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Optical constants at complex energies: local deconvolution

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Abstract: Causality implies that the optical constants of any material continue in the upper complex plane of photon energies or wavelengths as an analytic function. This is the basis for Kramers-Kronig dispersion relations to obtain ε_1 from ε_2, or n from k. However, there have not been attempts to explore this continuation. This research focuses on such continuation and on applications thereof. An interesting property has been found: optical constants progressively smoothen when entering the upper complex plane. The continuation to complex energies is found to result in an average of the optical constants with a Lorentzian weight function.

This optical-constant smoothening originated in a shift to the upper complex plane is naturally produced in optical constants that have been obtained by means of an optical instrument with a Lorentzian slit function. This smoothening results in reduced resolution through convolution with the slit function. A procedure that takes advantage of optical constants at complex energies is developed for optical-constant deconvolution. Deconvolution is performed locally, i.e., with no integration, and it consists in shifting the energy of the optical constants by an imaginary amount by means of a Taylor series expansion. The first correction term involves the derivative of the other optical constant. Even though deconvolution of optical constants measured with a Gaussian slit cannot be directly performed with the present method, an approach based on powers of the Lorentz function is also proposed. This procedure could be implemented as an analysis tool of a spectrophotometer or an ellipsometer; this tool would enable one to measure optical constants with a modest resolution and to improve it by post—processing them with the present scheme.

Keywords: optical constants, deconvolution, optical models, optical materials, optical properties of thin films

1. Introduction

Pairs of optical constants, such as ε_1 and ε_2 or n and k, can be set as a complex number $\varepsilon_1+i\varepsilon_2$ or $N=n+ik$ that is a function of a real variable, such as photon energy or wavelength. Such function is known to be the limit of an analytic (holomorphic) function defined in the upper complex half plane C^+, i.e., for photon energies that are complex numbers with a positive imaginary part. Hence the optical constants of a material can be continued in C^+, where they must be analytic (holomorphic). This is a consequence of causality and it is connected with the well-known Kramers-Kronig (KK) dispersion relations [1,2].

This extension of the spectral variable to a complex variable has been mostly used in the literature as a mere mathematical procedure to get to a convenient result, i.e., KK dispersion relations, which is applied at real (i.e., non-complex) photon energies. Hence the complex spectral variable is not used in practical calculations of ε_1 from ε_2 or n from k and vice versa.

There are few practical cases in which optical constants have been used in or proposed to extend to C^+. The optical constants at the imaginary axis have been used in the evaluation of the Casimir thermal effect [3]. Sum rules as a procedure to evaluate optical-constant consistency have been proposed for optical constants at imaginary energies [4]. Adachi proposed a procedure to avoid divergences in simple optical-constant models based in shifting photon energy along the imaginary axis [5]. However, there have not been attempts to explore the correspondence between optical constants on the real axis and on the complex plane.

This research presents an interpretation of the optical constants extended to complex photon energies. The shift of the optical constants from real to complex photon energies results in a certain average of the optical constants (mathematically, a convolution) with a Lorentzian weight function where the half-width of this weight function is given by the (positive) imaginary part of the photon energy. Hence moving from the real axis through C^+ results in a progressive optical-constant smoothening.

This shift of the optical constants into C^+ can find a practical application through improving spectral resolution on measured optical constants. Optical-constant resolution is limited by the optics of the optical instrument, by its detector resolution (for an array detector), and, in practice, by measurement time. The latter means that even if the optical system and detector enable higher resolution, an improved resolution may require an unavailable acquisition time. The obtained resolution can be improved by deconvolution of the measured optical constants. The present research proves that deconvolution can be obtained by local differentiation of the optical constants and a Taylor series expansion, i.e., with no integration. Such deconvolution could be implemented as a post-analysis tool of the optical instrument, such as a spectrophotometer or an ellipsometer. The procedure can be applied on an optical instrument with a Lorentzian slit function. Even though the present procedure does not apply to measurements with a Gaussian slit function, it can be done by means of approaching the Gaussian slit function with a power of the Lorentz function.

The paper is organized as follows. In section 2 we derive the equations to relate the optical constants at complex energies with a convolution, which is illustrated with figures of optical-constant smoothening. Section 3 presents the application of the continuation to complex energies for the local deconvolution of an optical function blurred with a Lorentzian slit function and with faster-decaying functions.

2. Extension of optical constants to complex photon energies

2.1 Interpretation of optical constants at complex photon energies

Let $\tilde{\varepsilon}(E) = \varepsilon_1(E) + i\varepsilon_2(E)$ be the dielectric function of a uniform material, where E is the real photon energy. We will initially assume that the material is non-conductor to avoid the divergence of $\tilde{\varepsilon}$ at E=0; the case of conductors is addressed below. By virtue of causality, the dielectric function is the limit on the real axis of an analytic (holomorphic) function in C^+ whose real and imaginary parts are Hilbert transform of each other. The latter is typically quoted as Titchmarsh theorem [6], whereas Labuda and Labuda [7] attribute such result originally to the combined work of Riesz [8] and of Payley and Wiener [9]. Let us convolve the dielectric function with a Lorentzian function. Using Cauchy's integral formula, it is straightforward to get to the following result:

$$\oint[\tilde{\varepsilon}(z) - 1]\frac{a/\pi}{a^2+(z-E)^2}dz = \int_{-\infty}^{\infty}[\tilde{\varepsilon}(E') - 1]\frac{a/\pi}{a^2+(E'-E)^2}dE' = \tilde{\varepsilon}(E + ia) - 1 \text{ (non conductors)} \quad (1)$$

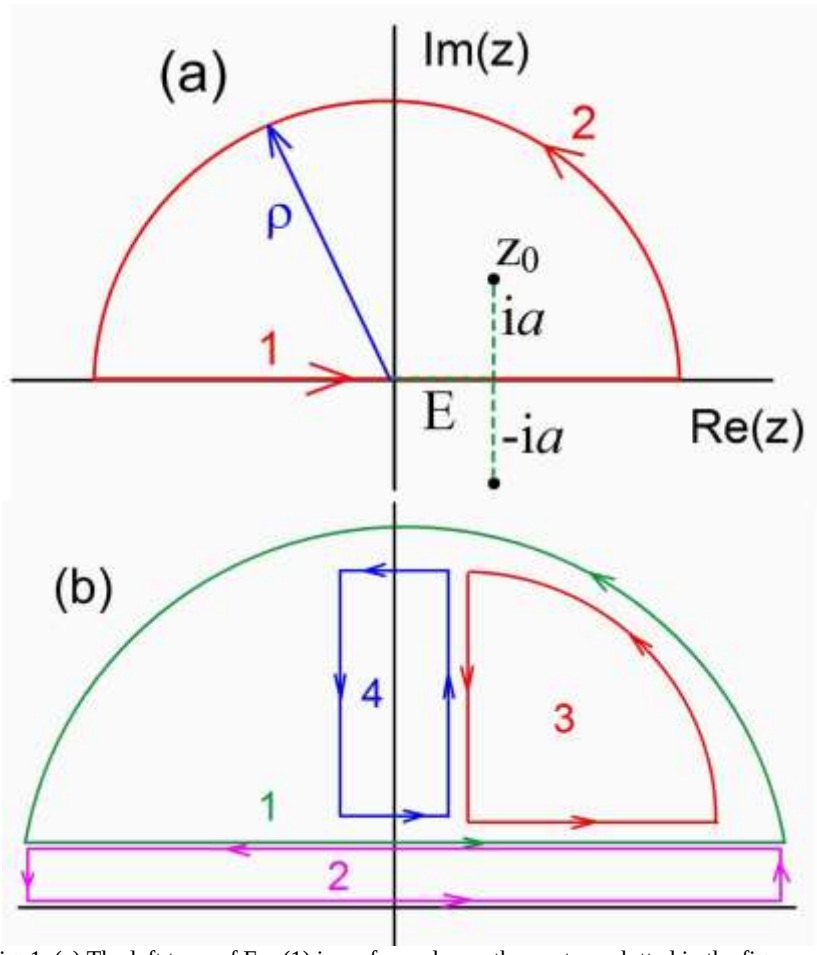

Fig. 1. (a) The left term of Eq. (1) is performed over the contour plotted in the figure, with z_0=E+ia being the only pole within the contour, which is extended to infinity. The other pole lies in the lower complex half plane at –ia. (b) Various integration contours to obtain sum rules.

where the integral contour is plotted in Fig. 1(a). In the limit of $\rho\to\infty$, path 2 of Fig. 1(a) gives no contribution due to function asymptotic convergence, so that the integral only needs to be calculated over path 1, which is the middle term of Eq. (1). The right term was obtained by calculating the residue at pole $z_0=E+ia$. To develop Eq. (1) it was used that $\tilde{\varepsilon}$ is an analytic function in C^+ and has no poles in the contour, including the real axis. The equality between the second and third term is known as the Poisson integral formula [10].

The convolution in Eq. (1) can be interpreted as an average of the dielectric function, with larger weight in the E±a range, although the integral extends to the whole spectrum. The right-hand side of Eq. (1) results in that this sort of average of the dielectric function can be calculated by just evaluating the dielectric function at the single complex energy E+ia. Equation (1) indicates that the continuation of optical constants to a domain with complex energies with positive imaginary part [right term of Eq. (1)] results in an average of the optical constant (through a convolution with a Lorentzian function) [middle term of Eq. (1)], in which the central energy of this average is the real part of the complex energy E and the half-width of this average is given by the imaginary part of the complex energy a.

Equation (1) can be applied iteratively. Let us do the same procedure on a line parallel to the real axis in C^+ at z=ia:

$$\int_{-\infty}^{\infty}[\tilde{\varepsilon}(E'+ia)-1]\frac{\delta/\pi}{\delta^2+(E'-E)^2}dE' = \tilde{\varepsilon}[E+i(a+\delta)]-1,\ a,\delta>0 \qquad (2)$$

which is obtained with the same procedure that Eq. (1). $\tilde{\varepsilon}(E+ia)$ with a>0 has no pole even for conductors (no pole at the shifted axis), so that Eq. (2) is valid for conductors and non-conductors. Eq. (2) expresses that the convolution with a Lorentz function through an axis shifted along the imaginary part of energy is additive.

The dielectric function of conductors involves a pole at E=0, which requires a modification of Eq. (1). The present formalism is adapted for metals with a pole at zero energy that conforms to the one of a Drude model and we overcome the presence of the pole by subtracting a term $i\sigma_{DC}\hbar/\varepsilon_0 E$ from the complex dielectric function, which finally results in:

$$P\int_{-\infty}^{\infty}[\tilde{\varepsilon}(E')-1]\frac{a/\pi}{a^2+(E'-E)^2}dE' + \frac{\sigma_{DC}\hbar a}{\varepsilon_0(E^2+a^2)} = \tilde{\varepsilon}(E+ia)-1 \qquad \text{(Drude-like conductors)} \quad (3)$$

where P stands for the integral in the sense of Cauchy principal value. There is an extra term that arises from the pole at E=0; this term involves the material DC conductivity σ_{DC} (in the International System of Units), along with Planck's reduced constant $\hbar$ and the permittivity of vacuum ε_0. The name "Drude-like metals" means here that the model is applied to metals with a pole at E=0 with the same functionality that in Drude model, and it does not mean that we are modifying Drude model.

The optical constants of materials at positive imaginary energies are used as a standard to calculate the thermal Casimir force because integration over the imaginary axis gives more robust results versus the oscillating results obtained when integrating over the real axis [3]. Let us particularize Eq. (3) at E=0 (pure imaginary energies) for the evaluation of optical-constants to calculate the Casimir thermal effect:

$$\int_{-\infty}^{\infty}[\varepsilon_1(E')-1]\frac{a/\pi}{a^2+E'^2}dE' = Re[\tilde{\varepsilon}(ia)]-1-\frac{\sigma_{DC}\hbar}{\varepsilon_0 a} \qquad \text{(Drude-like conductors)} \quad (4a)$$

$$P\int_{-\infty}^{\infty}[\varepsilon_2(E')]\frac{a/\pi}{a^2+E'^2}dE' = Im[\tilde{\varepsilon}(ia)] = 0 \qquad \text{(Drude-like conductors)} \quad (4b)$$

Equation (4b) is null because of the dielectric-function parity properties at the imaginary axis. By applying the parity conditions on Eq. (4a):

$$2\int_{0}^{\infty}[\varepsilon_1(E')-1]\frac{a/\pi}{a^2+E'^2}dE' = Re[\tilde{\varepsilon}(ia)]-1-\frac{\sigma_{DC}\hbar}{\varepsilon_0 a} \qquad \text{(Drude-like conductors)} \quad (5)$$

Equations (4b) and (5) are combined in:

$$\tilde{\varepsilon}(ia)-1 = Re[\tilde{\varepsilon}(ia)]-1 = \frac{\sigma_{DC}\hbar}{\varepsilon_0 a} + \frac{2a}{\pi}\int_0^{\infty}\frac{[\varepsilon_1(E')-1]}{a^2+E'^2}dE' \quad \text{(Drude-like conductors)} \quad (6)$$

Equation (6) is valid also for non-conductors by just taking σ_{DC}=0. By starting with Eq. (3) with an added E' factor in the integrand and by particularizing at E=0, the following is obtained:

$$\tilde{\varepsilon}(ia)-1 = Re[\tilde{\varepsilon}(ia)]-1 = \frac{2}{\pi}\int_0^{\infty}\frac{E'\varepsilon_2(E')}{a^2+E'^2}dE' \qquad \text{(Drude-like conductors)} \quad (7)$$

which is a well-known dispersion relation [11] that is used in the calculation of the thermal Casimir effect. The usual Eq. (7) could be replaced with Eq. (6) in the calculation of the thermal Casimir effect, although it requires knowledge of σ_{DC}.

From Eqs. (6) and (7) the following identity is obtained:

$$a\int_0^\infty \frac{[\varepsilon_1(E')-1]}{a^2+E'^2}dE' + \frac{\pi\sigma_{DC}\hbar}{2\varepsilon_0 a} = \int_0^\infty \frac{E'\varepsilon_2(E')}{a^2+E'^2}dE' \qquad \text{(Drude-like conductors) (8)}$$

The above developments can be reproduced by replacing $\tilde{\varepsilon} = \varepsilon_1 + i\varepsilon_2$ with $\tilde{N}$= $\tilde{\varepsilon}^{1/2}$=$n$+i$k$. Operating with $\tilde{N}$ instead of $\tilde{\varepsilon}$ involves a simplification: for a conductor with a Drude sort of divergence at E=0, there is no need for the added term of Eq. (3) because the divergence of $\tilde{N}$ at $E\rightarrow 0$ involves only the square root of energy. Hence one gets to:

$$\int_{-\infty}^{\infty}\left[\tilde{N}(E')-1\right]\frac{a/\pi}{a^2+(E'-E)^2}dE' = \tilde{N}(E+ia)-1 \quad \text{(both Drude metals and dielectrics) (9)}$$

In order for the latter to be valid for conductors, the integral must be understood as the Cauchy principal value. The identity given in Eq. (8) can be also obtained in terms of $\tilde{N}$:

$$a\int_0^\infty \frac{[n(E')-1]}{a^2+E'^2}dE' = \int_0^\infty \frac{E'k(E')}{a^2+E'^2}dE' \qquad (10)$$

which is valid also for conductors, and it was obtained by King [4]. More identities are obtained by means of the shift of the standard sum rules to C^+. The new sum rules are presented in Appendix A.

For the dielectric function and for the refractive index, the interpretation of the continuation from real to complex energies as an average through a range given by the imaginary part of energy was obtained with no assumption other that the optical-constant function must converge on the real axis and be analytic in C^+, which are always assumed in the context of Titchmarsh theorem [6]. Hence the same interpretation can be made for other analytic functions in C^+, such as the inverse of the dielectric function $(1/\tilde{\varepsilon})-1$, the inverse of the refractive index, $\left(1/\tilde{N}\right)-1$, or the single-interface reflectance logarithm {with the pair ln(r)+iθ in an adequately subtracted form to avoid divergence [2]}. Regarding conductors with a pole at E=0, $\tilde{N}-1$, $(1/\tilde{\varepsilon})-1$, and $\left(1/\tilde{N}\right)-1$ satisfy Eq. (1) with no extra term because either they have no pole (the latter two functions) or the divergence only goes with the square root of energy (the former function).

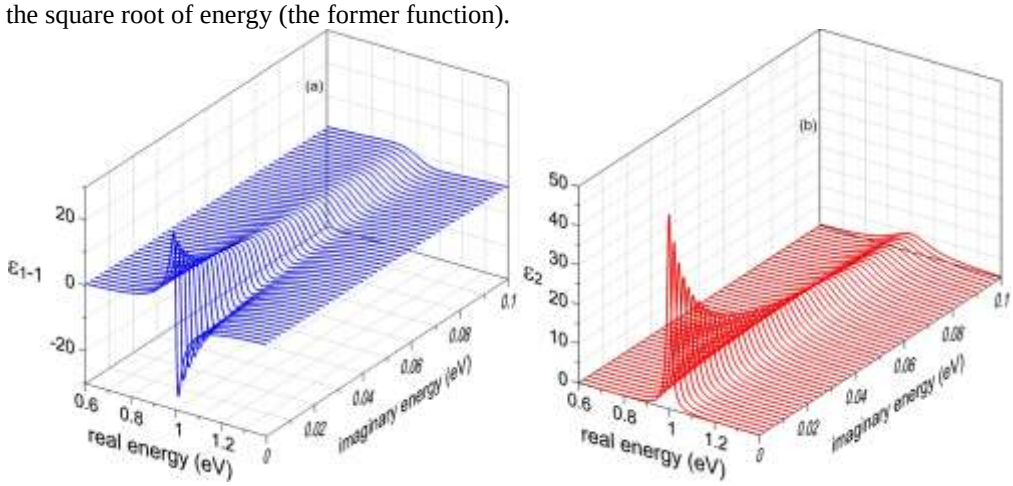

Fig. 2. ε_1-1 (a) and ε_2 (b) plot of a Lorentz oscillator versus complex energy. Oscillator central energy: 1 eV. Oscillator width: 0.01 eV. One plotted line every 0.002 eV imaginary energy.

2.2 Examples of optical constants at complex photon energies

Equation (1) helps interpret optical constants at complex energies. Figure 2 displays complex continuation for a simple function: a Lorentz oscillator. The figure displays that the convolution with a Lorentzian function results in a progressive curve smoothening with

increasing a, the imaginary part of energy. Equation (1) says that the behaviour observed for a Lorentz oscillator is a general property of any causal optical constants.

Let us see how the pole at E=0 for conductors affects optical-constant continuation in C^+. The case of a material with a pole at E=0 is exemplified in Fig. 3; the displayed function consists in the oscillator plotted in Fig. 2 plus a Drude-model term. ε_2 has a pole at E=0 for a=0, which is the standard pole of conductors. ε_2 smoothens with increasing a in the same way that the oscillator or any dielectric, i.e., by averaging through the convolution with a Lorentz function starting at a=0. However, ε_1 has an extra contribution that diverges for $a \to 0$ (but is not present at a=0), which is the expression of the extra term of Eq. (3) and (4a): contrarily to ε_2, smoothening does not start at a strictly 0, but once a>0. After that, ε_1 smoothens in the same way that ε_2.

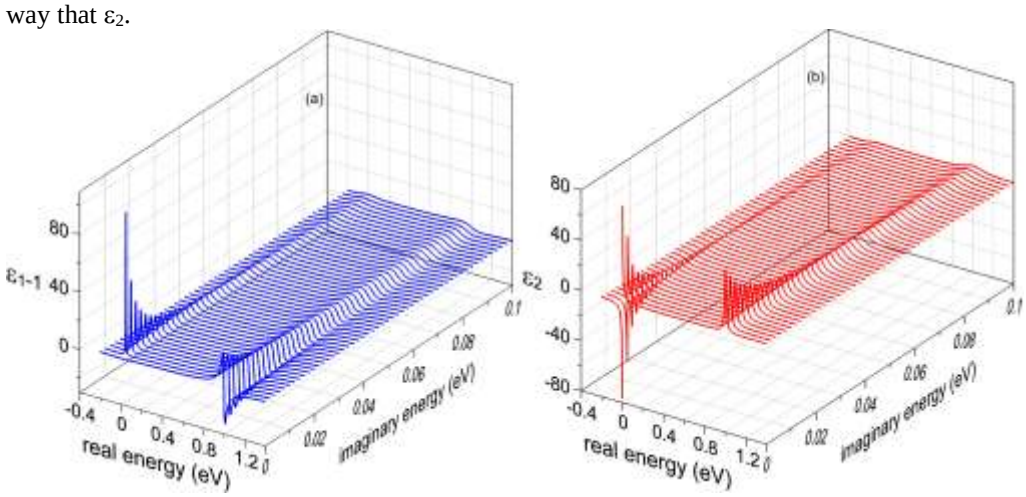

Fig. 3. ε_1-1 (a) and ε_2 (b) versus complex energy for a Lorentz oscillator (central energy=1 eV, width=0.01 eV) plus a Drude-model term $\tilde{\varepsilon} = 1 - E_p^2/E(E + iE_\tau)$, with E_p=0.2 eV and E_τ=0.2 eV. One plotted line every 0.002 eV imaginary energy.

It has been shown that shifting from the real axis into C^+ results in optical-constant smoothening. One may wonder if there might exist situations in which the optical constants do not follow such smoothening trend. Appendix B demonstrates that the observed smoothening is the only possible extension of the optical constants to C^+.

3. Local deconvolution of optical constants

3.1 Deconvolution with Lorentzian function

The present interpretation of optical constants at complex energies can be inversely applied: if we start with optical constants that are known to be a convolution with a Lorentzian weight function of known width, the optical constants can be deconvolved by just calculating them at an energy with a reduced imaginary part. This could be the case for measurements performed with an optical instrument that has a Lorentzian slit function, and the present deconvolution could be used to remove the slit-resolution limiting effect. If $\tilde{\varepsilon}_c(E)$ stands for the complex dielectric function obtained with a procedure that involves a convolution with a Lorentzian function of half-width a, then the present sort of deconvolution can be performed by recalculating the dielectric function at the energy shifted by -ia:

$$\tilde{\varepsilon}(E) = \tilde{\varepsilon}_c(E - ia) \tag{11}$$

where $\tilde{\varepsilon}(E)$ represents the correct deconvolved dielectric function. Eq. (11) assumes that the material is a dielectric; if it were a conductor, it would be more convenient to work, for

instance, with the refractive index to avoid an extra term to deal with the pole at E=0. Equation (11) is exact. Since the dielectric function is analytic, we can calculate $\tilde{\varepsilon}(E)$ through a Taylor series expansion. In the expansion, we will assume that the convergence radius extends to the axis of real energies:

$$\tilde{\varepsilon}(E) = \tilde{\varepsilon}_c(E) - ia\tilde{\varepsilon}_c{}'(E) - \frac{a^2}{2}\tilde{\varepsilon}_c{}''(E) + \frac{ia^3}{6}\tilde{\varepsilon}_c{}'''(E) + O(a^4) \qquad (12)$$

The number of primes stands for the derivative order. For the present sort of deconvolution we only need a local knowledge of the convolved dielectric function, and its first or, if necessary, higher-order derivatives in the real direction, and we do not need to know the function in an extended spectral range, as it is required in usual deconvolution processes. For such a deconvolution we replace the need to know the function in an extended (virtually infinite) range with the need to precisely know the function locally in order to take first or higher order derivatives. In a typical case of deconvolution, a is expected to be small. To first order, the evolution of ε_1 (ε_2) in the imaginary direction depends on the derivative of ε_2 (ε_1) in the real direction; hence to first order, deconvolution of the optical constants through shifting energy by a small imaginary amount only depends on the derivative of the other optical constant. When going to higher expansion orders, the roles of the same and the other constant are alternating, so that even (odd) orders involve even (odd) derivatives of the same (other) constant.

The presented local deconvolution with constant a can be generalized to a Lorentzian function with a half-width that is any function of photon energy by just replacing a in Eq. (12) with $a(E)$; in order to apply this procedure for local deconvolution, we need to know the half-width of the Lorentzian function at the photon energy E at which we perform the deconvolution.

Figure 4 compares a starting function that is a Lorentz oscillator both before and after convolving it with a Lorentz function of half-width a=0.03 eV, and also after deconvolution with Eq. (12) to 1st and 2nd order. Whilst first-order deconvolution gets to a significant approach to the original function, the second-order deconvolution results in an excellent match. Obviously, the larger a, the larger the effect of convolution and the worse the recovery for a given deconvolution order.

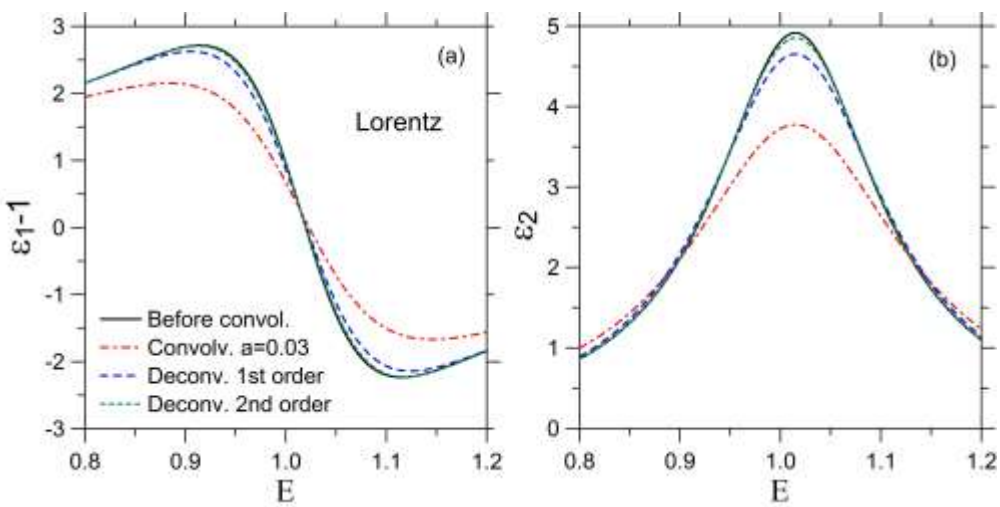

Fig. 4. ε_1-1 (a) and ε_2 (b) as a function of E (real part of complex energy) for a Lorentz oscillator (central energy=1 eV, width=0.2 eV). The starting function before convolution is plotted, along with the function convolved with a Lorentzian function with a=0.03 eV, and later deconvolved with Eq. (12) to 1st and 2nd order.

The deconvolution process based on Eq. (12) could be implemented in spectrophotometers and ellipsometers with Lorentz slit function for optical-constant measurements as a tool to improve spectral resolution. This would enable measuring with a somewhat larger slit width and hence saving acquisition time, in situations where the required resolution is either not possible to obtain or it requires too long a measurement time.

3.2 Convolution and deconvolution with functions faster than Lorentzian

Along with Lorentzian, Gaussian is the other function that is often assumed to convolve optical functions. Convolution with a Gaussian function cannot be addressed with the present scheme; this extends to the Voigt profile, because it involves a Gaussian function.

A Gaussian profile involves a lower weight in the convolution of distant photon energies compared with the Lorentzian profile. In this respect, powers of the Lorentzian function reduce the weight of distant energies compared with the standard Lorentz function and they progressively approach a Gaussian profile by increasing the power. Hence the convolution with a Gaussian function can be approached with the convolution with powers of the Lorentzian function, and the latter can be performed with the present scheme of deconvolution.

Equation (1) can be immediately adapted to include powers of the Lorentzian function by integrating a function with multiple-order poles; below are the square and the cube:

$$\frac{2a^3}{\pi}\int_{-\infty}^{\infty}\frac{[\tilde{\varepsilon}(E')-1]}{[a^2+(E'-E)^2]^2}dE' = \tilde{\varepsilon}(E+ia)-1-ia\tilde{\varepsilon}'(E+ia) \quad \text{(Lorentz square)} \quad (13)$$

$$\frac{8a^5}{3\pi}\int_{-\infty}^{\infty}\frac{[\tilde{\varepsilon}(E')-1]}{[a^2+(E'-E)^2]^3}dE' = \tilde{\varepsilon}(E+ia)-1-ia\tilde{\varepsilon}'(E+ia)-\frac{a^2}{3}\tilde{\varepsilon}''(E+ia) \quad \text{(Lorentz cube)} \quad (14)$$

Every time we increase the power of the Lorentz function in the integrand denominator, a new term with a further-order derivative is added. The above expressions are exact, i.e., they are not truncated expansions.

Let us see how we can use these equations to deconvolve experimental data, i.e. to recover $\tilde{\varepsilon}(E)$ out of $\tilde{\varepsilon}(E+ia)$ and its derivatives. Let $\tilde{\varepsilon}_c(E)$ be the optical constants obtained with a slit half-width a that were measured with an n-power Lorentzian slit function; $\tilde{\varepsilon}_c(E)$ is what was really measured [left term of Eqs. (13) and (14)], and it is related to $\tilde{\varepsilon}(E+ia)$ according to the right term of Eqs. (13) and (14) for n=2 and 3, respectively. We want to extract $\tilde{\varepsilon}(E)$ out of $\tilde{\varepsilon}_c(E-ia)$; the latter will be obtained from a Taylor series expansion. For n=2:

$$\tilde{\varepsilon}(E) = \tilde{\varepsilon}_c(E) - \frac{a^2}{2}\tilde{\varepsilon}_c''(E) - \frac{ia^3}{3}\tilde{\varepsilon}_c'''(E) + O(a^4) \quad (15)$$

For n=3:

$$\tilde{\varepsilon}(E) = \tilde{\varepsilon}_c(E) - \frac{a^2}{6}\tilde{\varepsilon}_c''(E) - \frac{a^4}{72}\tilde{\varepsilon}_c''''(E) + O(a^5) \quad (16)$$

Again, even terms correspond to even derivatives of the same optical constant, whereas odd terms correspond to odd derivatives of the other optical constant. Figure 5 compares an original function that is a Lorentz oscillator, both before and after convolving it with a Lorentz square function of half-width a=0.03 eV, and also after deconvolution with Eq. (15) to 2nd order. In this case the first correction involves a derivative (second derivative) of the same optical constant, so that this expansion to the first term in a can be applied even if the other optical constant were unknown. Figure 6 plots the same that Fig. 5 except for a Lorentz cube function deconvolved with Eq. (16) to 2nd and 4th order. For Lorentz cube, the first term in the deconvolution is again the second-order derivative of the same optical constant. It is seen that the larger the Lorentz power, the lower the effect of the convolution for a given a; on the other hand, it is obtained that the larger the Lorentz power, the larger the need of a

higher-order expansion. For the Lorentz cube, a reasonable approach of ε_2 requires 4th order expansion.

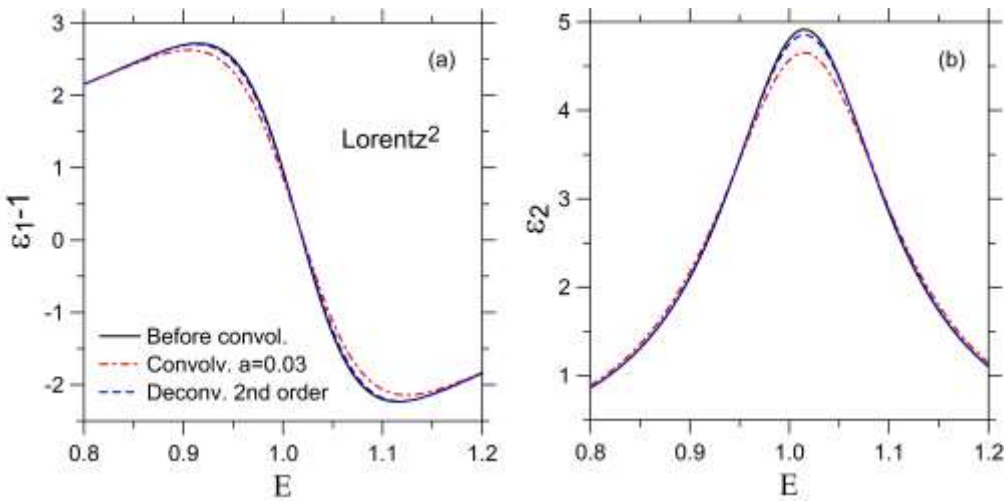

Fig. 5. ε_1-1 (a) and ε_2 (b) as a function of E (real part of complex energy) for a Lorentz oscillator (central energy=1 eV, width=0.2 eV). The original function before convolution is plotted, along with the function convolved with a Lorentz square function with a=0.03 eV, and later deconvolved with Eq. (15) to 2nd order.

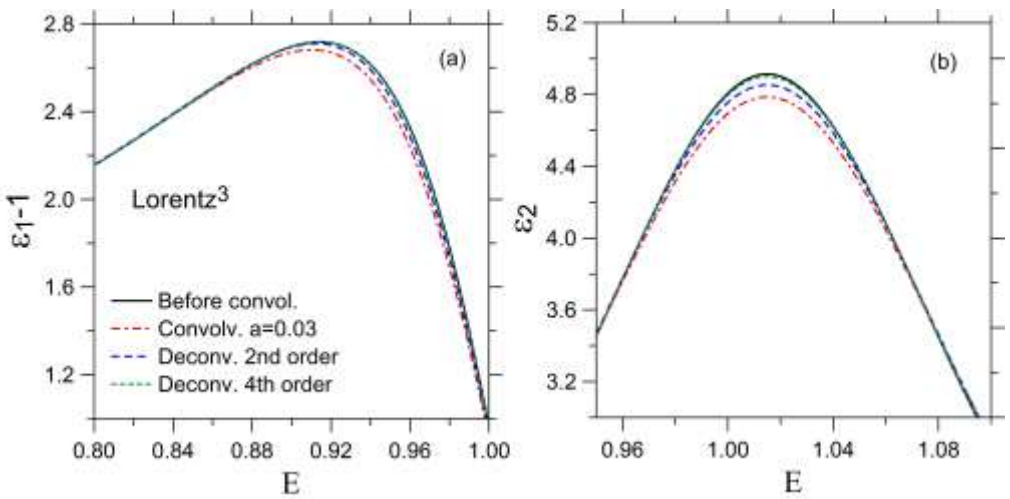

Fig. 6. ε_1-1 (a) and ε_2 (b) as a function of E (real part of complex energy) for a Lorentz oscillator (central energy=1 eV, width=0.2 eV). The original function before convolution is plotted, along with the function convolved with a Lorentz cube function with a=0.03 eV, and later deconvolved with Eq. (16) to 2nd and 4th order.

Other than Lorentzian and Gaussian profiles, Voigt profile is another popular convolution function. It consists in a convolution of a Lorentzian and a Gaussian function. Voigt profile involves middle weight of distant energies between slow-decaying Lorentzian profile and fast-decaying Gaussian profile. Voigt profile cannot be addressed with the present scheme either. Again, we can approach Voigt profile by replacing the Gaussian function with a Lorentz power and by later convolving it with a standard Lorentz function: $\text{Lorentz}^n * \text{Lorentz}^1$, where * stands for convolution. If our dielectric function were convolved with such a function, we could successively apply Eq. (1) to account for the standard Lorentz function and Eqs. (13) or (14) to account for Lorentz square or cube, respectively. Let a and b

be the half-widths of the standard Lorentz and of the Lorentz power function, respectively. For example, for a Lorentz cube we get from Eq. (1) and (14):

$$[\tilde{\varepsilon}(E') - 1] * L^1(a) * L^3(b) = \tilde{\varepsilon}(E + ia + ib) - 1 - ib\tilde{\varepsilon}'(E + ia + ib) - \frac{b^2}{3}\tilde{\varepsilon}''(E + ia + ib) \qquad (17)$$

Equation (17) is exact, i.e., it is not a truncated expansion.

Deconvolution procedures operating through derivatives have been reported in the literature [12,13,14,15]; they were developed to improve the resolution of spectrophotometric measurements. These procedures operate by performing a Fourier transform through series expansion. These procedures deal with a real function, which is the spectrum intensity versus energy. In those procedures and for some convolution functions, such as the Lorentzian function, the odd terms in the expansion do not only involve direct derivatives of the spectrum; they also involve derivatives of a certain integral (its Hilbert transform) over the spectrum, which results in missing the property of being local. The procedure developed here was applied to deconvolve not a spectrum but the optical constants. In the expansion of the optical constants, being a complex function whose real and imaginary parts are Hilbert transforms of each other, the odd terms naturally appear as the other optical constant. Eqs. (1), (13), and (14) express that convolution can be performed through differentiation. For the case of the Lorentzian function as well as the Lorentz powers, the convolution is performed exactly, and not as a series expansion. To our knowledge, this is the first time that deconvolution of optical constants has been performed through local derivatives.

As a distinct application of Eq. (1), it could be used to calculate integrals of functions that are divided by the term (a^2+x^2); such a function must satisfy the requirement that its Hilbert transform exists. Eqs. (13) and (14) could be also used to calculate integrals when the function (with the same requirement) is divided by $(a^2+x^2)^n$ terms, with n=2 and 3, respectively.

Conclusions

A simple interpretation for the optical constants at complex energies has been given. The extension to complex energies corresponds to a convolution with a Lorentzian function that results in a progressive optical-constant spectral smoothening into the imaginary energy axis. For Drude-like conductor materials, continuation of the optical constants to complex energies requires an extra term for the dielectric function; such extra term also progressively reduces for increasing energy, both its real and its imaginary part.

The optical constants at pure imaginary energies, which are required in the calculation of the Casimir thermal effect, can be also calculated by integration of ε_1 (or n) and a new identity was found for the integral of the dielectric function over the spectrum.

The interpretation of convolution through the continuation to complex energies enables a procedure to improve the resolution of optical constants. The procedure can be applied when the optical constants were measured using an instrument with a Lorentz slit function. The convolution introduced by the instrument results in a shift of the optical constants in the imaginary energy direction; the convolution range is given by the size of this shift. The desired optical constants at the non-shifted energy can be retrieved through Taylor expansion involving local derivatives, and the first term corresponds to the first derivative of the other optical constant. Hence deconvolution can be performed with no integration. This deconvolution procedure could be implemented as an analysis tool to improve spectral resolution in optical instruments that are used to obtain optical constants.

Even though Gaussian convolution cannot be performed with the present scheme, it can be approached with powers of the Lorentz function, and the present procedure can be adapted for

convolution with the latter function. The convolution with a Lorentz power function also results in an energy shift of the optical constants in the imaginary direction, and, additionally, it involves as many optical-constant derivatives as the excess of the Lorentz power above 1. Optical-constant deconvolution for a Lorentz-power slit function can be also performed through Taylor series expansion from the shifted optical constants, which starts in the second-order term. The convolution of a single Lorentz function with a Lorentz power, as an approach to a Voigt profile, can be also addressed with the present scheme.

Appendix A. Sum rules on optical constants at complex energies

Sum rules can be applied to optical constants at energies shifted to C^+. Inertial sum rule results in [16,17]:

$$\int_0^\infty \{Re[\tilde{\varepsilon}(E+ia)]-1\}dE = 0 \tag{A1}$$

It is immediately obtained along contour No. 1 of Fig. 1(b). The circular arc (in the limit of infinite radius) provides no contribution to the integral due to the asymptotic convergence requirement. For a>0, the integral is 0 even for Drude-like conductors.

Regarding f-sum rule, it can be immediately extended too:

$$\int_0^\infty EIm[\tilde{\varepsilon}(E+ia)]dE = \frac{1}{2}\pi E_p^2 \tag{A2}$$

where $E_p=\hbar\omega_p$ stands for the plasma energy of the material and involves its total number of electrons. The ordinary f-sum rule is represented by Eq. (A2) when a=0. Here there is no divergence for conductors due to the factor E in the integrand, so that Eq. (A2) is valid both for conductors and non-conductors, as it is the case for the standard f-sum rule. To demonstrate it, let us integrate $E[\tilde{\varepsilon}(E)-1]$ along contour No. 2 in Fig. 1(b). The two short lines at the edges, having a finite length, provide no contribution to the integral due to the standard requirements of $\tilde{\varepsilon}$ on asymptotic convergence and parity. Hence integration of the imaginary part along a shifted axis equals integration on the real axis (when the path has the same direction, such as from -∞ to +∞) since there is no singularity.

Summarizing, the two ordinary sum rules are also satisfied on any straight line parallel to the real axis, since no assumption was made for a to be small.

Let us see more sum rules. By following contour 3 of Fig. 1(b) and by taking the radius of the 90°-arc to infinity, we get to the following identity:

$$\int_E^\infty [\tilde{\varepsilon}(E'+ia)-1]dE' = i\int_a^\infty [\tilde{\varepsilon}(E+ia')-1]da' \tag{A3}$$

It is valid also for a=0. For non-conductors, it is valid also for E=0; otherwise, an extra term originated at the pole needs to be added. Following contour 4 of Fig. 1(b) and by upwards extending the vertical lines to infinity, the following identity is obtained:

$$\int_0^E Re[\tilde{\varepsilon}(E'+ia)-1]dE' = \int_a^\infty Im[\tilde{\varepsilon}(E+ia')]da' \tag{A4}$$

where parity of the dielectric function was applied. Eq. (A4) can be also obtained from Eqs. (A1) and (A3). For non-conductors, it is valid also for a=0. All the above sum rules also apply on $\tilde{N}(E)$, which does not required an extra term for conductors as expressed above.

Appendix B. The convolution with Lorentzian function as the unique extension in the complex plane

Equation (1) states that the extension of the optical-constant function from the real axis to C^+ consists in the convolution with a Lorentzian function. This results in a smoothening of any feature present on the real axis, as in the example of Fig. 2. Let us answer the following

question for an optical-constant function: could there exist a different way to span C^+, such as with a more abrupt behaviour?

Let $\tilde{F}_1(E)$ and $\tilde{F}_2(E)$ be two optical-constant functions which are identical on the real axis except, at most, at some discrete energies (i.e., a null set), where they have a finite difference; hence $\tilde{D}(E) = \tilde{F}_2(E) - \tilde{F}_1(E)$ is mostly 0 on the real plane. Let $\tilde{G}_1(E + ia)$ and $\tilde{G}_2(E + ia)$ be the analytic functions in C^+ whose boundary limits on the real energy axis are $\tilde{F}_1(E)$ and $\tilde{F}_2(E)$, respectively, so that:

$$\tilde{F}_i(E) = \lim_{a \to 0} \tilde{G}_i(E + ia) \text{ for } i=1,2 \tag{B1}$$

$\tilde{G}_1$ and $\tilde{G}_2$ must be analytic in C^+ if they are to describe the optical constants of a material. Let us apply Cauchy integration on $\tilde{H} = \tilde{G}_2 - \tilde{G}_1$:

$$\oint \frac{\tilde{H}(z)}{z - z_0} dz = 2\pi i \tilde{H}(z_0) \tag{B2}$$

where z_0 is any complex number in C^+. We perform the integration along the same contour plotted in Fig. 1.

$$2\pi i \tilde{H}(z_0) = \int_{-\infty}^{\infty} \frac{\tilde{D}(E')}{E' - z_0} dE' + \lim_{\rho \to \infty} \int_0^{\pi} \frac{\tilde{H}(\rho e^{i\theta})}{\rho e^{i\theta} - z_0} i\rho e^{i\theta} d\theta \tag{B3}$$

In the first term of the right-hand side, $\tilde{H}$ was replaced with $\tilde{D}$ since the latter must be the limit of the former when Im(z) approaches 0, as stated by Titchmarsh theorem; such term is zero (path 1 in the contour) because the integrand is zero except for a null set of energies. The second term of the right-hand side (path 2 in the contour) is also zero, because both $\tilde{G}_1$ and $\tilde{G}_2$ individually integrated along such a path would result in a null integral due to the standard convergence requirements on the optical-constant function, as stated by Titchmarsh theorem. When the material is a Drude-like conductor, the optical function has a pole at E=0; however, $\tilde{F}_1$ and $\tilde{F}_2$ were assumed essentially identical, so that the pole is avoided by subtraction, and hence the integral of path 1 can be performed all the way through E=0.

The above result means $\tilde{H}(z_0) = 0$ for any arbitrary z_0 in C^+, hence $\tilde{G}_2(z_0) = \tilde{G}_1(z_0)$ in C^+, both for dielectrics, Drude-like conductors, and materials with a null set of finite discontinuities on the real axis. If we now assume that $\tilde{G}_1$ has been obtained through Lorentzian convolution using Eq. (1) [or using Eq. (3) for a Drude-like conductor], then the present result states that any function $\tilde{G}_2$ describing the optical constants of a material in C^+ must equal the Lorentzian convolution of its boundary on the real axis. Therefore, moving upwards in C^+ necessarily results in the smoothening expressed by Eqs (1) or (3). In other words, the optical constants of a material on the real axis cannot be the limit of two different analytic functions in C^+. The present unicity result reminds the concept of unique analytic continuation. However, analytic continuation starts with an analytic function on an open set of the complex plane [18], whereas the real axis is not an open set in C; furthermore, in general the optical constants of a material at real energies $\tilde{F}_1$ and $\tilde{F}_2$ are not strictly required to be analytic on the real axis. In the present case, a result that resembles the unicity of analytic continuation has been obtained starting with a non-necessarily analytic function defined in a non-open set because of the additional requirements by virtue of Titchmarsh theorem: the function must be analytic in C^+, the function on the real axis must be the limit of the function in C^+, and it must converge fast enough.

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5.2 Disclosures

The authors declare no conflicts of interest.

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