The effects of He addition on the performance of the fission-fragment excited Ar/Xe atomic xenon laser

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The intrinsic power efficiency of the atomic xenon laser depends upon the electron density because of the mixing of the laser levels by electron collisions while the electron density in high-pressure particle-beam excited plasmas increases with increasing gas temperature. Therefore, in order to reduce the amount of electron collisional mixing when operating at high-energy loadings (> 100's J/1-atm) mixtures having a high-heat capacity are required. In particle-beam excited Ar/Xe mixtures, which typically yield the highest intrinsic laser efficiencies, increasing the gas pressure to increase the heat capacity is not always practical due to the high-stopping power of the gas mixture. For this reason we have experimentally and theoretically investigated adding He to Ar/Xe mixtures in studies of a fission-fragment excited atomic xenon laser. Adding He increases the heat capacity without appreciably perturbing the favorable kinetics resulting in efficient operation of the laser in Ar/Xe mixtures. We find that when adding He to Ar/Xe mixtures the dominant laser transition switches from 1.73 to 2.03 μm without significantly decreasing the efficiency. The laser pulse length also increases, an effect attributed to a lowering of both the electron temperature and gas temperatures.

I. INTRODUCTION

The atomic xenon laser operates in the near infrared at several wavelengths between 1.7 and 3.6 μm, predominantly on transitions between the $5d$ and $6p$ manifolds.[1-7] (see Fig. 1). Intrinsic laser efficiencies as high as 5% have been obtained using binary gas mixtures composed of rare gases and less than a few percent of xenon.[3,6] Ar/Xe mixtures have provided the best efficiency and under optimum conditions operate predominantly on the 1.73 μm transition $\{Xe(5d[3/2]_1 - 6p[5/2]_2)\}$. In Ar/Xe mixtures the 2.03 μm transition $\{Xe(5d[3/2]_1 - 6p[3/2]_1)\}$, which dominates in He/Xe mixtures, is virtually absent. It has been suggested that a rapid collisional radiative cascade following electron-ion dissociative recombination of $ArXe^+$ efficiently populates the $Xe(5d[3/2]_1)$ level.[8] Quenching of the common lower laser level of the 1.73 and 2.63 μm transitions $\{Xe(6p[5/2]_2)\}$ by collisions with argon is sufficiently fast that there is little competition between these lines with high-power excitation (> kW's/cm^3). Heavy particle collisions are therefore responsible for both efficiently populating the upper laser levels and emptying the lower laser levels, which explains why the laser spectrum is a sensitive function of the gas mixture.[2]

When operating at high-power deposition such that the fractional ionization exceeds $\sim 10^{-5}$, electron collisional mixing (ECM) of the $6s$, $6s'$, $6p$, and $5d$ manifolds may reduce or quench laser oscillation.[8,9] This suggests that to obtain high specific laser energy one must pump at a moderate rate for long periods of time to insure that the critical fractional ionization is not exceeded. A problem, however, occurs at high-energy loading (> 100's J/1-atm) when the gas temperature increases by many 100s K. The electron density increases with increasing gas temperature because the rate constants for atomic ion association to dimer ions[10] and their subsequent dissociative recombination,[11] which is the predominant electron loss process, decrease with increasing gas temperature. Therefore, laser oscillation may be quenched by ECM simply by energy loading the gas. One therefore would like to operate with gas mixtures having a high-heat capacity which, for rare gases, requires high gas pressures. Due to the relatively short range of energetic particles in Ar, simply increasing the total gas pressure of Ar/Xe mixtures in particle-beam excited systems is often not practical.

One is therefore motivated to increase the heat capacity of Ar/Xe mixtures by adding a lower atomic weight rare gas such as He. The xenon laser using He/Xe mixtures, though, is less efficient than Ar/Xe mixtures. Using He/Xe mixtures results in both a lower laser efficiency (< 1%)and a different spectrum (mainly 2.03 and 3.37 μm). One questions, then, whether the favorable performance obtained in Ar/Xe mixtures can be realized by adding He to a particle-beam-excited mixture. A cursory examination of the pertinent rate coefficients predicts that the dominant ion in He/Ar/Xe mixtures having $He/Ar \leqslant 1$ is still $ArXe^+$. Therefore *adding* He to Ar/Xe mixtures

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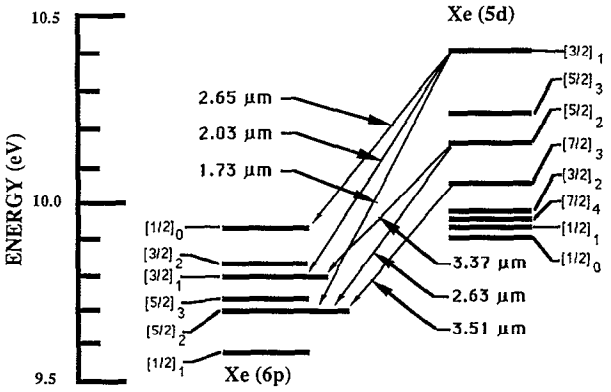


FIG. 1. Partial energy level diagram of Xe showing some of the laser transitions between the 5*d* and 6*p* manifolds.

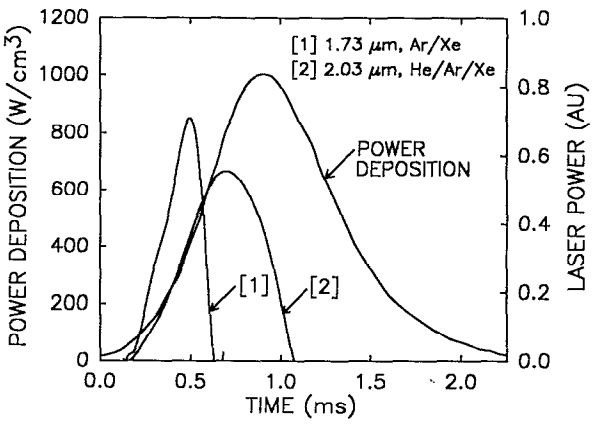


FIG. 2. Power deposition and laser power as a function of time during a pumping pulse from the SPR III nuclear reactor. The pulse averaged power deposition is 500 W/cm^3. Laser oscillation using an Ar/Xe mixture (520 Torr) is predominantly at 1.73 μm, and terminates earlier than that using a He/Ar/Xe mixture (1040 Torr). The predominant line for the He/Ar/Xe mixture is 2.03 μm.

would not be expected to significantly interfere with the excitation mechanism of the 5*d* manifold. Furthermore, He is not likely to significantly modify the quenching of the 6*p* manifold by Ar. Fission-fragment pumping of He/Ar/Xe mixtures has shown good efficiency for lasing at 2.03 μm rather than 1.73 μm.[6] It should be noted, though, that substitution of He for argon in discharge-excited Ar/Xe mixtures causes a significant reduction in laser energy.[7]

In this paper we report on experimental and theoretical studies of the effect of adding He to Ar/Xe mixtures during fission fragment excitation. We find that adding He to Ar/Xe mixtures does not detrimentally affect the laser efficiency when the He/Ar ratio is >0.3–0.4 and results in longer laser pulse lengths. This is attributed to an increase in the heat capacity of the mixture which reduces the electron density at high-energy loading and a lowering of the electron temperature upon adding helium. The dominant transition, however, switches from 1.73 to 2.03 μm. We show that this trend is due to differences in quenching rates of the various 6*p* levels by helium and argon.

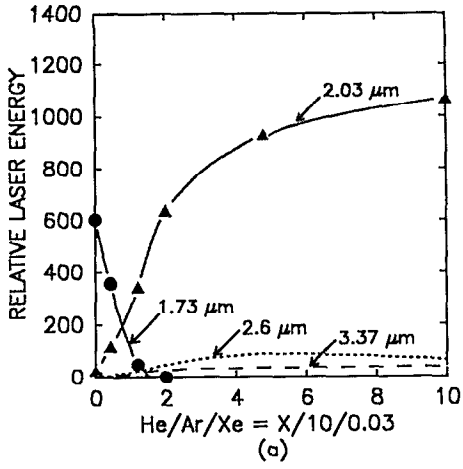
II. FISSION FRAGMENT EXCITATION OF He/Ar/Xe MIXTURES

The xenon laser was pumped using fission-fragment excitation with the Sandia Pulsed Reactor III (SPR III) facility. The apparatus is very similar to that described in Ref. 6 and therefore will be only briefly discussed here. The active pumped volume of the laser cell is $60\times1\times7$ cm^3 and the cell is equipped with Brewster angle windows made of quartz. Fission fragments from $^{235}UO_2$ foils lining the cavity are generated by neutron pulses from the reactor having pulse lengths of 0.5–1.5 ms FWHM. The power deposition is obtained by normalizing the slow-neutron signal observed with a Reuter-Stokes cobalt detector to the energy deposition obtained by gas pressure rise.[6] Power depositions of 10's to 100's W/cm^3 are obtained for gas pressures of 0.5–1.5 atm. This wall-pumping scheme can give rise to medium motion which causes the medium to look like a converging lens in one dimension.[12] The optical cavity is a stable-unstable resonator designed to minimize the effect of this time-dependent index-of-refraction variation. The resonator consists of a spherical primary mirror ($R=4$ m) and a cylindrical secondary ($R=-1.65$ m) with a 45° scraper mirror that has a 3-cm feedback hole. The primary and secondary mirrors were gold coated while the scraper mirror was aluminum coated.

Laser energy was measured with a calorimeter while adding helium to an Ar/Xe mixture of 99.7/0.3. The Ar/Xe components were kept at a fixed pressure of 520 Torr. The average power deposition by fission fragments was kept constant at 500 W/cm^3. This results in a total energy deposition of approximately 500 J/l and a temperature rise of 1000 K for an initial pressure of 1 atm. The power deposition and laser pulses for Ar/Xe and He/Ar/Xe mixtures are shown in Fig. 2 for a He/Ar ratio of 1/1 at a total pressure of 1040 Torr. The laser energy spectrum is shown in Fig. 3(a), and the laser energy, power and pulse length are shown in Fig. 3(b) as functions of helium fraction. When the He/Ar ratio exceeds 0.1–0.2, the dominant laser transition switches from 1.73 to 2.03 μm. The total laser power drops by approximately half upon addition of 10% He but then increases at higher He concentrations to a value slightly below that obtained with Ar/Xe. The total laser energy has a small decrease when initially adding He and then increases above the value obtained in the Ar/Xe mixture. The length of the laser pulse, as shown in Figs. 2 and 3(b), increases with He addition.

The fact that the average laser power (energy/pulse length) is constant for He/Ar >0.2–0.3 while the laser energy increases implies that the improvement in the total laser energy comes from the increase in laser pulse length. The increase in pulse length, we suggest, is likely a result of the increase in heat capacity, which gives smaller increases in gas temperature and electron density, which delays termination of laser oscillation by ECM.

The 2.03-μm transition has a higher oscillator strength than the 1.73-μm transition with which it shares a common



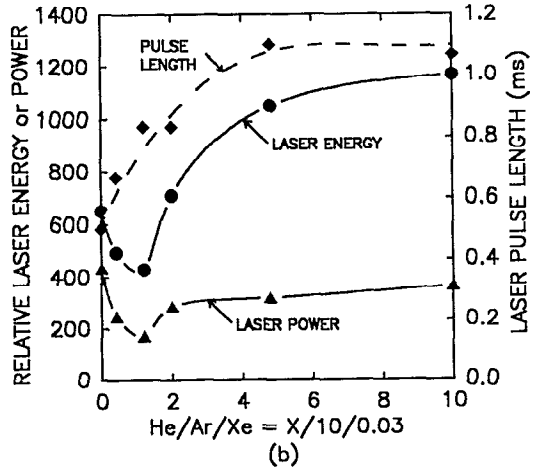


FIG. 3. Experimental laser energy/power for adding helium to an Ar/Xe mixture. The Ar pressure is 518 Torr. (a) Wavelength resolved laser energy; (b) Total laser energy, laser power, and laser pulse width. The average power deposition is constant at 500 W/cm^3. The laser pulse energy decreases with addition of small amounts of He, but ultimately increases when the laser transition switches from 1.73 to 2.03 μm. The increase in laser energy is largely a result of an increase in the laser pulse width.

upper laser level.[13] Earlier modeling efforts suggested that the 1.73-μm transition oscillates in Ar/Xe mixtures because of preferential quenching of its lower laser level by Ar, and the fact that it has a more favorable ratio of degeneracies.[8] This allows the 1.73 μm transition to saturate the Xe($5d[3/2]_1$) level. The change in laser spectrum from 1.73 to 2.03 μm upon addition of He suggests that the lower level of the 2.03-μm transition {Xe($6p[3/2]_1$)} is selectively quenched by He. It also suggests that the lower laser level of the 1.73 μm transition Xe($6p[5/2]_2$) may be selectively pumped by a collisional cascade from the higher lying levels of the Xe($6p$) manifold.[7]

III. MODEL RESULTS AND ANALYSIS

Laser performance was simulated using a detailed kinetics model which includes individual levels of the $6p$ and $5d$ manifolds of xenon in addition to the lumped states Xe($6s$), Xe($6s'$), Xe($7s/7p$), and Xe** (higher radiating states), and laser fluxes for transitions at 1.73, 2.03 , 2.63, 2.65, and 3.37 μm. The model, based on that described in Ref. 8 for Ar/Xe mixtures, was modified to include the necessary plasma chemical reactions for addition of He. Although rate coefficients are available for most of the plasma chemical reactions relating to the total species balance in He/Ar/Xe[14,15] mixtures, little information is available for the quenching of the individual excited states of xenon by helium. The plasma chemical reactions set an upper limit on the laser efficiency, while neutral quenching of excited states in large part determines the laser's spectrum.

TABLE I. Rate coefficients [10^{-11}cm^3/s] for quenching of Xe ($6p$).

Level	Ar	Ref.	He	Ref.
$6p[1/2]_0$[a]	14.0	b	2.0	d
$6p[3/2]_2$	4.7	b	0.17	d
$6p[3/2]_1$	1.0	b	7.5	c
$6p[5/2]_3$	2.6	b	0.7	d
$6p[5/2]_2$	8.2	b,c	0.65	c
$6p[1/2]_1$	0.6	b	0.2	d

[a]Including branching to $5d$ manifold.
[b]J. Xu and D. W. Setser, J. Chem. Phys. **92**, 4191 (1990).
[c]W. J. Alford, IEEE J. Quant. Electron. **26**, 1633 (1990).
[d]J. Xu and D. W. Setser, J. Chem. Phys. (submitted).

State-to-state quenching rates for quenching of the $6p$ levels by argon have been measured by Xu and Setser.[16] Rate coefficients for quenching of Xe($6p[5/2]_2$) and Xe($6p[3/2]_1$) levels by helium have been recently measured by one of the authors (W.J.A.) using a multiphoton laser-induced-fluorescence technique. Those results are reported in detail elsewhere.[17] The remaining coefficients for the quenching of Xe($6p$) by He levels were obtained from Ref. 18 (see Table I). As the branching for the products of these reactions are not known, we assumed that the quenching is to the $6p$ or $6s$ level directly lower in energy (see discussion below). The exception is for Xe($6p[1/2]_0$) which has branching to the $5d$ manifold.[18] Other quenching processes and branching ratios were semi-empirically derived by hypothesizing a quenching scheme, incorporating the scheme into the model, and comparing to experiment. The reaction scheme and branching ratios so generated are not unique, but they do help in explaining or hypothesizing the processes responsible for the observed laser behavior. Some of these coefficients are discussed below.

In Ar/Xe mixtures the $5d$ manifold is predominantly populated by dissociative recombination of ArXe$^+$ followed by collisional and radiative cascade. Due to its high-excitation energy and low-binding energy, it is not expected that dissociative recombination of HeXe$^+$ and its cascade will populate the same states as recombination of ArXe$^+$. The fact that laser oscillation is obtained on Xe($7p$–$7s$) transitions in He/Xe mixtures but not in Ar/Xe mixtures supports this conjecture. The fact that total laser power is somewhat independent of He concentration in He/Ar/Xe mixtures, at least for He/Ar ratios of 0.3–1.0, suggests that HeXe$^+$ is not playing a dominant role in the recombination kinetics. We estimated that the

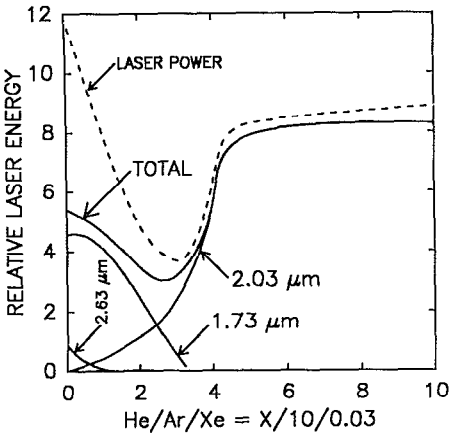


FIG. 4. Calculated laser energy as a function of helium concentration in an He/Ar/Xe mixture for the conditions of Fig. 3. The experimental results are generally reproduced with the exception that the calculated minima in laser energy and power occur at a larger He fraction.

rate constant for $Xe^+ + He + He \rightarrow HeXe^+ + He$ is 1×10^{-32}cm^6/s, which is 10 times smaller than that for $Xe^+ + Ar + Ar \rightarrow ArXe^+ + Ar$.[8] The rate decreases with increasing gas temperature as $T_g^{-1.5}$. The dissociative recombination rate is given by $3.5\times10^{-7}[1 - \exp(-180/T_g)]T_e^{-0.5}$ cm^3/s, where T_e is the electron temperature[8]. We also estimated that heavy-particle collisions of $5d$ levels by He are not selective and result in thermalization of the manifold to the gas temperature with a rate constant of 10^{-12} cm^3/s. The one exception is our estimation that the Xe($5d[5/2]_2$) level is preferentially quenched by He with a rate constant of 10^{-11} cm^3/s. This process was found to be necessary to supress oscillation at 3.37 μm at high He fractions, a transition which is not experimentally observed to be important. In the absence of this quenching, the 3.37 μm transition would carry approximately half of the laser power when He/Ar = 1.

Laser parameters were computed by specifying a gas mixture, power deposition and energy loading, which determines the gas temperature, and calculating quasi-cw laser power. These conditions are obtained in approximately 10 μs. These results were then convolved with the experimental power deposition profile, which extends for >1 ms, to yield laser parameters as a function of time. These values were then averaged over time to yield pulse averaged quantities. The computed laser energy spectrum and pulse-averaged laser power are shown in Fig. 4. The experimental trends are generally reproduced. Laser power first decreases with He addition, and then settles to nearly a constant value. The laser pulse energy also first decreases and then increases with increasing He addition, a consequence largely of the longer laser pulse. The laser spectrum also changes from predominantly 1.73 to 2.03 μm. The behavior of the 2.63 μm transition is not well reproduced. The predicted minimum in laser power and the switch in the laser line also occur at a He addition of 0.2–0.3, similar to that observed in a discharge[7], whereas these events occured at a He addition of 0.1–0.2 in fission-fragment-excited systems. These last few items are quite sensitive to the electron temperature which is rapidly changing for He additions of <0.5 (see discussion below). Higher electron temperatures tend to decrease 2.63-μm laser power at low-He fractions and increase it at high-He fractions, while lower electron temperatures shift the minima in laser energy and power to lower He fractions.

The switch in the laser line from 1.73 to 2.03 μm results from a combination of at least two processes. First, the rate coefficient for the quenching of Xe($6p[3/2]_1$), lower laser level for 2.03 μm, by He is large (7.5×10^{-11} cm^3/s). This level is presumably quenched one level down to the Xe($6p[5/2]_3$).[17] Second, this level also has a reasonably large rate coefficient for quenching to the Xe($6p[5/2]_2$), lower laser level of the 1.73 μm transition, by argon.[16] The switch in the predominant laser line from 1.73 to 2.03 μm (which share a common upper laser level) is largely explained by the selective quenching of Xe($6p[3/2]_1$) by helium and cascade of population to the Xe($6p[5/2]_2$) by collisions with argon. Also, the decrease in electron temperature which occurs with He addition (see below) initially decreases gain, which tends to favor the 2.03 μm transition.

The lengthening of the laser pulse is largely due to a decrease in the fractional ionization with increasing He addition which reduces the rate of ECM. This decrease in fractional ionization results from two causes. First the higher heat capacity of the mixture reduces the gas temperature for a given energy loading and therefore reduces the rate of increase in the electron density. Therefore a higher energy loading can be sustained (that is, longer pumping) before ECM terminates the laser pulse. The second cause is related to the electron temperature.

The initial decrease in laser energy and power upon addition of He is largely due to a rapid decrease in electron temperature. In rare-gas particle-beam-excited plasmas, the electron temperature, T_e, is determined by the rate of electron thermalization below the first electronic excitation threshold and the energy at which electrons are removed by dissociative recombination. Since the former energy is $\geqslant 10$ eV in He/Ar/Xe mixtures and the latter is $\leqslant 1$ eV, the rate of momentum transfer by electrons in the range of 1–5 eV is most important in determining T_e. This rate, proportional to $2m_e\sigma_m/M$ (m_e is the electron mass, M is the atomic mass, and σ_m is the momentum-transfer cross section) is approximately 10–100 times larger in He/Ar/Xe mixtures compared to Ar/Xe mixtures. This results from the smaller mass of He and its larger momentum-transfer cross section.[19] The electron temperature therefore decreases with increasing He fraction, as shown in Fig. 5. This decrease in T_e is initially detrimental to laser performance because it reduces the electron density and reduces the rate of electroionization which recirculates energy between Xe($6s$) and Xe^+.[1–3] In the example shown in Fig. 5, the power deposition is too large to sustain quasi-cw laser oscillation in an Ar/Xe mixture because ECM quenches the inversion. The lowering of the electron density with He addition eventually allows oscillation to occur. At sufficiently high-energy loading when the electron density

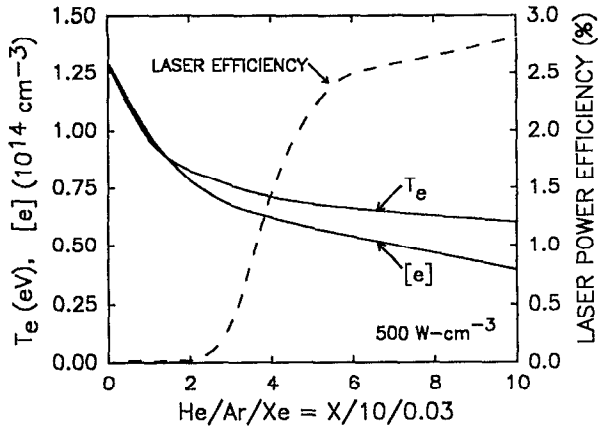


FIG. 5. Electron temperature, density, and laser power efficiency predicted by the model for room temperature excitation of He/Ar/Xe mixtures. The power deposition is 500 W/cm^3 and the Ar partial pressure is held fixed at 520 Torr (10 psia). Both electron temperature and density decrease with helium addition. Laser oscillation, quenched by ECM at low-He fraction, eventually occurs when the electron density decreases sufficiently.

would normally increase due to the increasing gas temperature, the reduced efficiency of electroionization which results from the lower electron temperature is advantagous as it prevents a catostrophic increase in electron density.

The motivation for adding He to Ar/Xe mixtures was to raise the heat capacity in order to increase the maximum permissable energy loading of the gas. We modeled the influence of helium addition in this respect by holding the gas density fixed (for a given mixture) and varing the gas temperature. In this way we simulated laser operation at different energy loadings. In Fig. 6 we show the laser power efficiency as a function of energy loading for Ar/Xe = 99.7/0.3 (520 Torr at 300 K) and for adding an equal portion of He (total pressure 1040 Torr) for pumping rates of 300 W/cm^3. This pumping rate yields almost the same laser efficiency in both mixtures at room temperature but it is near the maximum quasi-cw pumping that the Ar/Xe mixture can sustain before electron collisional mixing quenches laser oscillation. Therefore only a small increase in electron density due to gas heating can be tolerated and laser oscillation in the Ar/Xe mixture is quenched by an energy loading of < 50 J/l or a temperature rise of 150 K. The He/Ar/Xe mixture can sustain oscillation to an energy loading of 400 J/l, or a temperature rise of 580 K before electron collision mixing terminates oscillation.

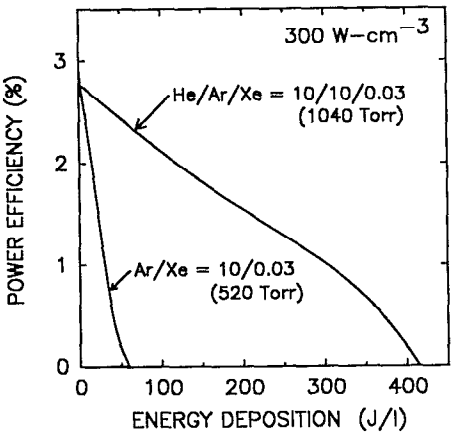


FIG. 6. Calculated quasi-steady state laser power efficiency as a function of energy loading for Ar/Xe = 10/0.03 mixtures (520 Torr or 10 psia) and He/Ar/Xe = 10/10/0.3 (1040 Torr or 20 psia). The power deposition is 300 W/cm^3. The increase in gas temperature is 2 K/(J/l-atm). The Ar/Xe mixtures are quenched at low-energy deposition due to electron collision mixing of the laser levels which increases with increasing gas temperature. The He/Ar/Xe mixtures are less sensitive to this effect due to their higher heat capacity and lower electron temperature.

IV. CONCLUDING REMARKS

We have experimentally and theoretically studied the effect of adding He to Ar/Xe mixtures in fission-fragment excited atomic xenon lasers. The addition of He most likely results in selective quenching of the lower laser level of the 2.03-μm transition and selective pumping of the lower level of the 1.73 transition. This causes a switch in the predominant laser transition from 1.73 μm to 2.03 μm, but does not significantly alter the excitation sequence of the upper laser levels. The electron temperature decreases with increasing He addition and this decrease is detrimental to laser performance at low-pump rates and energy loading. At sufficiently high-pump rates ($>$ 10's–100's W/cm^3) and energy loading ($>$ 10's–100's J/l), though, this decrease in electron temperature is advantageous due to the regulating effect it has on the increase in electron density which occurs with increasing gas temperature. The use of He/Ar/Xe mixtures therefore provides the means whereby higher energy loading of the gas may be obtained by lowering the gas temperature and ultimately the electron temperature and density, and hence reducing the effects of electron collision mixing.

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[1] S. A. Lawton, J. B. Richards, L. A. Newman, L. Specht, and T. A. DeTemple, J. Appl. Phys. **50**, 3888 (1979).
[2] N. G. Basov, V. A. Danilychev, A. Yu. Dudin, D. A. Zayarnyi, N. N. Ustinovskii, I. V. Kholin, and A. Yu. Chugunov, Sov. J. Quantum Electron. **14**, 1158 (1984).
[3] N. G. Basov, A. Yu. Chugunov, V. A. Danilychev, I. V. Kholin, N. N.Ustinovskii, and D. A. Zayarnyi, IEEE J. Quantum Electron. **QE-19**, 126 (1983).
[4] A. Suda, B. L. Wexler, B. J. Feldman, and K. J. Riley, Appl. Phys. Lett. **54**,1305 (1989).
[5] P. J. M. Peters, M. Qi-Chu, and W. J. Witteman, Appl. Phys. B **47**, 187 (1988).
[6] W. J. Alford and G. N. Hays, J. Appl. Phys. **65**, 3760 (1989).
[7] K. Komatus, E. Matsui, F. Kannari, and M. Obara (unpublished).

[8] M. Ohwa, T. J. Moratz, and M. J. Kushner, J. Appl. Phys. **66**, 5131 (1989).
[9] E. L. Patterson, G. E. Samlin, P. J. Brannon, and M. J. Hurst IEEE J. Quantum Electron **26**, 1661 (1990).
[10] R. E. Mickens, Chem. Phys. Lett. **121**, 334 (1985).
[11] T. F. O'Malley, Phys. Rev. **185**, 101 (1969).
[12] D. R. Neal, J. R. Torczynski, and W. C. Sweatt (to be published).
[13] M. Aymar and M. Coulombe, At. Data and Nucl. Data Tables **21**, 537 (1978).
[14] M. Ohwa and M. Obara, J. Appl. Phys. **63**, 1306 (1988).
[15] H. Hokazono, K. Midorikawa, M. Obara, and T. Fujioka, J. Appl. Phys. **56**, 680 (1984).
[16] J. Xu and D. W. Setser, J. Chem. Phys. **92**, 4191 (1990).
[17] W. J. Alford, J. Quantum Electron. **26**, 1633 (1990).
[18] J. Xu and D. W. Setser (unpublished).
[19] Y. Itikawa, At. Data Nucl. Data Tables **21**, 69 (1978).