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# Real-Time ellipsometric spectra without systematics errors

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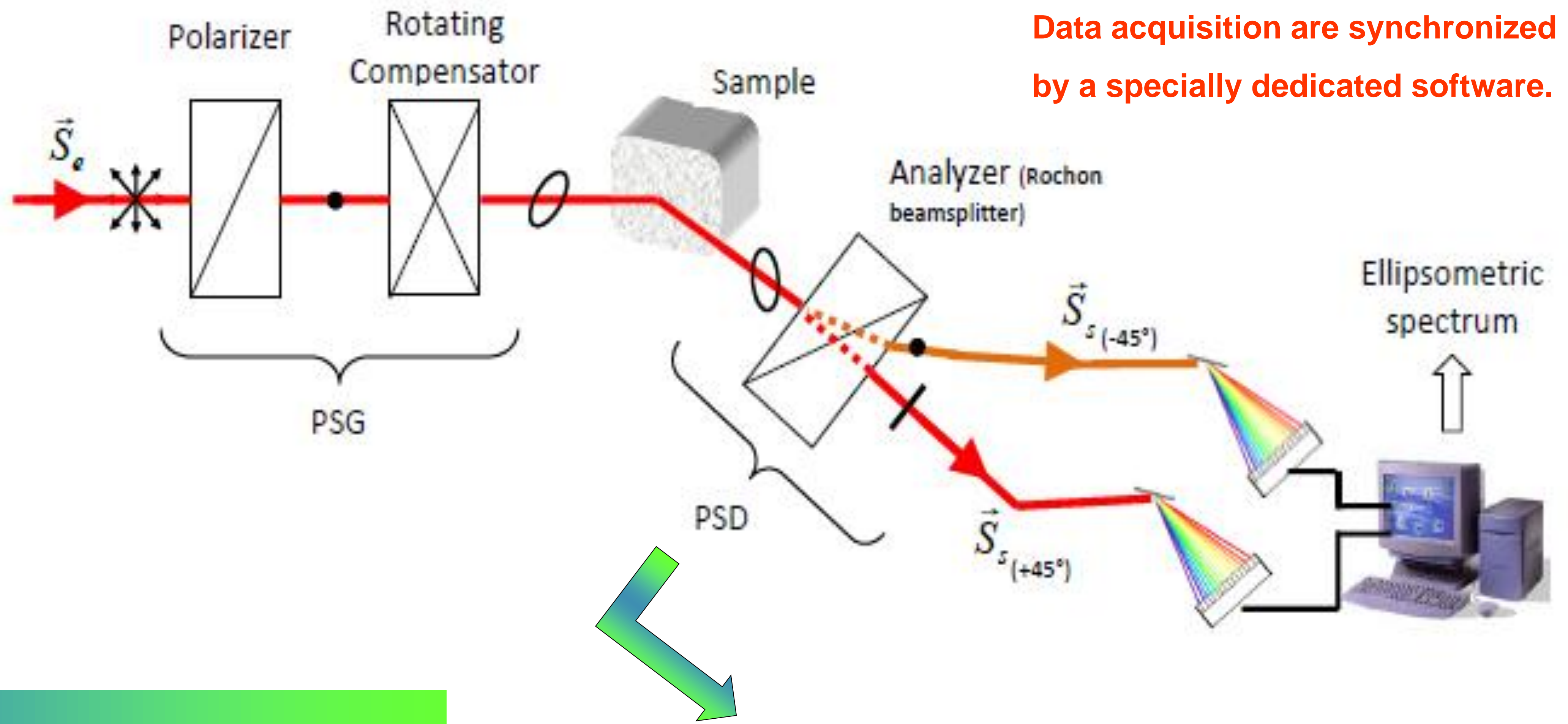
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In real-time measurement, it is difficult to reduce systematic errors on spectra of ellipsometric angles  $\Psi$  and  $\Delta$  by the mean of two measurements at two analyzer positions with the incidence plane  $+45^\circ$  and  $-45^\circ$  [1-2]. Generally, the analyzer is mounted on a motorized rotation stage. The rotation of the optical component from  $-45^\circ$  to  $+45^\circ$  requires few seconds, and this is incompatible with dynamic measurements.

## Real-time Spectroscopic Rotating Compensator Ellipsometer (RTSRCE)

- Based on a rotating compensator technology [3] using two detectors in the visible domain (400-800nm).
- The analyzer is replaced by a fixed Rochon birefringent beamsplitter which splits the incidence light wave into two perpendicularly polarized waves, one oriented at  $+45^\circ$  and the other one at  $-45^\circ$  according to the plane of incidence.
- The two emergent optical signals are analyzed by two identical CCD detectors.
- The final spectrum is the result of the two averaged  $\Psi$  and  $\Delta$  spectra acquired by both detectors.
- $\Psi$  and  $\Delta$  spectra are acquired without systematic errors on a spectral range fixed from 400 to 800nm. The acquisition time can be adjusted down to 25ms



Data acquisition are synchronized by a specially dedicated software.

## The systematic errors

The detected light has the following form:

$$I = I_0 \left[ a_0 + \sum_n (a_{2n} \cos 2nC + b_{2n} \sin 2nC) \right]$$

where  $C = \omega t$  is the angle of the fast axis of the compensator at time  $t$ .

The ellipsometric parameters of the sample are deduced by:

$$\tan \Delta = \frac{\alpha_{2c} \sin 2P - \alpha_{2s} \cos 2P}{\alpha_{4c} \sin 2P - \alpha_{4s} \cos 2P} \left( \frac{1 - \cos \delta_c}{2 \sin \delta_c} \right)$$

$$\tan \Psi = \frac{K \cdot \text{sgn}A \cdot \sin 2A (1 \pm \sqrt{1 + 1/K^2})}{(1 + \cos 2A)}$$

where

$$\frac{1}{K} = \frac{\sqrt{(\alpha_{2c}^2 + \alpha_{2s}^2)(1 - \cos \delta_c)^2 / \sin^2 \delta_c + 4(\alpha_{4c} \cos 2P - \alpha_{4s} \sin 2P)^2}}{2(\alpha_{4c} \cos 2P + \alpha_{4s} \sin 2P)}$$

with  $\text{sgn}A = 1$  for  $A > 0$  and  $\text{sgn}A = -1$  for  $A < 0$ .

All errors independent of their origins, propagate through the Fourier coefficients  $a_0, a_{2n}$  and  $b_{2n}$ . The systematic errors due to the positioning of the optical elements can be reduced by a suitable calibration procedure, but other errors due to the imperfection of the components affect the measurements. They are evaluated by calculating the Jacobian matrix relating  $\delta \Delta$  and  $\delta \Psi$  to the errors  $\delta \alpha_0, \delta \alpha_{2n}$  and  $\delta b_{2n}$ .

For example, the contribution to  $\Delta$  resulting from  $\delta \alpha_2$  is derived from equation of  $\tan \Delta$  by first differentiation with respect to  $\alpha_2$ :

$$\delta \Delta_{\alpha_2} = \frac{1 - \cos \delta_c \cos^2 \Delta \sin 2P}{2 \sin \delta_c \alpha_4 \sin 2P - \beta_4 \cos 2P} \delta \alpha_2$$

Taking into account that  $\alpha_2$  and  $\delta \alpha_2$  are proportional to  $a_2$  and  $\delta a_2$  [i.e.  $\alpha_n = G a_n$  and  $\beta_n = G b_n$ ,  $G$  may be considered as the overall gain and is assumed to be real and frequency independent] we obtain:

$$\delta \Delta_{a_2} = -\text{sgn}A \frac{\sin 2P \cos \Delta}{\sin \delta_c \sin 2P} \delta a_2$$

$\delta a_2$  is evaluated using a Taylor expansion of  $S$ . At the first order, the Stokes vector of the detected light is:

$$S_f = S_f^0 + \sum_k \frac{\partial S_f}{\partial \lambda_k} \delta \lambda_k = S_f^0 + \sum_k (\delta S_f)_k$$

where  $S_f^0$  is the Stokes vector for the ellipsometer without any imperfect elements. For a given perturbation  $\lambda$ , the second element of the second term allows to obtain the corresponding Fourier coefficients  $\delta a_0, \delta a_{2n}$  and  $\delta b_{2n}$ . The perturbation of the Stokes vector caused by an error is given by replacing the ideal matrix by the corresponding error matrix  $\delta M$ .

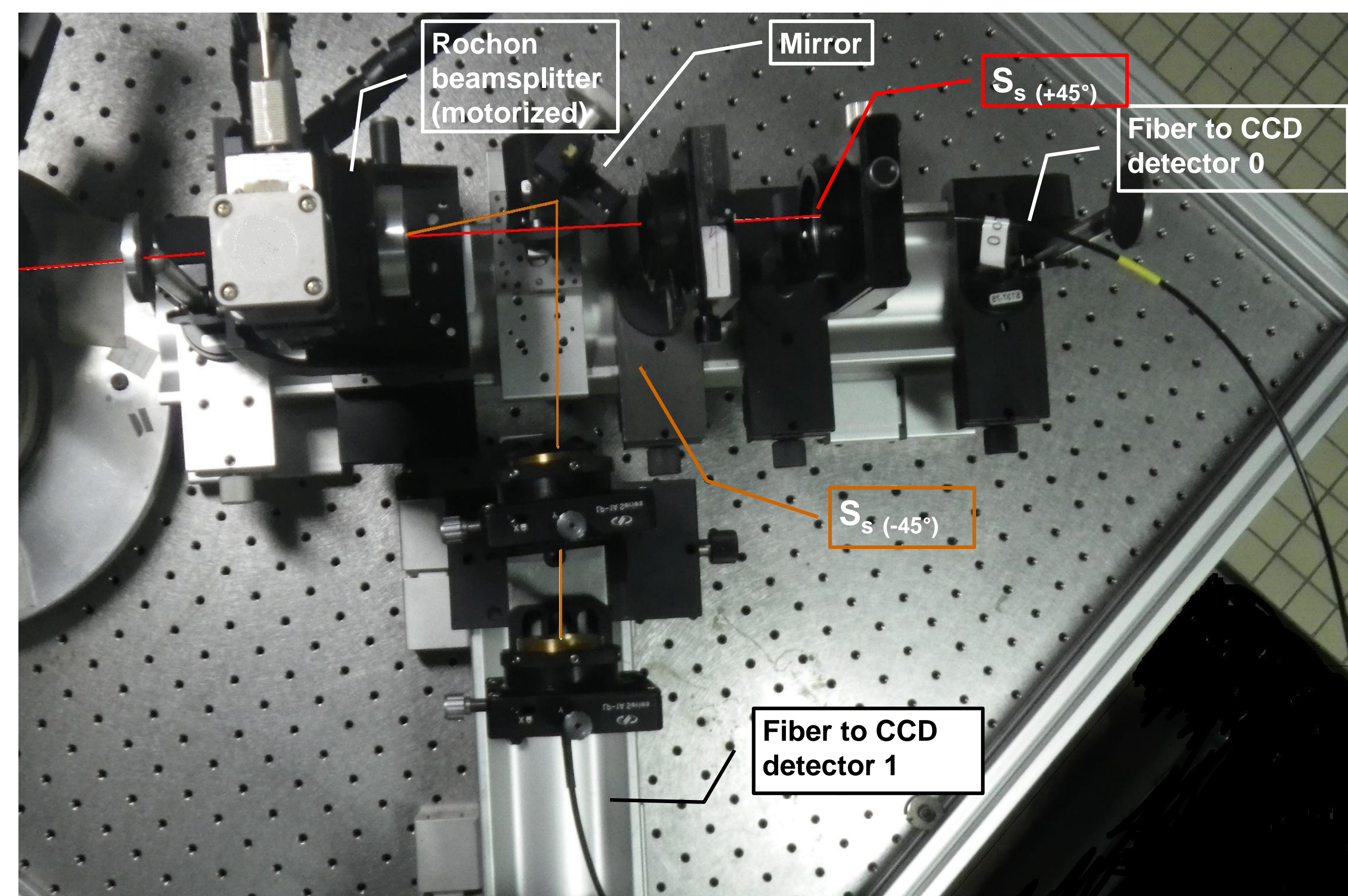
Azimuthal errors:  $\delta A, \delta P, \delta C$   
Element imperfections:  $\gamma A, \gamma P, \gamma C$

Systematic errors in RCE without Rochon beamsplitter with  $A = 45^\circ$  or  $A = -45^\circ$ .

Origin	$\delta \Psi$	$\delta \Delta$	Origin	$\delta \Psi$	$\delta \Delta$
$\delta A$	$- \text{sgn}A \sin 2\Psi$	0	$\gamma A$	0	$- \text{sgn}A \sin 2\Psi$
$\delta P$	$- \text{sgn}A \cos \Delta$	$2 \text{sgn}A \sin \Delta \cos 2\Psi$	$\gamma P$	$\text{sgn}A \cos 2P \sin \Delta \cos^2 2\Psi - \sin 2P \cos \Delta \cos 2\Psi \sin 2\Psi$	$2(-\sin 2P \cos^2 \Delta + \text{sgn}A \cos 2P \cos \Delta \cot \Psi)$
$\delta C$	$2 \text{sgn}A \cos \Delta$	$-2 \text{sgn}A \sin \Delta \cot 2\Psi$	$\gamma C$	0	0

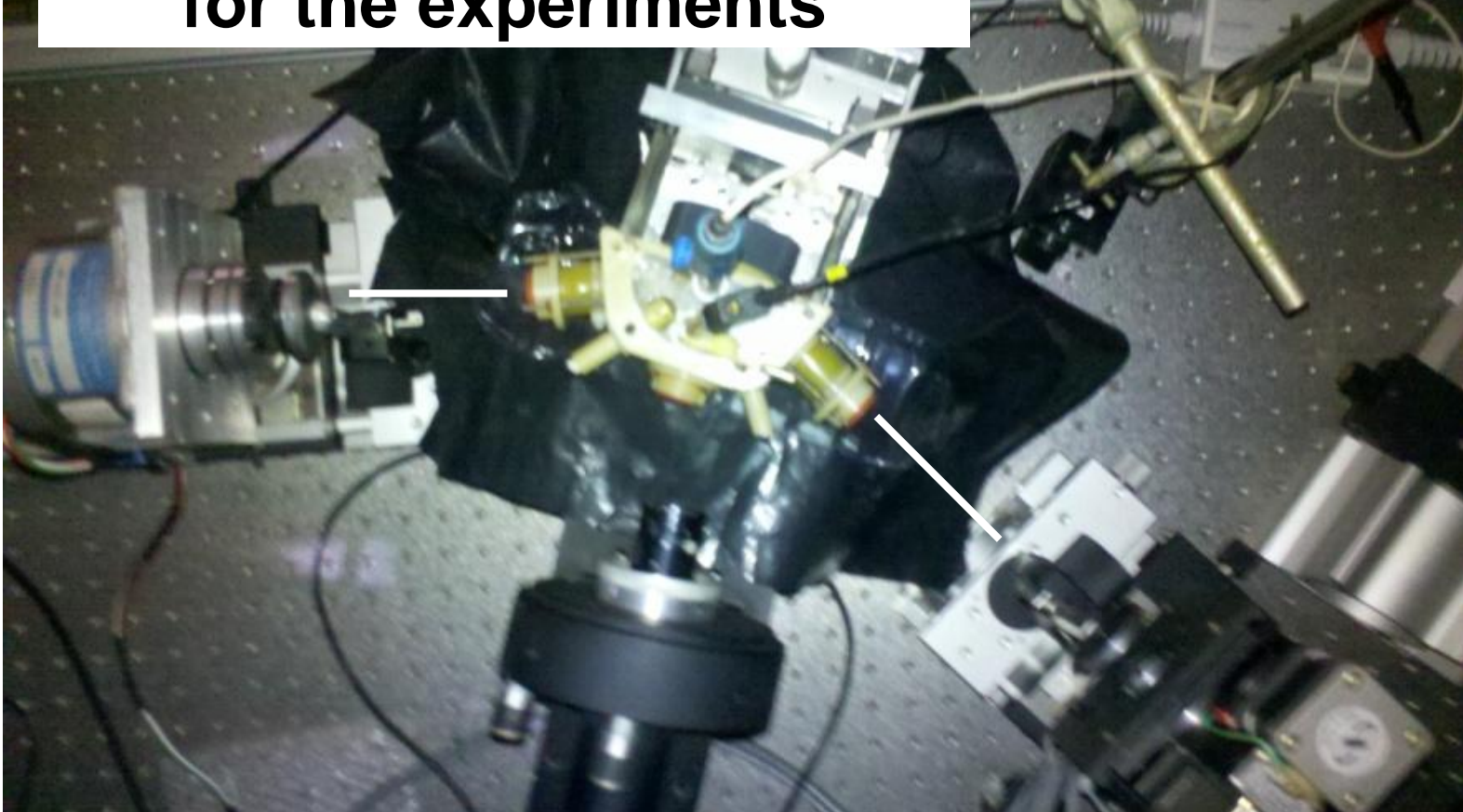
Systematic errors in RCE with Rochon beamsplitter at  $A = 45^\circ$ .

Origin	$\delta \Psi$	$\delta \Delta$	Origin	$\delta \Psi$	$\delta \Delta$
$\delta A$	0	0	$\gamma A$	0	0
$\delta P$	0	0	$\gamma P$	$-\sin 2P \cos \Delta \cos 2\Psi \sin 2\Psi$	$-2 \sin 2P \cos^2 \Delta$
$\delta C$	0	0	$\gamma C$	0	0



## In situ ellipsometric monitoring of electrocrystallization of Bismuth telluride films

Electrochemical cell used for the experiments

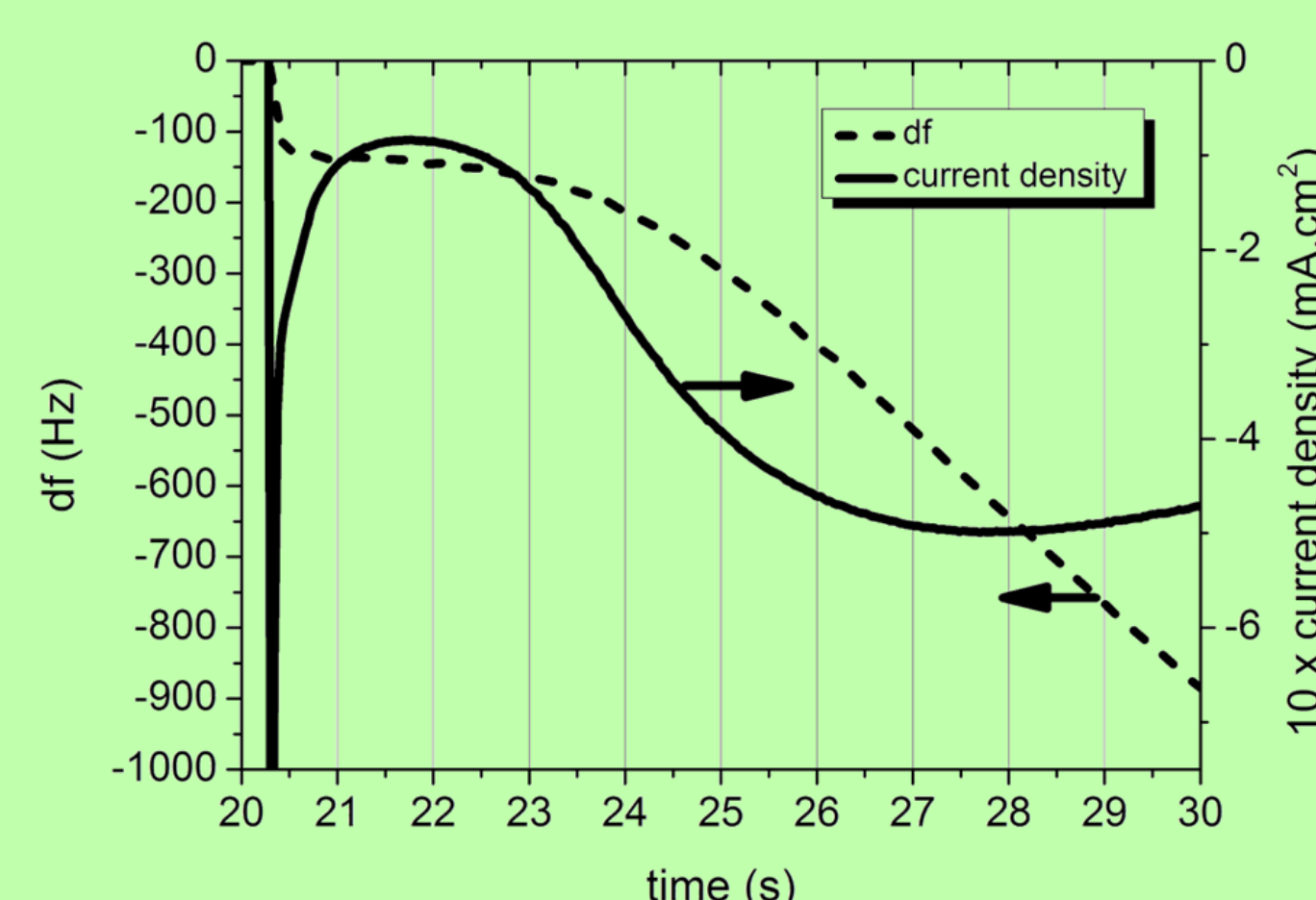
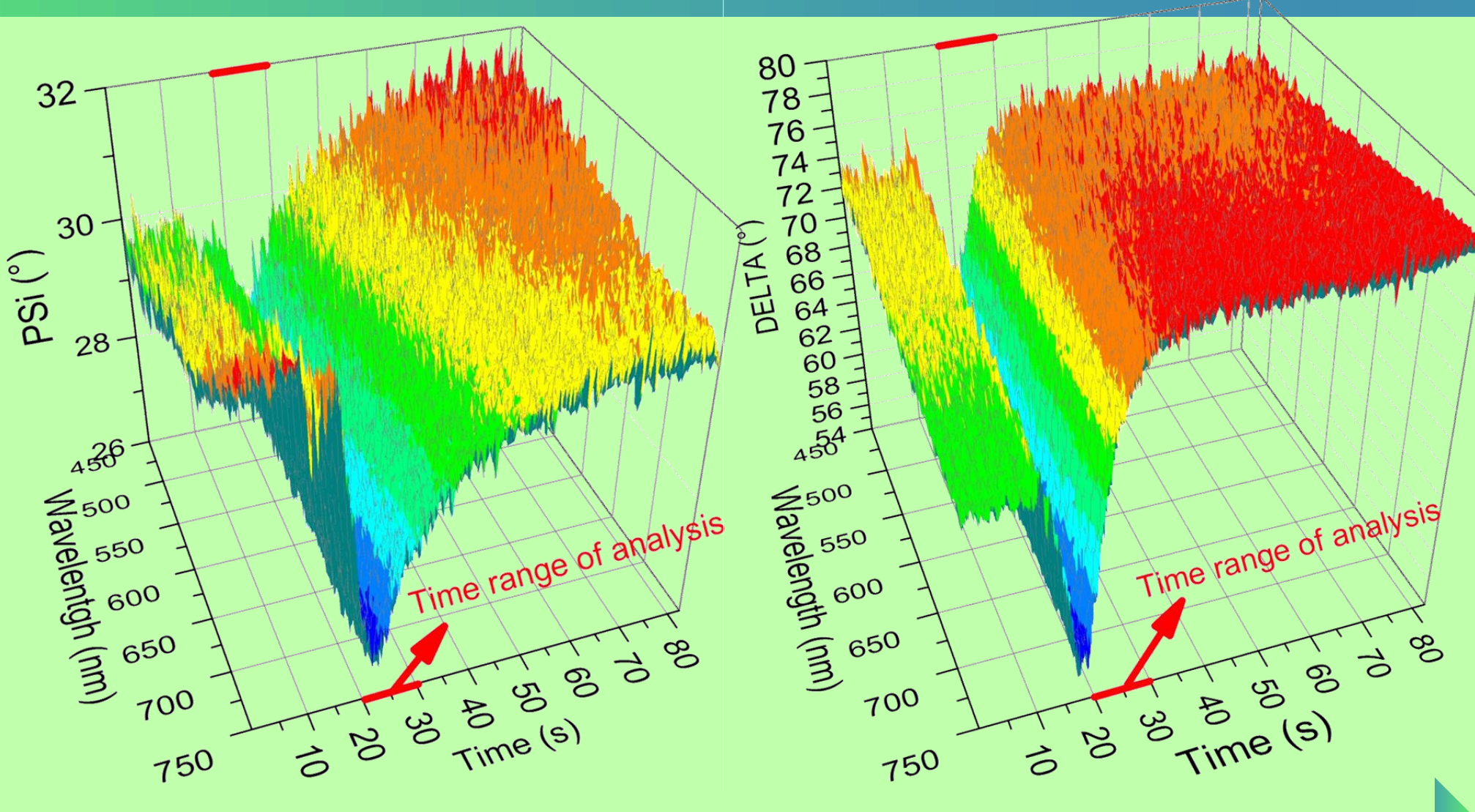


Simultaneous and synchronous records of :

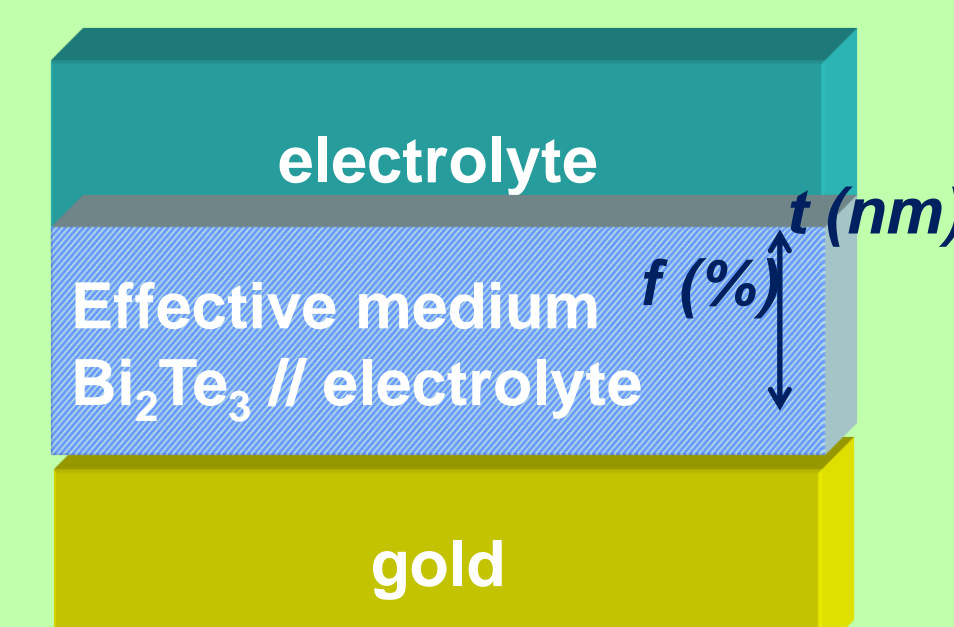
- Optical data:  $\Psi$  and  $\Delta$  spectra
- Gravimetric data: Electrochemical Quartz Crystal Microbalance
- Coulometric data: Potentiostat

Acquisition times :

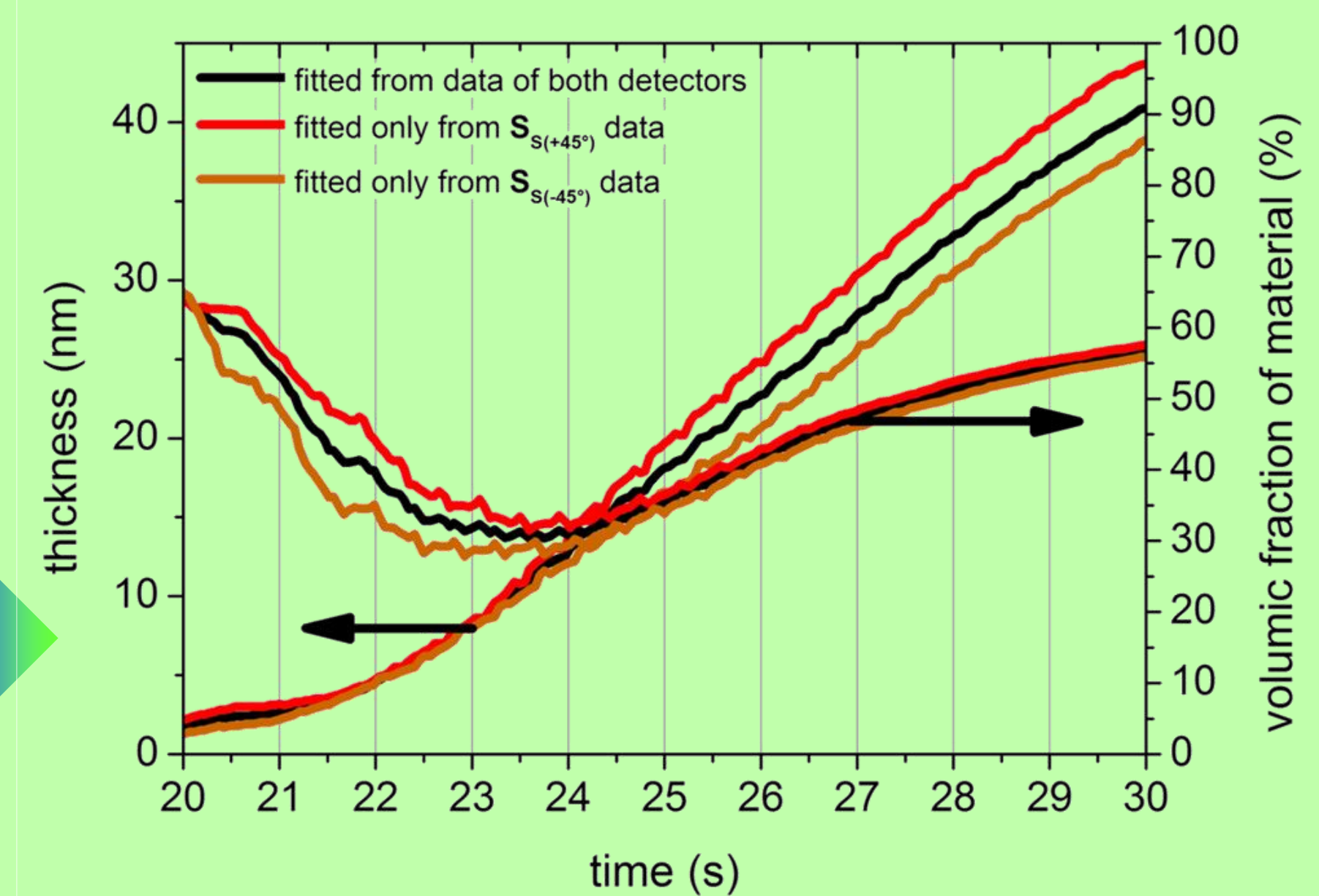
SE : 50 ms/EQCM : 160 ms/ potentiostat : 100 ms



## Optical model



Fitted parameters  
Material volumic fraction  $f$  (%)  
thickness  $t$  (nm)



	Time	Thickness (nm)	Volumic fraction (%)
Fitted only from $S_{s(+45^\circ)}$ data		4.9	43.4
Fitted only from $S_{s(-45^\circ)}$ data	22s	4.8	33.5
Fitted from data of both detectors		4.7	38.6
Fitted only from $S_{s(+45^\circ)}$ data		35.8	52.5
Fitted only from $S_{s(-45^\circ)}$ data	28s	30.6	50.3
Fitted from data of both detectors		32.9	50.5

- References
- [1] R. Kleim, L. Kuntzler and A El Ghemaz, J. Opt. S. Am. A 11:2550-2558 (1994).
  - [2] J. Lee, P. Rovira, I. An and R. Collins, Review of Scientific Instrument 72:1742-1754 (1998).
  - [3] L. Broch, L. Johann, N. Stein, A. Zimmer and R. Beck, Review of Scientific Instrument 78:064101 (2007).

