TRANSIENT REFRACTIVE INDEX IN A XeF LASER

by

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20. Abstract Continued

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Transient Refractive Index in a XeF Laser

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INTRODUCTION

Excimer lasers, which provide radiation in the visible and ultraviolet regions of the spectrum, are scalable to high energy. Because high energy per pulse is available, excimer lasers have many potential applications. Several applications require excellent beam quality.

As with many gas lasers, beam quality depends strongly on the refractive index field within the lasing gas. The processes of pumping and power extraction may generate spatial gradients of the index of refraction. Further, high energy lasers employ flowing gases as a means of eliminating heat; the volumetric release of heat is a consequence of laser inefficiency. Excimer lasers circulate the gases which constitute the laser medium.

Excimer lasers are inherently a pulsed device. When the energy is deposited by the e-beam, the static temperature of the gas may increase by 30%, driving strong acoustic waves and even shock waves with Mach number the order of 1.15. The impact of the waves on the index of refraction must be eliminated between pulses. Acoustic waves are phenomena which excimer lasers share with other pulsed electrical lasers.

For lasers operating with vibrational transitions in the infrared, e.g., CO$_2$ and DF, the index of refraction can be modeled adequately in terms of the Gladstone-Dale constant and
the variation in mass density.\textsuperscript{4} For an excimer laser, the refraactive index does not depend solely on mass density. As the discussion in Section I demonstrates, the index of refraction depends on both the mass fraction of chemical species and the populations for each of the chemical species in addition to mass density. Hence, to understand variations in index of refraction, the variations in chemical species and populations must be understood. Section I also discusses the methods to calculate the index of refraction.

Variations in the index of refraction cause distortion of phase at the output aperture of the laser.\textsuperscript{9,10} A nonuniform distribution of phase causes a severe degradation in beam quality. One approach to calculating the index of refraction and its impact on beam quality is to use the Ladenburg formula which is based on the Kramers' quantum-theoretical dispersion formula.\textsuperscript{11} The Ladenburg formula requires extensive knowledge of oscillator strengths and photoionization cross sections for molecules in the ground state as well as in excited states. Sources of data are discussed in Section II. Additional knowledge concerning the rise and fall of chemical species and the populations also is needed. A computer program yielding the information on species concentrations and populations is discussed in Section II and III.
Results of calculations of the index of refraction are presented in Section IV. Combining the information from Sections III and IV, an estimate of the influence of a spatial variation in input power density on beam quality is made in Section V.

I. INDEX OF REFRACTION

A. Calculation of index of refraction

The development in this section follows closely that of Fano and Cooper.\(^{12}\) The dielectric constant, \(\varepsilon\); complex index of refraction, \(\tilde{n}\); and electric susceptibility, \(\chi_e\), are related by

\[
\varepsilon = \tilde{n}^2 = (n + i\kappa)^2 = 1 + 4\pi\chi_e
\]

(1)

The real part of \(\tilde{n}\) is of interest and is given by

\[
n = \text{Re}[1 + 4\pi\chi_e]^{1/2} = \text{Re}[1 + 2\pi\chi_e]
\]

(2)

Fano and Cooper use the susceptibility in the form

\[
\chi_e = \frac{e^2 N}{m} \sum_s \frac{f_s}{\omega_s^2 - \omega^2 - i\gamma_s \omega}
\]

(3)

where \(e\) is the charge of an electron, \(m\) is mass of an electron, \(N\) is the number density of molecules, \(f_s\) is the absorption oscillator strength, \(\omega_s\) is the frequency, and \(\gamma_s\) is the damping. Further, Fano and Cooper point out that when photoionization processes occur, the form of Eq. (3) should be
\[
\sum_s \text{discrete} = \sum_s + \int_I^{\infty} \frac{h \omega_s}{2\pi} \\\ (4)
\]

In Eq. (4), \(I\) is the lowest ionization threshold. Fano and Cooper discuss the influence of autoionization.

A relation exists between the photoionization cross section and the spectral density of oscillator strengths which is \(df/dE\). \(E\) is the energy in appropriate units. Berkowitz\(^{13}\) gives the relation which is

\[
\sigma = \frac{\pi e^2 h}{mc} \frac{df}{dE} \\\ (5)
\]

Combining Eqs. (1) to (5) yields

\[
n - 1 = \frac{2\pi Ne^2}{m} \sum_s \frac{f_s (\omega_s^2 - \omega^2)}{(\omega_s^2 - \omega^2)^2 + \gamma_s^2 \omega^2} + \frac{cNh}{2\pi^2} \int_I^{\infty} \frac{\sigma(E_s^2 - E^2) dE_s}{(E_s^2 - E^2)^2 + \hbar^2 \gamma_s^2 E_s^2} \\\ (6)
\]

In Eq. (6), \(c\) is the velocity of light, and \(h\) is Planck's constant. The integral has been written in terms of energy, \(E = h\omega/2\pi = \hbar \omega\). Frequently cross section data are given in terms of energy instead of frequency; hence, the form of Eq. (6) is more convenient.

When \(\gamma_s\) is equated to zero, Eq. (6) reduces to Eq. (1) of Liggett and Levinger.\(^{14}\) This check provides confidence in the
algebraic manipulation involved in developing Eq. (6). Retaining $\gamma_s$ in Eq. (6) avoids mathematical difficulty when $\omega = \omega_s$ or $E = E_s$.

Equation (6) is correct for $N$ molecules in the ground state. If the gas is excited, the influence of excited species must be included. Excited species occur in excimer lasers. To develop an analogous equation to Eq. (6), consider the hypothetical atom shown in Fig. 1. For ease of writing, $\gamma_s$ is equated to zero. Define subscripts as follows: $i$, initial state; $f$, final state; $u$, upper level; and $l$, lower level.

The oscillator strength is always written as $f_{if}$, i.e., the first subscript is the initial state. Consequently, $f_{ul}$ is emission, and $f_{lu}$ is absorption. In accordance with convention and the need for compatible equations, $f_{ul}$ is negative, whereas $f_{lu}$ is positive. See Aller or Kemble. Not every author uses the same notation. For example, Mitchell and Zemansky use $f_{fi}$ for subscripts.

Having discussed notation, the seven terms implied by Fig. 1 are written explicitly

\[
2n - 1 = \frac{2\pi e^2}{m} \left[ \frac{f_{01}N_0}{\omega_{01}^2 - \omega^2} + \frac{f_{12}N_1}{\omega_{12}^2 - \omega^2} + \frac{f_{21}N_2}{\omega_{21}^2 - \omega^2} + \frac{f_{10}N_1}{\omega_{10}^2 - \omega^2} \right] \\
+ \frac{chN_0}{2\pi^2} \int_0^\infty \frac{\sigma_0 dE_0}{E_0^2 - E^2} + \frac{chN_1}{2\pi^2} \int_1^\infty \frac{\sigma_1 dE_1}{E_1^2 - E^2} + \text{etc.}
\]  

(7)
The subscript \( c \) identifies continuum; \( c \) occurs in the integrals.

The relation between emission and absorption oscillator strengths is given by Aller\(^{15} \) as

\[
f_{ul} = - \frac{g_u}{g_l} f_{lu}
\]  

(8)

where \( g_l \) is the degeneracy of the lower state. Using Eq. (8), Eq. (7) can be rewritten as

\[
\begin{align*}
n - 1 & = \frac{2\pi e^2}{m} \sum_{i=0} \sum_{k=i+1} f_{ik} \frac{N_i}{\omega_{ik}^2} \left[ 1 - \frac{N_k}{N_i} \frac{g_i}{g_k} \right] \\
& + \frac{ch}{2\pi} \sum_{s=0} N_s \int_{I_s} \frac{\sigma \, dE}{E_{sc}^2 - E^2}
\end{align*}
\]  

(9)

In Eq. (9), \( i \) is a lower state, and \( k \) is an upper state. Obviously, Eq. (9) involves only absorption oscillator strengths. The quantity \( I_s \) is illustrated in Fig. 1.

The similarity between the double sum in Eq. (9) and the gain equation is apparent. The similarity will now be discussed.

B. Index of refraction from gain

The influence of the lasing species upon the index of refraction can be determined from the gain. Define \( \mu = 2\omega c / c \) as the gain coefficient. From Eq. (9), the index of refraction due to the lasing species can be written as
\[ n - 1 = \frac{\pi e^2}{m \omega} \frac{(\omega_{ik} - \omega)f_{ik} N_i}{(\omega_{ik} - \omega)^2 + (\gamma_{ik}/2)^2} \left[ 1 - \frac{N_k g_i}{N_i g_k} \right] \]  

(10)

To obtain Eq. (10), the approximation

\[ \omega_{ik}^2 - \omega^2 \approx 2\omega(\omega_{ik} - \omega) \]  

(11)

was used. Equation (10) is for the lasing transition. The gain coefficient is

\[ \mu = \frac{\pi e^2}{m c g_k} \frac{\gamma_{ik} f_{ik} N_i}{(\omega_{ik} - \omega)^2 + (\gamma_{ik}/2)^2} \left[ 1 - \frac{N_k g_i}{N_i g_k} \right] \]  

(12)

Forming the ratio of \( n - 1 \) to \( \mu \) gives

\[ \frac{n - 1}{\mu} = \frac{(\omega_{ik} - \omega) c g_k}{\omega \gamma_{ik}} = \frac{(\omega_{ik} - \omega) \lambda g_k}{2\pi \gamma_{ik}} \]  

(13)

If the laser is tuned to the line center, i.e., \( \omega_{ik} = \omega \), the index of refraction due to the lasing molecule is zero. When \( \omega_{ik} \) does not equal \( \omega \), Eq. (13) can be used to estimate \( n - 1 \) given \( \mu \). Equation (13) applies to the case of natural line broadening. Other equations can be derived for other line broadening.

C. Sensitivity of index of refraction to specie concentration

Replace a typical term in the summation of Eq. (9) with the term shown in Eq. (10). Multiply the term in square brackets in Eq. (10) by \( N_i / g_i \). To find the sensitivity of \( n - 1 \) to species concentration \( N_i \), differentiate yielding
\[
\frac{\partial n}{\partial N_i} = \frac{\pi e^2}{m \omega} \frac{(\omega_{ik} - \omega) g_i f_{ik}}{(\omega_{ik} - \omega)^2 + (\gamma_{ik}/2)^2} + \frac{ch}{2\pi^2} \int_{I_S}^{\infty} \frac{g_s dE_{sc}}{E_{sc}^2 - E^2}
\]

Equation (14) is useful in determining the relative importance of a particular state or electronic configuration, i.e., the \(i\)-th state, of a specific chemical species.

D. Dependence of index of refraction on mass fraction

Based on the discussion to this point, one additional summation should occur in Eq. (9). As written, Eq. (9) is for a single chemical species. The index of refraction should be calculated by summing over the contributions of each chemical species. The third summation is accomplished by replacing \(N_i\) in Eq. (9) by \((N_i/N_\lambda)(N_\lambda/N)\). The quantity \(N_i/N_\lambda\), which is the fraction of species \(\lambda\) in state \(i\), reflects the influence of populations. \(N_\lambda\) is the number density of species \(\lambda\); and, hence, \(N_\lambda/N\) is the mole fraction \(X_\lambda\) of species \(\lambda\) in the gas mixture.

As written, Eq. (9) provides the contribution of species \(\lambda\) to the index of refraction; identify that contribution by the symbol \(T_\lambda\). The index of refraction for a gas mixture having \(X_\lambda\) mole fractions and populations \(N_i/N_\lambda\) is

\[
n - 1 = N \sum_\lambda X_\lambda T_\lambda
\]

The total number density is \(N\).
To ascertain the dependence of \( n \) on mass density, introduce the partial density \( \rho_\lambda \) which is \( N_\lambda m_\lambda \); \( m_\lambda \) is the mass of one molecule of species \( \lambda \). A mass fraction \( Y_\lambda = \rho_\lambda / \rho \) can be included in Eq. (15). When the changes are made, Eq. (15) becomes

\[
 n - 1 = \rho \sum_\lambda \frac{Y_\lambda T_\lambda}{m_\lambda}
\]  

Equation (16) shows that \( n - 1 \) is proportional to \( \rho \); however, Eq. (16) further demonstrates that \( n - 1 \) depends on mass fraction of each species. Since \( T_\lambda \) involves a summation over populations [refer to Eq. (9)], \( n - 1 \) also depends on populations.

Based on the preceding discussion, \( \Delta n/n \) depends not only on \( \Delta \rho/\rho \) but also on populations and mass fractions. A better choice for specifying the constraint on optical medium inhomogeneity is \( \Delta n/n \).

To estimate the maximum allowable \( \Delta n/n \), assume the maximum allowable phase shift at the output aperture is \( \lambda/20 \). The geometrical length of the optical path in the laser cavity is \( L \). The reference index of refraction is \( n_0 \). Under these conditions

\[
 \frac{\Delta n}{n_0} = \frac{n_0 - n}{n_0} = \frac{\lambda}{(20)(L)}
\]  

(17)
Inserting typical values for an excimer laser, the maximum allowable $\Delta n/n$ is $2 \times 10^{-8}$.

II. BACKGROUND DATA FOR CALCULATION OF INDEX OF REFRACTION

A. Typical species concentrations

During an interval extending from the early part of the pulse to the end, a steady state condition prevails for many of the chemical species. Lasing occurs during the steady state period. Table I gives the number density for 30 chemical species for an e-beam driven excimer laser with initial concentrations Ne:Xe:NF$_3$:0.992:0.006:0.002. The results shown in Table I were calculated using an excimer laser kinetics computer program developed by Johnson, Palumbo, and Hunter.  

The chemical species in Table I are listed in order of decreasing number density. Initial mass density of gases can be inferred from the number densities. The information shown is for a time of 250 nanoseconds from initiation of e-beam pulse. At 250 nanoseconds, steady state has been attained.

The dominant molecule is Ne with a number density about 200 times greater than Xe. The index of refraction for the gas mixture should be dominated by Ne. However, other chemical species present in trace amounts may contribute significantly due to large oscillator strength or due to resonance at laser
TABLE I. Number densities for thirty chemical species in a XeF excimer laser.

<table>
<thead>
<tr>
<th>Species</th>
<th>Ne</th>
<th>9.7E19</th>
<th>Xe</th>
<th>2.3E13</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe</td>
<td>5.8E17</td>
<td>Ne$_2^+$</td>
<td>2.2E13</td>
<td></td>
</tr>
<tr>
<td>NF$_3$</td>
<td>1.8E17</td>
<td>Ne$_3^+$</td>
<td>2.0E13</td>
<td></td>
</tr>
<tr>
<td>NF$_2$</td>
<td>9.0E15</td>
<td>XeF*</td>
<td>1.6E13</td>
<td></td>
</tr>
<tr>
<td>F</td>
<td>7.9E15</td>
<td>XeF</td>
<td>1.3E13</td>
<td></td>
</tr>
<tr>
<td>F$_2$</td>
<td>3.0E15</td>
<td>NF$_3^+$</td>
<td>9.9E12</td>
<td></td>
</tr>
<tr>
<td>hv</td>
<td>2.1E14</td>
<td>Xe*</td>
<td>9.7E12</td>
<td></td>
</tr>
<tr>
<td>F$^-$</td>
<td>9.7E13</td>
<td>Xe$_2^+$</td>
<td>8.4E12</td>
<td></td>
</tr>
<tr>
<td>F$^*$</td>
<td>9.3E13</td>
<td>NeF*</td>
<td>5.3E12</td>
<td></td>
</tr>
<tr>
<td>Ne$^*$</td>
<td>8.3E13</td>
<td>Ne$_2^*$</td>
<td>5.2E12</td>
<td></td>
</tr>
<tr>
<td>e$^-$</td>
<td>4.9E13</td>
<td>Xe$_2^*$</td>
<td>1.7E11</td>
<td></td>
</tr>
<tr>
<td>NF</td>
<td>3.6E13</td>
<td>N</td>
<td>9.3E10</td>
<td></td>
</tr>
<tr>
<td>Ne$^+$</td>
<td>3.1E13</td>
<td>Xe$_3^+$</td>
<td>7.8E10</td>
<td></td>
</tr>
<tr>
<td>NeXe$^+$</td>
<td>3.0E13</td>
<td>NF$_2^+$</td>
<td>1.7E09</td>
<td></td>
</tr>
<tr>
<td>F$_2^*$</td>
<td>2.4E13</td>
<td>N$_2$F$_2$</td>
<td>4.3E06</td>
<td></td>
</tr>
</tbody>
</table>

Note: hv is the photon density cm$^{-3}$ for wavelength of 352 nm. Units for number density are cm$^{-3}$.
wavelength. Initial efforts, which are reported in this paper, focus on Ne. Fortunately, considerable spectroscopic information exists for the ground and excited states of Neon.\textsuperscript{18-35} Since Ne is the lasing molecule in the ubiquitous HeNe laser, research in the early and mid 1960's was directed toward thorough understanding of Ne.

B. Oscillator strengths

The energy level diagram for Neon is shown in Fig. 2. The energy levels are shown for each electronic configuration. Figure 2 serves as a reference for subsequent tables which are arranged in the form of matrices.

Table II is a matrix which shows the references used to obtain oscillator strengths. For example, data for the transition 3s $\rightarrow$ 3p can be found in references 18, 20, 22, 26-29, 33, and 34. The reason that abundant data exist for 3s $\rightarrow$ 3p is that the HeNe lasers employ this transition. When an "X" occurs in Table II, the transition does not occur due to selection rules.

Not all data are given conveniently as oscillator strength. The data may be in the form of lifetime, Einstein coefficient $A_{\text{uv}}$, or matrix element. All data, except for 3s $\rightarrow$ 4p transition, were converted to oscillator strength for an array.
TABLE II. Matrix for references used to obtain oscillator strengths.

*References for \(3s^+ \rightarrow 3p\) transition are 16, 20, 22, 26, 27, 28, 29, 33, and 34.
Methods for changing line or multiplet f-numbers to array values are given in Aller and Condon and Shortley.

When a blank occurs in Table II, the oscillator strength was estimated by extending the continuum or by extrapolation of plots of f-number versus principal quantum number, n.

The 3s → 4p transition for Neon has lines in resonance with XeF laser radiation in 351 to 355 nm wavelength interval. Consequently, Eq. (9) must be evaluated line-by-line. Large values of n - 1 may occur due to lines in the 3s → 4p array. The array oscillator strength is obtained from Gruzdev and Loginov, while the relative line strengths can be determined from Table 4 of Semenov and Strugach.

C. Photoionization

According to Eq. (9), photoionization influences the magnitude of n - 1. Data are available for the photoionization cross section of Neon in the ground state. These data were used to evaluate the integral in Eq. (9).

For Neon in the 1s^22s^22p^53s (metastable) configuration, the photoionization cross section can be obtained from McCann and Flannery or from Hazi and Rescigno.

For other configurations of Neon, information about σ could not be found. One approach to supply the missing data is to calculate σ using procedures outlined in references 13 and 37-41. The calculations for σ were beyond the scope of the
present study. The contributions of $\sigma_s$ due to higher energy configurations of Neon have not been included.

III. XeF EXCIMER LASER

A. Typical conditions

Four aspects of excimer lasers are relevant to beam quality as follows: number density of various chemical species, populations, atomic and molecular data for oscillator strength and photoionization cross sections, and the flow of gases in the laser channel. Both number density and populations involve a detailed knowledge of kinetics.$^{17,42}$ For the calculations of this paper, the inhomogeneities due to waves are not included. Suppression is a separate and severe problem.$^{7,43-46}$

Table I provides information about species. Table III gives conditions for a typical XeF laser with flow.

B. Transient conditions

Power extraction occurs for a period the order of a microsecond. Variations in mass density, $\rho$, occur on a time scale, $\tau$, the order of

$$\tau = \frac{d}{a}$$

(18)

where $d$ is a characteristic length and $a$ is the acoustic velocity which is 300 to 400 m/sec. Since $\tau$ is several hundred microseconds, changes in $\rho$ do not occur during the pulse of e-beam and lasing.
TABLE III. Typical conditions in XeF flowing excimer laser

<table>
<thead>
<tr>
<th>Initial Gas Composition, Mole Fractions</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Ne</td>
<td>95.5</td>
</tr>
<tr>
<td>Xe</td>
<td>4.3</td>
</tr>
<tr>
<td>NF&lt;sub&gt;3&lt;/sub&gt;</td>
<td>0.2</td>
</tr>
</tbody>
</table>

| Initial Gas Temperature, °K                     | 300     |
| Initial Gas Pressure, Atmospheres               | 3-4     |
| Temperature Rise in Cavity due to Pulse, °K     | 100     |
| Pulse Length, Microseconds                      | 0.5-2   |
| Flow Mach Number                                | 0.5     |

Another factor which is related to flow is turbulence. Turbulence can affect adversely beam quality. The influence of turbulence is ignored. Consequently, the results of this paper are applicable to a laser which has perfect damping between pulses.

IV. CALCULATION OF INDEX OF REFRACTION

A. Plasma and free electrons

The contribution to the index of refraction of free electrons in the plasma is

\[ n^2 = 1 - \frac{\omega_p^2}{\omega^2} \]  

(19)

where \( \omega_p \) is the plasma frequency given by
\[ \frac{\omega^2}{p} = \frac{n_e^2}{\varepsilon_0 m_e} = \left[ 5.63 \times 10^4 \sqrt[n_e]{n_e} \right]^2 \] (20)

In the third term of Eq. (20), \( n_e \) had units of \( \text{cm}^{-3} \). As an example, from Table I, \( n_e = 4.9 \times 10^{13} \) giving \( \omega_p = 4 \times 10^{11} \). At a wavelength of 352 nm, the value of \( \omega \) is \( 5.4 \times 10^{15} \). Hence \( n - 1 \) due to electrons has a value of \( 3 \times 10^{-7} \). When \( n_e \) approaches \( 1 \times 10^{17} \), the optical properties of the free electrons become important.

B. Index of refraction of ground state Neon

Using Eq. (6) with \( \gamma_s = 0 \), the oscillator strengths from references 14, 21, 26, 32, and 33, the photoionization cross section from reference 14, and the wavelengths from reference 24, Etchechury \(^{49}\) calculated \( n - 1 \) to have a value of \( 1.0006705 \) at 352 nm. The value compares favorably with that given by Liggett and Levinger. \(^{14}\)

Of interest is the size of the photoionization integral relative to the sum of discrete terms in Eq. (6). Table IV summarizes the values of \( n - 1 \) due to photoionization. At 352 nm, the photoionization integral contributes about 9% of the overall value of \( n - 1 \).

C. Index of refraction of metastable Neon

To calculate \( n - 1 \) due solely to \( \text{Ne}(3s) \), Eq. (6) is used. The summation includes both emission, e.g., \( 3s \rightarrow 2p \),
TABLE IV. Contribution of photoionization to n - 1 for ground state Neon.

<table>
<thead>
<tr>
<th>Energy, Rydbergs</th>
<th>Wavelength, nm</th>
<th>(n - 1) x 10^5</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>914</td>
<td>5.835</td>
</tr>
<tr>
<td>0.2</td>
<td>457</td>
<td>5.864</td>
</tr>
<tr>
<td>0.3</td>
<td>305</td>
<td>5.915</td>
</tr>
<tr>
<td>0.4</td>
<td>228</td>
<td>5.988</td>
</tr>
<tr>
<td>0.5</td>
<td>183</td>
<td>6.085</td>
</tr>
<tr>
<td>0.6</td>
<td>152</td>
<td>6.211</td>
</tr>
<tr>
<td>0.7</td>
<td>130</td>
<td>6.370</td>
</tr>
<tr>
<td>0.8</td>
<td>114</td>
<td>6.568</td>
</tr>
<tr>
<td>0.9</td>
<td>102</td>
<td>6.818</td>
</tr>
<tr>
<td>1.0</td>
<td>91</td>
<td>7.134</td>
</tr>
</tbody>
</table>

and absorption, e.g., 3s → 3p, terms. Only one integral is necessary for photoionization which is for 3s → continuum. Equation (9) could be used if all N_i or N_s except N_{3s} were zero.

Etchechury\(^49\) calculated n - 1 due only to Ne(3s). Table V shows the data for calculating (n - 1) due to discrete transitions. For transitions 3s → 2p, 3p, 5p, the individual line transitions are replaced by an equivalent array. However, for the transition 3s → 4p, each line must be accounted for.
Some lines in the $3s \rightarrow 4p$ array are resonant with the XeF photons.

**TABLE V. Information for discrete transitions**

<table>
<thead>
<tr>
<th>Transition</th>
<th>Array Oscillator Strength, $f_A$</th>
<th>Energy, Rydbergs</th>
<th>Number of Terms</th>
</tr>
</thead>
<tbody>
<tr>
<td>$3s \rightarrow 2p$</td>
<td>-0.0482</td>
<td>1.230</td>
<td>One using $f_A$</td>
</tr>
<tr>
<td>$3s \rightarrow 3p$</td>
<td>1.080</td>
<td>0.146</td>
<td>One using $f_A$</td>
</tr>
<tr>
<td>$3s \rightarrow 4p$</td>
<td>0.0106</td>
<td>0.259</td>
<td>Thirty using $f_L$</td>
</tr>
<tr>
<td>$3s \rightarrow 5p$</td>
<td>0.00129</td>
<td>0.302</td>
<td>One using $f_A$</td>
</tr>
<tr>
<td>$3s \rightarrow np$</td>
<td>-</td>
<td>-</td>
<td>Included in continuum integral for $n &gt; 5$</td>
</tr>
</tbody>
</table>

Note: $f_A$ means oscillator strength for an array; $f_L$ means oscillator strength for a line.

For the transitions $3s \rightarrow np(n > 5)$, the effect is included in the integral. An equivalent cross section is determined from the spectral density of oscillator strength. References 12 and 40 discuss the method.

Table V gives the array oscillator strength, $f_A$, for $3s \rightarrow 4p$ resonant transition. To obtain the line oscillator strength, $f_L$, for each of the 30 lines, the table of relative line strengths from reference 31 can be used. The results are shown in Table VI. $K$ is an index for the upper levels (4p) while $M$ is an index for the lower levels (3s).
TABLE VI. Relative line strengths (upper number) and line oscillator strengths (lower number) for 3s → 4p transition.

<table>
<thead>
<tr>
<th>K</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
<th>9</th>
<th>10</th>
<th>M</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3p₁₀</td>
<td>3p₉</td>
<td>3p₈</td>
<td>3p₇</td>
<td>3p₆</td>
<td>3p₅</td>
<td>3p₄</td>
<td>3p₃</td>
<td>3p₂</td>
<td>3p₁</td>
<td></td>
</tr>
<tr>
<td>1s₂</td>
<td>.056</td>
<td>0</td>
<td>0</td>
<td>.139</td>
<td>.833</td>
<td>.667</td>
<td>.833</td>
<td>.111</td>
<td>.333</td>
<td>.333</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>.0015</td>
<td>J</td>
<td>.0036</td>
<td>.0216</td>
<td>.0173</td>
<td>.0216</td>
<td>.0029</td>
<td>.0066</td>
<td>.0066</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1s₃</td>
<td>.056</td>
<td>0</td>
<td>0</td>
<td>.278</td>
<td>0</td>
<td>.333</td>
<td>0</td>
<td>0</td>
<td>.333</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>.0044</td>
<td>J</td>
<td>J</td>
<td>.0216</td>
<td>J</td>
<td>.0259</td>
<td>J</td>
<td>J</td>
<td>.0259</td>
<td>J</td>
<td></td>
</tr>
<tr>
<td>1s₄</td>
<td>.250</td>
<td>0</td>
<td>1.375</td>
<td>.625</td>
<td>.083</td>
<td>0</td>
<td>.208</td>
<td>.222</td>
<td>.125</td>
<td>.111</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>.0065</td>
<td>J</td>
<td>.0356</td>
<td>.0162</td>
<td>.0021</td>
<td>.0054</td>
<td>.0057</td>
<td>.0032</td>
<td>.0029</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1s₅</td>
<td>.694</td>
<td>2.333</td>
<td>.292</td>
<td>.097</td>
<td>.750</td>
<td>0</td>
<td>.625</td>
<td>0</td>
<td>.208</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>.0108</td>
<td>.0361</td>
<td>.0046</td>
<td>.0016</td>
<td>.0116</td>
<td>.0097</td>
<td>J</td>
<td>.0032</td>
<td>J</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Note: Paschen notation is used to identify levels. The four lines which are resonant with XeF radiation are underlined.
Using the information from Table VI, a graph of \((n - 1)\) due only to \(3s + 4p\) transition was calculated. Figure 3 shows four resonances in the XeF lasing region. Also shown in Fig. 3 are the wavelengths for the line centers and the specific transitions. For the specific transitions, Paschen notation is used.

At a wavelength of 352 nm and a Ne(3s) density of \(2.68 \times 10^{13} \text{ cm}^{-3}\), Etchechury calculated \((n - 1)\) due solely to Ne(3s). The calculated result is \(n - 1 = -7.79E-10\). At the same wavelength and number density for Ne(3s), the contribution of photoionization to \(n - 1\) is \(3.4E-12\). The percentage contribution of photoionization is 0.4% at 352 nm.

Later, the ratio \((n - 1)/N\) will be used; the ratio is the index of refraction minus unity per molecule. In terms of this ratio, the results for Ne(3s) at 352 nm are as follows: overall \(n - 1\) is \(-2.907E-23\) and due to photoionization, \(+1.27E-25\).

Table VII summarizes the contributions to \((n - 1)\) of the various transitions. The table has been calculated for a number density equal to Loschmidt's number, and the values of the wavelength have been selected for XeF.
<table>
<thead>
<tr>
<th>Number of Terms</th>
<th>Transition</th>
<th>(n - 1) x 10^6</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>351.09 nm</td>
</tr>
<tr>
<td>One array</td>
<td>3s → 2p</td>
<td>-3.3</td>
</tr>
<tr>
<td>One array</td>
<td>3s → 3p</td>
<td>-2370.9</td>
</tr>
<tr>
<td>Thirty lines</td>
<td>3s → 4p</td>
<td>2230</td>
</tr>
<tr>
<td>One array</td>
<td>3s → 5p</td>
<td>5.3</td>
</tr>
<tr>
<td>One integral</td>
<td>3s + continuum</td>
<td>3.8</td>
</tr>
<tr>
<td><strong>Total for (n - 1)</strong></td>
<td></td>
<td>-135.1</td>
</tr>
</tbody>
</table>
D. Index of refraction of Neon in higher energy levels

Using Eq. (6), n – 1 was calculated. Information concerning the photoionization cross section for energy levels above 3s could not be found. The trend in σ between ground state Neon and metastable Neon is a large reduction; σ is a factor of ten less for Ne(3s) than for Ne(2p⁶). If the trend in the magnitude of σ continues, the contribution of the continuum should be small.

For Ne(3p) and higher energy levels, the energy of XeF photons occurs above the ionization threshold. Hence, the integration of the continuum has both a positive and a negative contribution, i.e., the denominator of the integrand in Eq. (6) switches from negative to positive as energy increases.

In Eq. (6), the damping term was equated to zero. All transitions between excited configurations can be represented in a matrix as shown in Table VIII. Each element in the matrix is the contribution to n – 1 per molecule for the transition represented by the matrix element. As an example, consider 3d → 4p transition which is absorption; assume N₃d is 3E11 cm⁻³. The contribution of 3d → 4p is

\[
(n - 1) = \left(\frac{n - 1}{N}\right)N \times 10^{-27} = (-7695)(3 \times 10^{11})(10^{-27}) = -2.3 \times 10^{-12}
\]
TABLE VIII. Table of \((n-1)/N\) for each array for excited Neon.

<table>
<thead>
<tr>
<th>INITIAL CONFIGURATION</th>
<th>2p</th>
<th>3m</th>
<th>3p</th>
<th>4p</th>
<th>5d</th>
<th>5p</th>
<th>5d</th>
<th>6s</th>
</tr>
</thead>
<tbody>
<tr>
<td>2p</td>
<td>E</td>
<td>-204</td>
<td>E</td>
<td>-26.7</td>
<td>E</td>
<td>-4.3</td>
<td>E</td>
<td>-8.8</td>
</tr>
<tr>
<td>3m</td>
<td>A</td>
<td>409</td>
<td>E</td>
<td>29,160</td>
<td>E</td>
<td>K*</td>
<td>E</td>
<td>3,890</td>
</tr>
<tr>
<td>3p</td>
<td>A</td>
<td>-87,674</td>
<td>E</td>
<td>33,316</td>
<td>E</td>
<td>39,029</td>
<td>E</td>
<td>12</td>
</tr>
<tr>
<td>4p</td>
<td>A</td>
<td>53.3</td>
<td>A</td>
<td>30,001</td>
<td>E</td>
<td>55,186</td>
<td>E</td>
<td>38,039</td>
</tr>
<tr>
<td>5d</td>
<td>A</td>
<td>43.8</td>
<td>A</td>
<td>-65,049</td>
<td>A</td>
<td>-7,695</td>
<td>A</td>
<td>-90,034</td>
</tr>
<tr>
<td>5p</td>
<td>A</td>
<td>17.6</td>
<td>A</td>
<td>-1,297</td>
<td>A</td>
<td>-18,395</td>
<td>A</td>
<td>-13,985</td>
</tr>
<tr>
<td>6s</td>
<td>A</td>
<td>22.9</td>
<td>A</td>
<td>-7,958</td>
<td>A</td>
<td>-63,398</td>
<td>A</td>
<td>-13,985</td>
</tr>
<tr>
<td>5d</td>
<td>A</td>
<td>12.4</td>
<td>A</td>
<td>-2,906</td>
<td>A</td>
<td>-8,418</td>
<td>A</td>
<td>-13,985</td>
</tr>
<tr>
<td>6s</td>
<td>A</td>
<td>12.4</td>
<td>A</td>
<td>-2,906</td>
<td>A</td>
<td>-8,418</td>
<td>A</td>
<td>-13,985</td>
</tr>
</tbody>
</table>

* resonance - requires special attention
Transitions above the diagonal in Table VIII are emission as denoted by the letter E. Transitions below the diagonal in Table VIII are absorption as denoted by the letter A. When the columns are summed, the contributions to \( n - 1 \) per molecule for all transitions are included. For the example above, the contribution of Ne(3d) is

\[
(n - 1) = (31324)(3 \times 10^{-11})(10^{-27}) = 9.4 \times 10^{-12}
\]

In a similar manner, all transitions can be calculated and summed if the populations are known.

Table VIII was calculated for a wavelength of 352 nm. The results are not sensitive to wavelength except for the two resonances indicated by \( R^* \). The two resonant transitions are between 3s and 4p either in emission or absorption.

V. CALCULATION OF NUMBER DENSITIES AND POPULATIONS

Cole \cite{42} modified the computer program of Johnson, Palumbo, and Hunter \cite{17} to yield populations of excited Neon. The program prior to Cole's modification calculated Ne*, which is the population of excited Neon irrespective of energy level. Cole divided Ne* into two groups: metastable Neon, i.e., Ne(3s), and one number representing all higher levels.

As an example, the following reaction generates Ne*,

\[
\text{XeNe}^+ + e^- \rightarrow \text{Ne}^* + \text{Xe}
\] (21)
with a rate coefficient 6E-08. The reaction given by Eq. (21) was replaced by two reactions

\[ \text{XeNe}^+ + e^- \rightarrow \text{Ne}(3s) + \text{Xe} \]  
(22)

and

\[ \text{XeNe}^+ + e^- \rightarrow \text{Ne}(Z) + \text{Xe} \]  
(23)

In Eq. (23) Ne(Z) represents Ne in any energy level above 3s. The reaction rate coefficients for Eqs. (22) and (23) were 3.60E-08 and 2.40E-08 respectively.

The volumes by McDaniels, et al., were helpful in estimating the branching ratios; however, many of the choices were arbitrary. The modified computer code can be altered to reflect improvements in rate constants.

To distribute the Ne(Z) populations among Ne(3p), Ne(4p), Ne(4s), etc., a Boltzmann distribution was assumed. The temperature, which varied as time evolved, was determined from the ratio of populations Ne(3s)/Ne(2p^6). Knowing the temperature, the energy level diagram, and the degeneracies of the levels, the Ne(Z) could be distributed.

When the populations were determined, the results shown in Table VIII were used to calculate \((n - 1)\) for each electronic configuration. Finally, all contributions from each electronic configuration were summed to obtain \((n - 1)\).
The calculations were performed twice, once with a current density of 10A/cm² and again with 11A/cm² for the e-beam. The e-beam voltage was constant. The energy deposited in the gas varied by 10%.

A quantity \((n - 1)/(n - 1)_0\) was determined as a function of time. The quantity \((n - 1)_0\) is the value at the start of the e-beam pulse. For a wavelength away from any of the \(3s \rightarrow 4p\) resonances, \((n - 1)/(n - 1)_0\) changed from unity at zero time to 0.999825 for low power and 0.999810 for high power. The fractional change is 1.5E-05. In terms of phase shift, \(\delta\), a laser cavity with length, \(L\), less than 35 meters will have \(\delta/\lambda\) less than 0.1.

For the case of a wavelength near a resonance in the \(3s \rightarrow 4p\) transition, the consequences are different. A length of laser cavity of only 0.1116 meters yields \(\delta/\lambda\) equal to 0.1. Recall the power variation was 10%.

Details of the calculations are given in Cole's thesis.42

VI. CONCLUSIONS

Neon, which is a dominant molecule in a XeF excimer laser, has transitions in the \(3s \rightarrow 4p\) array which are resonant with laser photons. Operation of the XeF laser near one of the resonances could cause extreme sensitivity to small variations in power density.
ACKNOWLEDGMENT

The authors wish to thank the Defense Advanced Research Projects Agency for funding this study. LCOL Rett Benedict sponsored the research. Dr. Robert Behringer, ONR Office, Pasadena, monitored the research effort, and Dr. William Condell of Physics Branch, ONR, assisted with expediting the contract. The authors want to thank both Dr. Condell and Dr. Behringer for their help.

Dr. Louis Palumbo provided the NRL computer program for laser kinetics. Further, he helped to "debug" the computer program for the NPS computer. His outstanding cooperation and assistance are gratefully acknowledged.
FIG. 1. Energy level diagram for a hypothetical atom used to derive equation for $n - 1$. 
FIG. 3. Plot of $n - 1$ as a function of wavelength. The quantity plotted is contribution of $3s \rightarrow 4p$ transition in Neon.


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31. R. I. Semenov and B. A. Strugach, "Determination of Coupling coefficients for the np\textsuperscript{5}n's and np\textsuperscript{5}n'p configurations from experimental data," Optics and Spectroscopy 24, 258-262 (1968).

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34. R. S. F. Chang and D. W. Setser, "Radiative lifetimes and collisional deactivation rate constants of excited Ne(2p\textsuperscript{5}3p) states," Department of Chemistry, Kansas State University, Manhattan, KS, 66506.


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