

The Role of Halogen Donors in Discharge Instability of Rare-Gas Halide Excimer Lasers*

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Received 10 May 1989/Accepted 25 July 1989

Abstract. Experimental results on striation instability in a self-sustained discharge-pumped XeCl laser are presented. The presence of HCl proves to be critical to the evolution of this particular mode of instability. The role of halogen donors in discharge instabilities of RGH lasers is discussed. That mixtures containing HCl will be susceptible to striation instability and those with F₂ or NF₃ will not is concluded.

PACS: 34.80, 42.55.H, 52.80

Great strides have been taken toward further development of self-sustained discharge-pumped rare-gas halide (RGH) lasers. Scaled-up XeCl lasers of 22 liters active volume [1], microsecond optical pulse [2] and operation pressure up to 13 atm [3] have recently been realized with corresponding improvements in laser performance. Yet at the same time, discharge instability was found to be the major impediment to continual scaling-up efforts. While XeCl lasers have shown good promise, similar attempts to scale-up rare-gas fluoride lasers (viz. ArF, KrF, and XeF etc.) have so far borne little fruit. Accumulated evidence over the past few years indicates that instability onset is much more rapid in fluoride based RGH lasers [4, 5]. So rapid indeed is the instability growth that hundred nanosecond optical duration rare-gas fluoride lasers are rarely reported.

Discharge instability of RGH excimer lasers was first investigated by Daugherty et al. [6] for an *e*-beam sustained KrF laser. Discharge instability manifested itself as current runaway in these experiments. Lasing ceased when the instability occurred. The cause of the instability was attributed to halogen fuel (F₂ in this case) burn-up during the long pulse discharge which led to an unbridled electron production by two-step ionization. The presence of halogen donor in the laser gas mixture should therefore stabilize the discharge.

More recent study on self-sustained discharge-pumped XeCl laser, however, seemed to indicate a contradictory role of halogen donors. In their study of a long pulse XeCl discharge laser, the excimer laser group of Oxford University observed that lasing terminated prematurely before the pump pulse [7, 8]. Simultaneous with the gradual demise of lasing, the growth of the filamentation mode of instability was also in evidence. Most significantly, the onset of filamentation instability was found to advance with increasing HCl concentration. Long pulse lasing of XeCl laser makes necessary gas mixtures lean in HCl [4, 7]. A theory based on the local halogen depletion was subsequently proposed to directly tie the growth of the filamentation mode of instability with halogen donors [9]. It is presumably applicable to most rare-gas halide discharge lasers.

Most recently, we reported the observation of the striation mode of instability in a XeCl laser [10]. A normally flat-topped XeCl laser pulse was induced to display self-oscillation under high pressure or long preionization delay conditions. This type of discharge instability was termed striation instability because of the strong resemblance it bears to the striated discharge of low pressure glow. It is also to be distinguished from the filamentation mode which precipitates the discharge to collapse and lasing to demise. In the sections to follow, we present experimental results that indicate striation instability can also be induced by varying the concentration of HCl, the halogen donor used in the gas mixture. Reviewing investi-

* This work is based on experiments done when the author was at UCSD

gations performed on RGH laser instability, the role of halogen donors seems far from clear.

We attempt in this paper to resolve some of the apparent conflicts of discharge instability in RGH lasers. Work done on discharge instability of molecular lasers in general will be reviewed briefly, and the difference between *e*-beam-sustained and self-sustained lasers will be discussed. After the presentation of experimental results, a qualitative description of striation instability in XeCl lasers will be given.

1. Discharge Instability in Molecular Lasers

The importance of discharge instability was recognized soon after the demonstration of the first high power molecular lasers (CO and CO₂ lasers). In typical CO and CO₂ laser discharges, the instability usually took the form of constriction and/or striation [11, 12]. Constriction causes the discharge to collapse and the extinction of the laser signal, while in a striated discharge, the optical signal will be modulated or sharply depressed. Both the discharge current and voltage may also exhibit modulated structure. Both modes have their root in an imbalance of electron generation and decay which may develop during the discharge. The striation mode, in particular, is closely connected with negative ion production and is sometimes called the attachment instability. Nighan pointed out in [12] that a necessary condition for the striation mode to occur is:

$$\frac{\partial \alpha_a}{\partial T_e} > 0, \quad (1)$$

where $\alpha_a = k_a n_a$ is the attachment coefficient and T_e is the electron temperature; k_a and n_a are respectively the attachment rate constant and number density of the attaching molecule. Such a condition is satisfied in typical CO₂ laser discharge where the major attaching agent is the CO₂ molecules. For the attachment coefficient α_a of the reaction:



is a rapidly increasing function over the region of T_e of interest during CO₂ laser operation.

2. Instability in *e*-Beam-Sustained and Self-Sustained RGH Lasers

An initial study of discharge instability in RGH lasers was performed on an *e*-beam-sustained KrF laser [6]. Since α_a for the halogen fuel (F₂ or NF₃) used in a KrF laser exhibits monotonically decreasing behaviour of T_e [13], in accordance with (1), discharge striation was not observed. Rather, the onset of instability was marked by a glow to filamentation transition and

runaway of the discharge current was in evidence. Based again on the concept of balancing electron production and decay, Daugherty et al. [6] derived a stability criterion for RGH lasers:

$$\alpha_a > 2\alpha_i^*, \quad (3)$$

where $\alpha_i^* = k_i^* n_m$ is the two-step ionization coefficient with k_i^* as the ionization rate constant and n_m the metastable number density. Hence, it follows from (3) that the role of halogen donor in RGH laser discharge should be a stabilizing one. High halogen concentration and large attachment rate constant should be beneficial to discharge stabilization.

After the early exploratory stage of RGH discharge lasers, the mainstream of development appears to have shifted to the self-sustained version of discharge pumping. The self-sustained XeCl discharge laser, in particular, has shown good promise to be further scaled-up both temporally ($\geq 1 \mu\text{s}$) and volumetrically ($\geq 20 \text{ l}$), whereas rare-gas fluoride discharge lasers of more than 50 ns duration are seldom reported. One of the major obstacles to the continual scale-up effort is again discharge instability.

Investigators from different laboratories have studied discharge instability in RGH lasers both theoretically and experimentally [4, 5, 7–9, 14–17]. Experimental evidence so obtained seems to agree that the onset of filamentation in XeCl is advanced by increasing HCl concentration and the growth of instability is much more rapid in rare-gas fluoride lasers [4, 5, 14–18]. It then appears that (3) cannot satisfactorily explain the adverse effect of halogen donors on discharge instability. To directly account for the prominent role of halogen donors, the Oxford group proposed the local halogen depletion model of instability [14]. Coutts and Webb [9] subsequently derived the following equation to describe the growth of filamentation:

$$1/\tau^2 \sim k_a^2 n_{e0} n_{\text{H.D.}}, \quad (4)$$

where τ is the timescale of growth of filamentation. In (4), τ is expressed as a function of attachment rate constant, electron number density n_{e0} and halogen donor number density $n_{\text{H.D.}}$. According to (4), the filamentation growth is more rapid in halogen-rich and intensely pumped (thus high n_{e0}) laser discharge. Strongly attaching halogen donors such as F₂ also cause a fast growth of instability.

Although (4) appears to agree qualitatively with experimental observations, the definition of τ [9] seems arbitrary. The attachment rate constant k_a is sensitive to electron mole fraction and the gas composition, and it can vary substantially from system to system. Moreover, since k_a changes with time during the discharge, its treatment as a constant in (4) needs to

be justified. Initial interest in discharge instability of self-sustained pumped RGH lasers was aroused by the sudden demise of the laser pulse well before the termination of the pump pulse [7]. Termination of the lasing process is marked by the onset of filamentation instability, which is often characterized by the growth time of n_{eo} [8, 14]. Equation (4) predicts that the instability growth time is inversely proportional to the square root of n_{eo} . Consequently, the laser pulse duration should decrease as n_{eo} (or equivalently, current density) increases. Such drastic reduction in laser pulse duration was indeed reported in [2]. In fact, the characteristic electron number density growth time τ was equated to the laser pulse duration in [16]. If we allow the equation of laser pulse duration for τ , the laser pulse duration should follow the same functional relationship. We recently reported on XeCl laser behavior under intense discharge pumping [19]. High small signal gain ($\geq 0.63/\text{cm}$) was measured at a power deposition rate of $\sim 45 \text{ MW}/\text{cm}^3$, which is about two orders of magnitude higher than that normally employed to excite RGH lasers. Kinetics model calculations (carried out by T. Ishihara at UCSD) also predict that n_{eo} can reach 10^{17} cm^{-3} during the discharge (an order of magnitude estimate of n_{eo} can also be inferred from current density j measurements in [19]). Direct application of (4) would therefore yield a laser duration about one tenth of that under normal discharge pumping. For laser gas mixture containing 2 Torr of HCl, however, the laser duration still amounts to 40 ns (FWHM) under such intense discharge pumping [19].

Unlike e -beam-sustained discharges where the operating E/n is controlled by the externally applied electric field, for self-sustained RGH laser discharges the operating E/n is determined by the discharge conditions and in particular, the gas mixtures. So whereas the ionization rate and attachment rate can be independently adjusted in e -beam-sustained lasers, those in self-sustained lasers cannot. Indeed, the following condition must hold for self-sustained RGH laser discharge:

$$\alpha_a = \alpha_i^* \quad (5)$$

so that electron loss and production are balanced. However, the balance reached in self-sustained RGH laser discharge is an unstable one. Small perturbation of the system (*viz.* a slight local increase of electron number density) will lead to its collapse. Perturbation analysis of an RGH laser system was performed [6] and a stability criterion (3) was established. It is clear from (5) that (3) will not be satisfied in self-sustained discharge. Hence, self-sustained discharge cannot remain stable for long. Owing to the absence of any damping process, no sooner a slight local increase in

electron number density appears than it will be amplified according to the prevailing ionization rate. Consequently, the initially small increase in electron number density eventually grows into local current runaway. Filamentation or arcing thus occurs. The growth of the local electron number density is determined by the ionization rate. Therefore, long pulse stable discharge occurs only when the ionization rate is low. It is also clear from (5) that the two-step ionization rate is proportional to halogen concentration. It then follows that when gas mixtures of high halogen concentration are particularly susceptible to discharge instability, mixtures lean in halogen donors are favorable to long pulse lasing, in agreement with previous observations [4, 7, 15, 16].

Thus far, the discussion has been concentrated on the filamentation mode of discharge instability. But the striation mode has also been reported recently [10]. One of the distinctive features of striation instability has been the self-oscillatory waveforms [11, 12]. In next section, experimental results on striation instability in XeCl laser will be presented, to be followed by a discussion on the origin of this particular mode of instability.

3. Experimental

The experiments were performed using a small discharge-volume device driven by a cable-array pulse-forming line (PFL). Details of the device and detection

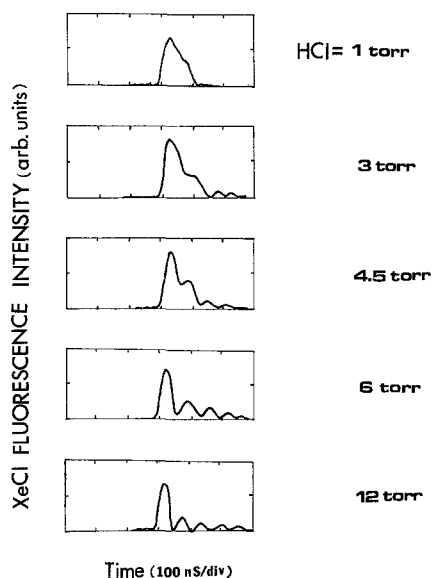


Fig. 1. XeCl fluorescence waveform at various HCl concentrations. The gas mixture has 200 Torr of Xe, 7 atm of Ne. The oscilloscope traces are triggered by the voltage of the preionizer. The preionization delay time is seen to be about 300 ns for all these traces

instrumentation for optical signals were reported previously in [3] and [10], and the detection of discharge current and voltage was reported in [20].

The gas mixtures employed in these experiments consist of 200 Torr of Xe, 7 atm of Ne and HCl from 1 to 12 Torr. XeCl $B \rightarrow X$ fluorescence waveforms at different HCl concentrations are shown in Fig. 1. The oscilloscope was triggered externally by the X-ray preionizer so that the oscilloscope traces of XeCl fluorescence is at Δt after the firing of the preionizer. Δt is fixed at $\sim 0.3 \mu\text{s}$ for all the experimental results presented in this paper. At 1 Torr of HCl, the fluorescence waveform has roughly the shape of a triangular pulse of 80 ns duration (FWHM). With increasing HCl concentration (3 and 4.5 Torr), the peak of the fluorescence gains in amplitude while several satellites begin to appear following the primary hump. At 6 Torr of HCl, the waveform is well modulated in the sense that the fluorescence intensity, after reaching its initial peak, retreats to near the base line, and rises back to a secondary maximum approxi-

mately 30% of the initial peak. The same modulated waveform continues when HCl concentration is raised to 12 Torr where the modulation is 100%. Similar to the waveforms observed in [10], the induced modulation is only approximately periodical with a period of roughly 70 ns.

Discharges using gas mixtures of HCl concentration higher than 15 Torr often collapse into several bright arcs with a complete loss of fluorescence signal. The blue green fluorescence of the triatomic excimer Xe_2Cl was found to share similar characteristics with that of XeCl, except that the secondary maxima of the Xe_2Cl fluorescence are of considerable amplitude, at times amounting to 60% of the initial peak.

Discharge voltage V and current I were also measured as a function of HCl concentration to allow further study of the discharge behavior. Typical V , I waveforms are shown in Fig. 2 for a gas mixture of 200 Torr of Xe, 7 atm of Ne and HCl at 4.5 Torr. The discharge voltage remains roughly constant for some 60 ns after the initial gas breakdown, (the voltage during this quasi-steady stage is termed self-sustained voltage V_{ss}) whereupon it drops to about one fifth of V_{ss} for another 80 ns as the HCl concentration is being depleted under the rather intense pumping (discharge current density $j = 1.3 \text{ kA/cm}^2$). The voltage turns negative thereafter. All the time, XeCl excimers are being generated as the fluorescence pulse is seen to last up to 250 ns (foot to foot). Both the breakdown voltage and V_{ss} increase with HCl concentration (Fig. 3), while the discharge current is insensitive. High frequency ripple attributable to stray capacitance and inductance of the connection cables [10] is visible on the current trace. The period of the ripple remains at 30 ns throughout the experiments.

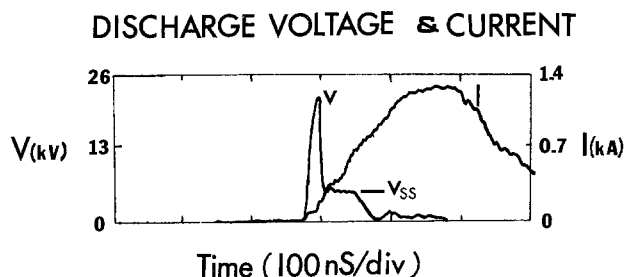


Fig. 2. Discharge voltage V and current I waveforms for a mixture of 4.5 Torr of HCl, 200 Torr of Xe and 7 atm of Ne. V_{ss} is the self-sustained voltage for the initial 60 ns right after the gas breakdown. High frequency ripple riding on the current trace is visible

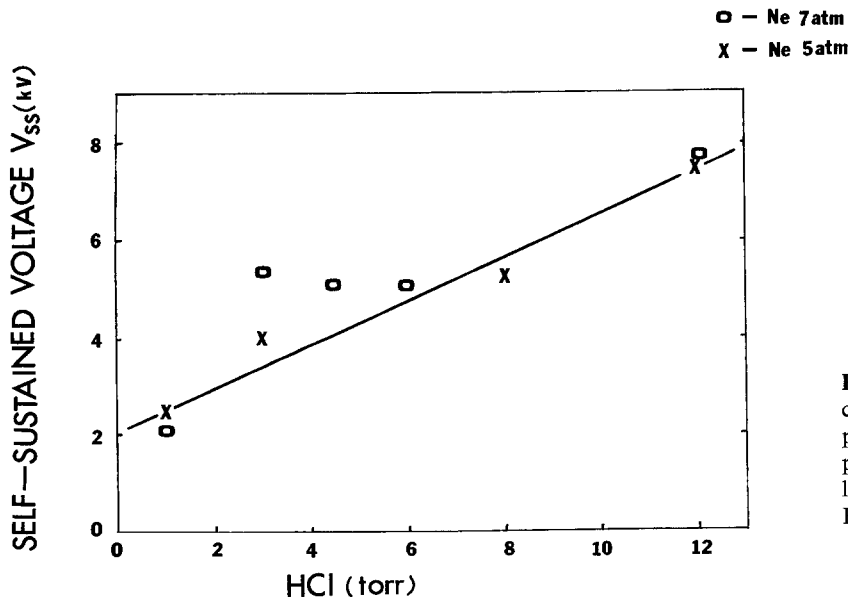


Fig. 3. V_{ss} plotted as a function of HCl concentration at two different Ne pressures but a constant Xe partial pressure of 200 Torr. The solid straight line is only to guide the eye. The effect of HCl concentration is obvious

4. The Origin of Striation Instability

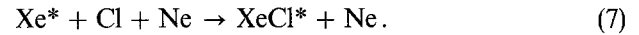
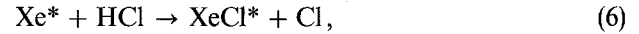
The necessary condition for striation to occur in molecular lasers was given in (1). Striation has not been observed in rare-gas fluoride lasers because the fluorine donors (F_2 or NF_3) are strongly attaching agents whose attachment rate constant k_a are decreasing functions of T_e . This is so because a crossing of the terms of the fluorine donor molecule and the negative ion occurs near the minimum of the potential curve of the donor molecule [21]. But k_a for HCl is an increasing function of T_e for $T_e \leq 0.8$ eV [13]. Employing experimental conditions similar to those of typical XeCl lasers, Letardi and Zheng recently measured the attachment rate constant k_a of HCl in electron swarm experiments [22]. It was found that k_a increases with E/n up to 0.73×10^{-17} V cm² where it reaches a maximum of about 1.1×10^{-10} cm³/s. For XeCl lasers operating below $(E/n)_{ss} = 0.73 \times 10^{-17}$ V cm², condition (1) can be satisfied. The striation instability apparently requires some time to develop so that in the case of lean HCl content, self-oscillation is not observed (Fig. 1); for fluorescence, pulse duration is limited by HCl burn-up. On the other hand, in richer mixtures, self-oscillation is evident some tens of ns into the discharge when $(E/n)_{ss}$ declines further. Such is the case during the long pulse discharge of the XeCl laser in Fig. 1.

4.1. Attachment Rate Constant at Low E/n

Conventional XeCl discharge lasers operate typically at $(E/n)_{ss}$ of about $0.3\text{--}1 \times 10^{-16}$ V cm² [23]. However, Champagne et al. reported $(E/n)_{ss}$ at 0.9×10^{-17} V cm² for a wide aperture XeCl laser [1]. As indicated in Fig. 3 (the solid straight line serves only to guide the eye), the self-sustained voltage V_{ss} increases with HCl concentration, but is otherwise rather insensitive to buffer gas pressure. Because of the high gas pressure employed in the experiments, the operating $(E/n)_{ss}$ is rather low. According to Fig. 3, $(E/v)_{ss}$ amounts to only about 3.2×10^{-17} V cm² for the discharge of 4.5 Torr of HCl during the initial 60 ns of the self-sustained stage. It drops to $\sim 0.6 \times 10^{-17}$ V cm² for the next 80 ns as HCl is being converted to negative chlorine ion. Condition (1) is thus satisfied. Because of the positive dependence of k_a on T_e in XeCl laser discharge, any slight increase in T_e results in an increase of attachment rate, which hastens the electron decay. At the same time, the ionization rate also increases. The two competing electron neutralization processes thus drive the discharge to self-oscillation. When the HCl concentration is varied in our experiments, self-oscillation is induced in XeCl laser discharge.

It is of interest to note that value of $(E/n)_{ss}$ as low as 0.6×10^{-17} V cm² is apparently sufficient for the continual generation of XeCl excimers as the fluorescence

pulse lasts up to 250 ns (foot to foot), albeit with a wavy appearance. At such low $(E/n)_{ss}$, there should be an abundance of Xe* in the discharge. Moreover, because of the burn-up of HCl, the supply of Cl^- will dwindle. On the other hand, atomic chlorine should also be plentiful as a result of the rapid radiative decay of XeCl* and the subsequent dissociation of the ground state. Instead of the ionic formation channel which requires Xe⁺ and Cl^- as precursors, formation of XeCl* could proceed via the neutral channels [4] in the late stage of discharge when $(E/n)_{ss}$ is low:



Under essentially the same discharge conditions, long pulse XeCl lasing with a modulated temporal structure had in fact been reported [3], even though the cause of the modulation was not explained.

In the next two sections, we point out two further mechanisms that might also be responsible for the striation instability in XeCl laser discharge. The first one is the negative differential conductivity (NDC) in gas mixtures with HCl as an impurity. The second one is the attachment-vibration instability.

4.2. Negative Differential Conductivity

It is well known that negative differential conductivity (NDC) can cause oscillation in gases [24] or semiconductors [25]. For instance, the Gunn effect microwave oscillator is a manifestation of NDC in semiconductors [26]. Self-oscillation of voltage and current can occur even in the presence of series ballast resistance [25]. Investigators of NDC in gases often relate its occurrence to the presence of a Ramsauer-Townsend minimum in the momentum transfer cross section, and some inelastic collisional processes in the vicinity of that minimum. But such a combination may not be a necessary condition [27]. In a series of drift chamber experiments, Lando et al. reported observation of NDC for Ar/HCl mixtures irradiated by an electron beam [28]. The gas mixtures used typically consist of a fraction of one percent of HCl, and several atmospheres of Ar. The effective E/n in the experiments ranges from less than 1 Td ($= 10^{-17}$ V cm²) to 12 Td. Such conditions resemble that of XeCl lasers.

The period t of the oscillation induced by NDC can be approximated by [25, 29]:

$$t \cong d/V_d, \quad (8)$$

where d is the electrode spacing and V_d is the electron drift velocity. Since V_d is not measured in our experiments, a calculated value of 8×10^6 cm/s will be used instead [30]. With $d = 1$ cm in our experiments, t is then 125 ns, which is in the same order of magnitude as our measured value of about 70 ns. It is interesting to note that using essentially an identical technique,

electrical conductivity measurements were again performed on Ar/F₂ mixtures [31]. NDC was not found. In Ar/F₂ mixtures, V_d is essentially an increasing function of E/n .

4.3. Attachment-Vibration Instability

In a theoretical study of XeCl laser discharge instability, Eletsii called attention to the strong dependence of k_a of HCl on vibrational excitation [32]. Dissociative attachment of HCl($v=1$) is 38 times more effective than HCl($v=0$) [33]. An increase of electron number density n_{e0} causes the vibrational excitation rate to rise. Consequently, electron decay via dissociative attachment becomes more rapid as more HCl($v=1$) becomes available from the increase in n_{e0} . As pointed out in Sect. 4.1, self-oscillation occurs as the electron temperature T_e enhanced dissociative attachment rate competes with the ionization rate. In the same vein, the competing processes of vibrational enhanced dissociative attachment and two-step ionization could also bring about self-oscillation in XeCl laser discharge. Attachment-vibration instability is again not applicable to fluorine-based gas mixtures because vibrationally excited fluorine molecules exhibit reduced electron affinity [21].

Stability criteria were derived in [32] and it was shown that most XeCl laser discharges will be unstable as a result of attachment-vibration instability. The oscillation period t was also derived:

$$t \cong 1/[4(d\alpha_a/d\varepsilon)k_v n_{e0}]^{1/2}, \quad (9)$$

where ε is the vibrational quanta per molecule and k_v is the HCl $v=0$ to $v=1$ vibrational excitation rate constant. Using parameters typical for our experiments, we estimate $t \cong 50$ ns.

5. Summary

We present experimental results of striation instability in a self-sustained discharge-pumped XeCl laser. Variation in the concentration of hydrogen chloride is effective in inducing instability because it directly causes a change in E/n and a rise and fall in electron temperature. In addition, both NDC and attachment-vibrational instability are candidates as sources of the instability. But whilst computation based on NDC overestimates the oscillation period t , that based on attachment-vibrational instability underestimates it. Since we note both in [10] and here that the self-oscillation is not exactly periodical, the observed instability may be caused jointly by several physical mechanisms. In relation to rare-gas fluoride lasers, since F₂ and NF₃ are strong attaching agents in comparison to HCl, the growth of the filamentation mode of instability will be fast. Furthermore, F₂ and NF₃ exhibit neither NDC nor attachment enhance-

ment due to vibrational excitation, so it seems unlikely that striation instability will occur in rare-gas fluoride lasers. Filamentation instability is expected to prevail in fluoride-containing gas discharges.

Acknowledgements. During the experimental portion of this work, the author was supported in part by U.S. National Science Foundation, Grant Number: CBT-8219521. Some of the references were obtained through SPI internal funds.

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