ISOTOPE DEPENDENCE OF GAS LASER INTENSITY PROFILES

by

Gerald A. Royce

A Thesis Submitted to the Faculty of the
COMMITTEE ON OPTICAL SCIENCES

In Partial Fulfillment of the Requirements
For the Degree of

MASTER OF SCIENCE

In the Graduate College

THE UNIVERSITY OF ARIZONA

1971
STATEMENT BY AUTHOR

This thesis has been submitted in partial fulfillment of requirements for an advanced degree at The University of Arizona and is deposited in the University Library to be made available to borrowers under rules of the Library.

Brief quotations from this thesis are allowable without special permission, provided that accurate acknowledgment of source is made. Requests for permission for extended quotation from or reproduction of this manuscript in whole or in part may be granted by the head of the major department or the Dean of the Graduate College when in his judgment the proposed use of the material is in the interests of scholarship. In all other instances, however, permission must be obtained from the author.

SIGNED: [Signature]

APPROVAL BY THESIS DIRECTOR

This thesis has been approved on the date shown below:

[Signature]
MURRAY SARGENT III
Assistant Professor of Optical Sciences
The advantage of using a gas containing just one isotope over a gas made up of many isotopes has been commonly realized in the commercial laser market where peak power and efficiency are important parameters. When single isotope gases are available the gases are used instead of the naturally occurring mixtures. Examples of these are the common usage of the isotope neon-twenty and the cadmium 114 in the helium-neon and the helium-cadmium lasers respectively.

Despite the obvious advantage of considering just single isotope gases, there have been a few papers on laser theory that have included facilities for handling isotope mixtures. At the suggestion of my advisor, Dr. Murray Sargent III, I began this study on the effects of isotopes in laser media. After an extensive, but by no means complete search of the literature, I found no numerical calculations had been reported on isotope mixtures. The text that follows is a rework and simplification of the many isotope theory with numerical results.

Since we have written two papers and delivered one paper to a professional society on this subject while working together on a daily basis and with constant interaction, it is almost impossible to separate the contributions that Dr. Sargent made to this study from my own and no attempt will be made. Most of the background material, the change in emphasis, and the new material that appears in this thesis, particularly Chapter III, is due to my independent work.
I am indebted to Dr. Sargent for his suggestion of this topic, his persuasion that it should be done, and his enthusiastic response to every sign of progress no matter how minor. I am also grateful to my employer, the Navy Weapons Laboratory, Dahlgren, Virginia for the financial support that I received through their Full-Time Advanced Study Program, and the opportunity to undertake this work.
TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>LIST OF ILLUSTRATIONS</td>
<td>vi</td>
</tr>
<tr>
<td>ABSTRACT</td>
<td>viii</td>
</tr>
<tr>
<td>I. INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>II. SEMICLASSICAL THEORY FOR MULTIPLE ISOTOPE LASER</td>
<td>6</td>
</tr>
<tr>
<td>Single Isotope Medium</td>
<td>6</td>
</tr>
<tr>
<td>Many Isotope Medium</td>
<td>16</td>
</tr>
<tr>
<td>III. STRONG SIGNAL THEORY</td>
<td>27</td>
</tr>
<tr>
<td>IV. ISOTOPES IN THE ZEEMAN LASER</td>
<td>40</td>
</tr>
<tr>
<td>APPENDIX A: INFLUENCE OF ISOTOPES ON INVERSION</td>
<td>48</td>
</tr>
<tr>
<td>APPENDIX B: SEMICLASSICAL SCALAR THEORY IN THE DOPPLER LIMIT</td>
<td>51</td>
</tr>
<tr>
<td>APPENDIX C: ISOTOPES OF LASER GASES</td>
<td>59</td>
</tr>
<tr>
<td>APPENDIX D: ZEEMAN LASER PARAMETERS</td>
<td>61</td>
</tr>
<tr>
<td>REFERENCES</td>
<td>63</td>
</tr>
</tbody>
</table>
# LIST OF ILLUSTRATIONS

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Single Isotope Energy Level Diagram</td>
<td>7</td>
</tr>
<tr>
<td>2</td>
<td>Self-Consistency Theory Flow Chart</td>
<td>8</td>
</tr>
<tr>
<td>3</td>
<td>Homogeneous and Inhomogeneous Broadening of Gas Media</td>
<td>15</td>
</tr>
<tr>
<td>4</td>
<td>Single Isotope Intensity-Detuning Profiles</td>
<td>17</td>
</tr>
<tr>
<td>5</td>
<td>Energy Level Diagram for Two Isotopes</td>
<td>20</td>
</tr>
<tr>
<td>6</td>
<td>Unsaturated Gain Versus Frequency for Two Neon Isotopes</td>
<td>21</td>
</tr>
<tr>
<td>7</td>
<td>Intensity-Detuning Profiles for Different Isotope Abundancies</td>
<td>23</td>
</tr>
<tr>
<td>8</td>
<td>Intensity-Detuning Profiles for Two Isotopes of Equal Abundancies</td>
<td>24</td>
</tr>
<tr>
<td>9</td>
<td>Intensity-Detuning Profiles for Near-Natural Neon Mixture</td>
<td>25</td>
</tr>
<tr>
<td>10</td>
<td>Illustration of $N(I,\Delta)$ to $I(N,\Delta)$ Interpolation</td>
<td>31</td>
</tr>
<tr>
<td>11</td>
<td>Comparison of Strong Signal Theory to Semiclassical Theory for Single Isotope Medium</td>
<td>32</td>
</tr>
<tr>
<td>12</td>
<td>Comparison of Strong Signal Theory to Semiclassical Theory for Two Equal Isotopes</td>
<td>33</td>
</tr>
<tr>
<td>13</td>
<td>Relative Excitation-Detuning Curves for Single Isotope</td>
<td>34</td>
</tr>
<tr>
<td>14</td>
<td>Relative Excitation-Detuning Curves for Two Equal Isotopes</td>
<td>35</td>
</tr>
<tr>
<td>15</td>
<td>Relative Excitation-Detuning Curves for Natural Neon</td>
<td>36</td>
</tr>
<tr>
<td>16</td>
<td>Strong Signal Intensity-Detuning Profiles for Single Isotope</td>
<td>37</td>
</tr>
<tr>
<td>17</td>
<td>Strong Signal Intensity-Detuning Profiles for Two Isotopes</td>
<td>38</td>
</tr>
<tr>
<td>18</td>
<td>Strong Signal Intensity-Detuning Profiles for Natural Neon</td>
<td>39</td>
</tr>
<tr>
<td>Figure</td>
<td>Description</td>
<td>Page</td>
</tr>
<tr>
<td>--------</td>
<td>---------------------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>19</td>
<td>Magnetic Field Splitting of Atomic Levels</td>
<td>41</td>
</tr>
<tr>
<td>20</td>
<td>Population Differences of Zeeman $\sigma^+$ and $\sigma^-$ Transitions</td>
<td>42</td>
</tr>
<tr>
<td>21</td>
<td>Magnetic Tuning Profiles for Single Isotope</td>
<td>44</td>
</tr>
<tr>
<td>22</td>
<td>Magnetic Tuning Profiles for Equal Mixture of Two Isotopes</td>
<td>45</td>
</tr>
<tr>
<td>23</td>
<td>Magnetic Tuning Profiles for Natural Neon</td>
<td>47</td>
</tr>
<tr>
<td>A1</td>
<td>Neon Excitation</td>
<td>49</td>
</tr>
</tbody>
</table>
ABSTRACT

The gain of a gas laser medium is dramatically altered by the mixing of atoms which belong to different isotopes of the active element. Compared with the single isotope gas, the maximum gain of an isotope mixture is reduced and the gain is frequency broadened.

In this thesis we determine the single mode output intensity as a function of cavity detuning for lasers having various isotope mixtures. Both the linearly polarized (Brewster-windowed) and Zeeman lasers are considered. The central tuning (Lamb) dip associated with single isotope medium gives way to more complicated tuning dependences such as multiple dips for certain combinations of isotope concentrations and excitation levels. Under axial magnetic fields, the isotope mixtures produce intensity profiles that have more structure than those for the single isotope medium.
Javan, Bennett and Herriot (1961) built the first gas laser utilizing a mixture of helium and neon. The system oscillated at 1.15\mu in the near infrared region of the spectrum and emitted a continuous, but weak, beam of highly monochromatic light. Since that early, but important, accomplishment, many gas lasers have been developed, some very rugged and powerful. Theories to predict and/or describe the performance of various laser devices have flowed in the mainstream of the rapidly developing technology. This present thesis is concerned with the effects of isotope mixtures on the intensity output of a laser. The discussion is aimed specifically, but not inevitably at the helium-neon laser operating in the red (632.8nm).

Perhaps the first experimental indication that isotopes influenced the operation of lasers can be found in the paper by McFarlane, Bennett and Lamb (1963). The initial attempt to observe the existence of a dip in the power output of a laser oscillating in a single axial mode as this mode is tuned through the line center of the laser transition was hampered by the presence of more than one isotope of neon in the discharge tube. They did report a slightly asymmetric curve of intensity versus tuning and suggested that the distortion was due to the mixture of Ne\(^{20}\) and Ne\(^{22}\) in natural neon in the ratio of 9:1. In the
same paper they reported the appearance of the dip (hereafter called Lamb dip) in the tuning profile of the same laser with 99.5% enriched Ne\textsuperscript{22}.

Before lasers, Naguoka and Mishima (1930) observed the isotope effect in the emission spectrum of neon. With natural neon gas they noticed that by cooling their discharge capillary to liquid air temperatures and operating at low neon pressures that they could detect the presence of a weak line on the shorter wavelength side of the strong neon line. The experiment required a high resolution Fabry-Perot interferometer where the mirror spacing was about four centimeters. At higher pressures or increased operating temperatures the individual structures of the lines became ill-defined as the lines broadened and merged.

Schrödinger (1926) derived an expression for the Rydberg constant $R$ for lines of element with atomic weight $M$ given by

$$ R = R_{\infty} \frac{M}{M + m} $$  \hspace{1cm} (1)

where

$$ R_{\infty} = \frac{2\pi^2 e^2 E^2 m}{h^3} $$

with $e$ and $E$ denoting the charges of electron and nucleus respectively, $m$ the mass of the electrons and $h$ the Planck's constant. In the case of interest, a simple calculation by putting $M = 20$ and 22 shows the difference in wavelength of Ne\textsuperscript{22} from Ne\textsuperscript{20} to be

$$ \Delta \lambda = 2.461 \times 10^{-6} \lambda $$  \hspace{1cm} (2)
or the change in frequency to be

\[ \Delta \nu = +2.461 \times 10^{-6} \nu. \]  \hspace{1cm} (3)

The calculated values derived using Equations 2 and 3 are not of the right order of magnitude for the neon isotopes, but they do correctly indicate the direction of the shift. To describe the isotope shift by just an altered Rydberg constant is quite questionable. The expression for \( R \) is only approximate, since no account was taken of the electric and magnetic moment of the nucleus in its derivation. In addition to changes in mass, there are also changes in the nuclear radius as one goes from one elemental isotope to another. The radius change is particularly important for the heavy elements (Herzberg 1944).

For the 632.8nm laser transition, the shift between the principle neon isotopes derived from Equation 3 is 0.0389cm\(^{-1}\) or approximately 1160 MHz. Naguoka and Mishima (1930) measured the shift to be 0.054cm\(^{-1}\) or 1620 MHz. More recent experimental observations have indicated lower values for the shift. Cordover, Jaseja and Javan (1965) measured the shift between two stabilized lasers, one filled with Ne\(^{20}\) and the other filled with Ne\(^{22}\). Through beating experiments and interferometer studies, they found the shift to be 875 ± 12 MHz. Schweitzer (1968) measured the shift by placing two gain tubes in his laser, each with a different neon isotope. By alternating which gain tube was lit and recording fringes with a scanning Fabry-Perot interferometer, he arrived at a frequency shift of 901 ± 20 MHz. Using an interferometer laser of the type described by Smith (1965), Zory (1966) was able to measure the
separation between the two Lamb dips of a laser filled with equal amounts of Ne\(^{20}\) and Ne\(^{22}\). Although the position of the dip minimum should be pushed in the direction of smaller gain and hence the measured shift was expected to be higher, the dips roughly indicated the position of the center frequency of the two isotopes and the reported separation between dip minima was 909 ± 15 MHz.

There have been a few theoretical discussions of the contributions of isotopes mixtures to lasers. Aronowitz (1965) and Whitney (1969) extended the standing wave laser theory of Lamb (1964) to the ring laser and described procedure to work with multiple isotope systems. Sargent, Lamb and Fork (1967a, 1967b) extended the Lamb model to cover the multi-isotope Zeeman laser. The approach of Sargent et al. (1967a) is used in the theoretical development that follows in the next chapters.

Chapter II is concerned with the theoretical results of Lamb (1964) for a single isotope laser operating single mode. An extension to handle many isotopes is then given and illustrated by the intensity versus cavity tuning curves for two active isotopes. In Chapter III, a rate equation theory is used to treat multi-isotope mixtures. Stenholm and Lamb (1969) have shown this approximate theory to be quite accurate, even at high intensities. A brief introduction to the Zeeman laser based on the work of Sargent et al. (1967a) is then provided in Chapter IV with results for one and two isotope media under the influence of an axial magnetic field. Pressure broadening effects as described by Gyorffy, Borenstein and Lamb (1968) are neglected for the most part,
although parameters used in the calculations reflected some pressure broadening.
CHAPTER II

SEMICLASSICAL THEORY FOR MULTIPLE ISOTOPE LASER

Single Isotope Medium

Before developing the many isotope theory, it is desirable to review briefly the semiclassical theory of Lamb (1964), which is directly applicable to single isotope laser. The theory requires that electromagnetic fields be treated as scalar classical quantities and the atoms be treated quantum mechanically. One can insure the applicability of this theory by using Brewster windows inside the laser cavity to cause the field to be linearly polarized, i.e., scalar. The atoms are considered to have two levels a and b between which laser action can take place (see Fig. 1). The separation between the two levels corresponds to a frequency \( \omega \). These levels are excited at rates \( \lambda_a \) and \( \lambda_b \), while the atomic decay constants are given by \( \gamma_a \) and \( \gamma_b \).

Lamb's model assumes a classical electromagnetic field \( E(r,t) \) inside of an optical resonator and an active medium of thermally moving atoms which acquire nonlinear electric dipole moments under the action of the field according to the laws of quantum mechanics. The corresponding macroscopic electric polarization of the medium \( P(r,t) \) acts as a source for a reaction electric field. The condition for laser oscillation is then that the assumed field to be just equal to the reaction field. The scheme is represented in Figure 2. The calculation is in
Fig. 1. Single Isotope Energy Level Diagram.

The separation between the energy levels a and b corresponds to a frequency $\omega$ where a and b decay with constants $\gamma_a$ and $\gamma_b$ respectively.
Fig. 2. Self-Consistency Theory Flow Chart.
three parts: (a) electromagnetic field differential equations are developed to relate \( E(r,t) \) to \( P(r,t) \), (b) quantum mechanical equations are used to obtain microscopic polarization for a single atom, and (c) statistical summation (averaging) is used to arrive at the macroscopic polarization.

Considering single mode, single frequency operation, the electric field in the laser is given by

\[
E(z,t) = \frac{1}{2} E(t) \exp[-i(\nu t + \phi(t))] U(z) + \text{complex conjugate}, \quad (4)
\]

where \( E(t) \) and \( \phi(t) \) are real, slowly varying (with respect to an optical cycle) amplitude and phase quantities, respectively, and \( \nu \) is the oscillation frequency of the laser. \( U(z) \) is the normal mode function of the cavity which we take as

\[
U(z) = \sin K z \quad (5)
\]

with the wave number

\[
K = \frac{n\pi}{L}, \quad (6)
\]

where \( n \) is a large integer and \( z \) is the axial coordinate.

From Maxwell's equations, one finds the self consistency relations

\[
\dot{E}(t) = -\frac{1}{2}(\nu/Q)E - \frac{1}{2}(\nu/\varepsilon_0) \text{Im} P(t) \quad (7)
\]

\[
\nu + \phi = \Omega - \frac{1}{2}(\nu/\varepsilon_0) E^{-1} \text{Re} P(t), \quad (8)
\]
where $Q$ is the quality factor of the cavity mode, the complex $P(t)$ denotes a slowly varying polarization, and $\Omega$ is the resonant frequency of the passive cavity.

For a single isotope medium, the complex polarization is given by

$$P(t) = 2 \rho \exp[i(\nu t + \phi)] \cdot \frac{2}{L} \int_0^L dz \int_{-\infty}^{\infty} dv \rho_{ab}(z,\nu,t), \quad (9)$$

where $\rho$ is the electric dipole moment matrix element for an atom. $\rho_{ab}(z,\nu,t)$ is an off-diagonal element of the density matrix and it is related to the transition probability of ensemble of atoms at place $z$ at time $t$ with $z$ component of velocity $\nu$ going from level $a$ to level $b$. Perturbation techniques are used to arrive at the microscopic contributions of $\rho(z,\nu,t)$ from which the macroscopic polarization $P$ is calculated. Specifically, first order perturbation gives

$$\text{Re} \ P^{(1)}(t) = - (\rho^2/\hbar \kappa u) \overline{N} Z_r(\nu - \omega)E \quad (10)$$

$$\text{Im} \ P^{(1)}(t) = - (\rho^2/\hbar \kappa u) \overline{N} Z_i(\nu - \omega)E. \quad (11)$$

Here $\overline{N}$ is called the average excitation defined by

$$\overline{N} = \frac{1}{L} \int_0^L dz \ N(z,t), \quad (12)$$

where $N(z,t)$ refers to an excitation density. The speed parameter $u$, not to be confused with the previous $U(z)$, is the average speed of the
atoms related to an effective temperature $T$ by

$$\frac{1}{2} M u^2 = k_B T,$$

(13)

where $M$ is the atomic mass and $k_B$ is the Boltzmann constant. The parameters $Z_T(\nu - \omega)$ and $Z_i(\nu - \omega)$ are the real and imaginary parts of what is commonly known as the complex plasma dispersion function. The function $Z$ is given by

$$Z(\nu - \omega) = iKu \int_0^\infty d\tau \exp(-\nu\tau - \frac{1}{4} (Ku)^2 \tau^2)$$

(14)

where

$$u = \gamma + i(\nu - \omega)$$

(15)

with

$$\gamma = \frac{1}{2} (\gamma_a + \gamma_b) + \gamma \text{ phase.}$$

(16)

The third order perturbation provides

$$\text{Re } P^3(t) = \frac{1}{8} \pi^{1/2} \frac{1}{N} [p^4/(\hbar^3 \gamma a \gamma b Ku)]$$

$$\cdot \left[ (\omega - \nu)L(\omega - \nu) \gamma^{-1} \right] E^3 \exp[-(\nu - \omega)^2/(Ku)^2]$$

(17)

$$\text{Im } P^3(t) = \frac{1}{8} \pi^{1/2} \frac{1}{N} [p^4/(\hbar^3 \gamma a \gamma b Ku)]$$

$$\cdot \left[ 1 + \gamma(\omega - \nu) \right] E^3 \exp[-(\nu - \omega)^2/(Ku)^2]$$

(18)
where the Lorentzian function

\[ L(\omega - \nu) = \frac{\gamma^2}{\gamma^2 + (\omega - \nu)^2} \]  \hspace{1cm} (19)

Using only contributions to third order

\[ P(t) = P^0(t) + P^3(t) \]  \hspace{1cm} (20)

one finds the amplitude relation from Eq. (7)

\[ \dot{E} = \alpha E - \beta E^3 \]  \hspace{1cm} (21)

where the linear net gain

\[ \alpha = -\frac{1}{2} (\nu/Q) + \frac{1}{2} \sqrt{\gamma} \left[ \frac{\rho^2}{\epsilon_0 \hbar K_u} \right] Z_i (\nu - \omega) \]  \hspace{1cm} (22)

and the self-saturation coefficient in the Doppler Limit \( \nu \ll K_u \)

\[ \beta = \frac{\nu}{16} \pi \frac{1}{\sqrt{\gamma}} \left[ \frac{\rho^2}{\epsilon_0 \hbar \gamma} \frac{ab^4}{\alpha' b' K_u} \right] \]

\[ \cdot \left[ 1 + L(\nu - \omega) \right] \exp\left[ -\left( \omega - \nu \right)^2 / K_u^2 \right] \]  \hspace{1cm} (23)

From Eq. (21), the steady state solution \( \dot{E} = 0 \) occurs for intensity

\[ E^2 = \frac{\alpha}{\beta} \]  \hspace{1cm} (24)

Expressing the coefficients \( \alpha \) and \( \beta \) in terms of a ratio \( \gamma^* = \frac{N}{N_0} \) called relative excitation, we find

\[ \alpha = \frac{1}{2} (\nu/Q) \left[ \frac{Z_i (\nu - \omega)}{Z_i (0)} \right] \gamma^* - 1 \]  \hspace{1cm} (25)
\[ \beta = \frac{\pi^2}{16} \left( \frac{\nu}{Q} \right) \left[ \frac{p^2 \gamma_\text{ab}}{(\hbar^2 \gamma_\text{a}\gamma_\text{b} \gamma_\text{i}(0))} \right] \]

\[ \cdot \left[ 1 + L(\nu - \omega) \right] \exp\left[ -\frac{(\nu - \omega)^2}{(\kappa \alpha)^2} \right] . \quad (26) \]

Here \( \overline{N}_T \) is the minimum excitation required to threshold oscillations in the cavity. For a single isotope medium, \( \overline{N}_T \) occurs for the cavity frequency \( \Omega \) tuned to the line center \( \omega \) of the atomic resonate curve. For \( \alpha = 0 \) and \( \nu - \omega = 0 \) in Eq. (22), we obtain

\[ \overline{N}_T = \epsilon_0 \frac{\hbar \kappa \alpha}{p^2 Q \gamma_\text{i}(0)} . \quad (27) \]

The expression for \( \alpha(\nu) \) as in Eq. (25) has the form of a truncated gaussian which is centered at \( \omega \) and that for \( \beta(\nu) \) as in Eq. (26) has the form of a biased lorentzian which also is centered at \( \omega \). The result of dividing \( \alpha \) by \( \beta \) to find intensity as in Eq. (24) gives a dip in the intensity versus tuning profile for large relative excitations, if \( \kappa \alpha \gg \gamma_{\text{ab}} \). The dip should occur for relative excitation somewhat above threshold, i.e., for relative excitation

\[ \eta > 1 + 2 \left( \frac{\gamma_{\text{ab}}}{\kappa \alpha} \right)^2 , \quad (28) \]

as can be shown by setting

\[ (\delta^2 I/\delta \nu^2) \nu - \omega > 0 . \]

To understand the occurrence of the dip, we first consider an atomic medium consisting of a single isotope. The frequency response of the atoms is not perfectly sharp but has a width owing to spontaneous
emission, collisions and other processes. This line broadening is called homogeneous, for all atoms are affected similarly. So for stationary gas atoms, the frequency response would have width due to homogeneous broadening alone which for low pressure the width is equal to $\gamma_{ab}$.

In a gas laser media, the atoms have velocities typically on the order of $10^5$ cm/sec which cause Doppler-shifted frequency responses of an inhomogeneous nature. Each atom responds most strongly to light that is at a frequency close to its up- or down-shifted response, as shown in Fig. 3. The overall response is therefore broadened. Since various atoms respond differently to incident light, this broadening is inhomogeneous. The line width is commonly referred to as the Doppler width $Ku$.

The interaction of a standing-wave electric field

$$E(z,t) = E_0 \cos(\nu t) \sin Kz,$$  \hfill (29)

with the atomic medium is quite frequency dependent. This field can be written as the sum of two oppositely directed running waves

$$E(z,t) = \frac{1}{2} E_0 [\sin(\nu t + Kz) - \sin(\nu t - Kz)] \hfill (30)$$

For $\nu > \omega$, the atoms that respond are those whose $z$ components of velocity cause $\nu$ to be Doppler downshifted to $\omega$. Because there are two running waves, two ensembles of atoms contribute to the laser action, each moving in the direction of one of the waves. The Lamb dip results because near central tuning ($\nu = \omega$) the atoms contributing to the laser
Fig. 3. Homogeneous and Inhomogeneous Broadening of Gas Media.

The individual atomic response curves are superimposed on the inhomogeneously broadened line for a possible laser medium. The homogeneous contribution is much narrower than the inhomogeneous contribution and, as such, the medium is said to be inhomogeneously broadened.
action are nearly stationary and comprise a single ensemble rather than
the pair contributing to detuned oscillation. The dip occurs for
$K_u \gg \gamma$ and sufficiently high excitation since there are less atoms con-
tributing to the laser action at line center than at some points off
resonance (see Fig. 4).

**Many Isotope Medium**

In the previous section we dealt with active atoms belonging to
one isotope only, for example, $^{20}\text{Ne}$ as the active atoms in the He-Ne
laser. In this section we will ultimately describe the case of two iso-
topes of the active gas, but first we will be more general and discuss
the many isotope theory.

Following the work of Sargent et al. (1967a), the contributions
of the various isotopes to the polarization can be treated separately
so that the total polarization is the sum

$$ P = \sum_j a_j P_j, \quad (31) $$

where $P_j$ is the contribution of the $j^{\text{th}}$ isotope and $a_j$ is the fractional
abundance of the $j^{\text{th}}$ isotope for which the normalization condition

$$ \sum_j a_j = 1 \quad (32) $$

must hold. Again only using contribution to the third order, the polari-
ization is given by

$$ P = \sum_j a_j P_j + \sum_j a_j P_j^{3j} \quad (33) $$

Fig. 4. Single Isotope Intensity-Detuning Profiles.

The curves are given by Eq. (24) where the intensity is \((\pi^{1/8}/8)(RE)^2(\gamma_0/\gamma)(\kappa^2\gamma_0^2\gamma)^{-1}\)
for \(K\omega = 1010 \text{ MHz, } \gamma = 80 \text{ MHz, } \gamma_0 = 1.01, 1.05, 1.10, 1.15, 1.2.\)
and the amplitude relation from Eq. (7) is still

\[ \dot{E} = \alpha E - \beta E^3, \tag{34} \]

but \( \alpha \) and \( \beta \) are found by summing over all the isotopes so that

\[ \alpha = \sum_j a_j \alpha_j = -\frac{1}{2} \nu/Q + \frac{1}{2} \nu \overline{N}[p^2/(\varepsilon_0 \hbar)] \cdot \sum_j (a_j/\kappa u_j) Z_i (\nu - \omega_j) \tag{35} \]

and

\[ \beta = \sum_j a_j \beta_j = \frac{\nu}{16} \pi^2 \overline{N}[\gamma_{ab} p^4/(\varepsilon_0 \hbar^3 \gamma_a \gamma_b)] \cdot \sum_j (a_j/\kappa u_j) (1 + \nu_j \nu - \nu) \exp\left[-(\omega_j - \nu)^2/\kappa u_j^2\right] \tag{36} \]

the steady state solution of Eq. (7) is again \( \dot{E} = 0 \) and

\[ E^2 = \frac{\alpha}{\beta}. \tag{37} \]

We now express \( \alpha \) and \( \beta \) in terms of the relative excitation parameter for the mixed isotope case, we find

\[ \alpha = \frac{1}{2} \nu \left[ \frac{\sum_j (a_j/\kappa u_j) \gamma_j Z_i (\nu - \omega_j)}{\sum_j (a_j/\kappa u_j) Z_i (\nu - \omega_j)} - 1 \right] \tag{38} \]

and

\[ \beta = \frac{\nu}{16} \pi^2 \gamma_{ab} [\gamma_2 (\gamma_a \gamma_b)] \sum_j (a_j/\kappa u_j) [1 + \nu_j \nu - \nu] \exp\left[-(\omega_j - \nu)^2/\kappa u_j^2\right] \left[ Q \sum_j (a_j/\kappa u_j) Z_i (\nu - \omega_j) \right] \tag{39} \]

where we have used

\[ \overline{N}_T = [Q (p^2/(\varepsilon_0 \hbar)) \sum_j (a_j/\kappa u_j) Z_i (\nu - \omega_j)]^{-1}. \tag{40} \]
where $\nu_k$ is the frequency for principle maxima of $\alpha$ and not necessary one of the line center frequencies $\omega_j$ as we had for the single isotope theory.

Using Eqs. (37), (38) and (39) the intensity can be expressed as

$$E^2 = \frac{\pi}{\Sigma_j} \left( \frac{\alpha_j/Ku_j}{\nu_k} \right) \frac{\Sigma_i}{\nu - \omega_i} - \frac{\Sigma_j}{\nu_k} \frac{\nu_{\alpha_i} - \omega_j}{\nu_{\alpha_i} - \omega_j} \frac{z_i^2}{z_j^2} \frac{(\nu_{\alpha_i} - \omega_j)^2}{(Ku)^2}. \quad (41)$$

The above expression is often modified to express the dimensionless intensity parameter

$$I_D = \frac{\pi}{\Sigma_j} \left( \frac{\alpha_j/Ku_j}{\nu_k} \right) \frac{\Sigma_i}{\nu - \omega_i} - \frac{\Sigma_j}{\nu_k} \frac{\nu_{\alpha_i} - \omega_j}{\nu_{\alpha_i} - \omega_j} \frac{z_i^2}{z_j^2} \frac{(\nu_{\alpha_i} - \omega_j)^2}{(Ku)^2}. \quad (42)$$

and in this paper the intensity is sometimes expressed as a fractional intensity

$$I_f = \frac{\pi}{4} I_D. \quad (43)$$

Now consider two active isotopes in the laser medium (see Fig. 5). The isotopes are characterized by slightly different energy levels and different line centers $\omega_1$ and $\omega_2$. Also the atoms of the heavier isotope tends to move more slowly and hence the Doppler width is slightly reduced. Specifically the average velocities are related by

$$u = \sqrt{M_1/M_2} \frac{u_1}{u_2}. \quad (44)$$

It is instructive to discuss the spread of gain for the equal abundance
Fig. 5. Energy Level Diagram for Two Isotopes.

The resonant frequencies differ by the isotope shift $\Delta_{\text{ISO}} = \omega_2 - \omega_1$. 
Fig. 6. Unsaturated Gain Versus Frequency for Two Neon Isotope.

Curves (solid) are due to the single isotopes drawn using the gaussian part of Eq. (25). Note they are centered about the $\omega_1$ and $\omega_2$ respectively. The resultant (dashed) using abundancies $a_1 = 0.5$ and $a_2 = 0.5$ gives a broader, lower profile than the single isotope curves. Assumed values are $K_u = 1010$ MHz, $\gamma = 80$ MHz, $\Delta_{150} = 875$ MHz.
case \( a_1 = a_2 = 0.5 \). In Fig. 6, we have sketched the gaussian part of the unsaturated gain for two isotopes. The resultant curve produced by multiplying by the appropriate abundancies and adding the individual gaussians shows how the gain is spread out and reduced.

In Fig. 7, the intensity (see Eq. 43) is plotted for a constant excitation level \( \bar{N} \) as the ratio of isotope abundancies is varied. The maximum obtainable intensity for the single isotope gas is much greater than that containing equal amounts of the two isotopes. The maximum obtainable intensity is greater for the neon twenty-two single isotope gas since its Doppler width is smaller. At low excitations, a single isotope gas may still have enough gain to oscillate even though an isotope mixture cannot.

In Figs. 8 and 9, the intensity output versus cavity detuning for natural and equal mixtures of isotopes is given as the relative excitation is varied. Note that there are two dips in the intensity curves of Fig. 9 at high excitation. These dips occur near the resonance frequencies \( \omega_1 \) and \( \omega_2 \), and are a direct result of the lorentzian dependencies of the single-isotope saturation coefficients \( \beta_1 \) and \( \beta_2 \). As in the case of the single dip, the double dip is due to the fact that at \( \omega_1 \) and \( \omega_2 \) a single ensemble of atoms replaces a pair contributing to detuned oscillation. We expect to observe the double dips at high excitation, but only when the contribution of one isotope in the region of the center frequency of the other isotope is small. The latter case usually means a large isotope shift such as that of the 632.8nm transition of neon. Criteria for dips are discussed in Appendix B. Since perturbation
Fig. 7. Intensity-Detuning Profiles for Different Isotope Abundancies.

Curves computed using Eq. (41) for fixed amount of excitation \( N \) corresponding to the relative excitation for the \(^{20}\text{Ne} \) medium \( \mathcal{N} = 1.2 \). The relative concentrations of the two isotopes are varied and the fractional abundance of the \(^{20}\text{Ne} \), indicated on the curves, go over the range 1 to 0, giving 100, 80, 50, 20, and 0% concentration in the total mixture. Here \( K_{u_1} = 1010 \) MHz, \( K_{u_2} = 969 \) MHz, \( \gamma = 80 \) MHz, and laser wavelength = 632.8nm.
Fig. 8. Intensity-Detuning Profiles for Two Isotopes of Equal Abundance.

Curves computed from Eq. (41) showing the effect of changing the amount of excitation. Here $^{20}\text{Ne}$ and $^{22}\text{Ne}$ are mixed equally ($a_1 = a_2 = 0.5$) and the relative excitations are 1.01, 1.1, 1.3, 1.7. Dips appear in the curves for the higher excitations. Other parameters are the same as in Fig. 7.
Fig. 9. Intensity-Detuning Profiles for Near-Natural Neon Mixture.

Curves computed from Eq. (41) showing the effect of changing the relative excitation for a near-natural mixture of neon ($a_1 = 0.9, a_2 = 0.1$). $\gamma = 1.01, 1.1, 1.3, 1.5, 1.7$. An asymmetric dip appears at the higher excitations. Other parameters are the same as in Fig. 7.
techniques valid for low values of the electric field were used, our theory is not quantitatively valid for the maximum intensities encountered ($\mathcal{E} = 1.7$). Nevertheless, it should be reasonably accurate for the lower intensities encountered in the dip regions, and it agrees qualitatively with the experiments of Zory (1966).
CHAPTER III

STRONG SIGNAL THEORY

The perturbation approach described in the previous chapter has much to commend it. An explicit expression for the intensity as a function of detuning and relative excitation is obtained and operation near threshold is quite accurately described. However, lasers operating far above threshold are inadequately depicted by the third order theory and consequently a more exact treatment is warranted. Stenholm and Lamb (1969) have given such a theory for single isotope media.

Their calculation is based on expansions of the population differences and in quadrature contributions to the polarization in Fourier series. In as much as the mathematics is quite involved, we begin with a primary result of the theory, namely, the imaginary part of the single mode polarization

$$\text{Im} P = -p^2 E \sum_{\text{ab}} \left[ \int_{-\infty}^{\infty} \text{d}v \, W(v) \mathcal{N}(v, \omega - v, I_D) [1 + I_D \mathcal{N}(v, \omega - v, I_D)]^{-1} \right], \quad (45)$$

where $\mathcal{N}(v, \omega - v, I)$ is a continued fraction. We will assume the velocity distribution is Maxwellian.

$$W(v) = \frac{1}{\pi} \frac{1}{\sqrt{\pi}} \exp \left(-\frac{v^2}{\nu^2}\right) \quad (46)$$

The introduction of Eq. (45) into the amplitude equation (see Eq. (7)),
gives for steady state \( \dot{E} = 0 \)

\[
\frac{1}{\bar{N}} = \frac{p^2 Q}{\varepsilon_0 \pi_{ab}} \int_{-\infty}^{\infty} W(v) \phi \phi [1 + I_D \phi \phi]^{-1}.
\] (47)

In terms of relative excitation, Eq. (47) is just

\[
\frac{\bar{N}_T}{\bar{N}} = \frac{1}{\bar{N}_T} = \frac{\int_{-\infty}^{\infty} dv W(v) \phi \phi(v, \omega - \nu, I_D)[1 + I_D \phi \phi(v, \omega - \nu, I_D)]^{-1}}{\int_{-\infty}^{\infty} dv W(v) \phi \phi(v, \omega - \nu_T, 0)}
\] (48)

where \( \nu_T = \omega \) for single isotope.

For a single isotope media, and for an extreme Doppler limit
\( (K_u = 40\gamma) \), Stenholm and Lamb (1969) show that a good approximation
(rate equation approximation) to the strong signal theory can be found
by truncating the continued fraction \( \phi \phi \) after the first term. The lowest
continued fraction uses the value

\[
\phi \phi = \frac{1}{2} D(\omega - \nu, K \nu)
\] (49)

where

\[
D(\omega - \nu, K \nu) = \gamma^2 \left[ \gamma^2 + (\omega - \nu + K \nu)^2 \right]^{\frac{1}{2}} \left[ \gamma^2 + (\omega - \nu - K \nu)^2 \right]^{\frac{1}{2}}
\] (50)

Now Eq. (47) can be written as

\[
\frac{1}{\bar{N}} = \frac{\int_{-\infty}^{\infty} dx e^{-x^2} D [1 + \frac{1}{2} I_D D]}{\int_{-\infty}^{\infty} dx e^{-x^2} D}
\] (51)

where \( x = \nu/u \).
For more than one isotope, the polarization is the sum

\[ P = \sum_j P_j \]

as before, and Eq. (45) becomes

\[ \text{Im}P = -\rho^2 \bar{N} (\hbar \gamma_{ab})^{-1} \sum_j \int_{-\infty}^{\infty} dv \, W_j(v) \frac{\partial \gamma_j}{\partial v} [1 + I_D \varphi_j] . \tag{52} \]

Again applying the steady state conditions \( \dot{E} = 0 \), Eq. (7), and the truncated continued fraction, we arrive at the many isotope solution for relative excitation

\[ \gamma^{-1} = \frac{\sum_j \int_{-\infty}^{\infty} dv \, W_j(v)D(\omega_j - \nu, K\nu)[1 + \frac{1}{2} I_D D(\omega_j - \nu, K\nu)]}{\sum_j \int_{-\infty}^{\infty} dv \, W_j(v)D(\omega_j - \nu_T, K\nu)} . \tag{53} \]

Eqs. (51) and (53) determine intensity only implicitly as contrasted with the third order theory which gives an explicit expression for intensity as in Eq. (41). Nevertheless, we can compute relative excitations for various values of intensity and detuning and then construct tuning curves from which values of intensity for constant relative excitation can be determined as indicated in Fig. 10.

A computer program was written to calculate the integrals of Eq. (53) for a number of intensity values including \( I_D = 0 \). This program also linearly interpolates to arrive at \( I_D(\gamma, \nu) \) values whenever \( \gamma \) is designated using the equation

\[ I = I_{K-1} + \frac{I_K - I_{K-1}}{\gamma_K - \gamma_{K-1}} (\gamma - \gamma_{K-1}) . \tag{54} \]
The calculations were performed on the University of Arizona CDC 6400 system using Fortran IV.

Basically the program uses a simple trapezoidal rule integration over a large velocity range. Since the most important contribution to the laser action occurs in the velocity interval of order $\gamma_{ab}/K$ around $V = |\omega_j - \nu|/K$, it is advantageous to center the integration at $V = |\omega_j - \nu|/K$ and go in both directions. A number of preliminary runs were made to determine a convenient integration range and step size that would give good agreement between the strong signal theory and the third order theory where they should agree, that is, $I = 0$. A good fit was found for integration over the interval

$$\frac{|\omega_j - \nu| - 80\gamma}{K} < V < \frac{|\omega_j - \nu| + 80\gamma}{K}, \quad (55)$$

with step size $dV = \gamma/(5K)$, (see Figs. 11 and 12).

The relative excitation versus detuning curves for constant intensities and for three different isotope mixtures are shown in Figs. 13, 14 and 15. The interpolated intensity is then shown in Figs. 16, 17 and 18 respectively.

As shown in Figs. 11 and 12, the computed intensity for a particular detuning and relative excitation is higher for the REA calculation than that obtained via the perturbation theory of Chapter II. These two figures show that there are quantitative as well as qualitative differences that arise through application of the two theories.
Fig. 10. Illustration of $\mathcal{M}(I, \Delta)$ to $I(\gamma, \Delta)$ Interpolation.
Fig. 11. Comparison of Strong Signal Theory to Semiclassical Theory for Single Isotope Medium.

Here intensity versus relative excitation curves computed from Eqs. (24) and (48) using fixed detuning values ($\Delta = 0$ and 477 MHz). The intensity values are a good fit at the low $\gamma$ but at high $\gamma$ the semiclassical curves (dashed) are lower than the REA curves (solid). $Ku = 1010$ MHz, $\gamma = 80$ MHz.
Fig. 12. Comparison of Strong Signal Theory to Semiclassical Theory for Two Equal Isotopes.

Here intensity versus relative excitation curves computed from Eq. (53) using fixed detuning values (Δ = 0 and 477 MHz). K\(u_1\) = 1010 MHz, K\(u_2\) = 969 MHz and γ = 80 MHz.
Fig. 13. Relative Excitation-Detuning Curves for Single Isotope.

Curves computed from Eq. (48) using $Ku = 1010$ MHz, $\gamma = 80$ MHz, $I_D = 0.0, 1.2, \text{ and } 2.4$. 
Fig. 14. Relative Excitation-Detuning Curves for Two Equal Isotopes.

Curves computed from Eq. (53) using $K_{u_1} = 1010$ MHz, $K_{u_2} = 969$ MHz, $\gamma = 80$ MHz, $I_D = 0.0, 1.2$ and $2.4$, $a_1 = a_2 = 0.5$. 
Fig. 15. Relative Excitation-Detuning Curves for Natural Neon.

Curves appropriate for near natural neon ($a_1 = 0.9$, $a_2 = 0.1$) computed using Eq. (53). $I_D = 0.0$, 1.09 and 1.99. Other parameters are the same as in Fig. 14.
Fig. 16. Strong Signal Intensity-Detuning Profiles for Single Isotope.

Curves given by Eq. (48) using values of relative excitation $\mathcal{N} = 1.1, 1.3, 1.5, 1.7, 1.9$. $K_u = 1010\ MHz$, $\gamma = 80\ MHz$. 
Fig. 17. Strong Signal Intensity-Detuning Profiles for Two Isotopes.

Here the abundancies are equal \((a_1 = 0.5 = a_2)\) and the curves are computed using Eq. (53) using values of relative excitation \(\mathcal{H} = 1.1, 1.3, 1.5, 1.7, 1.9\). \(K_1 = 1010\) MHz, \(K_2 = 969\) MHz, \(\gamma = 80\) MHz.
Fig. 18. Strong Signal Intensity-Detuning Profiles for Natural Neon.

Using abundancies appropriate for natural neon ($a_1 = 0.9$, $a_2 = 0.1$), the curves were computed from Eq. (53) using the same parameters as in Fig. 17.
CHAPTER IV

ISOTOPES IN THE ZEEMAN LASER

Now we consider an internal mirror laser (without Brewster windows) subject to a dc axial magnetic field. Since the electric field is no longer constrained to oscillate with a given polarization, the field is given by

$$\mathbf{E}(z,t) = \frac{1}{2} \left[ E_+ e_+ \exp[-i(\nu_+ t + \phi_+)] + E_- e_- \exp[-i(\nu_- t - \phi_-)] \right] U(z) + \text{complex conjugate}$$  \hspace{1cm} (56)

containing two orthogonal circular polarizations

$$\hat{E}_\mp = 2^{-\frac{1}{2}} (\hat{i} \pm \hat{j})$$  \hspace{1cm} (57)

Here the amplitudes $E_+$, $E_-$ and phases $\phi_+$, $\phi_-$ are slowly varying quantities for these polarizations, and $\nu_+$, $\nu_-$ are the corresponding frequencies.

The 6328 Å transition in Ne has angular momenta $J_a = 1$ and $J_b = 2$ as depicted in Fig. 19. The electric dipole selection rules allow $E_+$ to induce and derive gain from transitions for which the change in magnetic quantum number between upper and lower levels is +1. Similarly, $E_-$ involves transitions for which this change is -1. A dc magnetic field splits the Zeeman sub-levels, thereby increasing the frequency differences for the former transitions ($\Delta m = +1$) and decreasing
Fig. 19. Magnetic Field Splitting of Atomic Levels.

Two-state atomic transition with upper state having angular momentum $J_a = 1$, lower state $J_b = 2$. Magnetic sublevels are shown with different energies as if under the influence of an applied magnetic field. Selection rules for axial magnetic fields require that $\Delta m = \pm 1$. Contributions to $E_+ (E_-)$ are from $\Delta m = \pm 1 (-1)$. 

Fig. 20. Population Differences for Zeeman $\sigma^+$ and $\sigma^-$ Transitions.

As the magnetic field is increased, the population (gain) shifts so that $\omega_+$ is increased.
those for the latter. The corresponding population differences for the transitions are depicted in Fig. 20. The gains for $E_+$ and $E_-$ are proportional to these differences. For a gas of single isotope, the gain curve is a symmetric, truncated gaussian whose width is limited by the Doppler width of the gas.

The steady-state intensity

$$I_+ = E_+^2 = \frac{\alpha_+ / \beta_+}{1 - C}, \quad (58)$$

where the "effective" linear net-gain coefficient

$$\alpha_+^\star = \alpha_+ - \theta_+ \theta_- (\alpha_- / \beta_-), \quad (59)$$

$\theta_+ \theta_-$ is the cross saturation coefficient coupling the polarizations and the coupling parameter $C = \theta_+ \theta_- / (\beta_+ \beta_-)$, (see Appendix D for more detail description of Zeeman laser parameters). The intensity $I_-$ is given by Eq. (58) with $+$ replaced by $-$. In particular, for a single isotope medium tuned to line center, the intensities reduce to

$$I_+ = I_- = \frac{\alpha / \beta}{1 + \sqrt{C}}, \quad (60)$$

In Fig. 21 we show the magnetic dip due to a single isotope medium when the laser cavity is resonant at the zero field line center. The dip that we see is a consequence of both the decrease in the coupling between $E_+$ and $E_-$ as the magnetic field is increased and a Lamb dip in which the atomic line centers are shifted with respect to the cavity.
Fig. 21. Magnetic Tuning Profiles for Single Isotope.

Intensity $I_+$ (or $I_-$) curves versus magnetic field given by Eq. (34) due to a single isotope medium when the laser cavity is tuned to the zero field line center. $K_u = 1010$ MHz, $\gamma = 30$ MHz, $J_a = 1$, $J_b = 2$, $\gamma_2 = 1.2$. 
Fig. 22. Magnetic Tuning Profiles for Equal Mixtures of Two Isotopes.

Curves of intensity $I_+$ (dashed) and $I_-$ (solid) versus magnetic field given by Eq. (32) for equal mixtures of two isotopes. The laser is tuned to the zero field threshold frequency $v_T$, $Ku_1 = 1010$ MHz, $Ku_2 = 969$ MHz, $\gamma = 30$ MHz, $J_a = 1$, $J_b = 2$, $\mathcal{N} = 1.2$. 
frequency rather than the reverse relationship encountered in the scaler laser dip.

Considering next a mixture of two isotopes with equal abundances \((a_1 = a_2 = 0.5)\), the threshold frequency \(v_T\) (cavity tuning that requires the least amount of excitation to cause laser oscillation) lies roughly halfway between the line centers of the isotopes. As shown in Fig. 22, the magnetic tuning dip for the cavity tuning \(v_T\) with this mixture is not as deep as that for the single isotope laser tuned to its \(v_T\). This is due to the fact that \(v_T\) differs from both \(\omega_1\) and \(\omega_2\) by more than the decay rate \(\gamma\), yielding operation outside the Lamb dip regime. Hence the magnetic tuning dip results from only the decrease in coupling \(C\) between the polarizations. Note that the \(I_+\) curve does not coincide with the \(I_-\) curve. This is due to the slightly different Doppler widths of the isotopes which result in an asymmetric total gain curve.

Finally, considering a mixture appropriate for natural neon \((a_1 = 0.9, a_2 = 0.1)\) and the corresponding cavity tuning \((v_T = \omega_1 + 47\text{MHz})\) we see in Fig. 23 that the magnetic tuning profile contains a dip with more structure than we have seen before because the small velocity saturation (Lamb dip) and the polarization coupling achieve their maximum values at different magnetic field strengths.
Fig. 23. Magnetic Tuning Profiles for Natural Neon.

Curves of intensity versus magnetic field given by Eq. (32) for natural neon (90% $^{20}$Ne and 10% $^{22}$Ne). The laser is tuned to zero field frequency $\nu_T = \omega_1 + 47$ MHz, $Ku_2 = 918$ MHz, $Ku_1 = 963$ MHz, $\gamma = 30$ MHz, $\gamma' = 1.2$, $J_a = 1$, $J_b = 2$. 
In the main part of this thesis, we assumed that the atoms of the neon isotope would have the same probability of being excited. A brief and non-rigorous discussion on the effects of isotopes on the excitation of the medium will follow in this section.

Consider the excitation mechanism for the 6328 Å helium-neon laser transition (see Fig. A1). The dominate excitation is due to "collisions of the second kind", where $^1S$ metastable helium atoms collide with neon atoms in the ground state and transfer energy so that the neon atoms are pumped to the $3S_2$ level. The energy of the $^4$He in the $^1S$ level is 20.61 ev and $^{20}$Ne in the $3S_2$ level has energy equal to 20.66 ev. From our knowledge of isotope shift (Chapter I), the corresponding levels for $^3$He and $^{22}$Ne will be lower for the former and higher for the latter. In particular, the energy difference between $^{22}$Ne and $^{20}$Ne is $\hbar \nu$ shift or

$$ (6.6 \times 10^{-34} \text{ Joules-sec})(875 \times 10^6 /\text{sec}) \approx 4 \times 10^{-6} \text{ ev}. $$

It is generally felt (private communication, A. D. White 1970) that the relatively small separation between neon levels has little effect on the probability of energy transfer from helium to neon and hence that our assumption is reasonable.
HELIUM

\[ \begin{align*}
1s & \quad 20.61 \text{ ev} \\
3s & \quad 19.82 \text{ ev}
\end{align*} \]

GROUND STATES

NEON

\[ \begin{align*}
1s & \quad 20.66 \text{ ev} \\
3s & \quad 20.66 \text{ ev} \\
3s & \quad 19.79 \text{ ev} \\
& \quad 6328 \text{ Å} \\
& \quad 1.15 \mu \\
& \quad 18.70 \text{ ev}
\end{align*} \]

Fig. A1. Neon Excitation
White and Gordon (1963) did demonstrate that there was a substantial increase in the power output of a He-Ne laser when \(^{3}\)He was substituted for \(^{4}\)He. On first glance, this observation seems erroneous since the \(^{4}\)He \(^{1}\)S level more closely matches the \(3S_{2}\) level of the neon isotopes and quantum mechanics predicts that the better the match the more chance for energy transfer. The answer lies not in the transfer of energy between helium and neon, but rather in the excitation of the helium isotopes. A characteristic of the helium discharge is that the electrons are more energetic on the average in the \(^{3}\)He gas and \(^{3}\)He atoms have a larger collision cross-section for the electrons than \(^{4}\)He. The combination of these two facts leads to the result that under similar conditions sufficiently more \(^{3}\)He atoms will be excited to the \(^{1}\)S level than \(^{4}\)He so that \(^{3}\)He is a better excitor even though it is less resonant with the neon.
APPENDIX B

SEMICLASSICAL SCALAR THEORY IN THE DOPPLER LIMIT

Given an electric field in the laser

\[ E(z, t) = \frac{1}{2} E(t) \exp \left[ -i(\nu t + \phi) \right] U(z) + \text{complex conjugate} \text{,} \quad (B1) \]

one can find from Maxwell's equations the self-consistency equations

\[ \dot{E}(t) = -\frac{i}{2} \left( \frac{\nu}{Q} \right) E - \frac{1}{2} \left( \frac{\nu}{\epsilon_0} \right) \text{Im}P(t) \quad (B2) \]

\[ \nu + \dot{\phi} = \Omega - \frac{1}{2} \left( \frac{\nu}{\epsilon_0} \right) E^{-1} \text{Re} \ P(t) \text{,} \quad (B3) \]

where \( \nu \) is the oscillation frequency of the laser. \( E(t), \phi(t) \) and the complex \( P(t) \) are slowly varying amplitude, phase, and polarization quantities, and \( Q, \Omega \) and \( U(z) \) are the \( Q \), the resonant frequency and the normal mode function of the passive cavity, respectively. For our purposes, we take \( U(z) = \sin(Kz) \) with the wave number \( K = \Omega/c \).

For a single isotope medium, the complex polarization

\[ P(t) = 2 \exp[i(\nu t + \phi)] \frac{2}{L} \int_{0}^{L} dz \int_{0}^{\infty} dv \rho_{ab}(z, v, t) \text{,} \quad (B4) \]

where the population matrix \( \rho(z, v, t) \) provides a quantum mechanical description of an ensemble of atoms at place \( z \), at time \( t \), and with \( z \) component of velocity \( v \). The polarization for the mixed isotope medium is given by a summation over the isotopes

\[ P(t) = 2 \exp[i(\nu t + \phi)] \sum_{j} \frac{2}{L} \int_{0}^{L} dz \ U(z) \rho_{ab}(j, z, v, t) \text{,} \quad (B5) \]
where the isotope fractional abundancies $a_j$ satisfy the normalization condition

$$\sum_j a_j = 1 \quad (B6)$$

and where $\rho(j,z,v,t)$ is the population matrix for the jth isotope. Hence to find the complex polarization for the self-consistency equations, Eqs. (B2) and (B5), we need only add contributions already calculated for the single isotope medium. For the latter, one has (in the Doppler limit $Ku \gg \gamma$)

$$P(j,t) = \rho^{(1)} + \rho^{(3)} = -\sqrt{\pi} \rho \frac{2}{N(\gamma_h Ku)}^{-1} \exp[-(\omega_j - \nu)^2/(Ku_j)^2] \cdot \{1 - 1/8 \rho^2 \gamma_{ab} (\gamma_h)^2 \gamma_a \gamma_b}^{-1} \{1 + L(\omega_j - \nu)\} \quad (B7),$$

where $\gamma = \gamma_{ab} + \gamma$ phase.

Combining with the self-consistency equations, one has the amplitude and frequency determining equations

$$\dot{E} = \alpha E - \beta E^3 \quad (B8)$$

$$\nu + \phi = \Omega + \sigma - \rho E^2 \quad (B9)$$

where the linear net-gain coefficient

$$\alpha = \sum_j \frac{1}{\epsilon} \sqrt{\rho} \frac{2}{N(\gamma_h \epsilon_0)}^{-1} \exp[-(\omega_j - \nu)^2/(Ku_j)^2] - \frac{1}{2} (\nu/Q), \quad (B10)$$

the self-saturation coefficient

$$\beta = \frac{1}{16} \sqrt{\pi} \rho \frac{4}{N} \gamma_{ab} (\gamma_h \epsilon_0 \gamma_a \gamma_b \gamma)^{-1} \cdot \sum_j \frac{1}{\epsilon} \sqrt{\rho} \frac{2}{N(\gamma_h \epsilon_0)}^{-1} \exp[-(\omega_j - \nu)^2/(Ku_j)^2] \{1 + L(\omega_j - \nu)\} \quad (B11),$$
the linear pulling term $\alpha$ vanishes in the Doppler limit and the self-pushing term

$$p = \frac{1}{16} \sqrt{\pi} \int p \, N_{ab} (\mu K_0 \gamma_a \gamma_b)^{-1} \sum (a_j / u_j) \exp\left[\frac{(\omega_j - \nu)^2}{(K u_j)^2}\right]$$

$$\cdot \left[\frac{(\omega_j - \nu)}{\gamma}\right] \cdot (\omega_j - \nu).$$

In the steady state ($\dot{E} = 0$), Eq. (B8) can be solved to yield the intensity

$$I = \frac{E^2}{\alpha/\beta} = \frac{(\sum a_j \alpha_j)}{(\sum a_j \beta_j)}.$$  

(B13)

In particular for a two isotope medium,

$$I = \frac{A_1 a_1 + A_2 a_2}{A_1 \beta_1 + A_2 \beta_2}.$$  

(B14)

This is different from what one might have expected, namely, that the intensity is the sum of the individual intensities

$$\sum_j a_j \frac{\alpha_j}{\beta_j}.$$  

It is convenient to express the coefficients $\alpha_j$ and $\beta_j$ in terms of threshold excitation. This excitation is defined as the minimum excitation for which oscillation can occur. For a single isotope medium, this occurs for central tuning ($\nu = \omega$). For a two isotope medium with equal amounts of either isotope, the minimum excitation occurs for a tuning between the isotopes. The condition for threshold is that the linear net-gain coefficient

$$\alpha = 0,$$  

(B15)
that is,

$$\sqrt{\pi} p^2 (\hbar \kappa_e)^{-1} \bar{N}_T \sum (a_j/u_j) \exp[-(\omega_j - v_T)^2/(\kappa u_j)^2] = Q^{-1}, \quad (B16)$$

where $\bar{N}_T$ and $v_T$ are the threshold excitation and oscillation frequency, respectively. Solving for $\bar{N}_T$,

$$\bar{N}_T = \left\{ \sum (a_j/u_j) \exp[-(\omega_j - v_T)^2/(\kappa u_j)^2] \right\}^{-1} \hbar \kappa_e \kappa (p Q \sqrt{\pi})^{-1}. \quad (B17)$$

In general this must be solved numerically. It can be solved analytically in two cases of interest: the single and double isotope media.

For the first ($a_1 = 1, a_2 = 0, j > 1$),

$$\bar{N}_T = \frac{\hbar}{\kappa u} \exp\left\{ \frac{\Delta \omega}{2} \right\}. \quad (B18)$$

For the second with $a_1 = a_2 = \frac{1}{2}$,

$$v_T = \omega_1 + \frac{1}{2} \Delta_{\text{ISO}}, \quad (B19)$$

where $\Delta_{\text{ISO}}$ is the isotope shift. Here we assume for analytical simplicity that the Doppler widths for both isotopes are the same. For this, Eq. (B17) reduces to

$$\bar{N}_T = \exp \left\{ \frac{\Delta_{\text{ISO}}^2}{(2\kappa u)^2} \right\} \hbar \kappa_e \kappa \left( p Q \sqrt{\pi} \right)^{-1}, \quad (B20)$$

in which we have set $u_1 = u_2$ for simplicity. Hence for this case,

$$\alpha_j = \frac{1}{2}(v/Q) \{ \gamma \exp[\Delta_{\text{ISO}}^2/(2\kappa u)^2 - (\omega_j - v)^2] - 1 \} \quad (B21)$$

where the relative excitation

$$\gamma = \bar{N}/\bar{N}_T. \quad (B22)$$
Furthermore, the self-saturation coefficient

\[ \beta_j = \frac{1}{16} \left( \frac{\nu}{\mathcal{Q}} \right)^p \gamma_{ab} (\gamma_a^2 \gamma_b^2)^{-1} \exp \left[ \frac{\Delta_{\text{ISO}}^2}{(2K\nu)^2} - \frac{(\omega_j - \nu)^2}{(K\nu)^2} \right] \cdot \left[ 1 + L(\omega_j - \nu) \right]. \]

(B23)

In terms of Eqs. (B21) and (B23), the intensity, Eq. (B13), simplifies to

\[ I = E^2 = \frac{e_1 + e_2 - 2\gamma_\Delta^{-1} e_\Delta}{C[(1 + L_1) e_1 + (1 + L_2) e_2]} \]

(B24)

where for typographical simplicity we have introduced the notation for the exponentials

\[ e_j = \exp \left[ - \frac{(\omega_j - \nu)^2}{(K\nu)^2} \right] \]  

(B25a)

\[ e_\Delta = \exp \left[ - \frac{\Delta_{\text{ISO}}^2}{(K\nu)^2} \right] \]  

(B25b)

and for the Lorentzian

\[ L_j = \gamma^2 \left[ \gamma^2 + (\omega_j - \nu)^2 \right]^{-1} \]  

(B26)

One result of interest is the occurrence of a double tuning dip, one dip centered at \( \omega_1 \) and one at \( \omega_2 \). The condition for such a configuration is that the change of slope of the intensity tuning curve at a resonance point be positive, that is that

\[ \left[ \frac{\delta^2 I}{\delta \nu^2} \right]_{\nu=\omega_j} > 0 \]  

(B27)
For the single isotope medium, this gives the previously published result,

$$\gamma > 1 + 2 \gamma^2 (Ku)^{-2}.$$  

(B28)

For the two isotope case considered above, the first derivative (with \(L_2 = 0\)) is

$$\frac{\delta I}{\delta \nu} = -2(Ku)^{-2} \left[ \frac{(\nu - \omega)e_1 + (\nu - \omega_2)e_2}{(1 + L_1)e_1 + e_2} \right] + \frac{2I}{(Ku)^2} \left[ (KuL_1/\gamma)^2 (\nu - \omega_1)e_1 + (1 + L_1)(\nu - \omega_1)(\nu - \omega_2)e_2 \right] \frac{1}{[(1 + L_1)e_1 + e_2]}$$

(B29)

The second derivative evaluated at \(\nu = \omega_1\) is

$$\left[ \frac{\delta^2 I}{\delta \nu^2} \right]_{\nu = \omega_1} = -\frac{2}{(Ku)^2} \left[ \frac{1 + e_2}{2 + e_2} \right]$$

(B30)

$$+ 2 \left[ \frac{1 + e_2 - 2\gamma^{-1} e_2^\Delta}{(2 + e_2)(Ku)^2} \right] \left[ \frac{(Ku)}{\gamma^2} \right] + 2 + e_2] .$$

Setting this expression greater than 0 and multiplying through by

$$\frac{1}{4} \gamma (Ku)^2 (2 + e_2),$$

one finds that

$$\gamma (1 + e_2) [(Ku/\gamma)^2 + 1 + e_2] - 2e_2 \Delta [(Ku/\gamma)^2 + 1 + e_2] > 0.$$  

(B31)
Solving this for the relative excitation, one has the dip condition

$$\mathcal{N} \geq 2e_\Delta \left[ \left( \frac{Ku}{\gamma} \right)^2 + 2 + e_2 \right] / \left[ \left( \frac{Ku}{\gamma} \right)^2 + 1 + e_2 \right]$$  \hspace{1cm} (B32)

Since we are assuming the Doppler limit \((Ku >> \gamma)\) is valid, this reduces to

$$\mathcal{N} > 2 \exp \left[ -\frac{\Delta_{ISO}^2}{2Ku} \right] .$$  \hspace{1cm} (B33)

For the Ne red line (6328 Å) in the He-Ne gas laser, the isotope shift is 875 MHz between \(^{20}\)Ne and \(^{22}\)Ne which give \(e_\Delta = 0.82\). Hence the double dip should appear for a relative excitation greater than 1.64.

This is, in fact, the case as shown in Fig. 8.

One might ask how valid the perturbation theory used in writing Eq. (7) is for such a relative excitation. For this, the saturation parameter,

$$I_S = \frac{\gamma_a \gamma_b}{(\gamma_a + \gamma_b)} (\gamma_{ab} / \gamma)$$

$$= \left[ \frac{e_1 + e_2 - 1}{(1 + L_1)e_1 + (1 + L_2)e_2} \right]$$  \hspace{1cm} (B34)

$$< \frac{e_1 + e_2 - 1}{e_1 + e_2}$$  \hspace{1cm} (B35)

For midtuning \((\nu = \nu_T)\) of Eq. (B19), this is

$$I_S = 1 - \frac{1}{2} \exp \left[ \frac{\Delta_{ISO}^2}{2Ku} \right] ,$$  \hspace{1cm} (B36)

or about 0.38 for the He-Ne laser in the red. This value is large.
\( I_s \) should be small compared to unity) and leads to as much as a 20% error in the numerical results. Nevertheless, the intensity formula, Eq. (814) should be qualitatively correct at this excitation for any tuning and quantitatively valid for low intensities encountered away from \( \nu_T \). Specifically in the tuning regions of the dips, the intensity should be fairly accurate.
<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>ATOMIC WEIGHT</th>
<th>NATURAL ABUNDANCE</th>
<th>POSSIBLE CONCENTRATED PURITY*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Helium</td>
<td>3</td>
<td>$1.34 \times 10^{-4}$</td>
<td>99 %</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>100 %</td>
<td>100</td>
</tr>
<tr>
<td>Neon</td>
<td>20</td>
<td>90.5</td>
<td>99.95</td>
</tr>
<tr>
<td></td>
<td>21</td>
<td>0.28</td>
<td>&gt; 50</td>
</tr>
<tr>
<td></td>
<td>22</td>
<td>9.21</td>
<td>99.5</td>
</tr>
<tr>
<td>Argon</td>
<td>36</td>
<td>0.34</td>
<td>&gt; 99.5</td>
</tr>
<tr>
<td></td>
<td>38</td>
<td>0.06</td>
<td>&gt; 25</td>
</tr>
<tr>
<td></td>
<td>40</td>
<td>99.6</td>
<td>100</td>
</tr>
<tr>
<td>Xenon</td>
<td>124</td>
<td>0.09</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>126</td>
<td>0.08</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>128</td>
<td>1.91</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>129</td>
<td>26.24</td>
<td>60</td>
</tr>
<tr>
<td></td>
<td>130</td>
<td>4.05</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>131</td>
<td>21.24</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td>132</td>
<td>26.93</td>
<td>38</td>
</tr>
<tr>
<td></td>
<td>134</td>
<td>10.52</td>
<td>27</td>
</tr>
<tr>
<td></td>
<td>136</td>
<td>8.93</td>
<td>99</td>
</tr>
<tr>
<td>Cadmium</td>
<td>106</td>
<td>1.22</td>
<td></td>
</tr>
<tr>
<td>ELEMENT</td>
<td>ATOMIC WEIGHT</td>
<td>NATURAL ABUNDANCE</td>
<td></td>
</tr>
<tr>
<td>-----------</td>
<td>---------------</td>
<td>-------------------</td>
<td></td>
</tr>
<tr>
<td>Cadmium</td>
<td>108</td>
<td>0.89</td>
<td></td>
</tr>
<tr>
<td></td>
<td>110</td>
<td>12.45</td>
<td></td>
</tr>
<tr>
<td></td>
<td>111</td>
<td>12.86</td>
<td></td>
</tr>
<tr>
<td></td>
<td>112</td>
<td>23.79</td>
<td></td>
</tr>
<tr>
<td></td>
<td>113</td>
<td>12.34</td>
<td></td>
</tr>
<tr>
<td></td>
<td>114</td>
<td>28.81</td>
<td></td>
</tr>
<tr>
<td></td>
<td>116</td>
<td>7.66</td>
<td></td>
</tr>
<tr>
<td>Mercury</td>
<td>196</td>
<td>0.16</td>
<td></td>
</tr>
<tr>
<td></td>
<td>198</td>
<td>10.02</td>
<td></td>
</tr>
<tr>
<td></td>
<td>199</td>
<td>16.92</td>
<td></td>
</tr>
<tr>
<td></td>
<td>200</td>
<td>23.10</td>
<td></td>
</tr>
<tr>
<td></td>
<td>201</td>
<td>13.22</td>
<td></td>
</tr>
<tr>
<td></td>
<td>202</td>
<td>29.72</td>
<td></td>
</tr>
<tr>
<td></td>
<td>204</td>
<td>6.84</td>
<td></td>
</tr>
<tr>
<td>Nitrogen</td>
<td>14</td>
<td>99.64</td>
<td></td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>0.36</td>
<td></td>
</tr>
<tr>
<td>Carbon</td>
<td>12</td>
<td>98.89</td>
<td></td>
</tr>
<tr>
<td></td>
<td>13</td>
<td>1.108</td>
<td></td>
</tr>
<tr>
<td>Oxygen</td>
<td>16</td>
<td>99.76</td>
<td></td>
</tr>
<tr>
<td></td>
<td>17</td>
<td>0.04</td>
<td></td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>0.20</td>
<td></td>
</tr>
<tr>
<td>Cesium</td>
<td>133</td>
<td>100</td>
<td></td>
</tr>
</tbody>
</table>

*Percentage of Total Elemental Gas
APPENDIX D

ZEEMAN LASER PARAMETERS

For brevity, a full description of the Zeeman laser parameters was not given in Chapter IV. Below we have written the equations relating $\alpha_\pm$, $\beta_\pm$, and $\theta_\pm$ to the gas laser parameter under the influence of axial magnetic field $H$. The summation over $a'(b')$ is the summation over the magnetic sublevel of the $a(b)$ levels. We have suppressed the subscript $j$ on the $\omega$'s and the $g$'s, where the $g$'s are the Lande $g$-factors.

The amplitude determining equation for the Zeeman laser is

$$E_\pm = E_\pm (\alpha_\pm - \beta_\pm E_\pm^2 - \theta_\pm E_\pm) \quad (D1)$$

with

$$\alpha_\pm = \sum_j \frac{a_j}{u_j} \sum_{a',b',a',b',\pm} (p_{a'b'})^2 Z_i \left[ (\omega_{a'b'} + i \omega_{a'b'}) \right] - \frac{\nu}{2Q_\pm} \quad (D2)$$

$$\beta_\pm = \left( \frac{\Gamma}{\Gamma a b} \right) \sum_j \frac{a_j}{u_j} \sum_{a',b',a',b',\pm} (p_{a'b'})^4 \left[ 1 + \gamma_{ab} \frac{2L(\omega_{a'b'} \nu_{a'b'})}{L(\nu_{a'b'})} \right] \quad (D3)$$

$$\theta_\pm = \left( \frac{\Gamma}{\Gamma a b} \right) \sum_j \frac{a_j}{u_j} \sum_{a',b',a',b',\pm} (p_{a'b'})^2 (p_{a'b'} \pm \nu_{a'b'})^2 \left( \gamma_{ab} \right)^{-1} \quad (D4)$$

same with $\gamma_{a \leftrightarrow b}$, $\delta_a \rightarrow \delta_b$ and $p_{a',\pm 2'b',\pm 2'b'}$. 

61
\[ v = \frac{1}{2}(v_+ + v_-), \quad (D5) \]

\[ \Gamma = vN \left( \epsilon_o \gamma iK \right)^{-1}, \quad \Gamma' = \frac{1}{4\pi} \frac{d_1}{h} \Gamma \quad , \quad (D6) \]

\[ L_{\alpha}(\Delta\omega) = \left[ \gamma_{\alpha}^2 + (\Delta\omega)^2 \right]^{-1}, \quad \alpha = a, b \quad (D7) \]

\[ L(\Delta\omega) = \left[ \gamma_{ab}^2 + (\Delta\omega)^2 \right]^{-1}, \quad (D8) \]

\[ \omega_{a' b'} = \omega_0 + (\mu_B/\hbar)H(g_a a' - g_b b'), \quad (D9) \]

\[ \delta_{\alpha} = (\mu_B/\hbar)Hg_{\alpha} + \frac{1}{2}(v_- - v_+), \quad (D10) \]

where \( \omega_0 \) is the zero magnetic field frequency of the jth isotope at its line center and \( \mu_B \) is the Bohr magneton.
REFERENCES


