Application of Thomas–Reiche–Kuhn Sum Rule to the Parametrization of JDOS of Hydrogenated Amorphous Silicon

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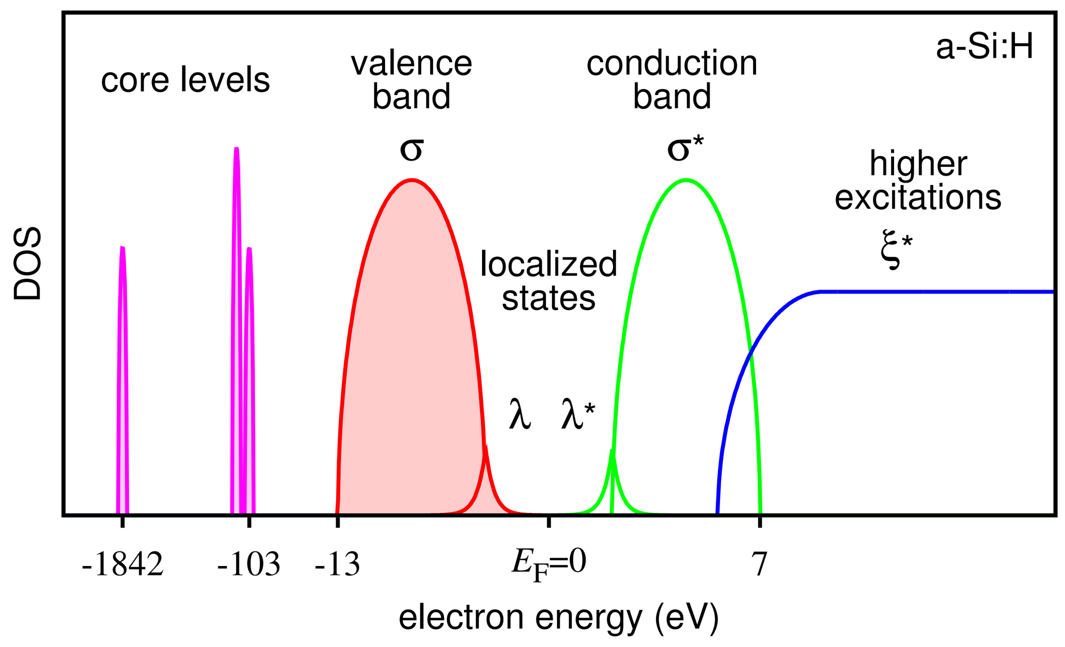
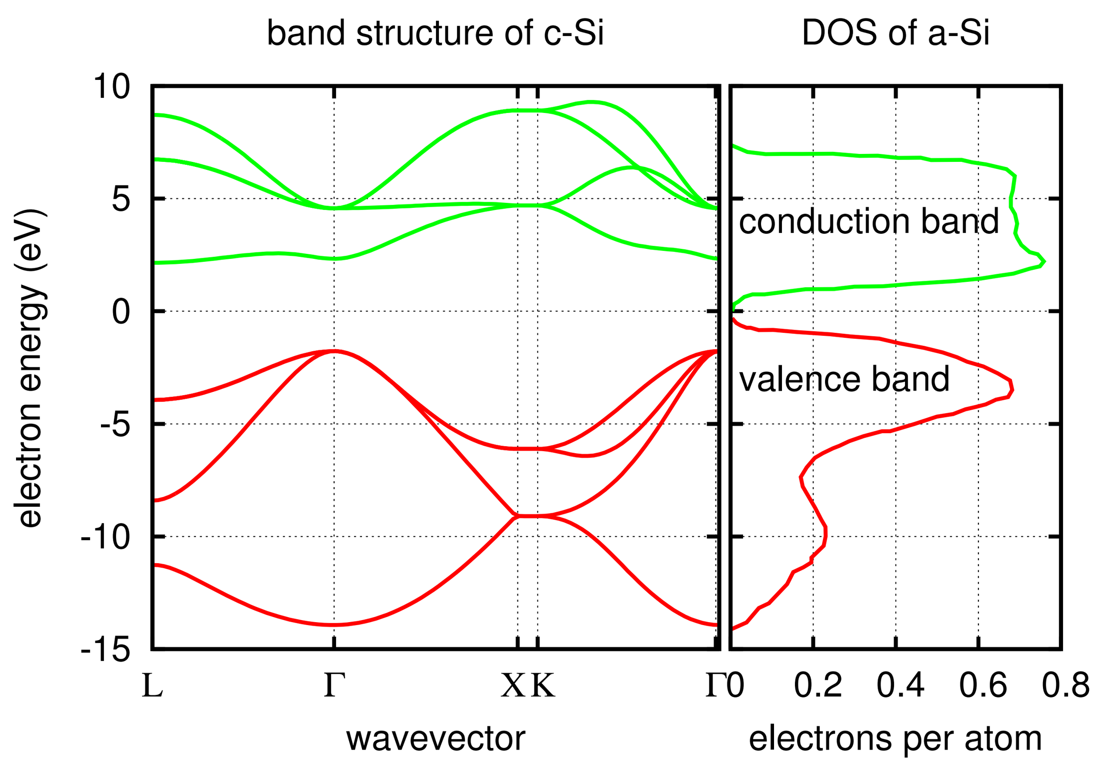
Electronic structure

	K	L	σ, λ, λ^*, σ^*	ξ^*
Si	1s	2s, 2p	3s, 3p	3d, 4s, 4p, …
H	–	–	1s	2s, 2p, 3s, …

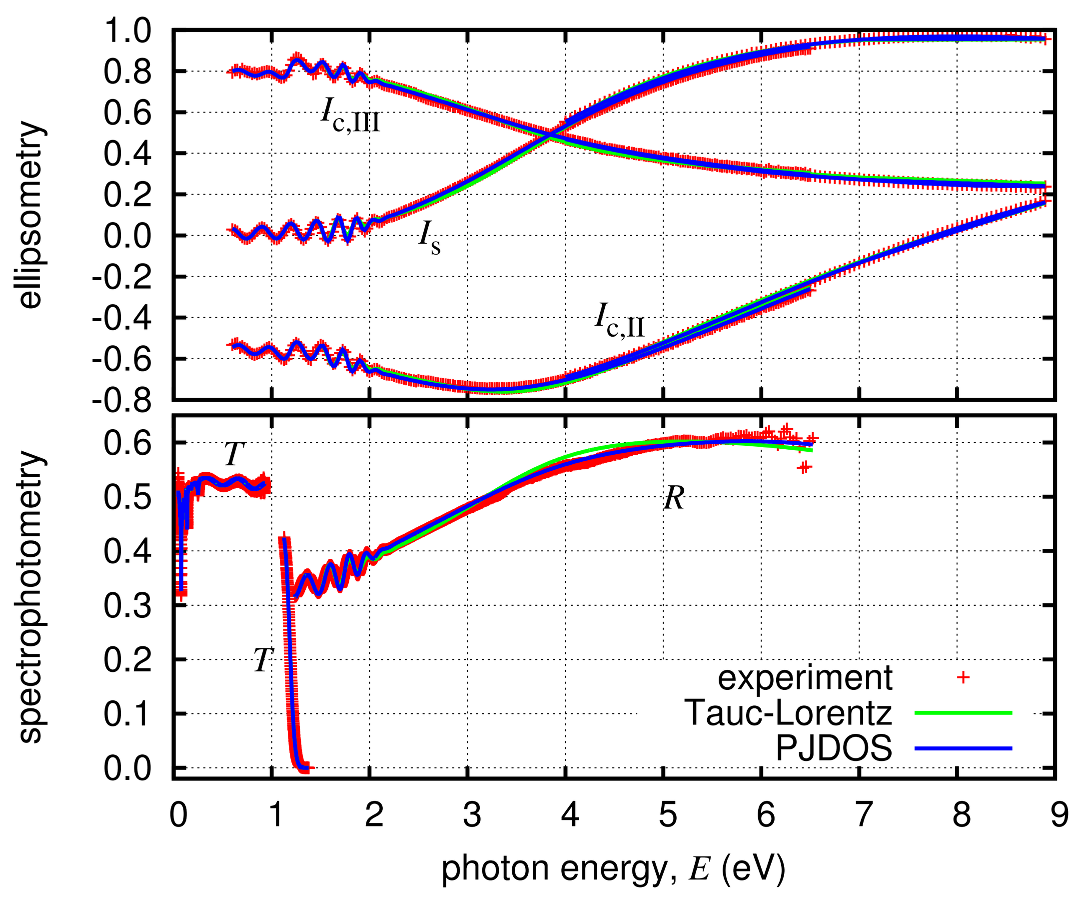


Schematic diagram of electronic structure of amorphous silicon.



Electronic structure of crystalline and amorphou silicon calculated using tight-binding method [1] and [2].

Experiment



Modeling

All individual contributions of the PJDOS model are modeled using analytical Kramers–Kronig consistent functions.

Interband transitions $\sigma \to \sigma^*$ (IBTL5)

Combination of parabolic bands with Lorentzian function.

$$\varepsilon_i(E) = \frac{J(E)}{E^2} = \begin{cases} N_{\sigma\sigma}\dfrac{(|E|-E_g)^2(|E|-E_h)^2}{CE[(E^2-E_c^2)^2+B_c^2E^2]} & \text{for } E_g < |E| < E_h \\ 0 & \text{otherwise,} \end{cases}$$

where C is normalization constant chosen that the following holds:

$$N_{\sigma\sigma} = \int_0^\infty E\,\varepsilon_i(E)\,dE$$

The real part is calculated from Kramers–Kronig integral:

$$\varepsilon_r(E) - 1 = \frac{2}{\pi}\int_0^\infty \frac{X\,\varepsilon_i(X)}{X^2-E^2}\,dX$$

It leads to cobinations of **logaritmic** and **rational** functions simillarly to the Tauc–Lorentz model (see poster devoted to DLC).

Excitations of electrons $\sigma \to \lambda^*$ and $\lambda \to \sigma^*$ – Urbach tail (UT4)

The absorption below the bad gap is modeled using exponential Urbach tail smootly extended by a second-order polynomial $P_2(E)$:

$$\varepsilon_i(E) = \frac{J(E)}{E^2} = \mathrm{sgn}(E)\times$$

$$\begin{cases} \dfrac{N_{\sigma\lambda}}{CE^2}\left[\exp\left(\dfrac{E-E_g}{E_u}\right) - \exp\left(-\dfrac{E_g}{2E_u}\right)\right] & \text{for } \dfrac{E_g}{2} < |E| < E_g \\ \dfrac{N_{\sigma\lambda}}{CE^2}\left[P_2(E) - \exp\left(-\dfrac{E_g}{2E_u}\right)\right] & \text{for } E_g \le |E| \le E_h \\ \dfrac{N_{\sigma\lambda}}{CE^2}\left[\exp\left(\dfrac{E_h-E}{E_u}\right) - \exp\left(-\dfrac{E_g}{2E_u}\right)\right] & \text{for } E_h < |E| < E_h + \dfrac{E_g}{2} \\ 0 & \text{otherwise.} \end{cases}$$

Integrations leads to **exponential integral** functions in $\varepsilon_r(E)$.

Excitations of electrons $\sigma \to \xi^*$ (HET2)

$$\varepsilon_i(E) = \frac{J(E)}{E^2} = \begin{cases} 0 & \text{for } |E| \le E_{g\xi} \\ N_{\sigma\xi}\dfrac{3E_{g\xi}(|E|-E_{g\xi})^2}{E^5} & \text{for } |E| > E_{g\xi} \end{cases}$$

$$\varepsilon_r(E) - 1 = N_{\sigma\xi}\frac{3E_{g\xi}}{\pi E^2}\left[a(E)\ln\left|1-\frac{E}{E_{g\xi}}\right| + b(E)\ln\left|1+\frac{E}{E_{g\xi}}\right| - c - \frac{d}{E^2}\right]$$

where

$$a(E) = -\frac{(E_{g\xi}-E)^2}{E^3}, \quad b(E) = \frac{(E_{g\xi}+E)^2}{E^3}, \quad c = \frac{2}{3E_{g\xi}}, \quad d = 2E_{g\xi}$$

Density of the valence electrons (optical electron density)

$$N_v = \int_0^\infty \sum_{j=\sigma,\lambda}\ \sum_{k=\lambda^*,\sigma^*,\xi^*} E\,\varepsilon_{i,j\to k}(E)\,dE \qquad (\mathrm{eV}^2)$$

$$N_v = N_{\sigma\sigma} + N_{\sigma\lambda} + N_{\sigma\xi}$$

Excitations of core electrons $\mathrm{K},\mathrm{L} \to \sigma^* + \xi^*$ (CEE2)

$$\varepsilon_i(E) = \frac{J(E)}{E^2} = \begin{cases} 0 & \text{for } |E| < E_k \\ N_k\dfrac{E_k}{E^3} & \text{for } |E| \ge E_k \end{cases}$$

$$\varepsilon_r(E) - 1 = \frac{N_k}{\pi E^3}\left[E_k \ln\left|\frac{E_k+E}{E_k-E}\right| - 2E\right]$$

Density of the K and L core electrons

$$N_K = 2N_v\frac{1-C_H}{4-3C_H} \quad \text{and} \quad N_L = 8N_v\frac{1-C_H}{4-3C_H}$$

where C_H is atomic concentration of hydrogen (for our sample $C_H = 0.11$)

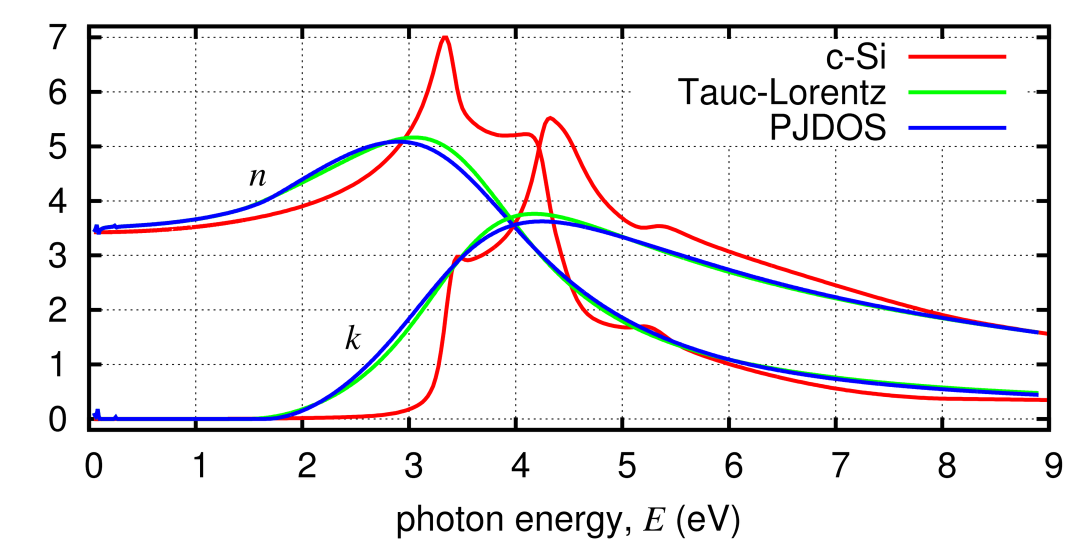
Phonon absorption peaks (GP3)

Modeled by Gausian peaks leading to **Dawson integrals** in $\varepsilon_r(E)$.

Tauc-Lorentz model

For renormalized Tauc-Lorentz model see poster devoted to DLC.

Results



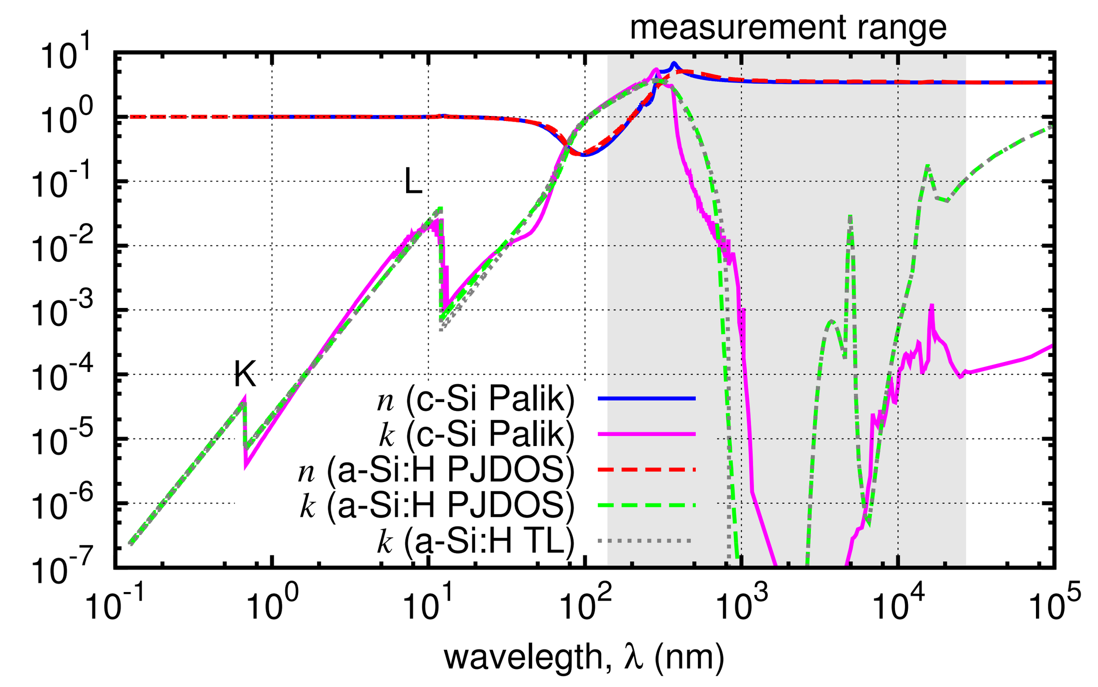
Optical constants of a-Si:H films determined using different models.

model	$N_{\sigma\sigma}$ (eV^2)	E_g (eV)	E_c (eV)	E_h (eV)	B_c (eV)	$N_{\sigma\xi}$ (eV^2)	$E_{g\xi}$ (eV)
Tauc–Lorentz	421.4	1.486	3.621	–	1.929	–	–
PJDOS	316.0	1.669	3.587	20*	2.355	102.6	5.878

model	$N_{\sigma\lambda}$ (eV^2)	E_u (eV)	E_K (eV)	E_L (eV)	C_H	χ
Tauc–Lorentz	0	–	1.842*	103*	0.11*	2.38
PJDOS	6.2	0.036	1.842*	103*	0.11*	1.38

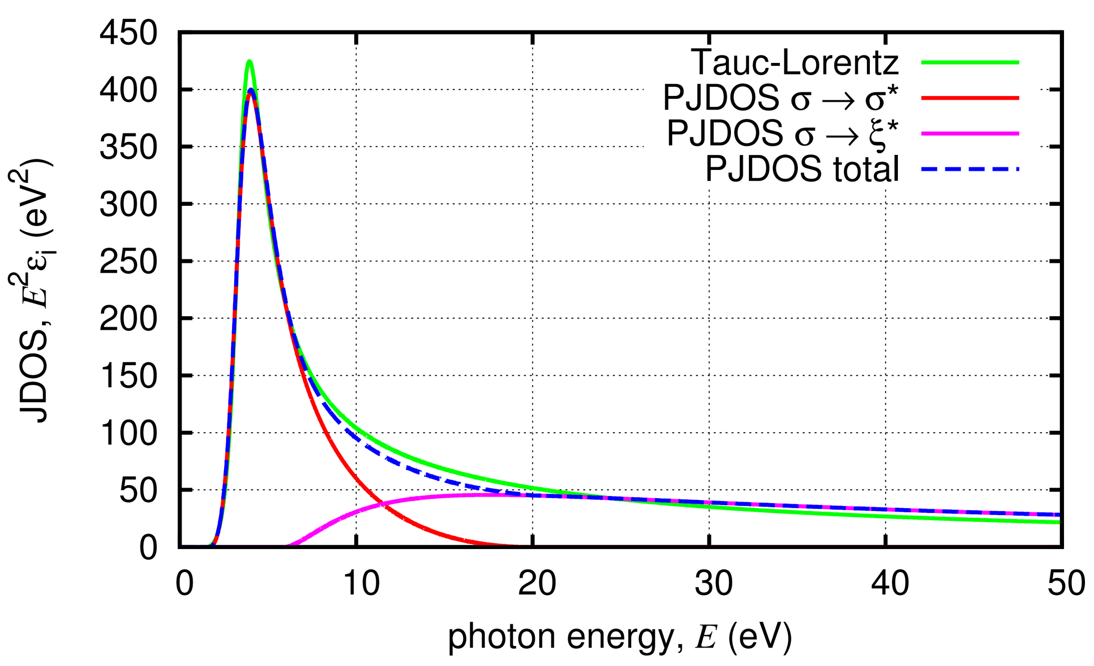
* fixed parameter
Quantity χ characterizes the disagreement between theoretical and experimental data (1 is optimum)

Comparison with c-Si



Log–log plot of optical constants of crystalline and amorphous silicon.

JDOS function of a-Si:H



Comparison of joint density of states determined using different models.

Total optical density of the electrons

$$N_e = N_v + N_K + N_L = N_v\frac{14-13C_H}{4-3C_H} \qquad (\mathrm{eV}^2)$$

Relation to plasma frequency ω_p

$$\text{for } E \to \infty, \quad \varepsilon_r(E) \approx 1 - \frac{2N_e}{\pi E^2} = 1 - \frac{(\hbar\omega_p)^2}{E^2} = 1 - \frac{\omega_p^2}{\omega^2}$$

$$N_e = \frac{\pi}{2}(\hbar\omega_p)^2$$

Optical density of atoms

$$N_a = \frac{N_e}{14-13C_H} = \frac{N_v}{4-3C_H} \qquad (\mathrm{eV}^2)$$

Real electron density

$$\mathcal{N}_e = 4.617\cdot 10^{26} N_e \qquad (1/\mathrm{m}^3)$$

Mass density of the a-Si:H films

$$\varrho = \mathcal{N}_a\Big[A_{Si}(1-C_H) + A_H C_H\Big]\,u \qquad (\mathrm{kg/m}^3)$$

$$\mathcal{N}_a = 4.617\cdot 10^{26} N_a$$

- $\mathcal{N}_a$ density of atoms (1/m^3)
- A_{Si} silicon atomic weight (28.09 g/mol)
- A_H hydrogen atomic weight (1.008 g/mol)
- u atomic mass unit ($1.6605\cdot 10^{-27}$ kg)

model	N_v (eV^2)	N_e (eV^2)	N_a (eV^2)	$\mathcal{N}_e$ (1/m^3)	$\mathcal{N}_a$ (1/m^3)	ϱ (kg/m^3)
Tauc–Lorentz	421.4	1443	114.8	$6.664\cdot 10^{29}$	$5.302\cdot 10^{28}$	2211
PJDOS	424.8	1455	115.7	$6.718\cdot 10^{29}$	$5.344\cdot 10^{28}$	2228

Compare with density of the c-Si: $\varrho = 2329\,\mathrm{kg/m}^3$

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References

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[2] H. C. Kang, J. Non-Cryst. Solids 261 (2000) 169–180.