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TIME AND SPACE DEPENDENCY OF
RADIATIVE TRANSFER IN MAGNESIUM

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FINAL report

Lawrence Livermore National Laboratory (#4294105)

Time and Space Dependence of Radiative Transfer in Magnesium

(Report submitted by J. Cooper)

As outlined in the previous report (see Appendix A) agreement between experiment and calculations in which partial redistribution of radiation is taken into account for radiative transfer in magnesium vapor is at the 10-15% level.¹ The major uncertainty was associated with a systematic (10%) increase in magnesium density during the (12 hours) experiment. In an attempt to explain this discrepancy a new cell was constructed in which the temperatures of the cell and "cold finger" could be independently varied over a large range. This cell showed very stable density measurements with a $\Delta\lambda^{-2}$ variation with respect to the magnesium resonance line, which indicated pure Rayleigh scattering and no evidence of impurities. In addition, it was not possible to form barium particulates even when the "cold finger" was hotter than the cell. Our conclusion, therefore, remains that we had an impurity problem (possibly MgO particulates) in the previous cell, and that, in that case, the initial density measurements were appropriate. We are writing up this work by publication.²

During the last 9 months we have mainly concentrated our efforts on examining the fundamental assumptions associated with the "standard" radiative transfer theory.

On the theoretical side, we noted that the Payne formulation for time dependent redistribution does not take into account the difference between the time scales for Rayleigh scattering (essentially instantaneous) and redistribution (at the radiative decay time Γ_{ul}^{-1}).³ A time dependent redistribution function $R(\nu, \nu'; t-t')$ is obtained:

$$R(\nu, \nu; t-t') = \Gamma_{ul} e^{-\Gamma_{ul}(\nu, \nu; t-t')} \phi_{\nu} \phi_{\nu}' + 2\delta(t-t') \frac{\Gamma_{ul}}{\Gamma_{ul} + \Gamma_E} \left\{ R_{II}(\nu, \nu') - \phi_{\nu} \phi_{\nu}' \right\}$$

For situations in which the time scale for processes of interest and the natural decay time are comparable, this could cause major differences in radiative transfer calculations.

The full angle-polarization-and time-dependent transfer equations have been obtained and are being written up for publication.⁴

Our main effort has pursued the detailed measurements of the radiation field using the sapphire probe technique (see Figures 1 and 2 and Appendix A). Unfortunately we could not obtain sufficient signal with the present set-up to frequency resolve the signal using the Fabry-Perot. Calculations indicated that these measurements were marginally possible, however, there were also difficulties in alignment with the existing apparatus, since the Fabry-Perot plates, which are highly reflecting at 285.2 nm (the Mg resonance line), are essentially transparent in the visible.

We were able to make high quality total intensity measurements,⁵ as shown in Figure 3. The polar plots associated with these measurements (Figure 4) show that the radiation field is highly anisotropic, although somewhat less so at late times. The angle-averaged theory predicts a rapid build up in intensity (see Figure 5), whereas the observations, especially for relatively large polar angles, show a build up on a time scale comparable to the population.

These results indicate the need for further detailed angular-dependent transfer theory and calculations.

References

1. A. Streater, Ph.D. Thesis, University of Colorado
2. A. Streater, J. Cooper, K. Burnett and W. Molander (to be published).
3. A. Streater, J. Cooper and W.J. Sandle, "On time-dependent radiative transfer," JQSRT (in press).
4. J. Cooper, A. Streater and D.J. Rees (in preparation).
5. A Streater, J. Cooper and K. Burnett (in preparation).

Figure 1 Schematic of the sapphire probe experiment.

Figure 2 Sapphire probe construction.

Note the interesting build-up of the signal to a maximum 50 ns after the laser pulse in the 45° case.

Figure 3

Signal at early and late times

Polar plots: Intensity vs. Angle

probe distance: 1 mm from initial excitation
pressure: 34 Torr Ar.

Early Times (39-88 ns)

Late times (88-425 ns)

More isotropic
at late times

probe distance: 2 mm from initial excitation
pressure: 34 Torr Ar.

Early times (39-88 ns)

Late times (88-425 ns)

Further away -
more concentrated
in forward
direction

Figure 4

Figure 5

Progress Report and Proposed Work

TIME-DEPENDENT RADIATIVE TRANSFER IN MAGNESIUM VAPOR

Introduction

After an initial pulsed excitation in a magnesium vapor, the excited state density has been measured as a function of time and space (1). The vapor is optically thick at frequencies in the center of the resonance line, so the excited state density spreads out slowly (compared to the natural decay rate of the excited state) as the radiation diffuses out of the initial excitation region. The vapor also contains argon atoms, which collisionally perturb the magnesium atoms. Line shape and frequency redistribution effects on the radiative transfer process are studied by varying the density of argon atoms in the vapor.

Results have been compared with a time-dependent partial redistribution radiative transfer theory (PRD theory), which is based on the Heasley-Kneer angle-averaged emission coefficient (1,2,3,4). A solution based on the assumption of complete redistribution, which ignores correlations between absorption and emission of light by the magnesium atoms (CRD theory), was also generated. The angle-averaged PRD calculations, which include the coherent Rayleigh scattering contribution, were found to fit the data more consistently than the CRD calculations.

An exploratory experiment has also been performed that measures the radiation field directly. One end of a small sapphire rod protrudes into the vapor, which acts as a light pipe to transport the light, unaltered in frequency, out of the vapor and into a detector. If the diameter of the probe

is less than the optical depth of the radiation, the presence of the probe should not significantly affect the radiative transport process. Large directional anisotropies in the radiation field were observed. The experiment also showed that a more detailed experiment, in which the light from the probe is resolved in frequency using a Fabry-Perot interferometer, is feasible.

Excited State Density Experiments

A laser pulse excites a column of magnesium atoms from the 3^1S ground state into the 3^1P excited level. In order to assure that the excitation is independent of position along the beam, and to prevent self-focusing effects, the frequency of the excitation laser is detuned slightly from resonance, and the 3^1P level is populated by absorption during collisions (5,6). The excitation is then allowed to diffuse out due to radiative transfer. At a later time, the excited state population is probed with a time-delayed probe laser (7), which brings excited 3^1P atoms to a higher 5^1S state. The subsequent fluorescence out of the higher 5^1S state is observed. This fluorescence is proportional to the excited state 3^1P density at the time and spatial position of the probe laser pulse. Figure 1 shows the relevant energy levels and transitions.

In the progress report submitted last year, spatial distributions of the excited state density at various times were presented. In this report we shall discuss the time-dependence of the excited state density in the central region of the initial excitation.

A schematic diagram of the apparatus is shown in Figure 2. In order to prevent changes in the characteristics of the probe beam upon changing the time delay between the excitation laser and the probe laser, the optical fiber delay used for time delays in the spatial distribution measurements described

in the previous progress report was replaced with a separate frequency-doubled YAG laser to pump the probe dye laser. The time delay was achieved electronically by timing the Q-switches of the two YAG lasers.

The density is measured by comparing off-resonance Rayleigh scattering off the magnesium vapor with Rayleigh scattering off a known amount of argon when the cell is cold and no magnesium is present (8).

Typical results for high and low perturber pressures are shown in Figure 3. Theoretical curves, using measured parameters, calculated using the Heasley Kneer emission coefficient (PRD theory), and calculated under the assumption of complete redistribution (CRD theory), are also shown. Note that at low perturber densities, frequency correlations theoretically have a greater effect. Data was also taken at higher and lower magnesium densities, and was repeated to test reproducibility. An analysis of the data indicates that the angle-averaged partial redistribution theory, which includes coherent Rayleigh scattering, fits the data better (under two different measures of fit) than the complete redistribution theory (1). As discussed below the major uncertainty of these results is related to the density measurement.

Radiation Field Measurements

In this experiment, which is depicted in figure 4, one end of a small diameter sapphire rod protrudes into the vapor, acting as a light pipe to probe the radiation field directly. In addition to a time and spatial dependence, the radiation field depends on direction, frequency, and polarization. The sapphire probe apparatus is capable of measuring the radiation field intensity as function of space, time, frequency, and direction.

The spatial and directional dependence is studied by changing the

position of the probe relative to the position of the excitation laser. The time dependence is monitored by recording the photomultiplier signal with a transient digitizer. The frequency of the light could be resolved using a Fabry-Perot interferometer, although in this exploratory experiment this was not done.

As in the excited state density experiments, the effect of collisions can be studied by varying the pressure of a perturber gas. As in previous experiments, argon was used.

As long as the magnesium density is kept low enough that the optical depth at line center is larger than the diameter of the probe, the presence of the probe should not significantly affect the radiative transfer process. The sapphire probe must be heated to a temperature slightly above the temperature of the magnesium sample in order to prevent vapor from condensing onto it, which would prevent light from entering. The construction of the probe assembly is shown in Figure 5.

Sample signals are shown in Figures 6a and 6b, which are keyed to the positions shown in Figure 7. Large directional anisotropies are observed (up to a factor of 3 or more), as shown in Figures 6a and 6b, where the distances from the excitation beam are roughly the same, but the direction of acceptance in figure 6a is significantly different than in figure 6b. This observation puts the isotropic radiation field assumption used in most radiative transfer calculations (including the calculations discussed in the preceding section) into question. Note also that in Figure 6b the intensity builds up in time, and then decays, which is not expected on simple theoretical grounds.

Future Proposed Work

The largest uncertainty in the excited state density experiments is the ground state magnesium density measurement. The Rayleigh scattering is found to increase by about 10% over a period of 12 hours, which introduces a systematic uncertainty into the density measurement. Experimental results at higher argon pressures suggest that the magnesium density is not actually increasing. The effect may be due to particles, perhaps of magnesium oxide, forming and being suspended in the convective currents of the heated vapor. Such particles can scatter light very efficiently, and can affect the Rayleigh scattering measurement of the magnesium density. Comparison of theory and experiment at high argon densities, where CRD should be valid, indicated that the initial density measurements are close to those expected. Further experimental work to determine the source of this increased scattering is proposed in order to more fully understand the experimental results.

Further radiation field measurements using the sapphire probe, in which the frequency is resolved using a Fabry-Perot interferometer, are also proposed. These frequency resolved measurements could determine whether the major "streaming" component is associated with photons in the line wings. Such measurements could thus answer interesting questions regarding the directional anisotropies which have been observed.

REFERENCES

- 1) Alan Streater, Ph.D. Thesis, University of Colorado (1985).
- 2) H.N. Heasley and F. Kneer, Ap.J. 203, 660-666 (1976).
- 3) J. Cooper, R.J. Ballagh, K. Burnett and D.G. Hummer, Ap.J. 260, 299 (1982).
- 4) W.E. Alley, J.Q.S.R.T. 30, 571 (1983).
- 5) A. Omont, E.W. Smith and J. Cooper, Ap.J. 175, 185 (1972).
- 6) K. Burnett and J. Cooper, Phys. Rev. A 22, 2044 (1980).
- 7) W. Molander, M. Belsley, A. Streater and K. Burnett Phys. Rev. A 29, 1548 (1984).
- 8) W.J. Alford, N. Anderson, K. Burnett, and J. Cooper, Phys. Rev. A 30, 2366 (1984).

Fig. 1. Energy level diagram showing the excitation laser, trapped fluorescence, probe laser, and observed fluorescence.

Fig. 2. Schematic of the experimental apparatus.

Fig. 3. Peak height results, for high and low pressure, with CRD and PRD solutions fit with an overall multiplicative factor.

Fig. 4. Schematic of the sapphire probe experiment. In the exploratory experiment described in the text, the Fabry-Perot interferometer was not used.

Fig. 5. Sapphire probe construction.

Fig. 6a.

Fig. 6b.

Fig. 7. Probe position Key for Fig. 6.