REDISTRIBUTION OF RADIATION IN THE ABSENCE OF COLLISIONS

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ABSTRACT

Redistribution and depolarization of near-resonant radiation was studied for the He 2 1P–3 1D line (668 nm) (and some data are also presented for Hz [656 nm]). These transitions have lower levels with natural line widths that are large compared with their upper-level natural widths, and, under experimental conditions, their collisional widths were also small. Our measurements of the ratio of Rayleigh to fluorescent intensities confirm the prediction that redistribution of radiation occurs in the absence of collisions for transitions having significant lower-level radiative widths. Depolarization rates by collisions with helium were also inferred from the measurements.

Subject headings: line formation — polarization — radiative transfer

I. INTRODUCTION

In stellar atmospheres one frequently has the situation in which scattering of radiation is important, so that the often-used approximation of complete redistribution of near-resonant radiation is invalid. Then one has to consider partial redistribution (Mihalas 1978). In calculations of partial redistribution it is usual to use a frequency redistribution function \( p(\omega_1, \omega_2) \) equivalent to that used by Zanstra (1941, 1946; see also Omont, Smith, and Cooper 1972, hereafter OSC; Mihalas 1978), namely,

\[
p(\omega_1, \omega_2) = \frac{\Gamma_u}{\Gamma_u + \Gamma_z} \delta(\omega_1 - \omega_2) + \frac{\Gamma_c}{\Gamma_u + \Gamma_c} f(\omega_2 - \omega_0),
\]

where \( \omega_1 \) and \( \omega_2 \) are the incoming and outgoing frequencies, \( \Gamma_u \) is the radiative width of the upper level, and \( \Gamma_c \) is the collisional width. The delta function corresponds to Rayleigh scattering. The function \( f(\omega_2 - \omega_0) = (\Gamma_c/2\pi)(\omega_2 - \omega_0)^2 + (\Gamma_c/2)^2 \) is a Lorentzian about the transition frequency \( \omega_0 \), with width equal to \( \Gamma_c = \Gamma_u + \Gamma_z \), and this second term of equation (1) represents the completely redistributed component.

However, equation (1) is appropriate only for resonance transitions for which the lower level is a ground state or an excited state metastable to radiative decay. As discussed by Cooper et al. (1982), this formula is invalid for subsidiary transitions for which the lower level has a significant decay rate \( \Gamma_z \). Under these circumstances, and when \( \Delta \omega = \omega_2 - \omega_0 \) is large compared with radiative and collisional widths, equation (1) becomes modified to qualitatively

\[
p(\omega_1, \omega_2) = \frac{\Gamma_u}{\Gamma_u + \Gamma_1 + \Gamma_c} \left[ \frac{1}{\pi} \frac{\Gamma_1}{(\omega_2 - \omega_0)^2 + \Gamma_1^2} \right] + \frac{\Gamma_c + \Gamma_1}{\Gamma_c + \Gamma_1 + \Gamma_u} f(\omega_2 - \omega_0),
\]

where the width of \( f(\omega_2 - \omega_0) \) now has increased by \( \Gamma_1 \) to \( \Gamma_1 = \Gamma_u + \Gamma_1 + \Gamma_c \).

The fact that radiation from the lower level of the transition may be trapped, so that the transition is optically thick, can cause repopulation of the lower level (an effect ignored by OSC). However, although this effect can produce changes in the width of the scattered Rayleigh component (Cooper and Ballagh 1978) (the first term in eq. [2]), and may also lead to a long effective lifetime for this lower level, it does not alter the redistributed term in any fundamental manner. Physically this is because any decay from the lower level destroys the optical dipole which gives rise to scattering.

Thus, even in the absence of collisions (\( \Gamma_z \to 0 \)), we still have redistribution. In this paper, we report experiments for which the lower levels of the transitions have significant radiative decay. Previous measurements (e.g., Carlsten, Szöke, and Raymer 1977; Kroop and Behmenburg 1980; Thomann, Burnett, and Cooper 1980; Behmenburg and Kroop 1983; Alford, Burnett, and Cooper 1983) have concentrated on ground states. In this experiment, we have concentrated principally on the He 1 2 1P–3 1D line (\( \lambda = 668 \) nm), although some work has been performed on Hz (\( \lambda = 656 \) nm) (Lombardi and Kelleher 1983).

The experimental apparatus is described in § II, the theoretical expectations are the subject of § III, the results are presented in § IV, and § V concludes with a discussion of the results and a comparison with theory. In particular, our basic conclusions concerning the role of the lower state in causing redistribution even in the absence of collisions are confirmed.

II. EXPERIMENT

A schematic diagram of the experimental apparatus is depicted in Figure 1. Linearly polarized light from a Nd:YAG pumped dye laser, tuned to the wing of the spectral line being studied, was incident along the axis of a 6 mm diameter positive column discharge. For the previously reported (Lombardi and Kelleher 1983) measurements on Hz, the discharge ran in a 30% mixture of H2 in He; for the He measurements, the gas was pure He. The current in the positive column, which was...
varied up to 30 mA, was large enough such that all the H₂ was dissociated (Weber 1979).

The bandwidth and power density of the laser were 0.1 cm⁻¹ and approximately 100 kW cm⁻², respectively. Amplified spontaneous emission from the laser in resonance with the spectral line under investigation did not make a measurable contribution to the redistribution signal. This was verified by blocking the grating in the dye laser cavity and by tuning the grating to a wavelength far from the spectral line. In both cases, the signal vanished, indicating that the signal observed is entirely attributable to narrow-band light, the wavelength of which is tuned by the grating.

The collection optics consisted of a collimating lens (L₁), a Polaroid to analyze the fluorescence (P), an interference filter (I.F.), and a Fabry-Perot étalon (F-P), a focusing lens (L₂), and an aperture (A). The signal from the photomultiplier (PMT) was amplified and detected by a boxcar averager with a 15 ns gate.

The bandwidth of the radiation was 0.1 cm⁻¹ and approximately 100 kW cm⁻², respectively. Amplified spontaneous emission from the laser in resonance with the spectral line under investigation did not make a measurable contribution to the redistribution signal. This was verified by blocking the grating in the dye laser cavity and by tuning the grating to a wavelength far from the spectral line. In both cases, the signal vanished, indicating that the signal observed is entirely attributable to narrow-band light, the wavelength of which is tuned by the grating.

The signal from the photomultiplier (PMT) is processed by an amplifier and a boxcar averager with a 15 ns gate.

III. THEORETICAL CONSIDERATIONS

This section is concerned with the calculation of the ratio of the redistributed (fluorescent) intensity, 〈F(ω₁, ω₂)〉, to the Rayleigh (〈R(ω)〉) or to the redistributed intensity (〈I(R)〉 or to the fluorescence (〈F(ω)〉) as a function of the angle αv.

The general formula for redistribution with lower-state interaction is difficult to work with (see OSC, eq. [53]). However, for detunings of the radiation (ω₁ − ω₂) large compared to the line widths, and for regions of interest such that (ω₁ − ω₂) (Rayleigh) or (ω₁ − ω₂) (fluorescence) are also comparable with the respective line widths, we obtain for the angle-averaged scattering

\[
\langle F(\omega_1, \omega_2) \rangle_{\text{angle av.}} \approx \frac{\mathcal{P}(\omega_1)}{\pi (\omega_1 - \omega_2)^2 + \gamma_{el}^2} + \left(\frac{2\gamma_{el}^2}{\Delta_{el}^2 + \gamma_{el}^2}\right) \mathcal{P}(\omega_2) \tag{3}
\]

This corresponds to the K = 0 multipole (Ballagh and Cooper 1977), and the notation of OSC is used. Here

\[
\mathcal{P}(\omega) = \frac{\gamma_{el}^2}{(\omega - \omega_0 - \Delta_{el}^2 + (\gamma_{el}^2)} + \gamma_{el}^2 \tag{4}
\]

with

\[
2\gamma_{el}^2 = \Gamma_u + \Gamma_l + 2(\gamma_{el}^2)_{el}
\]

where \(\gamma_{el}^2\) is the collisional line broadening width and

\[
\gamma_{el}^2 = \Gamma_l + (\gamma_{el}^2)_{el} = \Gamma_u + \kappa_u
\]

This results in the ratio of fluorescent (Iₕ) to Rayleigh (Iₙ) intensity

\[
I_R = \frac{I_I + Q_k + \kappa_i}{\Gamma_u + \kappa_u} \tag{5}
\]

(Minor modifications occur to these collisional rates, \(\kappa_i\), if inelastic collisions repopulating the levels are significant [Cooper and Ballagh 1978; Cooper et al. 1982].)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>30% H₂ in He</th>
<th>Pure He</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n_e) (cm⁻³)</td>
<td>4.5 × 10¹⁰</td>
<td>3.4 × 10¹⁰</td>
</tr>
<tr>
<td>(n_{H^2}) (cm⁻³)</td>
<td>8 × 10¹⁵</td>
<td>8 × 10¹⁵</td>
</tr>
<tr>
<td>(n_{H}) (cm⁻³)</td>
<td>7 × 10¹⁵</td>
<td>...</td>
</tr>
<tr>
<td>(T_e) (eV)</td>
<td>5.3</td>
<td>5.5</td>
</tr>
</tbody>
</table>

TABLE 1

Density and Temperature of Various Plasma Components for a Discharge Current of 27 mA

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With regard to polarization of the emitted fluorescence, fine-
and hyperfine-structure effects are quite subtle for H2 owing to
different summations over intermediate states of the \( F_{ij} \) and
\( F_{i'j} \) terms of OSC; see also Lombardi and Kelleher
1983). For the He 1 2 \(^{1}P-3 \(^{1}D\) line, however, the hyperfine
splitting is negligible and we can treat it as a simple \( p \rightarrow d \)
transition. Then the results of Ballagh and Cooper (1977) for
the fluorescent component may be used directly.

It is convenient to define the polarization parameter \( \beta \)
in terms of the intensity of scattered light polarized parallel to \( I_{\parallel} \)
and perpendicular to \( I_{\perp} \) the incident radiation:

\[
\beta = \frac{2(I_{\perp} - I_{\parallel})}{I_{\parallel} + 2I_{\perp}}. \tag{6}
\]

This quantity is related to the degree of linear polarization, \( p \), by

\[
\beta = \frac{4p}{3 - p}. \tag{7}
\]

Using Ballagh and Cooper (1977, eqs. [25] and [28]) for \( I_{\parallel} \)
\( (\epsilon_{1}, \epsilon_{2} = 1) \) and \( I_{\perp} \) \( (\epsilon_{1}, \epsilon_{2} = 0) \) (where \( \epsilon_{1} \)
and \( \epsilon_{2} \) are the polarization vectors), results in

\[
\beta = 7 \left( \frac{(2\gamma_{ad} / \gamma_{c}) - 1}{10 \left[ (2\gamma_{ad} / \gamma_{c}) - 1 \right]} \right) - 1
\]

\[
= \frac{7}{10} \left( \Gamma_{\Gamma} + Q_{\Gamma} + \kappa_{u} + \kappa_{i} - \gamma_{depol} \right) \frac{(\Gamma_{u} + \kappa_{u})}{(\Gamma_{u} + \gamma_{depol})}. \tag{8}
\]

The last form follows, since we can write \( \gamma_{c} = \Gamma_{\Gamma} + \gamma_{c} \), \( \Gamma_{u} + \gamma_{depol} \). The collisional term \( (\gamma_{c} / \Gamma_{u}) \), is the rate of destruction
of alignment, which we will refer to as depolarization collisions
\( (\gamma_{depol}) \).

To the extent that collisional rates are small compared with
\( \Gamma_{u} \) (but not necessarily \( \Gamma_{\Gamma} \)) and to the extent that \( \kappa_{i} \) is small
compared with \( \Gamma_{u} \), this equation takes on the particularly simple form

\[
\beta = \beta_{0} \frac{\Gamma_{u}}{\Gamma_{u} + \gamma_{depol}}, \tag{10}
\]

where \( \beta_{0} = 0.7 \).

It should be noted that the form of equation (10) is equivalent
to the initially prepared polarization \( \beta_{0} \) being destroyed by
subsequent depolarizing collisions during the lifetime \( \Gamma_{u} \).
We shall see this adequately describes the He 1 line under investigation.

Actually, the above formulae are only strictly valid for a steady
state situation, whereas we report experimental results due to a finite-duration pulse. However, when the pulse is
"adiabatic," the formulation developed by Kleiber et al. (1983)
can be used. In our context, "adiabatic" implies that the time
scale on which the pulse changes significantly is long compared to
\( \Delta^{-1} \), where \( \Delta = o_{2} - o_{1} \) is the laser detuning, which is the
case for these experiments. Then it is straightforward to show
(using Kleiber et al. 1983) that equations (5)–(10) may be
used with \( I_{\perp} \) and \( I_{\parallel} \) replaced by their time integrals for a given pulse
(i.e., \( \int I_{\perp} \) dt and \( \int I_{\parallel} \) dt). This is hardly surprising, since "steady
state" may be considered as a series of "adiabatic" pulses, which
turn on and off sufficiently slowly that no switching
transients of the atomic dipole are created. We note that \( \int I_{\perp} \)
dt is integrated for a given pulse, and the time integral must be
extended beyond the length of the pulse, since fluorescence

TABLE 2

<table>
<thead>
<tr>
<th>( (o_{2} - o_{1})/2\pi ) ( (\text{cm}^{-1}) )</th>
<th>Pressure ( \text{torr} )</th>
<th>( I_{\perp}/I_{\parallel} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>-3.0</td>
<td>0.4</td>
<td>27</td>
</tr>
<tr>
<td>-1.0</td>
<td>0.4</td>
<td>20</td>
</tr>
<tr>
<td>-1.0</td>
<td>1.0</td>
<td>17</td>
</tr>
<tr>
<td>-1.0</td>
<td>3.0</td>
<td>31</td>
</tr>
</tbody>
</table>

associated with it occurs also after the termination of the pulse.
For a short pulse there is little fluorescence during the pulse.
Then we can show, again using the method of Kleiber et al.
(1983), that for such a short pulse (i.e., \( \gamma T_{p} \ll 1 \), where \( T_{p} \) is
the pulse length)

\[
\beta(t) = \frac{2[I_{\parallel}(t) - I_{\perp}(t)]}{I_{\parallel}(t) + 2I_{\perp}(t)} = \frac{(2\gamma_{ad} - \gamma_{c})}{(2\gamma_{ad} - \gamma_{c})} \exp \left( -\frac{\gamma_{c}T_{p}}{\gamma_{ad}} \right). \tag{11}
\]

Note that this is entirely equivalent to equation (8) when we
integrate the numerator and denominator separately over time.

IV. RESULTS

a) \( I_{\perp}/I_{\parallel} \) Measurements

The ratio of fluorescence to Rayleigh intensity was measured for
the He 2 \(^{1}P-3 \(^{1}D\) line. The results are presented in Table 2.
The rather large scatter in these data is attributable to the use of
a 15 ns gate on the boxcar averager. While the Rayleigh
emission has the same temporal development as the laser pulse
(5 ns FWHM), the fluorescent intensity has a decay time deter-
mined by the radiative decay lifetime and by collisional
quenching. In the absence of collisions, the decay half-life of the
fluorescence is 15 ns. Consequently, not all of the fluorescent
light is collected, and the measured ratio tends to underesti-
mate \( I_{\perp}/I_{\parallel} \). In spite of the large uncertainty in \( I_{\perp}/I_{\parallel} \), it is pos-
sible to draw some conclusions regarding redistribution in the
presence of lower-level broadening. These conclusions are dis-
cussed in the following section.

b) Polarization Measurements

Detailed measurements were made of the polarization of the
He 2 \(^{1}P-3 \(^{1}D\) line with a pressure of 0.4 torr. The measured
time dependence of \( \beta \) is shown in Figure 2. The line drawn
through the points is a least squares fit to the data of the form

\[
\beta(t) = \beta_{0} \exp \left( -\gamma_{depol} t \right). \tag{11}
\]

We infer \( \beta_{0} = 0.64 \pm 0.07 \) and \( \gamma_{depol} = (5.2 \pm 0.5) \times 10^{7} \text{ s}^{-1} \).
The uncertainties quoted are obtained from the statistical
properties of the data, and do not include possible systematic
effects. It should be noted that the time is measured from the
peak of the laser pulse. Since the laser pulse has a finite
duration \( (T_{p} \approx 5 \text{ ns}) \), the atoms observed after a delay time \( t \) rep-
resent a range of delays. However, it is easy to verify that this
correction is of order \( (\gamma_{depol} T_{p})^{2} \), which is negligible for
\( \gamma_{depol} = 5.2 \times 10^{7} \text{ s}^{-1} \).

For completeness, we also give some results for H2 at a fixed
pressure (0.7 torr) and several laser detunings. The results are
given in Table 3. Since the polarization (when collisions are
unimportant) is expected to be independent of laser detuning for
detunings large compared to the fine-structure splitting (as
appears to be the case), the mean, weighted according to the
uncertainties of the data, is also tabulated.

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V. DISCUSSION

In the absence of collisions, equation (5) predicts $I_p/I_k = \Gamma_p/\Gamma_k$. These radiative rates are well known for the $2 \, ^1P$ and $3 \, ^1D$ levels (Wiese, Smith, and Glennon 1966), which imply that $I_p/I_k = 28.2$ (using $\Gamma_p = 6.4 \times 10^4$ and $\Gamma_k = 1.8 \times 10^5$). Considering the experimental difficulties mentioned in § IV, this is in very satisfactory agreement with the results presented in Table 2. However, we must estimate the other parameters involved before drawing definite conclusions.

From the laser excitation experiments of Dubreuil and Catherinot (1980), we infer an inelastic rate of $\kappa_e \approx 3 \times 10^9 \text{ s}^{-1}$ at 0.4 torr He. Stark broadening (also mainly inelastic) would contribute, according to the calculations of Griem (1964), about an order of magnitude less ($\kappa_e \approx 3 \times 10^5 \text{ cm}^{-1}$). The inelastic rates for the lower level are expected to be even smaller.

The collisional broadening rates, $(\tau_{el})_i$, for van der Waals and resonance broadening may also be estimated from the theoretical expressions of Griem (1964). The values for van der Waals and resonance broadening for He at 0.4 torr are calculated to be $2.4 \times 10^7$ and $1.2 \times 10^8 \text{ s}^{-1}$, respectively. This gives the total $(\tau_{el})_i$ as about $1.5 \times 10^8 \text{ s}^{-1}$. According to equation (5), this changes $I_p/I_k$ to about 32. This emphasizes the relatively weak pressure dependence in this low-pressure regime.

On the other hand, the above calculation is changed dramatically if $\Gamma_i$ is neglected, as would be the case if there were no fluorescence in the absence of collisions. Then, for He at 0.4 torr, $I_p/I_k \approx 4.4$ would be predicted. This result is definitely in disagreement with the experimental values of the ratio. The polarization measurements for the $2 \, ^1P-3 \, ^1D$ line are also in agreement with the predictions, since $\beta_0 = 0.64 \pm 0.07$ compares very favorably with the prediction of $\beta_0 = 0.7$. Further, $\gamma_{dep} = 5.2 \times 10^7 \text{ s}^{-1}$, which, as expected, is of the same order of magnitude as the van der Waals broadening (Ballagh and Cooper 1977) (because we would expect these collisions to dominate in the depolarization rate).

Although less quantitative, the Hz results are also in agreement with the expectations (Lombardi and Kelleher 1983); in particular, given a predicted $\beta_0$ of 0.53, the inferred $\gamma_{dep}$ of $1.8 \times 10^8$ (from Table 3) is entirely consistent with the estimated broadening rate of $2.4 \times 10^8 \text{ s}^{-1}$.

We conclude, therefore, that lower-state radiative decay is very important for determining redistribution in subsidiary lines, especially when the collisional rate is small compared with the dominant radiative rate.

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