Sum rules for the optical constants

Frederick W. King*

Chemistry Department, Queen's University, Kingston, Ontario, Canada K7L 3N6
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A number of sum rules for the optical constants, in particular, the refractive index of a nonconducting medium, are obtained. Some of the sum rule constraints are highly damped for large frequencies, exponentially in one particular case. Formal integral relationships for the index of refraction at complex frequencies are presented. Sum rules based on known experimental points for which $n(\omega)-1$ has zeros are indicated. An outline of a modified derivation of some recently presented sum rules for the optical constants is given.

I. INTRODUCTION

The Kramers-Kronig relations for linear optical properties have led to a number of useful sum rule constraints which these properties must satisfy. The underlying conditions for which these sum rules should hold are very general. The physical basis, the causality condition, determines the domain in which the generalized optical property is holomorphic, from which the Kramers-Kronig relations may then be deduced. A number of sum rules have been recently derived by employing the Kramers-Kronig relations and the appropriate asymptotic behavior of the particular optical properties at large frequencies. This asymptotic behavior may be readily determined by assuming at sufficiently high frequencies, the medium responds like a free electron gas.

This paper is concerned with the investigation of principally, the constraints on the generalized refractive index for a nonconducting medium. A number of surprisingly simple, but potentially very useful sum rules have been obtained for the generalized refractive index by Altarelli *et al.*[1,2] and Villani and Zimerman.[3] The simplest of these relations are

$$\int_0^\infty \omega\kappa(\omega)\,d\omega=\frac{\pi}{4}\,\omega_p^2, \tag{1}$$

$$\int_0^\infty [n(\omega)-1]\,d\omega=0, \tag{2}$$

$$\int_0^\infty [\,\omega\kappa(\omega)[n(\omega)-1]\,d\omega=0, \tag{3}$$

$$\cos\pi\beta\int_a^\infty \frac{[n(\omega)-1]\,d\omega}{(\omega-a)^\beta(\omega+a)^\beta}-\sin\pi\beta\int_a^\infty\frac{\kappa(\omega)\,d\omega}{(\omega-a)^\beta(\omega+a)^\beta}+\int_0^a\frac{[n(\omega)-1]\,d\omega}{(a-\omega)^\beta(a+\omega)^\beta}=0,\quad -\tfrac{1}{2}<\beta<1, \tag{4}$$

$$\int_0^\infty \omega^2[n(\omega)-1]^2\,d\omega=\int_0^\infty\omega^2\kappa^2(\omega)\,d\omega, \tag{5}$$

where $n(\omega)$ and $\kappa(\omega)$ are the real and imaginary parts, respectively, of the generalized refractive index, and ω_p is the plasma frequency. The first of these relationships, Eq. (1) is the well-known f sum rule, Eqs. (2) and (3) being derived by Altarelli *et al.*, and are designated as the ADNS sum rules, and the expressions given by Eqs. (4) and (5) have been obtained by Villani and Zimerman (designated VZ sum rules). Equation (4) contains the ADNS sum rule equation (2), as a special case; $\beta=0$.

In Sec. II we outline how some of the above sum rules may be obtained in an alternative manner from the Kramers-Kronig relations. Section III presents a number of formal relationships connecting the generalized refractive index on the complex imaginary frequency axis, with the refractive index at real frequencies. In Sec. IV we outline some strikingly simple and apparently previously unnoticed sum rules based on the zeros of $n(\omega)-1$. Section V deals with some generalizations of the above sum rules, which have the desirable property of being highly damped at large frequencies. Finally, in Sec. VI, we discuss possible extensions and limitations of the sum rules derived herein.

II. DERIVATION OF SUM RULES FROM THE KRAMERS-KRONIG RELATIONS

The basic equations from which most of the sum rules for the refractive index have been derived are the Kramers-Kronig relations

$$n(\omega_0)-1=\frac{2}{\pi}P\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{\omega^2-\omega_0^2}, \tag{6}$$

$$\kappa(\omega_0)=-\frac{2\omega_0}{\pi}P\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{\omega^2-\omega_0^2}. \tag{7}$$

From Eq. (6), it follows immediately that

$$\int_0^\infty[n(\omega')-1]\,d\omega'=\frac{2}{\pi}\int_0^\infty d\omega'\,P\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{\omega^2-\omega'^2} \tag{8}$$

and on interchanging the order of integration

$$\int_0^\infty[n(\omega')-1]\,d\omega'=\frac{2}{\pi}\int_0^\infty\omega\kappa(\omega)\,d\omega\,P\int_0^\infty\frac{d\omega'}{\omega^2-\omega'^2}, \tag{9}$$

and noting the relationship

$$P\int_0^\infty\frac{d\omega'}{\omega^2-\omega'^2}=-\frac{\pi^2}{4}\,\delta(\omega), \tag{10}$$

then

$$\int_0^\infty[n(\omega')-1]\,d\omega'=-\frac{\pi}{2}\int_0^\infty\omega\kappa(\omega)\delta(\omega)\,d\omega=0, \tag{11}$$

which is the ADNS sum rule, Eq. (2). The conditions for the interchange of the order of integration to obtain Eq. (9) need to be scrutinized carefully. The necessary requirement is that the integrand, a function of ω and ω', be summable over the plane $-\infty<\omega<\infty$, $-\infty<\omega'<\infty$. Similarly, from the Kramers-Kronig relation (7),

$$\int_0^\infty \frac{\kappa(\omega')\,d\omega'}{\omega'} = -\frac{2}{\pi}\int_0^\infty d\omega'\, P\int_0^\infty \frac{[n(\omega)-1]\,d\omega}{\omega^2-\omega'^2}, \qquad (12)$$

and by interchanging the order of integration, and employing Eq. (10), leads to the result

$$\int_0^\infty \frac{\kappa(\omega)\,d\omega}{\omega} = \frac{\pi}{2}[n(0)-1]. \qquad (13)$$

Depending on the behavior of $\kappa(\omega)$ as $\omega \to 0$, the integral in Eq. (13) may exist only in the sense of a principal values. Equation (13) is, of course, just the limit as $\omega_0 \to 0$ of the Kramers-Kronig relation, Eq. (6). It is worth pointing out that Eq. (13) is not applicable to metals where both $n(\omega)$ and $\kappa(\omega)$ behave as $\sim\omega^{-1/2}$ as $\omega \to 0$.

Similar derivations may be carried out by first squaring the appropriate Kramers-Kronig relations. From Eq. (7),

$$\frac{\kappa^2(\omega)}{\omega^2} = \frac{4}{\pi^2}\, P\int_0^\infty \frac{[n(\omega')-1]\,d\omega'}{\omega'^2-\omega^2} \times P\int_0^\infty \frac{[n(\omega'')-1]\,d\omega''}{\omega''^2-\omega^2}, \qquad (14)$$

and hence

$$\int_0^\infty \kappa^2(\omega)\,d\omega = \frac{4}{\pi^2}\int_0^\infty \omega^2\,d\omega\, P\int_0^\infty \frac{[n(\omega')-1]\,d\omega'}{\omega'^2-\omega^2} \times P\int_0^\infty \frac{[n(\omega'')-1]\,d\omega''}{\omega''^2-\omega^2}. \qquad (15)$$

Interchanging the order of integration as before, and making use of the result,

$$P\int_0^\infty \frac{\omega^2\,d\omega}{(\omega'^2-\omega^2)(\omega''^2-\omega^2)} = \frac{\pi^2}{4}[\delta(\omega'-\omega'')+\delta(\omega'+\omega'')] \qquad (16)$$

converts Eq. (15) into

$$\int_0^\infty \kappa^2(\omega)\,d\omega = \int_0^\infty [n(\omega')-1]\,d\omega' \times \int_0^\infty [n(\omega'')-1]\{\delta(\omega'-\omega'')+\delta(\omega'+\omega'')\}\,d\omega'', \qquad (17)$$

and hence

$$\int_0^\infty \kappa^2(\omega)\,d\omega = \int_0^\infty [n(\omega)-1]^2\,d\omega, \qquad (18)$$

a result noted by Altarelli and Smith.[2] In a similar manner, squaring Eq. (6) and integrating over all frequencies gives

$$\int_0^\infty \omega^2[n(\omega)-1]^2\,d\omega = \frac{4}{\pi^2}\int_0^\infty \omega^2\,d\omega\, P\int_0^\infty \frac{\omega'\kappa(\omega')\,d\omega'}{\omega'^2-\omega^2} \times P\int_0^\infty \frac{\omega''\kappa(\omega'')\,d\omega''}{\omega''^2-\omega^2}. \qquad (19)$$

Employing Eq. (16), we have the result

$$\int_0^\infty \omega^2[n(\omega)-1]^2\,d\omega = \int_0^\infty \omega^2\kappa^2(\omega)\,d\omega,$$

which is the VZ sum rule, Eq. (5). The sum rule, Eq. (13), is obtained from the Kramers-Kronig relations as follows. Multiplying Eq. (6) by (7) gives the result

$$\frac{\kappa(\omega)[n(\omega)-1]}{\omega} = -\frac{4}{\pi^2}\, P\int_0^\infty \frac{\omega'\kappa(\omega')\,d\omega'}{\omega'^2-\omega^2} \times P\int_0^\infty \frac{[n(\omega'')-1]\,d\omega''}{\omega''^2-\omega^2}, \qquad (20)$$

and hence

$$\int_0^\infty \omega\kappa(\omega)[n(\omega)-1]\,d\omega = -\frac{4}{\pi^2}\int_0^\infty \omega^2\,d\omega \times P\int_0^\infty \frac{\omega'\kappa(\omega')\,d\omega'}{\omega'^2-\omega^2}\, P\int_0^\infty \frac{[n(\omega'')-1]\,d\omega''}{\omega''^2-\omega^2}, \qquad (21)$$

which yields

$$\int_0^\infty \omega\kappa(\omega)[n(\omega)-1]\,d\omega = 0.$$

A number of additional results may be c' ined by considering higher powers of the generalized refractive index. The quantity $N^2(\omega)-1$, where $N(\omega)$ is the generalized refractive index, is a holomorphic function in the upper half-plane, from which it can be deduced, with the appropriate asymptotic behavior, that

$$N^2(\omega')-1 = \frac{P}{\pi i}\int_{-\infty}^\infty \frac{[N^2(\omega)-1]\,d\omega}{\omega-\omega'}, \qquad (22)$$

and hence

$$n^2(\omega')-\kappa^2(\omega')-1 = \frac{4}{\pi}P\int_0^\infty \frac{\omega\kappa(\omega)n(\omega)\,d\omega}{\omega^2-\omega'^2}, \qquad (23)$$

$$n(\omega')\kappa(\omega') = -\frac{\omega'}{\pi}\, P\int_0^\infty \frac{[n^2(\omega)-\kappa^2(\omega)-1]\,d\omega}{\omega^2-\omega'^2}. \qquad (24)$$

From Eqs. (9) and (23),

$$\int_0^\infty [n^2(\omega')-\kappa^2(\omega')-1]\,d\omega' = \frac{4}{\pi}\int_0^\infty d\omega'\, P\int_0^\infty \frac{\omega\kappa(\omega)n(\omega)\,d\omega}{\omega^2-\omega'^2} = -\pi\int_0^\infty \omega\kappa(\omega)n(\omega)\delta(\omega)\,d\omega = 0,$$

hence,

$$\int_0^\infty [n^2(\omega)-1]\,d\omega = \int_0^\infty \kappa^2(\omega)\,d\omega. \qquad (25)$$

Similarly, from Eqs. (9) and (24),

$$\int_0^\infty \frac{n(\omega)\kappa(\omega)\,d\omega}{\omega} = \frac{\pi}{4}[n^2(0)-1]. \qquad (26)$$

Equation (26) can also be recognized as the limit as $\omega' \to 0$ of the Kramers-Kronig relation, Eq. (23). Equation (26) is restricted to nonconductors. Sum rules for higher powers of the refractive index may also be obtained from the squares and the products of Eqs. (23) and (24). Thus from Eqs. (23) and (16), we get

$$\int_0^\infty [n^2(\omega)-\kappa^2(\omega)-1]^2\omega^2\,d\omega = 4\int_0^\infty \omega^2 n^2(\omega)\kappa^2(\omega)\,d\omega; \qquad (27)$$

from Eqs. (24) and (16), we obtain

$$\int_0^\infty n^2(\omega)\kappa^2(\omega)\,d\omega = \frac{1}{4}\int_0^\infty [n^2(\omega)-\kappa^2(\omega)-1]^2\,d\omega; \qquad (28)$$

and from Eqs. (16), (23), and (24), we get

$$\int_0^\infty \omega n(\omega)\kappa(\omega)[n^2(\omega)-\kappa^2(\omega)-1]\,d\omega = 0. \qquad (29)$$

III. GENERALIZED REFRACTIVE INDEX AT COMPLEX FREQUENCIES

The generalized refractive index of a medium may be expressed in the form

$$N^2(\omega) = 1 + \int_0^\infty \exp(i\omega\tau)G(\tau)\,d\tau, \qquad (30)$$

where the function $G(\tau)$ depends on the properties of the medium and the time, and ω is to be regarded as a complex frequency. From Eq. (30), it follows that for complex frequencies

$$N^2(-\omega^*)=N^{*2}(\omega). \tag{31}$$

For purely imaginary frequencies, $\omega=i\omega''$,

$$N^2(i\omega'')=N^{*2}(i\omega''), \tag{32}$$

and hence, on the purely imaginary frequency axis, $N^2(\omega)$ is real. A similar argument may be presented for the function $N(\omega)-1$. The function $N^2(\omega)-1$ obeys the relation[4]

$$\int_{-\infty}^{\infty}|N^2(\omega'+i\omega'')-1|^2\,d\omega<\text{const}\qquad(\omega''>0), \tag{33}$$

which allows by use of Titchmarsh's theorem, for $N(\omega)$ to be written in the form

$$N(\omega)=1+\int_0^\infty \exp(i\omega\tau)F(\tau)\,d\tau, \tag{34}$$

where $F(\tau)$ depends on the properties of the medium; from which it follows that $N(\omega)$ is real on the imaginary frequency axis and in particular

$$\kappa(i\omega'')=0 \tag{35}$$

for real ω''.

The simplest result for the refractive index at complex frequencies can be obtained by considering the integral of the function $[N(\omega)-1]/(\omega-i\omega_0)$ around a half-circle contour containing the real axis, and the upper-half complex frequency plane. For ω_0 real and greater than zero, and the notation simplified by designating ω as a real variable, we have the result

$$\omega_0\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{\omega^2+\omega_0^2}+\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{\omega^2+\omega_0^2}=\pi[n(i\omega_0)-1], \tag{36}$$

and considering the function $[N(\omega)-1]/(\omega+i\omega_0)$, we obtain

$$\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{\omega^2+\omega_0^2}=\omega_0\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{\omega^2+\omega_0^2}. \tag{37}$$

From Eqs. (36) and (37), the results for the refractive index on the imaginary frequency axis are

$$\frac{\pi}{2}[n(i\omega_0)-1]=\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{\omega^2+\omega_0^2}, \tag{38}$$

$$\frac{\pi}{2}[n(i\omega_0)-1]=\omega_0\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{\omega^2+\omega_0^2}. \tag{39}$$

From Eqs. (38) and (39) the following results for integrals along the imaginary frequency axis can be obtained:

$$\int_0^\infty\frac{[n(i\omega')-1]\,d\omega'}{\omega'}=\frac{2}{\pi}\int_0^\infty d\omega'\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{\omega^2+\omega'^2}, \tag{40}$$

on interchanging the order of integration, Eq. (40) becomes

$$\int_0^\infty\frac{[n(i\omega')-1]\,d\omega'}{\omega'}=\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{\omega}, \tag{41}$$

hence,

$$\int_0^\infty\frac{[n(i\omega)-n(\omega)]\,d\omega}{\omega}=0; \tag{42}$$

and from Eq. (38),

$$\int_0^\infty[n(i\omega)-1]\,d\omega=\int_0^\infty\kappa(\omega)\,d\omega. \tag{43}$$

A result analogous to Eq. (43), is well known for the dielectric constant.[5] Kramers-Kronig relations involving the refractive index on the imaginary axis may be derived by restricting the contour Γ to a circular arc in the first quadrant, then the integral

$$\int_\Gamma\frac{[N(\omega)-1]\,d\omega}{\omega-\omega'}$$

(ω' on the real positive axis) leads to the results

$$\omega'\int_0^\infty\frac{[n(i\omega)-1]\,d\omega}{\omega^2+\omega'^2}=\pi[n(\omega')-1]-P\int_0^\infty\frac{\kappa(\omega)\,d\omega}{\omega-\omega'}, \tag{44}$$

$$\int_0^\infty\frac{\omega[n(i\omega)-1]\,d\omega}{\omega^2+\omega'^2}=\pi\kappa(\omega')+P\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{\omega-\omega'}. \tag{45}$$

Sum rules which are more rapidly convergent for large frequencies can be generated by noting values of the derivatives of the generalized refractive index on the imaginary frequency axis. The integral

$$\oint\frac{[N(\omega)-1]\,d\omega}{(\omega^2+1)^2}$$

evaluated for a semicircular contour in the upper-half complex plane yields the result

$$\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{(\omega^2+1)^2}=\frac{\pi}{4}[n(i)-1]-\frac{\pi i}{4}\left(\frac{d[N(\omega)-1]}{d\omega}\right)_{\omega=i}; \tag{46}$$

and since

$$\left(\frac{d[N(\omega)-1]}{d\omega}\right)_{\omega=i}=\frac{4i}{\pi}\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{(\omega^2+1)^2}, \tag{47}$$

Eq. (46) simplies to

$$\int_0^\infty\frac{(1-\omega^2)[n(\omega)-1]\,d\omega}{(\omega^2+1)^2}=2\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{(\omega^2+1)^2}. \tag{48}$$

Equation (48) may be readily checked by differentiating the integrals in Eq. (37) with respect to ω_0, which leads to

$$\int_0^\infty\frac{\omega\kappa(\omega)\,d\omega}{(\omega^2+\omega_0^2)^2}=\frac{1}{2\omega_0}\int_0^\infty\frac{[n(\omega)-1](\omega_0^2-\omega^2)\,d\omega}{(\omega^2+\omega_0^2)^2}, \tag{49}$$

and thus reduces to Eq. (48) on setting $\omega_0=1$. Considering in a similar manner, the integral

$$\oint\frac{[N(\omega)-1]\,d\omega}{(\omega^2+1)^3},$$

leads to the result

$$\int_0^\infty\frac{[n(\omega)-1]\,d\omega}{(\omega^2+1)^3}=\frac{3\pi}{16}[n(i)-1]-\frac{3\pi i}{16}\left(\frac{d[N(\omega)-1]}{d\omega}\right)_{\omega=i}-\frac{\pi}{16}\left(\frac{d^2[N(\omega)-1]}{d\omega^2}\right)_{\omega=i}. \tag{50}$$

With the result

$$\left(\frac{d^2[N(\omega)-1]}{d\omega^2}\right)_{\omega=i}=\frac{4}{\pi}\int_0^\infty\frac{[n(\omega)-1](3\omega^2-1)\,d\omega}{(\omega^2+1)^3}, \tag{51}$$

Eq. (50) simplifies to the same result as was obtained by considering the integral

$$\oint \frac{[N(\omega)-1]\,d\omega}{(\omega+1)^2},$$

i.e.,

$$\int_0^\infty \frac{[n(\omega)-1](1-\omega^2)\,d\omega}{(\omega^2+1)^2} = 2\int_0^\infty \frac{\omega\kappa(\omega)\,d\omega}{(\omega^2+1)^2}. \qquad (52)$$

Equation (52) may be rearranged with the aid of Eq. (2) to the alternative form

$$2\int_0^\infty \frac{\omega\kappa(\omega)\,d\omega}{(\omega^2+1)^2} = -\int_0^\infty \frac{\omega^2(\omega^2+3)[n(\omega)-1]\,d\omega}{(\omega^2+1)^2}. \qquad (53)$$

IV. ZEROS OF THE FUNCTION $[n(\omega)-1]$

From the ADNS sum rule, Eq. (2), it is obvious that there must exist at least one zero of the function $[n(\omega)-1]$ [except in the trivial and unrealistic situation that $n(\omega)=1$ for all ω]. If this zero is designated by the point $\omega=\omega_0$, then from the Kramers-Kronig relationship, Eq. (6), we have the immediate result that

$$\int_0^\infty \frac{\omega\kappa(\omega)\,d\omega}{\omega^2-\omega_0^2}=0. \qquad (54)$$

The necessity of retaining the principal value in Eq. (54) depends on the behavior of $\kappa(\omega)$ at the point $\omega=\omega_0$.

From the Kramers-Kronig relations, subtracted dispersion relations for the real and imaginary parts of the generalized refractive index can be written

$$n(\omega')-n(\omega'')=\frac{2}{\pi}(\omega'^2-\omega''^2)P\int_0^\infty \frac{\omega\kappa(\omega)\,d\omega}{(\omega^2-\omega'^2)(\omega^2-\omega''^2)}, \qquad (55)$$

$$\kappa(\omega')-\kappa(\omega'')=\frac{2}{\pi}(\omega''-\omega')P\int_0^\infty \frac{[n(\omega)-1][\omega^2+\omega'\omega'']\,d\omega}{(\omega^2-\omega'^2)(\omega^2-\omega''^2)}. \qquad (56)$$

For any two given frequencies ω_0', ω_0'' $(\omega_0'\neq\omega_0'')$, which are both zeros of the function $[n(\omega)-1]$, or alternatively for which $[n(\omega_0')-n(\omega_0'')]$ vanishes, then from Eq. (55),

$$P\int_0^\infty \frac{\omega\kappa(\omega)\,d\omega}{(\omega^2-\omega_0'^2)(\omega^2-\omega_0''^2)}=0. \qquad (57)$$

Similarly, if there exist two distinct frequencies ω_1, ω_2 $(\omega_1\neq 0,\ \omega_2\neq 0)$ such that $[\kappa(\omega_1)-\kappa(\omega_2)]=0$, then Eq. (56) yields the result

$$P\int_0^\infty \frac{[n(\omega)-1][\omega^2+\omega_1\omega_2]\,d\omega}{(\omega^2-\omega_1^2)(\omega^2-\omega_2^2)}=0. \qquad (58)$$

The relationship Eq. (57) also follows directly from Eq. (54) when two zeros of the function $[n(\omega)-1]$ are known. In fact, for a set of distinct frequencies which are zeros of $[n(\omega)-1]$, a compact form for the sum rules can be written as

$$\int_0^\infty \omega\kappa(\omega)\,d\omega=\int_0^\infty \frac{\omega^3\kappa(\omega)\,d\omega}{\omega^2-\omega_0^2}=\int_0^\infty \frac{\omega^3\kappa(\omega)\,d\omega}{\omega^2-\omega_1^2}=\cdots, \qquad (59)$$

where $\omega_0, \omega_1, \cdots$ designate the zeros of $[n(\omega)-1]$. If we employ the f sum rule [Eq. (1)], Eq. (59) becomes

$$\frac{\pi}{4}\omega_p^2=\int_0^\infty \frac{\omega^3\kappa(\omega)\,d\omega}{\omega^2-\omega_0^2}=\int_0^\infty \frac{\omega^3\kappa(\omega)\,d\omega}{\omega^2-\omega_1^2}=\cdots. \qquad (60)$$

V. HIGHLY DAMPED CONSTRAINTS

The desirability of finding highly damped sum rule relations for the optical constants lies in the importance of deemphasizing certain frequency regions. For the high-frequency region, the optical constants are not ascertainable with high accuracy. The purpose of this section is to outline some possibly useful constraints for providing a consistency check of experimental values, while at the same time, damping the high-frequency results, so that inaccuracies in this region may be ignored. The results obtained make use of some of the formal relations for the refractive index at imaginary frequency.

The method of generating highly damped sum rules consists of considering the appropriate analytic exponential function $[\exp(a\omega)/(\exp(b\omega)+1)]\exp(i\delta\omega)$ $(\delta>0;\ b>a;\ a,\ b,\ \delta$ all real) multiplied by the function $[N(\omega)-1]$. Considering the integral

$$\oint \frac{\exp(a\omega)}{\exp(b\omega)+1}\exp(i\delta\omega)[N(\omega)-1]\,d\omega,$$

with the contour a semicircle in the upper half-plane,

$$\oint \frac{\exp(a\omega)}{\exp(b\omega)+1}\exp(i\delta\omega)[N(\omega)-1]\,d\omega = 2\pi i\Sigma\left(\text{residues at } \frac{n\pi i}{b},\ n=1,3,\cdots\right), \qquad (61)$$

then using Jordan's Lemma, and taking the limit $\delta\to+0$,

$$\int_{-\infty}^\infty \frac{\exp(a\omega)}{\exp(b\omega)+1}[N(\omega)-1]\,d\omega = -\frac{2\pi i}{b}\sum_{j=0}^\infty \exp\left[(ia\pi/b)(2j+1)\right]\left[N\left((2j+1)\frac{\pi i}{b}\right)-1\right]. \qquad (62)$$

Separating Eq. (61) into real and imaginary parts, leads to the results

$$\int_{-\infty}^\infty \frac{\exp(a\omega)}{\exp(b\omega)+1}[n(\omega)-1]\,d\omega = \frac{2\pi}{b}\sum_{j=0}^\infty \sin\left(\frac{\pi a}{b}(2j+1)\right)\left[n\left((2j+1)\frac{\pi i}{b}\right)-1\right] \qquad (63)$$

and

$$\int_{-\infty}^\infty \frac{\exp(a\omega)\kappa(\omega)\,d\omega}{\exp(b\omega)+1} = -\frac{2\pi}{b}\sum_{j=0}^\infty \cos\left(\frac{\pi a}{b}(2j+1)\right)\left[n\left((2j+1)\frac{\pi i}{b}\right)-1\right]. \qquad (64)$$

Some special cases of Eqs. (63) and (64) can be given. Taking the $\lim a\to+0$ for Eq. (63), leads to the result

$$\int_0^\infty [n(\omega)-1]\,d\omega=0,$$

which is the ADNS sum rule. Taking the $\lim a\to+0$ for Eq. (64) and making use of Eq. (38) leads to a trivial identity [both sides equal to $-\int_0^\infty \kappa(\omega)\tanh(\frac{1}{2}b\omega)\,d\omega$]. This identity can be proved easily for general a, b $(b>a)$ by employing Eq. (38) and the result

$$\sum_{j=0}^\infty \frac{\cos[(\pi a/b)(2j+1)]}{(2j+1)^2+b^2\omega^2\pi^{-2}} = \frac{\pi^2}{4b\omega}[\cosh a\omega\tanh\tfrac{1}{2}b\omega-\sinh a\omega]. \qquad (65)$$

The $\lim b \to a$ leads to similar results. An interesting special case is obtained for $2a=b$. From Eqs. (63) and (38) we have

$$\int_0^\infty [n(\omega)-1]\operatorname{sech}\left(\frac{b\omega}{2}\right)d\omega = \frac{4}{b}\sum_{j=0}^{\infty}(-1)^j\int_0^\infty \frac{\omega\kappa(\omega)\,d\omega}{\omega^2+(2j+1)^2\pi^2 b^{-2}} = \left(\frac{4b}{\pi^2}\right)\int_0^\infty \omega\kappa(\omega)\,d\omega \sum_{j=0}^{\infty}\frac{(-1)^j}{(2j+1)^2+b^2\omega^2\pi^{-2}}. \quad (66)$$

Now employing the result

$$\sum_{j=0}^{\infty}\frac{(-1)^j}{(2j+1)^2}=\beta(2)\ \text{[Catalan's constant (Ref. 6)]}, \quad (67)$$

where $\beta(2)\approx 0.91596$, and the inequaltiy

$$\frac{4b}{\pi^2}\beta(2)\int_0^\infty \omega\kappa(\omega)\,d\omega > \frac{4b}{\pi^2}\int_0^\infty \omega\kappa(\omega) \times\left(\sum_{j=0}^{\infty}\frac{(-1)^j}{(2j+1)^2+b^2\omega^2\pi^{-2}}\right)d\omega,\quad 0<b<\infty \quad (68)$$

allows the following inequality to be written:

$$b\beta(2)\pi^{-1}\omega_p^2 > \int_0^\infty [n(\omega)-1]\operatorname{sech}\left(\frac{b\omega}{2}\right)d\omega, \quad (69)$$

which is a principal result of this paper. The integral in Eq. (69) is very strongly damped at high frequencies. The special case $2a=b$ for Eq. (64) leads to a trivial identity.

A general extension of the above procedure consists of considering the situation in which the appropriate function has a singularity on the real axis, in addition to the singularities on the imaginary frequency axis. Consider the integral

$$\oint \frac{\exp(a\omega)\exp(i\delta\omega)}{\exp(b\omega)+1}\,\frac{[N(\omega)-1]\,d\omega}{\omega-\omega_0}$$

($\delta>0$; $b>a$: a, b, δ, and ω_0 real), with the contour taken as a semicircle in the upper half-plane, including the real axis, with the contour indented (into the upper complex plane) at the singularity on the real axis. The limit $\delta\to+0$ gives

$$P\int_{-\infty}^{\infty}\frac{\exp(a\omega)[N(\omega)-1]\,d\omega}{[\exp(b\omega)+1](\omega-\omega_0)} = -2\pi i\sum_{j=0}^{\infty}\frac{\exp[(a/b)\pi i(2j+1)]}{\pi i(2j+1)-b\omega_0}\left[N\left(\frac{\pi i}{b}(2j+1)\right)-1\right]+\frac{i\pi\exp(a\omega_0)}{\exp(b\omega_0)+1}[N(\omega_0)-1]. \quad (70)$$

The real and imaginary parts are

$$P\int_{-\infty}^{\infty}\frac{[n(\omega)-1]\exp(a\omega)\,d\omega}{[\exp(b\omega)+1](\omega-\omega_0)}+\frac{\pi\exp(a\omega_0)\kappa(\omega_0)}{[\exp(b\omega_0)+1]} = -2\sum_{j=0}^{\infty}\frac{\{n[(\pi i/b)(2j+1)]-1\}}{(2j+1)^2+b^2\omega_0^2\pi^{-2}} \times\left[(2j+1)\cos\left(\frac{\pi a}{b}(2j+1)\right)+b\omega_0\pi^{-1}\sin\left(\frac{\pi a}{b}(2j+1)\right)\right] \quad (71)$$

and

$$P\int_{-\infty}^{\infty}\frac{\kappa(\omega)\exp(a\omega)\,d\omega}{[\exp(b\omega)+1](\omega-\omega_0)}-\frac{\pi[n(\omega_0)-1]\exp(a\omega_0)}{[\exp(b\omega_0)+1]} = -2\sum_{j=0}^{\infty}\frac{\{n[(\pi i/b)(2j+1)]-1\}}{(2j+1)^2+b^2\omega_0^2\pi^{-2}} \times\left[(2j+1)\sin\left(\frac{\pi a}{b}(2j+1)\right)-b\omega_0\pi^{-1}\cos\left(\frac{\pi a}{b}(2j+1)\right)\right]. \quad (72)$$

Some interesting special cases can be obtained from Eqs. (71) and (72). Taking the $\lim a\to+0$ in Eqs. (71) and (72) leads to the results

$$P\int_0^\infty \frac{[n(\omega)-1]\omega\tanh(\frac{1}{2}b\omega)\,d\omega}{\omega^2-\omega_0^2}+\frac{\pi}{2}\kappa(\omega_0)\tanh\left(\frac{b\omega_0}{2}\right) = 2\sum_{j=0}^{\infty}\frac{(2j+1)\{n[(\pi i/b)(2j+1)]-1\}}{(2j+1)^2+b^2\omega_0^2\pi^{-2}}, \quad (73)$$

$$P\int_0^\infty \omega_0\frac{\kappa(\omega)\tanh(\frac{1}{2}b\omega)\,d\omega}{\omega^2-\omega_0^2}-\frac{\pi}{2}\tanh\left(\frac{b\omega_0}{2}\right)[n(\omega_0)-1] = -2b\omega_0\pi^{-1}\sum_{j=0}^{\infty}\frac{n[(\pi i/b)(2j+1)]-1}{(2j+1)^2+b^2\omega_0^2\pi^{-2}}. \quad (74)$$

Combining Eqs. (73) and (38) and employing the result

$$\tfrac{7}{8}\zeta(3)=\sum_{j=0}^{\infty}\frac{1}{(2j+1)^3} > \sum_{j=0}^{\infty}\frac{2j+1}{(2j+1)^2+b^2\omega_0^2\pi^{-2}}\,\frac{1}{(2j+1)^2+b^2\omega^2\pi^{-2}}, \quad (75)$$

where $\zeta(m)$ is Riemann's zeta function $[\zeta(3)\approx 1.202]$, leads to the result

$$P\int_0^\infty \frac{\omega\tanh(\frac{1}{2}b\omega)[n(\omega)-1]\,d\omega}{\omega^2-\omega_0^2} < \tfrac{7}{8}\zeta(3)b^2\omega_p^2\pi^{-2}-\tfrac{1}{2}\pi\kappa(\omega_0)\tanh\left(\frac{b\omega_0}{2}\right). \quad (76)$$

Combining Eqs. (73) and (39) leads directly to the inequality

$$\tfrac{1}{2}\pi\kappa(\omega_0)\tanh\left(\frac{b\omega_0}{2}\right)+P\int_0^\infty\frac{\omega\tanh(\frac{1}{2}b\omega)[n(\omega)-1]\,d\omega}{\omega^2-\omega_0^2}<0, \quad (77)$$

where the result

$$\sum_{j=0}^{\infty}\frac{1}{(2j+1)^2} > \sum_{j=0}^{\infty}\frac{(2j+1)^2}{(2j+1)^2+b^2\omega_0^2\pi^{-2}}\,\frac{1}{(2j+1)^2+b^2\omega^2\pi^{-2}} \quad (78)$$

and the ADNS sum rule, Eq. (2), have been employed. Equation (77) is a stronger inequality than Eq. (76) for the upper bound to the integral. Similar results can be derived from Eq. (74);

$$P\int_0^\infty \frac{\tanh(b\omega/2)\kappa(\omega)\,d\omega}{\omega^2-\omega_0^2} + \tfrac{15}{16}b^3\pi^{-3}\omega_p^2\,\zeta(4) > \tfrac{1}{2}\pi\omega_0^{-1}\tanh\left(\frac{b\omega_0}{2}\right)[n(\omega_0)-1] \qquad (79)$$

and

$$\omega_0 P\int_0^\infty \frac{\tanh(b\omega/2)\kappa(\omega)\,d\omega}{\omega^2-\omega_0^2} > \tfrac{1}{2}\pi\tanh\left(\frac{b\omega_0}{2}\right)[n(\omega_0)-1], \qquad (80)$$

where $\zeta(4)\approx 1.082$. Equation (80) is the stronger inequality for a lower bound to the integral

$$P\int_0^\infty \frac{\tanh(b\omega/2)\kappa(\omega)\,d\omega}{\omega^2-\omega_0^2}.$$

VI. DISCUSSION

The simplified procedure for derivation of many of the sum rules for the optical constants discussed in Sec. II is restricted only by the necessity of establishing the conditions of summability of the integrands, so that, the order of integration may be inverted. Summability encompasses the asymptotic behavior that has been assumed in the derivation of these sum rules by Altarelli *et al.*[1]

The establishment of results for the optical constants as a function of imaginary frequency are of little utility from the point of view of providing a constraint for the testing of experimental data. However, they do provide a route to other sum rules, which evolve from particular functional forms of the optical constants for which poles on the imaginary axis appear, as in the case of the integrals considered in Sec. V.

Constraints on the optical constants which arise from the zeros of the particular optical functions have possible wide utility. The majority of the known sum rule constraints, with the principal exceptions of Eqs. (1) and (2), relate the integrals of different optical constants over an infinite frequency interval. In order to test experimental data by such sum rules, both optical constants need to be experimentally accessible, or obtainable through indirect means (i.e., by Kramers-Kronig inversions). Sum rules of the type (54) circumvent this difficulty, since only experimental results in the vicinity of the zero(s) need to be determined for one of the optical constants, and not the entire frequency interval of the particular optical constant. This represents a considerable reduction in effort compared with determining data over large frequency intervals.

The key results of Sec. V, Eqs. (69), (77), and (80) provide useful results from the point of view of providing suitable criteria for the quality of optical data. The highly damped nature of Eq. (69) essentially eliminates the difficulty of obtaining data at high frequencies, or having only poor data available for this region. Attempts to obtain strongly damped integral constraints as a function of a single optical constant have not as yet met with success.

The generalization of the above sum rules for conductors can be carried out in a straightforward manner. Connections between the various optical constants, such as the dielectric constant, the conductivity, etc., also allow a number of sum rules to be readily obtained. Extensions of the asymptotic method and the above procedures to second-order Kramers-Kronig relations for nonlinear optical phenomena[7–9] appear to be possible and this will be the subject of a further investigation.

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*Present address: Physical Chemistry Laboratory, Oxford University, South Parks Rd., Oxford OX1 3QZ, U.K.

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