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LEVEL

RESEARCH IN GAS LASERS

T.F. Morse
Principal Investigator

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I INTRODUCTION

As noted in our renewal proposal, we have concerned ourselves with several problems, of both a theoretical and more immediately practical nature and believe we have made progress toward our understanding of several difficult problems. The main thrust of our present theoretical efforts has been toward a systematic and sound derivation of rate equations for a three-level molecular or atomic system. Such equations have immediate relevance to several schemes of laser gas diagnostics involving non-linear Raman processes, as well as newer diagnostic techniques associated with saturation laser induced fluorescence. These techniques are of importance to the non-perturbative measuring of the properties of gas flow fields over a wide temperature range [1]. We have, in this analysis, used a time-scaling approach that reduces, in the proper limits, to the two-level system rate equations derived from the density matrix by Carlsten [2].

The work on the rate equation descriptions of pulse propagation in an inhomogeneously broadened medium is complete, and is being prepared for submission to the Journal of Quantitative Spectroscopy and Radiation Transfer, and will be discussed below.

During the first and second weeks of this past September, I visited several laser institutes in Germany, and attended the Second International Conference on Gas Dynamic and Chemical Lasers, where I presented a paper on a novel technique for isotope separation [3]. During this conference, I had ample opportunity to discuss several new ideas on laser diagnostic techniques that show promise to lead toward fruitful experiments. One technique is concerned with the use of a laser doppler velocimeter in which measurements can be performed in the absence of a scattering center. In essence, the scheme uses a TEA CO_2 laser to create a microplasma - a gas breakdown at the focal point of the laser within a flowing gas. The radiation scattered coherently from the plasma is heterodyned to produce a beat signal that can be shown to be proportional to velocity. The theoretical development and preliminary experimental work on this has been under the aegis of the AFOSR. It is possible that such a technique might be applicable in a hostile combusion environment.

In conjunction with Prof. J. Cipolla, of the Department of Mechanical Engineering, Northeastern University, who is spending a sabbatical year working in our group at Brown, we have been working on some kinetic boundary problems of radiative transfer. In particular, the quenching of excited particles by wall interaction, can influence the interpretation of fluorescent measurements. The results of research on these topics will be commented upon in more detail in that which follows. These studies are pertinent to all processes in which radiation measurements are made on low to moderate pressure gases, and may have particular application to the technique of resonance saturation fluorescence.

II AFOSR SUPPORTED RESEARCH

A. Pulse Amplification - Attenuation in a Two-Level Gas

By considering the kinetic equations and equation of radiative transfer for a two-level gas, it has been possible to obtain a simple algorithm that

yields the pulse spreading or narrowing effects as the pulse propagates through either an amplifying or absorbing medium. The limit of pulse duration long compared with the coherence time has been chosen, so that true coherence effects such as self-induced transparency are absent. However, the system of rate equations is velocity dependent, so that doppler broadening mechanisms are included. A simple algorithm allows the calculation of pulse spreading in a two-level amplifying gas with mixed homogeneous - inhomogeneous broadening. Furthermore, this technique permits a simple calculation of the effects of pulse spreading due to inhomogeneous effects in a weakly absorbing gas. This would have particular application to problems of pulse propagation through a weakly attenuating atmosphere at high altitudes in the neighborhood of an absorption line, and would also encompass the more difficult problem of pulse propagation through an atmosphere of variable density [4].

B. Plasma Velocimeter

In this small scale experimental program, we have been attempting to determine if it is possible to utilize a plasma breakdown in air (caused by a focussed CO_2 TEA laser) to serve as a scattering center. The basic concept is that the tail of the pulse scattered from the plasma, (a one atmosphere plasma will be opaque to 10.6 µ radiation) can be heterodyned to give a beat signal proportional to velocity. Thus far, we have been able to determine that the magnitude of the scattered signal provides ample signal for our purposes at short ranges of interest. We have, however, encountered severe noise problems associated with the CO_2 laser discharge, and have been forced to take measures to assure proper isolation of the laser and the detector. It is hoped that within the near future we will meet with success in the actual heterodyning of the signal reflected from the plasma. It is anticipated that this technique may prove of some use in those situations in which doppler velocimetry is not possible due to a lack of a sufficient number of naturally occurring scattering centers [5].

C. Boundary and Kinetic Effects in Gas Lasers

This work has been developed in conjunction with Prof. J. Cipolla of Northeastern University, who is spending a sabbatical year at Brown. As a continuation of earlier studies on the optical excitation of gases [6,7], a resonance transition in a plane layer of gas pumped by an external source has been treated using kinetic theory. Our objective has been the proper assessment of transparent boundaries on the intensity of radiation and the effect of using the correct gas kinetic boundary condition on the particle velocity distribution functions. In this calculation, we have used previously formulated kinetic equations coupled with the equation of radiative transfer to describe a specialized gas model characteristic of visible resonant transitions in low density, high temperature gases. The treatment has been phenomenological throughout, neglecting both true coherence effects among the atomic states as well as phase phenomena in the radiation field. In the gas model chosen, the ground and the excited levels of the resonant transition are separated by a series of intermediate levels to which upper level particles may relax during spontaneous radiative decay. This artifice introduces a branching ratio, B_{10}, into the equations in

such a way that it may serve as a perturbation parameter. The use of this perturbation then enables us to generate approximate solutions for both the particle and photon distribution functions but places no restriction on either the form or intensity of the exciting radiation.

Detailed calculations have been completed for a one-dimensional slab of gas irradiated externally. The results of the perturbation analysis demonstrate the strong effect of a transparent solid boundary on both the spatial variation of the excited level density and on the frequency dependence of the radiation emitted from the slab in the normal direction. We find, in particular, that all quantities are dependent on the parameters δ and α, where δ is the slab thickness, in photon mean free paths, and α represents the mean number of photon mean paths travelled by an excited level particle before decay to any of its allowed terminal states. Large and small δ represent optically thick and thin conditions respectively. The relevance of α is that it measures the importance of particle streaming in de-exciting the gas. For α small, emissions occur over length scale small compared to the e-folding distance of the local radiation field; therefore, excitation and decay occur under nearly identical radiative conditions and such an approximation, ($\alpha = 0$) leads to velocity independent rate equations and (in the limit of two level atoms) to the Bibermann-Holstein [8,9] description of the transfer of resonant radiation. This is valid, however, only for locations far removed from boundaries, in the vicinity of which there always exists a thin layer in which the radiation field varies rapidly and in which streaming (particle) is important. Consequently, the limit $\alpha \to 0$ is singular. Our approximate solution demonstrates the nature of this singularity and shows its effect on the macroscopic gas properties. In particular, the excited level density exhibits non monotonic spatial behavior in layers near the boundary rather than the monotone decay characteristic of the $\alpha = 0$ limit. In addition, without a proper assessment of these features, the emission from the gas normal to the slab boundary can be in error by 30% or more in the near wings of the line, even for relatively small values of α. Perhaps a more striking effect on the emitted radiation is the non-monotonic frequency dependence (known as line reversal) that first occurs for optically thick slabs ($\delta \sim 10$), and for $\alpha \sim .1$, but is totally absent for $\alpha = 0$.

Additional research in this area is concerned with the use of laser induced fluorescence and the relationship of the fluorescence intensity to the kinetic state of the gas. The effects of a finite laser beam have recently been considered by Stern [10] in the optically thin limit ($\delta \to 0$), but without the presence of solid boundaries. Comparisons are also made with recent work on this problem employing a discrete velocity model [11]. Our work has progressed to the point at which we are presently preparing results for publication.

D. Time Scaling the Density Matrix to Obtain Three-Level Rate Equations

There are many examples, both in optical pumping as well as many diagnostic situations in a flowing gas in which a description incorporating the behavior of three atomic or molecular levels is mandatory. All optically pumped lasers are in this category, and there are many flow diagnostic techniques in which at least a three-level description is needed.

We have succeeded in formulating the density matrix equations in such a manner that for situations in which a pulse or pumping time is longer than a dephasing time (this has been done previously for the case of the two-level atom), equivalent rate equations result that explicitly contain contributions that are non linear in the field. These equations were developed previously on a purely phenomenological basis [12], however, it was believed that a derivation from first principles would be more convincing and perhaps lead to a wider usage. As with previous work on the density matrix, phenomenological collision terms have been introduced so that we may account for V-V, V-T, V-R and T-R collisions, each phenomena properly characterized by its own relaxation time for the diagonal elements. In the proper limits, these results reduce to those of Temkin and Panock [13]. In the matrix representation of these equations, the various contributions associated with the linear and non-linear phenomena become particularly clear. In the limit of strong pumping fields, the non-linear effects simplify and both homogeneously broadened and inhomogeneously broadened limits may be obtained. In addition, for the case in which we have a CW field, and in which level degeneracy in a gas is removed by a pulsed Stark field, we may exactly solve for the response of this system to the pulsed field. This case has been treated as a perturbation elsewhere, where only exponential time behavior in a perturbation to the third order in the [14,15] field is recovered. Our solution, for this physical situation, is valid to all orders in the field strengths, and, for the case of a strong CW field, particularly simple solutions result that exhibit the characteristic "ringing" in the approach to a steady state. It is expected that this material will be prepared for publication in the near future.

In conclusion, we think it fair to state that we have solved the difficulties in developing suitable model equations for radiating gases that exhibit non-linear two photon behavior. We have, through our matrix formalism, been able to solve exactly a problem previously treated by perturbation methods. It is planned that we will look toward using these equations on a variety of problems in the coming year. We should also mention with regard to our non-linear rate equations, that we may also plan to investigate the non-linear effects of the real part of the susceptibility in hopes of exploring whether any phase sensitive diagnostic techniques may be developed to supplant those that consider only the gain. Other authors have recently pursued this direction [17].

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Submitted by:

T. F. Morse

T.F. Morse
Professor of Engineering
Principal Investigator

Carl Cometta

Carl Cometta
Executive Officer
Division of Engineering

TFM:ga

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The work on the rate equation descriptions of pulse propagation in homogeneous media is complete and is being prepared for publication. The Pulse duration limit was chosen as long compared to the coherence time so that true coherence effects such as self-induced transparency is absent. However, the system of rate equations are velocity dependent so droppler broaden mechanisms are included. Attempts have continued to determine if it is possible to utilize a plasma break down in air to deduce velocity. Thus far, it has been determined that the magnitude of the scattered signal provides ample signal for short ranges. This →next page

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technique will prove useful where conventional doppler velocimetery is not possible due to the absence of naturally occurring scattering centers. The density matrix equations for three energy level gases have been formulated. For situations were the pulse or pumping time is longer than a dephasing time, the rate equations explicity contain contributions that are nonlinear.