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Irving Bigio

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ELECTRON-ATTACHING GASES IN LASER DISCHARGES:
ALTERED DISCHARGE PARAMETERS AND NEGATIVE-ION PRODUCTION

Irving J. Bigio.
University of California, Los Alamos Scientific Laboratory, Los Alamos, NM 87545.

Two very important parameters of a laser discharge which affect the excitation rates and
laser kinetics as well as the "quality" of the discharge are the electric-field-to-pressure
ratio, E/P, and the preionization. The addition
of an electron-attaching gas to the laser mix can
strongly modify both of these parameters. By
properly accounting for the changes in these
discharge parameters, changes in the laser
kinetics and emission can be correctly predicted.

Two exemplary cases are considered: the addition
of SF$_6$ to N$_2$ lasers, and preionization effects
in eximer lasers.

Several authors have described the effects of
adding SF$_6$ to the N$_2$ discharge laser operating
on the $C^3\Pi_u \to E^3\Pi_g$ transition.$^{1-4}$ The
increased power and modified pulse shapes have
been variously attributed to altered kinetic
mechanisms (e.g. quenching rates) and/or new
excitation pathways, with contradicting
explanations and experimental results.$^{2,3}$

In order to clarify the situation we have
carried out a theoretical and experimental study
which shows that the only important effect of
adding SF$_6$ to the nitrogen discharge is the
modification of electronic-state excitation rates
due to the increased E/P value. (The electro-
negative gas allows a higher voltage buildup
before breakdown and also results in a higher
impedance discharge.$^5$) The Boltzman transport
equation has been solved numerically for the
electron-impact excitation rates into the states
of interest, for different values of E/P.$^6$ The
distribution among vibrational levels is
determined from the Franck-Condon factors. Then
by using the temporal histories of the discharge
parameters, it is possible to predict the temporal
history of the population inversion, hence gain,
hence laser emission. Figure 1 compares the
theoretical and experimental results for the case
of a fast Blumlein-type discharge.

As further proof that only the E/P
modification (and not any kinetic mechanism) was
responsible for the laser effects, a totally
different electro-negative gas was used, CF$_4$,
which produced very similar results to the SF$_6$.

In eximer lasers pumped by a fast discharge,
efficient preionization of the gas is essential
for producing a uniform, arc-free discharge.
Typically a strong uv flash, produced by a string
of sparks, provides the preionization. However,
the effectiveness of the preionization appears to
be relatively independent of the time delay between the spark pulse and the main discharge. This was surprising since the gas mixtures typically contain a few torr of $F_2$, $NF_3$ or $BCl_3$ which have exceedingly fast dissociative-electron-attachment rates. Thus, essentially all the free electrons produced by the uv flash would disappear within $<10^{-7}$ seconds, whereas uniform discharges are produced even with delays exceeding one $\mu$sec.\(^7\)

Haia\(^8\) has suggested that the negative ions produced by dissociative-attachment reactions such as $F_2 + e + F + F^- \rightarrow F^+ + X + e$ have a low enough electron affinity (\(\sim 3\) eV) that when the main discharge field is applied they undergo collisional detachment: $F^- + X + F + X + e$. Thus, the negative ions themselves provide a secondary source of electrons right at the time of the main discharge.

To test this hypothesis experimentally we chose a totally different method of preionization: a radioactive $\alpha$-emitter, americium-241. The advantages of a radioactive source for this study are that it produces no electric fields which might alter the discharge parameters, and it is much easier to estimate the electron production rate. The experimental details are described in Ref. 9. For our experimental geometry, the relation which determines the density of electrons, $n_e$, is

$$\frac{dn_e}{dt} = \frac{AQ}{eR^2L} - \alpha n_e \beta n_i - \beta n_e$$

(1)

where $A$ is the total source activity, $Q$ is the number of electron-ion pairs produced per alpha particle, $R$ is the range of the alphas in the given gas, $L$ is the length of the strip source (= the length of the laser discharge region), $n_i$ is the ion density, while $\alpha$ and $\beta$ are the electron-ion recombination and electron-attachment coefficients, respectively. Since for the electronegative eximer gas mixtures $\beta >> \alpha n_i$, equation (1), at equilibrium, becomes

$$n_e = \frac{AQ}{eR^2L} \beta$$

(2)

For our parameters equation (2) gives us a value $n_e \lesssim 1$ cm$^{-3}$, which is obviously insufficient for stabilizing the discharge. However, since essentially all the electrons produced by the $\alpha$-source result in negative ions, $F^-$, the density of negative ions, $n_{F^-}$, is determined by

$$\frac{dn_{F^-}}{dt} = \frac{AQ}{eR^2L} \alpha (n_{F^-})^2$$

(3)

where the main loss mechanism for the $F^-$ is assumed to be three-body ion-ion recombination (rate denoted by $\alpha^-$): $F^- + X^+ + M \rightarrow$ neutral products. Thus, we get a negative-ion density $n_{F^-} \approx 5 \times 10^8$ cm$^{-3}$ at equilibrium. This density of low-affinity negative ions is sufficient to aid in stabilizing the main discharge for $E/P$ values $\gtrsim 100$ V-cm$^{-1}$-torr$^{-1}$, especially if the initial voltage risetime is fast (\(\sim 10^{-8}\) sec.).

In summary, whenever an electron-attaching gas is used in a laser discharge, it is wise to examine carefully the effects of increased $E/P$ and the role of negative-ions in the discharge kinetics.

References:
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