Rare-Gas-Halide Discharge Stability

M. R. Osborne*

Optics Section, Blackett Laboratory, Imperial College, London SW7 2BZ, UK

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Abstract. An experimental study of the dependence of the output pulse duration of a long-pulse, x-ray preionized excimer laser on the halogen donor molecule is reported. Results obtained at the XeCl* wavelength, using a variety of halogen donors, are in agreement with the halogen donor depletion model of discharge instability. The pulse durations of the rare-gas-fluoride lasers (KrF*, XeF*) have been found to be significantly shorter than would be predicted using this model and the XeCl* data mentioned above. Reasons for this inferior performance are proposed and investigated. In particular, multiple halide ion donation and electrode surface structure effects are discussed in detail.

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Recent advances in preionization techniques and discharge circuit design have greatly extended the output pulse duration available from self-sustained, discharge-pumped XeCl* lasers. Laser pulses of 200–300 ns duration have been produced in several laboratories [1–5], and are now available in a commercial system [6]. Indeed, Taylor and Leopold have demonstrated microsecond pulses, although at somewhat reduced efficiency [3]. A very noticeable feature of these long-pulse excimer lasers is their relatively poor performance on rare-gas-fluoride gas mixtures such as KrF*, which generally exhibit gains which are comparable to, or higher than, XeCl* in short pulse systems. KrF*, ArF*, and XeF* pulses of greater than 50 ns FWHM have seldom been reported, ruling out the use of these lasers in applications which require the reduced peak power of long pulse systems. This is particularly disappointing, as many of the potential applications benefit from either the greater spatial resolution and higher photon energies of ArF* (193 nm, 6.4 eV) and KrF* (248 nm, 5 eV) radiation, or the optical fibre compatibility of XeF* (351 nm) radiation.

The present paper describes an experimental study of the output pulse duration of a multi-wavelength x-ray preionized excimer laser. The results obtained are discussed in the light of the recently improved understanding of the factors governing the stability of the laser discharge [7–9].

1. Background

In excimer lasers subject to extend electrical excitation, collapse of the discharge volume has been found to limit the laser pulse duration [4, 5, 7–10]. Of the several discharge instability mechanisms identified, the one which is dominant in a well-designed laser is caused by local depletion of the halogen donor molecule [7]. This gives rise to the constriction of the discharge into narrow, filamentary channels on a timescale (1/τ) which is a function of the initial concentration of the halogen donor molecule [H.D.], the initial electron number density n_e, and the rate coefficient for electron dissociative attachment to the halogen donor, k [7, 9]. In the case of a laser excited from an idealized pulse-forming-line (PFL) circuit, the following simple expression can be obtained [7]

(1/τ)^2 ~ k^2 n_e [H.D.].

(1)

This scaling law has been experimentally tested previously on the present laser system operating on XeCl* [4, 8, 11]. Fluorescent emission from high-lying ionic levels of the rare gas (Xe II, Xe IV) were used as the diagnostic of the laser discharge. This provides a
precise, although subject to an arbitrary multiplicative factor, definition of the time $\tau$ and allows an accurate comparison between (1) and the experimental results. The exact relationship between the fluorescent emissions and the discharge character is discussed in [8].

Using this technique, the functional dependence of $(1/\tau)^2$ on both electron density ($n_e(0)$) and halogen donor pressure ($[\text{H.D.}]_0$) has been studied [11], and found to be in close agreement with (1) over a relatively wide (factor of $\sim$3) range of parameters. The range of parameters which can be used is limited by the need to restrict measurements to the "steady-state" period of the discharge, i.e. after the current rise time and before the onset of discharge collapse, in order to obtain quantitative results. This limitation is not too severe for previous investigations where $n_e(0)$ and $[\text{H.D.}]_0$ may be chosen at will as free parameters. However, as (1) shows, the corresponding range for the dissociative attachment coefficient, $k$, is only $\sqrt{3}$. Added to this is the fact that $k$ is not a free parameter, but a constant (for a given electron energy distribution) for each halogen donor. Furthermore, $k$ varies by as much as two orders of magnitude between possible halogen donors (e.g. between Cl$_2$ and CCl$_4$). These factors render the high-lying ion fluorescence diagnostic [8] inapplicable to quantitative studies of the dependence of discharge stability on the electron attachment rate, $k$, to the halogen donor although the correct qualitative trend between two halogen donors (HCl and CCl$_4$) has been reported previously [11].

2. Experiment

In light of the above difficulties, the laser output pulse duration was taken as the primary diagnostic. This has the benefits of being easily measured and of being a practically useful quantity, although it has the disadvantage of being rather more difficult to relate to the discharge character than the fluorescent emissions.

The results were obtained using the long-pulse x-ray preionized, PFL driven laser system described in [12] and used previously in [4, 11]. The electrical excitation was maintained at a constant level ($\pm$20%) for $\sim$180 ns [13], and the optical cavity consisted of a plane uncoated quartz flat output coupler, a plane, $\sim$100% Al/MgF$_2$ reflector and an aligned, quartz intra-cavity pressure window.

Figure 1 shows the laser pulse duration (FWHM) obtained from XeCl$^*$ and XeF$^*$ using neon buffered gas mixtures at 2 bar total pressure. Data obtained using KrF$^*$ did not differ by more than 10% from the XeF$^*$ data. The halogen donor pressure was fixed at 2Torr, and the xenon pressure at 15Torr (krypton 30Torr). The discharge current was adjusted via the PFL voltage to give an initial electron number density, $n_{e0}$, of $3.0 \pm 0.3 \times 10^{14}$ cm$^{-3}$ as calculated from

$$n_{e0} = J_0/qV_d,$$

where $J_0$ is the measured initial current density, $q$ the electronic charge and $V_d$ the predicted electron drift velocity in neon for the measured value of $E/N$ (ratio of the electric field to buffer gas number density). The electron energy distribution is assumed Maxwellian, with a mean value, calculated from $E/N$, of $2.2 \pm 0.3$ eV. These experimental conditions were chosen because they lead to reliable and reproducible operation with all of the halogen donors used. Longer duration output pulses are available with, for example, HCl by using greater total pressures, but this leads to difficulties when using CCl$_4$ and NF$_3$.

The dissociative attachment rate coefficients, $k$, in Fig. 1 are, with the exception of that for HCl(v), taken from the published literature [14, 15]. The large error bars displayed result from the dependence of the coefficients on the electron energy distribution, uncertainties in the published values of $k$, and the need to perform a degree of extrapolation of the published results to the electron energies applicable in the present work, e.g. for CHCl$_3$. The rate coefficient for HCl is an effective average, including contributions from several vibrationally excited states. The value used in Fig. 1 corresponds to that measured previously on the pre-