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TRANSIENT REFRACTIVE INDEX IN A XeF LASER

by

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September 1980

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Transient refractive index was calculated for an e-beam pumped XeF laser having initial concentrations Ne:Xe:NF_3::95.5:4.3:0.2. The calculation used Ne in excited states including the transition array 3s * 4p and all other constituents in ground state. Lines in 3s * 4p array may be resonant with XeF radiation. To obtain the transient populations for Ne, a modification was made to a Naval Research Laboratory computer code which typically accounts for 180-200 reactions involving 50-60 different species. The modified program included populations in 12 electronic configurations of Ne. Electron beam

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20. Abstract Continued

current was changed 10% at constant voltage to simulate the influence of a hibachi shadow. For a nonresonant laser wavelength, an optical pathlength in the laser, L, less than 35 meters will have distortion δ/λ less than 0.1; however, for a laser wavelength in resonance with Neon, L of only 0.11 meter will cause $\delta/\lambda = 0.1$. The difference in optical pathlength is δ.

delta

lamda

delta

delta/lambda

TABLE OF CONTENTS

TABLE OF CONTENTS Continued

LIST OF FIGURES

FIGURE

LIST OF TABLES

TABLE

Transient Refractive Index in a XeF Laser

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Transient refractive index was calculated for an e-beam pumped XeF laser having initial concentrations Ne:Xe:NF_3:: 95.5:4.3:0.2. The calculation used Ne in excited states including the transition array 3s → 4p and all other constituents in ground state. Lines in 3s → 4p array may be resonant with XeF radiation. To obtain the transient populations for Ne, a modification was made to a Naval Research Laboratory computer code which typically accounts for 180-200 reactions involving 50-60 different species. The modified program included populations in 12 electronic configurations of Ne. Electron beam current was changed 10% at constant voltage to simulate the influence of a hibachi shadow. For a nonresonant laser wavelength, an optical pathlength in the laser, L, less than 35 meters will have distortion δ/λ less than 0.1; however, for a laser wavelength in resonance with Neon, L of only 0.11 meter will cause $\delta/\lambda = 0.1$. The difference in optical pathlength is δ.

INTRODUCTION

Excimer lasers, which provide radiation in the visible and ultraviolet regions of the spectrum, are scalable to high energy.[1-3] 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.[4] The processes of pumping and power extraction may generate spatial gradients of the index of refraction.[5] Further, high energy lasers employ flowing gases as a means of eliminating heat;[6] 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.[7,8]

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.[4] For an excimer laser, the refractive 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.[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.[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, ε; complex index of refraction, $\tilde{n}$; and electric susceptibility, χ_e, are related by

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

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

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

Fano and Cooper use the susceptibility in the form

$$\chi_e = \frac{e^2N}{m} \sum_s \frac{f_s}{\omega_s^2 - \omega^2 - i\gamma_s\omega} \tag{3}$$

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, ω_s is the frequency, and γ_s is the damping. Further, Fano and Cooper point out that when photoionization processes occur, the form of Eq. (3) should be

$$\sum_{s} = \sum_{s}^{\text{discrete}} + \int_{I}^{\infty} \frac{h\,d\omega_s}{2\pi} \tag{4}$$

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

$$\sigma = \frac{\pi e^2 h}{mc} \frac{df}{dE} \tag{5}$$

Combining Eqs. (1) to (5) yields

$$\begin{aligned} n - 1 = {} & \frac{2\pi N e^2}{m} \sum_{s} \frac{f_s(\omega_s^2 - \omega^2)}{(\omega_s^2 - \omega^2)^2 + \gamma_s^2 \omega^2} \\ & + \frac{cNh}{2\pi^2} \int_{I}^{\infty} \frac{\sigma(E_s^2 - E^2)\,dE_s}{(E_s^2 - E^2)^2 + \hbar^2 \gamma_s^2 E^2} \end{aligned} \tag{6}$$

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 γ_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 γ_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, γ_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, $f_{u\ell}$ is emission, and $f_{\ell u}$ is absorption. In accordance with convention and the need for compatible equations, $f_{u\ell}$ is negative, whereas $f_{\ell u}$ is positive. See Aller[15] or Kemble.[16] Not every author uses the same notation. For example, Mitchell and Zemansky[11] use f_{fi} for subscripts.

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

$$n - 1 = \frac{2\pi e^2}{m}\left[\frac{f_{01}N_0}{\omega_{01}^2 - \omega^2} + \frac{f_{12}N_1}{\omega_{12}^2 - \omega^2} + \frac{f_{21}N_2}{\omega_{21}^2 - \omega^2} + \frac{f_{10}N_1}{\omega_{10}^2 - \omega^2}\right]$$

$$+ \frac{chN_0}{2\pi^2}\int_{I_0}^{\infty}\frac{\sigma_0 dE_{0c}}{E_{0c}^2 - E^2} + \frac{chN_1}{2\pi^2}\int_{I_1}^{\infty}\frac{\sigma_1 dE_{1c}}{E_{1c}^2 - E^2} + \text{etc.} \qquad (7)$$

The subscript c identifies continuum; c occurs in the integrals. The relation between emission and absorption oscillator strengths is given by Aller[15] as

$$f_{u\ell} = -\frac{g_\ell}{g_u} f_{\ell u} \tag{8}$$

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

$$n - 1 = \frac{2\pi e^2}{m} \sum_{i=0} \sum_{k=i+1} \frac{f_{ik} N_i}{\omega_{ik}^2 - \omega^2} \left[1 - \frac{N_k}{N_i} \frac{g_i}{g_k}\right]$$

$$+ \frac{ch}{2\pi^2} \sum_{s=0} N_s \int_{I_s}^{\infty} \frac{\sigma_s dE_{sc}}{E_{sc}^2 - E^2} \tag{9}$$

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 $\mu = 2\omega\kappa/c$ as the gain coefficient. From Eq. (9), the index of refraction due to the lasing species can be written as

$$n - 1 = \frac{\pi e^2}{m\omega} \frac{(\omega_{ik} - \omega) f_{ik} N_i}{(\omega_{ik} - \omega)^2 + (\gamma_{ik}/2)^2} \left[1 - \frac{N_k}{N_i} \frac{g_i}{g_k} \right] \quad (10)$$

To obtain Eq. (10), the approximation

$$\omega_{ik}^2 - \omega^2 \cong 2\omega(\omega_{ik} - \omega) \quad (11)$$

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

$$\mu = \frac{\pi e^2}{mcg_k} \frac{\gamma_{ik} f_{ik} N_i}{(\omega_{ik} - \omega)^2 + (\gamma_{ik}/2)^2} \left[1 - \frac{N_k g_i}{N_i g_k} \right] \quad (12)$$

Forming the ratio of n - 1 to μ gives

$$\frac{n - 1}{\mu} = \frac{(\omega_{ik} - \omega) c g_k}{\omega \gamma_{ik}} = \frac{(\omega_{ik} - \omega) \lambda g_k}{2\pi \gamma_{ik}} \quad (13)$$

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 ω_{ik} does not equal ω, Eq. (13) can be used to estimate n - 1 given μ. 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

$$\frac{\partial n}{\partial N_i} = \frac{\pi e^2}{m\omega} \frac{(\omega_{ik} - \omega) g_i f_{ik}}{(\omega_{ik} - \omega)^2 + (\gamma_{ik}/2)^2} + \frac{ch}{2\pi^2} \int_{I_s}^{\infty} \frac{\sigma_s dE_{sc}}{E_{sc}^2 - E^2} \tag{14}$$

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 $(N_i/N_\ell)(N_\ell/N)$. The quantity N_i/N_ℓ, which is the fraction of species ℓ in state i, reflects the influence of populations. N_ℓ is the number density of species ℓ; and, hence, N_ℓ/N is the mole fraction X_ℓ of species ℓ in the gas mixture.

As written, Eq. (9) provides the contribution of species ℓ to the index of refraction; identify that contribution by the symbol T_ℓ. The index of refraction for a gas mixture having X_ℓ mole fractions and populations N_i/N_ℓ is

$$n - 1 = N \sum_{\ell} X_\ell T_\ell \tag{15}$$

The total number density is N.

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

$$n - 1 = \rho \sum_\ell \frac{Y_\ell T_\ell}{m_\ell} \tag{16}$$

Equation (16) shows that n - 1 is proportional to ρ; however, Eq. (16) further demonstrates that n - 1 depends on mass fraction of each species. Since T_ℓ 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

$$\frac{\Delta n}{n_0} = \frac{n_0 - n}{n_0} = \frac{\lambda}{(20)(L)} \tag{17}$$

Inserting typical values for an excimer laser, the maximum allowable $\Delta n/n$ is 2×10^{-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.[17]

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.

Ne	9.7E19	Xe^+	2.3E13
Xe	5.8E17	Ne_2^+	2.2E13
NF_3	1.8E17	Ne_3^+	2.0E13
NF_2	9.0E15	XeF^*	1.6E13
F	7.9E15	XeF	1.3E13
F_2	3.0E15	NF_3^+	9.9E12
hν	2.1E14	Xe^*	9.7E12
F^-	9.7E13	Xe_2^+	8.4E12
F^*	9.3E13	NeF^*	5.3E12
Ne^*	8.3E13	Ne_2^*	5.2E12
e^-	4.9E13	Xe_2^*	1.7E11
NF	3.6E13	N	9.3E10
Ne^+	3.1E13	Xe_3^+	7.8E10
$NeXe^+$	3.0E13	NF^+	1.7E09
F_2^*	2.4E13	N_2F_2	4.3E06

Note: hν 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.[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 → 3p can be found in references 18, 20, 22, 26-29, 33, and 34. The reason that abundant data exist for 3s → 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 $A_{u\ell}$, or matrix element. All data, except for 3s → 4p transition, were converted to oscillator strength for an array.

2p	3s	3p	4s	3d	4p	5s	4d	4f	5p	5d	6s
☒ 2p	14, 21, 26, 32, 33	☒	14, 20, 26, 32	14, 32	☒	14, 32	14, 20, 32		☒	32	14, 32
	☒ 3s	*	☒	☒	23, 30, 33	☒	☒	☒	23	☒	☒
		☒ 3p	22, 26, 29, 31	23, 33	☒	22, 23, 26	23	☒	☒	23	22, 23, 26
			☒ 4s	☒	26, 30, 33	☒	☒	☒		☒	☒
				☒ 3d	26	☒	☒	32		☒	☒
					☒ 4p	30		☒	☒		30
						☒ 5s	☒	☒		☒	☒
							☒ 4d		26	☒	☒
								☒ 4f	☒		☒
									☒ 5p		
										☒ 5d	☒
											☒ 6s

TABLE II. Matrix for references used to obtain oscillator strengths.

*References for 3s → 3p transition are 18, 20, 22, 26, 27, 28, 29, 33, and 34.

Methods for changing line or multiplet f-numbers to array values are given in Aller[15] and Condon and Shortley.[36]

When a blank occurs in Table II, the oscillator strength was estimated by extending the continuum[12,14] 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,[32] while the relative line strengths can be determined from Table 4 of Semenov and Strugach.[31]

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.[14] These data were used to evaluate the integral in Eq. (9).

For Neon in the $1s^2 2s^2 2p^5 3s$ (metastable) configuration, the photoionization cross section can be obtained from McCann and Flannery[37] or from Hazi and Rescigno.[38]

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 σ_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, ρ, occur on a time scale, τ, the order of

$$\tau = \frac{d}{a} \tag{18}$$

where d is a characteristic length and a is the acoustic velocity which is 300 to 400 m/sec. Since τ is several hundred microseconds, changes in ρ do not occur during the pulse of e-beam and lasing.

TABLE III. Typical conditions in XeF flowing excimer laser

Initial Gas Composition, Mole Fractions	
Ne	95.5
Xe	4.3
NF_3	0.2
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.[47] 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[48] is

$$n^2 = 1 - \frac{\omega_p^2}{\omega^2} \tag{19}$$

where ω_p is the plasma frequency given by

$$\omega_p^2 = \frac{n_e e^2}{\varepsilon_0 m_e} = [5.63 \times 10^4 \sqrt{n_e}]^2 \qquad (20)$$

In the third term of Eq. (20), n_e had units of cm^{-3}. As an example, from Table I, n_e = 4.9E13 giving ω_p = 4E11. At a wavelength of 352 nm, the value of ω is 5.4E15. Hence n - 1 due to electrons has a value of 3E-07. When n_e approaches 1E17, 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 Ne(3s), Eq. (6) is used. The summation includes both emission, e.g., 3s → 2p,

TABLE IV. Contribution of photoionization to n - 1 for ground state Neon.

Energy, Rydbergs	Wavelength, nm	$(n - 1) \times 10^5$
0.1	914	5.835
0.2	457	5.864
0.3	305	5.915
0.4	228	5.988
0.5	183	6.085
0.6	152	6.211
0.7	130	6.370
0.8	114	6.568
0.9	102	6.818
1.0	91	7.134

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 → 4p array are resonant with the XeF photons.

TABLE V. Information for discrete transitions

Transition	Array Oscillator Strength, f_A	Energy, Rydbergs	Number of Terms
3s → 2p	-0.0482	1.230	One using f_A
3s → 3p	1.080	0.146	One using f_A
3s → 4p	0.0106	0.259	Thirty using f_L
3s → 5p	0.00129	0.302	One using f_A
3s → np	-	-	Included in continuum integral for $n > 5$

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

For the transitions 3s → 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 → 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 $3s \rightarrow 4p$ transition.

K	1	2	3	4	5	6	7	8	9	10	
	$3p_{10}$	$3p_9$	$3p_8$	$3p_7$	$3p_6$	$3p_5$	$3p_4$	$3p_3$	$3p_2$	$3p_1$	M
$1s_2$	.056	0	0	.139	.833	.667	.833	.111	.333	.333	4
	.0015	J		.0036	.0216	.0173	.0216	.0029	.0066	.0066	
$1s_3$	.056	0	0	.278	0	.333	0	0	.333	0	3
	.0044	J	J	.0216	J	.0259	J	J	.0259	J	
$1s_4$	.250	0	1.375	.625	.083	0	.208	.222	.125	.111	2
	.0065	J	.0356	.0162	.0021		.0054	.0057	.0032	.0029	
$1s_5$	.694	2.333	.292	.097	.750	0	.625	0	.208	0	1
	.0108	.0361	.0046	.0016	.0116		.0097	J	.0032	J	

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 2.68×10^{13} cm^{-3}, Etchechury[49] 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 VII. Summary of contributions to (n - 1) by various transitions for metastable Neon.

Number of Terms	Transition	$(n - 1) \times 10^6$					
		351.09 nm	351.15 nm	351.26 nm	353.18 nm	353.20 nm	353.23 nm
One array	3s → 2p	- 3.3	- 3.3	- 3.3	- 3.3	- 3.3	- 3.3
One array	3s → 3p	- 2370.9	- 2372.1	- 2374.3	- 2412.7	- 2430.7	- 2413.7
Thirty lines	3s → 4p	2230	7287	1135	- 428.6	- 422.2	- 412.7
One array	3s → 5p	5.3	5.3	5.3	5.1	5.1	5.1
One integral	3s → continuum	3.8	3.8	3.8	3.8	3.8	3.8
	Total for (*n* - 1)	- 135.1	4920.7	- 3503.5	- 2835.7	- 2847.3	- 2820.8

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($2p^6$). 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 N_{3d} 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.

FINAL CONFIGURATION	INITIAL CONFIGURATION											
	2p	3s	3p	4s	3d	4p	5s	4d	4f	5p	5d	6s
2p	☒	E -204	☒	E -26.7	E -4.3	☒	E -8.8	E -2.3	E	☒	E -1.2	E -4.1
3s	A 408	☒	E 29,160	☒	☒	E R*	☒	☒	☒	E -78.1	☒	☒
3p	☒	A -87,474	☒	E 33,316	E 39,029	☒	E 3,890	E 4,774	☒	☒	E 1,744	E 1,558
4s	A 53.3	☒	A -11,105	☒	☒	E 30,001	☒	☒	☒	E 659	☒	☒
3d	A 43.8	☒	A -65,049	☒	☒	E 12	☒	☒	E	E 8.8	☒	☒
4p	☒	A R*	☒	A -90,034	A -7,695	☒	E 55,186	E 38,039	☒	☒	E 5,051	E 4,679
5s	A 17.6	☒	A -1,297	☒	☒	A -18,395	☒	☒	☒	E 37,995	☒	☒
4d	A 22.9	☒	A -7,958	☒	☒	A -63,398	☒	☒	E	E 24,991	☒	☒
4f	☒	☒	☒	☒	A	☒	☒	A	☒	☒	E	☒
5p	☒	A 234	☒	A -1,977	A -5.3	☒	A -113,985	A -14,994	☒	☒	E 42,005	E 78,005
5d	A 12.4	☒	A -2,906	☒	☒	A -8,418	☒	☒	A	A -70,009	☒	☒
6s	A	☒	A -519	☒	☒	A -1,560	☒	☒	☒	A -26,002	☒	☒
	558	-87,444	-59,674	-58,722	31,324	-61,758	-54,918	27,817		-32,435	48,799	84,238

R* 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[42] modified the computer program of Johnson, Palumbo, and Hunter[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*,

$$XeNe^{+} + e^{-} \rightarrow Ne^{*} + Xe \qquad (21)$$

with a rate coefficient 6E-08. The reaction given by Eq. (21) was replaced by two reactions

$$XeNe^+ + e^- \rightarrow Ne(3s) + Xe \qquad (22)$$

and

$$XeNe^+ + e^- \rightarrow Ne(Z) + Xe \qquad (23)$$

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 McDaniel, et al.,[19] 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/cm^2 and again with 11A/cm^2 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 → 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, δ, a laser cavity with length, L, less than 35 meters will have δ/λ less than 0.1.

For the case of a wavelength near a resonance in the 3s → 4p transition, the consequences are different. A length of laser cavity of only 0.1116 meters yields δ/λ 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 → 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.

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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.

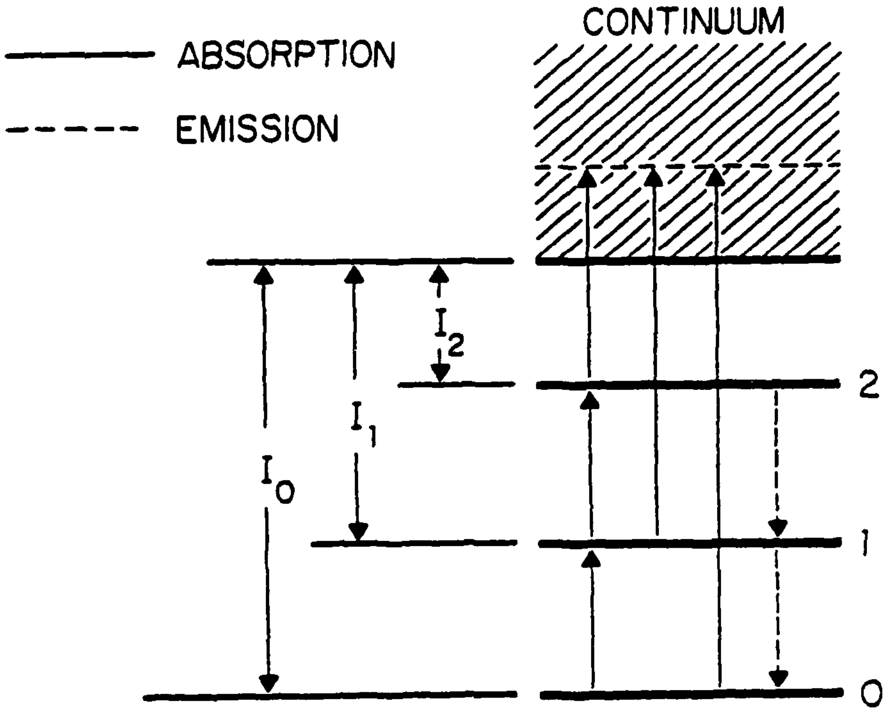

FIG. 1. Energy level diagram for a hypothetical atom used to derive equation for n - 1.

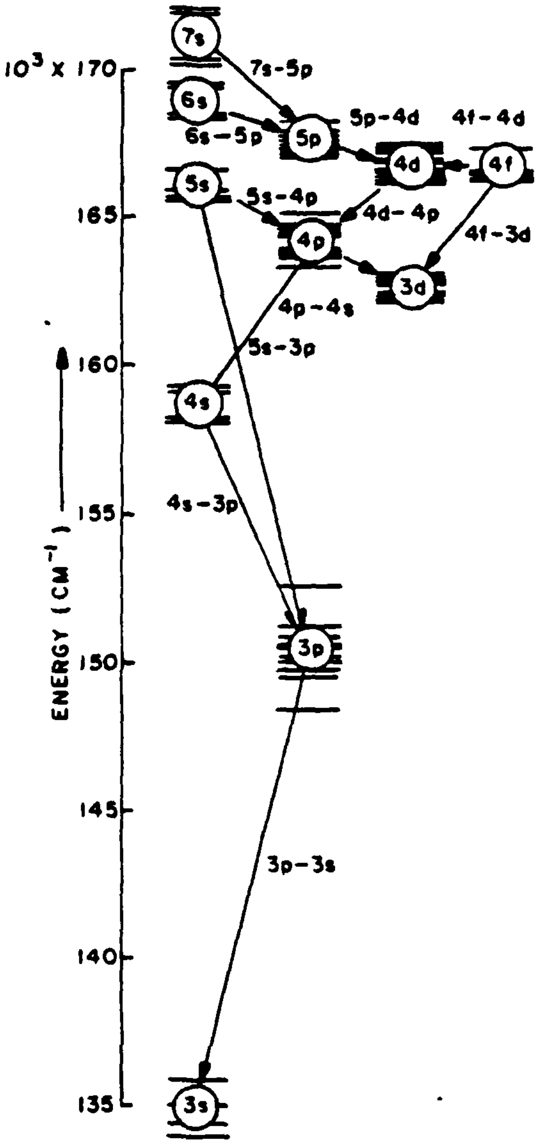

FIG 2. Energy level diagram for Neon. (From R. J. Pressley, *Handbook of Lasers*, Chemical Rubber Co., Cleveland Ohio, 1971[30])

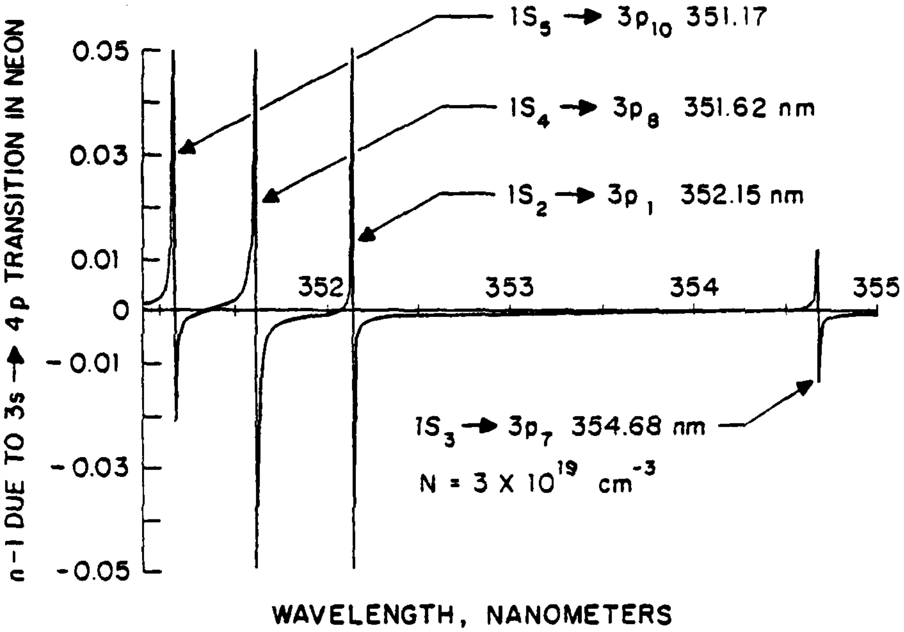

FIG. 3. Plot of n - 1 as a function of wavelength. The quantity plotted *is contribution of* $3s \rightarrow 4p$ transition in Neon.

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