Optical breakdown of Al vapour based on the detailed kinetic model with account for the basic collision and radiation-induced transitions

Optical breakdown : Analysis Baseline

The objective is to determine the threshold intensity $G^* = G^*(\eta\omega_l)$ versus laser wavelength. The analysis is performed for constant intensity laser pulse and the pulse duration is not restricted in advance.

It is postulated that optical breakdown have occurred in the medium if the $\nu_{en} < \nu_{ei}$ condition is fulfilled, ν_{en}, ν_{ei} are the frequencies of electron-neutral and electron-ion collisions.

The initial state of the metal vapor is defined by setting its temperature T_0 and density ρ_0 equal to the temperature and density at outer boundary of the Knudsen layer while the surface of the target is at the equilibrium boiling temperature : $T_0 = 0.2\text{eV}$, $N_0 = 6 \times 10^{18}\ \text{cm}^{-3}$, $N_e = 3 \times 10^{14}\ \text{cm}^{-3}$.

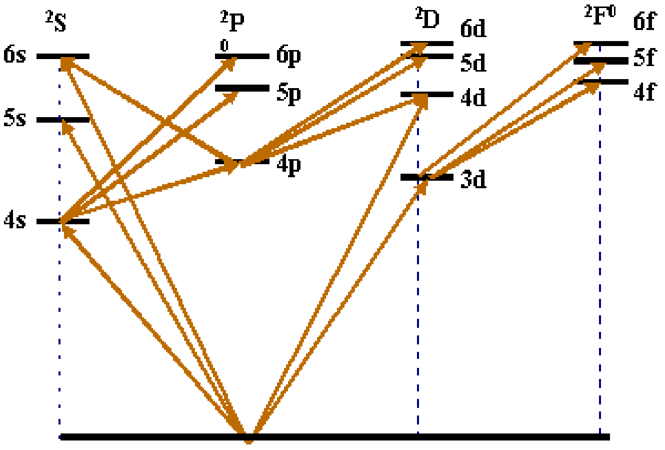
Gaseous medium is assumed to be spatially homogeneous, optically thin, no spatial effects are analysed.

Kinetics of laser-induced breakdown in Al vapour

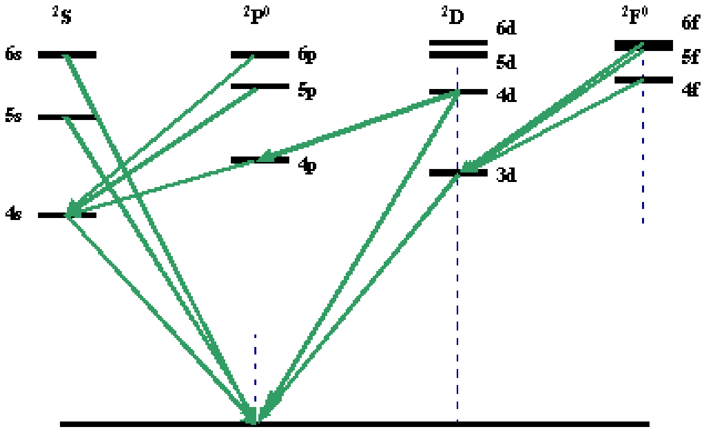
Type of transition	Participating particles	Rate coefficient
Electron impact ionization	$Al_m + e \rightarrow Al_i + e + e$	α_m [$\text{cm}^3\ \text{s}^{-1}$]
Three-particle recombination	$Al_i + e + e \rightarrow Al_m + e$	β_m [$\text{cm}^6\ \text{s}^{-1}$]
Collisional excitation/ de-excitation	$Al_m + e \leftrightarrow Al_n + e, n > m$	k_{mn}, r_{mn} [$\text{cm}^3\ \text{s}^{-1}$]
Spontaneous radiative decay	$Al_n \rightarrow Al_m + \hbar\omega_{mn},\ n > m$	A_{mn} [s^{-1}]
Laser-induced photoionization	$Al_m + \hbar\omega_l \rightarrow Al_i$	ν^l_m [s^{-1}]
Laser-induced photorecombination	$Al_i + e + \hbar\omega_l \rightarrow Al_m + \hbar\omega_l$	R^l_m [$\text{cm}^3\ \text{s}^{-1}$]
Laser-induced photoexcitation	$Al_m + \hbar\omega_l \rightarrow Al_n,\ n > m$	ν^l_{mn} [s^{-1}]
Continuum radiation photoionization	$Al_m + \hbar\omega_c \rightarrow Al_i$	ν^c_m [s^{-1}]
Continuum radiation hotorecombination	$Al_i + e + \hbar\omega_c \rightarrow Al_m + \hbar\omega_c$	R^c_m [$\text{cm}^3\ \text{s}^{-1}$]
Continuum radiation photoexcitation	$Al_m + \hbar\omega_c \rightarrow Al_n, n > m$	ν^c_{mn} [s^{-1}]

Breakdown kinetics

Electron configuration of Al atom taken into account by the model



Collisional transitions in Al atom



Radiative transitions in Al atom

Mathematical Model

Concentrations of neutral N_0, excited N_m, charged N_i particles and electrons N_e, $N_i \approx N_i$ are described by the system of rate equations:

$$\frac{dN_0}{dt} = -\sum_{j=1}^{M}(k_{0j}N_0 - r_{j0}N_j)N_e - (\alpha_0 N_0 - \beta_0 N_i N_e)N_e + \sum_{j=1}^{M} A_{j0}N_j(x) -$$

$$- \sum_{j=1}^{M} \nu^{c}_{0j}\left(N_0 - \frac{g_0}{g_j}N_j\right) - \nu^{\ell}_{01}\left(N_0 - \frac{g_0}{g_1}N_1\right) - \left(\nu^{l}_{0} + \nu^{c}_{0}\right)N_0 + \left(R^{c}_{0} + R^{\ell}_{0}\right)N_i N_e$$

$$\frac{dN_m}{dt} = \sum_{j=0}^{m-1}(k_{jm}N_j - r_{mj}N_m)N_e - \sum_{j=m+1}^{M}(k_{mj}N_m - r_{jm}N_j)N_e - (\alpha_m N_m - \beta_m N_i N_e)N_e +$$

$$+ \nu^{\ell}_{m-1,m}\left(N_{m-1} - \frac{g_{m-1}}{g_m}N_m\right) + \sum_{j<m}^{M} \nu^{c}_{jm}\left(N_j - \frac{g_j}{g_m}N_m\right) - \sum_{j>m}^{M} \nu^{c}_{mj}\left(N_m - \frac{g_m}{g_j}N_j\right) -$$

$$- \sum_{j=0}^{m-1} A_{mj}N_m + \sum_{j=m+1}^{M} A_{mj}N_j + \left(R^{c}_{m} + R^{\ell}_{m}\right)N_i N_e - \left(\nu^{c}_{m} + \nu^{\ell}_{m}\right)N_m, \qquad m = 1, \ldots, M$$

$$\frac{dN_i}{dt} = \frac{dN_e}{dt} = \sum_{m=0}^{M}(\alpha_m N_m - \beta_m N_i N_e)N_e + \sum_{m=1}^{M}\left(\nu^{c}_{m} + \nu^{\ell}_{m}\right)N_m - \sum_{m=1}^{M}\left(R^{c}_{m} + R^{\ell}_{m}\right)N_i N_e$$

Energy balance of the nonequilibrium plasma is characterized by two temperatures: T_e for the electrons and T_g for the heavy particles (atoms and ions). The equations are written as:

$$\frac{3d(N_e T_e)}{2dt} = \left(\left(\mu G - \frac{3}{2}\delta\left(T_e - T_g\right)\right)(\nu_{en} + \nu_{ei}) - \sum_{m=0}^{M} Q_{m,J} - \sum_{m=0}^{M-1} Q_{m,\Delta E}\right)N_e + Q_\Phi$$

$$\frac{3}{2}\frac{d}{dt}(N_g T_g) = \frac{3}{2}\delta\left(T_e - T_g\right)(\nu_{en} + \nu_{ei})N_e$$

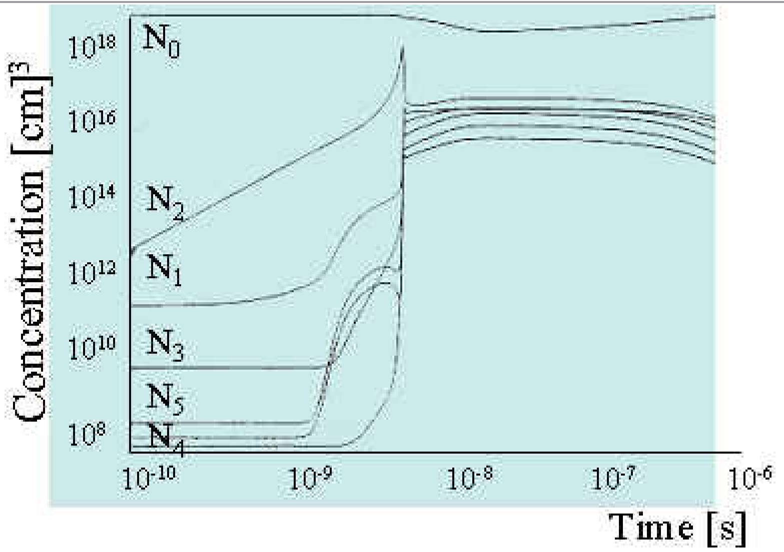
$$\delta = \frac{2m}{M} \qquad \mu = \frac{4\pi e^2}{mc\left(\omega^2 + (\nu_{en} + \nu_{ei})^2\right)} \qquad N_g = \sum_{m=0}^{M} N_m$$

$$Q_{m,J} = J_m(\alpha_m N_m - \beta_m N_i N_e)$$

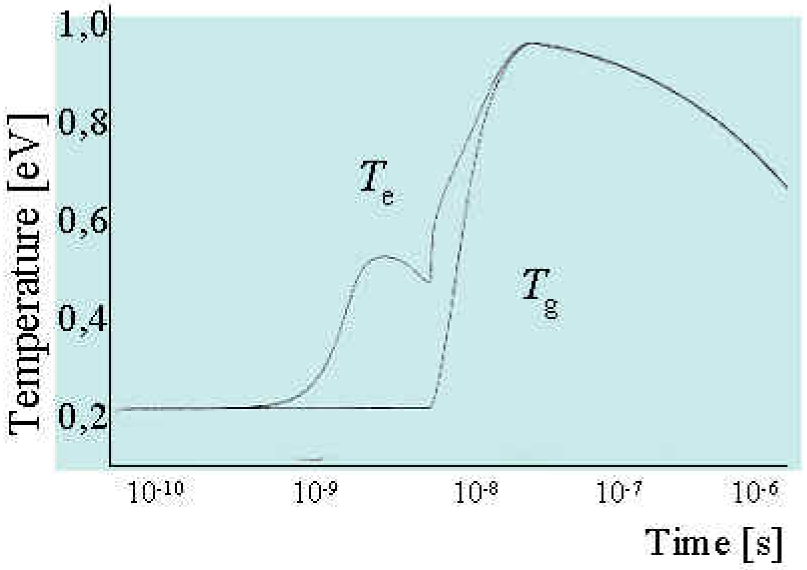
$$Q_{m,\Delta E} = \sum_{j=m+1}^{M} \Delta E_{jm}(k_{mj}N_m - r_{jm}N_j)$$

$$Q_\Phi = \sum_{m=0}^{M}\left[\left(\hbar\omega_\ell - J_m\right)\left(\nu^{\ell}_{m}N_m + R^{\ell}_{m}N_e N_i\right) + \left(\hbar\omega_c - J_m\right)\left(\nu^{c}_{m}N_m + R^{c}_{m}N_e N_i\right)\right]$$

Here : m, M are masses of electrons and atoms, ν_{en}, ν_{ei} are the frequencies of electron - neutral and electron - ion collisions, E_m denotes the excitation energy of the m-th state, J_0, J_m are ionization energy of ground and excited states.

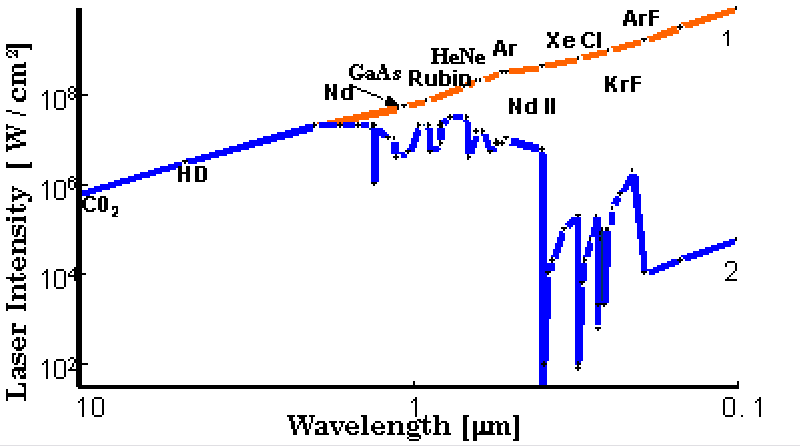


Excited state concentrations for laser intensity $G_0 = 2.10^7$ W/cm^2, wavelength λ = 0.248 µm and 30 ns pulse duration.



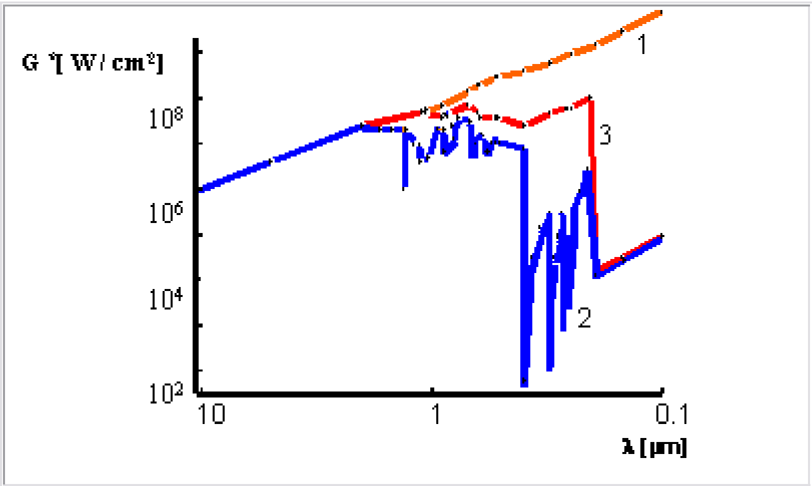
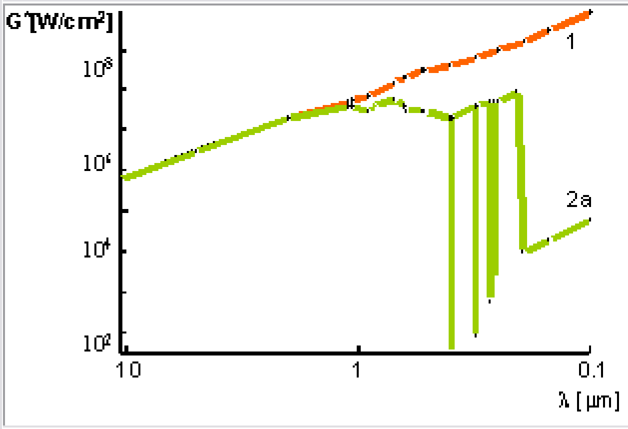
Temperatures of electrons T_e and heavy particles T_g for laser intensity $G_0 = 2.10^7$ W/cm^2, λ = 0.248 µm and 30 ns pulse duration.

Breakdown kinetics



Spectral dependence of the breakdown threshold intensity G* = G*(λ)

(l) : the model including only the collisional processes and the spontaneous decay, (the so-called classical model i.e. without photo-induced transitions) ;
(2) the complete model. Note that the duration of laser action is not restricted beforehand. Therefore the found values of G* are the minimum ones for the given wavelength.

	Spectral dependence of the breakdown threshold intensity : (1) the classical model ; (2) the complete model ; (3) the classical model + laser-induced photoionization.
	Spectral dependence of the breakdown threshold intensity : (1) the classical model ; (2a) the classical model + laser-induced photoionization + resonance photoexcitation

Conclusion

It is shown that in the UV- spectral range:

Resonance/ non-resonance photo excitation and ionization from the ground and first excited states are the major mechanisms of optical breakdown;
Non-resonance photo-excitation depends significantly on the broadened line profile formed by simultaneous contribution of several broadening mechanisms;
The threshold radiation intensities in the UV-range are 2-3 orders of magnitude lower then the ones predicted by the classical approach (i.e. without photo-induced transitions).