Technical Note

Lorentz Dispersion Model

Spectroscopic ellipsometry (SE) is a technique based on the measurement of the relative phase change of reflected and polarized light in order to characterize thin film optical functions and other properties. The measured data are used to describe a model where each layer refers to a given material. The model uses mathematical relations called dispersion formulae that help to evaluate the thickness and optical properties of the material by adjusting specific fit parameters.
This application note deals with the Lorentzian dispersion formula.

Note that the technical notes «Classical dispersion model» and «Drude dispersion model» are complementary to this one.

Theoretical model

The Lorentz classical theory (1878) is based on the classical theory of interaction between light and matter and is used to describe frequency dependent polarization due to bound charge. The bindings between electrons and nucleus are supposed to be similar to the that of a mass-spring system.

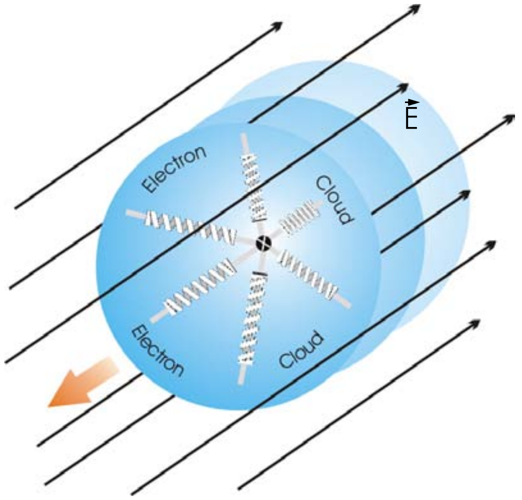


Fig. 1 Restoring force between the orbiting electron and the atomic centre (Ref. 4).

Electrons react to an electromagnetic field by vibrating like damped harmonic oscillators. The way the dipole replies to a submitted electric field is given by the following equation of motion of a bound electron:

$$m\cdot\frac{d^2\vec{r}}{dt^2}+m\cdot\Gamma_0\cdot\frac{d\vec{r}}{dt}+m\cdot\omega_t^2\cdot\vec{r}=-e\cdot\vec{E}_{loc}\qquad(1)$$

where:

- $m\, d^2r/dt^2$ is the acceleration force;
- $m\Gamma_0\, dr/dt$ is the viscous force; Γ_0 is the damping factor;
- $m\omega_t^2 r$ is the Hooke's force; m is the electronic mass and ω_t is the resonant frequency of the oscillator;
- $-e\,\mathbf{E}_{loc}$ is the local electric field driving force; e is the magnitude of the electronic charge and $\mathbf{E}_{loc}$ is the local electric field acting on the electron. The assumption is made that the macroscopic and local electric fields are equal and vary in time as $e^{i\omega t}$.

The solution to the previous equation yields the expression for the amplitude of oscillation **r** depending on the photon energy ω:

$$\vec{r}(\omega)=\frac{1}{m}\cdot\frac{-e\cdot\vec{E}_{loc}}{\left(\omega_t^2-\omega^2\right)+i\cdot\Gamma_0\cdot\omega}\qquad(2)$$

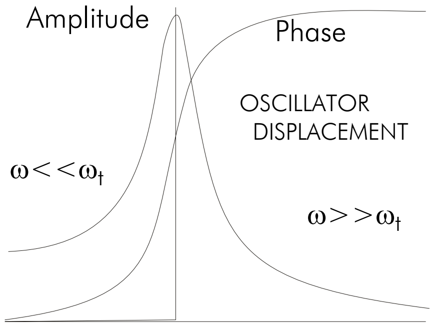


Fig. 2 The oscillator amplitude as a function of frequency (Ref. 5).

At low frequencies $\omega<<\omega_t$, the amplitude **r** has a medium finite value and is in phase with **E**.
At the resonance frequency $\omega\cong\omega_t$ the amplitude is imaginary and maximum when denominator is minimum. More, at $\omega\cong\omega_t$ there is a 90° phase shift between **E** and **r**.
At high frequencies $\omega>>\omega_t$, the amplitude **r** vanishes.

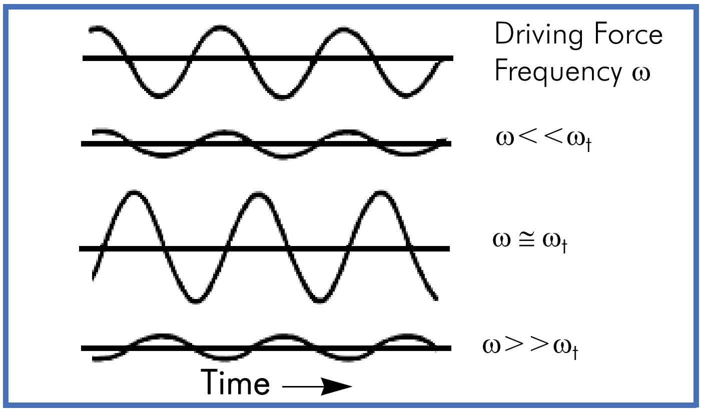


Fig. 3 The response of an oscillator amplitude to a periodic driving force depends on the resonance frequency (Ref 5).

The induced dipole moment μ is related to **r** through this relation:

$$\vec{\mu}(\omega)=-e\cdot\vec{r}(\omega)\Rightarrow\vec{\mu}(\omega)=\frac{e^2\cdot\vec{E}_{loc}}{m\cdot\left[\left(\omega_t^2-\omega^2\right)+i\cdot\Gamma_0\cdot\omega\right]}\quad(3)$$

The polarizability α(ω) is given by $\mu(\omega)=\alpha(\omega)E(\omega)$ where:

$$\alpha(\omega)=\frac{e^2}{m}\cdot\frac{1}{\left(\omega_t^2-\omega^2\right)+i\cdot\Gamma_0\cdot\omega}\quad(4)$$

Taking the sum of the single atom dipole moment over all atoms in a volume, it comes that the polarization per unit volume is given by:

$$P(\omega)=N\cdot\alpha(\omega)\cdot E(\omega)=\varepsilon_0\cdot\chi(\omega)\cdot E(\omega)\quad(5)$$

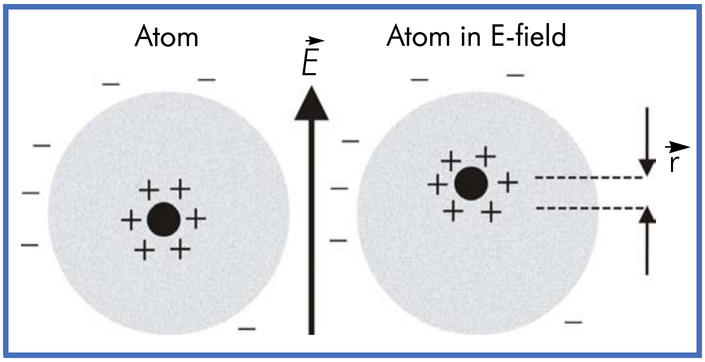


Fig. 4 Polarization of the electronic cloud due to external E-field. (Hecht, Ref. 3)

The susceptibility χ(ω) is deduced from the previous equation:

$$\chi(\omega)=\left(\frac{N\cdot e^2}{\varepsilon_0\cdot m}\right)\cdot\frac{1}{\omega_t^2-\omega^2+i\cdot\Gamma_0\cdot\omega}\quad(6)$$

where the prefactor ($Ne^2/\varepsilon_0 m$) is the plasma frequency squared ω_p^2.
The dielectric function is then given through this relation

$$\tilde{\varepsilon}(\omega)=1+\chi(\omega)=1+\frac{\omega_p^2}{\omega_t^2-\omega^2+i\Gamma_0\,\omega}\quad(7)$$

The limits ε_s and ε_∞ of the dielectric function respectively at low and high frequencies are given by:

$$\begin{cases}\varepsilon_s=\tilde{\varepsilon}(\omega\to 0)=1+\dfrac{\omega_p^2}{\omega_t^2}\\ \varepsilon_\infty=\tilde{\varepsilon}(\omega\to\infty)=1\end{cases}\quad(8)$$

The complex dielectric function can also be expressed in terms of the constants ε_s and ε_∞ by substituting equations (8) into (7) which yields the following equation:

$$\boxed{\tilde{\varepsilon}(\omega)=\varepsilon_\infty+\frac{(\varepsilon_s-\varepsilon_\infty)\cdot\omega_t^2}{\omega_t^2-\omega^2+i\cdot\Gamma_0\cdot\omega}\quad(9)}$$

where ε_s is defined as: $\varepsilon_s=\varepsilon_\infty+\dfrac{\omega_p^2}{\omega_t^2}$

Lorentz model describes radiation absorption due to inter-band transitions (quantum-mechanical interpretation). Interband transitions are transitions for which the electron moves to a final state corresponding to a different band without changing its **k**-vector in Brillouin's first zone.

Extension to multiple oscillators

If there is more than one oscillator, the dielectric function is assumed to be equal to the sum of contributions from individual oscillators. This situation fits better to the case of real materials.
In DeltaPsi2 software, the mathematical formulation used for a collection of:

- three Lorentz oscillators is:

$$\boxed{\tilde{\varepsilon}(\omega)=\varepsilon_\infty+\frac{(\varepsilon_s-\varepsilon_\infty)\cdot\omega_t^2}{\omega_t^2-\omega^2+i\cdot\Gamma_0\cdot\omega}+\sum_{j=1}^{2}\frac{f_j\cdot\omega_{0j}^2}{\omega_{oj}^2-\omega^2+i\cdot\gamma_j\cdot\omega}}$$

- N (N ≥ 1) Lorentz oscillators is:

$$\boxed{\tilde{\varepsilon}(\omega)=\varepsilon_\infty+\sum_{j=1}^{N}\frac{f_j\cdot\omega_{0j}^2}{\omega_{oj}^2-\omega^2+i\cdot\gamma_j\cdot\omega}}$$

Increasing the number of oscillators leads to a shift of the peaks of absorption toward the ultraviolet region.

The parameters of the equations

4 parameters may be used in the expressions of the single Lorentz oscillator but it may be possible to characterize the function with fewer coefficients.

Parameters describing the real part of the dielectric function

- The constant ε_∞ is the high frequency dielectric constant; it takes into account the contribution of high energy inter-band transition. Generally, $\varepsilon_\infty=1$ but can be greater than 1 if oscillators in higher energies exist and are not taken into account.
- The constant ε_s ($\varepsilon_s>\varepsilon_\infty$) gives the value of the static dielectric function at a zero frequency. The difference ε_s-ε_∞ represents the strength of the single Lorentz oscillator. The larger it is then the smaller the width Γ_0 of the peak of the single Lorentz oscillator.

Parameters describing the imaginary part of the single Lorentz oscillator dielectric function

- ω_t (in eV) is the resonant frequency of the oscillator whose energy corresponds to the absorption peak. When ω_t increases then the peak is shifted to higher photon energies. Generally, $1 \leq \omega_t \leq 20$.
- Γ_0 (in eV) is the broadening of each oscillator also known as the damping factor. The damping effect is due to the absorption process involving transitions between two states. On a graphic representing $\varepsilon_i(\omega)$, Γ_0 is generally equal to the Full Width At Half Maximum (FWHM) of the peak. As Γ_0 increases the width of the peak increases, but its amplitude decreases. Generally, $0 \leq \Gamma_0 \leq 10$.

Parameters describing the imaginary part of the multiple Lorentz oscillators dielectric function.

- f_j (j = 1, 2 … N) term is the oscillator strength present in the expression of the multiple Lorentz oscillator. As f_j increases then the peak amplitude increases, but the width of the peak γ_j decreases. Generally, $0 \leq f_j \leq 10$.
- ω_{0j} (in eV) (j = 1, 2 … N) is the resonant (peak) energy of an oscillator for a collection of several Lorentzian oscillators. It is similar to ω_t. Generally, $1 \leq \omega_{0j} \leq 8$.
- γ_j (in eV) (j = 1, 2 … N) parameter is the broadening parameter corresponding to the peak energy of each oscillator. It behaves like Γ_0. Generally, $0 \leq \gamma_j \leq 10$.

Limitation of the model

The Lorentz oscillator is not suitable for describing the properties (presence of gap energy and quantum effects) of real absorbing (amorphous, semiconductors) materials.

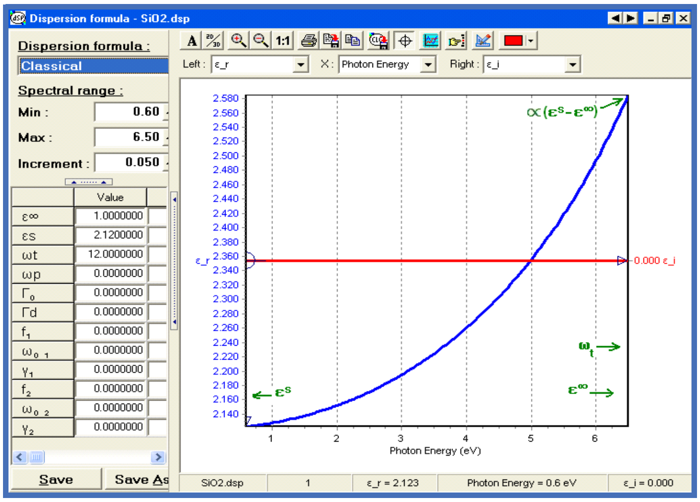
Parameter set up

Note that:

1. The Lorentzian dielectric function is available in the Classical dispersion formula in the DeltaPsi2 software.
2. The sign « $\propto$ » before a given parameter means that either the amplitude or the broadening of the peak is linked to that parameter.
3. For each multiple oscillator the graphs show the different contributions (in red dashed lines) of the N Lorentzian oscillators to the imaginary part of the Lorentz dielectric function (in red bold line).

> Transparent Lorentz function

- This function exhibits no absorption: $\Gamma_0=0$.
- This case corresponds to normal dispersion where $\varepsilon_r(\omega)$ increases with photon energy.



Dispersion formula : Classical

Spectral range :
Min : 0.60
Max : 6.50
Increment : 0.050

Left : ε_r
X : Photon Energy
Right : ε_i

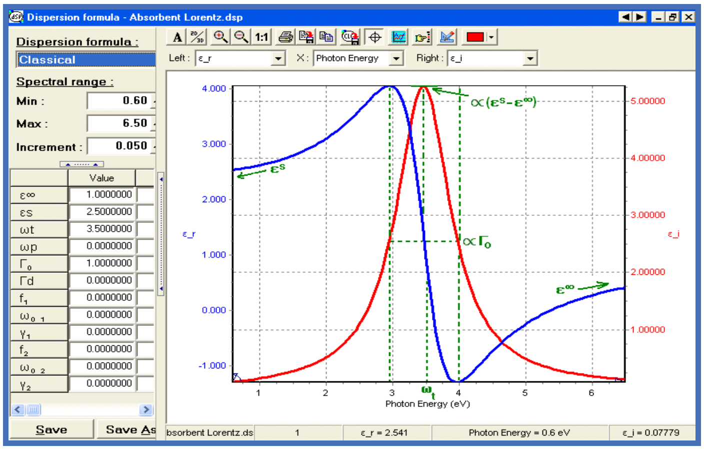
	Value
$\varepsilon\infty$	1.0000000
εs	2.1200000
ωt	12.0000000
ωp	0.0000000
Γ_0	0.0000000
Γd	0.0000000
f_1	0.0000000
$\omega_{0\ 1}$	0.0000000
γ_1	0.0000000
f_2	0.0000000
$\omega_{0\ 2}$	0.0000000
γ_2	0.0000000

SiO2.dsp | 1 | ε_r = 2.123 | Photon Energy = 0.6 eV | ε_i = 0.000

Dielectric function of SiO_2

> Absorbing Lorentz function

- This function exhibits absorption: $\Gamma_0 \neq 0$.
- The real part of the dielectric function increases with increasing frequency (normal dispersion) except for a region between [3eV - 4eV] where the dispersion becomes anomalous. The absorption peak is given by the imaginary part of the dielectric function ε_i (ω) and is always positive.



Dispersion formula : Classical

Spectral range :
Min : 0.60
Max : 6.50
Increment : 0.050

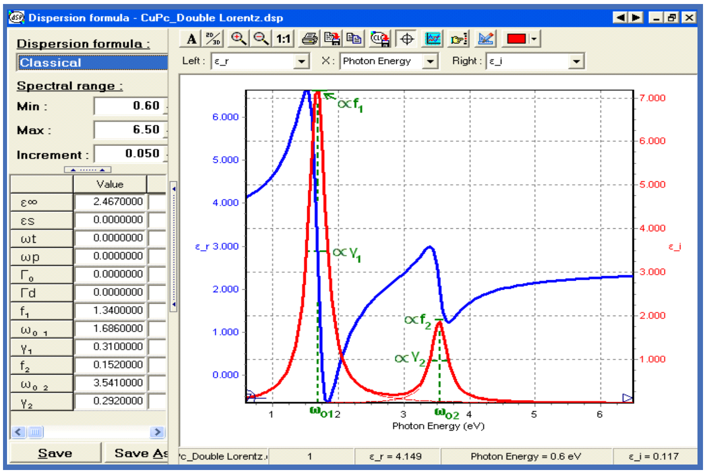
	Value
ε∞	1.0000000
εs	2.5000000
ωt	3.5000000
ωp	0.0000000
Γ_o	1.0000000
Γd	0.0000000
f_1	0.0000000
$\omega_{o\ 1}$	0.0000000
γ_1	0.0000000
f_2	0.0000000
$\omega_{o\ 2}$	0.0000000
γ_2	0.0000000

Left : ε_r X : Photon Energy Right : ε_i

bsorbent Lorentz.ds | 1 | ε_r = 2.541 | Photon Energy = 0.6 eV | ε_i = 0.07779

Representation of a Lorentz absorbing function

> Multiple oscillator Lorentz function



Dispersion formula : Classical

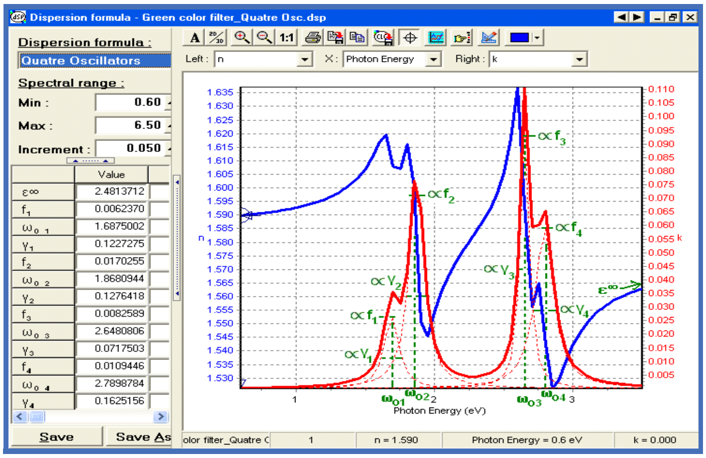
Spectral range :
Min : 0.60
Max : 6.50
Increment : 0.050

	Value
ε∞	2.4670000
εs	0.0000000
ωt	0.0000000
ωp	0.0000000
Γ_o	0.0000000
Γd	0.0000000
f_1	1.3400000
$\omega_{o\ 1}$	1.6860000
γ_1	0.3100000
f_2	0.1520000
$\omega_{o\ 2}$	3.5410000
γ_2	0.2920000

Left : ε_r X : Photon Energy Right : ε_i

'c_Double Lorentz.: | 1 | ε_r = 4.149 | Photon Energy = 0.6 eV | ε_i = 0.117

Dielectric function of CuPc described by 2 oscillators



Dispersion formula : Quatre Oscillators

Spectral range :
Min : 0.60
Max : 6.50
Increment : 0.050

	Value
ε∞	2.4813712
f_1	0.0062370
$\omega_{o\ 1}$	1.6875002
γ_1	0.1227275
f_2	0.0170255
$\omega_{o\ 2}$	1.8680944
γ_2	0.1276418
f_3	0.0082589
$\omega_{o\ 3}$	2.6480806
γ_3	0.0717503
f_4	0.0109446
$\omega_{o\ 4}$	2.7898784
γ_4	0.1625156

Left : n X : Photon Energy Right : k

olor filter_Quatre (| 1 | n = 1.590 | Photon Energy = 0.6 eV | k = 0.000

Dielectric function of a green colored filter described by 4 oscillators

Applications to materials

The Lorentz oscillator model is applicable to insulators. It describes well for example the behaviour of a transparent or weakly absorbent material (insulators, semiconductors). The spectral range of validity of the Lorentz formula depends on the material but usually the fit is performed over the region $\omega<\omega_t$ for the single Lorentz oscillator and $\omega<\omega_i$ in case of multiple oscillators where ω_i is the transition energy of the oscillator of highest order.

List of materials following single lorentz oscillator model

Materials	ε_∞	ε_s	ω_t	Γ_0	S. R. (eV)
AlAs	1.0	8.27	4.519	0.378	0 - 3
AlGaN	1.0	4.6	7.22	0.127	0.6 - 4
AlN	1.0	4.306	8.916	0	0.75 - 4.75
Al_2O_3	1.0	2.52	12.218	0	0.6 - 6
Al_xO_y	1.0	3.171	12.866	0.861	0.6 - 6
Aminoacid	1.0	1.486	14.822	0	1.5 - 5
Au disc	1.0	2.409	1.628	0.708	
Biofilm	1.0	2.12	12.0	0	1.5 - 5
CaF_2	1.0	2.036	15.64	0	0.75 - 4.75
CrO	0.687	3.1	8.0	1.694	
Red Color Filter	1.0	2.497	5.278	0	0.65 - 2
GaAs Ox.	2.411	3.186	5.855	0.131	0.75 - 4.75
GeO_x	1.0	2.645	16.224	0.463	0.6 - 4
H_2O	1.0	1.687	11.38	0	1.5 - 6
HfN	1.0	3.633	8.452	0	
HfO_2	1.0	2.9	9.4	3.00	1.5 - 6
HMDS	1.0	2.1	12.0	0.500	1.5 - 6.5
ITO	1.0	3.5	6.8	0.637	1.5 - 6
o - LaF_3	1.0	2.546	14.098	0.177	0.75 - 4.75
e - LaF_3	1.0	2.521	16.842	0.670	0.75 - 4.75
$LiGdF4{:}Eu^{3+}$	1.0	2.256	16.594	8.416	1,0 - 6.5
$LiNbO_3$	1.0	5.0	12.0	0	
LTO	1.0	2.204	13.784	0	
MgF_2	1.0	1.899	16.691	0	0.8 - 3.8
MgO	11.232	2.599	1.0	0	1.5 - 5.5
NBF3	1.0	2.503	13.911	0	
NiO	1.0	121.480	3.470	0.360	

List of materials following single Lorentz oscillator model

Materials	ε_∞	ε_s	ω_t	Γ_0	S. R. (eV)
PEI	1.0	2.09	12.0	0	0.75 - 4.75
PEN	1.0	2.466	4.595	0	1.5 - 3.2
PET	1.0	3.2	7.0	0	
PMMA	1.0	2.17	11.427	0	0.75 - 4.55
Polycarbonate	1.0	2.504	12.006	0	1.5 - 4
Polymer	1.0	2.3	12.0	5.0	0.75 - 4.75
PP	1.0	2.16	8.579	0.065	1.5 - 6.5
p-Si	1.0	12.0	4.0	0.5	1.5 - 5
Spincoated Polystryol	1.0	2.25	8.0	0	1.5 - 5
PTFE	1.0	1.7	16.481	0	1.5 - 6.5
PVC	1.0	2.304	12.211	0	1.5 - 4.75
Quartz	1.0	2.264	11.26	0	
Resist	1.0	2.189	10.814	0.334	1.5 - 6.5
Sapphire	1.0	3.09	13.259	0	1.5 - 5.5
a-Si : H	3.22	15.53	3.71	2.14	1.5 - 6
a-Si	3.109	17.68	3.93	1.92	1.5 - 6
SiC	3.0	6.8	8.0	0	0.6 - 4
SiN	2.320	3.585	6.495	0.398	0.6 - 6
Si_3N_4	1.0	5.377	3.186	1.787	1.5 - 5.5
SiO_2	1.0	2.12	12.0	0.1	0.7 - 5
SiO_2 doped As	1.0	2.154	11.788	0	1.8 - 5
SiO_xCH_y	1.0	2.099	13.444	0	1.45 - 2.75
SiON	1.0	2.342	10.868	0	0.75 - 3
SnO_2	3.156	3.995	4.786	1.236	
Ta_2O_5	1.0	4.133	7.947	0.814	0.75 - 4
TiO_x	0.290	3.820	6.50	0	0.6 - 3
YAG:Tb(10%)	1.0	2.545	10.342	0.793	1,0 - 6.5
Y_2O_3	1.0	2.715	9.093	0	1.55 - 4
ZrO_2	1.0	3.829	9.523	0.128	1.5 - 3

List of materials following multiple oscillator model

Materials / Parameters	CuPc	Green Color Filter	Pentacene/Si
ε_∞	1.800	2.481	1.834
f_1	0.140	0.00623	1.093
$\omega_{0,1}$	2.000	1.687	1.981
γ_1	0.130	0.122	0.207
f2	0.400	0.0170	-1.539*
$\omega_{0,2}$	3.700	1.868	1.982
γ_2	0.900	0.127	0.164
f3	0.140	0.00825	0.579
$\omega_{0,3}$	1.700	2.648	1.982
γ_3	0.150	0.0717	0.133
f4	0.130	0.0109	1.524
$\omega_{0,4}$	2.550	2.789	3.100
γ_4	0.950	0.162	11.005

References

1) H. M. Rosenberg, *The Solid State*, Oxford University Press
2) F. Wooten, *Optical Properties of Solids*, Academic Press (1972)
3) Eugene Hecht, *Optics*, Chap. 3, Hardcover (2001)
4) http://www.ifm.liu.se/~boser/surfacemodes/L3.pdf
5) langley.atmos.colostate.edu/at622/notes/at622_sp06_sec13.pdf