Dedicated to jubilee of
Vladimir Zakharov

High-temperature nonlinear phenomena in LNB crystals

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Physical background: Nonlinear optical material

$LiNbO_3$ crystals – Silicon of optics? Why?

- Cheap and robust ferroelectric, $T_c \approx 1200$℃, C_{3v} symmetry
- Transparent in the visible-to-infrared range ($\approx$ 4 eV forbidden gap)
- High $\chi^{(2)}$ nonlinearity, large electro-optic coefficients
- Domain engineering: PPLN structuring, photonic crystals, quasi-phase-matching, etc
- Sensitization by doping (Fe, Cu,...). Light-induced charge transport
- Generation of large electric fields and refractive index changes (reversible optical recording)

Serious drawback: Optical damage

Unwanted generation of strong nonlinear index changes and deterioration of light beams for $I > (10^3 - 10^4)$ W/cm^2

A closer look at light-induced charge transport

$$j_e = -\beta I + e\,\mu_e n_e E + eD_e \frac{\partial n_e}{\partial z}$$

$$\alpha = N_e \sigma_e$$ **Light absorption coefficient**

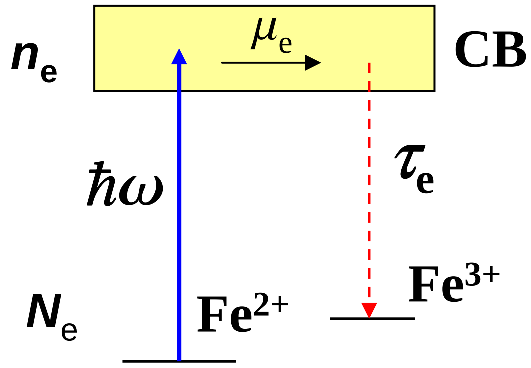
$$n_e = \frac{N_e \sigma_e I}{\hbar\omega}\tau_e$$ **Free electron concentration**

$$\beta = e\frac{N_e \sigma_e}{\hbar\omega} l_{pv}$$ **Photovoltaic coefficient**

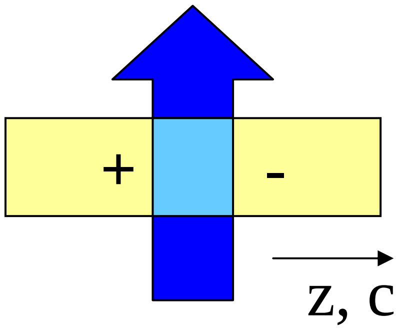
$$D_e = \frac{\mu_e T}{e}$$ **Einstein relation**

$$E_{pv} = \frac{l_{pv}}{\mu_e \tau_e} \approx 10^5 \text{ V/cm}$$ **Photovoltaic field**

$$\Delta n \approx 10^{-3}$$ **Index change**



$n_e << N_e$



Why high temperature?

An ultimate goal of electron engineering:

To remove photo-excitable electrons far below the background level, $N_e \ll 10^{15}$ cm^{-3}, i.e., to make much lower the Fermi level

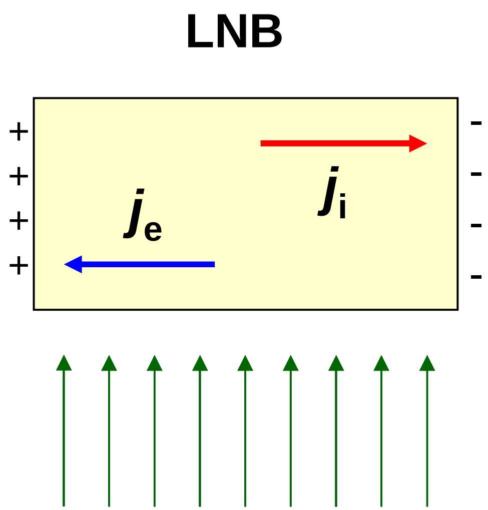
At room temperature electrons are the only charge carriers, $j_e = 0$ in steady state, and the arising electric field E_{pv} blocks the removal completely

For $T > 200$ ℃, ions (H^+, Li^+, …) become mobile: $j_i = e\,\mu_i(T)\,N_i\,E$
In the electric equilibrium $j_\Sigma = j_e + j_i = 0$, but $j_e \neq 0$
The electron removal persists

In other words, ions compensate the removed electrons and maintain the charge quasi-neutrality

A trivial example

Steady state



$$j_e + j_i = -j_{pv} + (\sigma_e + \sigma_i)E = 0$$

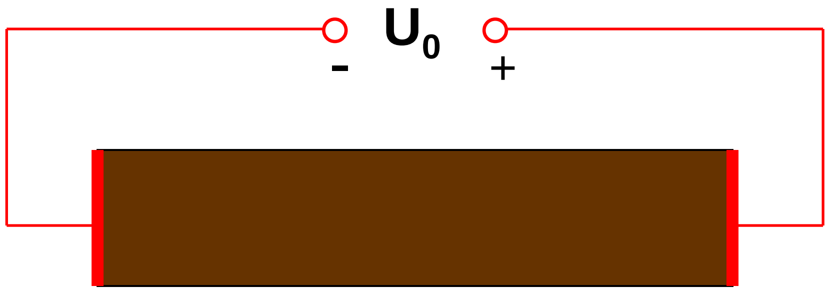
$$E = \frac{E_{pv}}{1 + \sigma_e / \sigma_i}$$

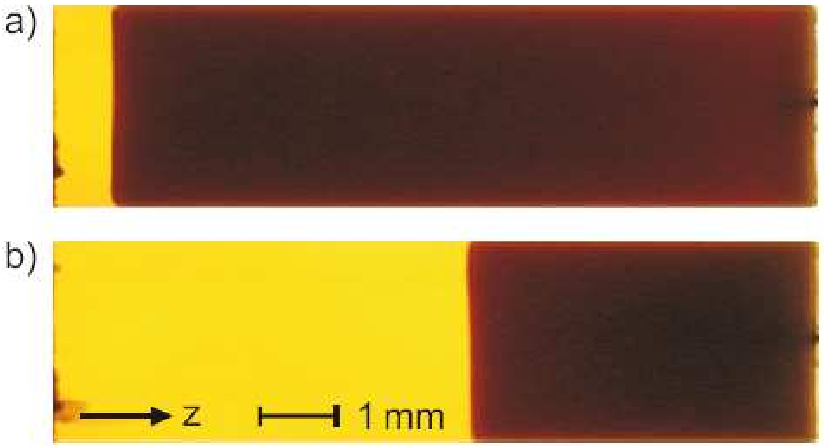
$$| j_{e,i} | = \frac{\sigma_e \sigma_i}{\sigma_e + \sigma_i} E_{pv}$$

There are permanent fluxes of electrons and ions to the right

Ultra-slow shock wave of electron density
(PRL 2008)

First experiments:





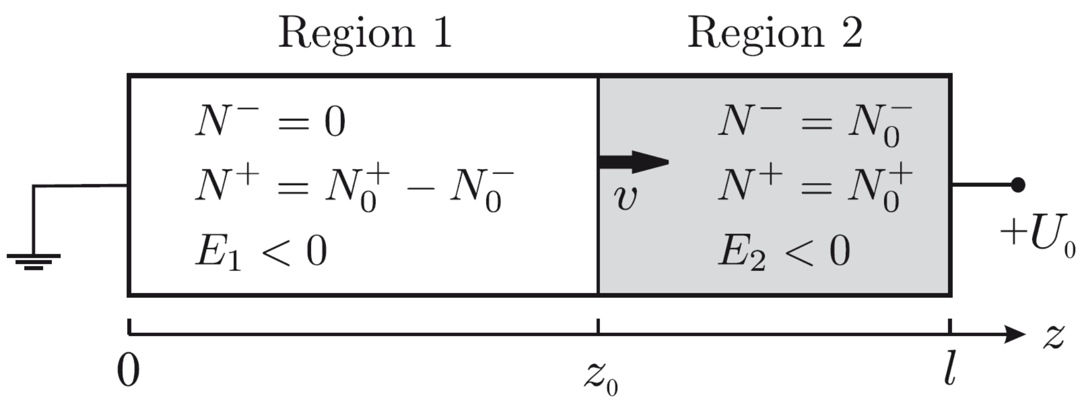
LNB:Fe, $N_e^0 \sim 10^{19}$ cm^{-3}

T = 600 ℃, U_0 = 1 kV

$t \sim$ 1 hour, w < 100 μm

Nobody observed things like this earlier

Model



Assumptions and notation:

$$N^- = N_e\,,\quad N^+ = N_i \;\; (N_{Li^+} \approx 10^{20}\ \mathrm{cm}^{-3})$$

$$j_\pm = e\mu_\pm N^\pm E,\quad \mu_\pm \propto \exp(-\varepsilon_\pm / T)$$

$$z_0 = z_0(t),\quad v = v(t)$$

$$E_0 = U_0 / l,\quad \xi = z_0 / l$$

Main physical requirement:

$$v = -\mu\_E_2$$

Main predictions:

$$v = \mu\_E_0 /(1 + a\xi)$$

$$\xi = \left(-1 + \sqrt{1 + 2at / t_0}\right)/a$$

$$a = (\mu\_ / \mu_+ + 1)/\left(N_0^+ / N_0^- - 1\right)$$

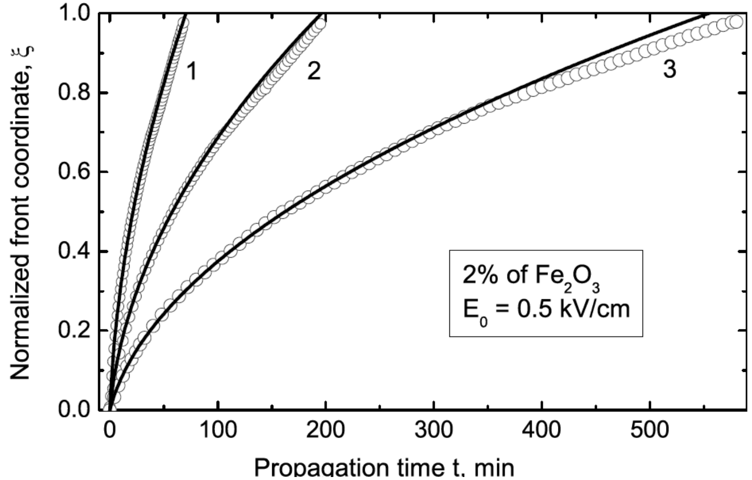
$$t_0 = l/\mu\_E_0$$

$$t_\Sigma = t_0(1 + a/2)$$

More predictions:

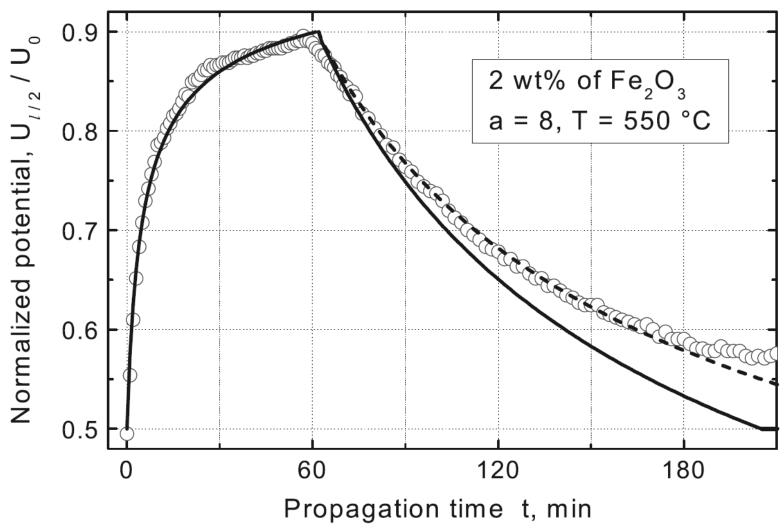
Time dependence of the potential $U_{l/2}(t)$

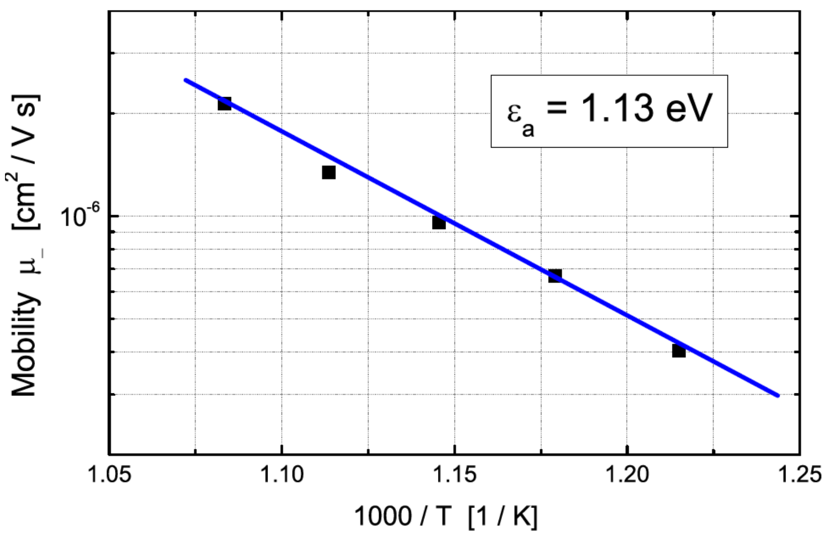
Comparison with experiment



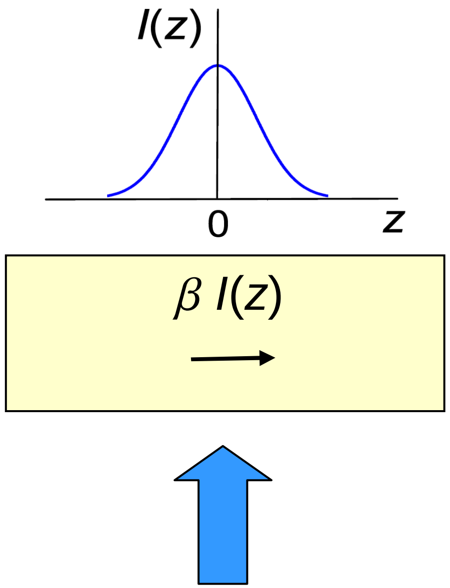
1: T = 650 ℃, a = 7.4, t_0 = 15 min
2: T = 600 ℃, a = 9.2, t_0 = 35 min
3: T = 550 ℃, a = 9.8, t_0 = 94 min

No doubts remain





Optical cleaning: How to remove electrons optically (NP-09)



$$\beta\, I(z)$$

The photovoltaic drift velocity

Initial set of equations

$$\frac{\partial N_e}{\partial t} + \frac{\partial J_e}{\partial z} = 0, \qquad \frac{\partial N_i}{\partial t} + \frac{\partial J_i}{\partial z} = 0$$

$$J_e = \underline{\beta\, I\, N_e} - \mu_e\, n_e E - D_e\, \partial n_e / \partial z$$

$$J_i = \mu_i\, N_i E - D_i\, \partial N_i / \partial z$$

$$\frac{\partial E}{\partial z} = \frac{4\pi}{\varepsilon}\left(N_i - N_e - N_i^0 + N_e^0\right)$$

$$n_e = \frac{s N_e I \tau_e}{\hbar\omega}$$

Zero-model of optical cleaning

Assume:

$$\sigma_e^0 << \sigma_i^0, \quad N_e^0 << N_i^0, \quad E << E_{pv}$$

A moving Gaussian light pattern
A moving coordinate frame

$$I = I_0\, e^{-(z-\mathrm{v}\,t)^2/z_0^2} = I_0\, e^{-\hat{z}^2/z_0^2}$$

$$\beta\, I = \mathrm{v}_0\, e^{-\hat{z}^2/z_0^2}, \quad \mathrm{v}_0 = \beta\, I_0$$

Single equation for N_e

$$\frac{\partial N_e}{\partial t} + \frac{\partial}{\partial \hat{z}} N_e \underbrace{\left(\mathrm{v}_0\, e^{-\hat{z}^2/z_0^2} - \mathrm{v}\right)}_{u(\hat{z})} = 0$$

To be solved semi-numerically by the characteristic method

$$\frac{d\hat{z}}{dt} = u(\hat{z}), \quad \hat{z}(0) = \xi$$

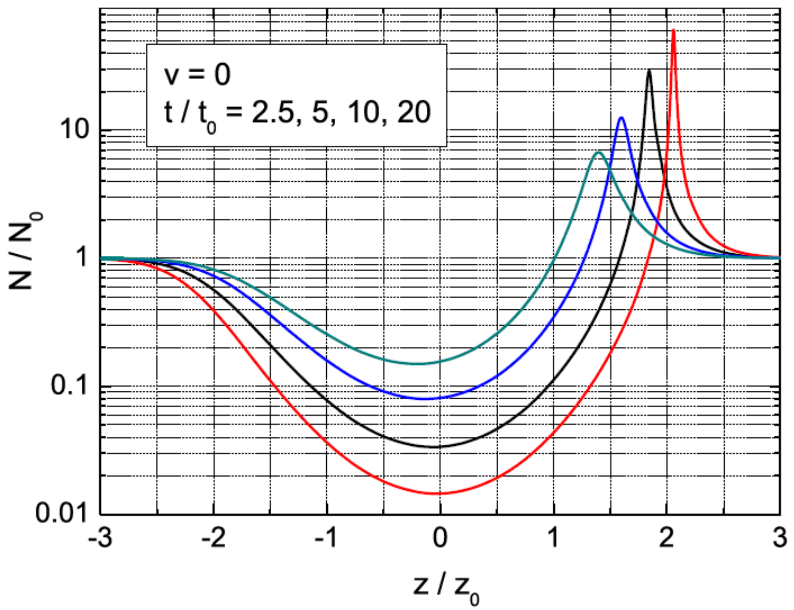
$$\frac{N_e(\hat{z},t)}{N_e^0} = \frac{u(\zeta(\hat{z},t))}{u(\hat{z})}$$

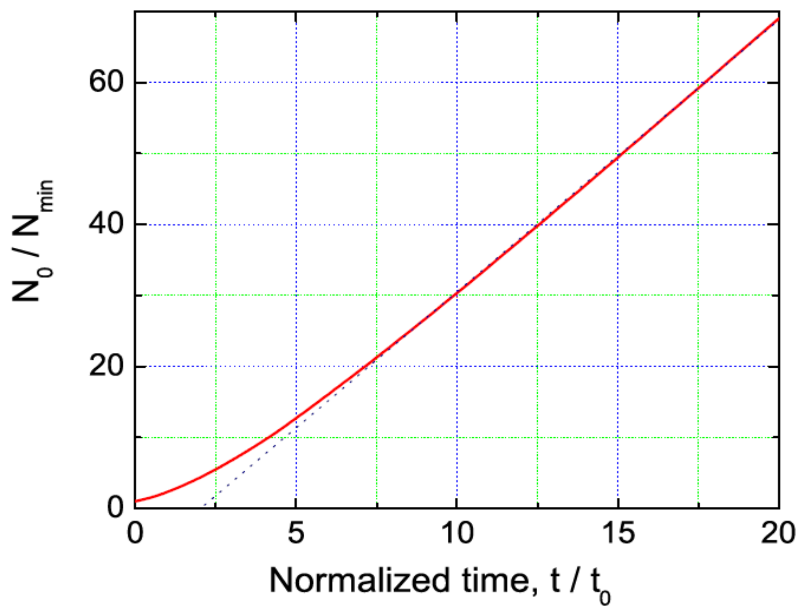
Characteristic drift time

$$t_0 = z_0 / \mathrm{v}_0 \propto z_0 / I_0$$

$t_0 \sim$ 1 hour, $\mathrm{v}_0 \sim$ mm/h

Results: Static light beam, v = 0

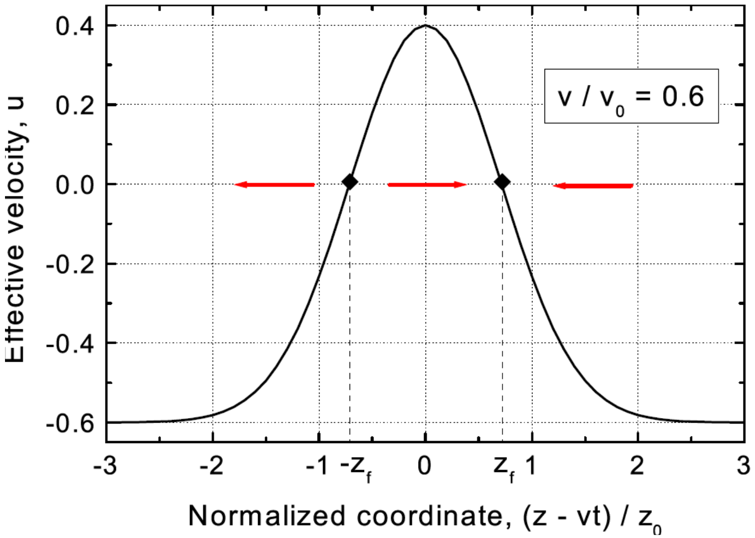




Works, but takes a long time

Results: Moving light beams

A different situation

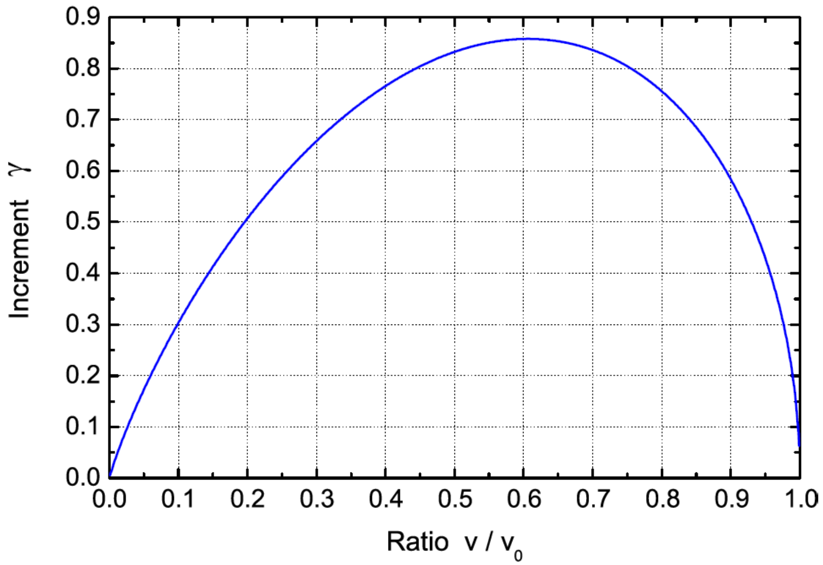


Two zero (fixed) points ($\pm z_1$) of the effective velocity profile *u*(*z*)

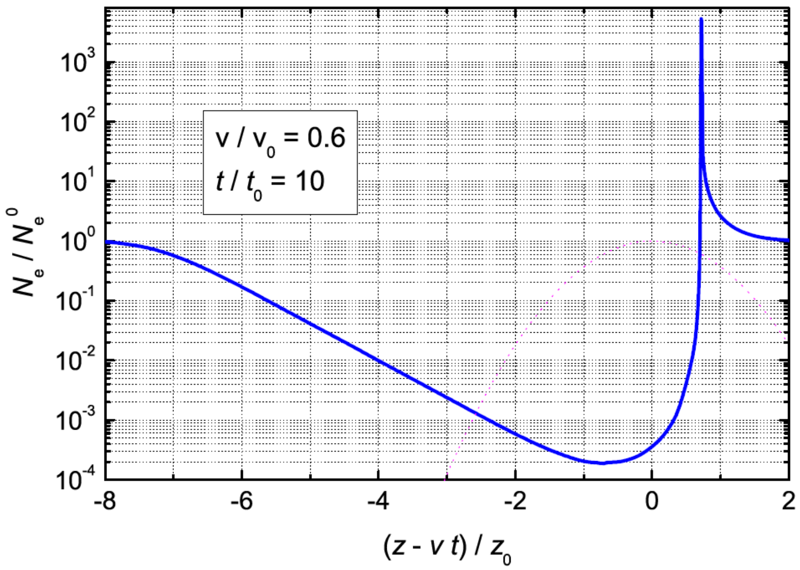
An exponential decrease/increase at the divergence/convergence points

$$N_e(\pm z_1, t) / N_e^0 = \exp(\pm \gamma\, t / t_0)$$

$$\gamma = 2\,(\mathrm{v}/\mathrm{v}_0)\,\sqrt{\ln(\mathrm{v}_0/\mathrm{v})}$$



Concentration profiles for moving beams

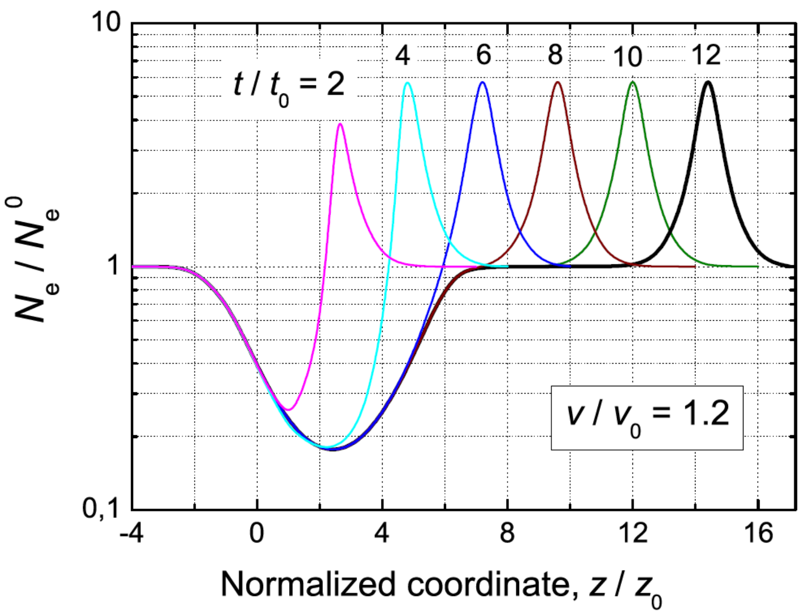


An exponential cleaning,

No profile breacking

A v > v_0 scenario

Abandoning of the cleaned area



Beyond the zero-model: Impact of nonlinear phenomena

Two parameters to control the nonlinear behavior

$$a = \sigma_e^0 / \sigma_i^0 < 1$$

$$b = N_e^0 / N_i^0 < 1$$

Zero-diffusion case

$$\frac{\partial N_e}{\partial t} + u(\hat{z}, N_e)\frac{\partial N_e}{\partial \hat{z}} = f(\hat{z}, N_e)$$

A quasi-linear equation

A small parameter to simplify theory

$$t_0 / t_d \approx 10^{-3} << 1$$

$$t_d = \varepsilon / 4\pi\, \sigma_\Sigma^0$$

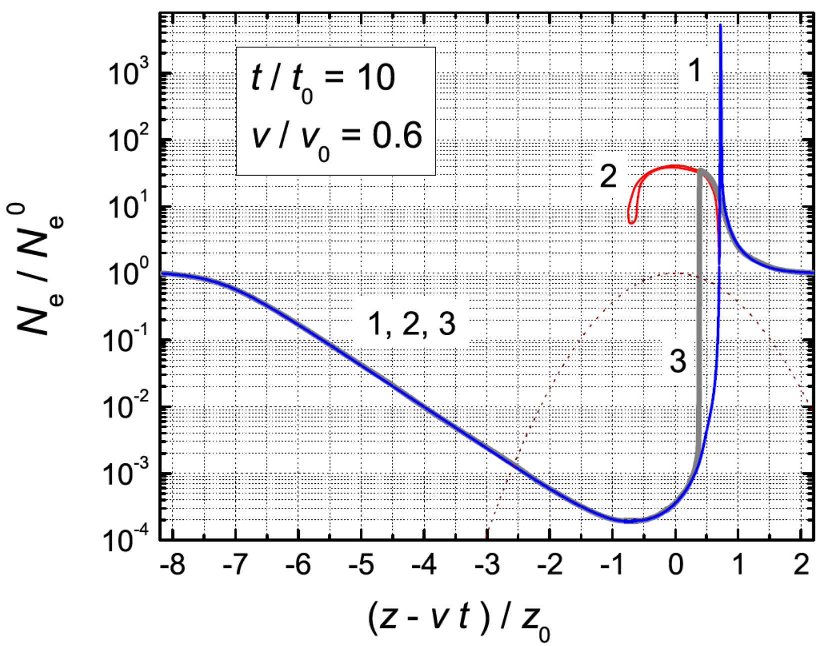
Local electric quasi-equilibrium

$$j_e + j_i \approx 0, \;\; E = E(N_e, N_i), \;\; N_i = N_i(N_e)$$

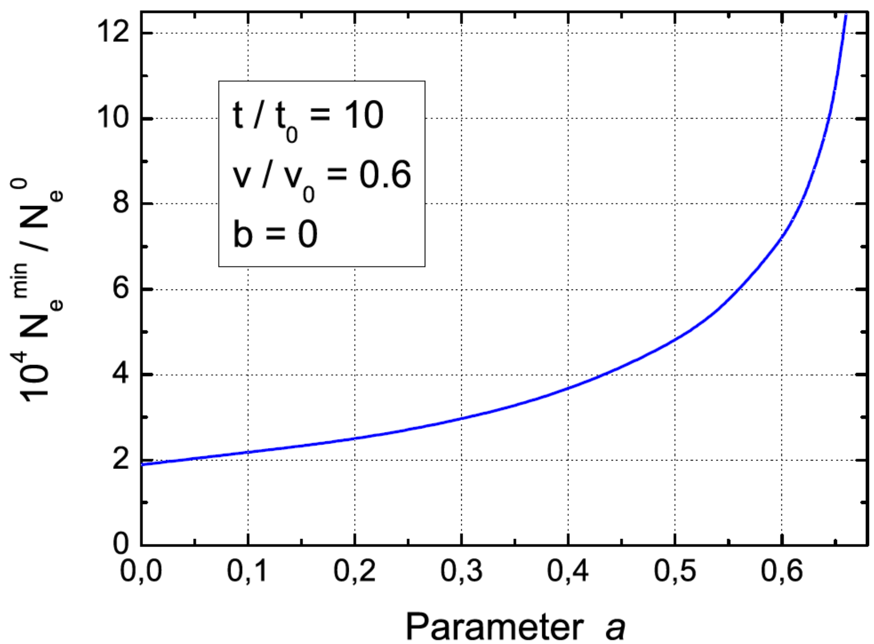
Can also be solved semi-analytically by the characteristic method

Nonlinear effects

$a = b = 0.01$



b = 0



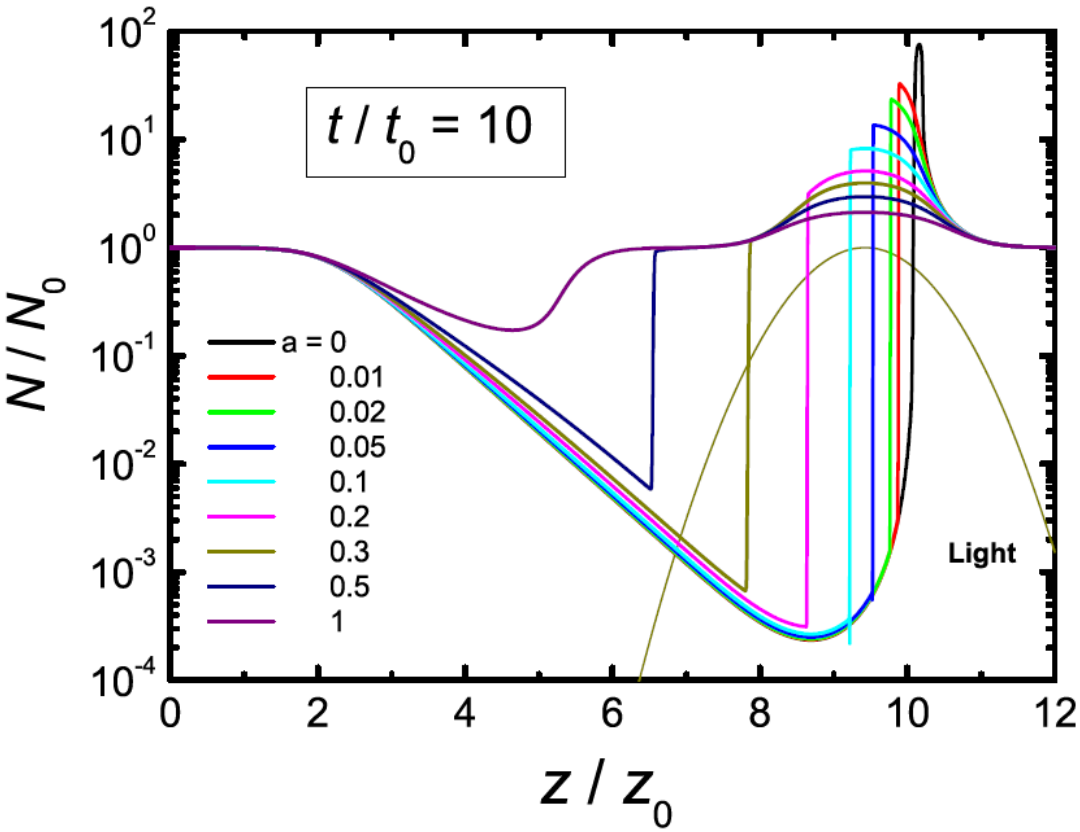
Breaking of the concentration profile

Stabilization by electron diffusion

Increase of N_{min}

Impairment of the cleaning performance

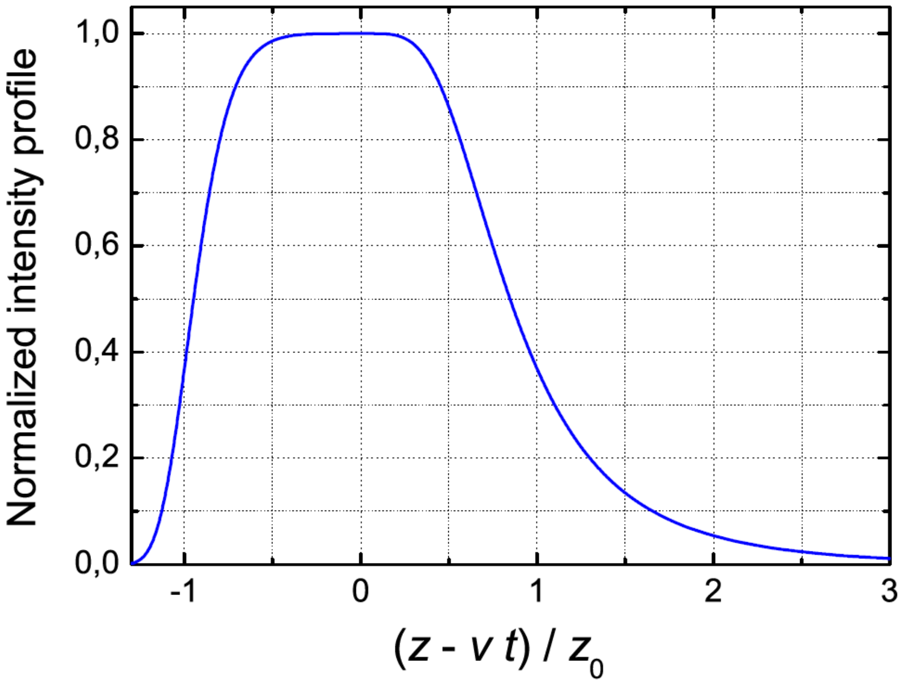
More nonlinear effects



b = 0

- **Shift of the discontinuity backwards**
- **Separation of the beam from the cleaned area**

More tricks



An asymmetric flat-top *I*-profile

This ensures:

- **Increasing of the exponential decay rate γ**
- **Separation of the zero (fixed) points**
- **Flattening of the concentration peak**
- **Suppression of the impact of nonlinear effects**

About experiment

At least four variable experimental parameters, I_0, z_0, v, $\sigma_i(T)$
Uncertainty of material parameters

At least two different cases:

Doped crystals ($N_e^0 > 10^{16}$ cm^{-3}). The profile $N_e(z)$ can be monitored
Via absorption scan. The model parameters are known (measurable).
Not useful to remove electrons up to ($10^{11} - 10^{12}$) cm^{-3}, but useful to
test the method

Undoped crystals (optically transparent). The initial concentration
$N_e^0 < 10^{15}$ cm^{-3} cannot be measured. Model parameters are uncertain.
Promising for applications

Long-time treatments, $t > 1$ day, $T = 180$ ℃, H^+ ions

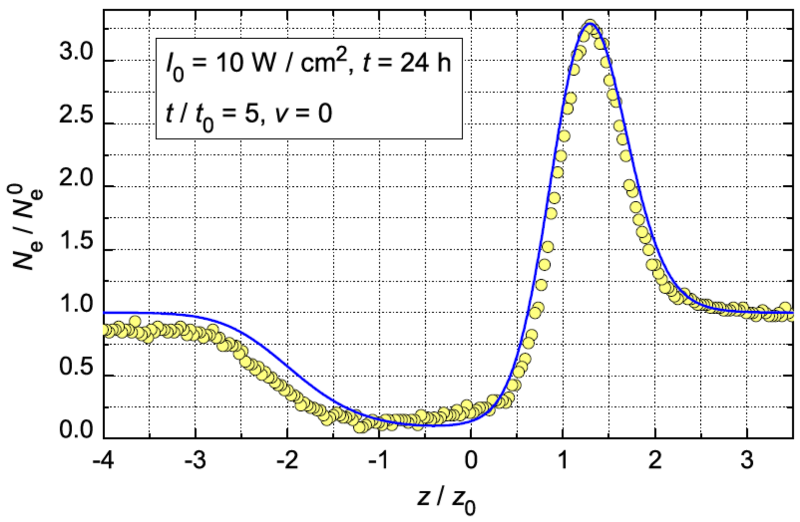
The cleaning performance is yet far from optimum

First experimental data

$N_e^0 \approx 6 \times 10^{16}$ cm^{-3}, $N_i^0 \approx 2 \times 10^{18}$ cm^{-3},

$z_0 \approx 70$ μm, $\lambda = 514$ nm, $I_0 = 10$ W/cm^2,

v = 0, $a \approx 0.5$, $b \ll 1$

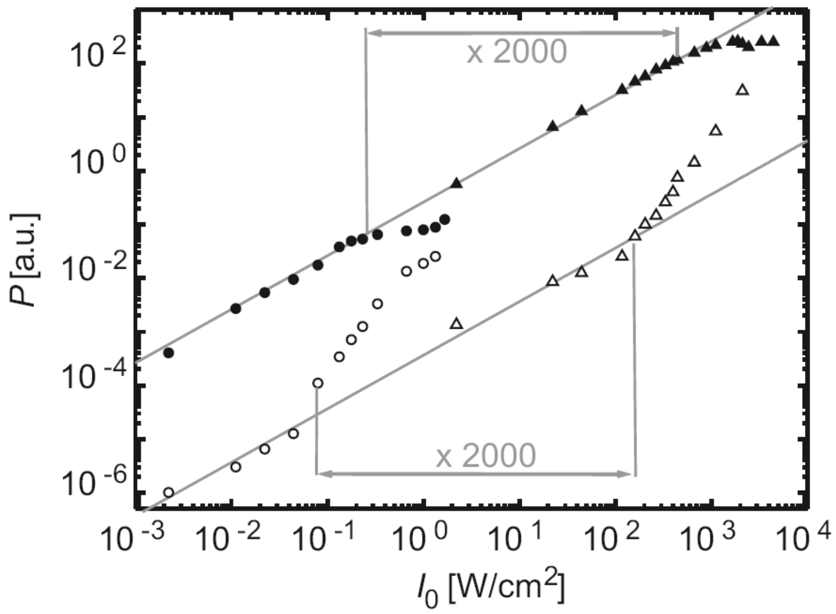


A 10 fold reduction of N_e

$z_0 \approx 42$ μm, $\lambda = 532$ nm, $I_0 = 15$ W/cm^2,

v = 3×10^{-3} mm/h, $a \approx 1$, $t = 340$ h,

v t = 1mm, $t / t_0 \approx 100$



$N_e(z)$ becomes non-measurably small

A 2000-fold increase of the threshold of optical damage

Thank you!