Electric field induced second harmonic generation (EFISH)

PřF:FB501 Plasma Diagnostics and Simulations

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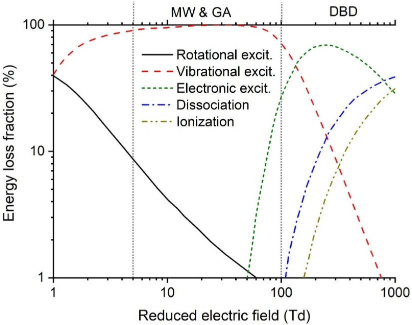
Motivation

Electric field determines, which plasmochemical processes take place in the plasma.
It determines how the coupled energy is distributed.

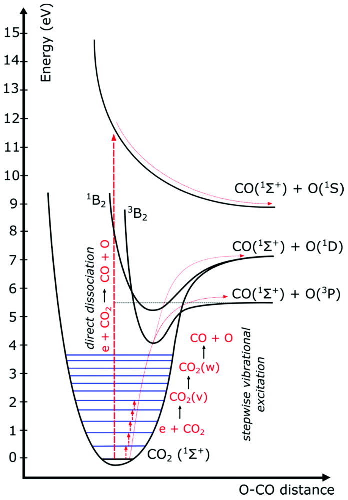


[A Bogaerts *et al*, ACS Energy Lett. 3 1013–27, 2018]: Fraction of electron energy transferred to different channels of excitation, as well as ionization and dissociation of N_2, as a function of the reduced electric field.

Motivation

Ex.: conversion $CO_2 \rightarrow C + O_2$: MW plasma has better energy efficiency than DBD

- MW: vibrational excitation + "leather climbing", dis. en. 5.5 eV
- DBD: electron excitation to repulsive state – 7-10 eV



[R Snoecks, A, Bogaerts, Chem. Soc. Rev., 2017,46, 5805-5863]

"Electric field" vs. "Reduced electric field"

- Electric field: E, $[\mathrm{V} \cdot \mathrm{m}^{-1}]$
- Reduced electric field: E/N, $[\mathrm{Td} = 10^{-21}\ \mathrm{V} \cdot \mathrm{m}^2]$
  - connected with the amount of energy gained by charged particle in the el. field between two collisions:

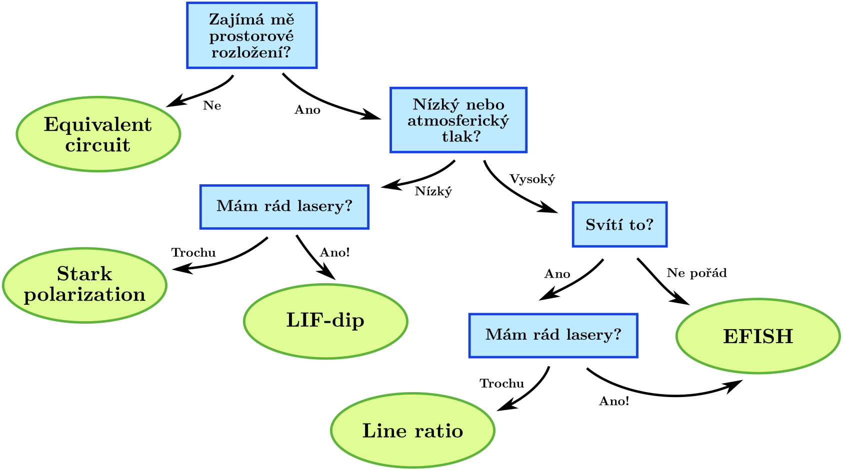
$$\varepsilon = q \cdot U = q \cdot \lambda \cdot E = \frac{q}{\sigma} \cdot \frac{E}{N}$$

  - λ [m] – mean free path between two collisions, $\lambda = \frac{1}{\sigma N}$
  - σ $[\mathrm{m}^2]$ – collisional cross section
  - N $[\mathrm{m}^{-3}]$ – concentration of particles

Diagnostic methods exhibition

- electrical probes:
    - equivalent circuit
    - capacitive probes
    - Pockels-effect-sensitive crystals
- optical emission spectroscopy:
    - Townsend coefficient
    - line intensity ratio (FNS/SPS)
    - Stark broadening, Stark polarization emission spectroscopy
    - bremsstrahlung (Z. Navrátil)
- laser-based methods:
    - CARS (Coherent anti-Stokes Raman scattering)
    - laser-induced fluorescence dip spectroscopy
    - EFISH

Diagnostic methods exhibition



Zajímá mě prostorové rozložení?

Ne

Ano

Equivalent circuit

Nízký nebo atmosferický tlak?

Nízký

Vysoký

Mám rád lasery?

Trochu

Ano!

Stark polarization

LIF-dip

Svítí to?

Ano

Ne pořád

Mám rád lasery?

EFISH

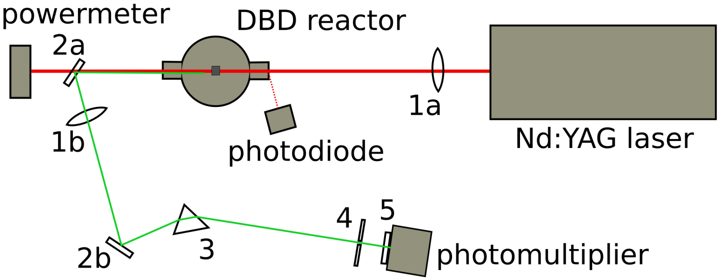
Trochu

Ano!

Line ratio

Electric field induced second harmonic generation (EFISH)

Electric field in a gas → polarisation of the gas → optical anisotrophy → second harmonics generation



Signal is week, coherent, line-integrated

A trip to the nonlinear optics

- nonlinear polarization:

$$P_i = \epsilon_0(\chi^{(1)}_{ij}E_j + \chi^{(2)}_{ijk}E_jE_k + \chi^{(3)}_{ijkl}E_jE_kE_l + \dots)$$

  - P_i – induced polarisation of the material
  - E_i – electric field intensity
  - $\chi^{(n)}$ – the *n*-th order nonlinear susceptibility tensor
- wave equation: $$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}$$
- second harmonics generation:

$$E_j = E_k = E_0\cos(\omega t)$$

$$P^{(2)}_i = \epsilon_0\chi^{(2)}_{ijk}E_jE_k = \epsilon_0\chi^{(2)}_{ijk}E_0^2\cos^2(\omega t) =$$

$$= \frac{1}{2}\epsilon_0\chi^{(2)}_{ijk}E_0^2 + \frac{1}{2}\epsilon_0\chi^{(2)}_{ijk}E_0^2\cos(2\omega t)$$

- $\chi^{(2)}_{ijk} = 0$ in a centrosymmetric media

A trip to the nonlinear optics - EFISH

EFISH:

$$E_j = E_k = E^{(\omega)}\cos(\omega t),\ \ E_l = E_{\mathrm{ext}}$$

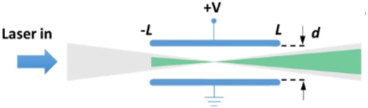
- induced polarization at the second harmonic frequency:

$$P_i^{(2\omega)} \sim \frac{3}{2}\chi^{(3)}_{ijkl}(-2\omega, 0, \omega, \omega)E_{\mathrm{ext}}(E^{(\omega)})^2$$

- signal intensity scales with the square of the induced polarization:

$$I^{(2\omega)} \sim \left[\chi^{(3)}E_{\mathrm{ext}}I^{(\omega)}\right]^2$$

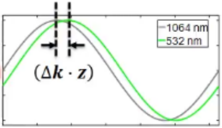
EFISH equations for homogeneous field



EFISH in homogeneous el. field $E = \frac{U}{d}$:

$$I^{(2\omega)} \propto \left[\alpha^{(3)} \cdot N \cdot I^{(\omega)} \cdot E_{\text{ext}} \cdot \frac{\sin(\Delta k L)}{\Delta k}\right]^2$$

- $I^{(2\omega)}$ – EFISH signal intensity
- $\alpha^{(3)}$ – third order nonlinear hyperpolarizability of the gas
- N – gas number density
- $I^{(\omega)}$ – intensity of the primary laser beam
- E_{ext} – external el. field (that's what we measure)
- $\Delta k = (2k_{\omega} - k_{2\omega})$ – wave vector mismatch
- L – interaction length

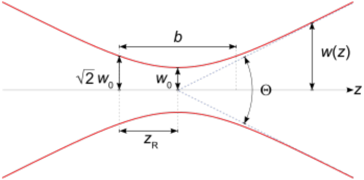


EFISH for inhomogeneous field

$$I^{(2\omega)} \propto \left[\alpha^{(3)} \cdot N \cdot I^{(\omega)}\right]^2 \cdot E_0^2 \cdot z_R \cdot \left| \int_{-\infty}^{\infty} E'_{\mathrm{ext}}(z') \cdot \frac{\exp(i \cdot \Delta k \cdot z_R \cdot z')}{[1 + i \cdot z']} \mathrm{d}z' \right|^2$$

$$E'_{\mathrm{ext}}(z') = \frac{E_{\mathrm{ext}}(z)}{E_0}$$

$$z' = \frac{z}{z_R}$$



And that's how we do it!

- If the gas composition, shape of the laser beam nor the spatial distribution of the external field $\frac{E_{\mathrm{ext}}(z)}{E_0}$ do not change during the whole measurement and calibration:

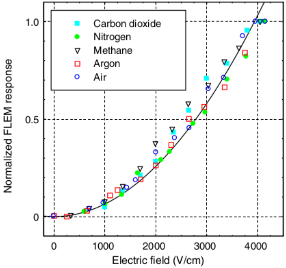
$$I^{(2\omega)} = A \cdot (I^{(\omega)})^2 \cdot E_0^2$$

- We find the calibration constant A by measurement of signal dependency on the known el. field (Laplacian field, sub-breakdown conditions)
- The desired el. field is determined from

$$E_0 = \frac{\sqrt{I^{(2\omega)}}}{\sqrt{A} \cdot I^{(\omega)}}$$

EFISH strengths

- high spatial and temporal resolution ($\sim$ laser pulse duration and shape)
- it works for most of gases
- nonresonant method, any laser wavelength
- sensitivity $\sim$ 1 kV/cm
- only one laser beam – simple alignment
- polarisation of the signal $\parallel E_{\text{ext}}$



[Dogariu et al, Phys. Rev. Applied 7, 024024 (2017)]

EFISH limitations

- line-integrated signal → uncertainty in signal origin; the signal can also originate surprisingly far from the focusal point
- unintuitive dependancy on L, difficult calibration if the shape of the field is unknown
- signal is dependent on the gas composition – forex. $\alpha^{(3)}_{\mathrm{vzduch}} \approx 20 \cdot \alpha^{(3)}_{\mathrm{He}}$
- for calibration, extrapolation of the field is often needed
- signal is dependent on frequency E_{ext}

EFISH dependancy on *L*

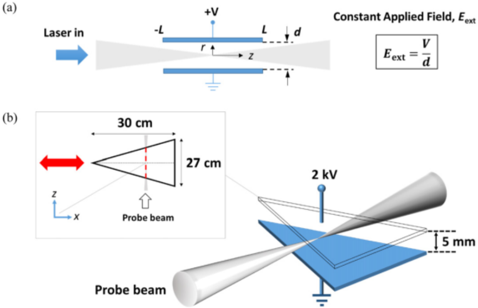


Figure 1. (a) Schematic of a canonical geometry typically used in E-FISH diagnosis. The top electrode is held at a potential of *V* with respect to the grounded bottom electrode. (While this illustration presents a focused probe beam, it should be understood that in the case of a plane-wave, the intensity of the probe beam is constant with respect to space and time.) (b) Corresponding schematic of the triangular-shaped, parallel-plate electrodes used for the experiments. By translating the electrodes (red arrow in figure inset), the effective electrode length (vertical red dashed line) seen by the E-FISH probe beam is varied.

[T L Chng et al Plasma Sources Sci. Technol. 29 (2020) 125002]

EFIAH dependancy on *L*

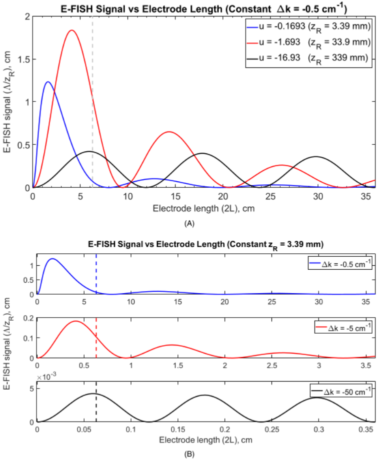


Figure 3. E-FISH signal in the modified plane-wave approximation. (a) Effect of electrode length, $2L$ on Λ (based on equation (4)) for three different z_R and $\Delta k = -0.5\ \mathrm{cm}^{-1}$ (note the logarithmic scale on the vertical axis). (b) Spatial (z) evolution of the E-FISH signal (given by equation (5)) in a constant external field, for $L = 6.4$ cm, and $z_R = 3.39$ mm. Gray and blue vertical dashed lines correspond to $z = \pm z_R$ and $\pm 5 z_R$ respectively.

[T L Chng et al Plasma Sources Sci. Technol. 29 (2020) 125002]

EFISH successfull results

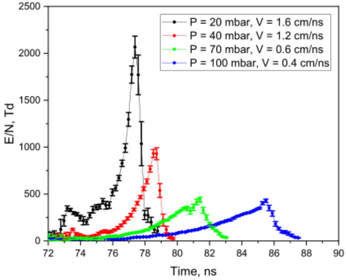


Figure 7. Reduced electric field measured in the front of a fast ionization wave discharge in nitrogen at $P = 20–100$ mbar, plotted on the same scale.

[T L Chng et al 2019 Plasma Sources Sci. Technol. 28 045004]

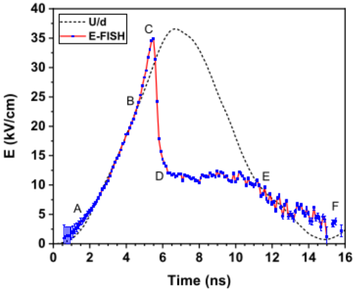
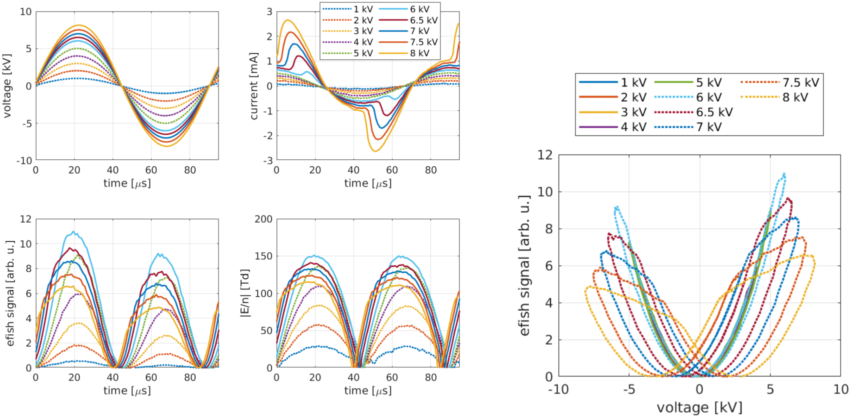


Figure 4. Longitudinal electric field temporal profile at 500 μm from the cathode obtained using the E-FISH technique (line+symbol line) compared with the electric field profile calculated as a voltage over gap length ratio (dashed line). He:N_2 = 5:1 mixture at 900 mbar, negative polarity discharge. The letters denote the boundaries of the regions with different field behavior; see text.

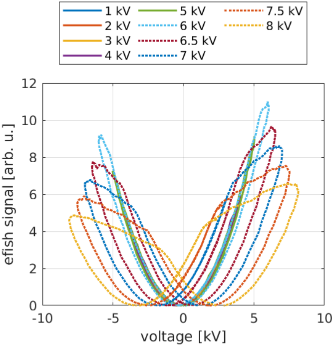
[N D Lepikhin et al 2021 J. Phys. D: Appl. Phys. 54 055201]

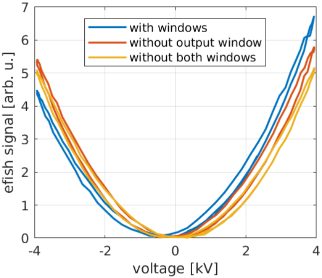
EFISH successfull results



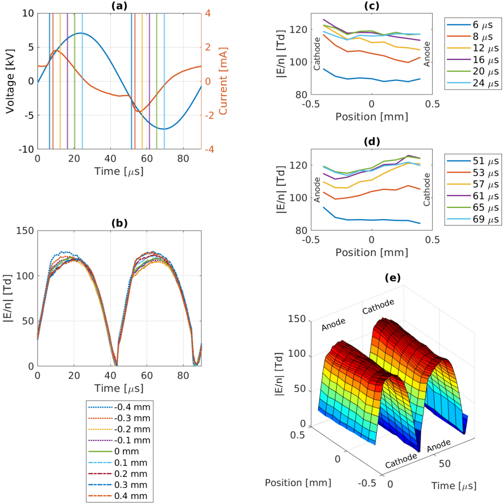
[M Mrkvičková et al 2023 Plasma Sources Sci. Technol. 32 065009]

EFISH successfull results

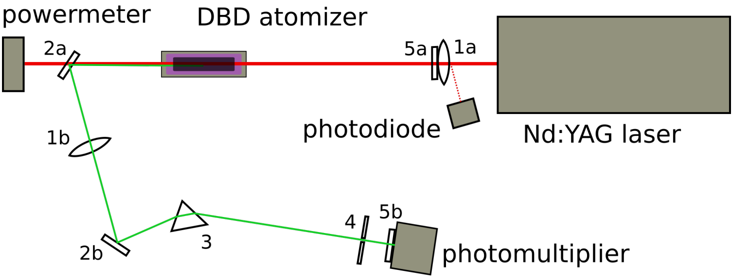




EFISH successfull results



Experimental part



Tasks:

- align the optical setup
- find the EFISH signal
- measure the dependency on the voltage amplitude or the discharge phase