Plasma Ionization Enhancement by Laser Line Radiation

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The possibility of increasing the degree of ionization in a plasma, by use of incident laser wavelengths matched to specific atomic transitions, is theoretically investigated. Such electron production occurs via a multistep process involving laser excitation of neutral atoms to electronic levels near the continuum and subsequent ionization of these atoms through collisions with thermalized plasma electrons. Detailed calculations for atomic oxygen, nitrogen, carbon, and cesium indicate that present state-of-the-art pulsed lasers are capable of raising 10^{14} to 10^{16} cm^{-3} electron densities by a factor of two. Use of laser-diode and laser-shock tube combinations for diagnostic studies of recombination processes is discussed briefly.

I. Introduction

THE production or perturbation of partially ionized plasmas by intense radiation is of significant interest. Relatively recent innovations, such as the high-intensity flash-tube[1] and the laser, have allowed experimental verification of particle-radiation interactions. In particular, the ultrahigh intensity of the laser has been used to ionize substances by causing localized electrical breakdown at the beam's focus.[2] Little attention, however, has been given to use of the narrow line-widths of lasers for excitation of discrete atomic or molecular transitions. Recently, Measures[3] theoretically investigated the possibility of using laser-induced selective excitation to pump atoms optically in a plasma into a higher state. He suggested that the increase in spontaneous emission from such a level could be used to diagnose plasma conditions. The small assortment of radiation wavelengths previously available in commercial lasers has prevented the matching of many atomic transitions to laser lines. However, recent advances in both fluid and solid-state lasing substances have made it possible to span the spectrum from the ultraviolet through the visible and into the near infrared.[4] As a consequence, it is feasible to use laser line radiation to excite atomic or molecular species and, with the increased power levels now available, to produce or enhance the ionization in a plasma. This process has a wide range of application, from diagnosing missile re-entry phenomena to providing space-charge neutralization in energy conversion devices.[5]

This paper discusses the possibilities of enhancing plasma ionization by employing laser line radiation to "pump" atoms into excited states, from which they are readily collisionally ionized. Such enhanced ionization could be created for short periods to study microscopic processes, such as recombination, in the plasma, or it could be used to produce long-lasting effects in specialized devices. OI, NI, and CI fall into the first category and are investigated in detail because of their astrophysical[6] and aerophysical importance in stellar and planetary atmospheres. CsI is analyzed because of its use in steady-state energy conversion devices.

We should emphasize that this method of selected laser line absorption differs from plasma production by "massive" excitation. In the latter technique, localized electrical breakdown occurs at the focus of the laser beam, with subsequent heating of the electrons by inverse bremsstrahlung.[2] The important parameter is the local field intensity at the focus, and the laser wavelength per se plays no significant role. The resulting plasmas are very dense, up to 10^{21} cm^{-3}, with particle energies of several hundred electron volts. However, the total volume of plasma initially excited is quite small, on the order of 0.1 cm.[3]

In contrast, plasma enhancement by discrete laser line absorption occurs in an interaction volume that is limited only by the physical size and attenuation of the laser beam. This volume would, in general, be several orders of magnitude larger than that produced by massive excitation, and therefore the incident radiation would be distributed among a larger number of particles. The "tunable" lasers required for discrete line absorption, however, will normally supply less power than the maximums available for massive excitation.

II. Radiation-Induced Excitation of Atomic Levels

The general features of plasma excitation via discrete laser lines can be most conveniently visualized if we consider laser pumping to counteract the collisional and radiative recombination processes. A minimum, or "bottleneck," in the rate of de-excitation of atoms as a function of the principal quantum number of the excited states in a collision-dominated plasma has been postulated by Byron et al.[7] and by Hinnov and Hirschberg[8] to limit the rate of such recombinations. On the basis of their analyses, the energy level associated with this bottleneck can be estimated for hydrogenic atoms.

The populations of the atomic energy levels above this bottleneck are in equilibrium with the free electron density. Consequently, absorption of laser radiation, which produces atomic excitation transitions crossing the bottleneck from below, will result in the production of additional electrons

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In order to maximize the efficiency of the laser ionization enhancement, therefore, it is necessary to choose a wavelength corresponding to a transition that crosses the bottleneck.

Associated with each transition is a spectral line profile determined by the lifetimes of the quantized atomic states, the thermal motion of the atoms, and interactions of the atoms with surrounding perturbers. The line shape indicates at exactly what wavelengths the absorption takes place and, therefore, how deeply the incident radiation can penetrate the plasma. In general, the profile of the line must be obtained by numerical evaluation of the Voigt function.[9,10]

Normally, the natural breadth of a line due to the lifetimes of states is negligible compared to the broadening produced by thermal and collisional processes. However, the ultrahigh radiation fields associated with strong pulsed laser sources may shorten the lifetimes sufficiently to cause significant natural broadening.[11]

Because of the significant electric fields produced by the charged particles in a plasma, the dominant perturbers are electrons and ions. However, the quasi-static interaction of heavy ions with atoms limits ionic perturbation effects, mainly to the wings of spectral lines.[12] Therefore, the shape of the central portion of an atomic line emanating from a plasma is predominantly affected by electron-neutral collisions.

Although the Voigt profile must, in general, be solved numerically, the dominance of the electron Stark broadening allows an analytical estimate of the volume absorption coefficient k_{ν_o} at the line center to be made.

$$k_{\nu_o} = k_o \exp\left[\left(\frac{\Delta\nu_c + \Delta\nu_N}{\Delta\nu_d}\right)^2 \ln 2\right] \operatorname{erfc}\left[\frac{\Delta\nu_c + \Delta\nu_N}{\Delta\nu_d} (\ln 2)^{1/2}\right] \quad (1)$$

For the conditions of this study, the natural broadening $\Delta\nu_N$ was found to be negligible in most instances and was not considered. Its effect would be to broaden the total line profile, thereby increasing the electron production. The constant k_o and the thermal and Stark half-widths (neglecting ionic contribution) are given in terms of the atomic and electronic masses m_a and m_e, the corresponding temperatures T_a and T_e, the electron and lower state atomic number densities N_e and N_l, and the absorption oscillator strength of the transition f_{lu}:

$$k_o = (2/\Delta\nu_d)(\pi \ln 2)^{1/2}(e^2/m_e c)N_l f_{lu} \quad (2)$$

Thermal half-width

$$\Delta\nu_d(s^{-1}) = (2/\lambda_o)(2kT_a \ln 2/m_a)^{1/2} \quad (3)$$

Stark half-width

$$\Delta\nu_c(s^{-1}) \simeq 2wN_e c/10^{24}\lambda_o^2 \quad (4)$$

for w in angstroms, N_e in cm^{-3}, c in cm s^{-1}, and λ_o in cm. The factor 10^{24} appears because of the normalization of w, calculated in angstroms for an electron density of 10^{16} cm^{-3}, to a single electron and the conversion of Å to cm. In the above relations, the constants e, c, and k represent the elementary charge, the speed of light in vacuum, and the Boltzmann constant, respectively, and λ_0 is the wavelength at the line center of the transition. The electron-impact half-width w is a slowly varying function of temperature and has been tabulated by Griem for various atomic transitions.[13] Approximate oscillator strengths have been derived for selected atoms by Griem[13] and specifically for Cs by Stone.[14] In addition to broadening, the line experiences a collisionally induced shift. Such shifts, in general a fraction of an angstrom, are also tabulated by Griem.[13]

If radiation with constant specific intensity I_o is incident on a plasma in which thermal and ionic broadenings are neglible with respect to electron-neutral Stark broadening, the absorption coefficient is given by a Lorentz function

$$k_\nu = k_{\nu_o}\{1 + 4[(\nu - \nu_o)/\Delta\nu_c]^2\}^{-1} \quad (5)$$

and the total absorption is reduced to analytic form. At a distance x in from the boundary of the plasma, the radiation absorbed across the entire line profile can be expressed in terms of an equivalent width W as[15]

$$W = \int_0^\infty \frac{I_o - I_\nu(x)}{I_o} d\nu = \int_0^\infty (1 - e^{-k_\nu x})d\nu = \pi A(\Delta\nu_c)xe^{-Ax}[J_o(iAx) - iJ_1(iAx)] \quad (6)$$

where $J_o(iAx)$ and $J_1(iAx)$ are the standard Bessel functions and the constant A is defined as

$$A = e^2 N_l f_{lu}/m_e c\Delta\nu_c \quad (7)$$

It is readily shown that as long as the absorption line is narrower than the frequency spread of the incoming beam, an increase in the Stark half-width results in an increase, at a given x, in the total absorption of radiation. Since a broadened line also allows greater uniform penetration of the radiation into the plasma, a large Stark width (within the confines of the incident beam) optimizes the radiation-particle interactions within the plasma.

III. Recombination Coefficients and Power Requirements for O, N, C, and Cs Plasmas

Theoretical and experimental data on total recombination rates involving excited as well as ground-state atoms are scant.[16] Bates et al.[17] have made theoretical calculations, based on Gryzinski's classical cross sections,[18] for the combined collisional and radiative recombination coefficients $\gamma(T_e, N_e)$ for hydrogen, hydrogenic ions, and pseudo-alkali atoms in an optically thin plasma. Their numerical results, indicating only slight sensitivity of the recombination rates to the species of singly charged ion involved, are in good agreement with Zgorzelski's results,[19] based on a somewhat different theoretical approach, and Aleskovskii's experimental values for collision-dominated Cs plasmas[20] (see Table 1). Dugan[21] has shown that Gryzinski's cross sections are applicable to alkalis, but may be less appropriate for nonhydrogenic atoms. However, Park[22] has recently shown experimentally that the recombination rate in a dense nitrogen plasma is also well predicted by the hydrogenic models. Further verification of the calculations of Bates et al. comes from some empirical data for the atomic absorption coefficients $a_\nu(T_e)$ in optically thin atomic O and N,[23,24] which are in general agreement with the theoretical results of Peach[25] and of Biberman and Norman.[26] This temperature-dependent absorption coefficient for bound-free transitions is defined as an averaged value with respect to the individual coefficients $\alpha_n(\nu)$ and population densities N_n of the nth atomic level:

$$\alpha_\nu(T_e) = \frac{1}{N}\sum_n \alpha_n(\nu)N_n(T_e) \quad (8)$$

N represents the total population density of atoms in all states.

With the use of Milne's relation between the individual absorption coefficients and their inverse recombination cross sections, an expression for the total radiative recombination coefficient $\beta(T_e)$ can be written

$$\beta(T_e) = \frac{2^{1/2}h^3 Q(T_e)\exp(E_i/kT_e)}{(m_e kT_e)^{3/2}\pi^{1/2}c^2 g_{io}} \int_0^\infty \alpha_\nu(T_e)\nu^2 \times \exp\left(-\frac{h\nu}{kT_e}\right) d\nu \quad (9)$$

where $Q(T_e)$ is the atomic partition function, E_i is the atomic

Table 1 Collisional-radiative recombination coefficients[a]

Coefficient	N_e, cm^{-3}			
	10^{11}	10^{12}	10^{13}	10^{14}
$\gamma^b(T_eN_e)$, cm^3 sec^{-1}	4.4×10^{-12}	1.4×10^{-11}	6.8×10^{-11}	3.8×10^{-10}
$\gamma^c(T_e,N_e)_{\rm H}$, cm^3 sec^{-1}	5.6×10^{-12}	1.7×10^{-11}	7.6×10^{-11}	5.2×10^{-10}
$\gamma^c(T_e,N_e)_{\rm alkali}$, cm^3 sec^{-1}	4.3×10^{-12}	1.4×10^{-11}	6.5×10^{-11}	4.9×10^{-10}

[a] $T_e = 3000$°K.
[b] Aleskovskii.[20]
[c] Bates et al.[17]

ionization potential, h is Planck's constant, and g_{io} is the degeneracy of the ground-state ion. Radiative recombination coefficients $\beta(T_e)$ were calculated from selected averaged values of $\alpha_\nu(T_e)$ in cm^2 (see Table 2); these coefficients, calculated here for the coronal limit,[27] show fair agreement with the hydrogenic values of Bates.

Information about recombination rates for atomic carbon is practically nonexistent. Praderie has done some theoretical analysis wherein he calculated the absorption coefficients at 6300°K using the quantum defect method.[28] Conversion of these coefficients by Eq. (9) to radiative recombination coefficients indicates agreement to within a factor of two with the hydrogenic coefficients calculated by Bates et al.[17] for transitions in the visible spectrum.

In view of the generally good correlation between existing information on collisional, as well as radiative, recombination rates and the hydrogenic predictions, it seems reasonable to assume that the hydrogenic estimates for the combined collisional-radiative recombination rates will be of the correct order of magnitude for all singly charged ionic species, and within a factor of two for most conditions of interest. Consequently, the analytical results of Bates et al. were adopted in evaluating the laser power requirements for specific ionization increases in collision-dominated singly charged O, N, C, and Cs plasmas.

We are now in a position to calculate the increased electron density produced by laser excitation. The recombination and excitation rates depend on the electron temperature, and therefore in the general case it is necessary to simultaneously determine the enhancement of both the electron temperature and the plasma density. The details of this calculation have been presented for other types of nonequilibrium plasmas[29-33] and are not repeated here.

Increases in the electron density and temperature are caused by the energy imparted to the plasma via the laser pumping. Initially, most of this energy produces additional electrons with possibly an associated loss of free-electron energy due to collisional ionization. Subsequently, however, the number of recombinations, as well as the de-excitation rate between the atomic levels involved in the laser pumping, increases. Since all such collisional de-excitations increase the free-electron energy, the electron temperature is expected to rise above its initial value.

The production of electrons by laser excitation requires that the atoms in the lower level of the laser pumped transition are continually replenished. Ultimately, this is accomplished by reducing the densities of the relatively highly populated ground state and levels near the ground state. Consequently, it is necessary to have sufficient atoms in low-lying levels to accommodate the increase in plasma ionization.

The strong radiation fields produced by the laser may result in a significant amount of stimulated emission in the plasma, which will decrease the effect of laser radiation absorption. The net rate of such absorptions for a radiation density $\rho(\nu_o)$ is

$$dN_u/dt = \rho(\nu_o)B_{lu}N_l(1 - g_lN_u/g_uN_l) \quad (10)$$

where the second term on the right-hand side represents stimulated emissions. B_{lu} is the Einstein coefficient for stimulated absorptions between the lower and upper levels of the laser-induced transition, and g_l and g_u are the associated degeneracies of these levels. It is evident from Eq. (10) that the ratio of N_u to N_l must be maintained small enough to prevent an excessive rate of stimulated emission.

The laser energy required to sustain an increased electron density must be sufficient to nullify the additional recombination that occurs (neglecting increases in the collisional ionization rate). If we assume 1) that the electron temperature is insignificantly affected by relatively small increases in the electron density, 2) that stimulated emissions can be neglected, and 3) that the increase in collisional ionization is small compared with the additional recombinations, the steady-state process is characterized by the rate of laser-induced absorptions per unit area $WI_o\omega/h\nu_o$ and by the additional recombination rate per unit area in a homogeneous plasma

$$[\gamma(T_e,N_{e_2})N_{e_2}^2 - \gamma(T_e,N_{e_1})N_{e_1}^2]x$$

The subscripts 1 and 2 refer to conditions prior to and during laser excitation, respectively, and ω is the solid angle of the incident radiation on the plasma. For a plasma 1 cm in depth, the required laser intensity incident on the plasma is

$$I \equiv I_o\omega \simeq [\gamma(T_e,N_{e2})N_{e2}^2 - \gamma(T_e,N_{e1})N_{e1}^2]h\nu_o/W \quad (11)$$

Figures 1, 2, and 3 are energy level diagrams for atomic O, N, and C adapted from Herzberg.[34] Figure 4, an energy-level diagram for atomic Cs is adapted from White.[35] Approximate positions of the recombination bottlenecks, for 1-ev OI, NI, and CI plasmas and for a 0.25-ev CsI plasma, are indicated by horizontal dashed lines. The corresponding energies approximate the $2s^22p^34s\,^5S^0$ state in O, $2s^22p^24s\,^4P$ state in N, $2s^22p3p\,^1D$ state in C, and 8^2P state in Cs. For each element, a number of multiplet transitions were chosen

Table 2 Radiative recombination coefficients

Coefficient	T_e, °K			
	10,500	11,000	12,000	13,000
$\beta^a(T_e)_{\rm O}$, cm^3 sec^{-1}	6.7×10^{-14}	7.3×10^{-14}	4.4×10^{-14}	4.7×10^{-14}
$\beta^b(T_e)_{\rm N}$, cm^3 sec^{-1}	2.8×10^{-13}	3.5×10^{-13}	3.7×10^{-13}	3.5×10^{-13}
$\beta^c(T_e)_{\rm H}$, cm^3 sec^{-1}	3.9×10^{-13}	3.8×10^{-13}	3.5×10^{-13}	3.3×10^{-13}

[a] Boldt.[23]
[b] Boldt.[24]
[c] Bates et al.[17] in limit $Ne \rightarrow 0$.

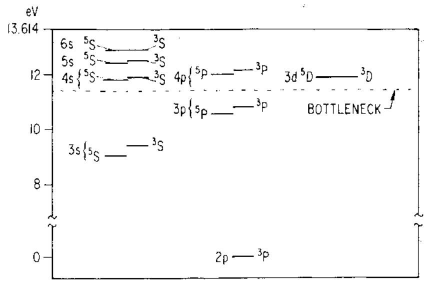

Fig. 1 Energy level diagram for OI terminating in the $^4S^0{}_{3/2}$ state (adapted from Ref. 34).

that satisfied the energy requirements, i.e., crossed the recombination bottleneck, and for which sufficient information was available[13,36,37] to allow calculation of the equivalent widths [see Eq. (6)]. Resultant laser power requirements necessary to double an electron density of 10^{16} cm^{-3} in 1-ev OI, NI, and CI plasmas are calculated from Eq. (11) and tabulated for the various multiplets in Table 3. Though these energy fluxes are large, indications are that they are available now, or will be in the near future, in pulsed-laser systems. Available lasing substances and presently obtainable power levels are discussed in Sec. IV. The situation is far less critical for atomic Cs (see Table 4), where diode conditions might call for doubling an electron density of 10^{14} cm^{-3} in a 0.25-ev plasma. The line half-widths $\Delta\lambda_c$, in Å, for the various transitions are also tabulated in Tables 3 and 4; they indicate that laser lines with half-widths of a small fraction of an angstrom could be used for the OI, NI, and CI plasmas. The total line-integrated intensities of such lasers would be correspondingly reduced.

The laser-induced transitions should increase the populations of all levels above the bottleneck. Consequently, a rough estimate of the maximum change in the electron temperature can be made by assuming that levels just above the bottleneck retain their initial populations as the electron density is doubled. Applying a modified form of the Saha relation to the population density N_u of such a state

$$\frac{N_e^2}{N_u} = \frac{2(2\pi m_e kT_e)^{3/2}}{h^3}\,\frac{Q_i(T_e)}{g_u}\exp\left[-\frac{E_i - E_u}{kT_e}\right] \quad (12)$$

prior to and during laser excitation, indicates that for the plasma conditions of this study the maximum possible change in the electron temperature will be less than 50%. In the preceding relation E_u is the energy of the level above the ground state and $Q_i(T_e)$ is the ionic partition function. Additional calculations suggest that the reservoir of atoms in the low-lying states, for the CsI, OI, and NI plasmas considered, should be sufficient to provide the additional electrons. However, the supply may be more marginal for the CI plasma.

In order to gain some insight into the magnitude of the stimulated emission rate, we considered the case where

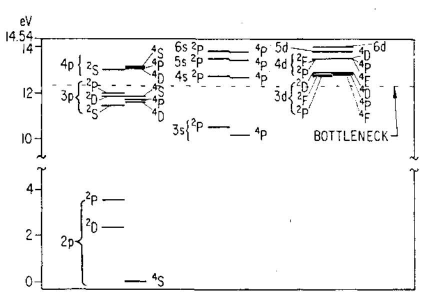

Fig. 2 Energy level diagram for NI terminating in the $^3P_0$ state (adapted from Ref. 34).

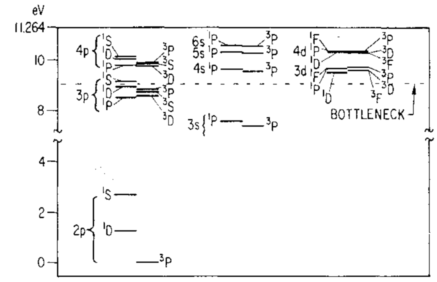

Fig. 3 Energy level diagram for CI terminating in the $^2P^0{}_{1/2}$ state (adapted from Ref. 34).

prior to laser excitation, the lower- and upper-population densities are approximately in equilibrium, and during the excitation process, the population of the lower level is relatively unaltered. Under these conditions, for a doubling of the electron density and a constant or increasing electron temperature, Eq. (12) and Boltzmann's relation suggest that

$$(g_l/g_u)N_{u_2}/N_{l_2} \lesssim 4(g_l/g_u)N_{u_1}/N_{l_1} \simeq 4\exp\left(-hc/\lambda_o kT_{e_1}\right) \quad (13)$$

where the subscripts 1 and 2 refer to populations prior to and during laser excitation, respectively. The resultant calculations indicate, from Eq. (10), that for the atomic transitions considered stimulated emission is negligible for CsI, and less than 33% of absorption for NI and CI. For OI, however, a maximum of 72% is reached. The conditions considered may be quite hypothetical, but they do point out that in some cases stimulated emissions may demand additional laser pumping and therefore somewhat larger power requirements.

IV. Laser Sources and Power Levels

The development of new lasing materials and efficient nonlinear optical media during the past few years has radically increased the range of wavelengths available for radiation-particle interaction. A review of these techniques and their application to line spectroscopy is given in Ref. 4.

Numerous semiconducting materials have been found to emit coherent radiation, thereby producing a host of new laser wavelengths. Frequency-changing methods, by which these wavelengths are altered through temperature, impurity, and composition variations of the lasing substance, by the application of external fields, or by nonlinear optical techniques, show sufficient promise that, if they are used in conjunction with liquid-dye lasers, a major portion of the spectrum from ultraviolet through infrared will eventually be totally covered. For example, GaAs, commercially available as a semiconductor laser, is temperature-tunable between 8000 and 9050 Å, with an electrical-to-radiation conversion efficiency of 80% at 77°K and a corresponding line half-width on the order of 1 Å. Greater wavelength coverage is provided by ternary compounds such as CdS_x-Se_{1-x}, which spans the spectral region 4900 to 6900 Å, and

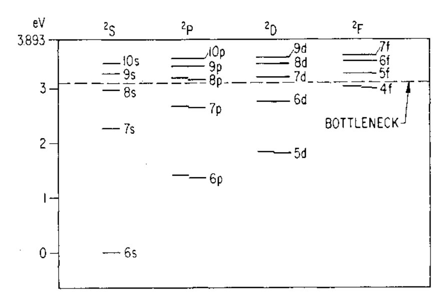

Fig. 4 Energy level diagram for CsI (adapted from Ref. 35).

Table 3 Laser power required to double an electron density of 10^{16} cm^{-3} in 1 − ev OI, NI, and CI Plasmas[a]

Species	Transition	λ_0, Å	$\Delta\lambda_c$, Å	I, W/cm²/Å
	$3^3S^0 - 5^3P$	3692.44	0.46	6.5×10^7
		6453.64		1.3×10^6
OI	$3^5P - 5^5S^0$	6454.48	1.09	7.7×10^5
		6456.01		5.5×10^5
	$3^3P - 5^3S^0$	7254.19	1.40	1.2×10^6
		7254.47		5.8×10^5
	$3^3P - 6^3S^0$	6046.26	2.84	6.1×10^6
		6046.46		3.0×10^6
		4137.63		2.2×10^7
	$3s^4P - 4p^4S^0$	4143.42	0.17	1.1×10^7
NI		4151.46		7.4×10^6
	$3s^4P - 4p^4P^0$	4214.73	0.15	2.5×10^7
		4223.04		1.8×10^7
	$3s^1P^0 - 4p^1S$	4932.00	0.45	4.2×10^5
	$3s^1P^0 - 5p^1P$	4371.33	0.54	1.7×10^6
CI	$3s^1P^0 - 5p^1D$	4268.99	0.84	1.6×10^6
	$3s^1P^0 - 5p^1S$	4231.35	1.09	2.6×10^6

[a] Only the strongest lines in each multiplet are included.

Table 4 Laser power required to double an electron density of 10^{14} cm^{-3} in a 0.25-ev CsI plasma

Transition	λ_0, Å	$\Delta\lambda_c$, Å	I, W/cm²/Å
5^2D–6^2F	7229	0.29	4.8×10^2
5^2D–5^2F	8081	0.11	1.3×10^2
5^2D–6^2F	7280	0.29	3.2×10^2
5^2D–7^2F	6871	0.56	6.5×10^2

$GaAs_xP_{1-x}$, which covers wavelengths between 6300 and 8450 Å.

Production of second and third[38] harmonics of the incident radiation can yield additional spectral lines via nonlinear interaction between intense laser beams and specific crystals or liquids, such as ammonium dihydrogen phosphate (ADP). In addition, nonlinear interaction between intense focussed lasers and Raman-active materials yields coherent radiation that is shifted in energy by some multiple of the Raman-bond energy. Resultant strong Stokes (S_1, shift to longer wavelengths) and weaker anti-Stokes (AS_1, AS_2, etc., shift to shorter wavelengths) lines are observed. The magnitude of these shifts is generally between 445 and 4160 cm^{-1}.

Application of parametric oscillation techniques, wherein optical media such as $LiNbO_3$ and $Ba_2NaNb_5O_{15}$ are employed, allows tuning of an incident wavelength within approximately the interval $1.4\lambda_{in} \leq \lambda_{out} \leq 4\lambda_{in}$ with conversion efficiencies ranging from 1% to 20%.[39]

Laser-pumped fluorescent dye solutions have exhibited lasing action over the entire spectral region from 4000 Å to 1.06 μ.[40–42] Wavelength tuning is easily achieved by variation of the organic solute concentration. Power levels up to 4 MW have been obtained over times equal to the pulse length of the exciting radiation (for Q-switched ruby or Nd lasers, this is usually 10 to 30 nsec). Flashlamp-pumped dye lasers of longer duration have also been successfully produced, yielding up to 2 J of wavelength-tunable laser output.

The previous discussions of various techniques indicate that wavelengths from 2500 Å to several microns should be available with proper laser tuning. This spread would cover the spectral region of interest for the OI, NI, CI, and CsI transitions considered in this study. Restriction of the lasers to Q-switched operation would yield output power levels of from 10^3 to 10^7 W/Å for pulse times between 10^{-8} and 10^{-7} sec.

V. Conclusions

Calculations were made for a cesium plasma at typical diode conditions and for substantially ionized oxygen, nitrogen, and carbon plasmas. The results indicate that presently available lasing techniques are capable of raising a 10^{14} cm^{-3} electron density in a 0.25-ev cesium plasma by more than a factor of two. However, doubling an electron density of 10^{16} cm^{-3} in 1-ev oxygen, nitrogen, or carbon plasmas requires laser power levels approaching the upper limits presently available. Though the capability for such plasma enhancement exists, sustaining these high electron levels would prove difficult.

Aside from simply enhancing the ionization, laser interaction studies can shed light on recombination phenomena. If the electron density is suitably monitored during the time of radiation-particle interaction, recombination rates can be measured. Furthermore, varying the laser-excited transition (or the plasma conditions) can aid in determining the nature and precise position of the recombination bottleneck and its dependence on the plasma parameters.

In any practical experiment, the manner of initial plasma production may differ, and the applicable diagnostic techniques may vary. For example, regions subjected to reflected shock waves may prove to be good sources of oxygen and nitrogen plasmas, and spectroscopic or probe techniques could be applied to monitor the electron density. However, carbon excitation may require special techniques, since carbon is a solid at room temperature and has a heat of sublimation approaching 6 ev.[43] Cesium is easier to deal with and readily ionizable. Interaction experiments involving Cs could be performed in a plasma diode.[5] Such an arrangement would have the advantage that the diode current would serve to monitor the electron density within the diode.

It must be remembered that the power requirements calculated in this study are based on simple estimates of recombination rates and bottleneck positions. Increases in collisional ionization and electron temperature during laser excitation are neglected, and both processes favor lower power levels. However, the authors hope that these basic calculations will stimulate interest in the possibilities of applying laser-particle interactions to basic research in recombination phenomena and will increase understanding of the line radiation intensities required and of lasing substances available to provide specific plasma conditions.

References

1 Wheaton, J. E. G., "Improvements in Design and Performance of the Large Aperture Lyman Flashtube," *Applied Optics*, Vol. 3, 1964, p. 1247.

2 Meyerand, P. G., Jr., "Laser Plasma-Production—A New Era of Plasmadynamic Research," *AIAA Journal*, Vol. 5, No. 10, Oct. 1964, p. 1730.

3 Measures, R. M., "Selective Excitation Spectroscopy and Some Possible Applications," *Journal of Applied Physics*, Vol. 39, 1968, p. 5232.

4 Dewey, C. F., Jr., "Applications of Lasers to Line Spectroscopy in Plasma Diagnostic Methods for Ionized Gas Flows" AFFDL-TR-68-170, 1968, Air Force Flight Dynamics Lab

5 Carpenter, J. R., "Experimental Investigation of Plasma Ionization Enhancement by Laser Line Radiation," M. S. thesis, 1969, Massachussetts Institute of Technology.

6 Aller, L. H., *The Atmospheres of the Sun and Stars*, 2nd ed., Ronald Press, New York, 1963, p. 417.

7 Byron, S., Stabler, R. C., and Bortz, P. I., "Electron-Ion Recombination by Collisional and Radiative Processes," *Physical Review Letters*, Vol. 8, 1962, p. 376.

8 Hinnov, E. and Hirschberg, J. G., "Electron-Ion Recombination in Dense Plasmas," *Physical Review*, Vol. 125, 1962, p. 795.

9 Unsöld, A., *Physik der Sternatmosphären*, 2nd ed., Springer-Verlag, Berlin, 1955, p. 261.

10 Davies, J. T. and Vaughan, J. M., "A New Tabulation of the Voigt Profile," *Astrophysical Journal*, Vol. 137, 1963, p. 1302.

[11] Oettinger, P. E., "Radiation-Induced Line Broadening of the Principal Series of Sodium," *Journal of Quantitative Spectroscopy and Radiative Transfer*, Vol. 10, 1970, p. 127.

[12] Wiese, W. L., "Line Broadening," *Plasma Diagnostic Techniques*, edited by S. L. Leonard and R. H. Huddlestone, Academic Press, New York, 1965, p. 275.

[13] Griem, H. R., *Plasma Spectroscopy*, McGraw-Hill, New York, 1964, pp. 363, 454.

[14] Stone, P. M., "Cesium Oscillator Strengths," *Physical Review*, Vol. 127, 1962, p. 1151.

[15] Goody, R. M., *Atmospheric Radiation. I. Theoretical Basis*, Oxford University Press, Oxford, 1964, p. 126.

[16] Marr, G. V., *Photoionization Processes in Gases*, Academic Press, New York, 1967, p. 130.

[17] Bates, D. R., Kingston, A. E., and McWhirter, R. W. P., "Recombination between Electrons and Atomic Ions. I. Optically Thin Plasmas," *Proceedings of the Royal Society (London), Ser. A*, Vol. A267, 1962, p. 297.

[18] Gryzinski, M., "Classical Theory of Electronic and Ionic Inelastic Collisions," *Physical Review*, Vol. 115, 1959, p. 374.

[19] Zgorzelski, M. J., "Reaction Rates and Energy Distributions During Ionization and Recombination Processes in Optically Thin Plasmas," 68-1, 1968, MIT Fluid Mechanics Lab., Cambridge, Mass.

[20] Aleskovskii, Y. M., "Investigation of Volume Recombination in a Cesium Plasma," *Soviet Physics JETP*, Vol. 17, 1963, p. 570.

[21] Dugan, J. V., Jr., "Calculation of Three-Body Collisional Recombination Coefficients for Cesium and Argon Atomic Ions With an Assessment of the Gryzinski Cross Sections," *Journal of Applied Physics*, Vol. 37, 1966, p. 5011.

[22] Park, C., "Measurement of Ionic Recombination Rate of Nitrogen," *AIAA Journal*, Vol. 6, No. 11, Nov. 1968, p. 2090.

[23] Boldt, G., "Rekombinations—und 'Minus'—Kontinuum der Sauerstoffatome," *Zeitschrift fur Physik*, Vol. 154, 1959, p. 319.

[24] Boldt, G., "Rekombinations—und 'Minus"—Kontinuum der Stickstoffatome," *Zeitschrift fur Physik*, Vol. 154, 1959, p. 330.

[25] Peach, G., "Continuous Absorption Coefficients for Non-Hydrogenic Atoms," *Monthly Notices of the Royal Astronomical Society*, Vol. 124, 1962, p. 371.

[26] Biberman, L. M. and Norman, G. E., "On the Calculation of Photoionization Absorption," *Optics and Spectroscopy*, Vol. 8, 1960, p. 230.

[27] Wilson, R., "The Spectroscopy of Non-Thermal Plasmas," *Journal of Quantitative Spectroscopy and Radiative Transfer*, Vol. 2, 1962, p. 477.

[28] Praderie, F., "Calcul de Quelques Sections de Photoionization Du Carbone Neutre," *Annales D'Astrophysique*, Vol. 27, 1964, p. 129.

[29] Fowler, R. H., *Statistical Mechanics*, 2nd ed., Cambridge University Press, Cambridge, 1936, Ch. 17.

[30] Kerrebrock, J. L. and Hoffman, M. A., "Nonequilibrium Ionization due to Electron Heating: II. Experiments," *AIAA Journal*, Vol. 2, No. 6, June 1964, p. 1080.

[31] Dugan, J. V., Jr., Lyman, F. A., and Albers, L. V., "Solution of the Boltzmann and Rate Equations for the Electron Distribution Function and State Populations in Non-equilibrium MHD Plasmas," *Electricity from MHD*, Vol. II, International Atomic Energy Agency, Vienna, 1966, p. 85.

[32] Nighan, W. L., "Electron-Transport Phenomena in Thermionic Converters," *Journal of Applied Physics*, Vol. 39, 1968, p. 223.

[33] Norcross, D. W., "Departures from LTE in the Thermionic Converter," 67-19, 1967, Sperry Rand Research Center, Sudbury, Mass.

[34] Herzberg, G., *Atomic Spectra and Atomic Structure*, Dover, New York, 1944, pp. 143, 144, 163.

[35] White, H. E., *Introduction to Atomic Spectra*, McGraw-Hill, New York, 1934, p. 87.

[36] Moore, C. E., "Atomic Energy Levels (Vol. I)," Circular 467, National Bureau of Standards, Washington, 1949, pp. 21, 24, 32, 35, 45, 47.

[37] Moore, C. E., "A Multiplet Table of Astrophysical Interest," T. N. 36, National Bureau of Standards, Washington, 1959, pp. 2–8.

[38] Geusic, J. E. et al., "Continuous 0.532μ Solid-State Source Using $Ba_2NaNb_5O_{15}$," *Applied Physics Letters*, Vol. 12, 1968, p. 306.

[39] Bjorkholm, J. E., "Efficient Optical Parametric Oscillation Using Doubly and Singly Resonant Cavities," *Applied Physics Letters*, Vol. 13, 1968, p. 53.

[40] Sorokin, P. P. and Lankard, J. R., "Stimulated Emission Observed from an Organic Dye, Chloro-aluminum Phthalocyanine," *IBM Journal of Research and Development*, Vol. 10, 1966, p. 162.

[41] Kagan, M. R., Farmer, G. I., and Huth, B. G., "Organic Dye Lasers," *Laser Focus*, Vol. 4, 1968, p. 26.

[42] Miyazoe, Y. and Maeda, M., "Stimulated Emission from 19 Polymethine Dyes—Laser Action Over the Continuous Range 710–1060 $m\mu$," *Applied Physics Letters*, Vol. 12, 1968, p. 206.

[43] Herzberg, G., *Molecular Spectra and Molecular Structure, I. Spectra of Diatomic Molecules*, 2nd ed., Van Nostrand, Princeton, 1950, p. 481.