## Experimental Elimination of Plasma EH'ects in a Gas-Loaded, Free-Electron Laser

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Saturated oscillation of a free-electron laser operating in the near infrared has been achieved with 200 Torr of H2 gas introduced into the optical cavity. Plasma effects which limited output power in previous gas-loaded, free-electron laser experiments have been eliminated through the use of a small doping fraction of an electron-attachment gas. It is necessary to flow the gas mixture through the oscillator cavity to avoid depletion of the dopant through dissociation. Measured gain is consistent with theory, as is the observed wavelength tuning of 0.7  $\mu$ m.

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Introduction of a gas with refractive index  $n$  into the wiggler chamber of a free-electron laser (FEL) modifies 'the synchonism condition to  $\frac{1}{2}$ 

$$
(n-1) + \frac{\lambda}{\lambda_w} = \frac{1 + a_w^2}{2\gamma^2},
$$
 (1)

where  $\gamma$  is the beam energy,  $\lambda_w$  is the wiggler period, and  $a_w$  is the dimensionless wiggler parameter.<sup>3</sup> Equation (1) shows that the wavelength of a gas-loaded FEL can be changed simply by the variation of the gas pressure. Therefore, a given wavelength can be produced with a lower beam energy when a gas is used, allowing the use of smaller and less expensive accelerators.

In a previous experiment,<sup>2</sup> an infrared FEL<sup>4</sup> was tuned from 4.15 to 3.77  $\mu$ m by the introduction of 100 Torr of  $H_2$ . The operating parameters for the FEL are shown in Table I. However, subsequent experiments with the same FEL filled with 100-200 Torr of hydrogen showed an unexpected rapid decline of peak laser power with pressure. Measured gains at pressures above 100 Torr were much lower than predicted by a model incorporating the effects of the emittance increase caused by scattering in the gas and in the boron nitride foil used to restrict the gas to the region of the wiggler.<sup>5,6</sup> At 200 Torr, only a slight enhancement over the spontaneousemission power level could be produced.

TABLE I. FEL operating parameters used in the gas-loaded FEL experiment.

| 108 cm           |
|------------------|
| $2.3 \text{ cm}$ |
| 4.18 $\mu$ m     |
| 0.98             |
| 73.6             |
| $7\pi$ mm mrad   |
| $1 \text{ mm}$   |
| $25 - 100$ A     |
| $0.5-2$ psec     |
| $350$ psec       |
| $2.7 \mu$ sec    |
| 15 Hz            |
|                  |

Reduced gain can be attributed to detrimental effects on electron beam propagation caused by interactions with the plasma produced by collisional ionization of the gas,  $7$  as well as to the dispersion of the plasma, which alters the group velocity of the optical wave. Previous experiments at similar hydrogen pressures implied that plasma effects would not impair FEL performance. No plasma effects were seen in an experiment in which the beam was propagated through <sup>1</sup> m of hydrogen at pressures from 1 to 950 Torr. $<sup>8</sup>$  However, FEL operation is</sup> sensitive to changes in beam trajectory too slight to have been detected in that experiment. The duration of the optical pulse observed in an earlier gas-loaded FEL experiment at 100 Torr was somewhat shorter than expected,<sup>2</sup> but this was attributed to a drift in electron beam energy over the macropulse.

Instabilities in beam propagation caused by plasma effects result from the low mass of the electrons in the plasma, $7$  and such instabilities can be eliminated by increasing the mass of the negative plasma ions. This is achieved through the use of a gas with a high cross section for electron attachment<sup>9</sup> as a dopant in the hydrogen. As an example, sulfur hexafluoride is an electronattachment gas often used to reduce arcing in highpower microwave guides.

Plasma electrons have an average kinetic energy of  $\approx$  25 eV after their formation through ionization, <sup>10</sup> which is subsequently dissipated through inelastic and which is subsequently dissipated through inelastic and<br>clastic collisions with the neutral background gas.<sup>11</sup> The average lifetime of a plasma electron is determined by the sum of the time needed for the electron to cool to an energy at which attachment becomes significant, plus the average time required before the electron is then captured. For a mixture of  $H_2$  with an electron-attachment doping gas, the plasma electron density is given by

$$
n_e = (S_{\text{H}_2} + S_{\text{dop}})(\tau_c + \tau_a),
$$
 (2)

where  $S_{\text{H}_2}$  and  $S_{\text{dop}}$  are the plasma density source rates in  $cm^{-3}/sec$  due to ionization of the hydrogen and dopant, respectively,  $\tau_c$  is the average cooling time, and  $\tau_a$  is the average attachment time.

In order to minimize  $\tau_c$ , it is desirable to use a gas to



FIG. 1. Plasma electron density as a function of the fraction of  $c$ -C<sub>4</sub>F<sub>8</sub> dopant added to H<sub>2</sub>. The equilibrium density without an attachment gas is determined by three-body recombination.

which relatively high-energy electrons can attach. Many perfluorcarbon molecules have higher attachment cross sections than  $SF_6$  for electron energies greater than 0.4 sections than  $SF_6$  for electron energies greater than 0.4<br>eV.<sup>12,13</sup> Of the nontoxic members of this family, the perfluorocarbon which can attach the highest-energy electrons is perfluorocyclobutane  $(c-C_4F_8)$ , with a significant attachment cross section for electrons up to 0.9 eV.

Figure l shows the plasma density calculated from Eq. (2) with use of  $c$ -C<sub>4</sub>F<sub>8</sub> as the dopant in H<sub>2</sub>. Without dopant, the equilibrium density is  $5 \times 10^{14}$  cm<sup>-3</sup>, determined by the three-body recombination rate, which is strongly dependent on the plasma temperature.<sup>14</sup> The plasma density declines linearly with doping fraction until the concentration of attachment gas is high enough so that  $\tau_a \approx \tau_c$  in Eq. (2). Beyond this concentration, the plasma density is determined mostly by the time required for ionized electrons to cool down to 0.9 eV, so that a minimum plasma density of  $2 \times 10^{11}$  cm<sup>-3</sup> is reached at a doping fraction of 0.8%, a decrease of over 3 orders of magnitude from the density without attachment gas. At even higher dopant concentrations,  $S_{\text{dop}}$  becomes larger than  $S_H$ , in Eq. (2), as the large c-C<sub>4</sub>F<sub>8</sub> molecules contribute more to the ionization rate than do the more numerous hydrogen molecules, and the plasma density begins to increase again.

In addition, the large attachment molecules contribute disproportionately to scattering of the electron beam. When the effect of this additional scattering on gain is



FIG. 2. Comparison of measured and calculated gain for the gas-loaded FEL. Operating parameters are  $\gamma = 74$ ,  $a_w^2 = 0.97$ , and  $\lambda_w = 2.3$  cm. The hydrogen fill gas is doped with 0.06% c-C4F8. Solid curve: calculated gain, which has been normalized to the value without gas but with the foil in place.

considered, the optimal doping fraction of  $c$ -C<sub>4</sub>F<sub>8</sub> is calculated to be only 0.1%.

Changes in the ambient gas temperature may be neglected in the evaluation of the performance of the electron-attachment gas. The hydrogen fill gas is maintained at an ambient temperature of  $T \approx 300$  K by the presence of the aluminum walls of the wiggler chamber. Further, transient temperature variations within one electron beam macropulse must be limited to  $\approx 1\% - 2\%$ in order to maintain a sufficiently stable index of refraction.<sup>15</sup> While the attachment cross section of  $c$ -C<sub>4</sub>F<sub>8</sub> for electrons below <sup>1</sup> eV has been shown to decrease with increasing temperature, <sup>16</sup> at this level of temperature stability it may be assumed to be essentially constant.

The experimental procedure with doped  $H_2$  was as described in Ref. 2, except for the addition of a separate gas inlet and outlet to allow gas flow through the wiggler chamber. Gain and wavelength were measured while the pressure was increased. Laser power increased greatly when a fractional concentration of  $c$ -C<sub>4</sub>F<sub>8</sub> was used. The optimal doping fraction of  $c$ -C<sub>4</sub>F<sub>8</sub> was found to be  $0.06\%$ , in good agreement with the theoretical prediction. Figure 2 shows excellent agreement between measured electronic gain and a theoretical curve predicted from a model including the effects of scattering in the gas.<sup>6</sup> Net gain was reduced from the plotted values by the cavity losses of 7%. Saturation was obtained up to 200 Torr, and higher gas pressures were not attempted because the gas retention foil had not been tested beyond 200 Torr. Table II gives the saturated power and the duration over which the optical signal was saturated at 0,

TABLE II. Saturated power levels in vacuum and at three different pressures of  $H_2$  doped with 0.06% c-C<sub>4</sub>F<sub>8</sub>. The duration of the electron beam macropulse was  $2.7 \mu$ sec.

| Pressure | Saturated power<br>(kW) | Duration of saturation<br>$(\mu$ sec) |
|----------|-------------------------|---------------------------------------|
| 0        | 6.5                     | 1.9                                   |
| 100      | 2.7                     | 1.1                                   |
| 150      | 2.4                     | 1.0                                   |
| 200      | 1.8                     | 0.8                                   |

100, 150, and 200 Torr. The buildup time increases with pressure because of decreasing gain, which reduces the duration of saturation.

The measured wavelengths are compared in Fig. 3 to the theoretical curve calculated from Eq. (1) and the dispersion relation for hydrogen.<sup>17</sup> The measured shift of  $\Delta\lambda$  =0.7  $\mu$ m agrees well with the expected value for wavelength tuning. While this is less than 20% tuning from the  $4.1\text{-}\mu\text{m}$  vacuum wavelength, it demonstrates the potential for much greater relative tuning. The absolute wavelength shift in a gas-loaded FEL can be written from Eq.  $(1)$  as

$$
\Delta \lambda = -\lambda_w (n-1) \tag{3}
$$

The shift  $\Delta\lambda$  is independent of the vacuum wavelength; therefore, tuning of 0.7  $\mu$ m is sufficient to tune an FEL operating at 1.0  $\mu$ m in vacuum all the way through the visible spectrum and down to 0.3  $\mu$ m in the ultraviolet. Below 0.3  $\mu$ m, the index of refraction is enhanced because of the hydrogen resonance near  $0.12 \ \mu m$ , <sup>17</sup> so that tuning from 0.3  $\mu$ m down to the resonant wavelength can be achieved with no further increase in gas pressure, as shown in Fig. 4 of Ref. 2.

Few improvements in present FEL accelerator technology are necessary to achieve tunability over this broad relative range from 1.0 to  $\approx$  13  $\mu$ m. Given the parameters for the Mark-III FEL in Table I, and if we assume an increase in beam energy to  $\gamma = 150$ , operation is possible at 1.0  $\mu$ m with a calculated gain of 22%. Given the feasible experimental improvement of placing the gas retention foil within 10 cm of the wiggler entrance, introduction of 200 Torr of hydrogen would then produce oscillation at 0.3  $\mu$ m with 19% electronic gain. In contrast to the vacuum FEL, gain in a gas-loaded FEL does not degrade severely as the wavelength is reduced, because the electron beam energy is allowed to remain constant. The presence of the gas can actually greatly increase the gain in an FEL utilizing a linear wiggler, as it allows operation in a new mode wherein the electrons travel at a velocity greater than the medium velocity of light.<sup>5</sup> This allows the possibility of operation near resonance at 0.13  $\mu$ m at a hydrogen pressure of only 150 Torr and with electronic gain of 72%.

In order to achieve the saturated laser power levels



FIG. 3. Measured optical wavelength vs hydrogen pressure. The theoretical curve is found from a simultaneous solution of Eq. (1) with the dispersion relation for hydrogen. The length of the bars on the measured data gives the FWHM of the optical power spectrum. The FEL linewidth is determined primarily by an energy slew over the electron beam macropulse.

shown in Table II, it is necessary to flow the gas mixture through the wiggler chamber. When gas flow is turned off, the peak output power obtainable at a given pressure begins to decline, decreasing by several orders of magnitude after a couple of minutes, as shown in Fig. 4. When fresh gas is again flowed, the power level rapidly returns to its saturated level.

The transient behavior without flow is attributable to fragmentation of the  $c$ -C<sub>4</sub>F<sub>8</sub> molecule as a result of collisions with the primary beam and because of dissociative attachment of plasma electrons. For molecules similar to  $c$ -C<sub>4</sub>F<sub>8</sub>, 1%-4% of attachment events are dissociato  $c$ -C<sub>4</sub>F<sub>8</sub>, 1%-4% of attachment events are dissociative.<sup>12,13</sup> A similar fraction of dissociative attachmen for  $c$ -C<sub>4</sub>F<sub>8</sub> leads to the conclusion that all of the parent attachment molecules in the wiggler chamber will be fragmented after 1-4 min of laser operation if the gas is not replaced, producing  $C_3F_5$ ,  $C_2F_3$ ,  $CF_3$ , and atomic fluorine,  $^{18}$  which do not have the high electron attachment cross section of  $c$ -C<sub>4</sub>F<sub>8</sub>.

The data shown in Fig. 4 were obtained with a macropulse repetition rate of 15 Hz. When the repetition rate was decreased to 1.5 Hz, the optical signal did not decrease after the flow of gas was stopped, but remained at the saturated level. This is attributable to the diffusion of unfragmented  $c$ -C<sub>4</sub>F<sub>8</sub> into the wiggler from the vacuum plumbing located at either end of the wiggler chamber, which is filled with a much larger volume of doped hydrogen than is contained in the wiggler itself. At the 15-Hz macropulse repetition rate,  $c$ -C<sub>4</sub>F<sub>8</sub> diffuses into the wiggler at a lower rate than it is dissociated, so that the partial pressure of attachment gas decreases over time.



FIG. 4. Transient behavior of gas-loaded FEL optical power when the flow of H<sub>2</sub> doped with 0.06%  $c$ -C<sub>4</sub>F<sub>8</sub> is stopped. The H2 pressure is 50 Torr. Power is normalized to the saturated power level obtained while doped H2 flowed. The electron beam macropulse repetition rate is 15 Hz.

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Wang, and R. H. Pantell, Rev. Sci. Instrum. 53, 749-757 (1982).

2J. Feinstein, A. S. Fisher, M. B. Reid, A. Ho, M. Ozcan, H. D. Dulman, and R. H. Pantell, Phys. Rev. Lett. 60, 18-20 (1988).

<sup>3</sup>T. C. Marshall, Free-Electron Lasers (Macmillan, New York, 1985), p. 20.

4S. V. Benson, J. M. J. Madey, J. Schultz, M. Mare, W. Wadensweiler, G. A. Westonskow, and M. Velghe, Nucl. Instrum. Methods Phys. Res., Sect. A 250, 39-43 (1986).

<sup>5</sup>M. B. Reid, J. Feinstein, R. H. Pantell, and A. S. Fisher, IEEE J. Quantum Electron. 23, 1539-1544 (1987).

 $6M$ . B. Reid, J. Feinstein, R. H. Pantell, and A. S. Fisher, in Proceedings of the Ninth International Free-Electron Conference, Williamsburg, VA, 14-18 September 1987 (to be published).

 ${}^{7}R$ . B. Miller, An Introduction to the Physics of Intense Charged Particle Beams (Plenum, New York, 1982).

8A. S. Fisher, R. H. Pantell, J. Feinstein, T. L. Deloney, M. B. Reid, and W. M. Grossman, Nucl. Instrum, Methods Phys. Res., Sect. A 250, 337-341 (1986).

<sup>9</sup>L. G. Christophorou, Electron-Molecule Interactions and their Applications (Academic, Orlando, FL, 1984).

<sup>0</sup>R. H. Garvey, H. S. Porter, and A. E. S. Green, J. Appl. Phys. 48, 4353 (1977).

<sup>11</sup>L. S. Kieffer, Joint Institute for Laboratory Astrophysics, Bouder, CO, Information Center Report No. 13, 1973 (unpublished).

<sup>2</sup>A. A. Christodoulides, L. G. Christophorou, R. Y. Pai, and C. M. Tung, J. Chem. Phys. 70, 1156 (1979).

<sup>3</sup>R. Y. Pai, L. G. Christophorou, and A. A. Christodoulides J. Chem. Phys. 70, 1169 (1979).

<sup>4</sup>H. S. W. Massey, E. H. S. Burhop, and H. B. Gilbody, Electronic and Ionic Impact Phenomena (Oxford Univ. Press, New York, 1974), Vol. 4, 2nd ed. p. 2153.

<sup>5</sup>M. B. Reid, Ph.D. dissertation, Stanford University, 1988 (unpublished), pp. 87-102.

 ${}^{6}L$ . G. Christophorou, R. A. Mathis, S. R. Hunter, and J. G. Carter, J. Appl. Phys. 63, 52-59 (1987).

<sup>7</sup>E. R. Peck and S. Huang, J. Opt. Soc. Am. 67, 1550-1554 (1977).

<sup>18</sup>C. Lifshitz and R. Grajower, Int. J. Mass Spectrom. Ion Phys. 10, 25-37 (1972).

<sup>&#</sup>x27;W. D. Kimura, J. A. Edighoffer, M. A. Piestrup, D. Y.