general general parameter form that was adopted for the susceptibility was:

\[ S \propto \frac{2+P}{\Delta_1 + iY_1} + \frac{1-P}{\Delta_2 + iY_2} \]

The absorption profile is proportional to the imaginary part of \( S \), i.e.

\[ (1+\delta_1)^{-1} + \left\{ \frac{1-\text{Re}P}{2+\text{Re}P} \right\} \frac{\gamma_1}{Y_2} \left( 1+\delta_2 \right)^{-1} \]

\[ - \frac{\text{Im}P}{(2+\text{Re}P)} \left\{ \frac{\delta_1}{(1+\delta_1^2)} - \frac{\gamma_1}{Y_2} \frac{\delta_2}{(1+\delta_2^2)} \right\} \]

A five parameter double Lorentzian of the form

\[ \tau(\lambda) = A \left\{ (1+\delta_1^2)^{-1} + \frac{\gamma_1}{2Y_2} (1+\delta_2^2)^{-1} \right\} \quad (4.6) \]

was used in the data reduction of the experimental optical depth \( \tau(\lambda) \) when non-Lorentzian contributions were ignored, and a seven parameter form

\[ \tau(\lambda) = A_1 \left\{ (1+\delta_1^2)^{-1} + \frac{A_2\gamma_1}{Y_2} (1+\delta_2^2)^{-1} + C \left[ \frac{\delta_1 (1+\delta_1^2)^{-1} - \frac{\gamma_1}{Y_2} \delta_2 (1+\delta_2^2)^{-1}}{1+\Delta_1^2} \right] \right\} \quad (4.7) \]

was used when the non-Lorentzian anti-symmetric terms were to be explicitly included. Wavelength dependence is contained in

\[ \delta_1 = (\lambda - \lambda_1)/\gamma_1 \]

and

\[ \delta_2 = (\lambda - \lambda_2)/\gamma_2, \]
where $\lambda_1$, $\gamma_1$ and $\lambda_2$, $\gamma_2$ are the peak positions and HWHM (in wavelength units) of the 5890Å and 5896Å lines respectively. Estimates of the best fit parameters were obtained by means of a computer minimisation (Fletcher and Powell, 1963; Temes and Calahan, 1967) of the quantity

$$F = \sum_{\text{channels}} (\tau(\lambda) - \overline{\tau}(\lambda))^2.$$ 

This least squares procedure weights each data point in proportion to the square of its optical depth. Although clearly false for large optical depths, for profiles with $\tau_{\text{max}} \leq 2.2$ and taking into account statistical counting uncertainties in both flashlamp background and absorption profiles the weighting procedure is statistically correct to within a factor of $\sim 2$. More complicated weights made the multi-dimensional surface for $F$ excessively intricate (see below). The computer method, starting from a user-input mesh, searches for the most negative gradient and follows this until it finds a local minimum in the multi-dimensional surface defined by $F$. The difficulty lies in determining when this is the global minimum. Two smoothing techniques were used in attempts to simplify the structure of this surface. Noise filtering using Fourier transform computer techniques was judged to be unsatisfactory due to significant noise contributions at frequencies close to those corresponding to the tops of the spectral profile peaks. Removal of this noise from the Fourier spectrum even by smooth sinusoidal truncations was considered to produce unacceptable decreases in spectral profile peak height. Smoothing using a moving least-squares technique (Savitzky and Golay, 1964) also tended to round off peaks excessively, but was otherwise satisfactory in determining a good initial guess for the line parameters before executing the computer reduction.
utilising unsmoothed data. Incorporating more complicated data point weighting schemes than that outlined above greatly increased program execution times and in any case in the analysis of several typical profiles produced solutions negligibly different from those obtained with the simpler procedures. To test whether the minima found in F were global minima, mesh searches of some representative spectra were made by evaluating F at every point on a finely spaced grid centred on the computer determined set of parameters. In every case examined no superior minimum was found, and, while not absolutely conclusive, this lends confidence in the final parameters. Results for the collision broadened $\frac{1}{2}-\frac{1}{2}$ width are presented in Table 4.1 and reduced $\frac{1}{2}-\frac{1}{2}$ widths for the 5890 Å line are plotted in Fig. 4.13. Experimental uncertainties for individual shots were ±6% for the temperature and ±7%, ±9% for the reduced $\frac{1}{2}-\frac{1}{2}$ widths of the 5890 Å line

Table 4.1: Measured $\frac{1}{2}-\frac{1}{2}$ widths

$[1$ channel $= 0.136 \AA , \text{Eq.}(4.6)]$

<table>
<thead>
<tr>
<th>Shot no.</th>
<th>$\rho_5$ ($10^{20} \text{cm}^{-3}$)</th>
<th>$T_5$ ($10^{3} \text{K}$)</th>
<th>5890 Å HHW (OMA channels)</th>
<th>5896 Å HHW (OMA channels)</th>
</tr>
</thead>
<tbody>
<tr>
<td>212</td>
<td>1.66</td>
<td>3.80</td>
<td>9.50</td>
<td>7.95</td>
</tr>
<tr>
<td>214</td>
<td>1.69</td>
<td>3.81</td>
<td>10.06</td>
<td>8.95</td>
</tr>
<tr>
<td>215</td>
<td>1.73</td>
<td>3.84</td>
<td>10.61</td>
<td>10.00</td>
</tr>
<tr>
<td>216</td>
<td>0.667</td>
<td>5.81</td>
<td>5.37</td>
<td>4.80</td>
</tr>
</tbody>
</table>
Fig. 4.13: Reduced half-half width of the Na D2 line broadened by He as a function of temperature. X this experiment; O Eckart (1975); • Baird et al. (1979)
Fig. 4.15: Raw and computer-fitted optical depths corresponding to Fig. 4.14 of p.75 (SH215)
Fig. 4.16: Values for ReP, ImP
and 5896 Å line respectively, due to uncertainties in shock speeds and reflected shock pressures as well as estimated uncertainties of ±2%, ±4% for the accuracy of the computer fitting program. For shot 212, noise on the pressure trace requires that these uncertainties be increased to ±8%, ±8% and ±10% respectively. The measured reduced $\lambda_2-\lambda_1$ width for the 5890 Å line at $(3.8 \pm 0.2) \times 10^3$ K is thus $(0.81 \pm 0.04) \times 10^{-20}$ Å cm$^3$ while the measured ratio for $\gamma_{5890}/\gamma_{5896}$ at this temperature is $1.13 \pm 0.04$. This value is in agreement with the measured ratio of $1.12 \pm 0.06$ at $(5.8 \pm 0.2) \times 10^3$ K and confirms the expectation that the broadening ratio should be independent of temperature.

For the above four shots, and also for five other shots where the gating time of ~9 µs covered reflected shock pressure changes too severe to include them in Table 4.1, computer estimated values for $ReP = (1 - 2A_2\gamma_2/\gamma_1) / (1 + A_2\gamma_2/\gamma_1)$ and $ImP = -C(2+ReP)$ were obtained. These are presented in Table 4.2 and Fig. 4.16.

### Table 4.2: Parameter values, Eq. (4.7)

<table>
<thead>
<tr>
<th>Shot no.</th>
<th>$p_5$ (approx)</th>
<th>ReP</th>
<th>ImP</th>
<th>C</th>
<th>Separation $\lambda_2-\lambda_1$ (channels, Eq.(4.6))</th>
</tr>
</thead>
<tbody>
<tr>
<td>195</td>
<td>1.1</td>
<td>-0.069</td>
<td>0.130</td>
<td>-0.068</td>
<td>44.6</td>
</tr>
<tr>
<td>205</td>
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<td>0.119</td>
<td>-0.063</td>
<td>44.0</td>
</tr>
<tr>
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<td>-0.102</td>
<td>0.071</td>
<td>-0.038</td>
<td>44.2</td>
</tr>
<tr>
<td>210</td>
<td>1.7</td>
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<td>0.037</td>
<td>-0.019</td>
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</tr>
<tr>
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<td>0.015</td>
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<td>0.7</td>
<td>-0.031</td>
<td>0.021</td>
<td>-0.011</td>
<td>44.6</td>
</tr>
</tbody>
</table>
In all cases, $|C| < 0.07$ and integrated over the entire profile the non-Lorentzian anti-symmetric term (modulus) contributed less than 1 part in $10^3$ to the overall profile area. Because of the difficulty in determining uncertainties in computer estimated parameters, the best estimate of uncertainties in ReP, ImP is the scatter for shots at the same conditions of $n_{\text{He}}$ (and T), in which case no consistent values can be given to ReP, ImP. Another line of reasoning leads to the same result. Suppose the general trend ReP $\approx$ -ImP, that is apparent at first glance at Fig. 4.16, is not merely some instrumental error but reflects some behavior of the overlapping Na D-lines. Then $C \approx \frac{\text{ReP}}{(2+\text{ReP})} \approx \frac{1}{2} \text{ReP}$. Because the degree of overlapping (and C) increases as $n_{\text{He}}$ increases, one would expect that shots having higher $n_{\text{He}}$ would also have larger ReP. However, shot 216 sits in the centre of the data distribution. It is much more likely that the distribution represents some instrumental error, firstly because the size of the terms is consistent with (integrated over the profile) statistical errors and secondly because sets 195, 205, 206 and 210, 211, 212, 214, 215, 216 were obtained on different days and the OMA was susceptible to a very small electronic drift in dispersion, focus and subtraction-equalisation controls on a day to day level.

On this basis, Eq. (4.6) rather than Eq. (4.7) was used to determine the quantities in Table 4.1 and the last column of Table 4.2. Absolute shift measurements were not practicable due to shot to shot irreproducibility of absolute wavelength on the OMA. Uncertainty in each of the values for $\lambda_2 - \lambda_1$ in Table 4.2 is $\pm 1.5$ channels. Given that the ungated D-line separation was $44.0 \pm 0.5$ channels and that no change in $\lambda_2 - \lambda_1$ was observed with changing $n_{\text{He}}$ or T, the last column of Table 4.2 is interpreted as demonstrating a small ($\sim 1\%$) change in dispersion between the non-gated and gated modes.
Comparison of the above data with that of Eckart and Baird et al. (Fig. 4.13) clearly discriminates in favour of Eckart. The value of 

\((0.81 \pm 0.04) \times 10^{-20} \text{Å cm}^3\) for the 5890 Å reduced ½−½ width at \(3.8 \times 10^3 \text{°K}\) is in exact agreement with the theoretical value of Lewis and McNamara (1977). The value \(\gamma_1/\gamma_2 = 1.13 \pm 0.04\) is in fair agreement with Eckart's result of \(\gamma_1/\gamma_2 = 1.0 \pm 0.2\) but is a far more accurate measurement (Eckart was limited to some 12 data points per shot).

The demonstration that non-Lorentzian terms are insignificant in treating the overlapping Na D-lines invalidates the explanation put forward by Baird et al. for the discrepancy between their results and those of Eckart (and this work). The most probable explanation is that the complex shock system formed at the entrance to their shock tube cookie-cutter test section was responsible for a (possibly non-uniform) slug of test gas having pressures, densities and temperatures slightly different to their calculated values. This would seem to justify the construction and testing of the shock tube used in the present work and described in sections 3.2 and 4.2.

These experimental results are being submitted as a paper for publication in the Journal of Physics B.

4.5 Experimental Limitations

With the shock tube operating at the well-diagnosed conditions described previously, the laser was tuned to the 5890 Å line and operated at 1 MW cm\(^{-2}\) with a slightly converging beam of FWHM \(\sim 1\) Å and jitter of \(\sim 0.3\) Å. A 400 ns gating time was chosen as the longest allowable if additional uncertainties due to a non-constant laser intensity were not to render the designed experiment totally non-quantitative. Relative discharge to discharge laser power was monitored using a fast Hewlett-Packard PIN diode observing scattered light from the rear laser
cavity mirror. To minimise spurious scattering of the laser, in addition to the beam dumping described earlier a series of stops was placed to prevent direct observation of any shock tube walls or unnecessary window faces. Despite these and other precautions, spurious scattering with no gas in the shock tube test section gave a signal at least \( \frac{1}{4} \) as large as that obtainable from the flashlamp or from the resonance fluorescence observed during a shot. The use of BK7 windows with their lower number of F-centre scatterers compared to quartz was not possible for more than one or two shots due to the pitting and fracturing of the BK7 surface after the passage of even one shock, particularly near the corner edges of the square shock tube. In addition, this spurious scattering was increased (though with the same approximate line shape) by an order of magnitude during a shot, whether or not the laser was tuned to one of the Na D-lines. Discharge to discharge variations in the laser line shape though small made subtraction of the scattered signal from the total observed signal not feasible. The results from a typical shot are shown in Fig. 4.17. Na number densities of \( \approx 3 \times 10^{13} \) cm\(^{-3} \) were estimated by running every second shot in absorption mode without the laser. Resonance fluorescence (at right angles) of the 5896 Å line at a level of the order of a hundred times that observed from emission in the absence of the laser indicated both saturation of the 5890 Å transition and lack of a high degree of ionization or coherent forward scattering. This lack is consistent with the expectations of section 3.1. The most likely cause of the enhanced spurious scattering is dust or other macroscopic particles swept off the shock tube side and rear walls and present in suspension and/or redeposited onto clean window surfaces after passage of the reflected shock but before the laser is fired. Efforts to reduce this, including machine polishing and washing in methanol of the entire shock tube including the rear wall
plug, were unsuccessful. The conclusion is that the designed experiment is not possible on the described shock tube.

At 400 ns gating duration, the OMA did not operate satisfactorily. In comparison with successful behaviour at 800 ns gating duration, the measured 5890 Å $\frac{1}{2}$-$\frac{1}{2}$ width was no longer in agreement with the reflected shock number density and temperature, and the measured 5890 Å to 5896 Å $\frac{1}{2}$-$\frac{1}{2}$ width ratio was as high as $\frac{3}{2}$. Additionally, dispersion at the edges was very different to that in the centre of the screen. The most probable cause is some sort of defocussing of the primary photoelectrons in their acceleration onto the silicon target chip, although no specific mechanism could be determined. Attempts to calibrate out this behaviour by deconvolving the observed profile with various plausible (possibly channel dependent) instrument functions were unsuccessful.

FOR CONCLUDING REMARKS,

ALL ADDENDIUM
Fig. 4.17: Laser scattering superimposed on flashlamp


Temes, G.C. and D.A. Calahan, P.Soc. IEEE, 55, 1832 (1967).
Wilson, A. and Y. Shimoni