Self-consistent optical-constant of materials for EUV multilayer coatings

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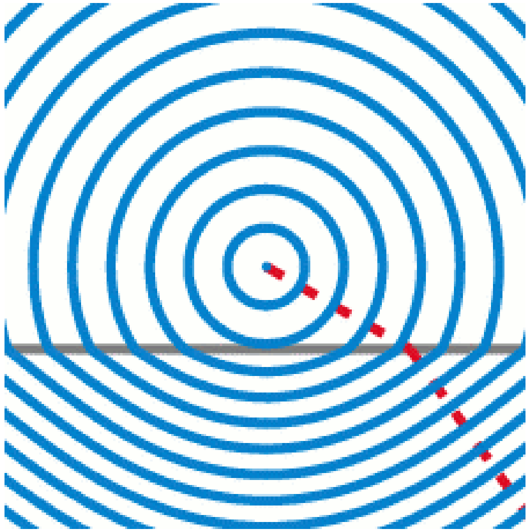
Refractive index= optical constants

* It describes how light propagates through an optical medium

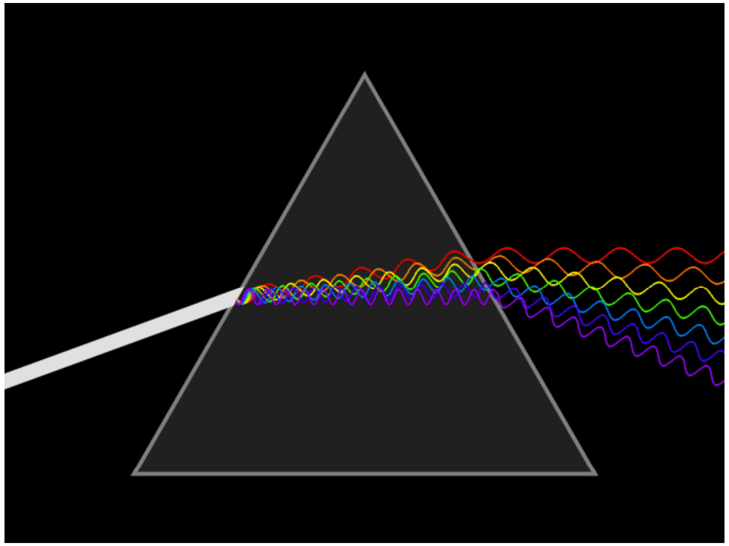


* It governs:

- refraction
- reflectance, transmittance, absorption
- optical path length
- group velocity

Refractive index=optical constants

- n: refraction - k: absorption
- N=n+ik: in the EUV, also k required
- dispersion: n(E), k(E)



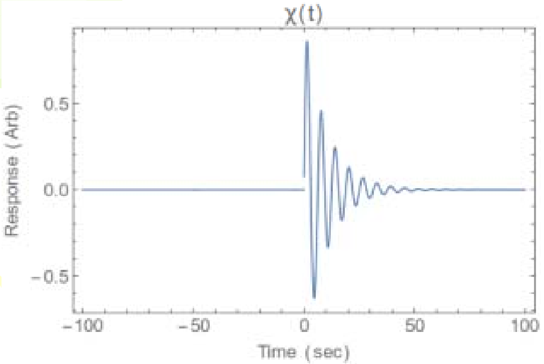
Self-consistent optical constants

Refraction (n) and absorption (k)
are not independent of each other

n(E), k(E) self-consistent:
when obtained with Kramers-Kronig analysis

$$n(E) - 1 = \frac{2}{\pi} P \int_0^\infty \frac{E' k(E')}{E'^2 - E^2} dE'$$

Causality and the Kramers-Kronig relations



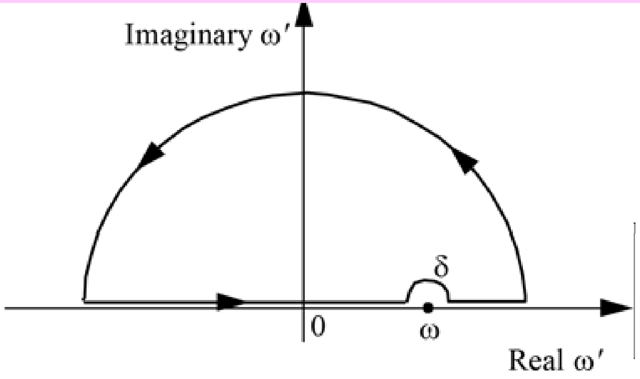
$$P(t) = \varepsilon_0 \int_{-\infty}^{+\infty} \chi(t-t')E(t')dt'$$

$$\chi(t<0)=0$$

Response can't precede the stimulus

$$\chi(\omega) = \int_{-\infty}^{+\infty} \chi(t)e^{-i\omega t}dt$$

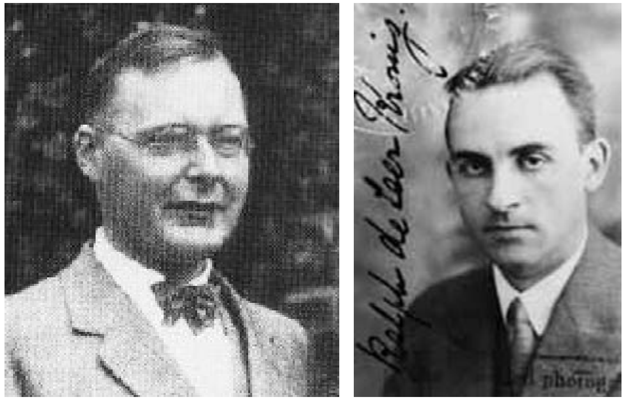
$\chi(\omega)=\varepsilon(\omega)-1$ has analytic continuation in C^+



$$\varepsilon_1(\omega)-1=\frac{2}{\pi}P\int_0^{\infty}\frac{\omega'\varepsilon_2(\omega')}{\omega'^2-\omega^2}d\omega'$$

$$\varepsilon_2(\omega)=-\frac{2\omega}{\pi}P\int_0^{\infty}\frac{\varepsilon_1(\omega')-1}{\omega'^2-\omega^2}d\omega'$$

Optical Constant calculation: Kramers-Kronig analysis



Provides (n,k) with a single measurement **in the whole spectrum**

*Two main options for KK analysis:

n, k

$$n(E)-1=\frac{2}{\pi}P\int_0^{\infty}\frac{E'k(E')}{E'^2-E^2}dE'$$

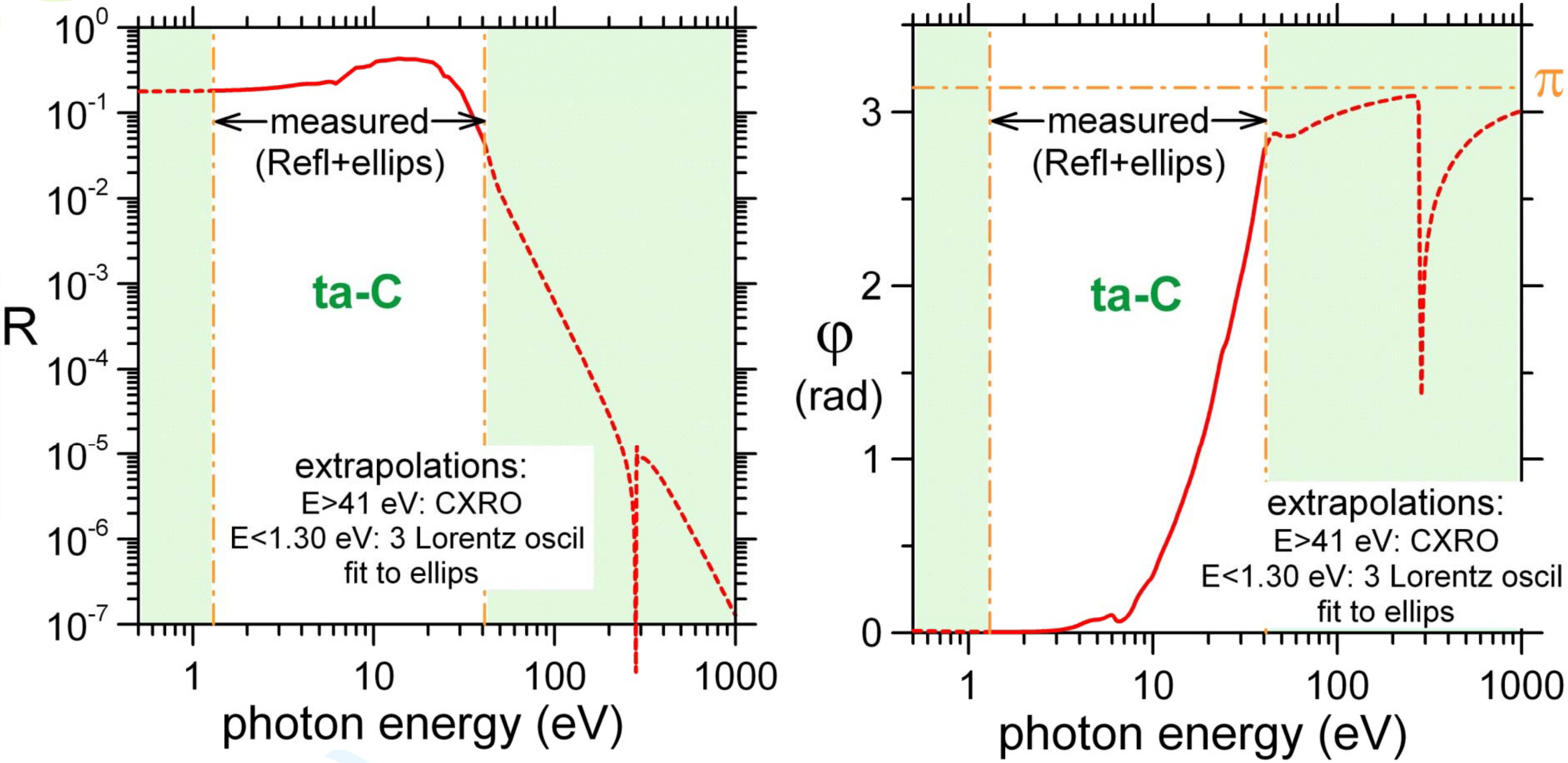
R, φ

$$r = R^{1/2}e^{i\varphi}$$

$$lnr = \frac{1}{2}lnR + i\varphi$$

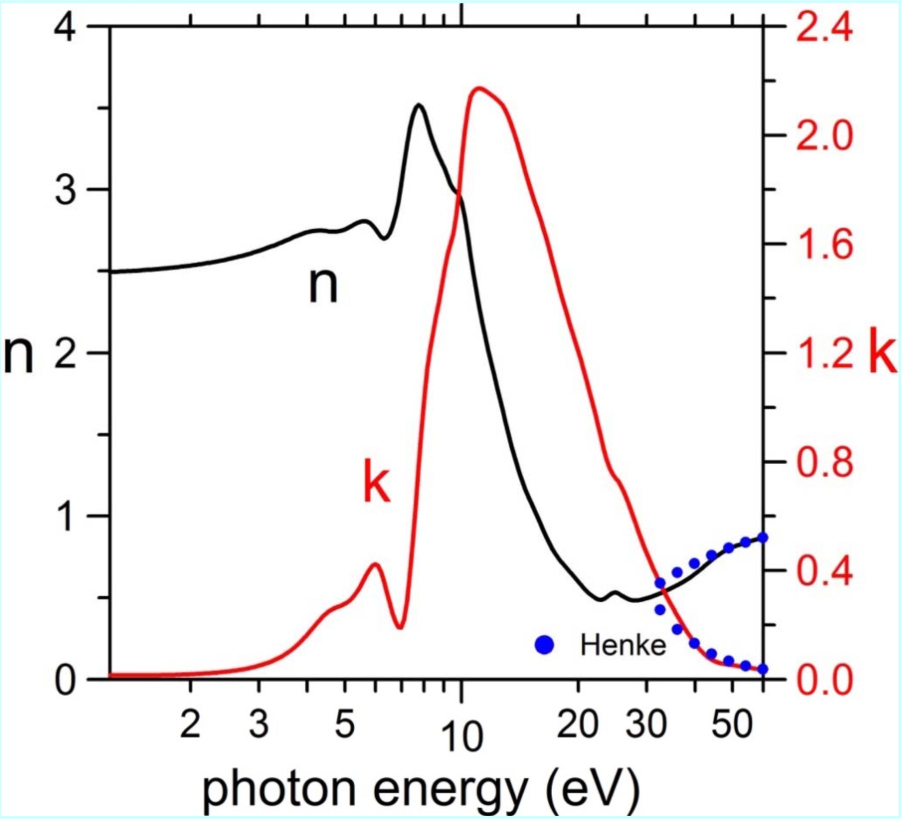
$$\varphi(E)=-\frac{E}{\pi}P\int_0^{\infty}\frac{\ln[R(E')]}{E'^2-E^2}dE'$$

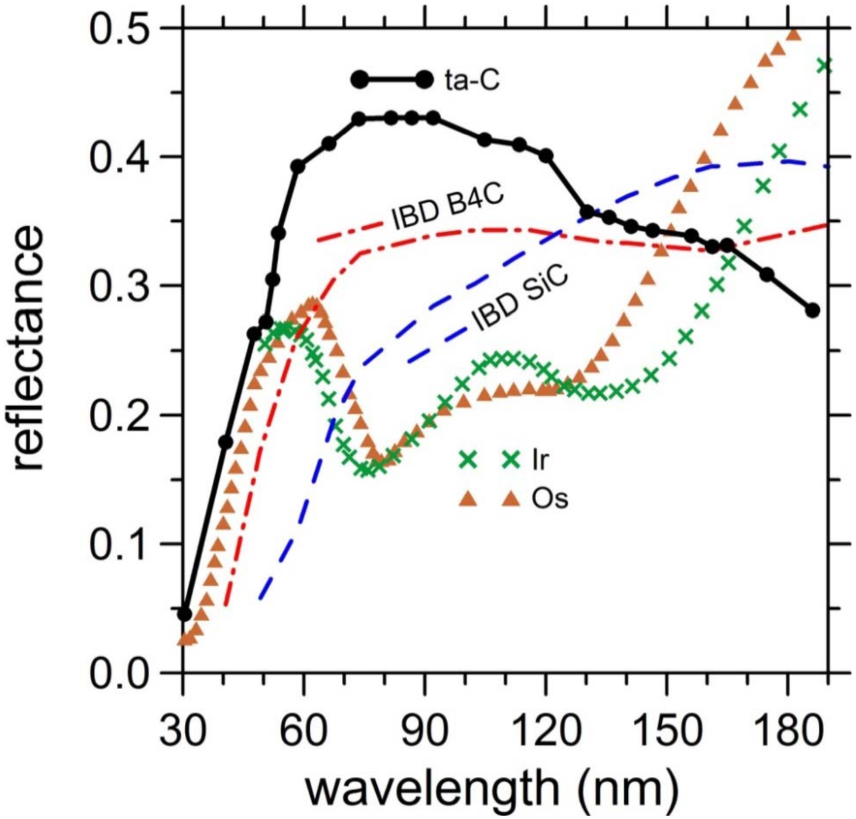
KK by reflectance: ta-C tetrahedral amorphous Carbon



Larruquert et al., Opt. Exp. **21**, 27537 (2013)

KK by reflectance: ta-C tetrahedral amorphous Carbon





From R, φ: n, k:

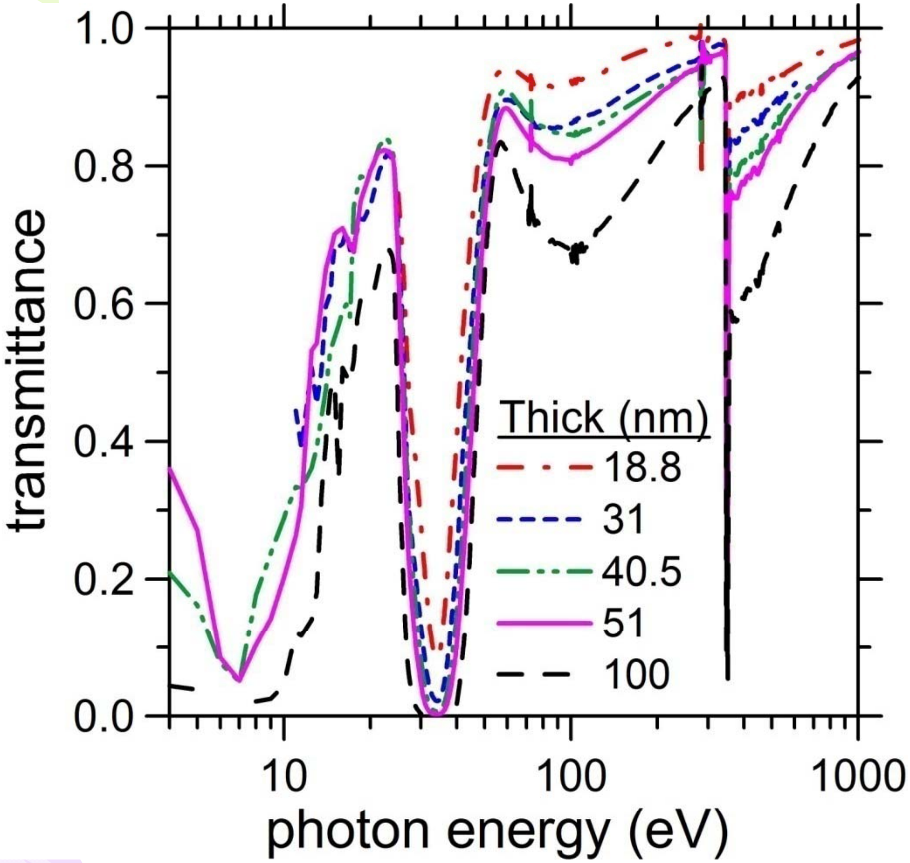
$$\boxed{R^{1/2}e^{i\varphi} = \frac{n-1+ik}{n+1+ik}}$$

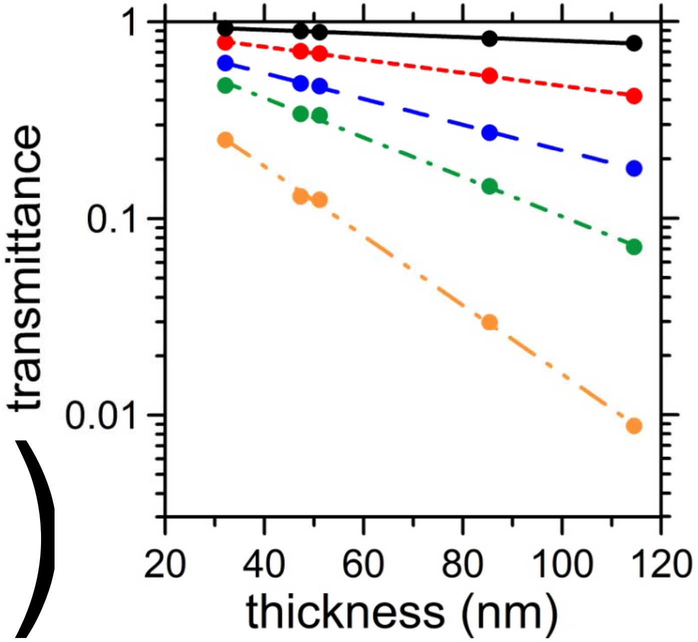
ta-C: coating with largest Refl in ~30-130 nm (9.5-41 eV)

Difficulty: film must be opaque

KK: n from k. **Ca**

substrate: C film supported on grid



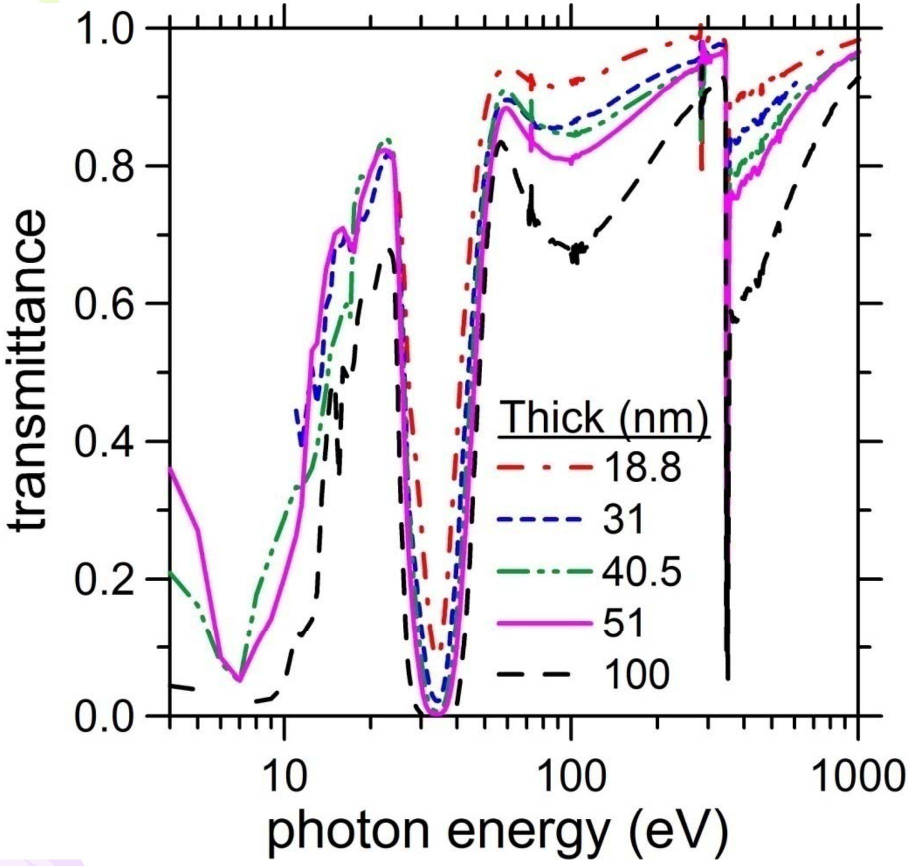


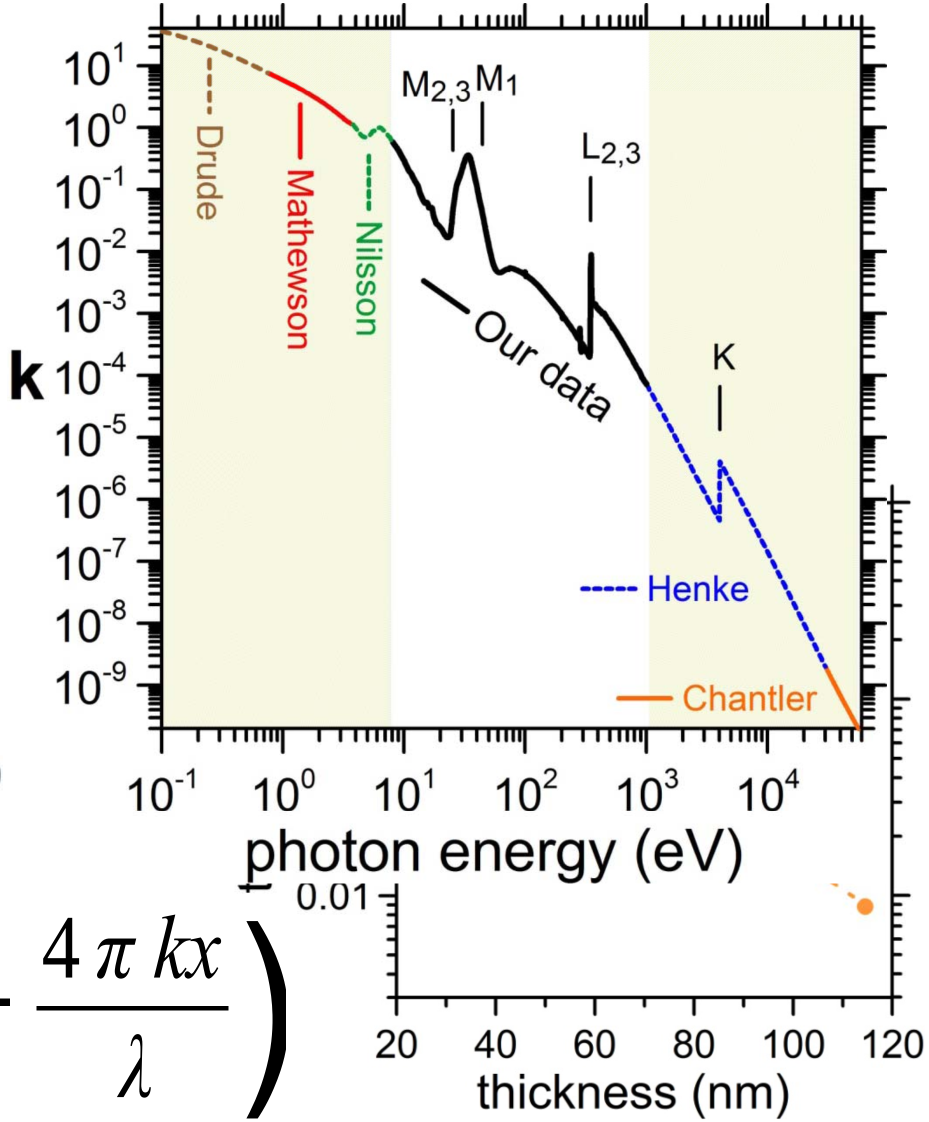
Measurements:
Transmittance vs
film thickness

$$T = \exp\left(-\frac{4\pi kx}{\lambda}\right)$$

KK: n from k. **Ca**

substrate: C film supported on grid



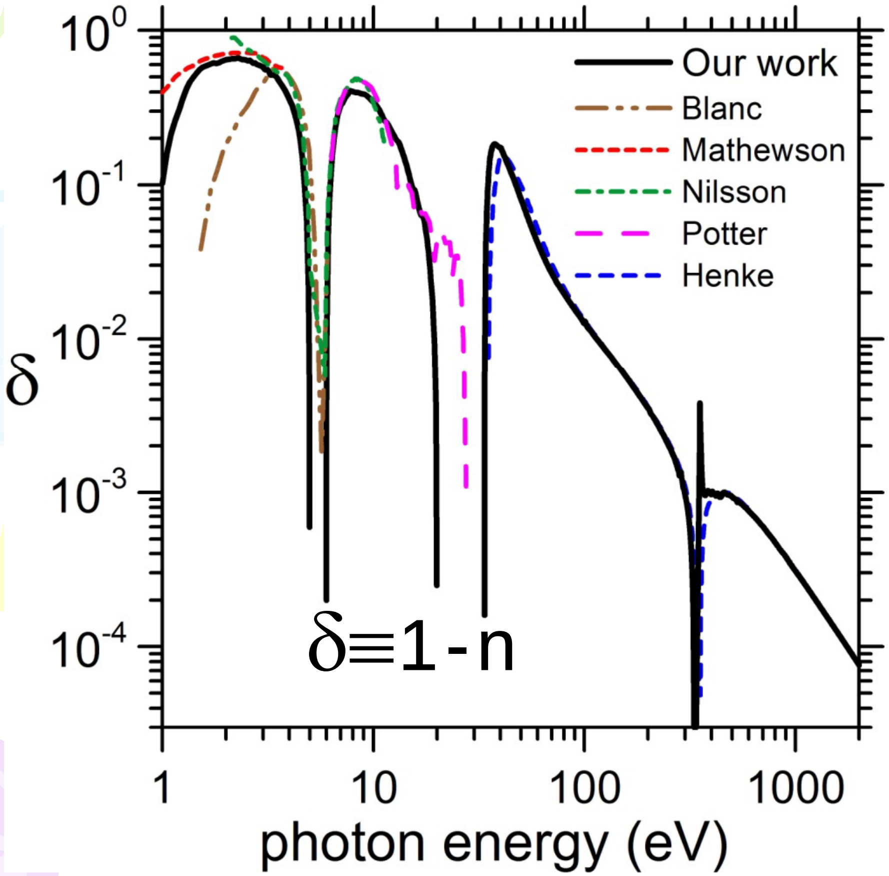


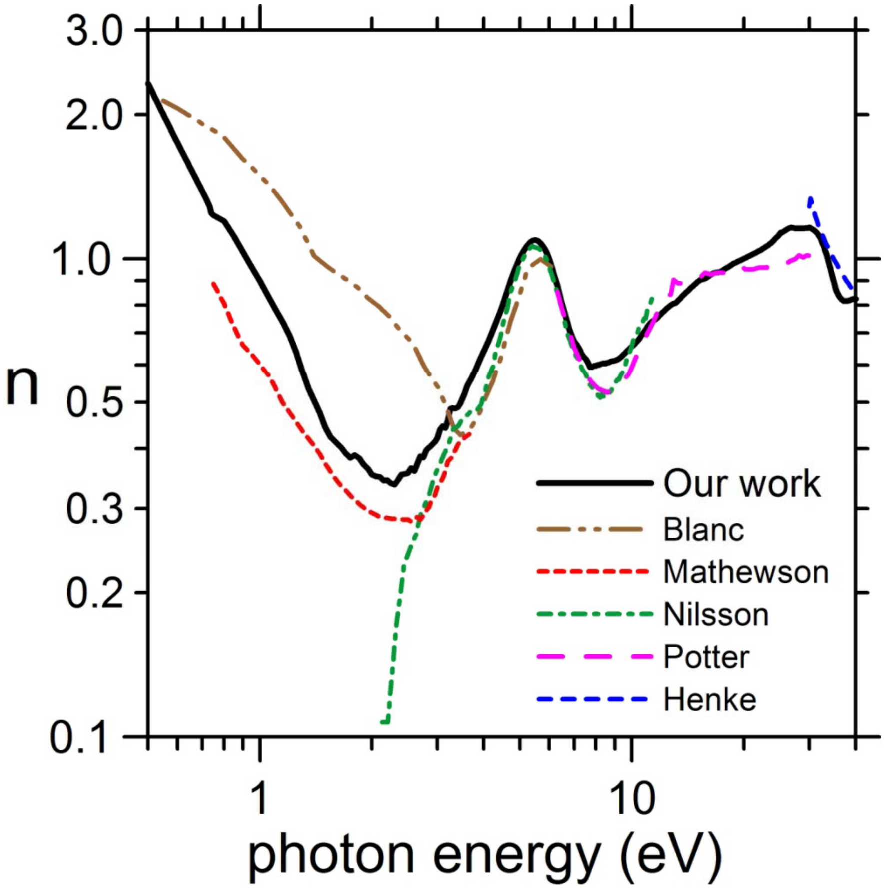
Measurements:
Transmittance vs
film thickness

$$T = \exp\left(-\frac{4\pi kx}{\lambda}\right)$$

KK: n from k. Ca

$n \equiv 1-\delta$

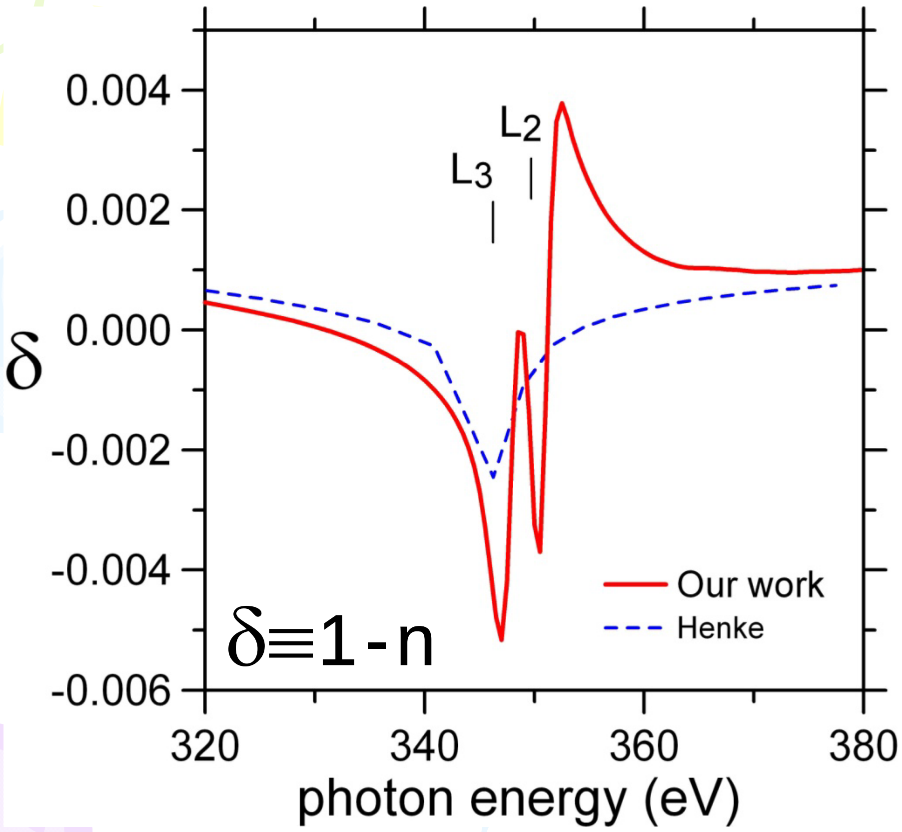


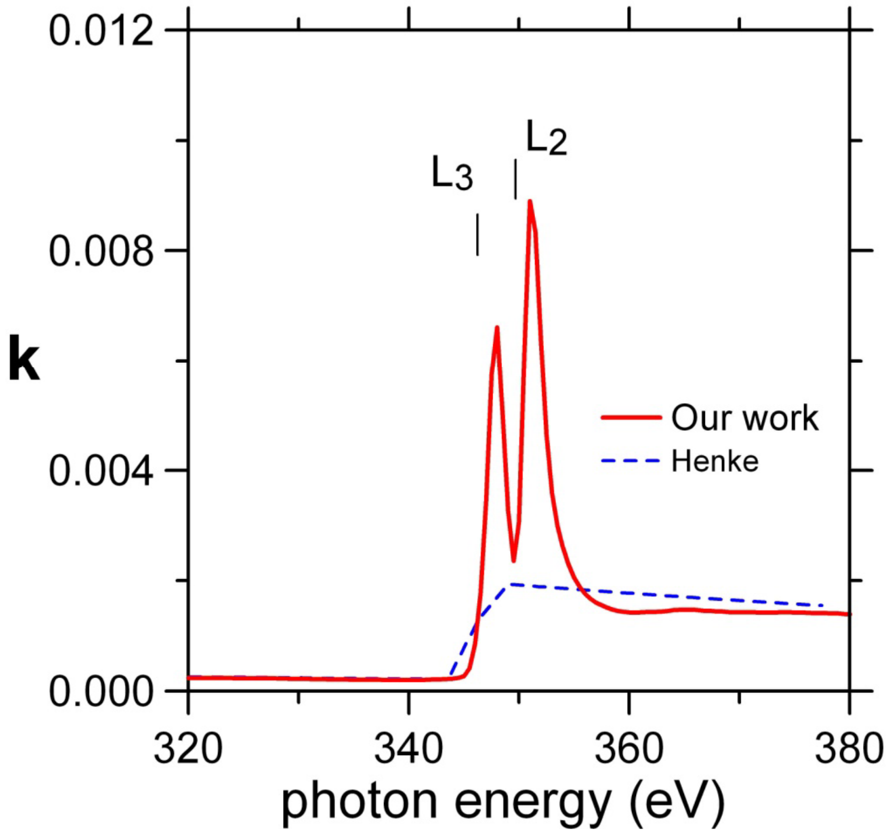


Rodríguez-de Marcos et al., Appl. Opt. **54**, 1910 (2015)

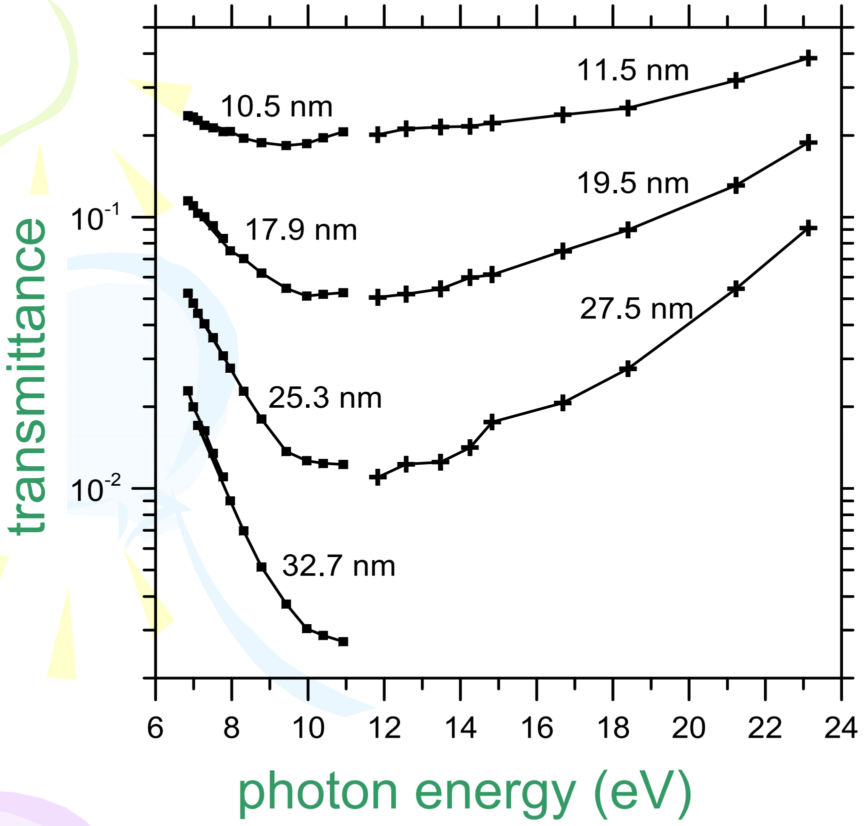
KK: n from k. Ca

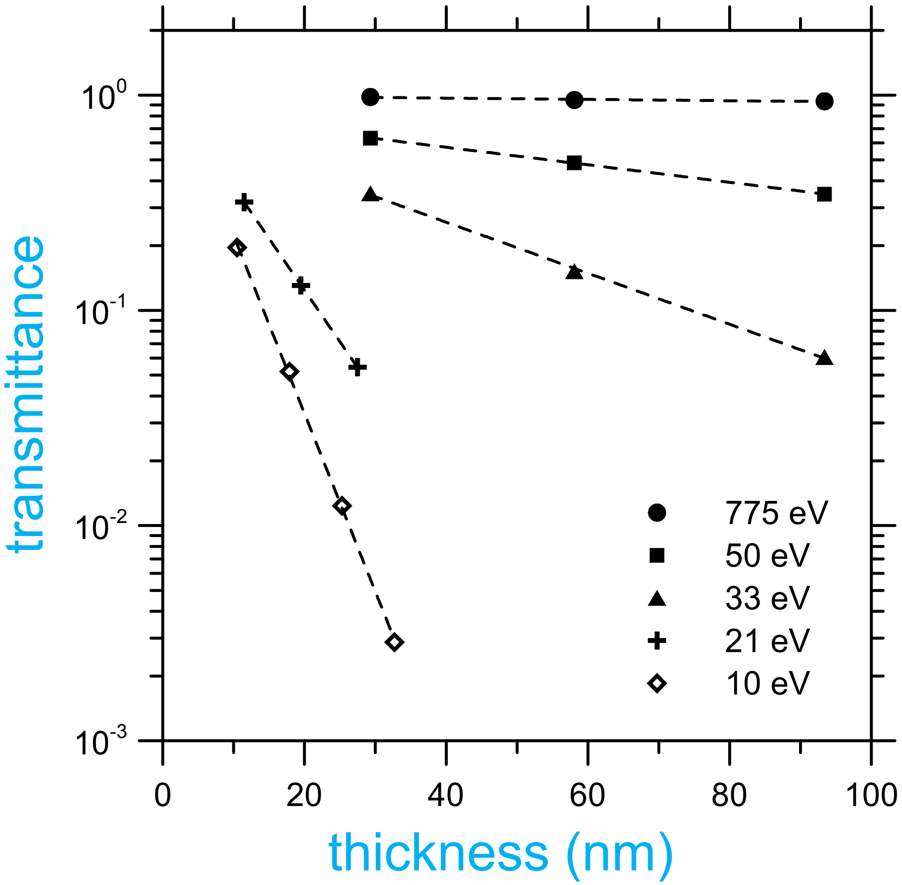
$L_{2,3}$ edge





n-k KK: **B**
k from transmittance vs film thickness

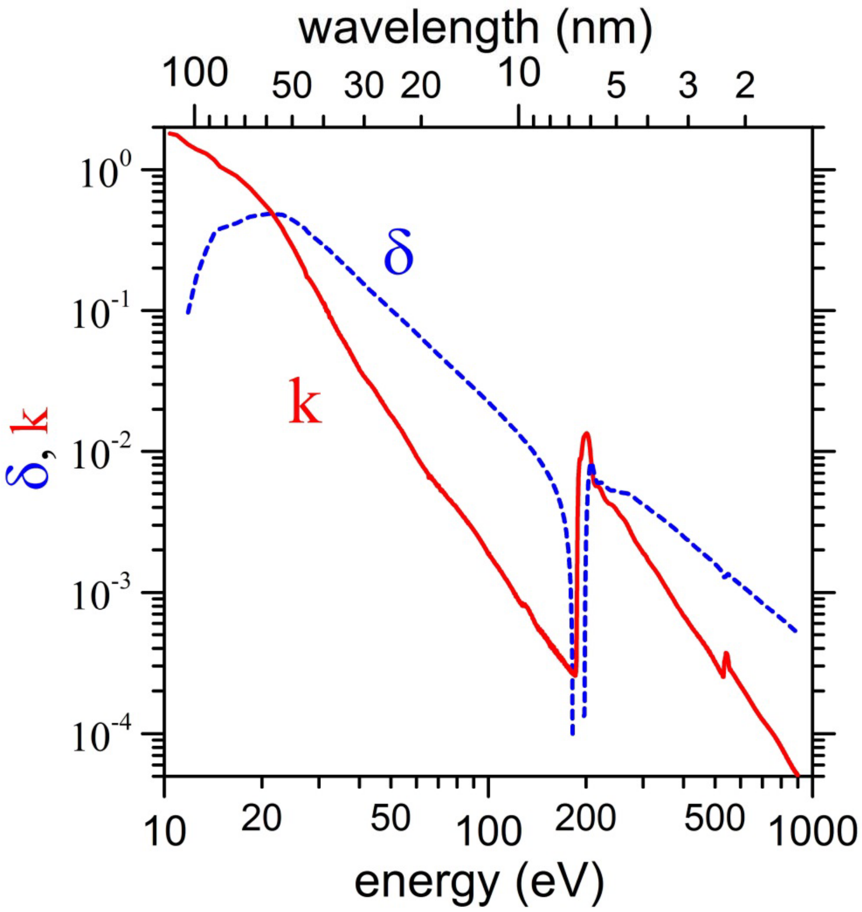


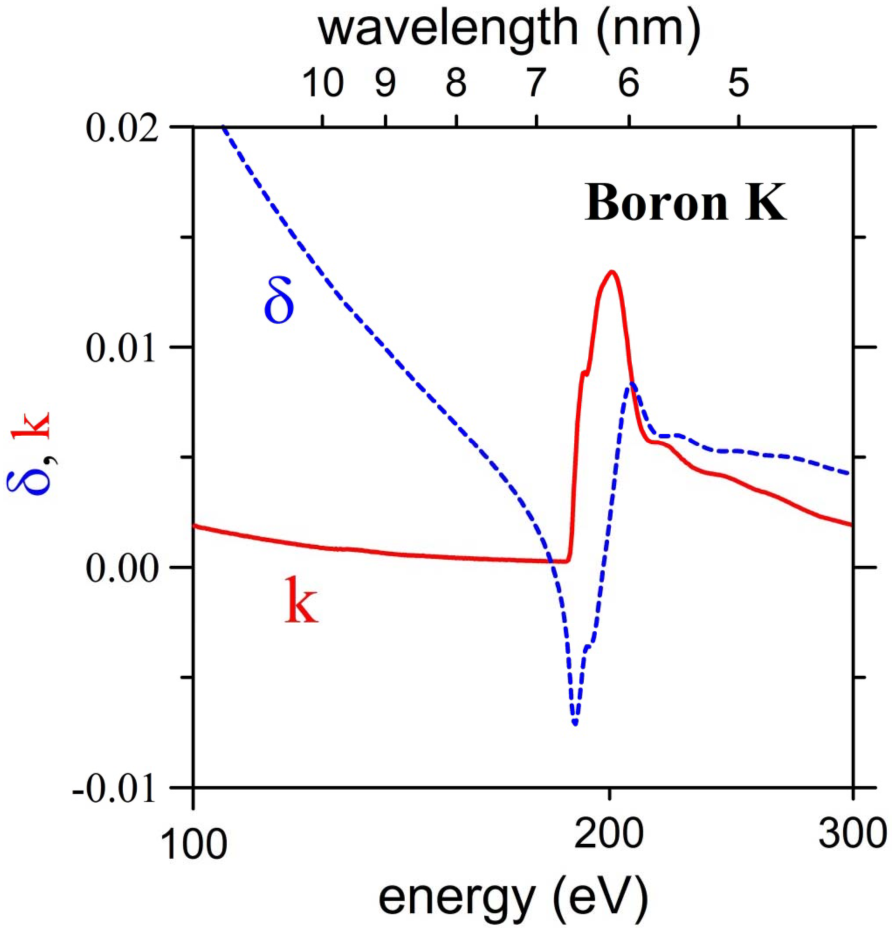


substrate: C film
supported on grid

$$\boldsymbol{T} = exp\left(-\frac{4\pi \boldsymbol{k} x}{\lambda}\right)$$

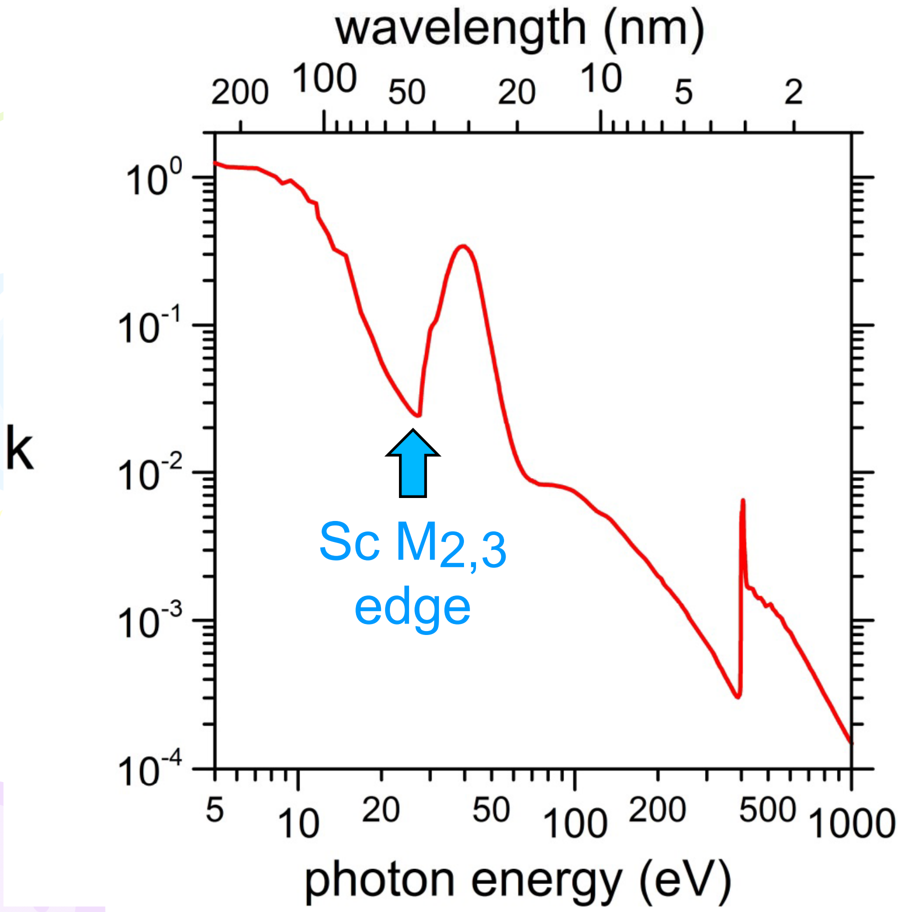
KK: n from k. **B**

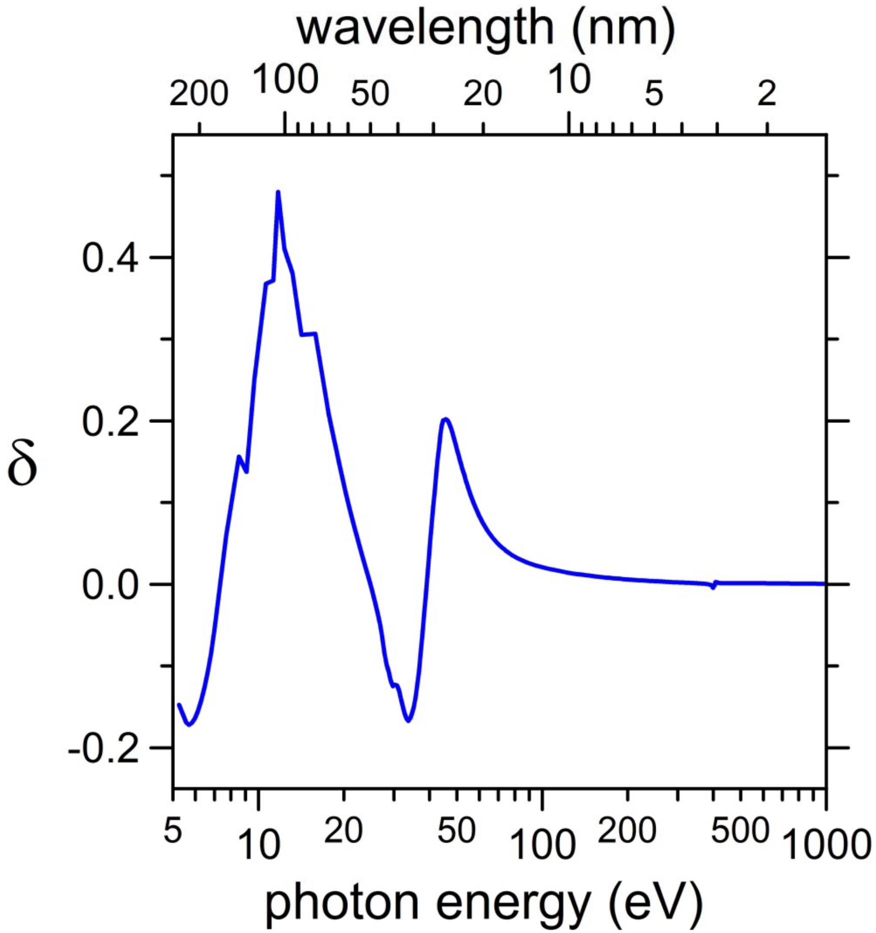




Fernández-Perea et al., J. Opt. Soc. Am. A. **24**, 3800 (2007)

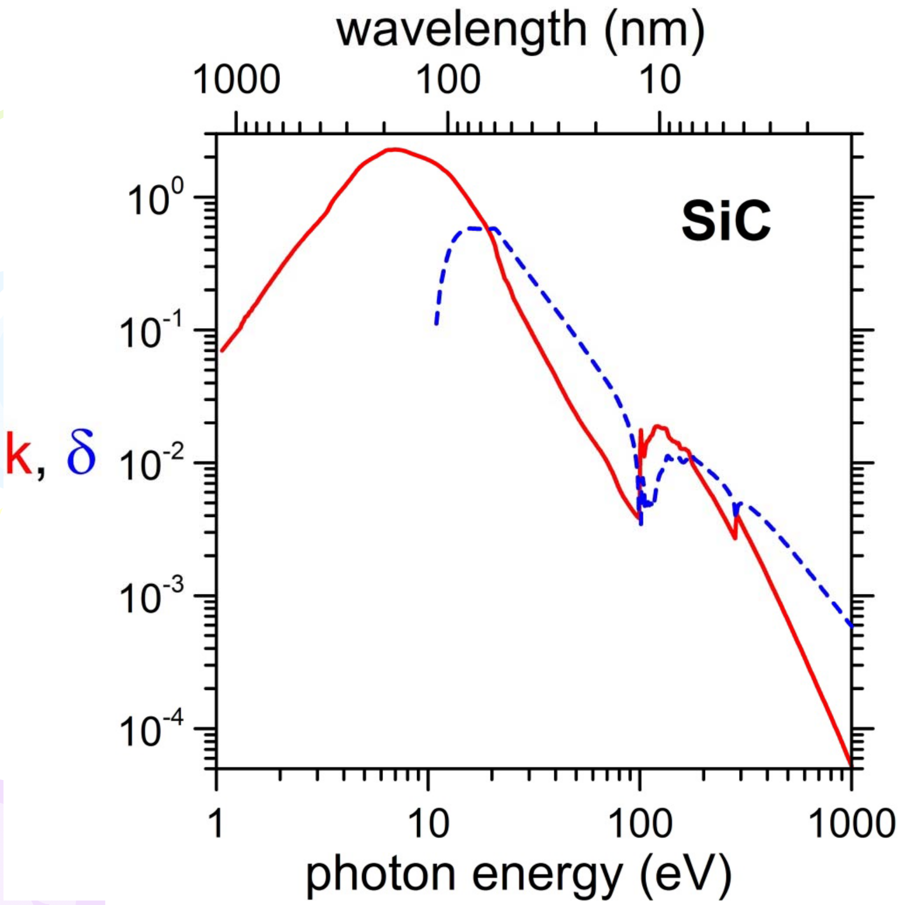
KK: n from k. **Sc**

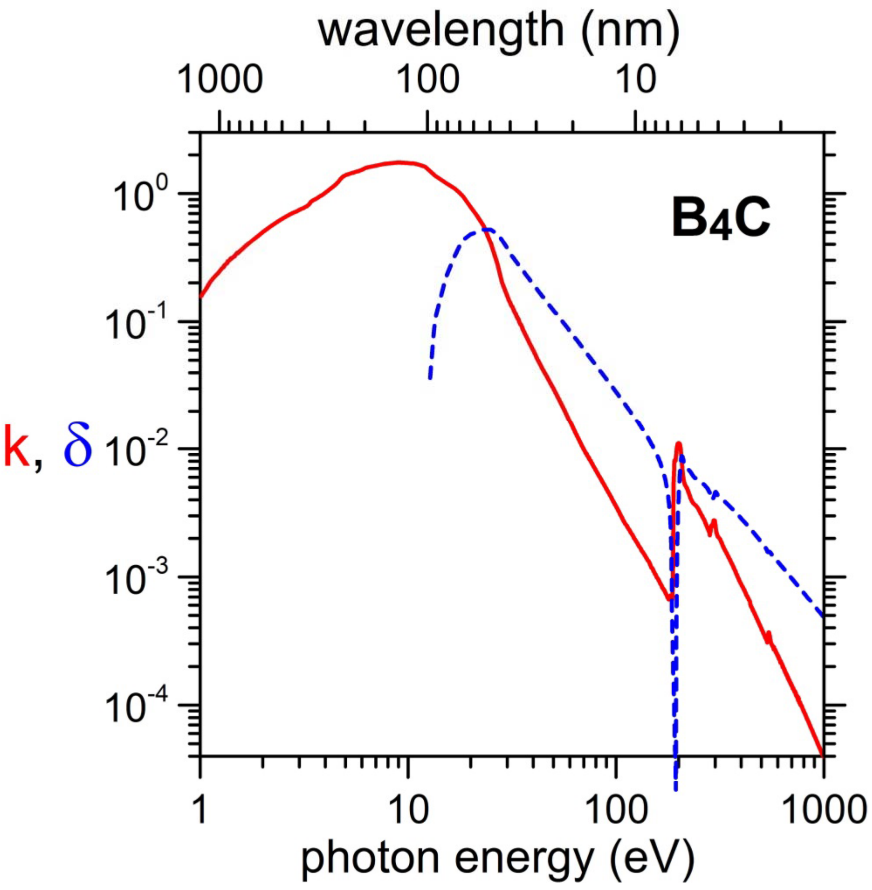




Fernández-Perea et al., J. Opt. Soc. Am. A. **23**, 2880 (2006)

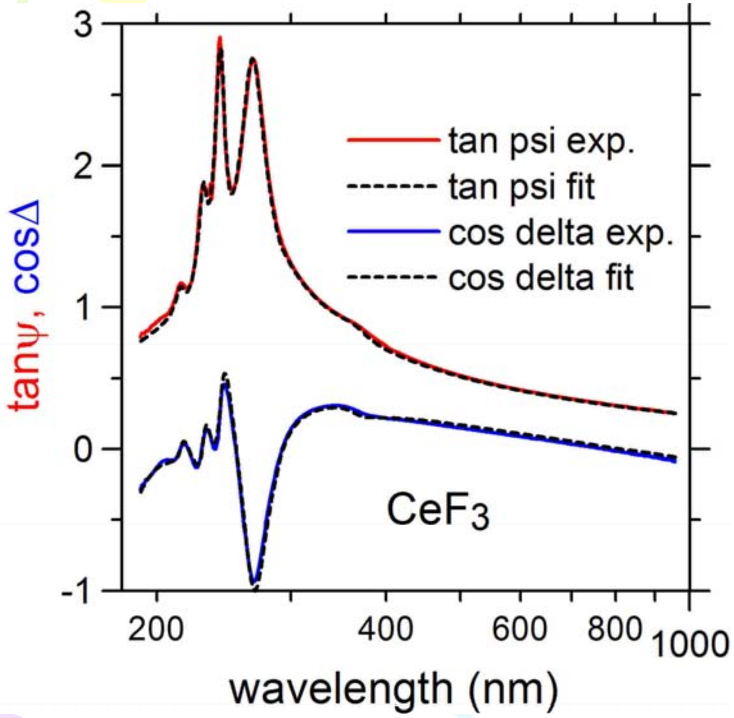
KK: n from k. **SiC and B_4C**

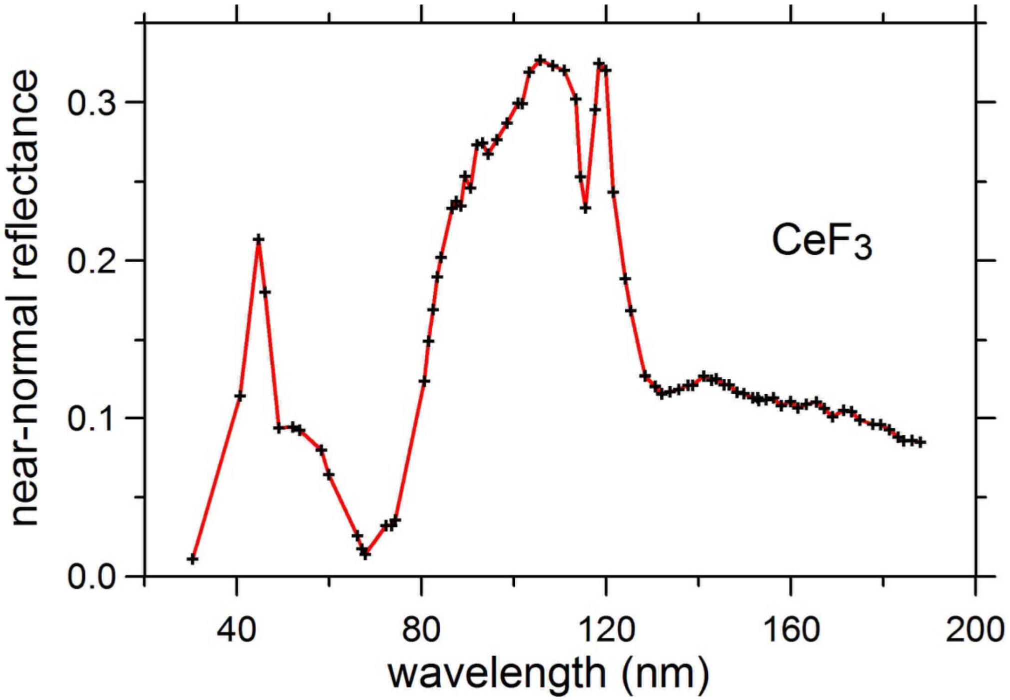


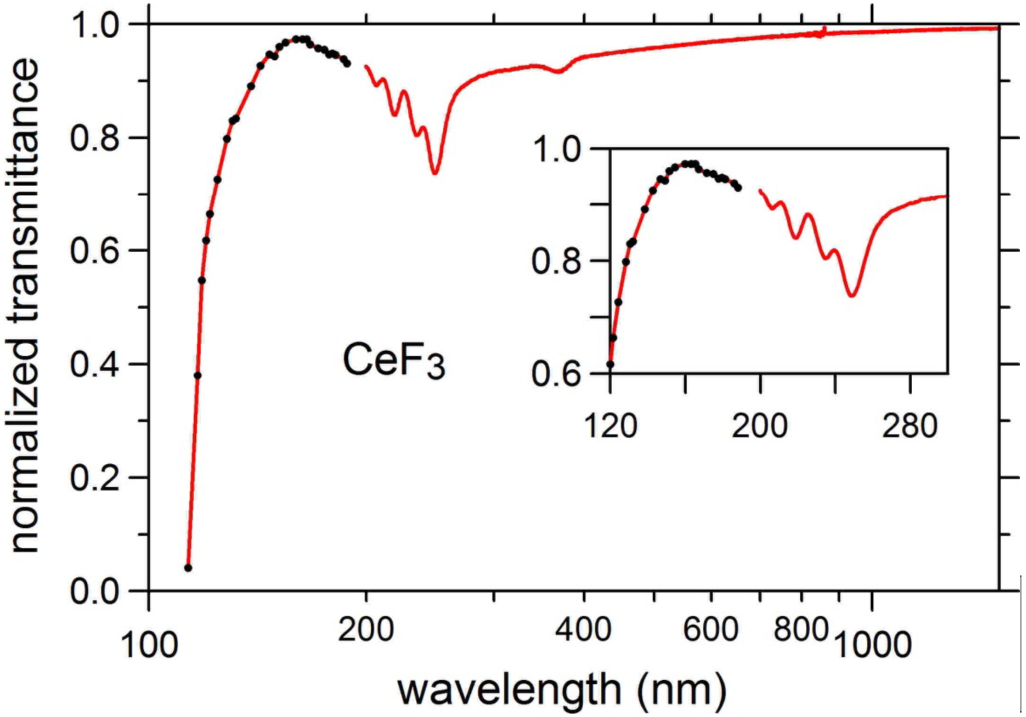


Larruquert et al., J. Opt. Soc. Am. A **28**, 2340 (2011),
ibid **29**, 117 (2012)

KK using R+T+Ellips: MgF2, LaF3, **CeF3**

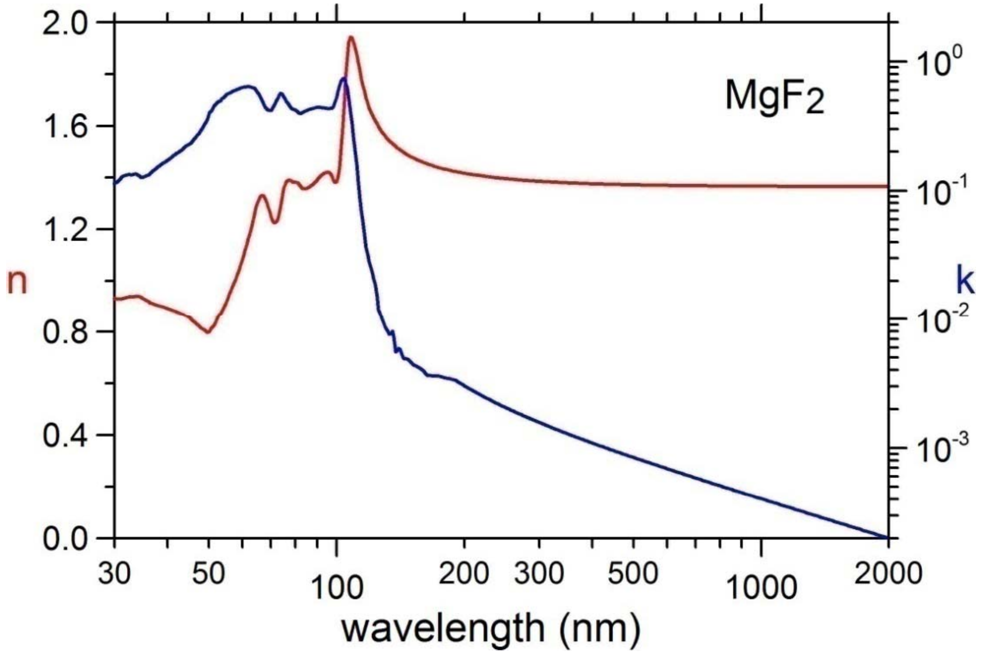


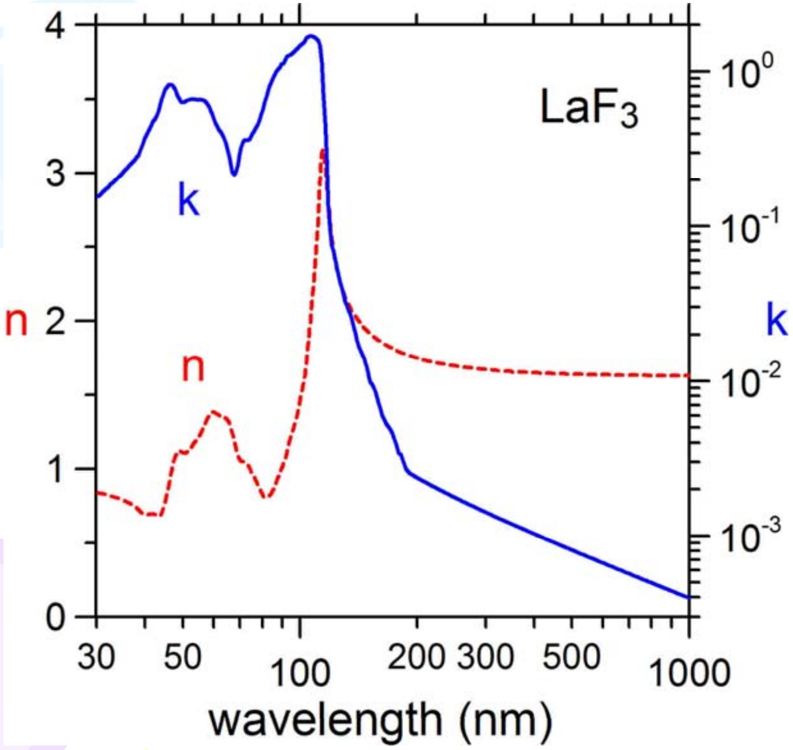


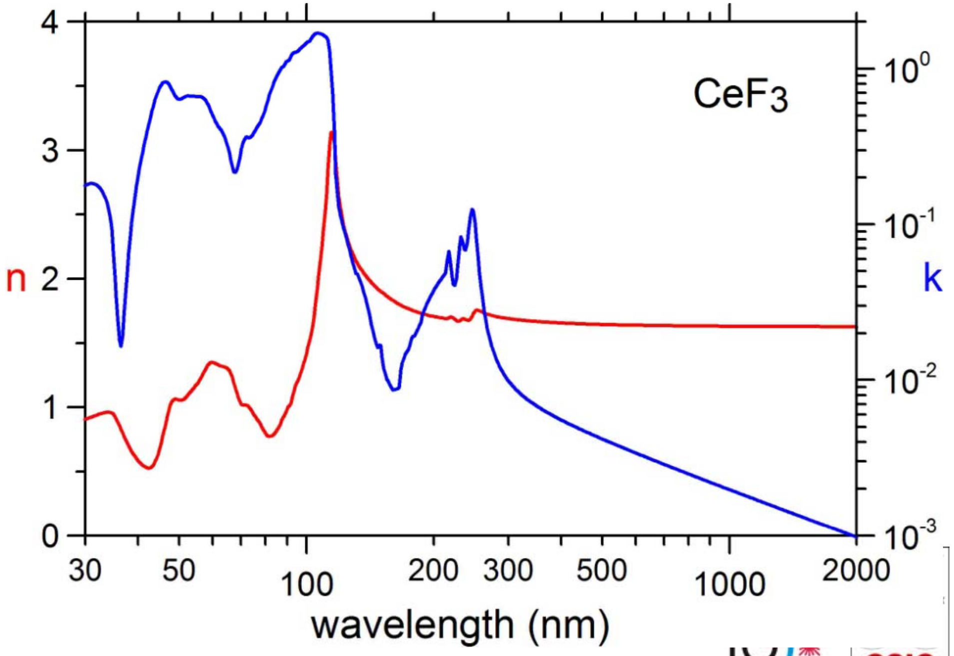


Rodríguez-de Marcos, Proc. SPIE **9627**, 96270B (2015)

KK using R+T+Ellips: MgF2, LaF3, CeF3







KK evaluation: global sum rules

f sum rule

$$\int_0^\infty E' k(E') dE' = \frac{\pi}{4} \frac{N_{el} e^2 h^2}{\varepsilon_0 m} \div Z$$

Z=atomic number

Inertial sum rule

$$\int_0^\infty \left[n(E) - 1 \right] dE = 0$$

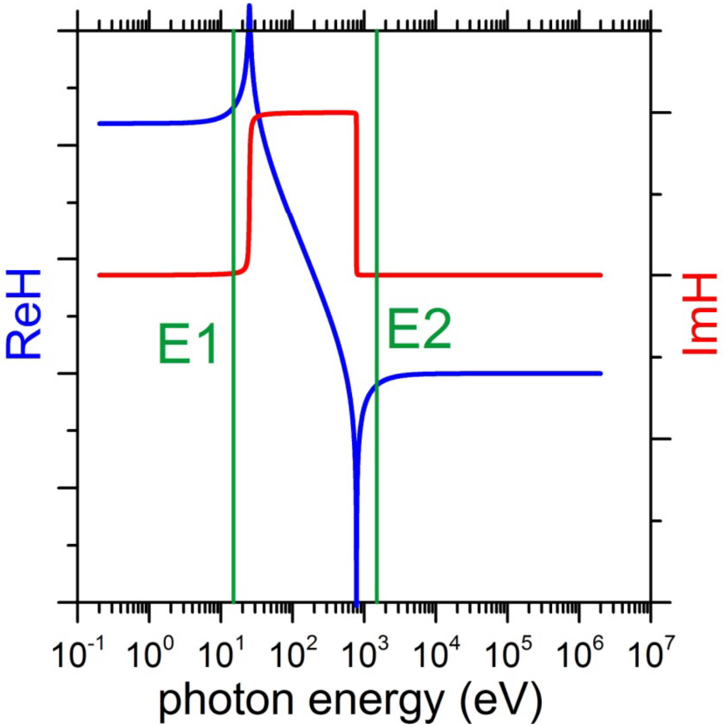
Evaluation parameter:

$$\zeta = \frac{\int_0^\infty \left[n(E) - 1 \right] dE}{\int_0^\infty \left| n(E) - 1 \right| dE}$$

$$|\zeta| < 0.005$$

Sum rules with window functions

- Increase weight in the desired spectral range [E1,E2]



KK evaluation: sum rules

f sum rule

$$n_{eff}=\frac{4\varepsilon_0 m}{Ne^2\hbar^2}\int_0^\infty E'k(E')dE'\div Z$$

Z=atomic number

global sum rules

Inertial sum rule

$$\int_0^\infty [n(E)-1]dE=0$$

local sum rules

$$n_{eff}=\frac{4\varepsilon_0 m}{Ne^2\hbar^2\left(E_1^{\ 2}-E_2^{\ 2}\right)}$$

$$\times\int_0^\infty E'^3\,\mathrm{Im}\{H_1(E')[N(E')-1]\}dE'\div Z$$

$$\int_0^\infty E'^{-2}\,\mathrm{Re}\{H_2(E')[N(E')-1]\}dE'=0$$

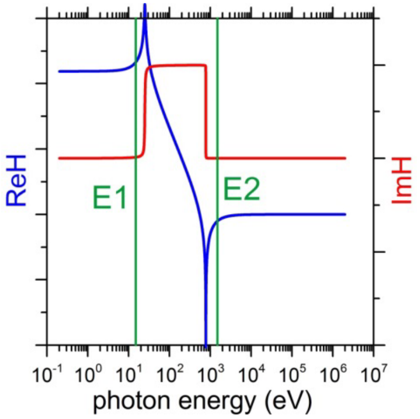
$$\int_0^\infty E'^{-1}\,\mathrm{Im}\{H_2(E')[N(E')-1]\}dE'=0$$

$$\int_0^\infty \mathrm{Re}\{H_2(E')[N(E')-1]\}dE'=0$$

$$\int_0^\infty E'\,\mathrm{Im}\{H_2(E')[N(E')-1]\}dE'=0$$

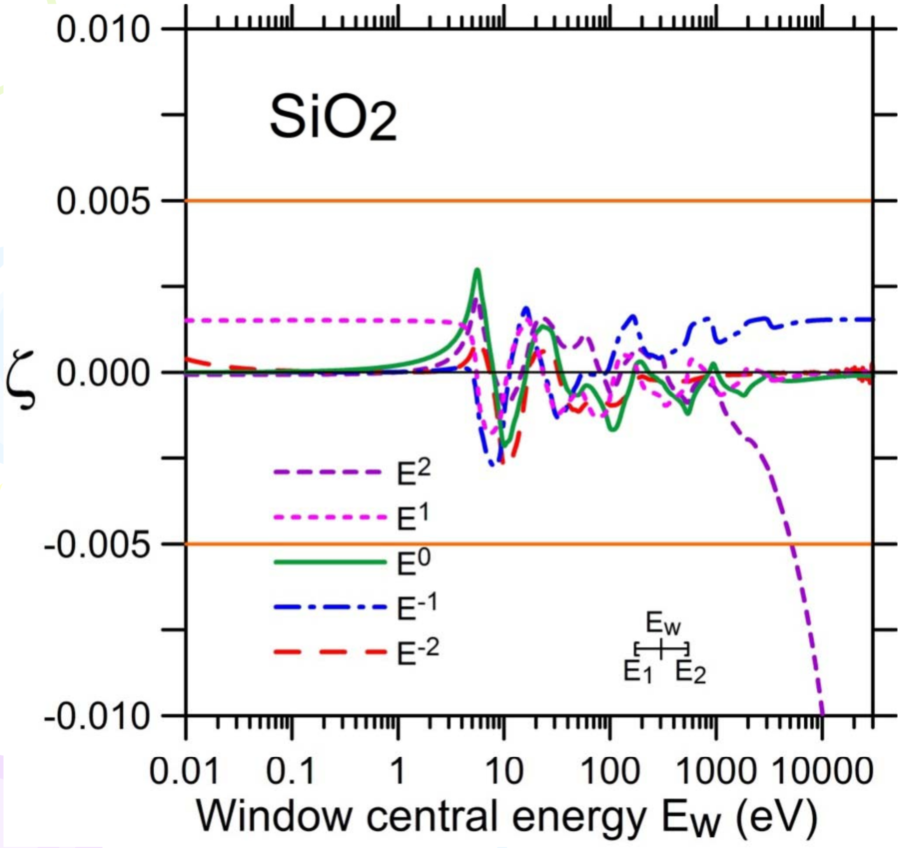
$$\int_0^\infty E'^2\,\mathrm{Re}\{H_2(E')[N(E')-1]\}dE'=0$$

Window function



J. Opt. **18**, 075606 (2016)

KK evaluation: sum rules with window functions



$$\int_0^\infty E'^2\,\mathrm{Re}\{H_2(E')[N(E')-1]\}dE'=0$$

$$\int_0^\infty E'\,\mathrm{Im}\{H_2(E')[N(E')-1]\}dE'=0$$

$$\int_0^\infty \mathrm{Re}\{H_2(E')[N(E')-1]\}dE'=0$$

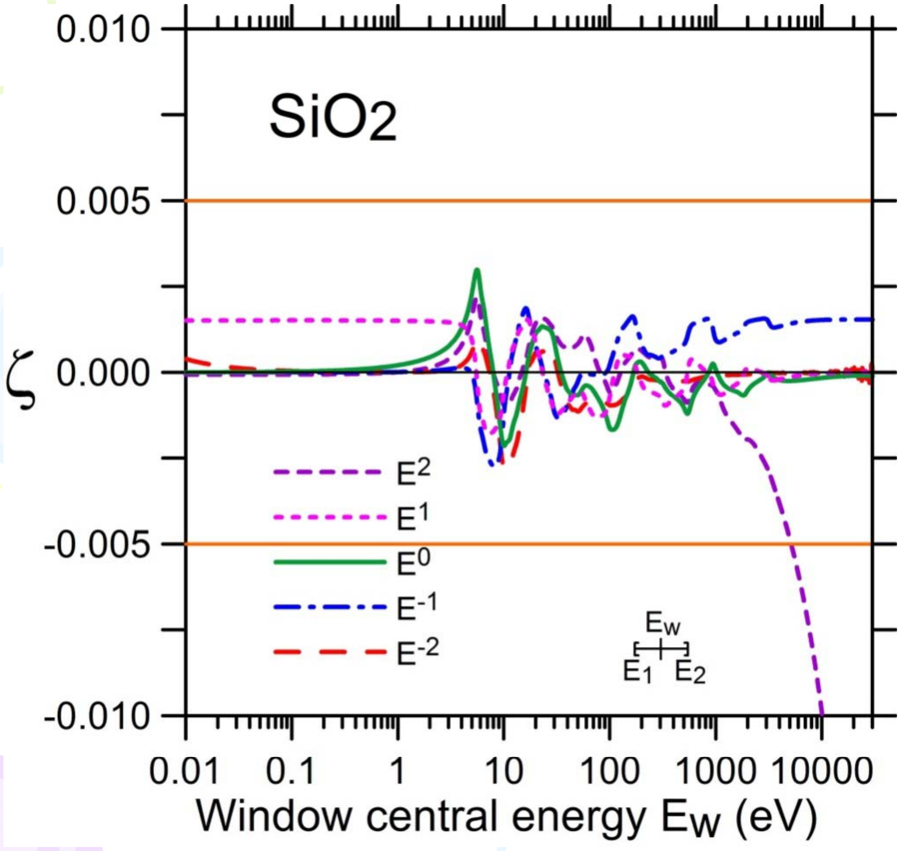
$$\int_0^\infty E'^{-1}\,\mathrm{Im}\{H_2(E')[N(E')-1]\}dE'=0$$

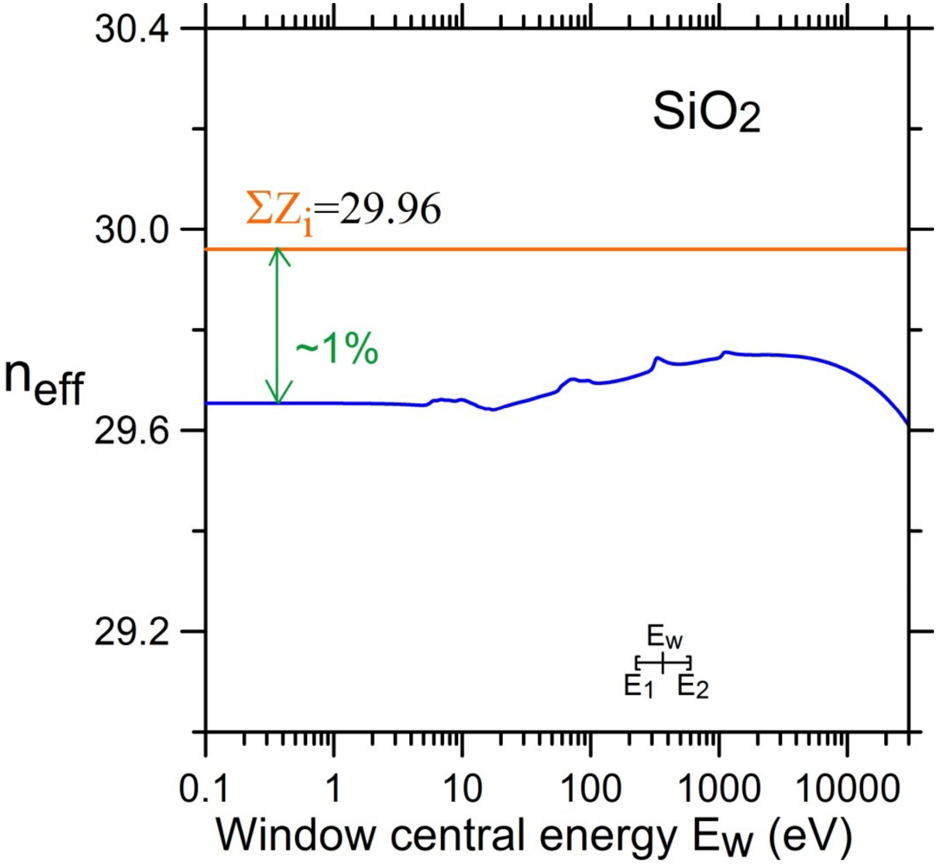
$$\int_0^\infty E'^{-2}\,\mathrm{Re}\{H_2(E')[N(E')-1]\}dE'=0$$

Inertial sum rules

Rodríguez-de Marcos, Opt. Mater. Express **6**, 3622 (2016)

KK evaluation: sum rules with window functions



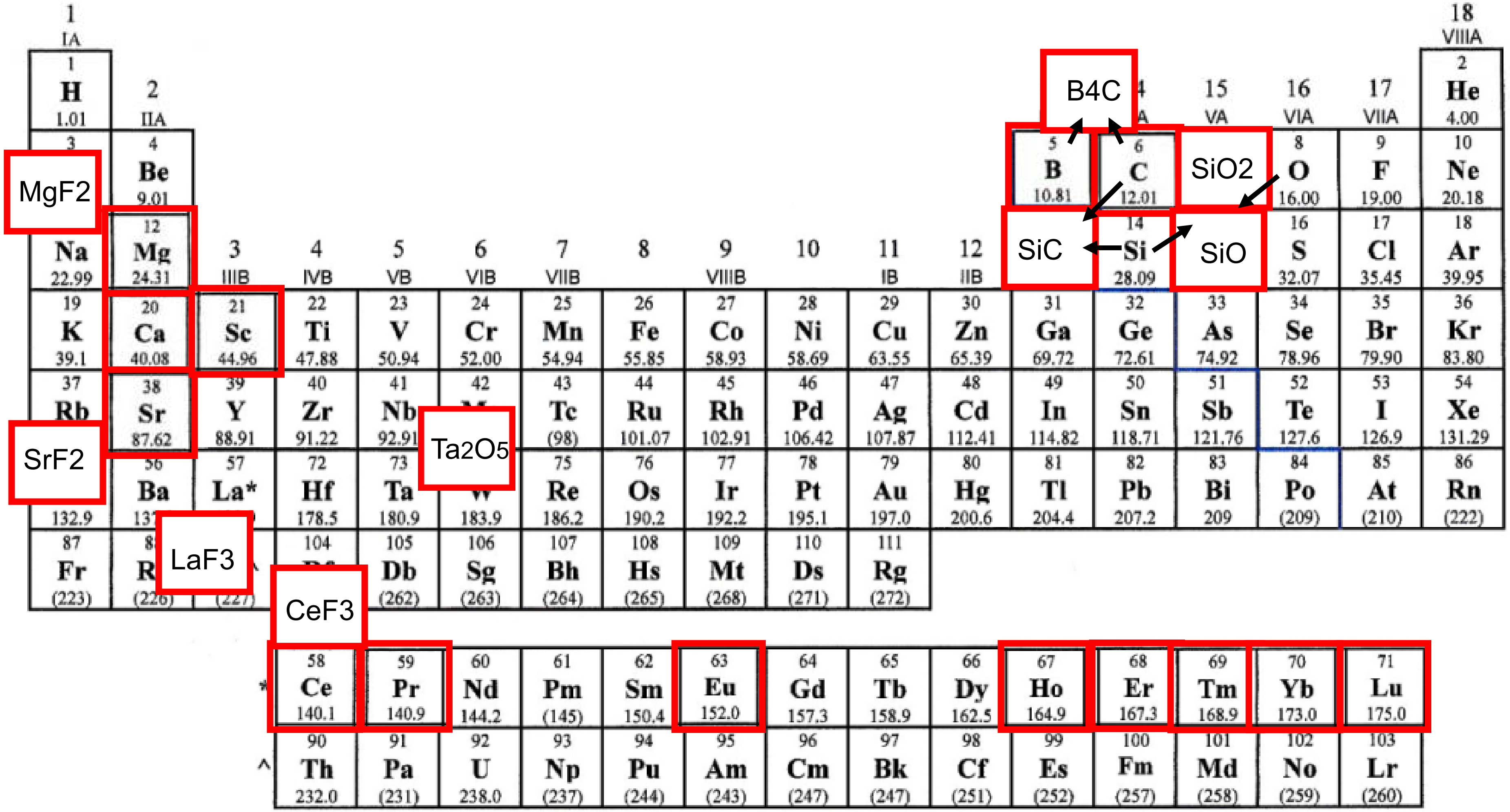


Inertial sum rules

f sum rule

Rodríguez-de Marcos, Opt. Mater. Express **6**, 3622 (2016)

Self-consistent characterization of materials



Reflectometer

- VUV Reflectometer for in-situ measurement of coatings
- Connected in UHV with deposition chambers in ISO-8 clean room
- λ main range: 40-200 nm
- Measurements in wider ranges at BEAR/Elettra and 6.3.2/ALS

Deposition of ML coatings

Knowledge of (n,k): to develop novel and efficient MLS

Broadband mirrors:

- Al/MgF2

- SiC; B4C

Transmittance filters:

- (Al/MgF2)$_n$ peaked at λ>120 nm

Narrowband mirrors:

-Al/LiF/SiC: peak at 102 nm

-Fluorides peaked at λ>120 nm

Polarizers:

- (Al/MgF2)$_n$ at λ>120 nm

75-cm diameter chamber in ISO-6 clean room to satisfy space requirements.

Can we measure (n,k) and develop coatings for your application?

Acknowledgments

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Collaborators in (n,k) determination:

- M. Fernández-Perea, M. Vidal-Dasilva, S. Garcia, A. P. Pérez-Marín
- A. M. Malvezzi, A. Giglia, S. Nannarone, F. Frassetto, L. Poletto, D. Garoli, N. Mahne, D. Bajoni
- R. Soufli, E. Gullikson, A. L. Aquila, F. Salmassi
- P. Martin, A. Bendavid