

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

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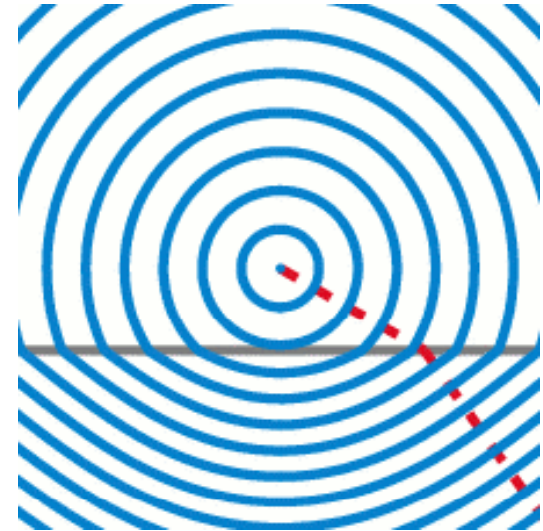
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Refractive index= optical constants

* It describes how light propagates through an optical medium

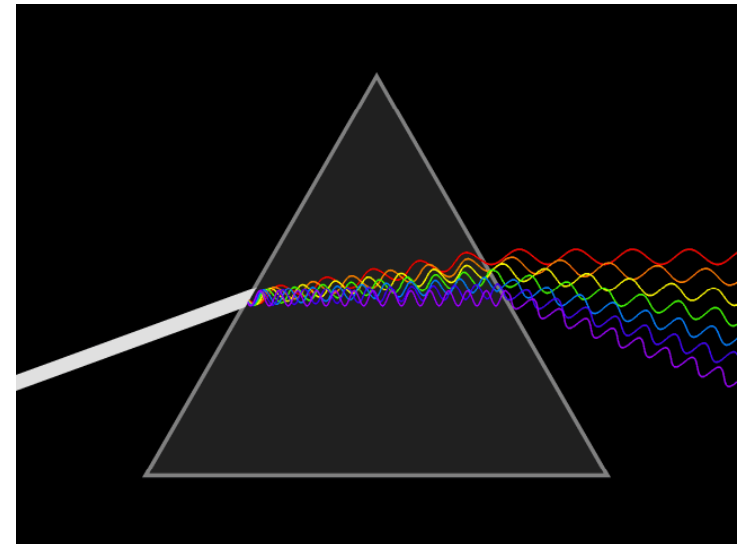


* It governs:

- 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)

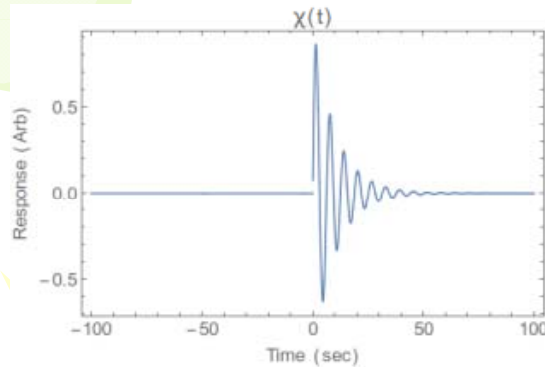
are not independent of each other

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



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

Response can't precede the stimulus

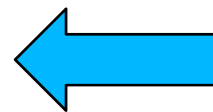


$$P(t) = \epsilon_0 \int_{-\infty}^{+\infty} \chi(t - t') E(t') dt'$$

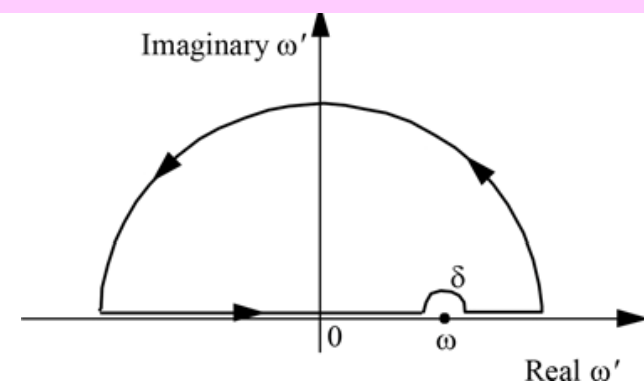
$$\chi(\omega) = \int_{-\infty}^{+\infty} \chi(t) e^{-i\omega t} dt$$

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

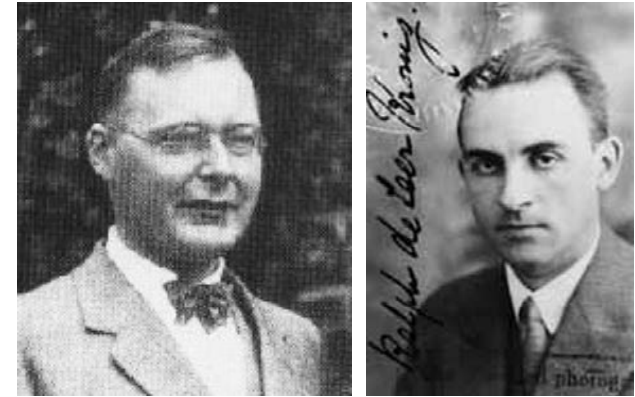
$$\epsilon_1(\omega) - 1 = \frac{2}{\pi} P \int_0^{\infty} \frac{\omega' \epsilon_2(\omega')}{\omega'^2 - \omega^2} d\omega'$$



$$\epsilon_2(\omega) = -\frac{2\omega}{\pi} P \int_0^{\infty} \frac{\epsilon_1(\omega') - 1}{\omega'^2 - \omega^2} d\omega'$$



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

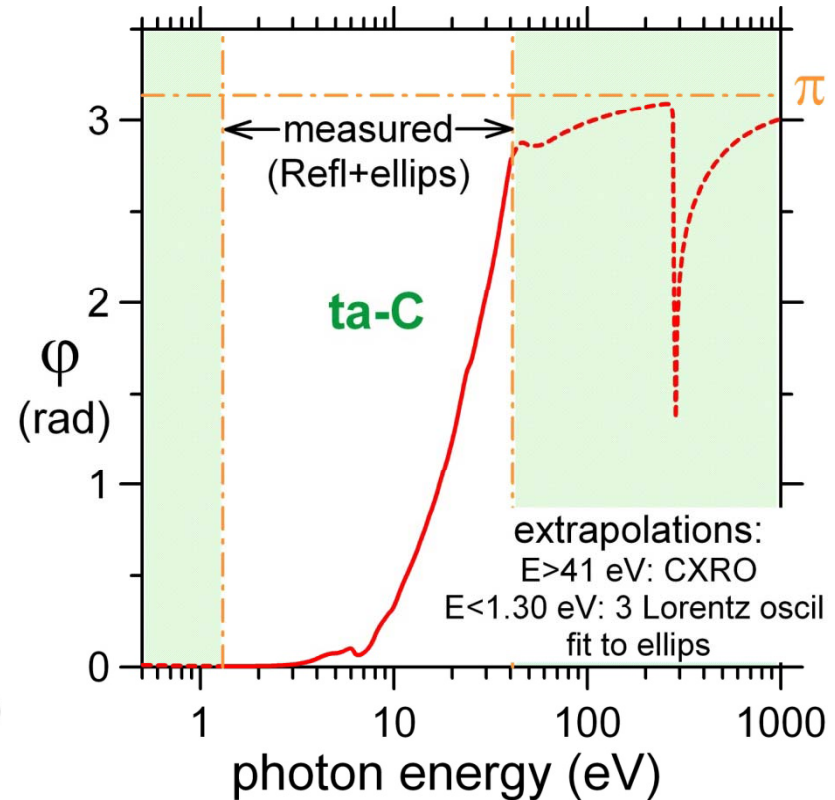
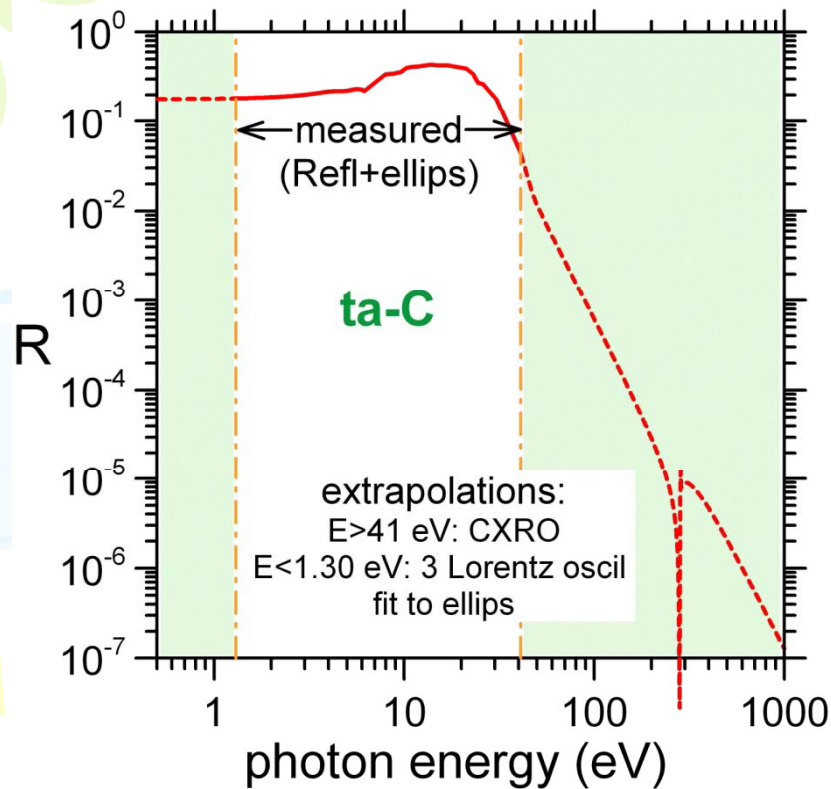
$$r = R^{1/2} e^{i\varphi}$$

$$\ln r = \frac{1}{2} \ln R + i\varphi$$

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

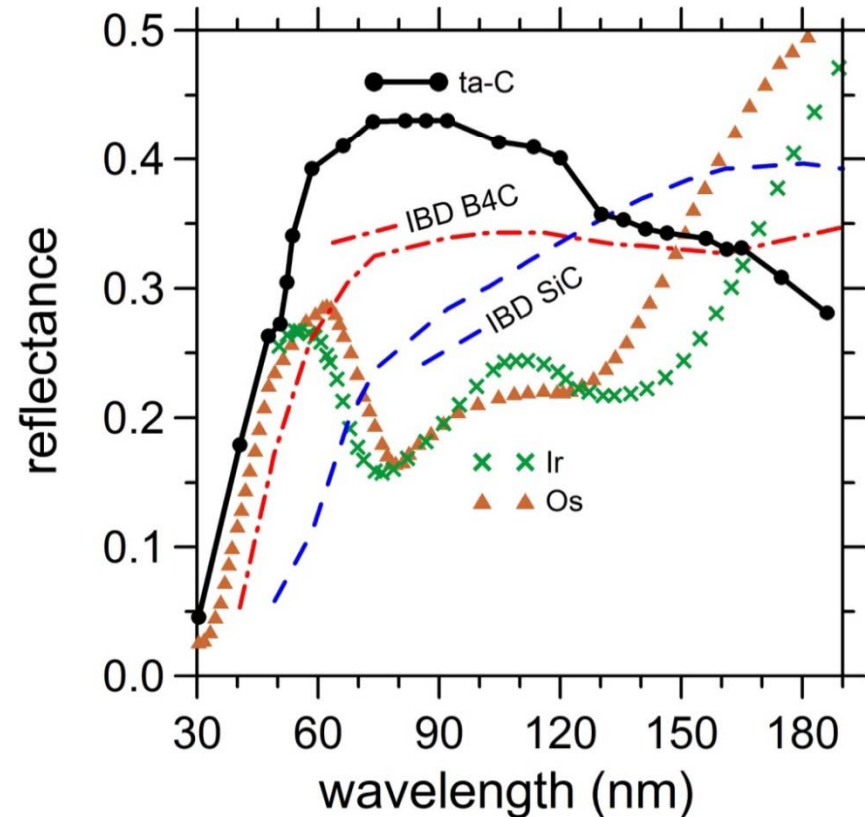
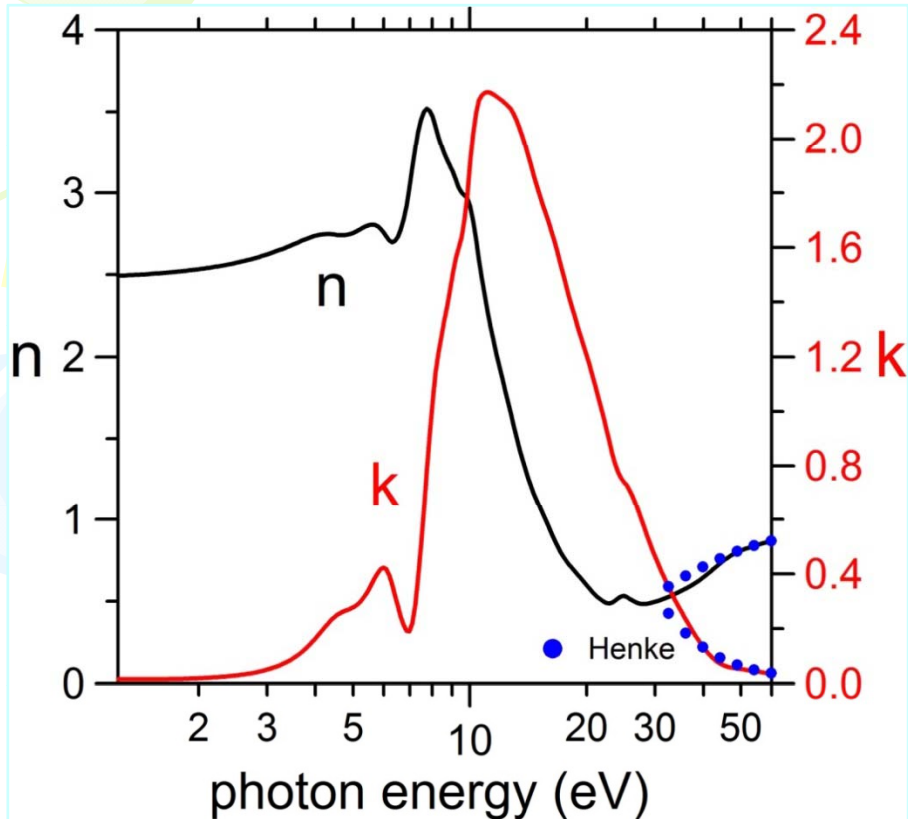


KK by reflectance: ta-C tetrahedral amorphous Carbon



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

KK by reflectance: ta-C tetrahedral amorphous Carbon



From R, φ : n, k :

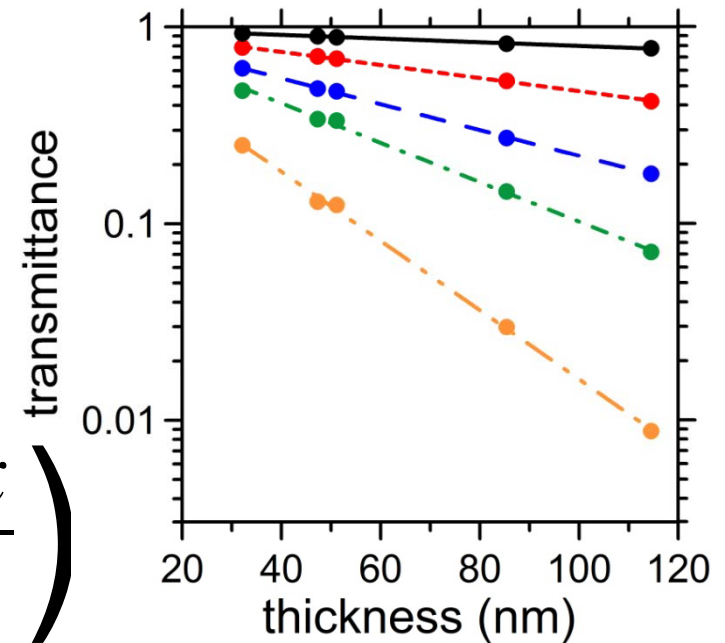
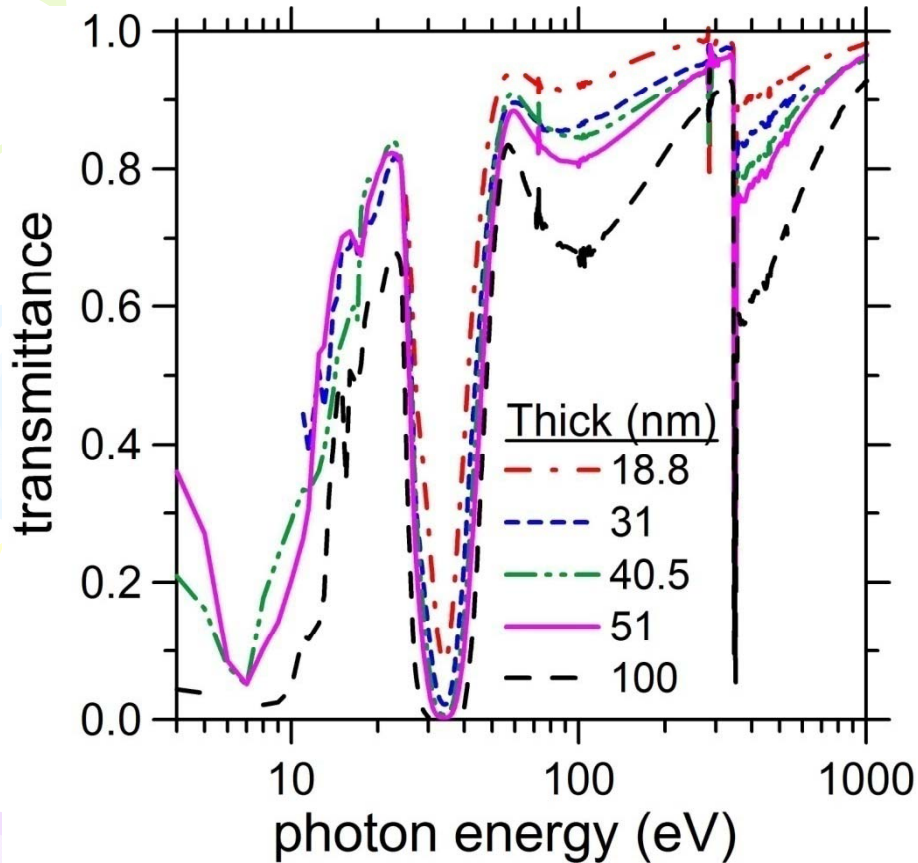
$$R^{1/2} e^{i\varphi} = \frac{n-1+ik}{n+1+ik}$$

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

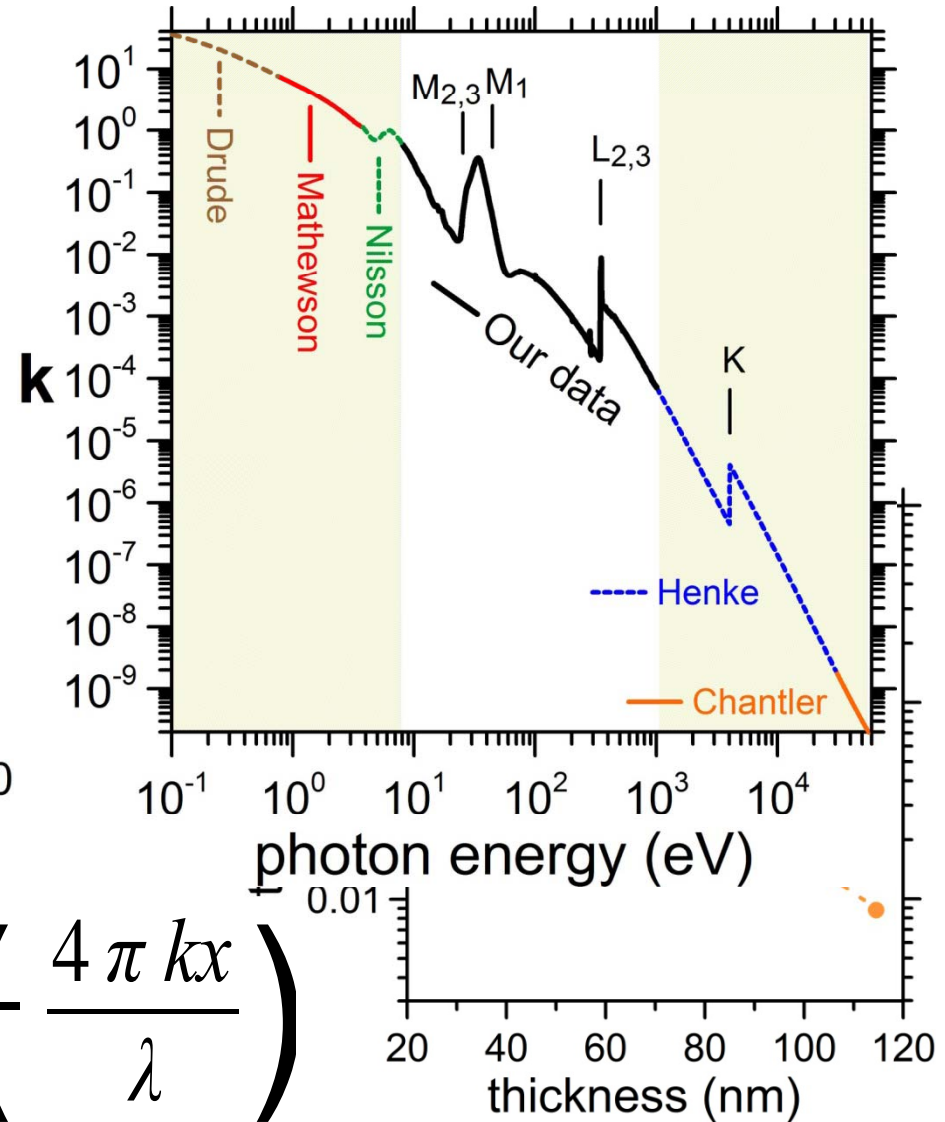
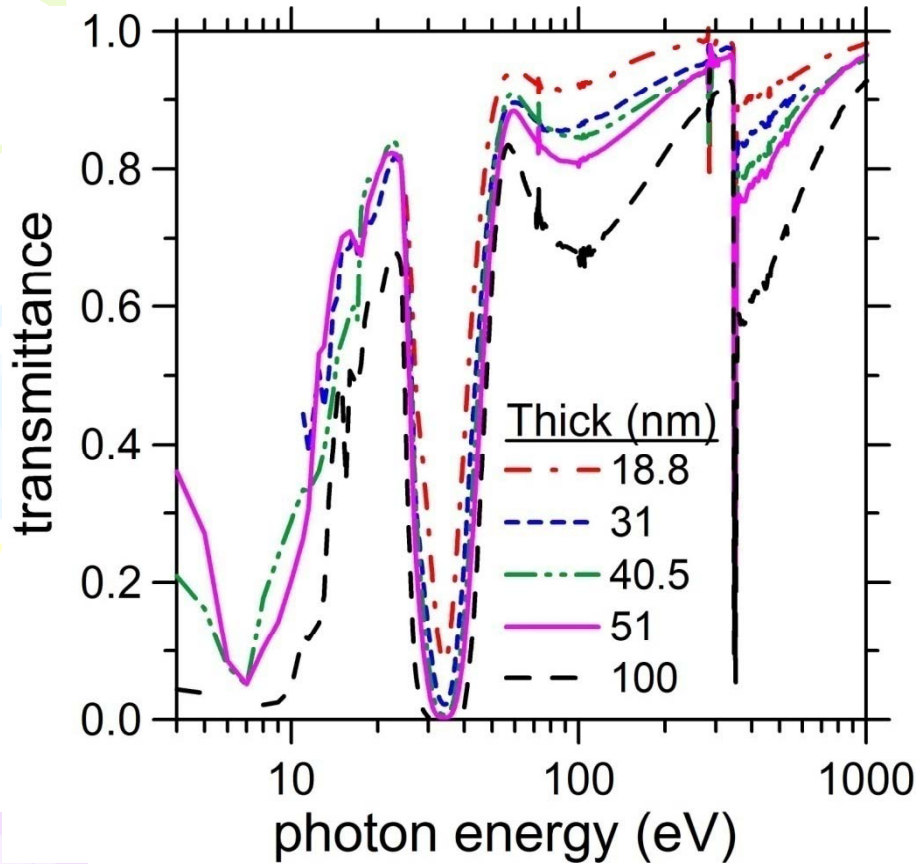


Measurements:
Transmittance vs
film thickness

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

KK: n from k. Ca

substrate: C film supported on grid

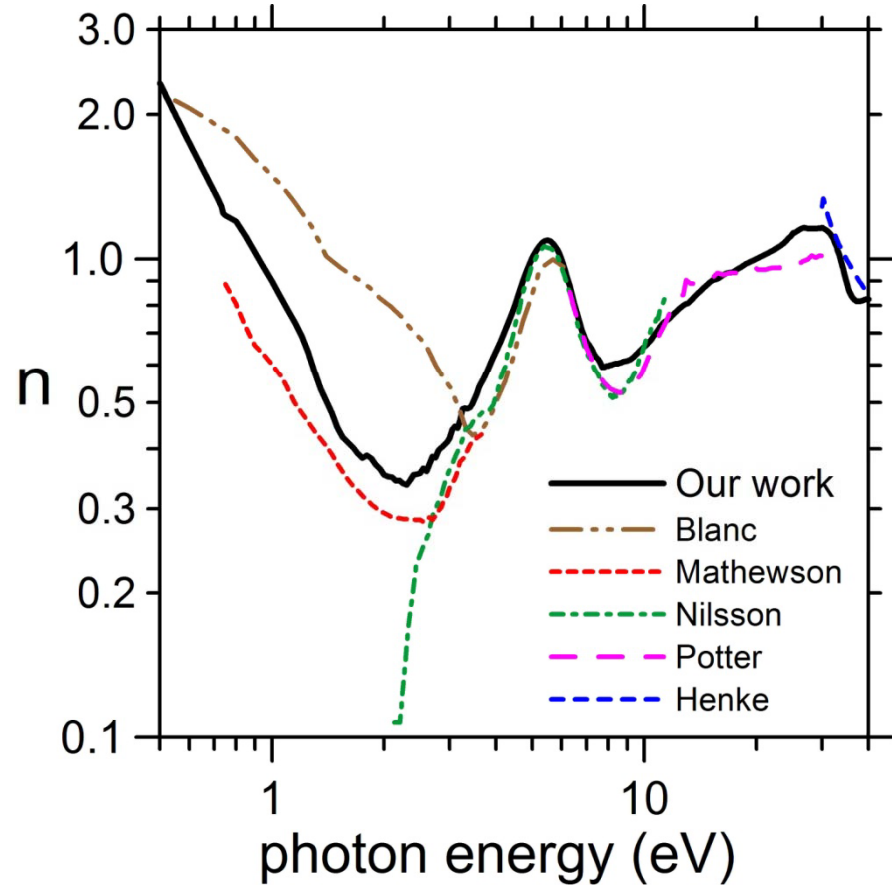
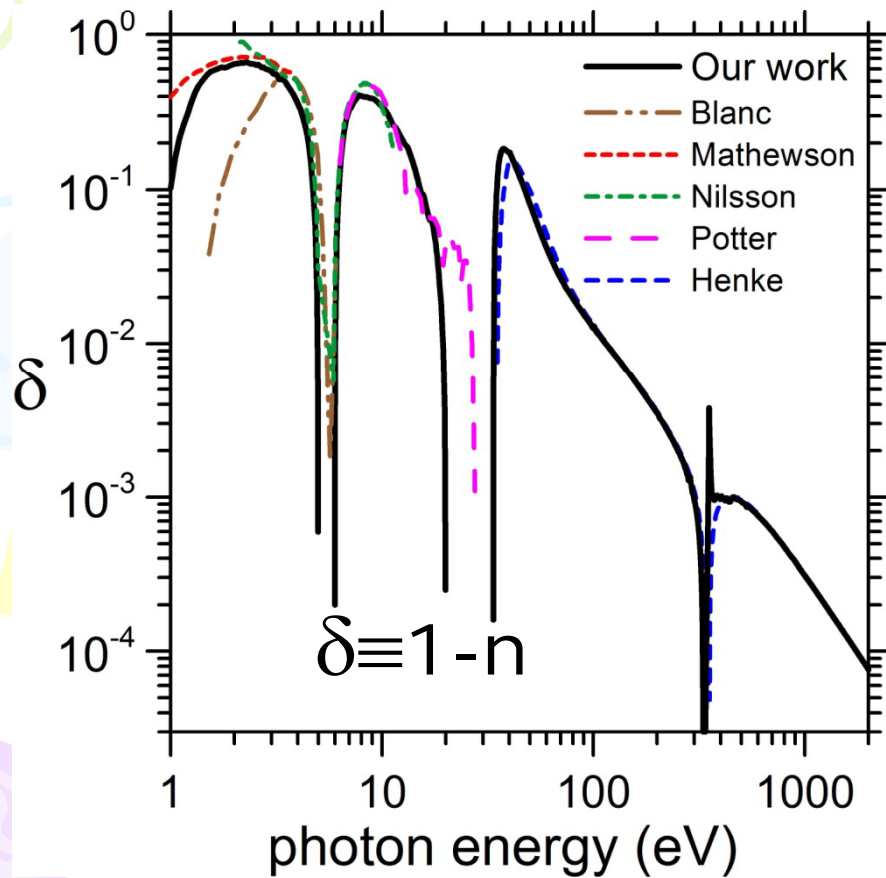


Measurements:
Transmittance vs
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$$T = \exp\left(-\frac{4\pi kx}{\lambda}\right)$$

KK: n from k. Ca

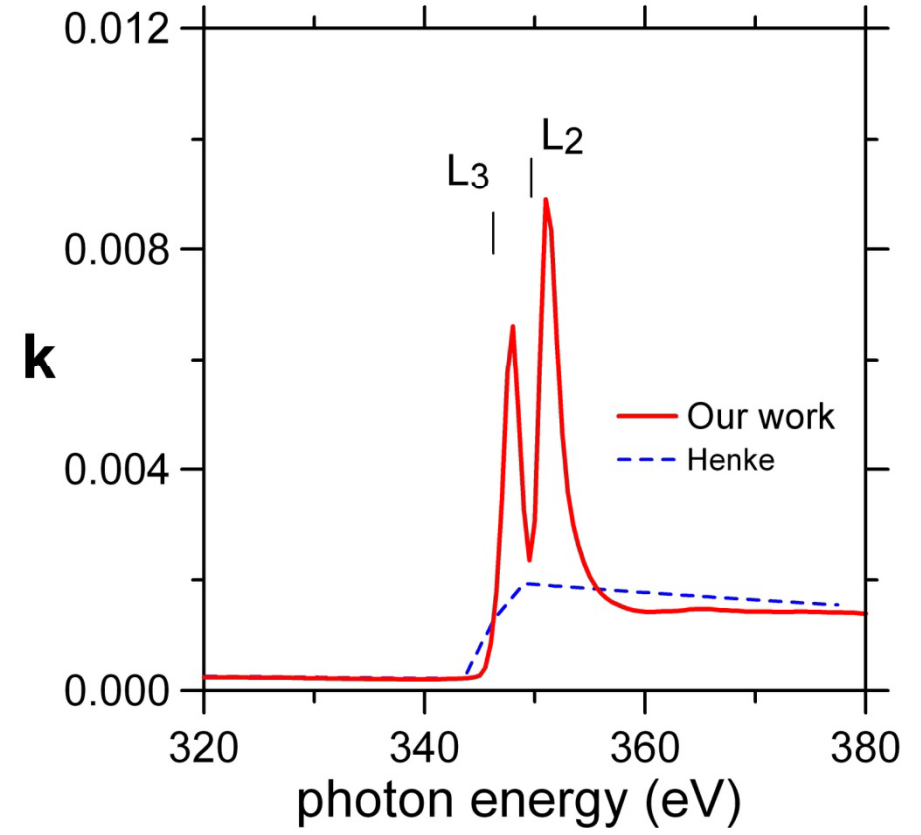
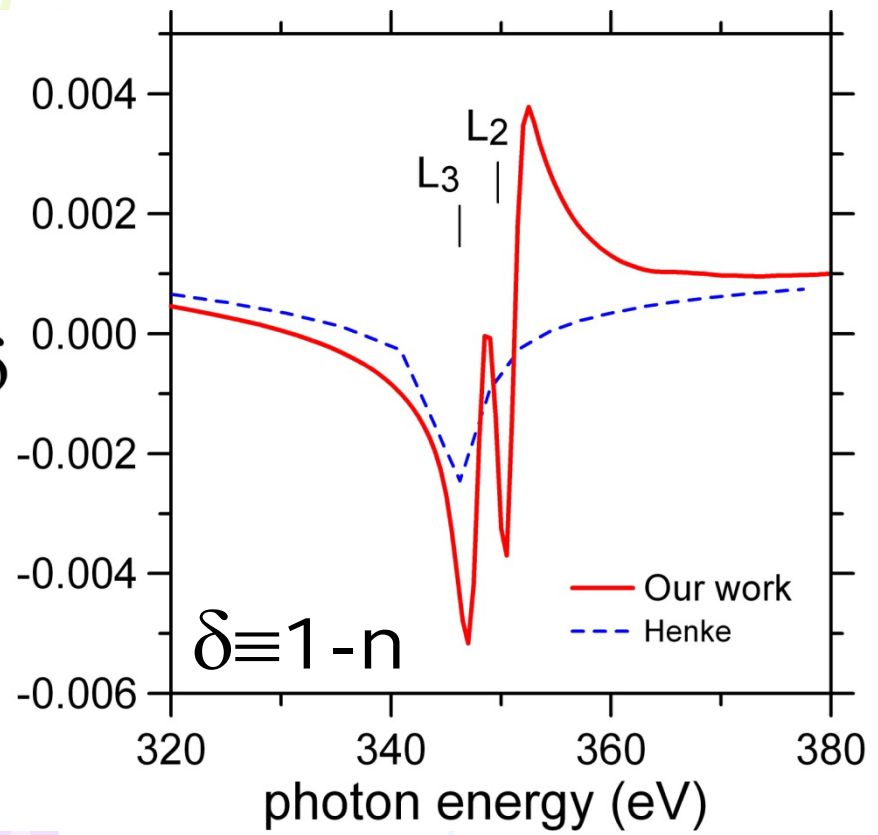
$$n \equiv 1 - \delta$$



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

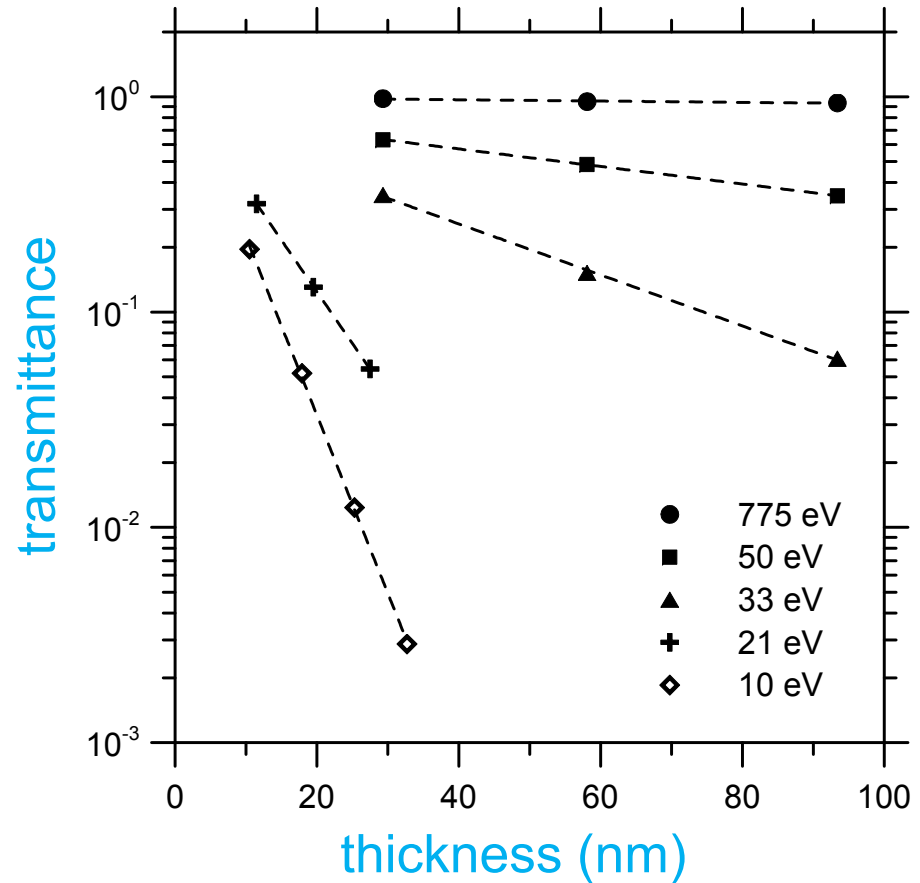
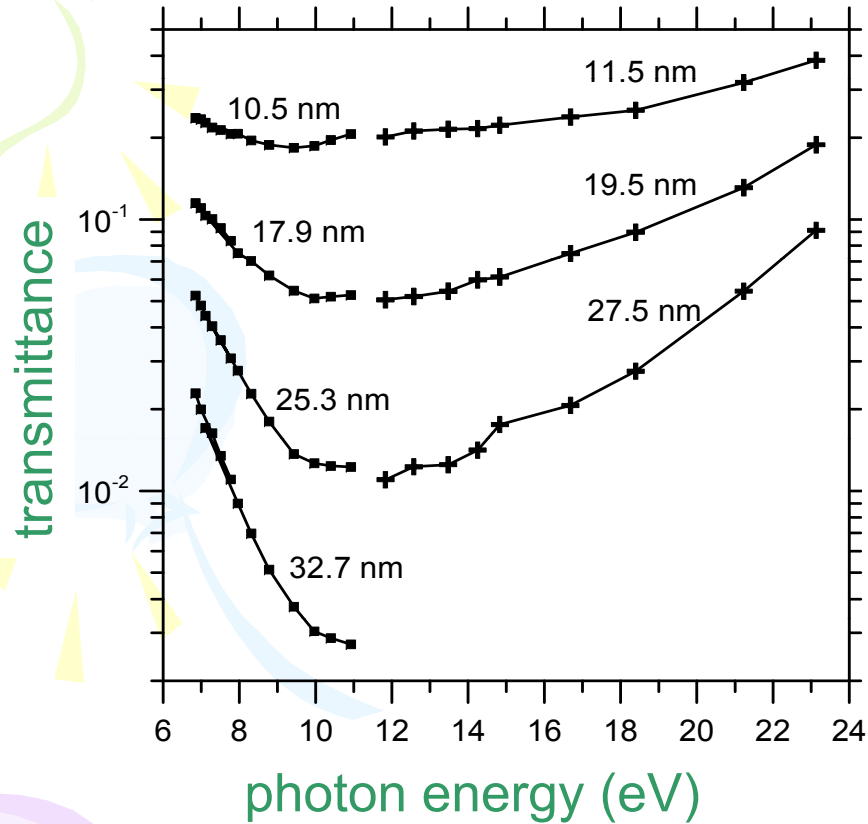
KK: n from k. Ca

L_{2,3} edge



n-k KK: B

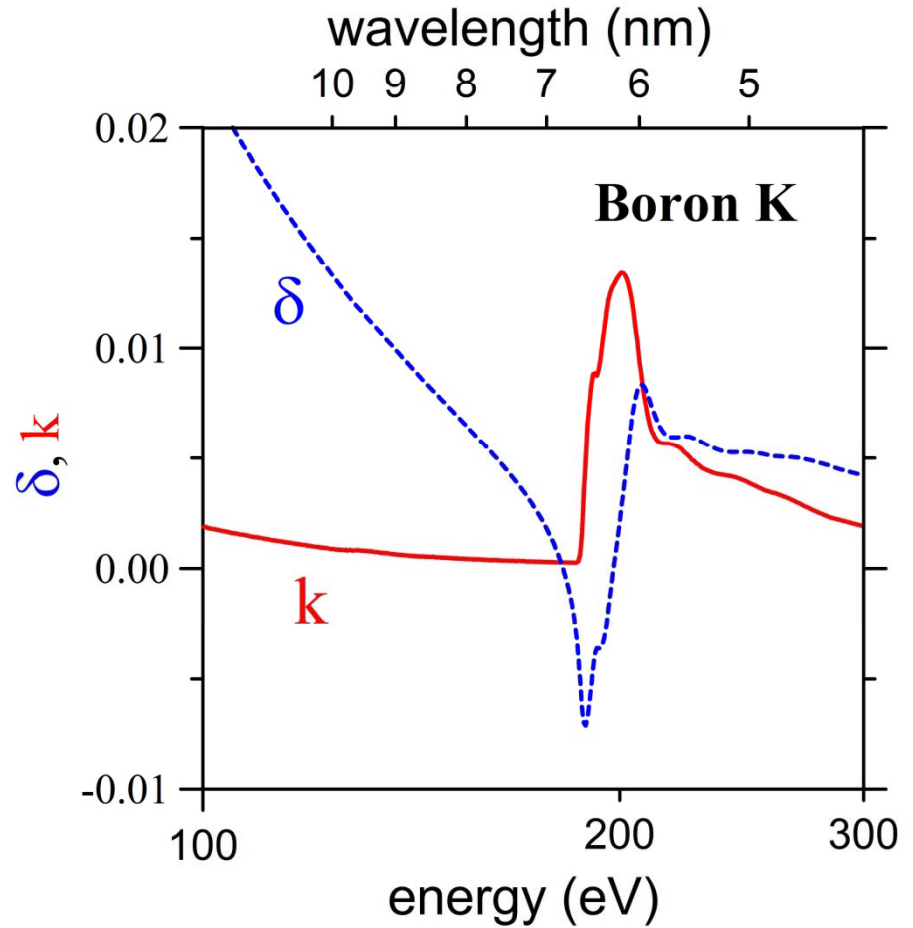
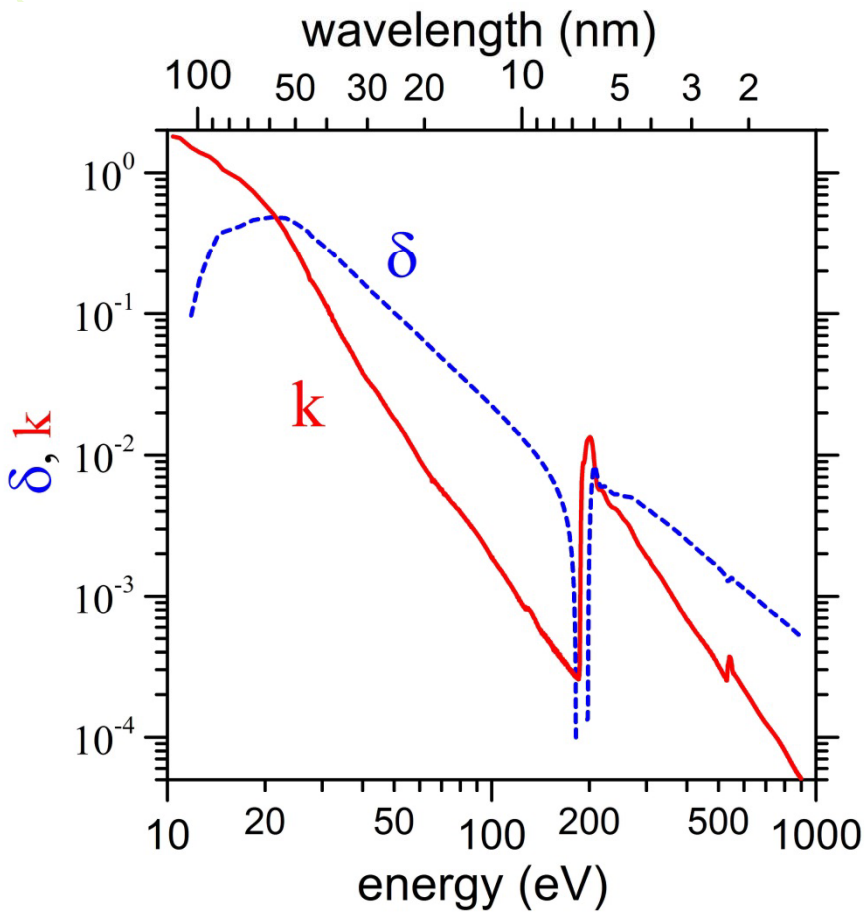
k from transmittance vs film thickness



substrate: C film
supported on grid

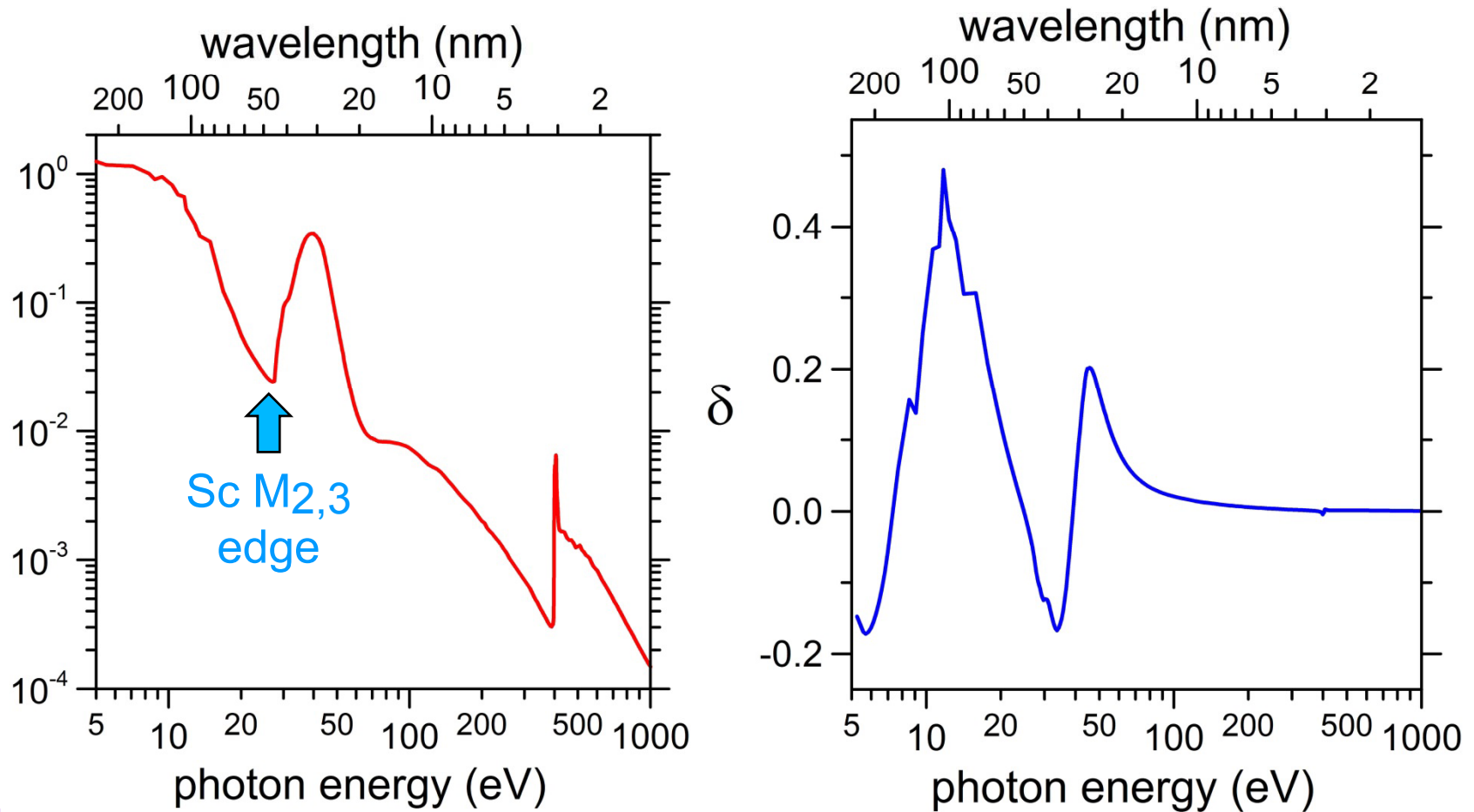
$$T = \exp\left(-\frac{4\pi kx}{\lambda}\right)$$

KK: n from k. B



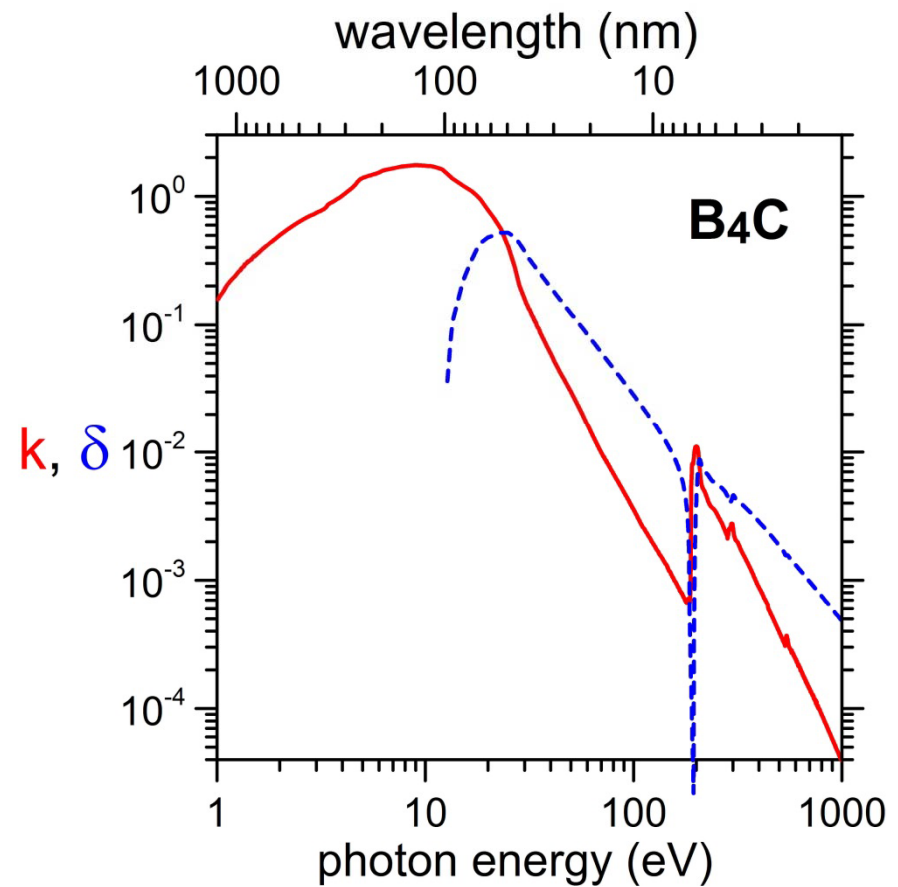
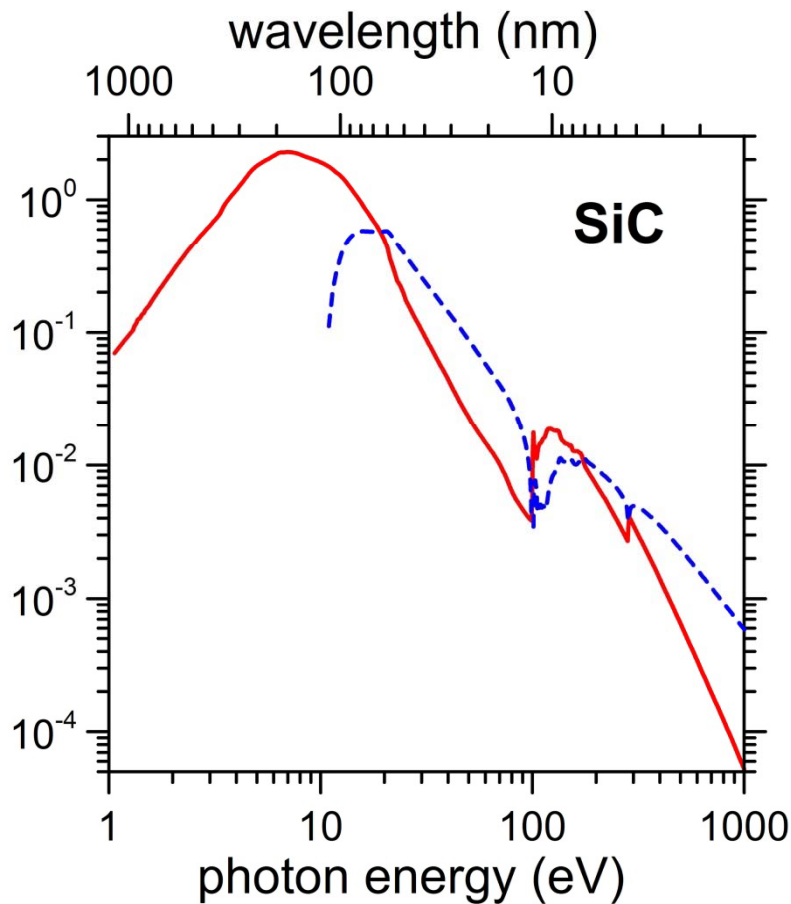
Fernández-Perea et al., J. Opt. Soc. Am. A. **24**, 3800 (2007)

KK: n from k. Sc



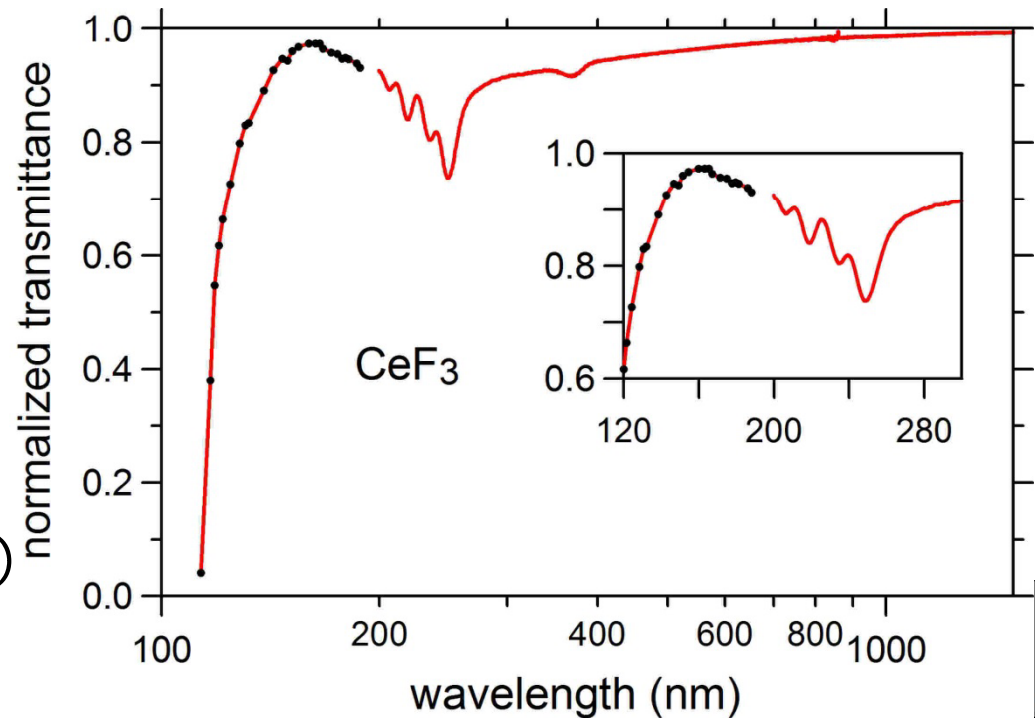
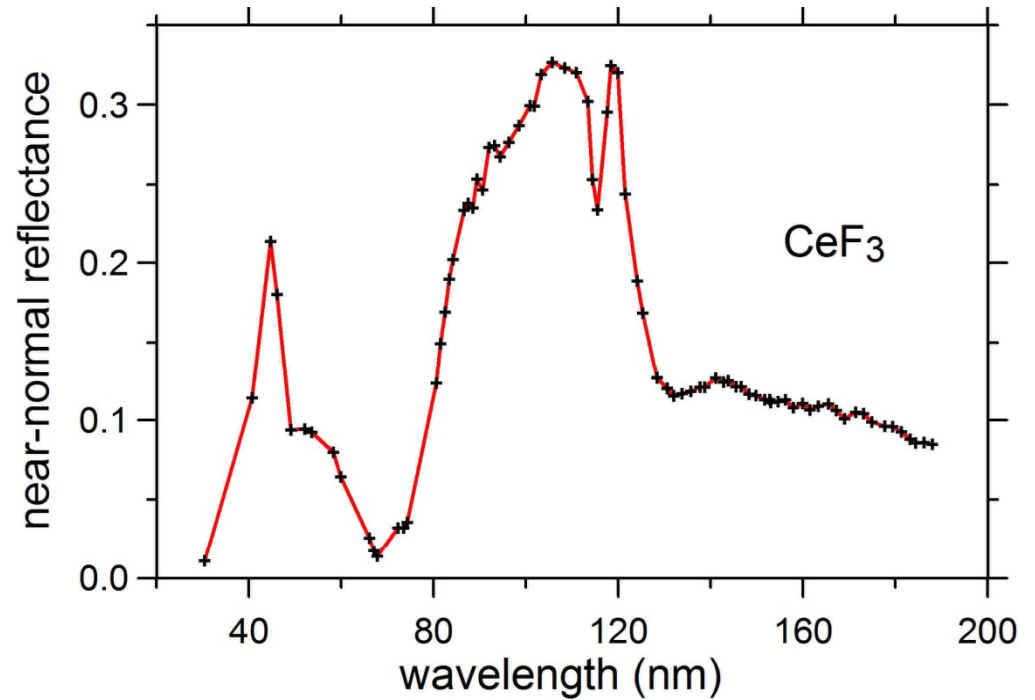
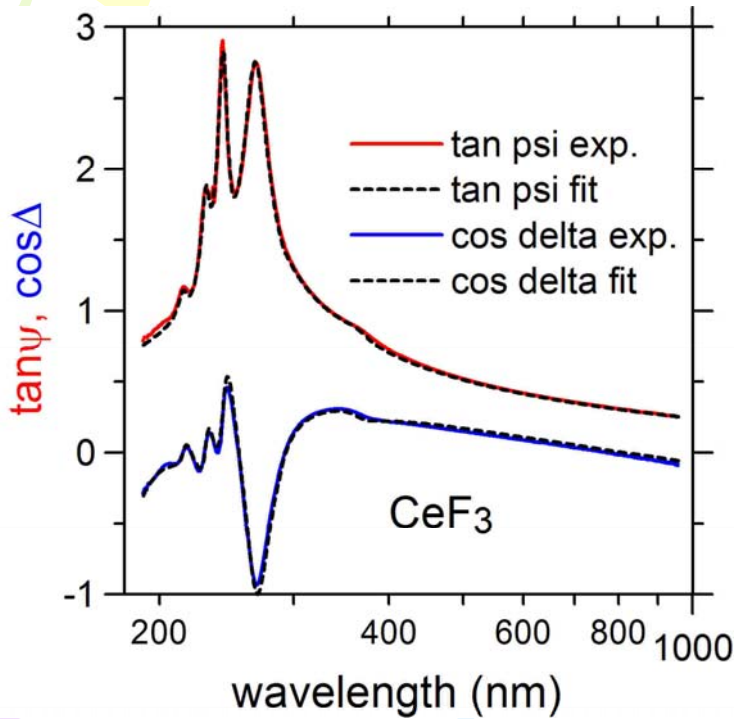
Fernández-Perea et al., J. Opt. Soc. Am. A. **23**, 2880 (2006)

KK: n from k. SiC and B₄C



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

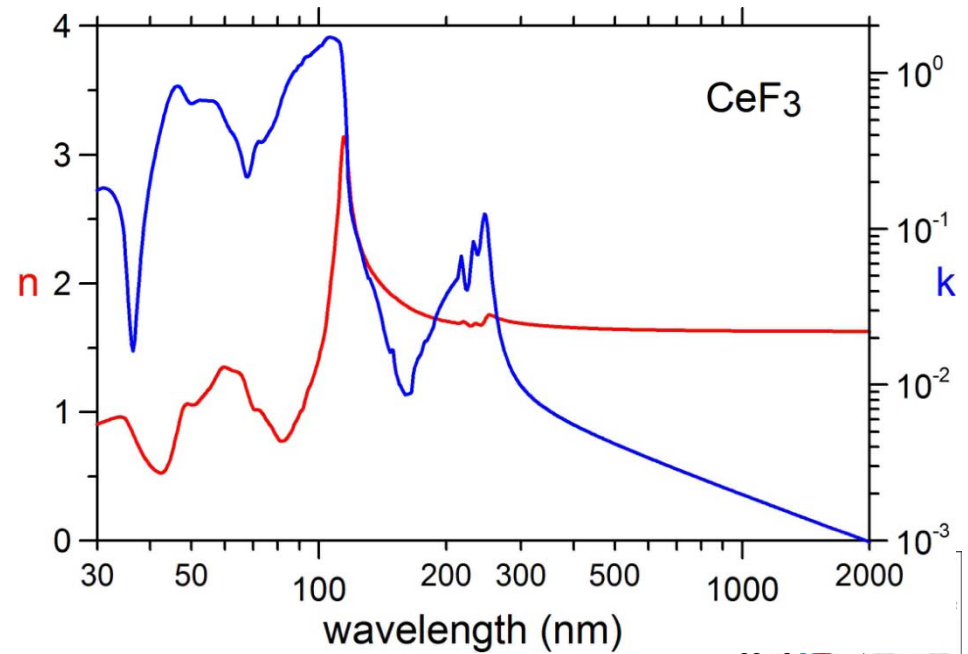
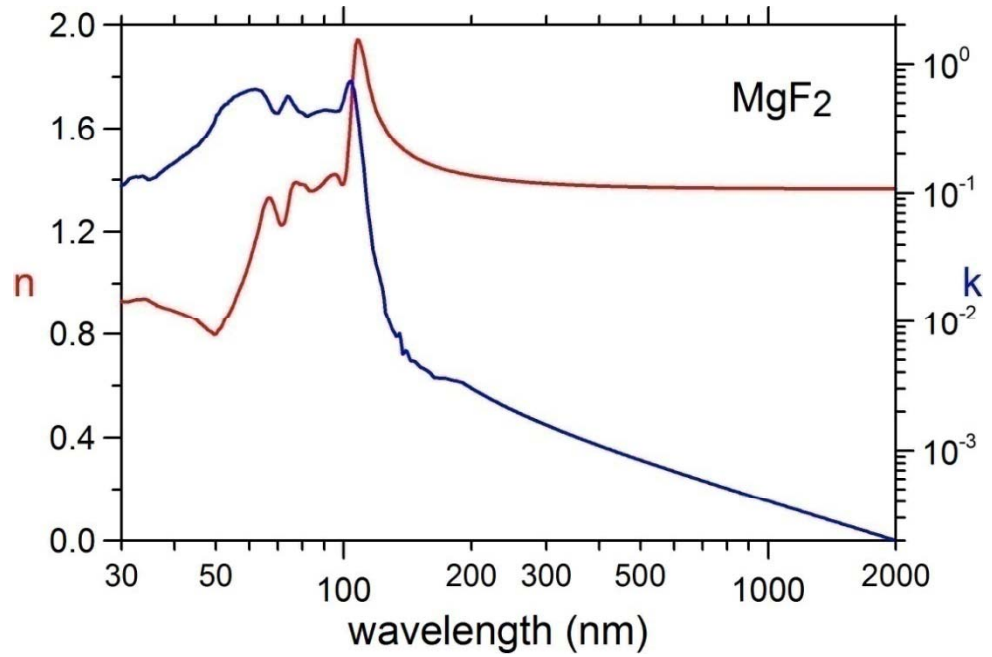
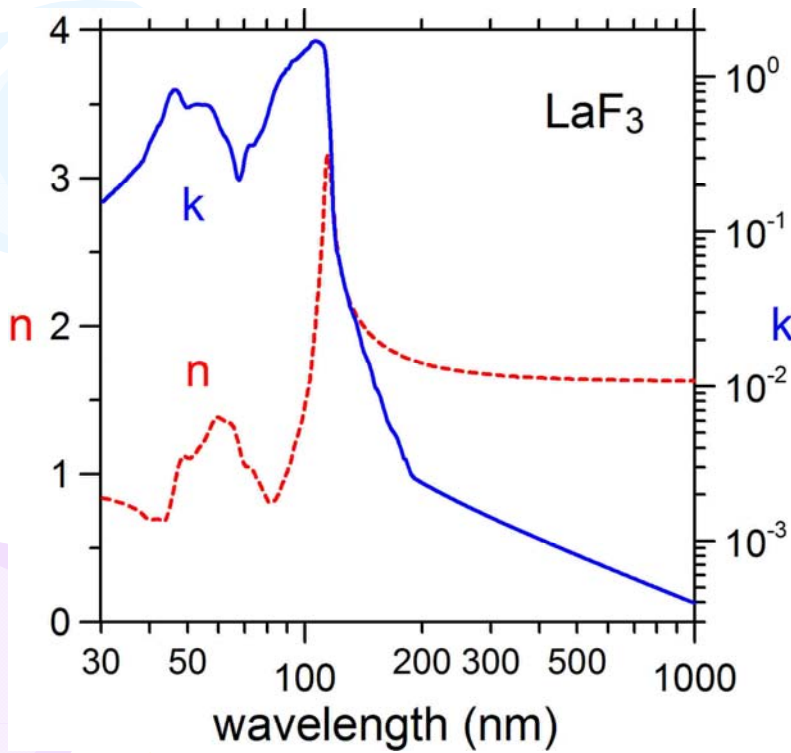
KK using
R+T+Ellips:
MgF2, LaF3, **CeF3**



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



KK using R+T+Ellips: MgF₂, LaF₃, CeF₃



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KK evaluation: global sum rules

f sum rule

$$\int_0^{\infty} E'' k(E) dE = \frac{\pi N_{ef} e^2 h^2}{4 \epsilon_0 m} \div Z$$

Z=atomic number

Inertial sum rule

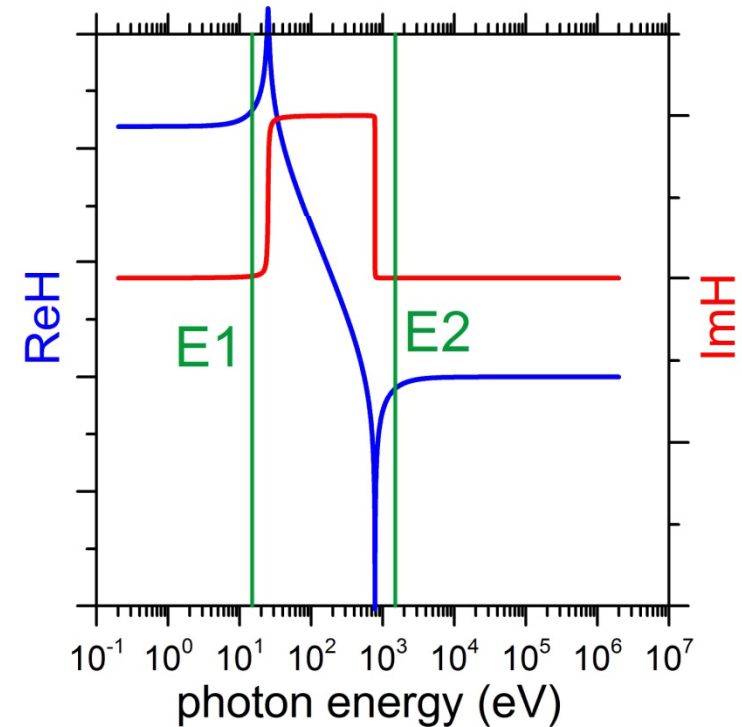
$$\int_0^{\infty} [n(E) - 1] dE = 0$$

Evaluation parameter:

$$\zeta = \frac{\int_0^{\infty} [n(E) - 1] dE}{\int_0^{\infty} |n(E) - 1| dE}$$
$$|\zeta| < 0.005$$

Sum rules with window functions

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



KK evaluation: sum rules

f sum rule

$$n_{eff} = \frac{4\epsilon_0 m}{Ne^2 \hbar^2} \int_0^\infty E' k(E') dE' \div Z$$

Z=atomic number

$$n_{eff} = \frac{4\epsilon_0 m}{Ne^2 \hbar^2} \left(E_1^2 - E_2^2 \right)$$

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

global
sum rules

Inertial sum rule

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

local
sum rules

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

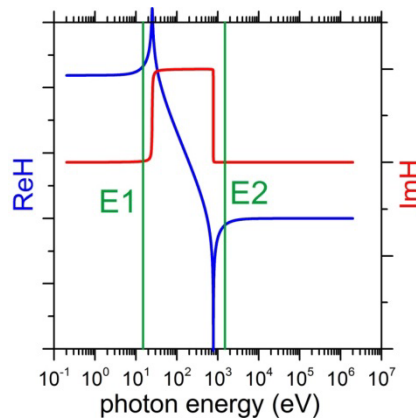
$$\int_0^\infty E'^{-1} \text{Im}\{H_2(E')[N(E')-1]\} dE' = 0$$

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

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

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

Window
function

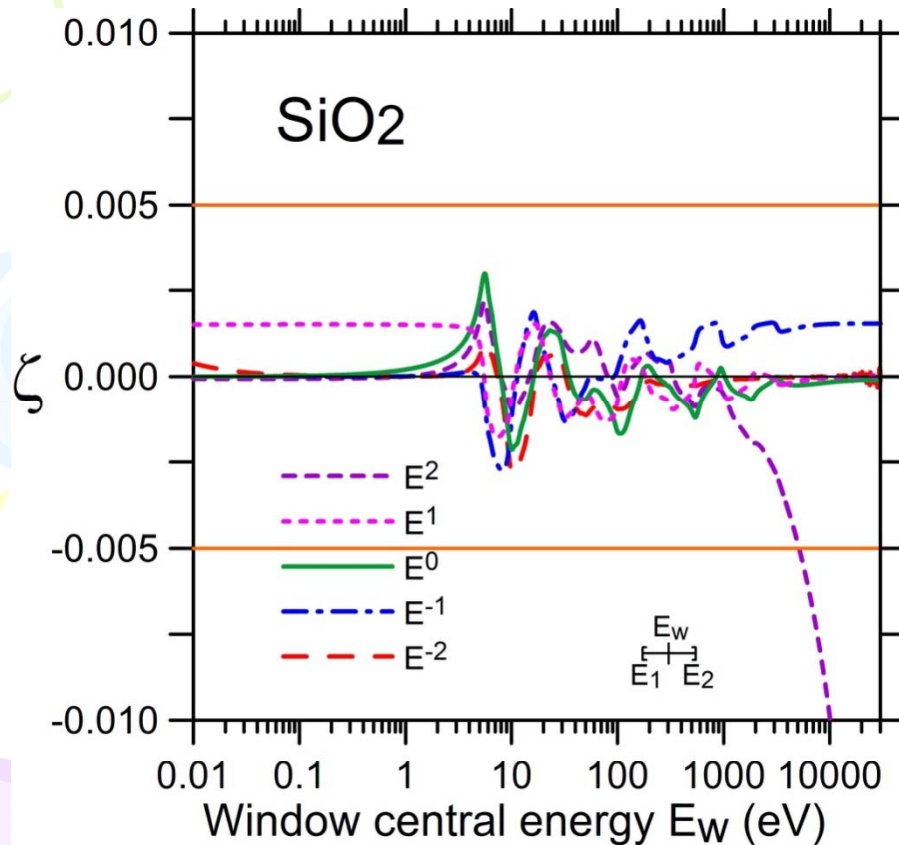


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J. Opt. **18**, 075606 (2016)



KK evaluation: sum rules with window functions



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

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

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

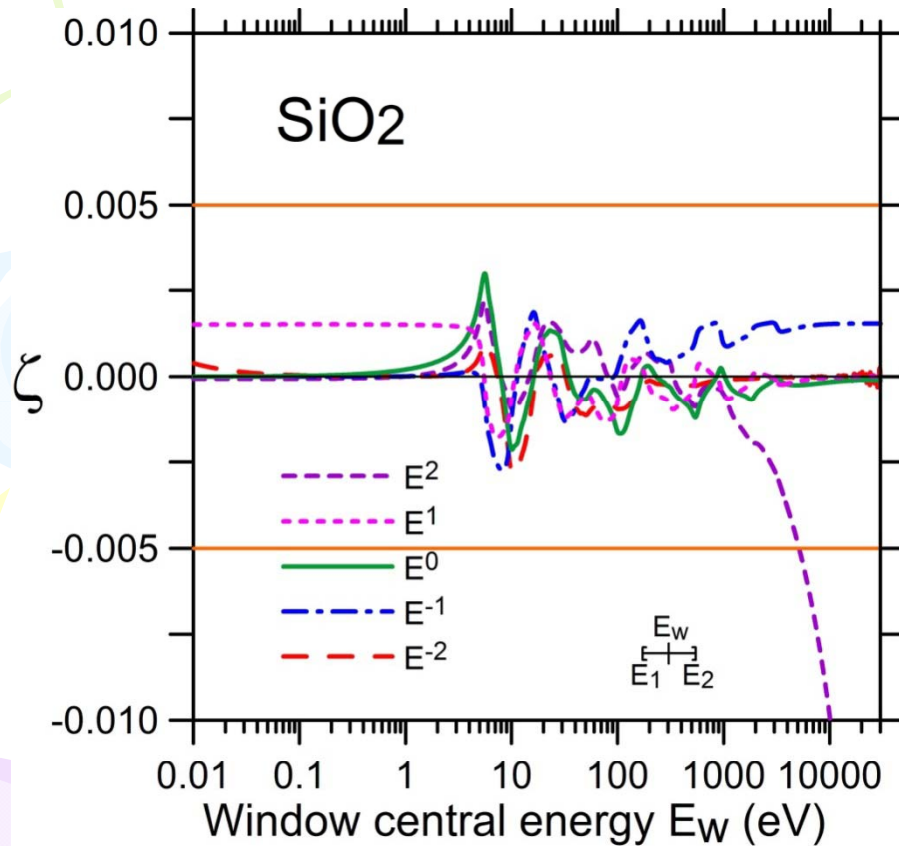
$$\int_0^{\infty} E'^{-1} \operatorname{Im}\{H_2(E')[N(E')-1]\}dE' = 0$$

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

Inertial sum rules

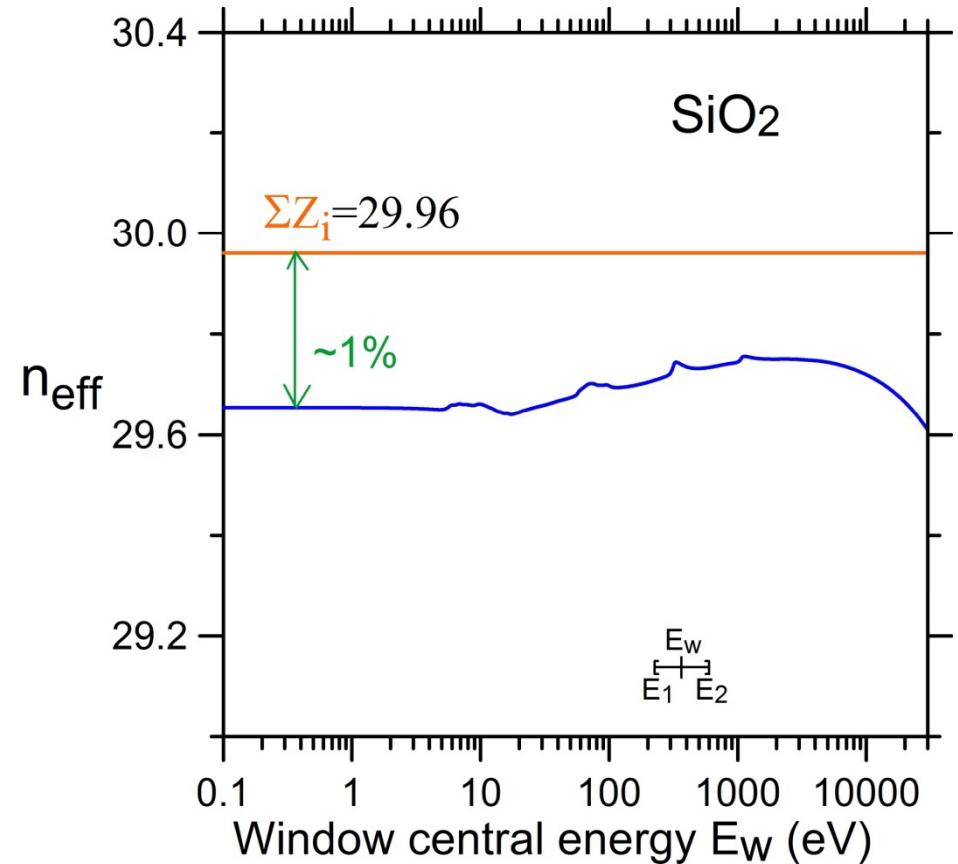
Rodríguez-de Marcos, Opt. Mater. Express **6**, 3622 (2016)

KK evaluation: sum rules with window functions



Inertial sum rules

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



f sum rule



Self-consistent characterization of materials

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
IA	IIA	IIIB	IVB	VB	VIB	VII B	VIII B	IX B	X B	IB	IIB	IIIA	IVA	VA	VIA	VIIA	VIIIA
1 H 1.01																	2 He 4.00
3 Li 6.94	4 Be 9.01											5 B 10.81	6 C 12.01	7 N 14.01	8 O 16.00	9 F 19.00	10 Ne 20.18
11 Na 22.99	12 Mg 24.31											13 Al 26.98	14 Si 28.09	15 P 30.97	16 S 32.07	17 Cl 35.45	18 Ar 39.95
19 K 39.1	20 Ca 40.08	21 Sc 44.96	22 Ti 47.88	23 V 50.94	24 Cr 52.00	25 Mn 54.94	26 Fe 55.85	27 Co 58.93	28 Ni 58.69	29 Cu 63.55	30 Zn 65.39	31 Ga 69.72	32 Ge 72.61	33 As 74.92	34 Se 78.96	35 Br 79.90	36 Kr 83.80
37 Rb 85.47	38 Sr 87.62	39 Y 88.91	40 Zr 91.22	41 Nb 92.91	42 Mo 95.94	43 Tc (98)	44 Ru 101.07	45 Rh 102.91	46 Pd 106.42	47 Ag 107.87	48 Cd 112.41	49 In 114.82	50 Sn 118.71	51 Sb 121.76	52 Te 127.6	53 I 126.9	54 Xe 131.29
55 Cs 132.9	56 Ba 137.3	57 La*	72 Hf 178.5	73 Ta 180.9	74 W 183.9	75 Re 186.2	76 Os 190.2	77 Ir 192.2	78 Pt 195.1	79 Au 197.0	80 Hg 200.6	81 Tl 204.4	82 Pb 207.2	83 Bi 209	84 Po (209)	85 At (210)	86 Rn (222)
87 Fr (223)	88 Ra (226)	89 Ac*	104 Db (262)	105 Sg (263)	106 Bh (264)	107 Hs (265)	108 Mt (268)	109 Ds (271)	110 Rg (272)								
		58 Ce 140.1	59 Pr 140.9	60 Nd 144.2	61 Pm (145)	62 Sm 150.4	63 Eu 152.0	64 Gd 157.3	65 Tb 158.9	66 Dy 162.5	67 Ho 164.9	68 Er 167.3	69 Tm 168.9	70 Yb 173.0	71 Lu 175.0		
		90 Th 232.0	91 Pa (231)	92 U 238.0	93 Np (237)	94 Pu (244)	95 Am (243)	96 Cm (247)	97 Bk (247)	98 Cf (251)	99 Es (252)	100 Fm (257)	101 Md (258)	102 No (259)	103 Lr (260)		

MgF₂
SrF₂
LaF₃
CeF₃
Ta₂O₅
B₄C
SiC
SiO₂
SiO

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 $\lambda > 120$ nm

Narrowband mirrors:


- Al/LiF/SiC: peak at 102 nm
- Fluorides peaked at $\lambda > 120$ nm

Polarizers:

- (Al/MgF₂)_n at $\lambda > 120$ 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?



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Collaborators in (n,k) determination:

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- 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