Causal models for Gauss-Lorentz response of solid media to radiative excitation

Jeremy Orosco and **Carlos F. M. Coimbra**

Department of Mechanical and Aerospace Engineering, Center for Energy Research,
University of California San Diego

UC San Diego JACOBS SCHOOL OF ENGINEERING Mechanical and Aerospace Engineering

CER Center for Energy Research

COIMBRA RESEARCH GROUP

Significance and objectives

The design and manufacture of optics and electronics depend critically on accurate knowledge of the **optical properties** of the materials involved. The optical properties of a given medium are determined by its **permittivity**, which has the following model structure for **linear optical media**:

$$\varepsilon(\omega) = \varepsilon_\infty + \chi(\omega)$$

$$\chi(\omega) = \sum_\phi \chi_\phi(\omega) + \sum_\beta \chi_\beta(\omega)$$

where χ is the electric **susceptibility**, resulting from **free** (ϕ) and **bound** (β) particle dynamics.

The **objectives of this work** are to provide accurate models:

1. for broad structures in **amorphous** media
2. for sharp electron transitions found in **metals**
3. that are physically consistent

These involve structures having both Gaussian and Lorentzian character. Existing models for these structures do not meet criteria for physical consistency and therefore do not represent materials that exist in nature. Unphysical models produce spurious results when utilized in certain theoretical frameworks.

Kramers-Kronig Relations (KKRs)

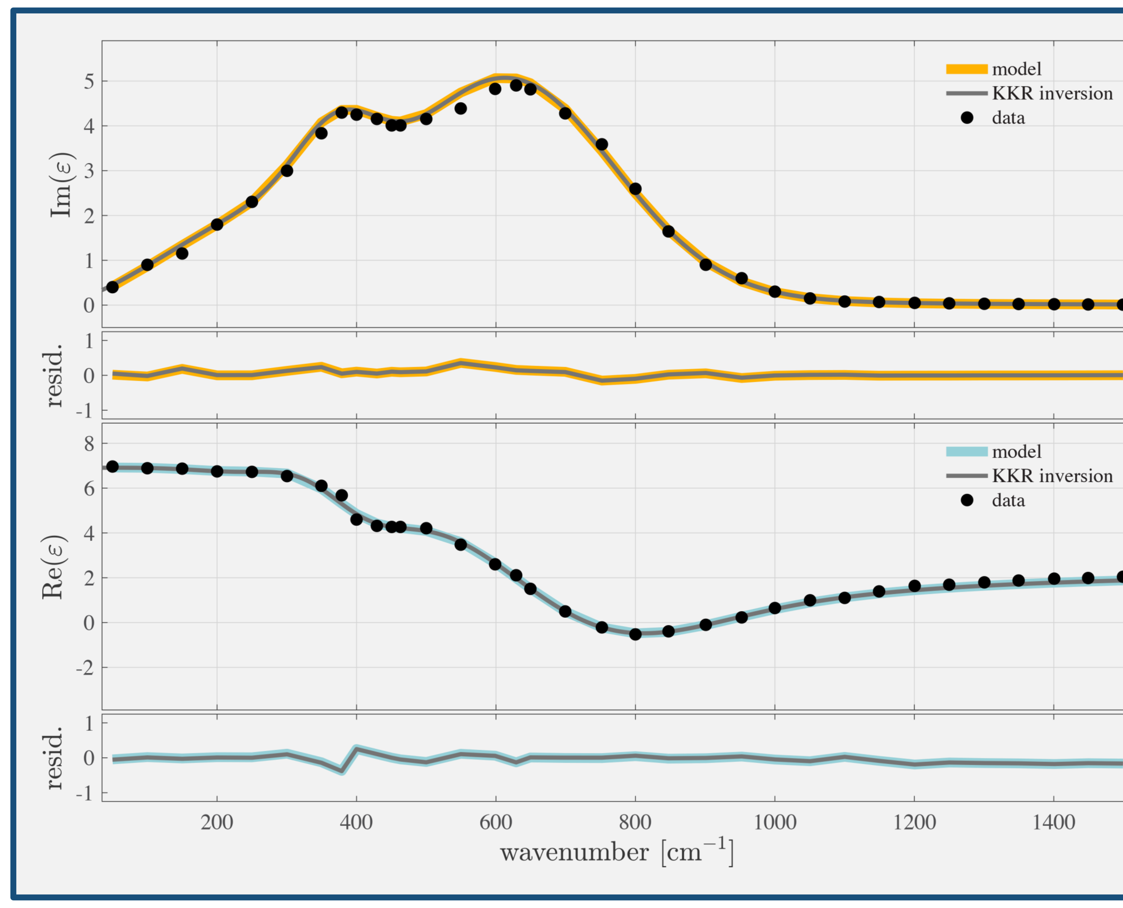
The KKRs constitute a mathematical formalism for assessing the **relativistic causality** of any susceptibility model corresponding to a **real-valued** time domain material response function (i.e., Green's function).

$$\chi'(\omega) = \frac{2}{\pi}\mathcal{P}\int_0^\infty \frac{\varpi\,\chi''(\varpi)}{\varpi^2-\omega^2}\,d\varpi$$

$$\chi''(\omega) = -\frac{2}{\pi}\mathcal{P}\int_0^\infty \frac{\omega\,\chi'(\varpi)}{\varpi^2-\omega^2}\,d\varpi$$

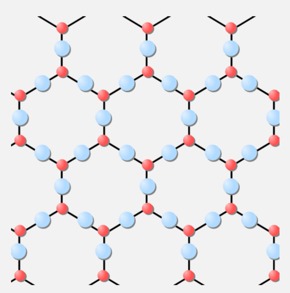
$$\chi'(\omega) \triangleq \mathbb{R}\mathrm{e}\{\chi(\omega)\},\quad \chi''(\omega) \triangleq \mathbb{I}\mathrm{m}\{\chi(\omega)\}$$

The causality requirement is satisfied if the model is **analytic** in the closed upper half complex plane. If, in addition, the model is **Hermitian**, then its inverse Fourier transform will be real-valued.



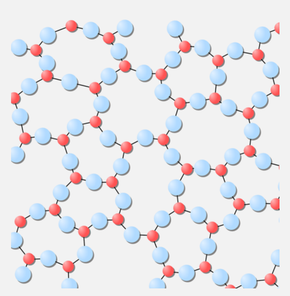
Infrared optical response of **amorphous alumina** (Al_2O_3) [1]. The **KKR transformed model** is included in the plots. The residual subplots show error for the model and for the numerical KKR inversion (both are computed with respect to the experimental data).

The model is obtained in 10 parameters (3 oscillators) leading to a relative RMSE of 0.08. The small residuals of the model inversion indicate that it has a **high degree of physical and internal consistency**.



crystalline

The ordered structure of crystalline models often permits a classical description as originally given by **Drude** (for the **free electron** behavior in metals) and **Lorentz** (for the oscillations of **bound particles**).



amorphous

The long range **lattice stochasticity** of amorphous media requires a different modeling approach. The behavior of the particles in such complex physical configurations gives rise to **broadened absorption** profiles.

Novel Gauss-Lorentz oscillator

The novel Gauss-Lorentz oscillator has the following **properties** [1,2]:

- strict adherence to KKRs
- asymptotic agreement with underlying classical theory
- internal-consistency
- 3-parameter KKR-compliant pure complex Gaussian oscillator for $\gamma = 0$

Previous models for modeling Gauss-Lorentz structures lack many (or, in some cases, all) of the noted properties.

$$\chi^*_\beta \triangleq \frac{\omega_p^2 f_\beta}{\omega_\beta^2}\left[\frac{s_w(z_+)+s_w(z_-)}{\chi_0}\right]$$

$$s_w(z) \triangleq i\,\pi\,w(z) + \exp(-z^2)\left[\log(z) + \log\left(-\frac{z^*}{|z|^2}\right) - i\,\pi\right]$$

$$z_\pm \triangleq (\pm\alpha_\beta - \omega_\beta)/\sqrt{2}\,\sigma_\beta$$

$$\alpha_\beta = \alpha'_\beta + i\,\alpha''_\beta \approx \sqrt{\omega^2 + i\,\gamma_\beta\,\omega}$$

$$\alpha'_\beta \triangleq (\omega/2)^{1/2}\left[(\omega^2+\gamma_\beta^2)^{1/2} + \omega\right]^{1/2}$$

$$\alpha''_\beta \triangleq (\omega/2)^{1/2}\left[(\omega^2+\gamma_\beta^2)^{1/2} - \omega\right]^{1/2} + \mu$$

$$D(z) \triangleq (\sqrt{\pi}/2)\exp(-z^2)\,\mathrm{erfi}(z),\quad w(z) \triangleq \exp(-z^2)\,\mathrm{erfc}(z)$$

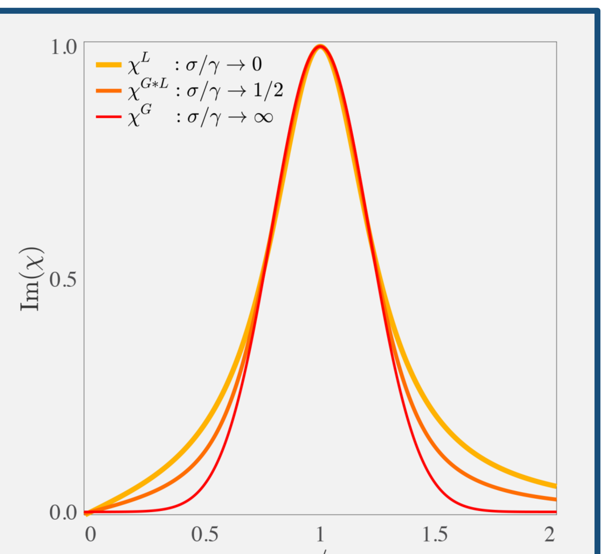
$$\chi_0 \triangleq -4\sqrt{\pi}\,D\left(-\frac{\omega_\beta}{\sqrt{2}\,\sigma_\beta}\right)$$

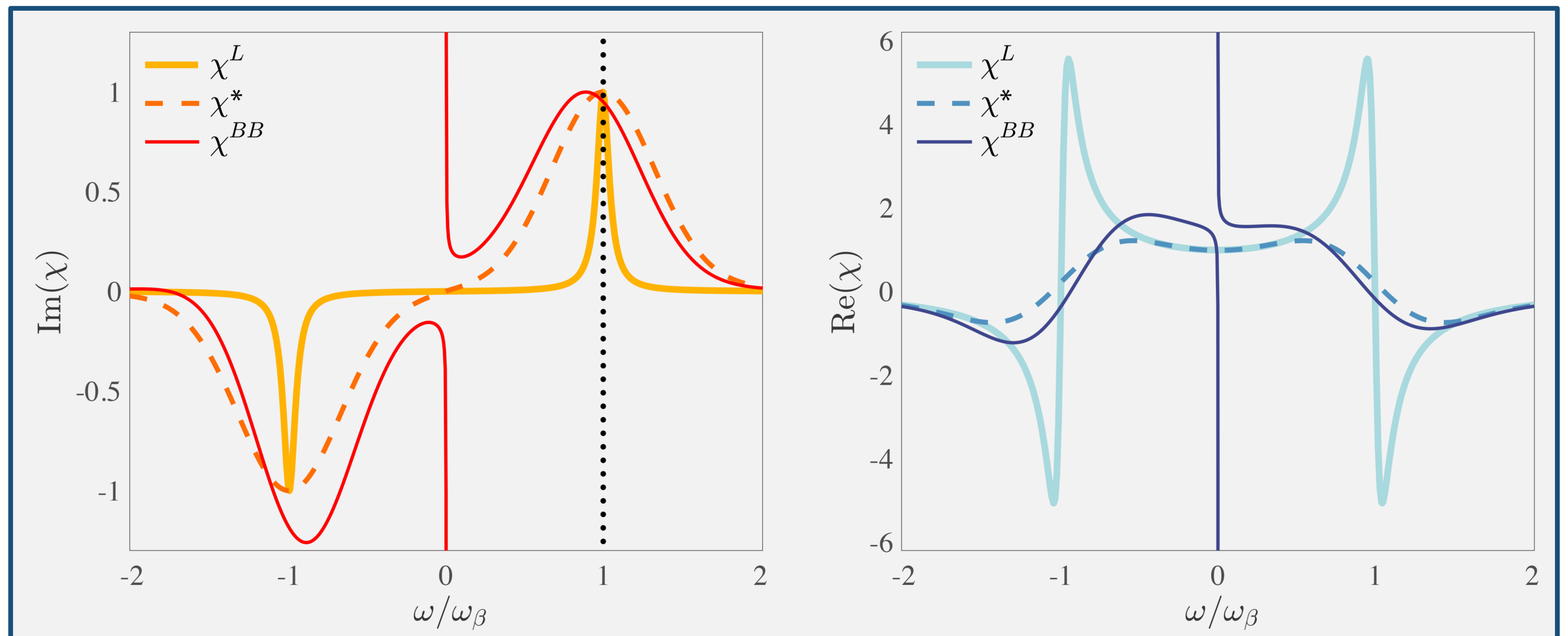
Gauss-Lorentz convolution approach

$$\chi_\beta^{G*L}(\omega) \triangleq \frac{\omega_p^2 f_\beta}{\sqrt{2\pi}\,\sigma_\beta}\int_{-\infty}^{\infty}\frac{\exp\left[-\left(\frac{y-\omega_\beta}{\sqrt{2}\,\sigma_\beta}\right)^2\right]}{\omega_\beta^2-\omega^2-i\,\gamma_\beta\,\omega}\,dy$$

ω_p: plasma frequency
f_β: oscillator strength
γ_β: Lorentzian broadening
σ_β: Gauss broadening

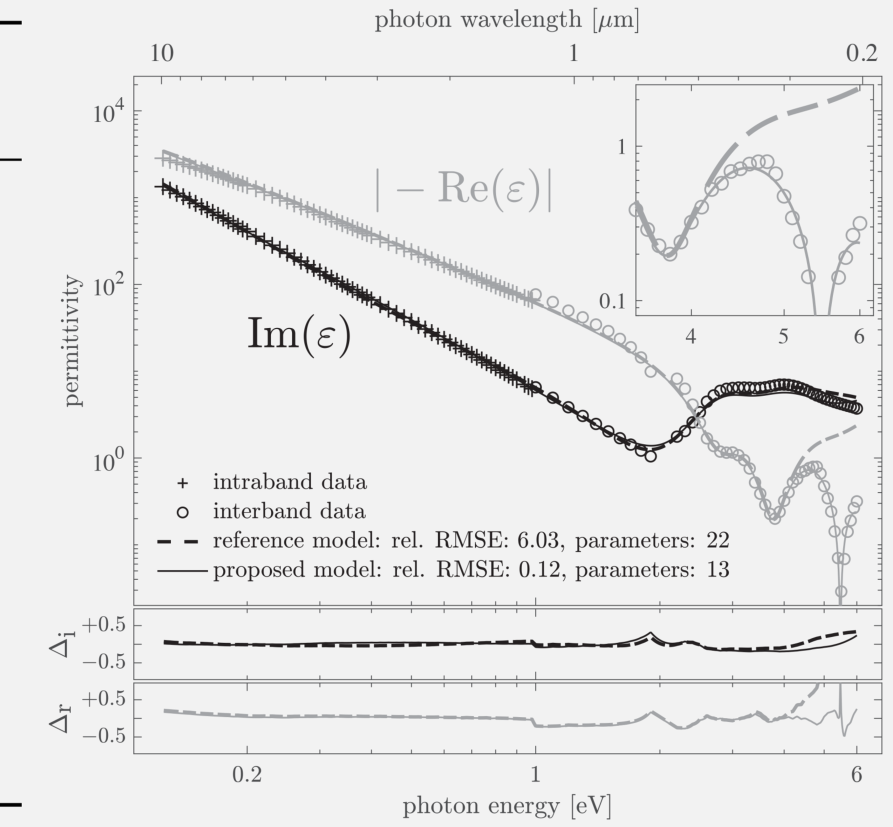
The Gauss-Lorentz convolution approach assumes an **infinite number of Gaussian distributed Lorentzian oscillators** about a critical point. The profiles have "tunable" Gaussian character (as indicated in the figure) that is capable of reproducing the broadened profiles.





Comparison of the novel oscillator (dashed lines), the Lorentz oscillator (thick solid lines), and the Brendel-Bormann (BB) oscillator (thin solid lines). The novel oscillator is self-consistent and exhibits good agreement with the asymptotics of the underlying classical theory. Since the novel oscillator is Hermitian and convergent in the closed upper half complex plane, it **adheres strictly to the KKRs** and therefore **generally represents materials which can exist in nature**. This is not the case for models utilizing the BB oscillator.

	rel. RMSE			
model	reference	proposed	ΔK	ΔN
Ag	4.10	0.19	1	9
Au	6.03	0.12	1	9
Cu	0.15	0.12	0	5
Al	0.08	0.07	1	4
Be	0.09	0.08	1	3
Cr	0.13	0.12	1	3
Ni	0.15	0.04	1	3
Pd	0.20	0.06	1	2
Pt	0.08	0.08	1	3
Ti	0.11	0.10	1	6
W	0.21	0.08	0	2



Models for several metallic films were generated using a novel **reductive modeling procedure** [2]. The table provides results for each material as compared with those from an often cited study using **identical source data**. The last two columns indicate model simplification by **reduction of oscillators** (ΔK) and/or **reduction of parameters** (ΔN). The plot provides a comparison for gold.

Conclusions

The novel Gauss-Lorentz oscillator **accurately reproduces broadened absorption** in amorphous materials and sharp electronic transitions in metals. It does so while **adhering strictly to established physical criteria** for real materials along with many other desirable properties. The model provides an excellent alternative to previously proposed unphysical models or models that less accurately reproduce the data.

Contact: Carlos F. M. Coimbra, ccoimbra@ucsd.edu

For more information, consult the following works (and the references therein):

[1] J. Orosco and C. F. M. Coimbra, "Optical response of thin amorphous films to infrared radiation," Phys. Rev. B **97**, 094301-1–094301-10 (2018).

[2] J. Orosco and C. F. M. Coimbra, "On the determination of optical properties for thin metallic films," Appl. Opt. (under review).

Author's website: http://acsweb.ucsd.edu/~jrorosco/