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, $T_c \approx 1200^\circ C$, $C_{3v}$ 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

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

\[ \alpha = N_e \sigma_e \]

\[ n_e = \frac{N_e \sigma_e I}{\hbar \omega} \tau_e \]

\[ \beta = e \frac{N_e \sigma_e}{\hbar \omega} l_{pv} \]

\[ D_e = \frac{\mu_e T}{e} \]

\[ E_{pv} = \frac{l_{pv}}{\mu_e \tau_e} \approx 10^5 \text{ V/cm} \]

\[ \Delta n \approx 10^{-3} \]

Light absorption coefficient

Free electron concentration

Photovoltaic coefficient

Einstein relation

Photovoltaic field

Index change
Why high temperature?

An ultimate goal of electron engineering:
To remove photo-excitable electrons far below the background level, $N_e << 10^{15}$ 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 $T > 200$ °C, 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$^{-3}$

$T = 600$ °C, $U_0 = 1$ kV

t ~ 1 hour, $w < 100$ μm

Nobody observed things like this earlier
### Model

**Assumptions and notation:**

\[
N^- = N_e, \quad N^+ = N_i \quad (N_{Li^+} \approx 10^{20} \text{ cm}^{-3})
\]

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

\[
z_0 = z_0(t), \quad v = v(t)
\]

\[
E_0 = U_0 / l, \quad \xi = z_0 / l
\]

### Main physical requirement:

\[
v = -\mu_- E_2
\]

### Main predictions:

\[
v = \frac{\mu_- E_0}{(1 + a \xi)}
\]

\[
\xi = \frac{-1 + \sqrt{1 + 2at / t_0}}{a}
\]

\[
a = \frac{\mu_- / \mu_+ + 1}{(N_0^+ / N_0^- - 1)}
\]

\[
t_0 = \frac{l}{\mu_- E_0}
\]

\[
t_\Sigma = t_0 \left(1 + a / 2\right)
\]

### More predictions:

- Time dependence of the potential \( U_{l/2}(t) \)
Comparison with experiment

1: $T = 650 \, ^\circ C$, $a = 7.4$, $t_0 = 15 \, \text{min}$
2: $T = 600 \, ^\circ C$, $a = 9.2$, $t_0 = 35 \, \text{min}$
3: $T = 550 \, ^\circ C$, $a = 9.8$, $t_0 = 94 \, \text{min}$

No doubts remain
The photovoltaic drift velocity

Initial set of equations

\[ \frac{\partial N_e}{\partial t} + \frac{\partial J_e}{\partial z} = 0, \quad \frac{\partial N_i}{\partial t} + \frac{\partial J_i}{\partial z} = 0 \]

\[ J_e = \beta I N_e - \mu_e n_e E - D_e \frac{\partial n_e}{\partial z} \]

\[ J_i = \mu_i N_i E - D_i \frac{\partial N_i}{\partial z} \]

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

\[ n_e = \frac{sN_e I \tau_e}{\hbar \omega} \]
Zero-model of optical cleaning

Assume:

\[ \sigma_e^0 \ll \sigma_i^0, \quad N_e^0 \ll N_i^0, \quad E \ll E_{pv} \]

A moving Gaussian light pattern
A moving coordinate frame

\[ I = I_0 e^{-\frac{(z-vt)^2}{z_0^2}} = I_0 e^{-\frac{\hat{z}^2}{z_0^2}} \]

\[ \beta I = v_0 e^{-\frac{\hat{z}^2}{z_0^2}}, \quad v_0 = \beta I_0 \]

Single equation for \( N_e \)

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

\[ t_0 = \frac{z_0}{v_0} \propto \frac{z_0}{I_0} \]

\[ t_0 \sim 1 \text{ hour}, \quad v_0 \sim \text{mm/h} \]
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

$$N_e(\pm z_1, t) / N_e^0 = \exp(\pm \gamma t / t_0)$$
$$\gamma = 2 \left( \frac{v}{v_0} \right) \sqrt{\ln\left(\frac{v_0}{v}\right)}$$
Concentration profiles for moving beams

An exponential cleaning,
No profile breaking

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

\[ a = \frac{\sigma_e^0}{\sigma_i^0} < 1 \]
\[ b = \frac{N_e^0}{N_i^0} < 1 \]

A small parameter to simplify theory

\[ t_0 / t_d \approx 10^{-3} \ll 1 \]
\[ t_d = \varepsilon / 4\pi \sigma_\Sigma^0 \]

Local electric quasi-equilibrium

\[ j_e + j_i \approx 0, \quad E = E(N_e, N_i), \quad N_i = N_i(N_e) \]

Zero-diffusion case

A quasi-linear equation

Can also be solved semi-analytically by the characteristic method
Nonlinear effects

\[ a = b = 0.01 \]

- Breaking of the concentration profile
- Stabilization by electron diffusion
- Increase of \( N_{\text{min}} \)
- Impairment of the cleaning performance
More nonlinear effects

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

$t / t_0 = 10$

$b = 0$
More tricks

An asymmetric flat-top profile

This ensures:
- Increasing of the exponential decay rate $\gamma$
- Separation of the zero (fixed) points
- Flattening of the concentration peak
- Suppression of the impact of nonlinear effects
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, $T = 180$ °C, H$^+$ ions

The cleaning performance is yet far from optimum
First experimental data

$N_e^0 \approx 6 \times 10^{16} \text{ cm}^{-3}$, $N_i^0 \approx 2 \times 10^{18} \text{ cm}^{-3}$,
$z_0 \approx 70 \mu\text{m}$, $\lambda = 514 \text{ nm}$, $I_0 = 10 \text{ W/cm}^2$,
$v = 0$, $a \approx 0.5$, $b << 1$

$N_e(z)$ becomes non-measurably small

A 10 fold reduction of $N_e$

$z_0 \approx 42 \mu\text{m}$, $\lambda = 532 \text{ nm}$, $I_0 = 15 \text{ W/cm}^2$,
$v = 3 \times 10^{-3} \text{ mm/h}$, $a \approx 1$, $t = 340 \text{ h}$,
$v t = 1 \text{ mm}$, $t / t_0 \approx 100$

A 2000-fold increase of the threshold of optical damage
Thank you!