Kinetic Processes in Noble Gas Ion Lasers: A Review

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I. Introduction

EXPERIMENTAL observation of laser action involving the excited states of noble gas ions was first reported in 1964 by Bridges, Gordon, Convert, Bennett, and others.[1-8] Since that time, a voluminous literature reporting on the operational characteristics, spectroscopy, and probable excitation mechanisms for these gaseous ion lasers has appeared from many different parts of the world. A representative, but by no means exhaustive, list of these research works is compiled here in Refs. 9-31. These intensive research efforts have also been accompanied by a rapid commercial development of practical noble gas ion lasers capable of operating continuously at the output power level of several watts from a single mode over a long period of time.

In comparison with gaseous atom lasers (e.g., the helium-neon laser,[32] which operates in the red and near-infrared region), and the continuous duty gaseous molecule lasers (e.g., the N_2-CO_2 laser,[33,34] and the HF, DF lasers,[35-37] which operates in the infrared), the noble gas ion lasers generally offer the advantage of shorter wavelength operation, ranging from the blue-green part of the visible spectrum up to the near ultraviolet (where photodetectors are generally more sensitive, which is an important consideration in many scientific research and communication applications). While not nearly as powerful nor as thermally efficient as the continuous duty molecular lasers, the noble gas ion lasers are at least as efficient as, and much more powerful than (by factors of the order of 10^3), the gaseous atom lasers. Even though the pulsed molecular nitrogen laser[38,39] does operate in the near ultraviolet at very high peak power, the noble gas ion lasers are more flexible; in the sense that they offer a wide selection of wavelengths in the visible and the near ultraviolet regions, and that they can be operated in both the continuous and the pulsed modes. (Further comparison between gaseous lasers and solid/liquid state lasers is out of the scope of the present paper. For a more extensive comparison, the readers are referred to Refs. 40 and 41 or other recent monographs on the subject.)

Because of the potential usefulness of noble gas ion lasers in aerospace science and technology, the operational characteristics and performance parameters for this type of laser are summarized here together with a general description of the spectroscopy and kinetic processes leading to the inversion mechanisms. The current status of quantitative theory development is then critically reviewed. Some interesting and yet unsolved plasma physics problems related to the question of ion laser performance at high current densities are also discussed.

II. General Operational Characteristics

While the operational characteristics of all noble gas ion lasers are quite similar, the argon-ion laser appears to be the one that has been studied most extensively. In a typical argon-ion laser,[3,10] which we shall consider representative of the group henceforth, the active plasma is produced in a discharge tube of several millimeter diameter and of length ranging from a fraction of a meter to one or two meters.[31] The discharge tube has been made out of quartz, ceramic, graphite, or even metal segments, with provision for cooling on the outside. The argon filling pressure for most efficient operation (or maximum output power) was found to lie in the range of 0.01-1 mm Hg, depending on the discharge tube diameter and on the mode of operation. Generally, such "optimum pressure" was found to vary inversely with the tube diameter, and the pulsed mode operates in the lower range of pressure than does the CW mode. While rf induction[13] is sometimes used to generate the electrical discharge, the discharge current (either d.c. or low-frequency

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a.c.) is more frequently supplied by electrodes located inside and at both ends of the discharge tube. The ends of the discharge tube are often connected to a common gas-filling reservoir, but otherwise sealed from the outside world, either by Brewster angle plates (to minimize transmission loss) or directly by reflecting mirrors which form the optical cavity for inducing laser oscillation.

For the d.c. driven argon-ion laser, the threshold current density required to initiate laser oscillation in a typical low-loss optical cavity is found to be of the order of 50 amps/cm^2 over a considerable range of discharge tube diameter. Immediately above such oscillation threshold, the laser output power from a given size discharge tube operating at a fixed filling pressure was observed[3] to increase rapidly with the discharge current I, at a rate like the sixth power of I. Well above threshold, the current-dependence of output power becomes less steep but still very strong, varying like I^4 to I^2 near the maximum current point, which is usually limited by such practical factors as power supply capacity, excessive ablation of discharge tube wall material by ion bombardment, heat removal rate of cooling system, etc., in the individual experiments. For a given size discharge tube operating at a given discharge current, the power output of the laser was observed to vary with filling pressure, with a single-valued maximum occurring at certain optimum pressure, as mentioned earlier. It was found that maximum laser power so obtained can be further increased by a significant factor (of the order of 2) through the application of an axial magnetic field of appropriate strength.

The optical gain of the noble gas ion lasers is generally sufficiently high so that most of the strong ion lines normally observed in emission spectra can be excited at sufficiently high current density. Wavelength selection of the laser output light is usually made either through variation of the spectral reflectance of the mirrors forming the optical cavity, or through introduction of suitable dispersive element (e.g., prism, diffraction grating) into the optical cavity.

The generation efficiency of the noble gas ion lasers (defined here as the laser output power divided by the electrical discharge input power) was found to be generally quite low, currently being in the range of 10^{-5} to 10^{-3}. As we shall see later in Section IV, this low efficiency is mainly due to the fact that 1) a relatively small fraction of the noble gas ions produced within the discharge has the chance of being excited to the upper laser state before reaching the discharge tube wall and getting lost through recombination with an electron; and 2) the energy per laser photon is a small fraction of the excitation energy and of the total energy required to produce an ion-electron pair within the discharge tube. From the point of view of economy, such low generation efficiency leaves much to be desired. Thus, major improvement on this particular operational characteristics shall be one of the most important objectives in noble gas ion laser research and development.

III. Spectroscopy of Noble Gas Ion Lasers

While the total number of spectral lines observed in noble gas ion laser oscillations is quite large (over a hundred in the visible and ultraviolet regions), only the stronger lines are of interest in most practical applications. A representative wavelength listing of these stronger lines, together with their identified ionic energy states involved in the laser transition, is extracted from the extensive compilation of Bridges and Chester in Ref. 9 and reproduced here in Table 1. For the Ar^+ lines, the spontaneous emission life time of the upper state[8,42] and the relative laser line intensity under typical operating conditions[43] are also tabulated. It is seen that,

Table 1 Strong lines observed in neon, argon, krypton, and xenon-ion lasers according to compilation of spectroscopic data in Ref. 9; for the Ar+ lines, spontaneous emission lifetimes are taken from Ref. 8, and relative intensity refers to single-line CW laser output in milliwatts from a typical commercial laser of a few watts total power rating (Ref. 43)

Ion	Best measured wavelength in Å	Transition: Upper state		Transition: Lower state	Spontaneous emission lifetime of upper state, nsec	Relative laser line intensity
Ne^+	3323.79 ± 0.06	$3p\ ^2P_{3/2}{}^0$	→	$3s\ ^2P_{3/2}$		
Ne^+	3378.33 ± 0.06	$3p\ ^2P_{1/2}{}^0$	→	$3s\ ^2P_{1/2}$		
Ne^+	3392.86 ± 0.06	$3p\ ^2P_{3/2}{}^0$	→	$3s\ ^2P_{1/2}$		
Ar^+	4545.04 ± 0.1	$4p\ ^2P_{3/2}{}^0$	→	$4s\ ^2P_{3/2}$	9.4 ± 0.5	30
Ar^+	4579.36 ± 0.16	$4p\ ^2S_{1/2}{}^0$	→	$4s\ ^2P_{1/2}$	8.8 ± 0.3	220
Ar^+	4657.95 ± 0.02	$4p\ ^2P_{1/2}{}^0$	→	$4s\ ^2P_{3/2}$	8.7 ± 0.3	90
Ar^+	4726.89 ± 0.04	$4p\ ^2D_{3/2}{}^0$	→	$4s\ ^2P_{3/2}$	9.8 ± 0.2	170
Ar^+	4764.88 ± 0.04	$4p\ ^2P_{3/2}{}^0$	→	$4s\ ^2P_{1/2}$		580
Ar^+	4879.86 ± 0.04	$4p\ ^2D_{5/2}{}^0$	→	$4s\ ^2P_{3/2}$	9.1 ± 0.6	1400
Ar^+	4965.09 ± 0.02	$4p\ ^2D_{3/2}{}^0$	→	$4s\ ^2P_{1/2}$		530
Ar^+	5017.17 ± 0.02	$(^1D)\ 4p\ ^2F_{5/2}{}^0$	→	$3d\ ^2D_{3/2}$	7.9 ± 0.2	290
Ar^+	5145.33 ± 0.02	$4p\ ^4D_{5/2}{}^0$	→	$4s\ ^2P_{3/2}$	7.5 ± 0.5	1500
Ar^{++}	3511.13 ± 0.06	$4p\ ^3P_2$	→	$4s\ ^3S_1{}^0$		60
Ar^{++}	3637.86 ± 0.04	$(^2D^0)\ 4p\ ^1F_3$	→	$(^2D^0)4s\ ^1D_2{}^0$		40
Kr^+	4619.17 ± 0.1	$5p\ ^2D_{5/2}{}^0$	→	$5s\ ^2P_{3/2}$		
Kr^+	4680.45 ± 0.06	$5p\ ^2S_{1/2}{}^0$	→	$5s\ ^2P_{1/2}$		
Kr^+	4762.44 ± 0.06	$5p\ ^2D_{3/2}{}^0$	→	$5s\ ^2P_{1/2}$		
Kr^+	4825.18 ± 0.06	$5p\ ^4S_{3/2}{}^0$	→	$5s\ ^2P_{1/2}$		
Kr^+	5208.32 ± 0.04	$5p\ ^4P_{3/2}{}^0$	→	$5s\ ^4P_{3/2}$		
Kr^+	5681.92 ± 0.04	$5p\ ^4D_{5/2}{}^0$	→	$5s\ ^2P_{3/2}$		
Kr^+	6471.0 ± 0.5	$5p\ ^4P_{5/2}{}^0$	→	$5s\ ^2P_{3/2}$		
Kr^{++}	3507.42 ± 0.06	$5p\ ^3P_2$	→	$5s\ ^3S_1{}^0$		
Kr^{++}	4067.36 ± 0.06	$(^2D^0)\ 5p\ ^1F_3$	→	$(^2D^0)5s\ ^1D_2{}^0$		
Kr^{++}	4131.38 ± 0.06	$5p\ ^5P_2$	→	$5s\ ^3S_1{}^0$		
Xe^+	5419.16 ± 0.06	$6p\ ^4D_{5/2}{}^0$	→	$6s\ ^4P_{3/2}$		
Xe^+	5971.12 ± 0.06	$(^1D)\ 6p\ ^2P_{3/2}{}^0$	→	$(^1D)6s\ ^2D_{3/2}$		
Xe^+	7989 ± 3	$6p\ ^4P_{1/2}{}^0$	→	$6s\ ^4P_{1/2}$		
Xe^+	8714 ± 3	$6p\ ^4D_{3/2}{}^0$	→	$5d\ ^2P_{3/2}$		
Xe^+	9697 ± 4	$6p\ ^4D_{3/2}{}^0$	→	$5d\ ^4P_{5/2}$		

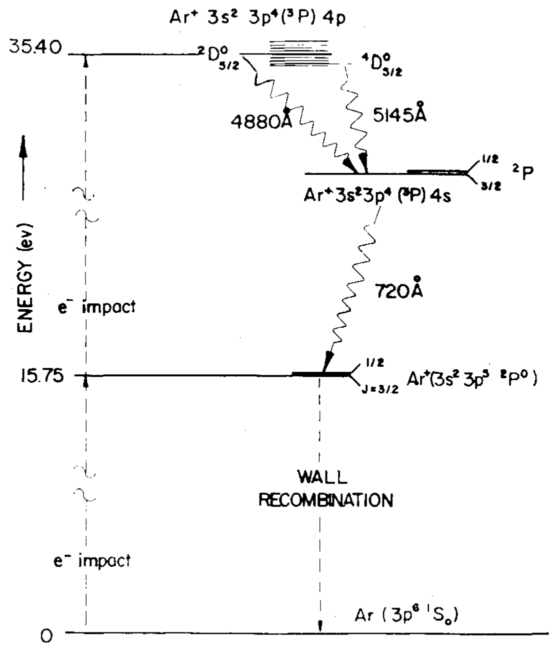

Fig. 1 Energy levels pertinent to the Ar^+ laser excitation mechanism.

with few exceptions, all the stronger lines for the singly-charged ions involved transition from one of the multiplet levels of an upper ionic configuration $np^4(n+1)p$ to one of the multiplet levels of a lower ionic configuration $np^4(n+1)s$, where n is the principal quantum number of the last filled p shell of the corresponding noble gas atom (i.e., $n = 2$ for neon, 3 for argon, 4 for krypton, and 5 for xenon). From the last two columns of Table 1, one may note that even though the stronger laser lines are also the strong lines observed in spontaneous emission,[9] the distribution of relative laser line intensity does not necessarily follow the distribution of spontaneous emission transition probability (or reciprocal lifetime). In fact, in the case of Ar^+, the two strongest lines at 4879.86 and 5145.33 Å often account for more than 60% of all the laser output power when the laser is operated in the multicolor mode. The reason is that the optical gain and power output of a laser line depend not only on its transition probability, but also on the population inversion density of the upper and lower states involved. The inversion mechanism, as well as the occupation number density for the upper and lower states under any given set of operating conditions, in turn, are governed by a number of competing kinetic processes and their specific excitation/de-excitation cross sections.

IV. Excitation and Inversion Mechanisms

The excitation/de-excitation processes responsible for generation of population inversion in argon-ion lasers have been extensively studied by Bennett et al,[7,8,18] Beigman et al,[21] Bridges and Halsted,[24] and Kitaeva et al.[29] While little attention has been given to specific excitation processes in other noble gas ion lasers, one may expect that the over-all inversion mechanism deduced for Ar^+ must be equally applicable to Ne^+, Kr^+ and Xe^+ under similar gas discharge conditions, on account of their spectroscopic similarities.

Referring to the energy level diagram depicted in Fig. 1 for the case of Ar^+ excitation, the authors in the aforementioned studies generally agreed that the primary inversion mechanism was due to the combined effect of a preferential electron-impact excitation of the upper $4p$ levels and a relatively short spontaneous emission life time of the lower $4s$ levels.[7] [Here, as elsewhere, the terms "$4p$ levels" and "$4s$ levels" refer to the various multiplet components of the $3s^23p^4(^3P)4p$ and the $3s^23p^4(^3P)4s$ configuration, respectively.] Under the typical low-pressure operating conditions, imprisonment of resonance line radiation (i.e., the 720 Å line connecting the $4s$ state and the ground state of Ar^+) is generally not very serious[24] and may further be alleviated by continuous Doppler shift accompanying the rapid radial acceleration of the positive ions by the plasma potential (see section V below). Thus, population inversion between the $4p$ and the $4s$ levels can be maintained at high current density (which implies high excitation/de-excitation rates for the lasing states) even in CW or quasi-CW operation.[7]

In regard to the detailed mechanism responsible for preferential excitation of the $4p$ levels, however, there appeared to be much less unanimous opinion among the different investigators. Altogether, four alternative paths for such preferential excitation have been suggested.[7,21,24] These are: 1) direct excitation by electron impact from the ground state $3s^23p^6\ ^1S_0$ of the neutral argon atom; 2) two-step excitation by electron impact through the ground state $3s^23p^5\ ^2P^0$ of the singly-charged argon ion Ar^+; 3) multistep electron impact excitation through some third configuration states of either Ar or Ar^+; and 4) radiative cascade into $4p$ from some upper radiating states excited by electron impact, either single-step or multistep, from the ground states of Ar and/or Ar^+.

The importance of path 1) previously cited was first pointed out by Bennett et al.,[7] who noted that according to the "sudden perturbation" theory, the excited state distribution produced in a fast electron collision of the type

$$e + \mathrm{Ar} \rightarrow (\mathrm{Ar}^+)^* + 2e \tag{1}$$

tends to favor the $3s^23p^4(^3P)4p$ configuration. Furthermore, the individual cross sections for direct excitation to the various multiplet levels of this configuration could be of magnitude competitive with the largest individual cross sections for exciting neutral levels by electron impact in the noble gases (i.e., cross sections of the order of 10^{-18} cm^2). Such large direct excitation cross sections were, indeed, confirmed by subsequent experimental measurements of Bennett, Mercer, Kindlmann, Wexler, and Hyman.[18]

In considering path 2, it has been argued[24] that if the ordinary dipole selection rule governing optical transitions in excited neutral atoms could be used to infer the ionic excitation cross sections in electron collision of the type

$$e + \mathrm{Ar}^+ \rightarrow (\mathrm{Ar}^+)^* + e \tag{2}$$

one would expect that excitation to the lower $4s$ levels should be more favorable than excitation to the upper $4p$ levels (since the latter configuration is of the same parity as the Ar^+ ground state). This would tend to produce a normal rather than an inverted population between the $4p$ and the $4s$ states. While no direct experimental measurement yet exists to either confirm or contradict such a conjecture, the theoretical calculations by Beigman et al[21] within their "Born-Coulomb" approximation seemed to suggest that the total excitation cross sections Q_i^* for the $4p$ and the $4s$ configurations may well be of the same magnitude. In fact, according to their numerical results, which are reproduced here in Table 2, excitation to the $4p$ configuration seemed to be slightly favored at high electron temperatures. (Though not explicitly stated in Ref. 21, the values of $\langle v_eQ_i^*\rangle$ were presumably based on a Maxwellian velocity average of $v_eQ_i^*$ at the indicated electron temperatures. Here, Q_i^* refers to the excitation cross section corresponding to the electron speed v_e.) Since the radiative life times for the lower $4s$ states are shorter than those for the upper $4p$ states,[7,8] an equal excitation probability for the two configurations would suffice for generation of population inversion. By assuming values of electron temperature and ion density consistent with those deduced from the experiments of Kitaeva et al.,[19,20] Beigman

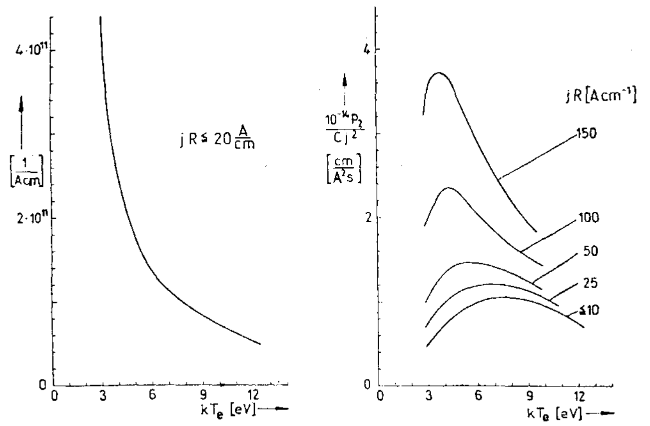

Fig. 2 Scaling relationships obtained by Herziger and Seelig for argon-ion laser: a) normalized electron density n_e/j; and b) normalized upper state pumping rate P_2/C_j^2, as function of electron temperature T_e for various values of jR (reproduced from Ref. 26; courtesy Springer-Verlag, Berlin).

and co-workers[21] have calculated the rates of various $(Ar^+)^*$ configuration excitations and concluded that such excitations by electron impact through the ground state of Ar^+ could, indeed, account for the total laser power output under certain experimental conditions.

The relative contributions of paths 3) and 4) toward excitations of the $4p$ and $4s$ configurations have also been examined by Beigman et al,[21] and more recently by Kitaeva et al,[29] using the same set of calculated values for $\langle v_eQ_i^*\rangle$ and experimentally determined electron temperature and ion density. The general conclusion was that contributions from these paths were relatively minor in comparison with the two-step process 2 under most of the experimental conditions considered.

In regard to the relative importance between path 1 and path 2, no quantitative comparison appeared to have been taken by the aforementioned investigators. The difficulty in making such quantitative comparison was, perhaps, mainly due to uncertainties about the neutral argon atom density under any given set of experimental conditions. Qualitatively speaking, however, it has generally been conceded that path 1 predominates under high electron energy, short-pulse conditions, while path 2 is likely to be more important under normal CW operating conditions at moderate electron temperatures.

V. Quantitative Theories

Development of quantitative theories for noble gas ion lasers clearly requires simultaneous consideration of both the plasma state and the excitation/de-excitation mechanisms. At the very low generation efficiency typical of existing noble gas ion lasers (see section II) where induced emission makes only a minor perturbation to the overall energy balance, the plasma state can be determined from suitable low-pressure gas discharge theories. However, in order to avoid excessive empiricism, such gas discharge theories must be based on fundamental plasma physics and ionization kinetics. While a comprehensive theory satisfying the aforementioned criteria does not yet exist for noble gas ion laser discharges, some recent works relevant to the development of such a theory have been reported by Herziger and Seelig,[26] Zarowin,[30] and by Chen and Lin.[44,45]

In the detailed treatment of Herziger and Seelig,[26] the plasma balance and energy equations for the low-pressure discharge were formulated in accordance with the celebrated work of Tonks and Langmuir.[46] Ionization kinetics was based on Maxwellian averaging of the electron impact ionization rate in which the energy dependence of the ionization cross section for Ar was approximated by a linear rise from the threshold potential. The electrical conductivity relating the current density to electric field strength in the partially ionized argon plasma was based on a linear combination of electron-atom and electron-ion collision probabilities, as first suggested by Lin, Resler, and Kantrowitz.[47] The ion number density n_i was assumed to be identical to the electron number density n_e in the quasi-neutral, singly-ionized plasma. From such formulation, Herziger and Seelig have been able to obtain some very useful scaling relationship between the ion number density and the discharge current density as a function of electron temperature that appeared to agree well with experimental observations[19,20] up to current densities of a few hundred amp/cm^2. By considering electron impact excitation through the ground state of Ar^+ (i.e., path 2 in Section IV) as the predominant excitation process and by assuming that a linear relationship exists between the velocity-averaged excitation probability $\langle v_eQ_i^*\rangle$ and the velocity-averaged ionization probability $\langle v_eQ_{a\rightarrow i}\rangle$, such that $\langle v_eQ_i^*\rangle = C\langle v_eQ_{a\rightarrow i}\rangle$, Herziger and Seelig have further obtained some scaling relationship between the upper state pumping rate P_2 (i.e., rate of excitation of Ar^+ to the $4p$ levels per unit volume per unit time), the current density j, the discharge tube radius R, and the electron temperature T_e. These scaling relationships are reproduced here in Fig. 2. The significance of the latter scaling relationship is that when the electron temperature T_e is transformed into neutral atom number density n_a (or N in Herziger and Seelig's notation) through the Tonks-Langmuir plasma balance equation, as illustrated here in Fig. 3, the optimum value of n_a for maximum pumping rate or laser output power at any discharge current density and tube radius can readily be determined.

As noted by Herziger and Seelig, it is generally difficult to relate the neutral atom density n_a within the electrical discharge to the total filling pressure p. As an approximation, they suggested the relationship

$$p = n_akT_a \tag{3}$$

where T_a is the neutral atom temperature within the discharge to be determined by other considerations. However, such an approximation appeared difficult to justify since the partial pressures from the electrons and ions are generally not negligible under the high current density condition typical of the noble gas ion laser discharge.

Another serious question concerning the adequacy of Herziger and Seelig's formulation,[26] perhaps, can be raised in regard to their extended application of the electrical conductivity (or electron mobility) formula

$$\sigma = e_0n_eb_e = An_ee_0^2/[(m_ekT_e)^{1/2}(n_a\bar{Q}_a + n_e\bar{Q}_i)] \tag{4}$$

into the high current density region. Here, e_0 and m_e denote, respectively, the electronic charge and mass; b_e the electron mobility (i.e., mean drift velocity per unit field

Table 2 Values of velocity-averaged excitation probability in 10^{-10} cm^3/sec to various $(Ar^+)^*$ configurations by electron impact on ground state Ar^+ according to the calculation of Beigman et al. (reproduced from Ref. 21)

Transition	Electron temperature, 10^4°K			
	3	5	8	10
$3p \rightarrow 4p$	0.18	3.0	14	23
$3p \rightarrow 3d$	3.0	44	190	300
$3p \rightarrow 4d$	0.078	2.4	16	28
$3p \rightarrow 5d$	0.0047	0.22	1.8	3.4
$3p \rightarrow 4s$	0.23	2.7	11	17
$3p \rightarrow 5s$	0.003	0.10	0.62	11
$3d \rightarrow 4p$	1900	2000	2000	1900
$4s \rightarrow 4p$	4800	6600	7700	8000

strength); k the Boltzmann constant; A a numerical constant close to unity (taken to be 0.865 in Ref. 26 and 0.532 in Ref. 47); $\bar{Q}_a$ the velocity-averaged momentum transfer cross section for electron-atom collisions; and

$$\bar{Q}_i = [\pi e_0^4/(3kT_e/2)^2] \ln(3kT_e/n_e^{\frac{1}{3}}e_0^2) \tag{5}$$

is the velocity averaged Coulomb cross section for singly-charged ions.[26,47] The above formulas, originally deduced[47] from the plasma conductivity theory of Spitzer and Härm,[48,49] can be expected to be valid only when the electric field is so weak that the mean electron drift velocity $\bar{v}_e$ remains sufficiently small in comparison with the electron sound speed $a_e \equiv (5kT_e/3m_e)^{1/2}$. At high electron drift velocities, the averaged collisional energy between an electron and an ion is given by

$$\bar{\mathcal{E}} = \tfrac{3}{2}kT_e + \tfrac{1}{2}m_e\bar{v}_e^2 = \tfrac{3}{2}kT_e(1 + \tfrac{5}{9}M_e^2) \tag{6}$$

where $M_e \equiv \bar{v}_e/a_e$ is the electron drift Mach number. Thus, the Colulomb cross section $\bar{Q}_i$ should be reduced by a factor like $[1 + (\frac{5}{9})M_e^2]^2$ from that given by Eq. (5) whenever M_e is not negligibly small in comparison with unity. Since the mean speed of the electrons is proportional to $(\bar{\mathcal{E}})^{1/2}$, the mean-free-time between electron-ion collisions at a fixed value of $\bar{Q}_i$ is reduced by a factor $[1 + (\frac{5}{9})M_e^2]^{1/2}$. These elementary considerations then lead one to conclude that at high electron drift velocities, the electrical conductivity for a highly-ionized, singly-charged plasma (i.e., one in which $n_a\bar{Q}_a \ll n_e\bar{Q}_i$) should be greater than that given by the Spitzer and Härm theory

$$\sigma_{\text{S.H.}} = \frac{9A}{4\pi} \cdot \frac{(kT_e)^{3/2}}{e_0^2 m_e^{1/2}} \left[\ln\left(\frac{3kT_e}{n_e^{1/3}e_0^2}\right)\right]^{-1} \tag{7}$$

by a factor like $[1 + (\frac{5}{9})M_e^2]^{3/2}$. This then leads to a divergence of the electrical conductivity with M_e^3 at high electron drift Mach number—a phenomenon called "electron runaway" in plasma physics.

The aforementioned simple anticipation of electron runaway at high drift velocity in a pure Coulomb plasma was well collaborated by the more elaborate theory of Yen,[50] who has carried out the calculation of electrical conductivity over the full range of electron drift Mach number for a fully-ionized, singly-charged plasma with a Maxwellian velocity distribution superimposed on the mean drift velocity $\bar{v}_e$, using the 13-moment method. Yen's theory, plotted here in Fig. 4 in the form of the normalized conductivity $\sigma/\sigma_{\text{S.H.}}$ as a function of M_e, shows considerably more structure than

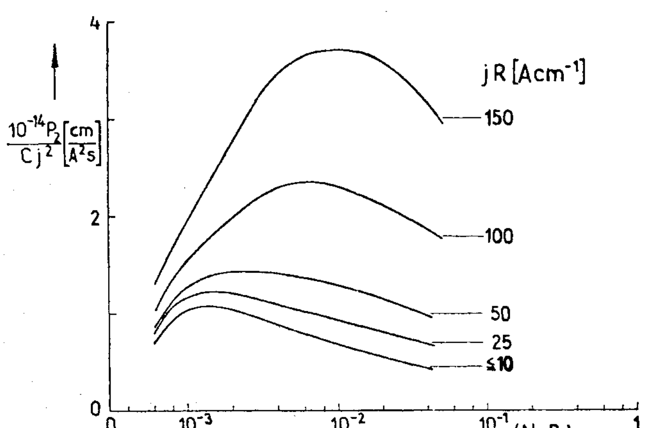

Fig. 3 Scaling relationship obtained by Herziger and Seelig for upper state pumping rate P_2, current density j, tube radius R, and neutral atom number density N (reproduced from Ref. 26, where $N_0 = 3.3 \times 10^{16}$ cm^{-3}, and $R_0 = 1$ cm; courtesy Springer-Verlag, Berlin).

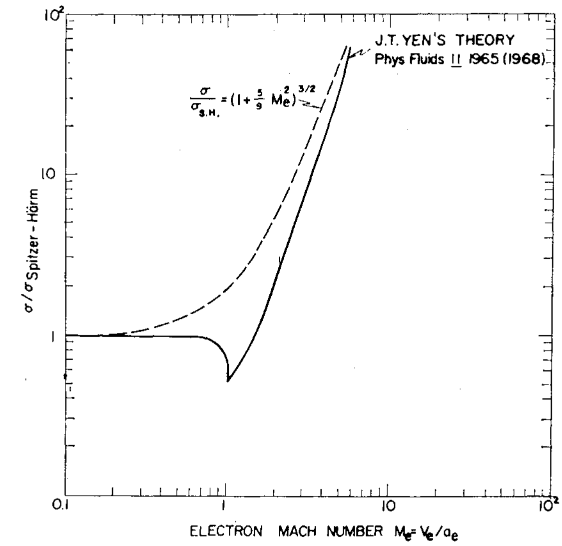

Fig. 4 Electrical conductivity for a fully ionized, singly charged plasma as a function of electron drift Mach number according to the theory of Yen (Ref. 50).

our simple curve $[1 + (\frac{5}{9})M_e^2]^{3/2}$ in the transonic range, i.e., the conductivity appears to dip sharply down near the sonic point, reaching a minimum value of about 0.5 at the sonic point, and then rises again toward the asymtote $(\sigma/\sigma_{\text{S.H.}}) \propto M_e^3$ at high Mach number. These features of Yen's theory seemed to have been observed in the recent experiments of Sakao and Sato.[51]

In a recent paper, Zarowin[30] showed some calculation of the electron drift velocity $\bar{v}_e$ in a partially-ionized argon plasma as a function of electron temperature and the degree of ionization $\alpha \equiv n_e/(n_a + n_e)$, again using Eq. (4) without allowing for the effect of electron drift Mach number on $\bar{Q}_i$ and σ. Essentially, he recovered Spitzer and Härm's formula [Eq. (7)] in the limit of negligible contribution from the electron-neutral collisions (i.e., $n_a\bar{Q}_a \ll n_e\bar{Q}_i$). From this formula, Zarowin proceeded to calculate the electron temperature T_e from values of σ deduced from Kitaeva and co-worker's experiments,[19] and then, using the other limit of Eq. (4) (i.e., $n_a\bar{Q}_a \gg n_e\bar{Q}_i$) and published values for $\bar{Q}_a$, estimated the upper bound for $\bar{v}_e$ and the lower bound for n_e at some typical current densities encountered in argon-ion laser discharges. Aside from the question of consistency, the significance of the upper and lower bounds so obtained for $\bar{v}_e$ and n_e remained doubtful on account of the inadequacy of Eq. (4) at high electron drift velocities, as discussed in the preceding paragraphs.

In the more recent three-temperature formulation reported by Lin and Chen,[44,45] an effort has been made to construct a more general theory of argon-ion laser discharge which may have a wider range of validity than that of Herziger and Seelig's theory.[26] In particular, to remove the uncertainty about the neutral atom number density n_a in the latter theory, Lin and Chen assumed that the total filling pressure p was equal to the sum of the partial pressures of electrons, ions, and neutral atoms for the partially-ionized, quasi-neutral plasma within the discharge tube, such that

$$p = p_e + p_i + p_a = n_ek(T_e + T_i) + n_akT_a \tag{8}$$

In order to allow for the possibility of high electron drift Mach number at high discharge current densities, Yen's conductivity theory[50] was used to relate the discharge current density j and field strength E, such that

$$j = e_0n_e\bar{v}_e = \sigma(T_e,\bar{v}_e)E \tag{9,10}$$

and

$$\sigma(T_e,\bar{v}_e) = Y(M_e)\sigma_{\text{S.H.}}(T_e) \tag{11}$$

where $Y(M_e)$ is the solid curve plotted in Fig. 4, which depends only on the electron drift Mach number $M_e \equiv \bar{v}_e/a_e = (3m_e/5kT_e)^{1/2}\bar{v}_e$. For an electron moving at a peculiar velocity $\mathbf{C}_e$ relative to its mean drift velocity $\mathbf{v}_e$, the resultant collisional velocity with a slowly moving neutral atom (or ion) is essentially $\mathbf{v}_e + \mathbf{C}_e$. Since the total electron impact ionization cross section for a neutral atom $Q_{a\rightarrow i}$ (or excitation cross section for an ion $Q_i{}^*$) depends on the total collisional energy[52]

$$\bar{\mathcal{E}} = \tfrac{1}{2}m_e|\mathbf{v}_e + \mathbf{C}_e|^2 \tag{12}$$

Lin and Chen further allowed for the effect of finite electron drift velocity on the ionization rate such that the total ionization frequency per electron in a plasma of n_a was evaluated according to the integral

$$\begin{aligned}\nu_i &= n_a \int |\mathbf{v}_e + C_e| Q_{a\rightarrow i}(|\mathbf{v}_e + \mathbf{C}_e|) f_e(\mathbf{C}_e,T_e) d\mathbf{C}_e \\ &\equiv n_a \langle v_e Q_{a\rightarrow i}\rangle\end{aligned} \tag{13}$$

where $f_e(\mathbf{C}_e,T_e)$ was the Maxwellian distribution function of electron peculiar velocity[53] at electron temperature T_e. (The justification for the assumption of an isotropic Maxwellian distribution for the electron peculiar velocity was based on the early works of Langmuir and Mott-Smith,[54] and the more recent works of Gabor, Ash and Dracott.[55–57])

By treating all the plasma properties as being uniform over the discharge tube cross section (or, alternatively, treating them as averaged quantities over the radial distributions of ion density and velocity already obtained by Tonks and Langmuir in Ref. 46), Eqs. (8–11), together with the plasma balance equation, the energy balance equation, and the two equations governing the ion temperature and neutral atom temperature, can then be considered as a closed set of eight algebraic equations for the eight unknowns n_e (or n_i), n_a, T_e, T_i, T_a, $\bar{v}_e$, σ, and E. These can readily be solved, at least numerically, for any given set of values for the four externally adjustable discharge parameters p, R, j, and T_w (where T_w is the discharge tube wall temperature).

In the preliminary calculations of Lin and Chen,[44,45] it was found that the ion temperature equation can readily be decoupled from six of the other seven equations since T_i is generally much lower than T_e and hence, contributes little in Eq. (8) and in the overall energy balance equation. The neutral atom temperature T_a, on the other hand, is strongly coupled to both the ion temperature T_i and the wall temperature T_w through charge-exchange and elastic collisions in the "free molecule" type flow. On account of the complicated manner in which the electron temperature T_e and drift velocity $\bar{v}_e$ entered into the conductivity and plasma balance equations, the simple scaling relationships discovered by Herziger and Seelig were generally vitiated at high discharge current densities.

The calculated electron temperature and ion density by Lin and Chen were found to be in good agreement with the experimental data of Labuda et al[24] and Kitaeva et al,[19,20,29] up to current densities of the order of 400 amps/cm^2. (It may be noted that the electron temperature data published by Kitaeva et al were actually obtained indirectly from ion temperature measurements via Kagan and Perel's theory,[58] and hence were subjected to some interpretive uncertainties.) The electron drift Mach number M_e corresponding to these experimental conditions were found to be subsonic but definitely not small in comparison with unity. At higher current densities, the electron drift velocity could become supersonic, and electron "runaway" could occur if other impeding effects, such as momentum loss due to collisions with residual neutral atoms, with the finite (non-Coulomb) core structure of the positive ions, and with the nonsteady plasma sheath at the tube wall,[55–57] etc., were indeed negligible.

From the plasma state determined by the three-temperature formulation just described, Chen and Lin[45] have also made calculations of the collisional excitation-rate-limited power output for CW argon-ion lasers, assuming different excitation mechanisms and using published values of the excitation cross sections. It was found that both the direct process 1 and the two-step process 2 discussed in the preceding section were capable of yielding the very steep current density dependence of the output power as observed in experiments. However, the absolute value of the direct excitation rate based on the measured cross sections of Bennett et al.[18] could account for only a small percentage of the experimentally observed output power density. On the other hand, the absolute value of the two-step excitation rate based on the calculated excitation probabilities by Beigman et al. (Table 2 and Ref. 21) from the ground state of Ar^+ was found to be quite adequate for explaining the experimentally observed argon-ion laser output power density.

VI. Summary

From the foregoing review and discussions, one may arrive at the following general conclusions about the status of our current understanding of noble gas ion lasers:

1) There exists a very large body of empirical information concerning the general characteristics and performance parameters of noble gas ion lasers. Of the different types of noble gas ion lasers, the steady-state (or CW) argon-ion laser appeared to be the one that has been most intensively and extensively studied.

2) The spectroscopy and general excitation mechanisms for noble gas ion lasers in general, and for argon-ion laser in particular, seems to be fairly well understood. However, some of the crucial excitation cross sections, such as those for electron impact excitation from the ground state of Ar^+ to various ionic configurations, have only been calculated from approximate theoretical models.[21] No experimental information seemed to be available about the excitation cross sections to specific multiplet levels from the ground state of Ar^+.

3) Classical gas discharge theories[46,58] appear to be adequate for approximate prediction of most of the plasma properties required for quantitative calculations of laser optical gain and power output at relatively low discharge current densities. In fact, the relatively simple formulation of Herziger and Seelig[26] in accordance with such classical theories, albeit deficient in the high current density regime, appeared to be quite satisfactory for correlation of experimental data and for limited scaling of various laser parameters for CW argon-ion lasers under the conditions of no externally applied magnetic field and modest discharge current densities.[27,31]

4) Extrapolation of existing theories[26,44,45] all indicated that the output power density and generation efficiency should continue to rise in a monotonic manner with increasing current density. Such trend is very significant from the point of view of practical noble gas ion laser development, since low generation efficiency has been the only major weakness in this otherwise ideal form of laser light source. However, it is not yet clear how far would such favorable trend continue. In fact, preliminary calculations by Lin and Chen[44] using normal plasma conduction theories[48,50] showed that the electron drift velocity may become supersonic at discharge current densities corresponding to the upper range of current densities encountered in some recent large-bore high-power ion laser experiments.[27,31] Such high electron drift velocity is likely to cause electrostatic or other modes of plasma instability,[59–62] which tends to destroy the smooth running condition of the laser. The onset of such plasma instability at very high current densities has indeed been qualitatively suggested by the noisiness of the laser output observed in some early pulsed argon-ion laser experiments.[7,63]

5) Multiple ionization may also be expected to play some role in determining the plasma state in the high current

density limit, but its quantitative effects on noble gas ion laser performance have not yet been assessed.

6) Most recent scaling relationships obtained by Chen[64] in accordance with the three-temperature formulation[44] indicate that the excitation-rate-limited power output per unit length of a CW laser should scale with the square of pR, the product of filling gas pressure and discharge tube radius. This is in the direction of favoring large-bore tubes for power generation, which is in agreement with the earlier observation of Herziger, Seelig et al.[27,31] However, deinversion due to trapping of the resonance line radiation (e.g., the 720 Å line connecting the 4s state and the Ar^+ ion ground state depicted in Fig. 1) at large values of pR is expected to set some practical limit on such scaling relationships. The resonance trapping limit in a four-level system typical of noble gas-ion lasers has been qualitatively discussed by Bennett,[63] but no quantitative analysis has yet been carried out.

7) Additional complications, such as the presence of externally applied magnetic fields,[17,24] the effect of "gas pumping"[24,65] on the pressure matching condition [Eq. (8)], etc., must also be taken into account in any detailed treatment of the laser discharge problem. Quantitative treatment of these additional complications is still very much lacking in the literature.

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