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Analysis of the UF_6-Xe Direct Nuclear-Pumped Laser

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The quenching role of UF_6 in noble gas nuclear-pumped lasers is examined. Detailed results are presented for the He-UF_6-Xe system. These results indicate that depletion of the atomic ion is the mechanism responsible for the observed behavior. Based on this, it is concluded that UF_6 is not compatible with noble gas lasers. In addition to identifying the quenching role of UF_6, some key rates whose rate constants are not known are identified.

Introduction

IF direct nuclear pumping is to become a viable pumping mechanism for high-power lasers, then a self-critical reactor using gaseous UF_6 must be constructed and a suitable lasing material found.[1] In such a reactor, pumping is achieved by the fission fragments from the $^{235}U(n,ff)FF$ reaction. Experiments with reactor-excited lasers, using UF_6 as a volume source of fission fragments in Xe-Ar mixtures,[2] indicated that lasing is possible at 2.65 μm for a total pressure of about 600 Torr. However, quenching resulted when the partial pressure of UF_6 exceeded 1 Torr.

The object of this investigation is to study the kinetics of nuclear pumping of noble gases by the $^{235}U(n,ff)FF$ reaction and to deduce from that the quenching role of UF_6. Although the experiment employed an Ar-Xe-UF_6 mixture, detailed calculations are carried for a He-Xe-UF_6 mixture. This is because more kinetic data are available for He; moreover, the pumping mechanism is not expected to depend on the buffer gas as long as it is a noble gas with ionization and excitation potentials higher than those for Xe. It will be assumed that He is the dominant substance in the system and, as such, is initially excited and ionized by fission fragments. The energy stored in the excited and ionized states of He is then transferred to the lasing material by charge transfer and Penning ionization. For noble gas lasers,[3,4] recombination of the atomic ion yields the upper laser level, while recombination of the molecular ion yields the lower laser level. Thus, there is an optimum noble gas concentration for which the power output is maximum.

Two possible mechanisms may be advanced for interpreting the termination of lasing observed in the experiments of Ref. 2. The first assumes that electron attachment is the dominant mechanism, while the second is based on the depletion of the atomic ion as a result of neutralization of Xe^+ by UF_6^- and F^-. The calculations indicate that the mechanism depends, to a large extent, on the neutralization rate of UF_4^+ and UF_6^- for which no measurements are available. For the case where the neutralization rate is comparable to rates involving complex ions,[5] the results were not consistent with available experiment. However, when lower rates were assumed, the results were consistent with the observation of Ref. 2. Calculations using the lower rate indicate that depletion of the atomic ion is the mechanism responsible for quenching.

Analytical Formulation

A typical experimental setup of a nuclear-pumped laser consists of a tube filled with fissionable material and a lasing medium surrounded by a moderator and placed in a fast-burst reactor.[3] The high-energy fission fragments, resulting from the interaction of thermal neutrons with the fissionable material, together with the primary and secondary electrons, ionize and excite the background gas. It is generally assumed that the fission fragments resulting from the $^{235}U(n,ff)FF$ reaction fall into two groups: a light group with an average mass number of 96, an average charge of 20 e, and an average initial energy of 98 MeV; and a heavy group with an average mass number of 140, an average charge of 22 e, and an average intitial energy of 67 MeV. Using this information, Wilson and De Young[6] derived the following approximate expression for the power density (w/cm^3) released by the $^{235}U(n,ff)FF$ reaction in representative laser tubes at pressures of the order of 1 atm:

$$P_d \simeq 4.2\times 10^{-16}\frac{\alpha(1-\alpha)p\phi_0}{1-\alpha+\beta\alpha} \tag{1}$$

where α is the UF_6 concentration, p the pressure in atmospheres, ϕ_0 the neutron flux, and β the ratio of the range of fission fragments in He to that in UF_6.

The analysis employed here follows closely that employed in Ref. 7. Thus, particles in different excited states are treated as different species and the multifluid conservation equations are used to describe the resulting plasma. It is shown in Ref. 7 that, for typical laser experiments, the pressure and temperature are essentially constant and the composition is obtained from the relation

$$R_s = 0 \tag{2}$$

where R_s is the production rate of species s resulting from nuclear and kinetic processes. Expressions for R_s follow from the important kinetic processes in the UF_6-He-Xe system and the rates of ionization and excitation of He. The latter follow from Eq. (1) and the appropriate w values, i. e., the energy required to create an excited or an ionized state.

Because we are interested in determining which Xe lines are capable of lasing, all $6s$, $6p$, and $5d$ states of Xe are considered together with the atomic and molecular ions. UF_6 is allowed to dissociate resulting in the formation of F, F^-, F_2, UF_6^-, UF_6^+, UF_5, UF_5^-, UF_4^+, and UF_4. All He excited states are lumped in a single state. The above, together with the electrons, the atomic and molecular helium ions, and the ground states of He, Xe, and UF_6 result in a system consisting of 34 different species. Excimers were not included in this investigation.

Hundreds of reactions involving the above constituents can take place in an UF_6-He-Xe system. However, because of the lack of rate data, especially those involving UF_6 and its dissociation products, only 110 simultaneous reactions are considered. As indicated previously, it is assumed that the power deposited generate ionized and excited states of He.

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Because of the high pressures of interest, He^+ is converted into He_2^+ according to the reaction

$$He^+ + 2He \rightarrow He_2^+ + He \quad (3)$$

with a rate[8] ranging from 6.78×10^{-32} to 10.7×10^{-32} cm^6/s. Both Xe and UF_6 have ionization potentials lower than the ionization and excitation potentials of He. Thus, energy is transferred from helium to Xe and UF_6 by charge transfer and Penning ionization. As a result, Xe^+ is formed according to the reactions[5,9,10]

$$He^* + Xe \rightarrow He + Xe^+ + e, [0.79 \times 10^{-9}] \quad (4)$$

$$He^+ + Xe \rightarrow He + Xe^+, [1.2 \times 10^{-10}] \quad (5)$$

$$He^+ + Xe + M \rightarrow He + Xe^+ + M, [10^{-29}] \quad (6)$$

$$He_2^+ + Xe \rightarrow He + He + Xe^+, [2.2 \times 10^{-10}] \quad (7)$$

$$He_2^+ + Xe + M \rightarrow He + He + Xe^+ + M, [2.4 \times 10^{-29}] \quad (8)$$

with M being a third body and quantities in square brackets indicating rate constants.

The situation involving UF_6 is not clear. In general, charge transfer reactions involving ions with large recombination energies and polyatomic molecules tend to produce mainly dissociative ion products. On the other hand, dimer ions have somewhat lower recombination energies and thus do not cause as much dissociative ionization. Thus, reactions involving He products and UF_6 are taken as

$$He^+ + UF_6 \rightarrow He + F_2 + UF_4^+, [10^{-9}] \quad (9)$$

$$He_2^+ + UF_6 \rightarrow He + He + UF_6^+, [10^{-9}] \quad (10)$$

In the absence of direct measurements, the above rates are estimates taken from Ref. 5.

Electron attachment resulting in the formation of UF_6^- is characterized by a rather low attachment rate.[11] Thus, it is generally believed that the formation of UF_6^- follows from the charge transfer reaction[12]

$$UF_5^- + UF_6 \rightarrow UF_6^- + UF_5, [1.4 \times 10^{-9}] \quad (11)$$

with UF_5^- being formed according to the reaction[11]

$$e + UF_6 \rightarrow UF_5^- + F, [5.0 \times 10^{-10}] \quad (12)$$

The rate indicated for the above reaction is that for thermal electrons with higher rates being observed for more energetic electrons. Another negative ion is formed by the dissociative detachment reaction[13]

$$e + F_2 \rightarrow F^- + F, [3.1 \times 10^{-9}] \quad (13)$$

With the presence of negative and positive ions in the system, the following mutual neutralization reactions are considered.

$$UF_6^- + Xe^+ \rightarrow UF_6 + Xe^*, [5 \times 10^{-7}] \quad (14)$$

$$F^- + Xe^+ \rightarrow F + Xe^*, [2.7 \times 10^{-7}] \quad (15)$$

$$UF_6^+ + UF_6^- \rightarrow UF_6 + UF_6, [5 \times 10^{-7}] \quad (16)$$

$$F^- + F + UF_4^+ \rightarrow UF_6, [3 \times 10^{-25}] \quad (17)$$

$$UF_6^- + UF_4^+ \rightarrow UF_6 + UF_4 \quad (18)$$

The above rates are estimates based on the summary presented in Chap. 24 of Ref. 5. Because the electron affinities of UF_6 and F are about 4.9 and 3.6 eV, respectively, the Xe* in reactions (14) and (15) has, on the average, an energy lower than the lower laser level. Because of this, Xe* in reactions (14) and (15) was taken as the ground state. As previously indicated, a series of rates for reaction (18) were employed ranging from 5×10^{-7} to 5×10^{-10}.

Just as helium atomic ions are converted into molecular ions, xenon molecular ions are formed according to the reaction[14]

$$Xe^+ + Xe + M \rightarrow Xe_2^+ + M, [2 \times 10^{-31}] \quad (19)$$

Recombination of the molecular and atomic Xe ions dictate which laser transition is possible. Although the overall rates are known, the recombination products are not. The rates for the various recombination processes involving xenon can be summarized as follows[15,5]:

$$Xe_2^+ + e \rightarrow Xe^* + Xe, [2.3 \times 10^{-6}(Te/300)^{-1/2}] \quad (20)$$

$$Xe^+ + e + e \rightarrow Xe^* + e, [0.7 \times 10^{-19}(Te/300)^{-4.5}] \quad (21)$$

$$Xe^+ + e + M \rightarrow Xe^* + M, [10^{-26}(Te/300)^{-2.5}] \quad (22)$$

$$Xe_2^+ + e + e \rightarrow Xe^* + M, [0.7 \times 10^{-19}(Te/300)^{-4.5}] \quad (23)$$

$$Xe_2^+ + e + M \rightarrow Xe^* + Xe + M, [10^{-26}(Te/300)^{-2.5}] \quad (24)$$

The two-body recombination reaction, Eq. (20), was studied by Shui et al.,[15] who measured the intensities of various lines in a recombining helium-xenon afterglow. A number of lines of interest could not be examined because of equipment limitations, but the intensities of lines involving $6p$ states were measured. For a plasma state where recombination and spontaneous emission are the dominant loss and production mechanisms, the total production rate is equal to the loss rate for a particular transition divided by the branching ratio for that transition. Given the intensities of several lines involving different states, one can then determine the relative production rates for these states. Using the intensity data of Ref. 15 and assuming that Xe_2^+ recombination produces p states only, the fractions of the total recombination rate populating the various states were established as follows: $6p[5/2]_3$, 0.40; $6p[3/2]_2$, 0.22; $6p[1/2]_0$, 0.17; $6p[3/2]_1$, 0.12; $6p[5/2]_2$, 0.03; and $6p[1/2]_1$, 0.03. In the absence of other measurements, similar fractions were employed in other reactions involving Xe_2^+ recombination.

Data involving Xe^+ recombination are nonexistent. Therefore, it is assumed in this study that the products of Xe^+ recombination are evenly distributed over $6p$ and $5d$ states. Obviously, other choices would yield different results.

Spontaneous emission reactions are important loss and production mechanisms for the noble gas-excited states. All transitions involving $6p$ and $5d$ states were incorporated in this work and their rates were taken from Ref. 16.

The final Xe states that require consideration are the $6s$ states. Reactions involving these states are taken as follows:

$$Xe(6s[3/2]_2) + Xe(6s[3/2]_2) \rightarrow Xe^+ + Xe + e, [0.5 \times 10^{-9}] \quad (25)$$

$$Xe(6s[3/2]_2) + Xe(6s[3/2]_1) \rightarrow Xe^+ + Xe + e, [0.5 \times 10^{-9}] \quad (26)$$

$$Xe(6s[3/2]_1) + Xe(6s[3/2]_1) \rightarrow Xe^+ + Xe + e, [0.5 \times 10^{-9}] \quad (27)$$

$$Xe(6s[3/2]_1) + M \rightarrow Xe(6s[3/2]_2) + M, [2.57 \times 10^{-13}] \quad (28)$$

$$Xe(6s[3/2]_2) + M \rightarrow Xe(6s[3/2]_1) + M, [1.39 \times 10^{-15}] \quad (29)$$

$$Xe(6s[3/2]_2)+M\rightarrow Xe+M,[2.01\times10^{-15}] \quad (30)$$

Rates of reactions (25-27) were estimated from Ref. 17, while those for reactions (28-30) were taken from Ref. 18.

The final rates employed in this model involved the recombination of flourine and UF_6 dissociation products

$$UF_5+F+M\rightarrow UF_6+M,[10^{-31}] \quad (31)$$

$$F+F+M\rightarrow F_2+M,[10^{-32}] \quad (32)$$

$$UF_4+F_2+M\rightarrow UF_6+M,[10^{-31}] \quad (33)$$

The above rates were estimated from Ref. 5.

The solution of the kinetic model gives the number densities of the species present in the plasma. Knowing the number densities, one can calculate the gain coefficient $\gamma(\nu)$ for the various transitions[7]

$$\gamma_{mn}=\frac{c^2 g_m}{8\pi\nu^2}A_{mn}\left[\frac{N_m}{g_m}-\frac{N_n}{g_n}\right]G(\nu) \quad (34)$$

where $G(\nu)$ is the shape factor, N_m and N_n are the number densities of the upper and lower levels, g the degeneracy, A_{mn} the Einstein coefficient for spontaneous transitions from level m to level n, c the speed of light, and ν the frequency. The shape factor is affected by both Doppler and pressure broadening. For pressures of interest here, pressure broadening is the dominant effect and, therefore,

$$G=\frac{2}{\pi}\sum_t \nu_{st} \quad (35)$$

where

$$\nu_{st}=\frac{2}{3}\left[2k\left(\frac{1}{m_s}+\frac{1}{m_t}\right)\right]^{1/2}Z_{st}N_t \quad (36)$$

k is the Boltzmann constant, Z_{st} the collision cross section, s the lasing gas, and t represents other gases in the system. The collision cross sections were calculated assuming a Lennard-Jones potential.[19]

Results and Discussion

Small signal gain coefficients were calculated for various mixtures and pressures and a flux of 3×10^{16} neutrons/cm^2, which is representative of fast burst reactors. No attempt was made to study the effects of neutron flux on gain coefficient because previous work[4,20] has indicated that the relationship is almost linear. Unless indicated otherwise, the pressure is 1 atm. The electron temperature Te is assumed to be 300 K.

Experiments involving pumping of Ar-Xe mixtures by the $^{235}U(n,ff)FF$ reaction[2] indicate that quenching takes place when the UF_6 fraction exceeds 1 in 600. Thus, if the kinetic model is consistent with experiment, one should expect that the gain coefficient will decrease with increasing UF_6 concentration. In an attempt to calculate the small signal gain coefficient, it became evident that it is rather sensitive to the neutralization reaction involving UF_6^- and UF_4^+; a reaction whose rate is not known. Because of this, a parametric study involving the neutralization rate of UF_6^- and UF_4^+ was undertaken. Figure 1 shows the gain coefficient for the 3.99 μm line (the line with the highest gain coefficient for the range of parameters investigated) as a function of UF_6 fraction for a rate coefficient of 5×10^{-7}, which is representative of neutralization of complex ions,[5] while Fig. 2 represents a similar plot for a rate coefficient of 5×10^{-10}. As seen from Fig. 1, the gain coefficient increases with UF_6 fractions exceeding

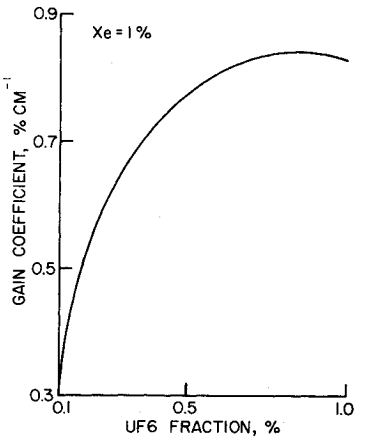

Fig. 1 Effect of UF_6 concentration of gain [high rate for reaction (18)].

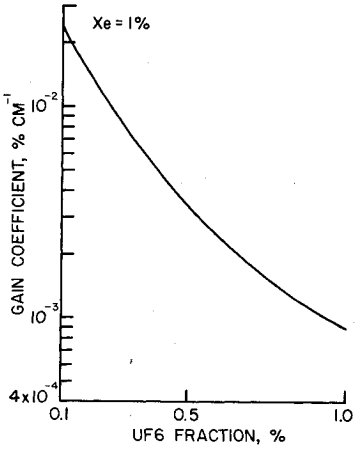

Fig. 2 Effect of UF_6 concentration on gain.

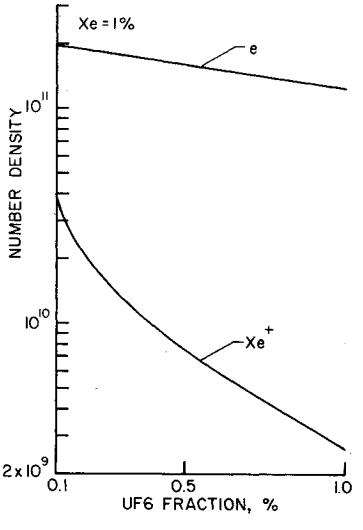

Fig. 3 Effect of UF_6 concentration on electron and X_e^+ number densities.

1/600; the fraction above which quenching took place.[2] This indicates that the neutralization rate coefficient for UF_4^+ and UF_6^- employed in generating Fig. 1 is unrealistic. On the other hand, for a neutralization rate coefficient of 5×10^{-10}, the gain coefficient decreases with UF_6 fraction in agreement with experimentally observed trends. Because of this, the neutralization rate for UF_6^- and UF_4^+ is taken as 5×10^{-10}, and the following results are based on this rate.

There are two possible explanations for laser quenching with increased UF_6 concentration. The first is based on increased electron attachment with the result that fewer electrons will be available for recombination with Xe^+. Such a mechanism will manifest itself by a decreased electron density with an increased UF_6 concentration. The other explanation

Table 1 Gain coefficients

Transition	λ, μm	γ, %cm^{-1}
$5d[1/2]_0 - 6p[1/2]_1$	3.99	0.241×10^{-1}
$5d[1/2]_1 - 6p[1/2]_1$	3.68	0.606×10^{-2}
$5d[1/2]_1 - 6p[3/2]_1$	12.27	0.75×10^{-3}
$5d[1/2]_1 - 6p[5/2]_2$	5.36	0.125×10^{-2}

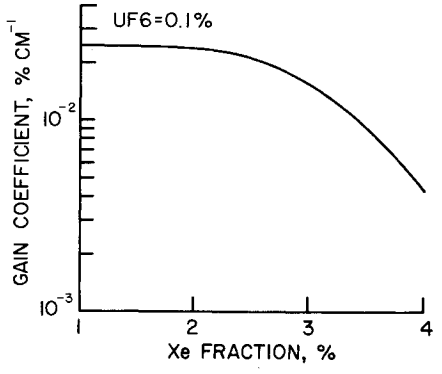

Fig. 4 Influence of xenon concentration on gain.

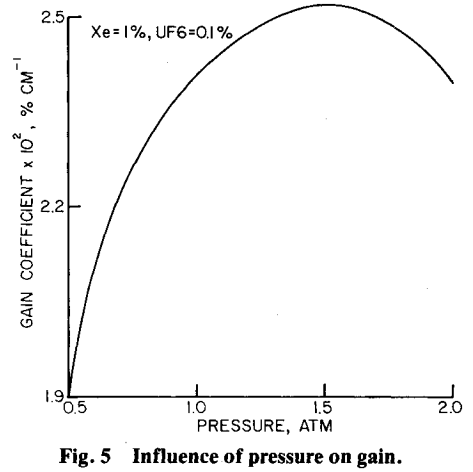

Fig. 5 Influence of pressure on gain.

involves the depletion of the atomic ion by recombination with F^- and UF_6^-. This, in turn, results in a decrease in the number density of the upper laser level. Plots of the number densities of electrons and Xe^+ as a function of UF_6 concentration are shown in Fig. 3. Figure 3 indicates rather clearly that the second mechanism is the one responsible for laser quenching.

The model incorporates all *d* and *p* states and thus is capable of predicting all possible *d* to *p* transitions. Table 1 shows a listing of all transitions that exhibited positive gain at a pressure of 1 atm and Xe and UF_6 fractions of 1 and 0.1%. The experiments of Ref. 2 indicated lasing on the 2.65 μm line, while the present analysis indicates that lasing should have taken place on the 3.99 μm line. This discrepancy is a result of the assumption that the products of Xe^+ recombination are evenly distributed over 6*p* and 5*d* states. Had we assumed that Xe^+ recombination resulted in the preferential pumping of the $5d[3/2]_1$ state, we would have predicted lasing on the 2.65 μm line. Because the 3.99 μm line is the only line that showed measurable gain for Xe concentration less than 4% and UF_6 concentrations less than 1%, the following discussion will be devoted to this line.

For a given pressure and Xe fraction, there is an optimum UF_6 concentration for optimum gain. This is because power deposition is proportional to UF_6 concentration. On the other hand, depletion of Xe^+ increases with UF_6 concentration. These two competing effects point to the existence of an optimum UF_6 concentration. Similarly, because Xe_2^+ increases with increased Xe concentration and because Xe_2^+ recombination yields the lower laser level, there is an optimum Xe fraction for a given power deposition and pressure. These considerations are responsible for the behavior indicated in Figs. 2 and 4.

Figure 5 shows the effect of pressure on the gain coefficient. Increasing the pressure results in an increase in Xe_2^+ and this results in an increase in the lower laser level. On the other hand, increasing the pressure results in more power deposition into the system. Thus, for a given mixture, there is an optimum pressure for optimum gain.

The results also indicate that optimum conditions are influenced by the rates employed in the model. This points to the need for reliable rate measurements involving UF_6 and its dissociation products. The same holds for the recombination products of Xe^+ and Xe_2^+.

Conclusions

The present calculations indicate that depletion of the atomic ion is the mechanism responsible for the observed quenching of laser action in noble gas lasers with increasing UF_6 concentrations. Because of this, UF_6 is not compatible with noble gas lasers.

Acknowledgment

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