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THE NITROGEN ION LASER

by

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The characterization and thermal scaling of the nitrogen ion laser pumped by charge transfer from He_3^+ is reported in this work. Intense laser emission in the violet at 427 nm has been observed and found to have a linewidth less than 0.3 Å. The pumping ions, He_2^+ and He_3^+, were produced by discharge of a fast-pulsed electron beam gun, APEX-1, into several atmospheres of a mixture of helium and nitrogen. Excitation current densities ranged from 1.1 to 2.5 KA/cm^2 at 1 MV over a 1 x 10 cm transverse geometry. Under these conditions, the

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efficiency of the emission of 427 nm laser radiation was found to be proportional to the total pressure raised to the 1.2 power. Efficiencies of 1.6 relative to the energy lost by the electron beam in the radiating volume have been achieved in volumes of 16 cc at room temperature. Outputs of 35 mJ have been obtained from the 16 cc working volume at 30 atm pressure under these conditions. Thermal scaling of the laser has been investigated and a strong inverse dependence of laser output on gas temperature was observed. At -20°C the output was found to increase to 80 mJ from the 16 cc. volume containing helium at a density giving a pressure of 35 atm. at room temperature. This corresponded to an output efficiency of 3% relative to the energy deposited by the electron beam. Quasi-cw operation was achieved under these conditions suggesting that much longer output pulses might be obtained with an e-beam pulse of greater duration. A peak power density of 320 MW/liter was achieved and intracavity circulating powers of over 1.2 GW/liter at intensities of 20 MW/cm^2 gave no evidence of either bottlenecking or photoionization of any of the species important to the kinetic chain pumping the laser.

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I. TECHNICAL REPORT SUMMARY

The objective of the research pursued under this contract is to continue the basic studies necessary to verify the feasibility of developing an electron beam excited helium plasma into a collisionally pumped laser emitting in the visible or near UV with 5-20% efficiency. Currently accepted theory[1,2,3,4] supported by the experimental results obtained to date indicate this to be a realistic goal. In fact, recent theoretical refinements[5] have led to increased, not decreased, estimates of photon yields and scaling studies tend to confirm this. All experimental parameterizations[6,7] of the helium-nitrogen, charge transfer pumping mechanism appear to be increasing toward an asymptote conforming to overall objectives. The developing kinetic model has benefitted from the characterization during the current reporting period of new component steps. It has reached a level of sophistication sufficient to predict the important phenomenon of thermal scaling in advance of its experimental observation.[8] Exploitation of this thermal scaling effect has raised peak rates of photon emission to 6.5 x 10^{20} photons/ℓ/sec, a value of photon intensity over four times greater than the best reported[9] for KrF excimer lasers. This "best value" of intensity emitted from the helium nitrogen laser corresponds to a peak power density of 320 MW/liter at an efficiency of 3.2%. Intracavity circulating powers have reached a peak of over 1.2GW/liter at an intensity greater than 20MW/cm^2 at 4278Å with no evidence of either bottlenecking or photoionization of any of the species important to the kinetic chain. During the current reporting period four-level operation[10] has been achieved and best values of power efficiencies (output/input) have been shown to be essentially constant at 3.2% for the duration of the pumping pulse. Achievements to date are approaching the overall objective of 10% and the limit on future improvement has not yet been found experimentally.

Even now, a scale-up at currently demonstrated levels of efficiency would yield 100KW average output power at 4278Å from a directly pumped e-beam laser operating with a 0.2 coul pulse at 100 Hz repetition rate discharging into a 100 liters of working volume. In such a system beam intensities could be kept below the foil survival limits for titanium. At present an efficiency of 3% could be projected but the developing kinetic model indicates that the strong dependence of efficiency on pressure and gas temperature will finally yield demonstrated efficiencies closely approaching the theoretical limit of 10%. While such projections are, of course, speculative, these are the implications of the most recent successes with the thermal scaling of the nitrogen ion laser discussed in the following material.

While it should be realized that the nitrogen ion laser is only the first example of the new class of e-beam charge-transfer lasers,[11] and that other similar systems offer the possibilities of even higher efficiencies and broader selections of output wavelengths, results observed to date for the emission of 4278Å laser radiation point to the nitrogen ion laser as a device of considerable significance and clearly confirm the importance of charge transfer reactions as laser pumping mechanisms.

II. KINETIC STUDIES

The concept of the collisional pumping of ion transitions in e-beam plasmas was first proposed by Collins, et al.,[11] in the course of work under this and a previous ARPA contract. The first example to be realized in this wholly new class of e-beam lasers was the nitrogen ion laser[12] pumped by charge transfer from He_2^+ as announced in 1974.

The continued successes[6] of the nitrogen ion laser has emphasized the importance of continued research on e-beam lasers pumped by these collisional transfer mechanisms of charge transfer, Penning ionization, and recombination. It appears these mechanisms represent the most efficient means of exploiting, for the production of visible laser radiation, the ionization deposited in a high pressure gas by an intense electron beam. Since over 80% of the energy of a relativistic electron beam can be stored either as ionization or metastables in the volume of high pressure helium, these elementary mechanisms for using this stored energy to produce an inversion of population makes possible high overall radiative efficiencies. A value approaching that of the absolute quantum efficiency of the transition can be expected. In the visible wavelength region, this would mean efficiencies between 5 and 20% provided the plasma constituents are successfully arranged to allow for the domination of the desired reaction channel.

The recent successes reported[6,8] for the helium-nitrogen charge transfer laser would seem to justify all of this optimism originally associated with the collisional pumping of ion lasers in helium. When this pumping process was discussed in the original proposal it was supposed that "an energy density of 5J/liter would be available to the lasing transition...so that peak powers of the order of a gigawatt per liter could be reasonably projected...." Achievements have clearly approached this goal and the limit on further improvement has not yet been found experimentally. Such agreement is perhaps more notable for the fact that the performance actually observed benefitted greatly from elementary kinetic processes which were completely unknown at the time of the original proposal. In fact, the primary problem in the development of efficient, high pressure gas lasers has consistently been this paucity of information about the elementary kinetic processes dominating this relatively new plasma environment. It is perhaps an equally important consequence of the work performed during this current reporting period that new insights have been obtained as well as some quantitative modelling, of previously unobserved kinetic processes of great importance in high pressure plasmas. These "new" processes[13] include termolecular charge transfer reactions leading to non-associative product channels, such as,

$$He_2^+ + N_2 + He \rightarrow N_2^+(B^2\Sigma_u) + 3He \quad , \qquad (1)$$

and capture-autoionization sequences for the quenching of the vibrational excitation of molecular ions,

$$N_2^{+}(v=1) + e \rightarrow N_2^{**}(v=1) \quad , \qquad (2a)$$

$$N_2^{**}(v=1) \rightarrow N_2^{+}(v=0) + e \quad , \qquad (2b)$$

where the double asterisk denotes an autoionizing molecular state. Both processes have now been recognized to be of critical importance to the high efficiency found for the helium-nitrogen charge transfer laser. In fact the latter sequence (2a) and (2b) has led to 4-level operation[10] of the helium-nitrogen laser on the 4278Å line indicating that a visible laser of exceptionally high average power could be pumped by charge transfer if development along those lines were funded.

Actually it is this operation as a 4-level laser that is the requisite condition for a particular laser system to be scalable in a practical sense. For such operation a laser medium must have an energy storage level, which is the level initially excited, two working levels, optically connected, and a final fourth level into which the population of the lower working level can be exothermically dumped following the stimulated emission.[10] It appears that of the e-beam lasers of current interest only the XeF and KrF excimer lasers together with the $He:N_2$ charge transfer laser are capable of 4-level operation. Because of the relatively high energy of the Coulomb potential of a molecule involving an inert gas ion above the repulsive ground state of the excimer system, it is likely that operation of inert gas excimer lasers will be confined to shorter wavelengths and the charge transfer lasers offers a complement valuable for operation at visible wavelengths.

Once 4-level operation has been established the limiting theoretical efficiencies for the direct e-beam pumping of collisional transfer lasers are quite similar to those of excimer lasers. Assuming one photon is ultimately extracted for each quantum stored in the upper level initially pumped and using the known values[14] of energy cost per quanta stored gives the "universal" laser model for e-beam excitation of inert gases shown in Fig. 1. Limiting efficiencies for KrF are shown together with those for likely charge transfer pairs. As can be seen, the limiting efficiency is primarily a function of operating wavelength. Actual efficiency depends upon the probability of achieving one photon per quanta originally stored and is difficult to estimate accurately from the paucity of data concerning elementary kinetic processes.

Since the actual laser performance is more a consequence of the probability of obtaining one photon per ion than it is a result of particularly favorable choice of gas pairs, it appears initially that charge transfer offers considerable advantages over other laser pumping mechanisms because of the large cross sections, 10^{-14} cm^2, characteristic of such processes. These values lead to reaction rates which are at least an order of magnitude larger than those characteristic of most

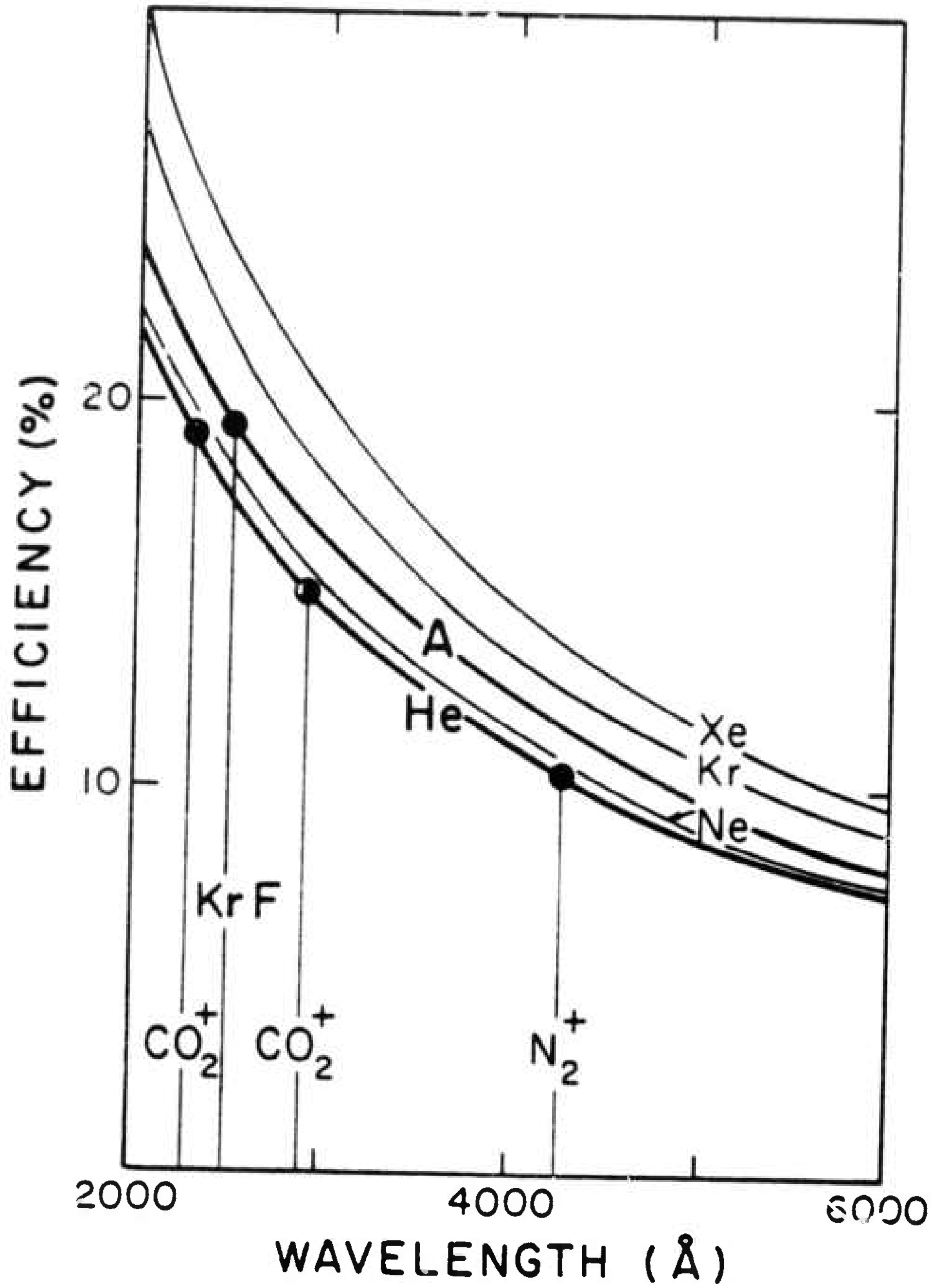

Figure 1: Graph showing the limiting efficiencies as functions of wavelength for excimer and charge transfer lasers operating in inert gases. Filled circles represent systems of current interest.

excitation transfer sequences involving neutral atomic and molecular species. As a consequence, the laser pumping reactions can be readily arranged to be the dominant process for loss of the ionization deposited by the electron beam. This can be done with relatively small concentrations of the gas to be excited which, in turn, means that chemical quenching of the final excited state population should be virtually negligible, as seems to be the case in the nitrogen ion laser.[7] For example, only 30 Torr represents the optimum concentration of N_2 in 20 atm. of helium.

When the helium-nitrogen charge transfer laser was first proposed by Collins, et al.,[11] it was expected that the inverting transition in N_2^+ would be pumped by the resonant transfer of energy from the diatomic helium ion, He_2^+ according to the well-known[15,16] bimolecular charge transfer reaction,

$$He_2^+ + N_2 \rightarrow N_2^+(B^2\Sigma_u) + 2He \quad . \qquad (3)$$

Laser output was first achieved in high pressure plasmas excited by electron beam discharges[12] and tended to confirm this, as did subsequent studies of scaling.[7] However, the parameterization of the efficiency of the laser output proved difficult to reconcile with the simple kinetic model first advanced. Data measured over a wide range of experimental parameters were found to be closely approximated by the empirical expression for the efficiency

$$\varepsilon = 6.5\%(P/100)^{1.2} \quad , \quad \text{(room temperature)} \qquad (4)$$

where P was the total gas pressure in atmospheres and 6.5% was the early theoretical limit on efficiency.[7] The validity of (4) was confirmed over a range of efficiencies varying from 0.3 to 1.6% as a consequence of changes in total pressure, fractional composition and mirror reflectivity. However, the data obtained during the current reporting period from the measurement of the rate coefficients of the elementary reactions involved in the pumping sequence suggested that additional pumping steps were involved. Details of these results are presented in the most recent manuscript submitted for publication,[13] together with a summary of rate coefficients obtained. They suggested, a posteriori, the importance of termolecular charge transfer reactions in the reaction chain.

Originally, because of the difficulties in establishing the predominance in a low-pressure ion source of the weakly bound molecular ions of the inert gases, charge transfer reactions involving these species proved less tractable to the generally successful flowing afterglow techniques which had served to measure most of the bimolecular charge-transfer rates important to aeronomy and related studies of planetary atmospheres.[16] Finally, in 1970 the ESSA group succeeded in measuring the bimolecular charge transfer rates for He_2^+, Ne_2^+ and Ar_2^+ reacting with Ne, Ar, Kr, NO, O_2, CO, N_2, and CO_2 under cryogenic conditions.[15]

At temperatures up to 200^{o}K the necessary predominance of the molecular ions could be established and rates generally approaching the theoretical limit were found. Conversely, reaction rate coefficients could not be determined at room temperatures. Since a strong dependence on temperature was not expected, a priori, at least for the highly probable exothermic reactions, the 200^{o}K values seemed sufficient for existing needs.

The current attention to these e-beam lasers has apparently stimulated a further interest in these ion-molecule reactions involving inert gas molecules. Partial cross-sections for charge transfer from He_2^+ into specific output channels of the reaction have been recently determined in an atomic beam apparatus.[17] Nevertheless, attempts to model quantitatively the kinetic sequences pumping high pressure lasers suffer from the paucity of rate coefficient data appropriate to atmospheric pressures. Efforts to describe the strong dependence given by (4) for laser outputs on gas pressure do not succeed when models are based upon extrapolations of available low pressure data and virtually all the rate coefficients appearing in the literature have been derived from data obtained at pressures below a few torr. The notable exception has been the work of Bourène and Le Calvé[18] which provided the first suggestion that reaction rate coefficients measured at high pressures can be significantly different from those derived from low pressure data.

Because of the restricted range of operating pressures available to the traditional flowing afterglow experiments, they were necessarily insensitive to the effects of termolecular reaction channels for which the three-body rate coefficient was less than about $3 \times 10^{-28} cm^6/sec$ whenever there was a parallel bimolecular channel having a cross-section near the Langevin limit. Such a generous limit is not helpful at atmospheric pressures where much smaller termolecular rates would still dominate.

Work supported by this contract has shown what are believed to be the first measurements of termolecular charge transfer reactions into non-associative product channels. These reactions are the termolecular analogs to the well-known[15] bimolecular charge transfer reactions involving helium molecular ions. In a sense they form parallel reaction channels, favored at high pressures, connecting the same reactant and product populations as the bimolecular ion-molecule reactions dominant at low pressures. For example, reaction (1) would be the termolecular analog of (3) and with a rate coefficient of only $10^{-29} cm^3/sec$ it would dominate in high pressure lasers operating at several atmospheres while being completely negligible in the conventional low pressure ion-molecule experiments. Evidentally, the possibility of termolecular reaction channels paralleling the bimolecular ones has not received much attention in the past and it appears that termolecular channels have been investigated[16] only for associative ion-molecule reactions for which the bimolecular analog is extremely unlikely.

In these initial measurements[13] performed during the current reporting period ion destruction frequencies have been determined as functions of helium pressure over the range from 300 to 1500 torr and as functions of the partial pressure of reactant from 50 to 400μ. Typical data are shown in Figure 2 which shows the time-dependence of the He_2^+ population and in Figure 3 which shows the dependence on pressure of the logarithmic destruction frequencies obtained from such population data. From this type of data pressure-dependent rate coefficients have been extracted and subsequently resolved into contributions from bimolecular and termolecular components for reactions of He_2^+ with Ne, N_2, CO, CO_2 and CH_4. The bimolecular components have been found to agree with the ESSA results[15] to within experimental error and in some cases the values determined here represent an improvement in precision. Values measured in this work for the bimolecular rate coefficients for charge transfer from He_2^+ are summarized in Table 1 together with previously reported values.

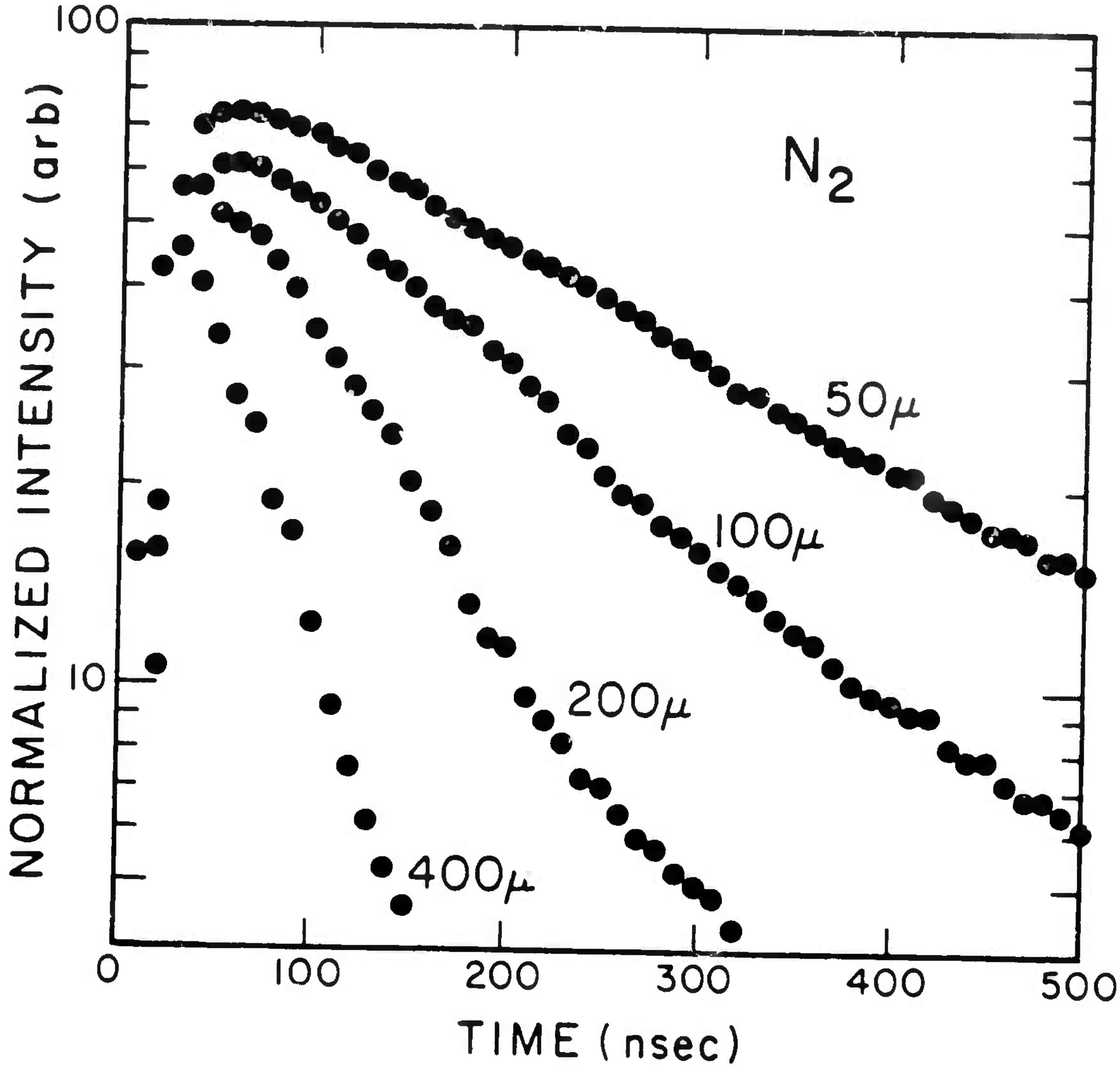

Figure 2: Graphs of the transient intensity at 4278Å measured in the afterglow of an intense electron beam discharge into 830 torr of helium containing the indicated partial pressures of nitrogen. The data have been normalized for presentation.

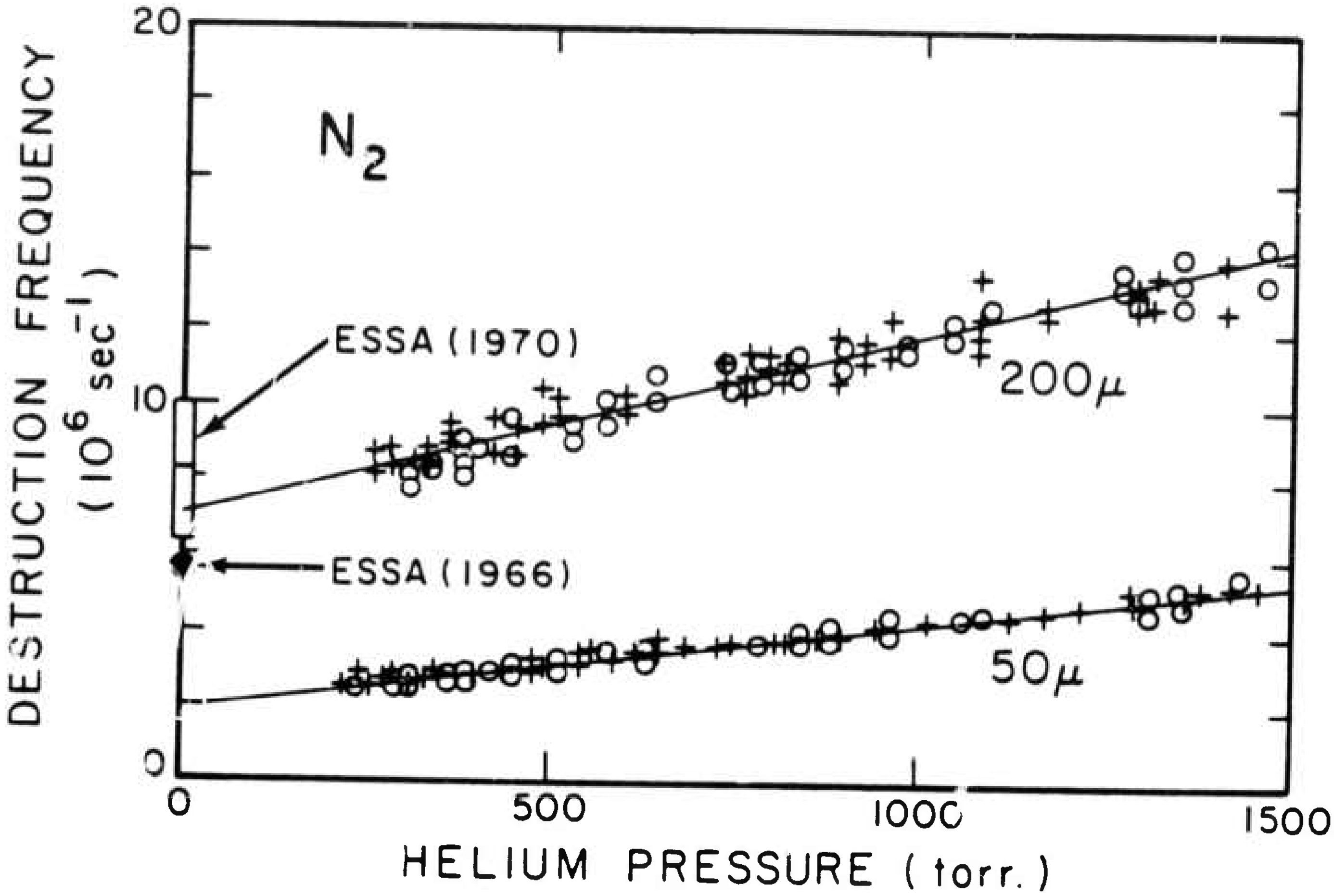

Figure 3: Graph of the measured destruction frequencies, ν, of the transient intensities of transitions to two different lower states from product $N_2^+(B\,^2\Sigma_u)$ molecules resulting from the charge transfer from He_2^+ as functions of helium pressure in the afterglow of an intense electron beam discharge. The type of symbol indicates the corresponding wavelength; (O) 4278Å; (+) 3914Å. Partial pressures of nitrogen are indicated and comparison values of ν computed from previously reported bimolecular rate coefficients are shown together with the reported experimental uncertainty where available.

Table 1

Summary of bimolecular charge transfer rate coefficients for reactions of He_2^+ measured in this work together with comparative values from the literature. Units are 10^{-10} cm^3/sec.

Reactant	This Work	ESSA[15]	BLC[18]	Villarejo[19]	Oskam[20]
N_2	11	13	6	2.2	
CO	11	14	5.3		
CO_2	16	18	13		
CH_4	5		5.5		
Ne	5	6	1.5	1.2	1.5

More important, however, is the identification and measurement of rate coefficients for the termolecular charge transfer reactions of the form,

$$He_2^+ + He + X \rightarrow \text{Products.} \tag{5}$$

The sensitivity of the method has been sufficient to detect termolecular components as small as $2 \times 10^{-30} cm^6/sec$ and values were found to range widely from this threshold value for Ne to $67 \times 10^{-30} cm^6/sec$ for CO_2. The size of these termolecular rates not only serves to explain the anomalous pressure dependence of the output from charge transfer lasers but also suggest the general importance of three-body ion-molecule reactions in higher pressure plasmas. Values obtained in this work are found in the following table.

Table 2

Summary of termolecular charge transfer rate coefficients measured in this work for reactions of He_2^+ with the reactants listed. Units are $10^{-30} cm^6/sec$.

Reactant	Rate Coefficient
N_2	16 ± 3
CO	36 ± 8
CO_2	67 ± 12
CH_4	5 ± 2
Ne	2 ± 2

When rapid, these termolecular reactions offer parallel kinetic paths for the destruction of the He_2^+ ions in a helium plasma. The destruction frequency expected, then, in a given plasma must be expected to contain contributions from both bimolecular and termolecular reactions. As a result, the primary reaction paths for molecular helium ions can differ widely in high and low pressure plasmas and afterglows. This suggests the need to represent the pumping sequence of the helium nitrogen laser more generally as follows:

ION FORMATION,

$$e + He \rightarrow He^{+} + e \quad , \qquad (6a)$$

$$He^{+} + 2He \rightarrow He_2^{+} + He \quad , \qquad (6b)$$

$$He_n^{+} + 2He \rightleftarrows He_{n+1}^{+} + He \quad , \qquad (6c)$$

where $n \geq 2$.

CHARGE TRANSFER PUMPING,

$$He_n^{+} + N_2 \rightarrow N_2^{+}(B^2\Sigma_u) + nHe \quad , \qquad (7a)$$

$$He_n^{+} + He + N_2 \rightarrow N_2^{+}(B^2\Sigma_u) + (n+1)He \quad , \qquad (7b)$$

where $n \geq 2$.

STIMULATED EMISSION,

$$N_2^{+}(B^2\Sigma_u) + h\nu \rightarrow N_2^{+}(X^2\Sigma_g)_{v=1} + 2h\nu \quad , \qquad (8)$$

CAPTURE-AUTOIONIZATION,

$$N_2^{+}(X^2\Sigma_g)_{v=1} + e \rightarrow N_2^{**}(v=1) \quad , \qquad (9a)$$

$$N_2^{**}(v=1) \rightarrow N_2^{+}(X^2\Sigma_g)_{v=0} + e \quad , \qquad (9b)$$

where the double asterisk indicates an autoionizing level.

Having this basic kinetic data available makes the construction of a comprehensive kinetic model relatively straightforward. Provided the partial pressure of minority constituent is not too great for reactions (6b) and (6c) to go to completion, the ionization will convert from He^+ to He_2^+ and He_3^+, thus making available the charge transfer reactions (7a) and (7b).

Determination of the actual output energy available in the resulting population of the $N_2^+(B^+\Sigma_u)$ state requires knowledge of the branching ratio between the possible output channels from the transfer reaction. It has been discussed in previous contract reports[6] that the occurrence of gain in the (0,0) vibrational component of the $N_2^+(B^2\Sigma_u \rightarrow X^2\Sigma_g)$ electronic transition requires at the minimum, 50% of the N_2^+ product population be pumped into the upper $B^2\Sigma_u$ state. The strength of the laser oscillations actually observed in this transition indicates, rather, that over 75-80% and possible ∿100%, of the product ions are in the upper $B^2\Sigma$ state. Thus it is reasonable to approximate the energy available in the product population as being potentially one output photon per molecular ion formed by (6b) and (6c).

In the final step, the capture-autoionization sequence, (9a) and (9b), unique to molecular ions, recently has been shown[6,8] to lead to 4-level operation, at least on the 4278 Å line. This has made possible quasi-cw operation of the B → X electron transition of N_2^+ at 3% efficiency.

Finally, it is important to recognize that other potentially limiting mechanisms such as chemical quenching are minimized in the charge transfer scheme described here. In this fact lies the primary advantage in this mechanism. It results largely from the large cross sections, 10^{-14} cm^2, characteristic of such processes. These values for the charge transfer process are at least an order of magnitude larger than those characteristic of most other excitation transfer reactions involving neutral atomic and molecular species. This means much smaller concentrations of the gas to be excited can be used which, in turn, means that chemical quenching of the final excited state population should be virtually negligible, as seems to be the case in the nitrogen ion laser actually realized.

III. CHARGE TRANSFER LASER PHENOMENOLOGY

As mentioned in the previous section, it was first demonstrated almost two years ago[11] that resonant charge transfer held considerable promise as a potential laser pumping mechanism. Direct measurements of gain obtained with a tunable dye laser were reported at that time. Lasing was reported a few months later.[12] Outputs were small, 9 KW, but efficiencies were around 1.8%. However, excited volumes were quite small, 0.63 cm^3, and the first concern upon receipt of the electron beam source at our facility was to determine the scalability of this result to larger volumes more nearly corresponding to the use of the entire e-beam cross section.

The subsequent series of experiments reported[7] served to raise outputs and lower efficiencies slightly to 1.6%. Complicated effects resulting from the non-linearity of the reaction sequence tended to confuse the early phenomenology.

This report concerns the resolution of many of these effects. As has already been discussed in the preceding technical report, output variation with mirror reflectivity, gas composition, and problems of energy deposition from the beam have been considered. These will be reviewed here only briefly for convenience. The introduction of the termolecular charge transfer reactions into the pumping sequence during the current reporting period has served to resolve the main outstanding problem of the strongly non-linear dependence of output power on pressure.

Finally, during the current reporting period, quasi cw operation was achieved under a variety of operating conditions, thus pointing the way toward much longer output pulses and hence, higher pulse energies. These results are discussed in the following material and the implications in Section IV.

The excitation of the charge transfer plasmas used throughout this research was produced by the APEX-1 electron beam device acquired under this contract. It was constructed by Systems, Science & Software of Hayward, California and is a fast pulse, sheet beam gun emitting 100 KA pulses of 1 MeV with a 1 x 10 cm transverse cross section. As used currently, pulse shapes are nearly triangular with 20 nanosecond FWHM and optionally with the fall time controlled by a shorting electrode. During the experimental series reported here, the anode-cathode spacing in the output diode was increased to give a larger diode impedance and consequently peak currents between 10 and 20 KA were obtained. Larger currents were not attempted as the operation under those conditions was rendered difficult by problems of foil survivability.

The afterglow chamber used in these experiments was the ELAC-1 device described previously.[6] It consisted of a laser cavity mounted to a foil support assembly and contained in a cylindrical high pressure vessel with axis of symmetry along the axis of beam propagation. The assembly was constructed of UHV-grade stainless steel with windows and gas handling connections made with Varian-type copper shear seals. The laser cavity consisted of a pair of dielectric mirrors which were mounted to allow angular alignment, spaced with 14 cm invar rods, and contained in the pressure vessel with the optical axis coincident with the longer 10 cm transverse dimension of the e-beam.

In operation the system was pressurized with 1 to 35 atmospheres of a mixture of helium and nitrogen. Useful partial pressures of nitrogen ranged from 2 to 120 Torr. Excitation was provided by the electron beam from APEX entering through a supported, 0.002-in. thick titanium foil window and propagating in a direction perpendicular to the optical axis.

Three laser lines have been excited in mixtures of helium and nitrogen pumped by charge transfer from He_2^+ and He_3^+. Each corresponds to transitions from the same upper vibration state, v = 0, of the $B^2\Sigma_u^+$ electronic state to different lower vibration states of the $X^2\Sigma_g^+$ electronic state of the N_2^+ molecular ion. The three lines and their respective vibrational transitions are: the 3914 Å (0,0), the 4278 Å (0,1), and the 4709 Å (0,2). With the proper mirror set, each has been excited individually. The most work has been done on the (0,1) transition at 4278 Å, but since each has the same upper state, those same results should be roughly characteristic of all with the exception of the (0,0) transition which self-terminates at very early times.

An initial examination of the raw data relating pulse energy to e-beam deposition did not reveal a trend suggesting the nature of the dependence of output on the various diverse experimental parameters. In fact, a highly degenerate system was found for which the same output was achieved from quite different experimental arrangements. Although the plane-parallel optical cavity generally appears, the most attractive in terms of analysis, a priori, in fact this is only the case for CW oscillation after stable cavity modes have developed. The transient response of such cavities to self-excitation is quite complex. This problem was considered in detail[21] during the previous reporting period and an analytical procedure was developed to describe the time-dependent growth of the plasma volume interacting with the cavity fields. A ray tracing program was written to deconvolute the time-dependent laser intensity observed for a given measured average beam divergence in order to obtain the average field-plasma interaction volume and the fraction of the energy extracted from the plasma emitted into the output beam in comparison with that "walking off" the mirrors.

As reported previously[21] this analysis served to show that all of the available energy was being extracted from the plasma by the fields even at relatively low intracavity powers and that the only effect of the varying mirror coefficients was to determine the rate at which that energy was extracted and whether it was routed into the transmitted beam or walked off the mirrors. This was a very important conclusion because it implied competing losses were negligible.

To confirm further the concept that the total energy extracted was independent of mirror parameters, an effort was made to obtain a cavity geometry with low walk-off. Though the unfolding program was only valid for plane parallel cavities, it could be reasonably assumed that for hemispheric cavities with large angular aperture compared to the output beam divergence, ray stability existed for an appreciable region around the cavity axis. For such cavities walk-off could be expected to be negligible in comparison with plane-parallel cavities. For the same plasma conditions, then it could be expected that the energy emitted from a hemispheric cavity would roughly equal the total extracted in either and this was found to be the case experimentally. Thus, the deconvolution of the laser pulses was found to reduce the multivariate dependence of all the data obtained to date from plane cavities to a form dependent upon a single parameter, the pressure, and to yield a value in agreement with the output measured from hemispheric cavities, assumed to be lossless.

To determine the efficiency of the extraction of output energy the deposition of electron beam energy into the laser cavity must be accurately calculated. An essential factor in understanding of the energy deposition from the beam lies in the fact that scattering and stopping power do not have the same dependence[16] on atomic number, Z. As a consequence, it is possible to have a situation in which a considerable fraction of beam energy is stopped without appreciable scattering or conversely that the scattering is so great that the simple approximation of the product of the stopping power and penetration depth seriously underestimates energy deposition, even for small fractional losses of beam energy.

The almost singular case of the light inert gases excited at beam currents below 20KA falls into the first category, and simplifying assumptions exist which render the problem tractable. Subject to limitations on the product of gas density and beam penetration depth, discussed in previous work,[21,22] the problem can be resolved into that of the differential energy loss in the gas of β-particles in a beam, the morphology of which is completely determined by the foil window through which the beam has entered. For a titanium foil, .002" thick, the following bounds on the domain of gas transparency were obtained,

$$PX\ (\text{Helium}) \leq 273\ \text{atm. cm} \tag{10a}$$

$$PX\ (\text{Argon}) \leq 4.3\ \text{atm. cm}\quad , \tag{10b}$$

where P is the gas pressure in atmospheres and X is the penetration depth of the beam into the gas.

It can be seen from these results that, whereas the simplifying assumptions break down for argon (Z=18) at an inch of penetration at two atmospheres, they remain valid in helium (Z=2) over the entire span of parameters required for practical operation of a small test laser device. For example, at 30 atmospheres pressure, the simplified model is valid to at least 9.1 cm depth of penetration which is sufficient to describe about a half liter volume excited by a 1 x 10 cm electron beam of divergence characteristic of transmission through a .002" titanium foil window at 1 MeV for currents less than 20 KA. At higher currents, the "drag e.m.f."[23] resulting from the return currents should be considered.

A complete analysis for helium including the dependence of stopping power and foil scattering on time-dependent beam energy has been presented previously.[21,22] It has lead to an average power deposition constant of 17.3 Megawatts/ℓ/atm/KA on the leading and falling edge of the pulse and a value of 18.05 MW/ℓ/atm/KA on the plateau. However, in view of the uncertain detail of the time-dependence of the beam such analysis is excessively tedious. A re-examination of the beam scattering as evidenced by burn patterns on plastic targets has led to an equivalent but simpler expression for the average power disposition of

$$17.2\ \text{Megawatts}/\ell/\text{atm}/\text{KA} \quad , \qquad (11)$$

on the beam axis at the depth of penetration of the cavity center. This value was used for the calculation of efficiencies throughout the remainder of the work reported here.

The actual values of current density in the electron beam were measured with a calibrated Faraday cup which replaced the pressure vessel and laser cavity. Particular attention was paid to relative timing and cable lengths so that the temporal relation between beam current and laser output could be determined subsequently. Output from the Faraday cup was directly recorded with a 519 oscilloscope. Laser performance data has now been collected over a broad span of experimental parameters. Total pressures have ranged from 1 to 35 atmospheres with partial pressures of nitrogen varying from 2 to 120 Torr. As discussed immediately above, the variation of cavity constants affected the rate of energy extraction from the e-beam plasma, but had little effect on its total. To within rather unrestrictive limits the deconvolution program was able to reduce the parameterization of the laser performance to dependence upon a single variable, the total pressure. The limits bounding this parameterization are primarily a consequence of two effects.

1) If the ratio of nitrogen to helium is so excessive that reactions (6b) and (6c) cannot go to completion, laser output will be drastically reduced or prevented altogether.

2) If the combination of mirror loss and gas composition is such as to delay the onset of lasing until beam current is beginning to decrease at the end of the e-beam pulse, outputs will again be reduced or terminated.

Evidently, in this case, the excited state chemistry is altered by the termination of the beam and competing processes such as recombination with the cooling electrons become important.

Except for data obtained under those conditions, most of the data could be reconciled with a very simple parameterization of the total energy extracted from some standard plasma volume.

The largest laser outputs found at 4278Å in the raw data were from the hemispheric cavity having 27% transparency and the average volume appropriate to that data was 16.2 cm^3. This was chosen as the "standard volume." Since walk-off was assumed zero for the hemispheric cavities, the largest laser outputs (from the 20 - 35 atm. data) simply correspond to unscaled measurements of the total energy emitted into the laser output beam. For the purposes of parameterization, other measurements were scaled to obtain the total energy extracted from a 16.2 cm^3 volume of the plasma, so that

$$E_x = (1 + \overline{W}) \times (16.2/\overline{V}) \times E_e \quad , \qquad (12)$$

where E_e is the energy emitted by the cavity into the laser beam, E_x is the total energy extracted from the plasma by the fields, $\overline{V}$ is the average volume from which it was extracted, and $\overline{W}$, the average ratio of energy walking-off the mirrors to that emitted into the beam. As mentioned above, for hemispheric cavities, $\overline{W}$ was assumed to be zero.

The resulting summary of measurements is presented in Figure 4. The total energy extracted from the 16.2 cm^3 of the charge transfer plasma is plotted as a function of total gas pressure. Data all corresponds to excitation at the level of 76 mJ/atm by a standard discharge pulse containing 275 μ coul of integrated current. The earlier attempts[22] to empirically fit the data had noted it grouped more closely around a line of slope 2.2. Since the energy input deposited by the beam varied linearly with pressure, the consequent efficiency of energy extraction could be modeled to vary with the 1.2 power of the pressure. An extrapolation of the fit to the data appeared to intersect the theoretical limit around 100 atm. so that the efficiency was conveniently expressed as

$$E = 6.5\% \; (P/100)^{1.2} \quad . \qquad (13)$$

The best output at room temperature was found to be 36 mJ at 28.7 atm. which corresponded to an efficiency of 1.6%.

As mentioned previously[7,21] the Franck-Condon factors for the (0,0) and (0,2) transitions between the same electronic states are sufficiently favorable that the thresholds could be individually attained with the proper mirror sets.

The 3914 Å component was found to self-terminate in about 2 nanoseconds and gave a measure of the time required for the lower laser state to "fill". Since the lower state of the 4278 Å differs only in

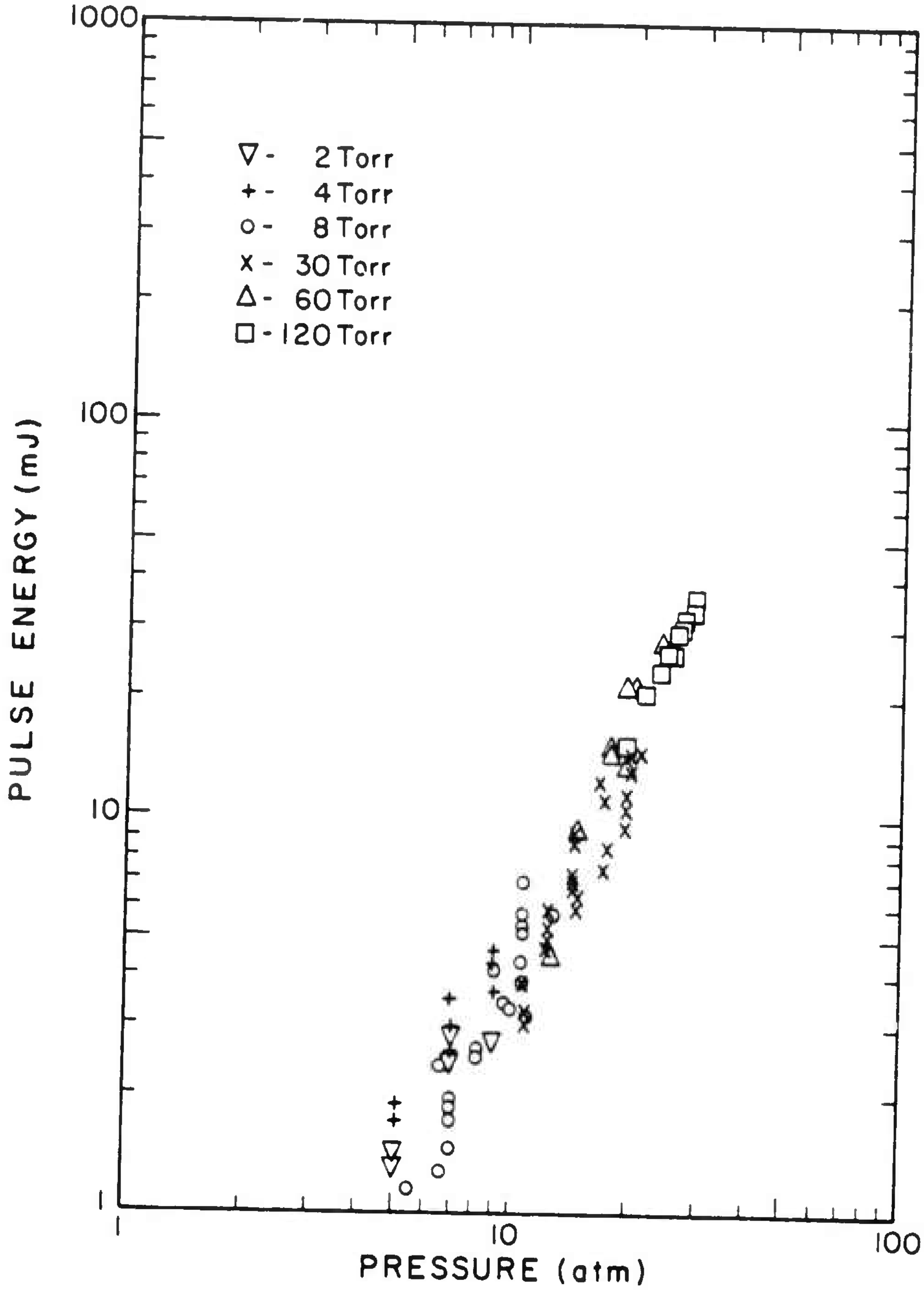

Figure 4: Summary plot of total laser pulse energy emitted from a 16 cm^3 volume as a function of total gas pressure. The integrated e-beam current corresponds to 275 μ coulomb for each case. Data points represent output at 4278 Å and different partial pressures of N_2 are indicated by the shape of the data point.

vibrational quantum number, it has the same degeneracy and should "fill" to terminate the 4278 Å transition in a comparable time. That it does not, as seen in the figure, is strong evidence for the existence of an unblocking process tending to quench the vibrational excitation of the lower, v"=1, state of 4278 Å transition. This is the process leading to the quasi-cw operation discussed below.

The problem of determining the absolute energy deposition from the electron beam in the radiating volume was particularly acute in the case of the 3914 Å emission, because the laser output in this case occurred so early in the course of the e-beam pulse. Because of the drastic difference in the pulse shape of the 3914 Å transition the growth of the interaction volume[7] in the laser cavity was quite different as was evidenced by the considerably increased divergence of the output beam. This, consequently, offered the best test of the deconvolution procedure because variance of divergence and interaction volume had been relatively small in the case of the (0,1) transition at 4278 Å. For the self-terminating (0,0) transition at 3914 Å average interaction volumes were found to vary from 3.4 to 14 cm^3 which was sufficient to more than mask other systematic variation of laser output with experimental parameters such as pressure and current.

However, when corrected and plotted on the "master curve" of laser performance, shown in Figure 4, the data of the gross extraction at

3914 Å "fit" smoothly both the theory and experimental data for output at 4278 Å.

These results on other transitions in the helium-nitrogen laser support three important conclusions. First is that the deconvolution procedure was realistic and defensible and thus that the energy available for extraction by the fields can be represented by a simple "master performance curve" as seen in Figure 4. Second is that since threshold is indeed achieved for the (0,0) component of the B → X electronic transition, the branching ratio of the output channels of the pumping reactions (7a) and (7b) strongly favors the upper B state and hence branching in the output channels poses no obstacle to the extraction of one photon per molecular helium ion. Finally, the third point is that the energy pumped into the B state of N_2^+ is effectively stored until the fields resulting from the spontaneous emission build up enough to start the stimulated emission ultimately extracting the energy stored in the inversion. Even though the 3914 and 4278 Å outputs start at substantially different times in the life of the plasma, the same integrated extraction efficiency is achieved. That this energy is not extracted in the (0,2) transition at 4709 Å is a consequence of the relatively small transition probability and hence slow accumulation of spontaneous emission at this wavelength necessary to initially excite the cavity oscillations. Evidentally the cooling of the plasma at the end of the e-beam discharge destroys the stored energy by recombination.

Either a longer duration e-beam pulse or an external source of cavity excitation would be necessary to extract the stored energy at 4709 Å.

Most of the advances in laser performance obtained during the current reporting period resulted from the thermal scaling of the output. Recognizing that reactions (6b), (6c), (9a), (9b) and most probably (7b) should have an inverse dependence on gas temperature, it was expected that the overall laser output would be strongly temperature dependent. Consequently it appeared that the theoretical limit of 10.5% efficiency shown in Figure 1 might best be approached through thermal adjustment of the kinetic sequence.

Unfortunately the existing laser device was not designed for thermal cycling and this together with the relatively high thermal conductivity of helium made the accurate control of gas temperature very difficult. Best control was obtained by mounting the device on a 30 cm drift tube which was then connected to the electron beam gun with an additional foil assembly. When the drift tube was filled to a rather critically defined pressure of nitrogen (∿500μ) about half the electron beam current could be conducted to the laser device. Under these conditions the more limited thermal conductivity reduced the constant thermal flow from the gun and afforded some control over temperature in the laser.

Cooling in that arrangement was accomplished by circulating cold liquid nitrogen vapor through a heat exchanger attached to the pressure vessel containing the laser device and the average gas temperature in the cell was obtained by measuring the pressure as the system cooled.

Since the laser cavity was positioned closest to the source of the thermal flow into the heat exchange, the actual temperature in the laser cavity was necessarily higher than the average gas temperature. Hence, the following data presented underestimates the effect on laser output of reduced gas temperatures by underestimating those temperatures. However, the variation of laser output with changes in average gas temperature in the pressure vessel could be obtained. Figure 5 shows the effect of continuously cooling the gas prior to the electron beam discharge at a nominal current down the drift tube of 11KA. As can be seen factors of improvement of as large as 10 were achieved for a 42°C decrease in average gas temperature. As might be expected, the limiting temperature appears not to have been reached at -20°C.

In an attempt to verify that the thermal enhancement observed was not limited to excitation at the low current available from the drift tube, the laser device and liquid N_2 heat exchanger were connected directly to the electron beam gun. Though not as remarkable as the effects on the outputs from the drift tube configuration shown in the previous figure, the effects of a similar decrease in average temperature on discharges at higher currents and pressures were substantial. "Best" outputs at each pressure were increased by a factor of approximately 2 for a 40 to 50°C decrease in the average gas temperature which in this configuration even more seriously under-estimated the actual gas

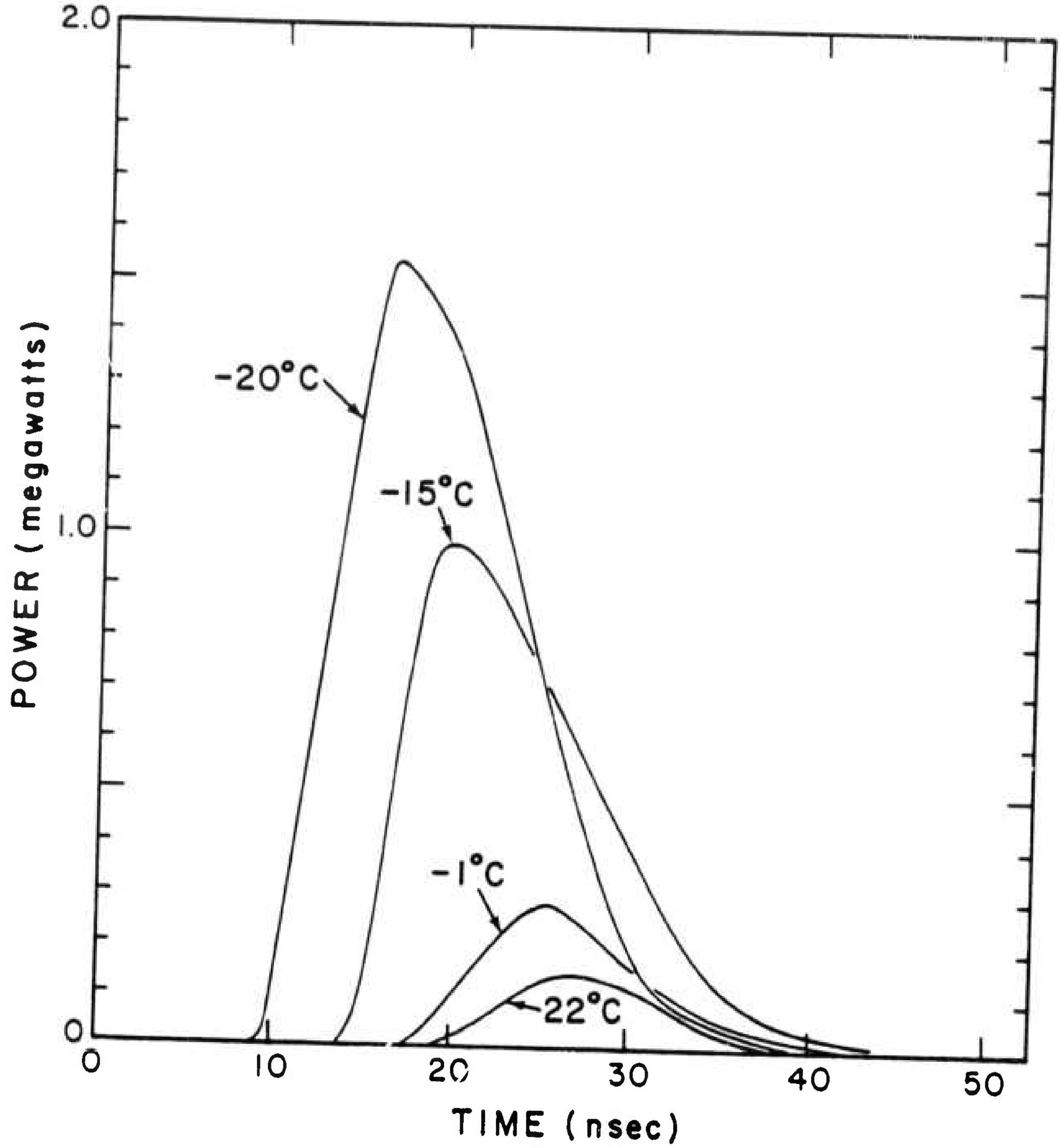

Figure 5: Thermal scaling of the helium-nitrogen charge transfer laser. Plotted parametrically as a function of gas temperature are time resolved power measurements of the violet line at 4278 Å. Data are shown for the discharge of 190 μ coulomb into 21 atm. pressure of helium containing 60 Torr of nitrogen. The time scale has been normalized so that the zero corresponds to the beginning of the e-beam current output.

temperature in the laser cavity. It was found that the low temperature data could be parameterized in an analogous manner to eq. (13) by the following empirical expression for the efficiency over the 10 - 35 atm. pressure range examined,

$$\varepsilon(-20°C) = 10.5\%(P/80)^{1.2} \quad , \qquad (14)$$

where P is the equivalent gas pressure at room temperature, as in the expression for the efficiency at room temperature, eq. (13) and 10.5% is the revised theoretical limit on efficiency shown in Figure 1.

The actual dependence of laser output on gas temperature is, in fact, more complex than might be inferred from eq. (14). In part, this results from the effect of cooling on the quasi-cw operation of the laser. Probably through an enhancement of reaction (9a) in the kinetic sequence, cooling of the gas tends to initiate quasi-cw operation at a lower operating pressure than would otherwise be possible. This is best seen in Figures 6a and 6b which show the time dependence of the power emitted at a relatively low pressure for nominal discharge currents of 15 and 20KA, respectively. To facilitate the estimation of the time dependence of the efficiency for the emission of the output power relative to the input power to the cavity, a constant fraction of the input power has been shown by the dashed curves.

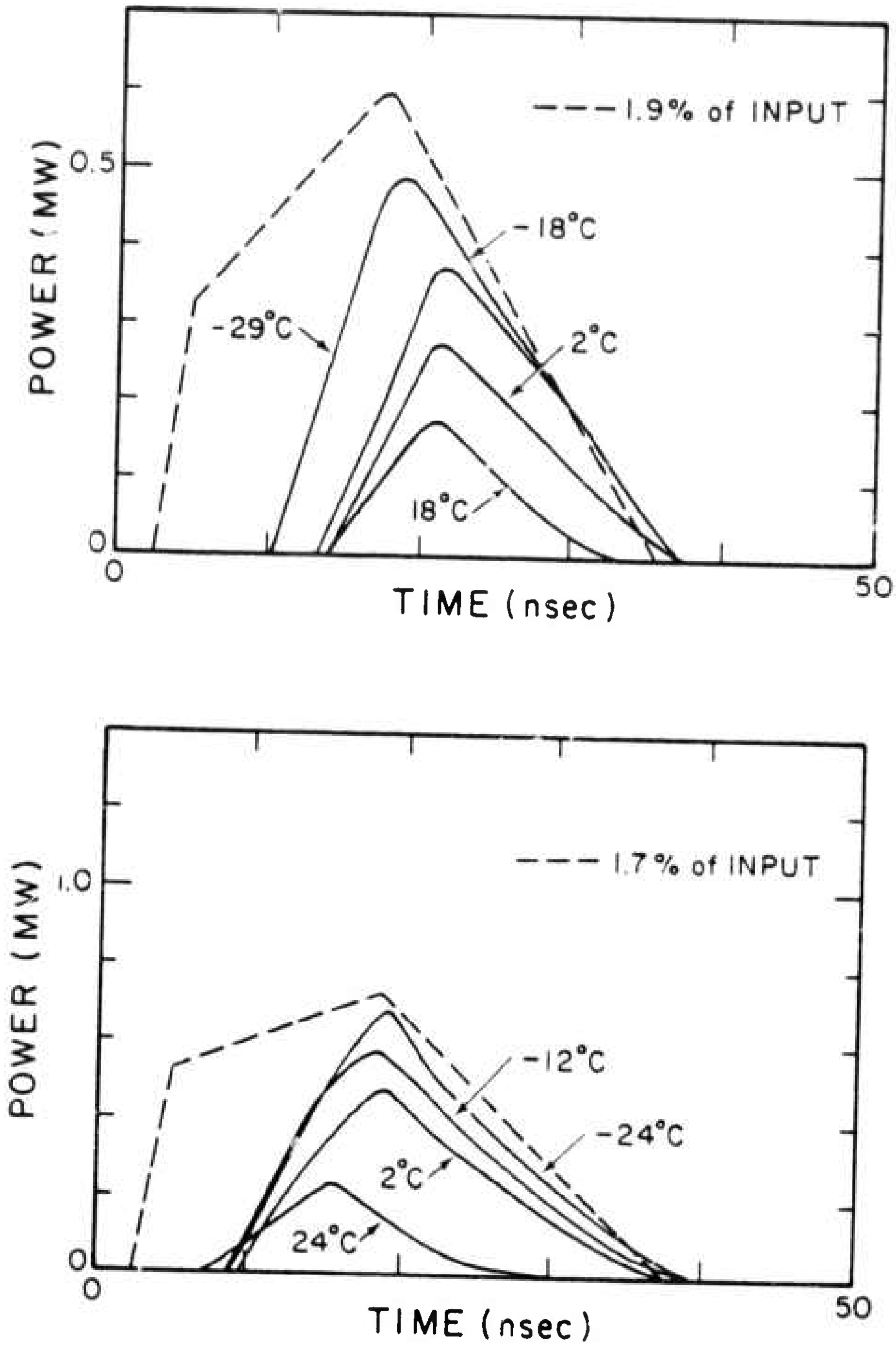

Figure 6: Plot of the time-resolved laser power emitted at 4278Å from an electron beam discharge into 7.7 atm pressure of helium containing 15 torr of N_2 at the gas temperatures shown. The dashed curves show the time dependence of a constant fraction of the corresponding input power deposited in the laser cavity. (a) Upper curves: Data for nominal 15KA e-beam current, (b) Lower curves: Data for nominal 20KA e-beam current.

It can be immediately seen that in neither case is quasi-cw operation in evidence at room temperatures. In both cases the output is seen to terminate before the input. However, as the temperature was decreased, two effects were noticed. First, quasi-cw operation was established for later times and the constancy of the power efficiency improved with decreasing temperature. Secondly, at sufficiently low temperature, around -20°C, the onset of threshold occurred earlier with operation at constant efficiency being more rapidly attained. From the standpoint of efficiency with respect to input power, the constant level of 1.9% achieved after onset of threshold at 7.7 atm. pressure and 15KA excitation current (Fig. 6a) is considerably in excess of the value of 0.6% consistent with the parameterization given by eq. (14).

The same general behavior was found at higher pressures as shown in Fig. 7 for the excitation of 11 atm. of gas mixture at a nominal 20KA of beam current. These results suggest that the efficiency most characteristic of the laser performance is the steady state value of efficiency reached with respect to input power. At least the range of times available to these experiments it appears that at low temperatures the laser output can be expected at that level of efficiency for as long as it is pumped by the electron beam. Such operation is of extreme importance as it points the way toward much longer output pulses to be obtained from longer discharge pulses.

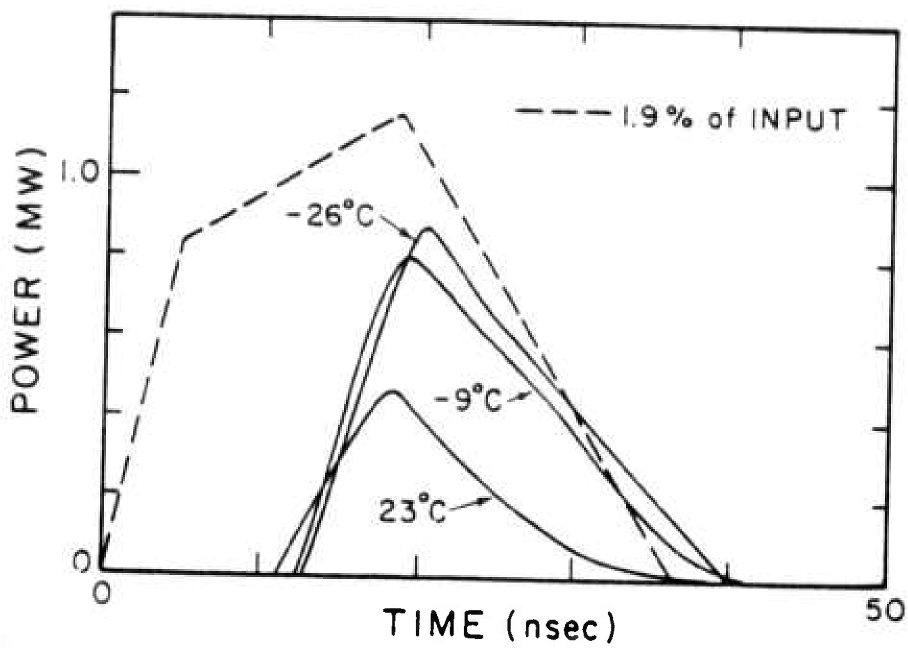

Figure 7: Plot of the time-resolved laser power emitted at 4278Å from an electron beam discharge into 11 atm pressure of helium containing 30 torr of N_2 at the gas temperatures shown. The dashed curves show the time dependence of a constant fraction of the corresponding input power deposited in the laser cavity for a nominal e-beam current of 20KA.

Finally this quasi-cw operation characteristic of a 4-level laser was observed at the highest pressures which could be accommodated by the existing pressure vessel. As seen in Fig. 8 operation at a steady-state power efficiency of 3% was achieved at $-20^{\circ}C$ for an average gas density corresponding to a pressure of 35 atmospheres at room temperature. The corresponding pulse energy was 80mJ and represented a peak power density of

$$P = 320 \text{ Megawatts/liter} \quad , \qquad (15a)$$

an energy density of

$$E = 5 \text{ Joules/liter} \quad , \qquad (15b)$$

at an efficiency,

$$\varepsilon = 3\% \quad . \qquad (15c)$$

Intracavity circulating powers reached a peak of over 1.2 GW/liter at an intensity greater than 20 MW/cm^2 with no evidence of either bottlenecking or photoionization of any of the species important to the kinetic chain.

A pragmatic comparison of these results from the collisional transfer pumping mechanism with that of the KrF excimer can perhaps be made on the basis of best performance to date and tends to place these results in perspective. Such a summary is shown in Table 3.

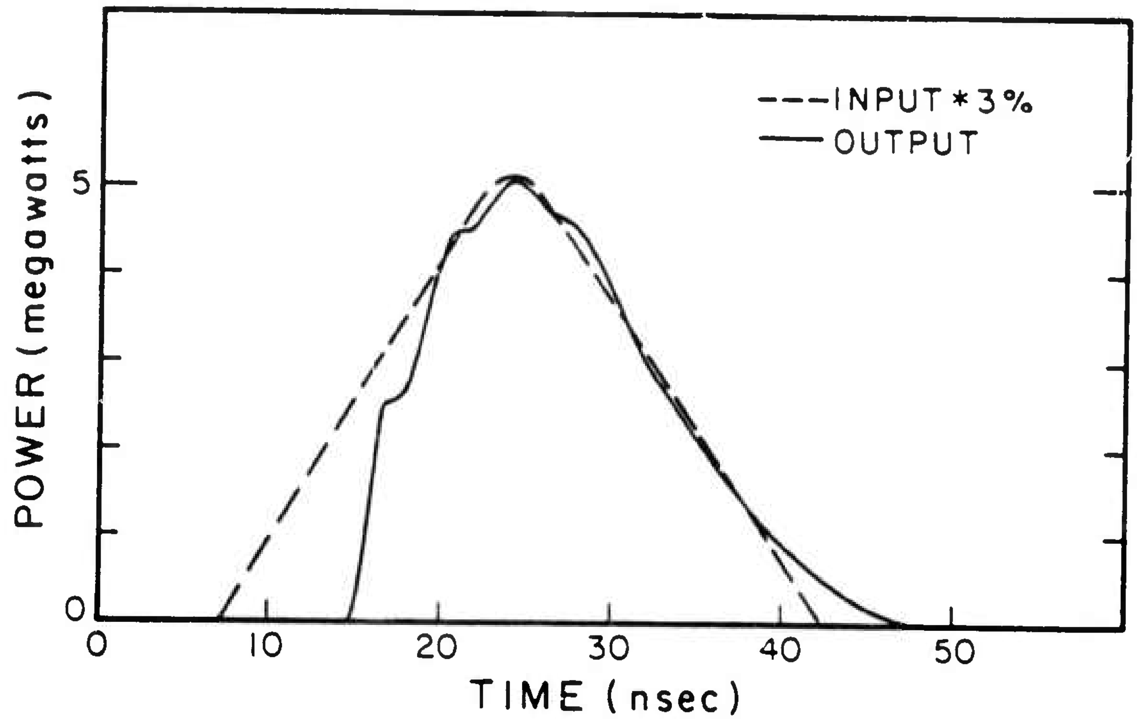

Figure 8: Plot of data showing quasi-cw operation of the helium-nitrogen laser. The solid curve shows output power at 4278 Å emitted from an electron beam discharge into 35 atmospheres pressure of helium containing 120 Torr of nitrogen. The dashed curve shows 3% of the corresponding power deposited in the laser cavity.

Table 3

Comparison of the best performance achieved to date for collisional transfer and inert gas-halide excimer lasers.

	Pulse Width (nsec.)	Energy (J/ℓ)	Photon Intensity (10^{20} photons/ℓ/sec)
Charge Transfer[6] ($He:N_2$)	17	5	6.5
High Efficiency Excimer[9] (KrF)	125	15	1.5

That larger outputs are reported for KrF lasers directly excited by e-beams can clearly be seen to result from a combination of higher energy per photon and longer excitation pulses available on e-beam machines used in the KrF studies. Both lasers operate as 4-level systems and hence, emit until the end of the excitation pulse.

It can be reasonably expected that further thermal enhancement will raise these levels for the helium-nitrogen charge transfer laser even closer to theoretical limits and will offer the future possibility of very long output pulses or even cw operation at relatively high saturation levels.

IV. IMPLICATIONS

More immediate implications concern the further thermal scaling of the current nitrogen charge-transfer laser. It now appears that the theoretical efficiency of 10.5% for the 4278 Å transition will be attained at operating pressures around 40 atm. at sufficiently low temperatures. Even at the relatively low operating charges of 275 μ coul currently used, this will yield an output of 20 J/liter. A value of 5 J/liter has already been attained. Since quasi-cw operation has been achieved an increase in e-beam pulse duration from a nominal 20 nsec to 100 nsec should bring the value projected at 40 atm to 100 J/liter. Funds to support procurement of a longer pulse-forming line for APEX have been requested in order to verify this projection.

Even at the present "best levels of efficiency," a kilojoule, laser at 4278 Å could be built with current technology. Assuming direct e-beam pumping at a level of 60% of the maximum foil loading, 90 liters could be pumped with a 200 nsec pulse to give 1KJ output at 4278 Å. The successful conclusion of the thermal scaling studies would serve to raise the output to 3KJ per pulse at the appropriate operating temperature.

While it should be realized that the helium-nitrogen laser is only the first example of the new class of e-beam charge transfer lasers, results observed to date for this system clearly confirm the importance of charge transfer reactions as laser pumping mechanisms.

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