

Direct Nuclear Pumping by a Volume Source of Fission Fragments

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A detailed kinetic model is presented for the analysis of nuclear pumped lasers when the pumping is a result of a volume source of fission fragments. The results of the model are employed to study a $^3\text{He-Xe}$ laser. For the range of the pressures, neutron fluxes, and mixtures considered, the gain and power calculations are in good agreement with experiment. Moreover, based on these calculations, it appears that collisional recombination is the dominant pumping mechanism.

Introduction

DIRECT nuclear pumping (DNP) by a volume source of fission fragments was first demonstrated by Jalufka et al.,¹ using a mixture of ^3He and Ar. Since that time, such pumping was demonstrated at a number of laboratories using various mixtures.² Because the neutron mean free path, which is determined by the absorption cross section and pressure, is somewhat long, this form of DNP can result in uniform stable plasmas at high pressures. This is the type of plasma needed for high-power lasers.

The object of this investigation is to study the performance characteristics of this new type of laser system. This entails the development of a detailed and self-consistent kinetic model capable of predicting the behavior of the plasma generated by the interaction of the fission fragments with the background gas. As an illustration, the interaction of fission fragments resulting from the $^3\text{He}(n,p)^3\text{H}$ reaction in $^3\text{He-Xe}$ mixtures is considered. Direct nuclear pumping of a $^3\text{He-Xe}$ laser was demonstrated by De Young et al.³ and Mansfield et al.⁴

In the problem under consideration, both the heavy particles (protons and tritons) and the electrons generated by the interaction of these fission fragments with the background gas play important roles in creating the plasmas; thus, their effects must be considered simultaneously. When calculating possible laser transitions resulting from nuclear pumping one has to treat particles in different quantum states as different species and use the multifluid equations to describe the resulting system. However, before one can formulate the multifluid equations, one has to decide on a kinetic model which incorporates all of the important reactions in the system. For noble gas mixtures, the types of reactions that need to be considered are those given in Ref. 5. The rates of reactions involving electrons and fission fragments require, for their calculations, energy distribution functions, which have to be calculated from appropriate Boltzman equations and cross sections. The remaining rates have to be determined from experiment.

The analysis presented here considers all He excited states with principal quantum number of 5 or less excluding the F states, $5d$, $6s$, $6s'$, $6p$, and $7p$ Xe excited states, ground state of Xe, four excited states of Xe_2 to allow for the formation of

dimers, He^+ , He_2^+ , Xe^+ , Xe_2^+ , and e . Solution of the multifluid equations yields, among other things, the number densities of the preceding species and intensities. This makes it possible to calculate the gain coefficient and power output as a function of pressure, neutron flux, and mixture ratio for a given cavity. The results are compared with available experiments.⁶ Considering the uncertainties in the various rates and cross sections, the predictions are deemed to be in good agreement with experiment.

Analytical Formulation

In nuclear pumping experiments, tubes filled with fissionable material and a lasing medium are surrounded by a moderator and placed in a fast-burst reactor. When the thermalized neutrons interact with the fissionable material, high-energy fission fragments are created and these ionize and excite the background gas. The resulting population inversion follows from excitation by the fission fragments and the resulting electrons and recombination of the ions of the background gas. When ^3He is the fissionable material, the fission fragments are protons with initial energies of 0.57 Mev and tritons with initial energies of 0.19 Mev.

By treating particles in the various excited states as different species, one can utilize the multifluid conservation equations to describe the plasma generated by $^3\text{He}(n,p)^3\text{H}$ reaction. For all of the DNP experiments carried out to date, the steady-state approximation is appropriate. In this approximation, the effects of gradients are assumed negligible. As shown in Ref. 5, such an approximation reduces the species conservation equations, momentum, and energy equations to, respectively,

$$R_s = 0 \quad (1)$$

$$p = \text{const} \quad (2)$$

$$T = \text{const} \quad (3)$$

where R_s is the production rate of species s resulting from both nuclear and kinetic processes, p is the pressure, and T is the gas temperature. To determine R_s , one needs to specify the important kinetic processes in the $^3\text{He-Xe}$ mixture. The types of reactions considered are those indicated in Ref. 5. Reactions pertaining to He and appropriate rates or cross sections are given in Ref. 7, while reactions pertaining to Xe or Xe-He are summarized in the Appendix.

Reactions involving fission fragments or electrons require, as a first step in calculating their rates, appropriate energy distribution functions. The fission fragments' distribution functions are determined from the "neutron transport" version of the Boltzman equation. Assuming an isotropic

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source Q_0 and the $P-1$ approximation, the resulting equation for the flux can be written as⁸

$$-\nabla \cdot (D \nabla \phi) + \sigma \phi = \int \sigma_s (E' \rightarrow E) \phi dE' + Q_0 \quad (4)$$

where ϕ is the heavy particle flux, σ_s is the differential scattering cross section, σ is the total cross section (scattering and charge transfer), $D(E, r)$ is the diffusion coefficient, and E is the energy. The inclusion of a capture cross section in Eq. (4) implies that, once the protons and tritons are neutralized, the resulting particles do not participate in additional ionization and excitation. This will underestimate the effects of direct ionization and excitation by both the fission fragments and electrons. The assumption is reasonable however, if the range of the fission fragments is of the order of the tube radius. Employing a slab-geometry, and assuming a solution of the form

$$\phi = \sum \sin \frac{m\pi x}{\ell} h_m(E) \quad (5)$$

where ℓ is the slab width, one finds that Eq. (4) reduces to a system of uncoupled integral equations which were solved numerically. In addition to providing the ionization and excitation rates, the preceding flux is used in calculating the distribution function of the primary electrons; such a distribution function is needed in the electron Boltzmann equation.⁹

The electron distribution function is obtained from a solution of the electron Boltzmann equation. The equation employed is an extension to a gas mixture of the equation developed in Ref. 9 and can be written as

$$\begin{aligned} & -\frac{v}{3} \frac{\partial}{\partial x_i} \left[-\frac{v}{\Sigma \nu_s} \frac{\partial f_0}{\partial x_i} + \frac{eE_i}{m\Sigma \nu_s} \frac{\partial f_0}{\partial v} \right] \\ & + \frac{eE_i}{3mv^2} \frac{\partial}{\partial v} \left[v^2 \left(-\frac{v}{\Sigma \nu_s} \frac{\partial f_0}{\partial x_i} + \frac{eE_i}{m\Sigma \nu_s} \frac{\partial f_0}{\partial v} \right) \right] \\ & + \frac{m}{v^2} \frac{\partial}{\partial v} \left[\sum \frac{\nu_s}{M_s} v^2 \left(v f_0 + \frac{kT}{m} \frac{\partial f_0}{\partial v} \right) \right] \\ & + \frac{1}{v} \sum_s N_s \sum_j [v'^2 Q'_{sj} f'_0 - v^2 Q_{sj} f_0] + \left(\frac{\partial f_0}{\partial t} \right)_c = 0 \end{aligned} \quad (6)$$

$$\frac{1}{2} m v'^2 = \frac{1}{2} m v^2 + \frac{1}{2} m v_{sj}^2 \quad (7)$$

v is the velocity, e is the electronic charge, m is the electronic mass, M_s is the mass of species s , N_s is the number density of species s , ν_s is the collision frequency with species s , $\frac{1}{2} m v_{sj}^2$ and Q_{sj} are the excitation energy and excitation cross section of level j of species s , and $(\partial f_0 / \partial t)_c$ is the source term resulting from primary and secondary ionization and recombination.

The method of solution of Eq. (6) is essentially that employed in Ref. 9. There is, however, one major complication, resulting from the fact that in this problem there are four ions— Xe^+ , Xe_2^+ , He^+ , and He_2^+ —whose fractions are not known a priori. The fractions of the ions are determined from a solution of the kinetic model, which, in turn, depends on the electron distribution function. Thus, when dealing with a gas mixture, one cannot ignore the coupling between the electron Boltzmann equation and the kinetic model; such coupling results invariably in a rather lengthy iterative procedure.

For a given cavity, use of the threshold condition

$$\gamma = -\ln(r_1 r_2) / 2L \quad (8)$$

where γ is the gain coefficient, r_1 and r_2 are the reflectivities of the mirrors, and L is the length of the cavity, provides the

additional condition needed to calculate the intensity for a given transition.¹⁰ Because the experiments of Ref. 6 considered the 2.027- μ line only, this line was the only line allowed to lase in the present calculations. To calculate the total power, one needs to know the manner in which the intensity varies with area. If the beam is Gaussian, then, for all practical purposes, an area whose diameter is three times the spot size will pass all beam power.¹¹ Thus,

$$P = (3w_s/2)^2 I \quad (9)$$

where P is the power, I is the transmitted intensity, and w_s is the spot size

$$w_s = (b\lambda/\pi)^{1/2} \quad (10)$$

where λ is the wavelength. The quantity b is the equivalent confocal radius and is given by

$$b = [4d(R_1 - d)]^{1/2}, \quad d = L(R_2 - L) / (R_1 + R_2 - 2L) \quad (11)$$

In Eq. (11), R_1 and R_2 are the radii of curvature.

Results and Discussion

The foregoing model is used to study a $^3\text{He-Xe}$ laser, and the results are compared with the experiments of De Young et al.⁶ For a given pressure, temperature, neutron flux, mole fraction, and cavity, one can predict the number densities of the various species and quantum states considered and intensities. This, in turn, yields the gain coefficients and power output. For the calculations presented here, the gas temperature is assumed constant at 300K, the pressure ranged from 400 Torr to 3 atm, while the neutron flux ranged from 10^{16} to 10^{17} neutrons/cm² s. Xenon fractions from 0.1 to 10% were employed.

Figure 1 shows a diagram for the kinetic model employed in this study. As indicated earlier, 59 different states and species were incorporated in this study. Thus, although the 5d, 6p, etc., states are indicated in the diagram as one block, each of the states belonging to 5d, 6p, etc., was treated as a distinct state. This is necessary if one desires to predict lasing on specific lines. It is seen from Fig. 1 that the model allows for direct excitation by the protons and electrons and energy transfer from the metastable and other excited He states. As a result of this energy transfer, Xe^+ and Xe_2^+ ions are formed. When these ions recombine, they produce 6p and 7p and possibly some other states.^{12,13} The cross sections for direct electron excitation are such that rate coefficients are about one to two orders of magnitude higher than those for He. Therefore, in spite of the low Xe fraction employed, direct excitation and recombination should be considered simultaneously when modeling DNP lasers.

Although some information is available regarding Xe_2^+ recombination, similar information is not available for Xe^+ . To infer which states would result when Xe^+ recombines, calculations were carried out in which Xe^+ recombination products were assumed to be, respectively, 6p and 7p states; 6p, 7p, and 5d states; and finally 5d states only. The results, which are summarized in Table 1, were compared with available experiments.^{4,6} Using hole coupled mirrors,⁴ lasing in $^3\text{He-Xe}$ mixtures was achieved for Xe fractions of 1% or less on two lines: the 3.6518 μ , which corresponds to the $7p[1/2]_1 - 7s[3/2]_2$ transition, and the 2.026 μ , which corresponds to the $5d[3/2]_1 - 6p[3/2]_1$ transition. In addition, De Young et al.⁶ measured a gain coefficient of 2%/cm for the 2.026- μ line for the following conditions: a pressure of 1 atm, a neutron flux of 3.23×10^{16} n/cm² s, and a Xe fraction of 1/2%. As is seen from Table 1, the first assumption results in the disappearance of the 2.026- μ line, while the third results in a rather low gain coefficient for the 3.6518- μ line and a rather high gain coefficient for the 2.026- μ line. On the other hand,

Figure 6 shows the effect of Xe fraction on power output. The behavior indicated in the figure, which is typical of available experiment^{4,6} can be traced to the dominant ion in the system. At low ($<0.01\%$), intermediate ($\approx 0.5\text{--}1\%$), and high ($>5\%$) Xe fractions, the dominant ions are He_2^+ , Xe^+ , and Xe_2^+ , respectively. Recalling that Xe^+ is allowed to recombine into $6p$, $7p$, and $5d$ states, while Xe_2^+ recombines into $6p$, $7p$ states only,^{12,13} one can see that the power output increases with Xe^+ , which together with direct excitation is

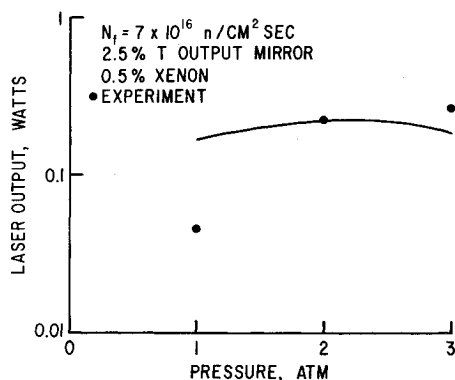


Fig. 5 Power output vs pressure.

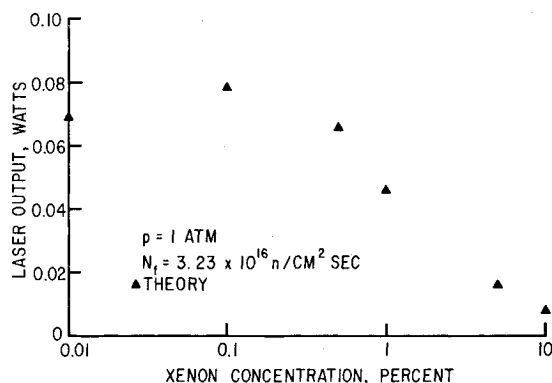


Fig. 6 Effect of Xe fraction on power output.

responsible for populating the upper level, and decreases with Xe_2^+ , which populates the lower laser level.

Concluding Remarks

The results presented here point out the need for accurate rate data and more experiments over a wide range of operating conditions. The fact that the predictions of this model are in good agreement with experiment suggests that the kinetic model presented here is basically correct. The calculations performed here assume a clean system. Because in practice impurities such as N_2 and water vapor are always present, there is a need to modify the present model to allow for the presence of such impurities.

Appendix

When one attempts to predict possible laser transitions in a given laser system, one needs to consider all of the states that may affect or contribute to the various transitions. In practice, lack of rate data makes the inclusion of all pertinent states impossible. For Xe, reliable rate data are nonexistent. Thus, the states considered are those in which some "reasonable" estimate of the excitation cross sections can be made. Thus, the excitation cross sections for the $6p$ and $7p$ states were taken from Ref. 14 despite the fact that some questions were raised regarding the accuracy of these measurements.¹⁵ The excitation cross sections for the optically allowed states $6s'[1/2]_1$, $6s[3/2]_1$, $5d[3/2]_1$, and $5d[1/2]_1$ were determined by the method of Ganas and Green.¹⁶ The measurements of Kuprianov¹⁷ were used to give the excitation cross sections for the metastable states $6s[3/2]_2$ and $6s'[1/2]_0$ with the absolute values determined from Borst.¹⁸ The shapes of the cross sections for the states $5d[1/2]_0$, $5d[7/2]_4$, $5d[3/2]_2$, $5d[5/2]_2$, $5d[5/2]_3$, and $5d[7/2]_3$ were assumed to be similar to those of argon¹⁵ with comparable J values. Their peaks were determined by assuming that the ratios of peaks of d and p states in argon and xenon are similar.

Experimental values for Einstein coefficients for spontaneous emission were used for $6p$ and $7p$ states.¹⁹ Calculated values for some of the other states are taken from Ref. 20. Those not available in Refs. 19 and 20 were calculated using the method of Statz et al.^{21,22}

The excited states produced by the recombination of Xe^+ are assumed to be $6p$, $7p$, and $5d$, while those produced by the recombination of Xe_2^+ are assumed to be $6p[5/2]_3$, $6p[1/2]_0$, $6p[3/2]_2$, $6p[3/2]_1$, $7p[5/2]_3$, $6p[5/2]_2$, and $6p[1/2]_1$ states in a proportion obtained from the coefficients of spontaneous emission and the reported intensities.¹³ Rates involving Xe_2 were taken from Ref. 23. Other pertinent Xe and Xe-He rates were taken from Refs. 24-32.

Proton ionization and excitation cross sections of xenon were taken from Refs. 33 and 34, while those for He were taken from Refs. 35-38. Proton charge transfer cross sections are given in Refs. 39 and 40.

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