Alternative approach to the derivation of dispersion relations for optical constants

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Abstract
The derivation of dispersion relations for linear optical constants is considered
starting from the representation of an optical property as a Herglotz function.
One form of the Kramers–Kronig relations is determined directly and the second
is obtained using elementary properties of the Hilbert transform. Application
to the complex refractive index is considered.

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1. Introduction

There continues to be considerable interest in the refinement and utilization of dispersion
relations. Recent advances have included applications to optical properties [1–12], to problems
in acoustics [13–17] and other developments [18–22].

The historical approach to the derivation of dispersion relations for optical properties
relied on establishing that the complex optical function was analytic in the upper half of the
complex angular frequency plane. Utilizing physical models of an optical property allowed
the asymptotic behaviour of the optical property for large complex angular frequencies to
be determined. From the analyticity and asymptotic behaviour, a pair of Hilbert transform
relations can be established connecting the real and imaginary parts of a complex optical
property. The Hilbert transform pair is then converted to a positive frequency spectral range
by employing crossing symmetry conditions for the optical property. Details on the approach
can be found in [2].

The purpose of this paper is to examine the development of dispersion relations for optical
constants, starting from the properties of Herglotz functions. The standard approach to the
derivation of the dispersion relations proceeds directly from the notion of causality and a
contour integration technique. The causality requirement also enters the present work, by
defining the domain for which the optical property is an analytic function.

2. Some basic properties of the Hilbert transform

A number of properties of the Hilbert transform will be required in the following, and these are collected together in this section. Discussion of these results can be found in a number of sources, see, for example, [23, 24]. The Hilbert transform on $\mathbb{R}$, the real line, is defined by

$$Hf(x) = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{f(y)\,dy}{x-y}, \qquad \text{for} \quad x \in \mathbb{R}, \tag{1}$$

where P signifies that the Cauchy principal value is taken. The Hilbert transform operator will be denoted by H throughout this work. Equation (1) is often written in the form

$$Hf(x) = \lim_{\varepsilon \to 0+} H_\varepsilon f(x), \tag{2}$$

where

$$H_\varepsilon f(x) = \frac{1}{\pi} \int_{|x-t|>\varepsilon} \frac{f(t)}{x-t}\,dt. \tag{3}$$

The function $H_\varepsilon f$ is sometimes referred to as the *truncated* Hilbert transform of f. From the definition in equation (1), it is straightforward to show that the Hilbert transform of an even function yields an odd function, and the Hilbert transform of an odd function gives an even function. This is the *parity* property of the Hilbert transform operator.

The class of Lebesgue integrable functions on the real line will be denoted by $L^p(\mathbb{R})$ and abbreviated when there is no risk of confusion by L^p. If f belongs to the class $L^p(\mathbb{R})$ for $1 < p < \infty$, then

$$\int_{-\infty}^{\infty} |Hf(x)|^p \,dx \leqslant \{\Re_p\}^p \int_{-\infty}^{\infty} |f(x)|^p \,dx, \tag{4}$$

where $\Re_p$ is the Riesz constant, which depends only on p. For $p = 2$, $\Re_p = 1$ and equality holds in equation (4). The best value of the constant $\Re_p$ for $1 < p < \infty$ is known to be

$$\Re_p = \begin{cases} \tan\left(\frac{\pi}{2p}\right), & 1 < p \leqslant 2 \\ \cot\left(\frac{\pi}{2p}\right), & 2 \leqslant p < \infty. \end{cases} \tag{5}$$

The constant is *best* in the sense that for any particular f belonging to the class $L^p(\mathbb{R})$, there is no constant less than $\Re_p$ for which equation (4) holds true.

An important property of the Hilbert transform is the inversion property, also referred to as the iteration property. If $Hf(x) = g(x)$ implies $Hg(x) = -f(x)$, then

$$H^2 f(x) \equiv H(Hf)(x) = -f(x), \quad \text{a.e.} \tag{6}$$

The abbreviation a.e. stands for almost everywhere. If $f \in L^p$ for $p > 1$, then $g \equiv Hf \in L^p$. Using this result, it is clear that functions belonging to the class L^p (for $p > 1$) satisfy equation (6). If an additional assumption is made for the case $p = 1$, that is, if $f \in L$ and $Hf \in L$, then functions of the class L are also included. It is not difficult to find examples for which $f \in L$ but $Hf \notin L$, hence the requirement in the preceding statement that both f and $Hf \in L$.

If $g(x) = Hf(x)$, then

$$H\{xf(x)\} = xg(x) - \frac{1}{\pi} \int_{-\infty}^{\infty} f(t)\,dt, \tag{7}$$

which is the *moment formula* for the Hilbert transform.

An important example is the Hilbert transform of a constant c. The result is

$$Hc = 0. \tag{8}$$

3. Representation of Herglotz functions

In this section an alternative approach to the derivation of the Kramers–Kronig relations is considered. The derivation is carried forward with a fairly general optical property $\mathcal{O}$, with the principal restriction being that the imaginary part of $\mathcal{O}$ satisfies the condition

$$\operatorname{Im} \mathcal{O}(\omega) \geqslant 0. \tag{9}$$

Since the imaginary component of $\mathcal{O}$ typically represents the dissipative behaviour of the system, then the preceding result is satisfied. Some particular choices for the function $\mathcal{O}$ will be considered in the following section. The approach revolves around the use of Herglotz functions, which are now introduced.

The following result of Herglotz [25] is key in what follows. If $g(w)$ is analytic in $|w| < 1$, and $\operatorname{Im} g(w) \geqslant 0$ in the same domain, then the function $g(w)$ admits the integral representation

$$g(w) = \mathrm{i} \int_0^{2\pi} \frac{\mathrm{e}^{\mathrm{i}\theta} + w}{\mathrm{e}^{\mathrm{i}\theta} - w} \, \mathrm{d}\beta(\theta) + C, \tag{10}$$

where $\beta(\theta)$ is a non-decreasing bounded function and C is a real constant. A function satisfying these two conditions is called a Herglotz function. A conformal mapping using

$$z = \mathrm{i} \frac{1 + w}{1 - w} \tag{11}$$

converts the interior of the disc to the upper half complex plane. Employing the change of variables

$$t = -\cot \frac{\theta}{2}, \qquad \frac{t - \mathrm{i}}{t + \mathrm{i}} = \mathrm{e}^{\mathrm{i}\theta}, \tag{12}$$

using $f(z) = g(w)$ and $\alpha(t) = \beta(\theta)$, leads to the result

$$f(z) = Az + \int_{-\infty}^{\infty} \frac{1 + tz}{t - z} \, \mathrm{d}\alpha(t) + C, \tag{13}$$

where A is a positive real constant. The first term on the right-hand side of this result arises from the possible jump of $\beta(\theta)$ at $\theta = 0$ and $\theta = 2\pi$; see [26–28] for some further comments on the derivation. Additional background on Herglotz functions can be found in the sources [29–32]. The result in equation (13) is used to arrive at a Kramers–Kronig connection between the real and imaginary parts of a general linear optical property. For general background on the Kramers–Kronig relations for optical properties, the reader is directed to the books [2, 12].

Let $\omega_z = \omega_r + \mathrm{i}\omega_i$ denote a complex angular frequency. If $\mathcal{O}(\omega_z)$ satisfies equation (9) and is analytic in the upper half complex frequency plane, then it is a Herglotz function, so that

$$
\begin{aligned}
\mathcal{O}(\omega_z) &= A\omega_z + \int_{-\infty}^{\infty} \frac{1 + \omega'\omega_z}{\omega' - \omega_z} \, \mathrm{d}\alpha(\omega') + C \\
&= A(\omega_r + \mathrm{i}\omega_i) + C + \int_{-\infty}^{\infty} \frac{\left[(1 + \omega'\omega_r)(\omega' - \omega_r) - \omega'\omega_i^2 \right] + \mathrm{i}\omega_i(\omega'^2 + 1)}{(\omega' - \omega_r)^2 + \omega_i^2} \, \mathrm{d}\alpha(\omega').
\end{aligned} \tag{14}
$$

The effects of spatial dispersion for the optical property are ignored. Furthermore, consideration is restricted to isotropic media; the non-isotropic case can be dealt with in a similar fashion by considering the appropriate tensor components of the optical property. In equation (14) and the following development, the frequency factors are rendered dimensionless by dividing each by 1 Hz, when this is necessary for dimensional considerations. If the constraint that $\alpha(\omega)$ is differentiable everywhere is imposed, then the Stieltjes integral can be

written using $d\alpha(\omega) = \alpha'(\omega)\,d\omega$. Taking the limit $\omega_i \to 0$, using the definition of the Dirac delta distribution

$$\pi\delta(x) = \lim_{a \to 0} \frac{a}{a^2 + x^2}, \tag{15}$$

and denoting ω_r by ω, equation (14) can be recast as

$$
\begin{aligned}
\mathcal{O}(\omega) &= A\omega + C + \lim_{\omega_i \to 0} \int_{-\infty}^{\infty} \frac{\left[(1 + \omega'\omega)(\omega' - \omega) - \omega'\omega_i^2\right]}{(\omega' - \omega)^2 + \omega_i^2}\alpha'(\omega')\,d\omega' \\
&\quad + \lim_{\omega_i \to 0} \int_{-\infty}^{\infty} \frac{\mathrm{i}\omega_i(\omega'^2 + 1)}{(\omega' - \omega)^2 + \omega_i^2}\alpha'(\omega')\,d\omega' \\
&= A\omega + C + P \int_{-\infty}^{\infty} \frac{1 + \omega'\omega}{\omega' - \omega}\alpha'(\omega')\,d\omega' \\
&\quad + \mathrm{i}\pi \int_{-\infty}^{\infty} \delta(\omega' - \omega)[\omega'^2 + 1]\alpha'(\omega')\,d\omega' \\
&= A\omega + C + P \int_{-\infty}^{\infty} \frac{1 + \omega'\omega}{\omega' - \omega}\alpha'(\omega')\,d\omega' + \mathrm{i}\pi(1 + \omega^2)\alpha'(\omega). \tag{16}
\end{aligned}
$$

The imaginary part of this equation yields

$$\operatorname{Im}\mathcal{O}(\omega) = \pi(1 + \omega^2)\alpha'(\omega). \tag{17}$$

From the preceding formula $\alpha'(\omega)$ is determined, and then inserting the result into equation (16) leads to

$$\operatorname{Re}\mathcal{O}(\omega) = A\omega + C - \frac{1}{\pi}P \int_{-\infty}^{\infty} \frac{(1 + \omega'\omega)\operatorname{Im}\mathcal{O}(\omega')\,d\omega'}{(\omega - \omega')(1 + \omega'^2)}. \tag{18}$$

Since only continuous linear optical properties are of interest in the present discussion, equation (17) provides the necessary justification for imposing the previously employed constraint that $\alpha(\omega)$ is differentiable everywhere.

Suppose that $\mathcal{O}(\omega)$ satisfies a crossing symmetry constraint of the form

$$\mathcal{O}(-\omega) = -\mathcal{O}^*(\omega), \tag{19}$$

where $*$ denotes the complex conjugate. The imaginary component of $\mathcal{O}(\omega)$ is therefore an even function and the real part is an odd function. Equation (18) can be recast as

$$\operatorname{Re}\mathcal{O}(\omega) = A\omega + C - \frac{2\omega}{\pi}P \int_{0}^{\infty} \frac{\operatorname{Im}\mathcal{O}(\omega')\,d\omega'}{\omega^2 - \omega'^2}. \tag{20}$$

If instead of equation (19) assume $\mathcal{O}(\omega)$ satisfies the crossing symmetry constraint

$$\mathcal{O}(-\omega) = \mathcal{O}^*(\omega), \tag{21}$$

then equation (18) reduces to

$$\operatorname{Re}\mathcal{O}(\omega) = A\omega + C - \frac{2(1 + \omega^2)}{\pi}P \int_{0}^{\infty} \frac{\omega'\operatorname{Im}\mathcal{O}(\omega')\,d\omega'}{(\omega^2 - \omega'^2)(1 + \omega'^2)}. \tag{22}$$

Resolving the integrand of the last integral into partial fractions leads to

$$\operatorname{Re}\mathcal{O}(\omega) = A\omega + C - \frac{2}{\pi}\int_{0}^{\infty} \frac{\omega'\operatorname{Im}\mathcal{O}(\omega')\,d\omega'}{1 + \omega'^2} - \frac{2}{\pi}P \int_{0}^{\infty} \frac{\omega'\operatorname{Im}\mathcal{O}(\omega')\,d\omega'}{\omega^2 - \omega'^2}. \tag{23}$$

The constants in equation (20) can be fixed as follows. The limit $\omega \to 0$ leads to

$$C = \operatorname{Re}\mathcal{O}(0), \tag{24}$$

and dividing both sides of equation (20) by ω and taking the limit $\omega \to \infty$ gives

$$A = \left. \frac{\text{Re } \mathcal{O}(\omega)}{\omega} \right|_{\omega \to \infty}. \tag{25}$$

Inserting these results into equation (20) leads to

$$\text{Re} \left\{ \mathcal{O}(\omega) - \mathcal{O}(0) - \omega \left\{ \frac{\mathcal{O}(\omega)}{\omega} \right\}_{\omega \to \infty} \right\} = -\frac{2\omega}{\pi} P \int_0^\infty \frac{\text{Im } \mathcal{O}(\omega') \, \mathrm{d}\omega'}{\omega^2 - \omega'^2}. \tag{26}$$

In a similar manner equation (23) can be recast as

$$
\begin{aligned}
\text{Re} &\left\{ \mathcal{O}(\omega) - \mathcal{O}(0) - \omega \left\{ \frac{\mathcal{O}(\omega)}{\omega} \right\}_{\omega \to \infty} \right\} \\
&= -\frac{2}{\pi} P \int_0^\infty \frac{\omega' \, \text{Im } \mathcal{O}(\omega') \, \mathrm{d}\omega'}{\omega^2 - \omega'^2} - \frac{2}{\pi} P \int_0^\infty \frac{\text{Im } \mathcal{O}(\omega') \, \mathrm{d}\omega'}{\omega'} \\
&= -\frac{2\omega^2}{\pi} P \int_0^\infty \frac{\text{Im } \mathcal{O}(\omega') \, \mathrm{d}\omega'}{\omega'(\omega^2 - \omega'^2)},
\end{aligned} \tag{27}
$$

and it is assumed that the integral $P \int_0^\infty \frac{\text{Im } \mathcal{O}(\omega') \, \mathrm{d}\omega'}{\omega'}$ is convergent.

In an important paper, Weaver and Pao [33] considered in a similar analysis the use of Herglotz functions for the discussion of the complex wave number in linear wave propagation in homogeneous and inhomogeneous media. In their work, these authors point out that it is not clear why the α term should be treated as differentiable. This would particularly be the case for inhomogeneous media. Another issue raised by these authors is that only one of the Kramers–Kronig pairs is obtained from this type of analysis. Using a lengthy complex variable analysis, they obtain the second dispersion relationship for the complex wave number. In the following section, we show that some basic properties of the Hilbert transform can be employed to obtain the second dispersion relationship in a straightforward manner. A justification for the differentiability of the α function is also given.

4. Applications

This section considers some applications of the results just obtained. The focus will be linear optical properties, of which the refractive index is employed as a representative example. An identical approach can be employed to obtain ordinary dispersion relations for nonlinear optical properties in the case of harmonic generation. For the case of multiple-variable dispersion relations for nonlinear optical properties, the situation becomes rather complicated, and this topic is not pursued in the present work. The reader interested in investigating the approaches to dispersion relations for nonlinear properties might consult the references [2, 12, 34–44, 46, 47].

Let $\mathcal{O}(\omega) = N(\omega) - 1$, where $N(\omega)$ is the complex refractive index, which is expressed in terms of the normal refractive index $n(\omega)$ by the relation $N(\omega) = n(\omega) + i\kappa(\omega)$ and $\kappa(\omega)$ determines the dissipation in the system. The complex refractive index is analytic in the upper half complex frequency plane, and $\text{Im } \mathcal{O}(\omega) = \kappa(\omega)$, which satisfies equation (9). With this particular choice of $\mathcal{O}(\omega)$, it is clear from equation (17) why $\alpha(\omega)$ can be treated as a differentiable function, since $\kappa(\omega)$ is differentiable for all angular frequencies for an insulator. The exceptional point for conductors is $\omega = 0$. A similar situation applies for the choices given later for $\mathcal{O}(\omega)$; however the point $\omega = 0$ for the latter two cases no longer presents any difficulties for conductors. The function α' is positive because of energy dissipation in the system, a result following directly from equation (17). From the result that

$\text{Re}\,\mathcal{O}(\omega) = n(\omega) - 1$, which has the asymptotic behaviour $n(\omega) - 1 = O(\omega^{-2})$ as $\omega \to \infty$, then

$$\omega^{-1}\,\text{Re}\,\mathcal{O}(\omega)|_{\omega \to \infty} = 0. \tag{28}$$

For the choice $\mathcal{O}(\omega) = N(\omega) - 1$, the crossing symmetry condition given by equation (21) applies, and from equation (27) it follows that

$$n(\omega) - n(0) = -\frac{2}{\pi} P \int_0^\infty \frac{\omega' \kappa(\omega')\,\mathrm{d}\omega'}{\omega^2 - \omega'^2} - \frac{2}{\pi} \int_0^\infty \frac{\kappa(\omega')\,\mathrm{d}\omega'}{\omega'}. \tag{29}$$

In this form, the result applies to insulators; conductors are excluded because of the behaviour of $\kappa(\omega)$ as $\omega \to 0$. Equation (29) can be rewritten as

$$n(\omega) - n(0) = -\frac{2\omega^2}{\pi} P \int_0^\infty \frac{\kappa(\omega')\,\mathrm{d}\omega'}{\omega'(\omega^2 - \omega'^2)}, \tag{30}$$

which is a single subtracted form of the dispersion relation for the refractive index. Singly, and more generally, multiply-subtracted dispersion relations lead to improved convergence for the numerical evaluation of optical constants [12].

Consider the choice $\mathcal{O}(\omega) = \omega N(\omega)$, then equation (9) is satisfied, $\text{Re}\,\mathcal{O}(0) = 0$ and $\omega^{-1}\,\text{Re}\,\mathcal{O}(\omega)|_{\omega \to \infty} = \lim_{\omega \to \infty} n(\omega) = 1$. Since equation (19) applies in this case, equation (26) leads to

$$n(\omega) - 1 = -\frac{2}{\pi} P \int_0^\infty \frac{\omega' \kappa(\omega')\,\mathrm{d}\omega'}{\omega^2 - \omega'^2}, \tag{31}$$

which is the standard form of the Kramers–Kronig connection relating $n(\omega) - 1$ to $\kappa(\omega)$. The alternative choice $\mathcal{O}(\omega) = \omega\{N(\omega) - 1\}$ satisfies equation (9), $\text{Re}\,\mathcal{O}(0) = 0$ and $\omega^{-1}\,\text{Re}\,\mathcal{O}(\omega)|_{\omega \to \infty} = 0$, with the result that equation (31) is obtained from equation (26).

The Kramers–Kronig relations come in pairs, and to obtain the second formula the following straightforward approach using some of the basic properties of the Hilbert transform can be employed. Equation (18) can be written as

$$\text{Re}\,\mathcal{O}(\omega) = A\omega + C - Hf(\omega) - \omega Hg(\omega), \tag{32}$$

with

$$f(\omega) = \frac{\text{Im}\,\mathcal{O}(\omega)}{(1 + \omega^2)}, \tag{33}$$

and

$$g(\omega) = \omega f(\omega). \tag{34}$$

The following constraints are imposed: (i) $A = 0$, which supposes that $\text{Re}\,\mathcal{O}(\omega)$ has a suitable asymptotic behaviour as $\omega \to \infty$; (ii) $\mathcal{O}(-\omega) = \mathcal{O}^*(\omega)$, which is a common situation for many optical properties; (iii) $g(t) \in L^p(\mathbb{R})$ for $1 < p < \infty$ and (iv) $\text{Re}\,\mathcal{O}(\omega) \in L^q(\mathbb{R})$ for $1 \leqslant q < \infty$. The latter requirement is sufficient to ensure that the Hilbert transform of $\text{Re}\,\mathcal{O}(\omega)$ exists; in order to ensure that it is bounded, the condition on q must be changed to $1 < q < \infty$. From equation (32) it follows that

$$H\,\text{Re}\,\mathcal{O}(\omega) = H[C] - H^2 f(\omega) - H[\omega Hg(\omega)]. \tag{35}$$

Let $h(\omega) = Hg(\omega)$, then the moment formula of the Hilbert transform can be applied to give

$$H[\omega h(\omega)] = \omega Hh(\omega) - \frac{1}{\pi} \int_{-\infty}^\infty h(\omega)\,\mathrm{d}\omega. \tag{36}$$

If condition (ii) applies, then $\text{Im}\,\mathcal{O}(\omega)$ is an odd function and so from equations (33) and (34) $g(\omega)$ is an even function. Recalling that the Hilbert transform of an even function is an odd

function, then $h(\omega)$ is odd, and so equation (36) simplifies to give

$$H[\omega h(\omega)] = \omega H^2 g(\omega). \tag{37}$$

Condition (iii) allows the inversion formula of the Hilbert transform to be applied as

$$H^2 f(\omega) = -f(\omega), \qquad H^2 g(\omega) = -g(\omega), \tag{38}$$

and recalling that the Hilbert transform of a constant is zero, equation (8), then equation (35) can be rewritten as

$$H \operatorname{Re} \mathcal{O}(\omega) = f(\omega) + \omega g(\omega), \tag{39}$$

which simplifies on inserting the definitions of f and g to yield

$$\operatorname{Im} \mathcal{O}(\omega) = H \operatorname{Re} \mathcal{O}(\omega). \tag{40}$$

Employing condition (ii) gives

$$\operatorname{Im} \mathcal{O}(\omega) = \frac{2\omega}{\pi} P \int_0^\infty \frac{\operatorname{Re} \mathcal{O}(\omega')\, \mathrm{d}\omega'}{\omega^2 - \omega'^2}, \tag{41}$$

which is the standard form of one of the Kramers–Kronig relations.

The second formula of the Kramers–Kronig pair can be obtained in a different manner. On taking the real part of equation (16), multiplying both sides of the resulting equation by $(1 + \omega^2)^{-1}$ and applying the Hilbert transform yields

$$\operatorname{Im} \mathcal{O}(\omega) = c_1 + c_2\omega + H[\operatorname{Re} \mathcal{O}(\omega)], \tag{42}$$

where c_1 and c_2 are constants. These constants can be fixed in the same manner as described previously. If equation (19) holds, then

$$\operatorname{Im} \mathcal{O}(\omega) = \operatorname{Im} \mathcal{O}(0) + \omega \left[\frac{\operatorname{Im} \mathcal{O}(\omega)}{\omega} \right]_{\omega \to \infty} + \frac{2}{\pi} P \int_0^\infty \omega' \operatorname{Re} \mathcal{O}(\omega') \left\{ \frac{1}{\omega^2 - \omega'^2} + \frac{1}{\omega'^2} \right\} \mathrm{d}\omega', \tag{43}$$

which assumes that $\int_0^\infty \frac{\operatorname{Re} \mathcal{O}(\omega')\, \mathrm{d}\omega'}{\omega'}$ is convergent, and if equation (21) applies, then

$$\operatorname{Im} \mathcal{O}(\omega) = \operatorname{Im} \mathcal{O}(0) + \omega \left[\frac{\operatorname{Im} \mathcal{O}(\omega)}{\omega} \right]_{\omega \to \infty} + \frac{2\omega}{\pi} P \int_0^\infty \frac{\operatorname{Re} \mathcal{O}(\omega')\, \mathrm{d}\omega'}{\omega^2 - \omega'^2}, \tag{44}$$

which assumes that $\lim_{\omega \to 0} \left\{ \omega P \int_0^\infty \frac{\operatorname{Re} \mathcal{O}(\omega')\, \mathrm{d}\omega'}{\omega^2 - \omega'^2} \right\}$ is zero.

As an application of equation (41) let

$$\mathcal{O}(\omega) = N(\omega) - 1, \tag{45}$$

and note that

$$\omega^{-1} \operatorname{Re} \mathcal{O}(\omega)|_{\omega \to \infty} = \omega^{-1} \{ n(\omega) - 1 \}|_{\omega \to \infty} = 0. \tag{46}$$

Conditions (i)–(iv) are satisfied, so that equation (41) yields

$$\kappa(\omega) = \frac{2\omega}{\pi} P \int_0^\infty \frac{\{ n(\omega') - 1 \}\, \mathrm{d}\omega'}{\omega^2 - \omega'^2}, \tag{47}$$

which is the well-known Kramers–Kronig connection between $\kappa(\omega)$ and $n(\omega) - 1$.

To treat the nonlinear case for harmonic generation the function $\mathcal{O}(\omega)$ is identified with the nth order susceptibility $\chi^{(n)}(p_1\omega, p_2\omega, \ldots, p_n\omega)$, where p_k are suitably selected integer values. The development of dispersion relations for $\chi^{(n)}(p_1\omega, p_2\omega, \ldots, p_n\omega)$ runs along the same lines as discussed for the refractive index.

5. Discussion

The reader may have wondered if this approach to the dispersion relations for the optical constants avoids the use of causal arguments. The answer is no, since it is necessary to work with an optical function that is analytic in the upper half of the complex frequency plane. The analyticity of the optical functions follows from a causality argument [47]. In the case of a choice like $\mathcal{O}(\omega) = N(\omega) - 1$, a much more involved argument is required, since $N(\omega) - 1$ does not correspond to any physically realizable response function [32].

Appropriate knowledge of the asymptotic behaviour of the optical property as $\omega \to \infty$ is also required. Realistic physical models of the behaviour of the optical properties provide the necessary information on the real frequency axis. The Phragmén–Lindelöf theorem of complex analysis allows the asymptotic behaviour in the upper half of the complex frequency plane to be determined from the behaviour on the real axis.

By appropriate selection of the function $\mathcal{O}$, dispersion relations for powers of optical properties can be established. The resulting formulae typically have less practical value for data analysis, since the real and imaginary components of the optical constant are intermingled. However, such formulae can be utilized to derive sum rule constraints for optical properties.

The conformal transformation step taken at equation (11) can be circumvented. It is possible to opt for a trigonometric expansion approach, leading to a conjugate series representation for the real and imaginary parts of an optical property [48–50].

In summary, an alternative to the standard approach of using contour integration and the Cauchy integral formula for deriving the Kramers–Kronig relations for linear optical properties has been considered. The derivation still exploits the analytic behaviour of the property under consideration. Only elementary properties of the Hilbert transform are required to obtain the second formula of the Kramers–Kronig pair, from the Herglotz representation of the optical property as a Herglotz function.

Acknowledgment

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