Integral inequalities for optical constants

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Some upper bound integral inequalities are derived for various optical constants. Weight functions which damp out the high frequency spectral domain are given special emphasis. The implications for testing experimental data are discussed.

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I. INTRODUCTION

The purpose of this paper is to consider integral inequalities of the form

$$\int_{\alpha}^{\beta} W(\omega_0,\omega)\mathscr{O}(\omega)d\omega < A(\alpha,\beta,\omega_0), \qquad (1)$$

where $\mathscr{O}(\omega)$ denotes a particular optical constant and $W(\omega_0,\omega)$ is some convenient weight function. In the present work the interval $[\alpha,\beta]$ will be taken as $[0,\infty)$ and $A(0,\infty,\omega_0)$ will be abbreviated to $A(\omega_0)$. The situation for general limits α and β, which is certainly a case of considerable interest and practical application, appears somewhat more difficult if a discussion free of additional assumptions about the specific nature of the medium is required. The topic is currently under investigation.

The motivation for this study was twofold. A considerable amount of work has been carried out on sum rules for optical constants.[1–7] Almost all these results may be written in the form

$$\int_0^{\infty} w(\omega_0,\omega)\mathscr{O}(\omega)d\omega = B(\omega_0). \qquad (2)$$

There are severe restrictions on the choice of weight function $w(\omega_0,\omega)$ which allow $B(\omega_0)$ to be determined explicitly. In order for a relationship like Eq. (2) to be useful in the analysis of experimental data, it is highly desirable that, for values of the optical constants which are experimentally inaccessible, the resulting contributions to the integral be as small as possible. Usually, the most inacessible spectral range for optical constant measurements is the high energy domain.

Problems in data analysis could be reduced to a minimum by the simple expedient of choosing a weight function vanishing very rapidly at high frequencies; for example, an appropriate gaussian function would be suitable. This has the effect of producing for the optical constant of interest what is essentially a "band-limited" function. Unfortunately, for weight functions of this particular form, the function $B(\omega_0)$ cannot be determined (at least by the procedures used in Refs. 1–7).

A commonly employed approach is to augment the available experimental data with semiempirical extrapolations to the high frequency range. In those particular cases where the spectral range for the optical data is rather restricted, which is common in practice, it becomes somewhat difficult to ascertain whether any inaccuracy in satisfying Eq. (2) rests with the experimental data or the extrapolation procedure, or both. In such cases Eq. (2) is of restricted utility for testing experimental data.

The alternative viewpoint, which is taken in this work, is to start with a highly damped weight function and then determine the upper bound $A(\omega_0)$. Errors arising from neglect of experimental data beyond some upper frequency can be reduced significantly by choosing weight functions which vanish rapidly as $\omega\rightarrow\infty$. The utility of Eq. (1) is then governed by the magnitude of the deviation between the constant $A(\omega_0)$ and the true value of the integral in question. When such deviations are small for a particular choice of weight function, Eq. (1) may be more useful in practical applications than Eq. (2), for reasons mentioned above.

The constant $A(\omega_0)$ is not in general determined as the best possible bound in this work. Work to determine the best possible bounds would certainly clarify the relative advantages of Eq. (1) and (2) in data analysis.

II. INEQUALITIES FOR THE REFRACTIVE INDEX

In this work we have restricted the assumptions on the mathematical properties of the optical constants to those employed in the derivation of Eq. (2). Stated briefly, the generalized optical constant is an analytic function in an appropriate domain, and some assumption about the asymptotic behavior as $\omega\rightarrow\infty$ is used to derive Eq. (2). Usually the asymptotic behavior is taken from the free electron gas model. With such general assumptions on the behavior of the optical constants, it is to be expected that there are relatively few general inequality relations.

If we denote the real and imaginary parts of the generalized refractive index by $n(\omega)$ and $\kappa(\omega)$, respectively, then

$$\int_0^{\infty} \omega^{1/2}\kappa(\omega)^{1/2}\exp(-\pi^3\omega^2/128\omega_0^2)\,d\omega \leqslant \omega_0^{1/2}\omega_p \qquad (3)$$

and

$$\int_0^{\infty} [\omega\kappa(\omega)n(\omega)]^{1/2}\exp(-\pi^3\omega^2/128\omega_0^2)\,d\omega \leqslant \omega_0^{1/2}\omega_p, \qquad (4)$$

where ω_p is the plasma frequency and ω_0 is some arbitrary frequency. Equation (3) is derived in the following manner. Starting from the Buniakowsky–Schwartz inequality, and for some weight function $W(\omega_0,\omega)$,

$$\int_0^{\infty} \omega^{1/2}\kappa(\omega)^{1/2}W(\omega_0,\omega)\,d\omega \leqslant \left[\int_0^{\infty}\omega\kappa(\omega)\,d\omega\int_0^{\infty}W(\omega_0,\omega)^2\,d\omega\right]^{1/2}. \qquad (5)$$

If we choose

$$W(\omega_0,\omega) = \exp(-\pi^3\omega^2/128\omega_0^2) \tag{6}$$

and use the result[2]

$$\int_0^\infty \omega\kappa(\omega)\,d\omega = \tfrac{1}{4}\pi\omega_p^2, \tag{7}$$

then Eq. (3) follows directly. An equivalent procedure yields Eq. (4) if the sum rule[2]

$$\int_0^\infty \omega\kappa(\omega)n(\omega)\,d\omega = \tfrac{1}{4}\pi\omega_p^2 \tag{8}$$

is employed.

In Eq. (3) the gaussian weight function is very highly damped as $\omega\to\infty$, so errors resulting from omission of data should be negligible for the high frequency spectral range. It should also be noted that Eq. (3) has an advantage over Eq. (4), since only experimental data for a single optical constant is required to utilize Eq. (3), whereas Eq. (4) requires both the dispersive and dissipative data for the generalized refractive index. Inequality constraints like Eq. (4) therefore do not provide information on the quality of the experimental data for the dispersive and dissipative modes separately.

An inequality similar to Eq. (3) can be derived for the refractive index. The first step is to introduce a second weight function of the form $(\omega^2+\omega_\alpha^2)^{-1/2}$. A related idea is well known in the derivation of certain types of integral inequalities, such as generalizations of Carlson's inequality.[8] We have

$$\int_0^\infty n(\omega)^{1/2}W(\omega_0,\omega)\,d\omega$$
$$= \int_0^\infty [(\omega^2+\omega_\alpha^2)^{1/2}W(\omega_0,\omega)n(\omega)^{1/2}\,d\omega/(\omega^2+\omega_\alpha^2)^{1/2}]$$
$$\leqslant\left\{\int_0^\infty (\omega^2+\omega_\alpha^2)W(\omega_0,\omega)^2\,d\omega\int_0^\infty [n(\omega)\,d\omega/(\omega^2+\omega_\alpha^2)]\right\}^{1/2}. \tag{9}$$

If we make use of the relation[6]

$$\int_0^\infty \frac{n(\omega)\,d\omega}{\omega^2+\omega_\alpha^2} - \frac{\pi}{2\omega_\alpha} = \frac{1}{\omega_\alpha}\int_0^\infty \frac{\omega\kappa(\omega)\,d\omega}{\omega^2+\omega_\alpha^2}, \tag{10}$$

then

$$\int_0^\infty n(\omega)^{1/2}W(\omega_0,\omega)\,d\omega$$
$$\leqslant\left[\int_0^\infty (\omega^2+\omega_\alpha^2)W(\omega_0,\omega)^2\,d\omega(\pi/2\omega_\alpha)(1+\omega_p^2/2\omega_\alpha^2)\right]^{1/2}, \tag{11}$$

where the inequality

$$\int_0^\infty \omega\kappa(\omega)\,d\omega/(\omega^2+\omega_\alpha^2) < \pi\omega_p^2/4\omega_\alpha^2 \quad (\omega_\alpha>0) \tag{12}$$

has been employed. For the choice

$$W(\omega_0,\omega) = e^{-\omega^2/2\omega_0^2}, \tag{13}$$

$$\int_0^\infty n(\omega)^{1/2}W(\omega_0,\omega)\,d\omega$$
$$<(\pi^{3/4}/4\omega_\alpha)[(\omega_0/\omega_\alpha)(2\omega_\alpha^2+\omega_0^2)(2\omega_\alpha^2+\omega_p^2)]^{1/2} \quad (\omega_\alpha>0). \tag{14}$$

Since ω_0 is arbitrary in Eq. (13), we may choose $\omega_0=\omega_\alpha$ and obtain

$$\int_0^\infty n(\omega)^{1/2}W(\omega_\alpha,\omega)\,d\omega < \frac{3^{1/2}}{4}\pi^{3/4}(2\omega_\alpha^2+\omega_p^2)^{1/2}, \tag{15}$$

with weight function $W(\omega_\alpha,\omega) = \exp(-\omega^2/2\omega_\alpha^2)$. The case $\omega_\alpha=\omega_p$ leads to the simple bound

$$\int_0^\infty n(\omega)^{1/2}\exp(-\omega^2/2\omega_p^2)\,d\omega < \tfrac{3}{4}\pi^{3/4}\omega_p. \tag{16}$$

The best possible bound for Eq. (14) can be obtained by determining the minimum of the right-hand side with respect to ω_α. We find the sharpest bound when ω_α is given by

$$\omega_\alpha = \tfrac{1}{2}\{\omega_p^2+\omega_0^2+[\omega_p^4+\omega_0^4+14\omega_p^2\omega_0^2]^{1/2}\}.^{1/2}$$

The above derivation may be carried through in an analogous manner with $n(\omega)^{1/2}$ replaced by $[\omega\kappa(\omega)]^{1/2}$ to yield

$$\int_0^\infty [\omega\kappa(\omega)]^{1/2}\exp(-\omega^2/2\omega_\alpha^2)\,d\omega$$
$$<(3^{1/2}/4)\pi^{3/4}\omega_\alpha^{1/2}\omega_p. \tag{17}$$

The above approach leads to a final result similar to that discussed at the start of this section; however, the final bound is less sharp. This is immediately apparent on writing Eq. (17) in the equivalent form

$$\int_0^\infty [\omega\kappa(\omega)]^{1/2}\exp(-\pi^3\omega^2/128\omega_0^2)\,d\omega < (\tfrac{3}{2})^{1/2}\omega_0^{1/2}\omega_p. \tag{18}$$

This is not surprising, since the procedure of introducing the additional weight factor $(\omega^2+\omega_\alpha^2)^{-1/2}$ does not lead to optimum bounds.

For the particular choice of weight function

$$W(\omega_0,\omega) = (\omega^2+\omega_\alpha^2)^{-1} \tag{19}$$

the following simple inequality for the refractive index is obtained:

$$\int_0^\infty [n(\omega)\,d\omega/(\omega^2+\omega_0^2)] < (\pi/2\omega_0)n(0) \quad (\omega_0>0), \tag{20}$$

where $n(0)$ is the refractive index at zero frequency. The bound indicated is of interest only for the case of insulators, since for conductors $n(\omega)\sim\omega^{-1/2}$ as $\omega\to 0$. The proof of Eq. (20) follows directly from the Kramers–Kronig relation connecting the real and imaginary parts of the refractive index:

$$n(\omega_0)-1 = (2/\pi)P\int_0^\infty [\omega\kappa(\omega)\,d\omega/(\omega^2-\omega_0^2)] \tag{21}$$

and for $\omega_0=0$

$$n(0)-1 = (2/\pi)\int_0^\infty [\kappa(\omega)\,d\omega/\omega]$$
$$>(2/\pi)\int_0^\infty [(\omega\kappa(\omega)\,d\omega/(\omega^2+\omega_0^2)], \tag{22}$$

and using Eq. (10) leads to Eq. (20). It is to be noted that inequality (20) involves only a single optical constant and is therefore suitable for analysis of experimental data.

The inequality (20) can be used to derive the following inequality (using the approach indicated above):

$$\frac{[\int_0^\infty W(\omega_0,\omega)n(\omega)^{1/2}\,d\omega]^2}{\int_0^\infty W^2(\omega_0,\omega)(\omega^2+\omega_\alpha^2)\,d\omega} < \frac{\pi n(0)}{2\omega_\alpha}, \quad \omega_\alpha > 0, \tag{23}$$

where $W(\omega_0,\omega)$ is any suitable weight function. Equation (23) depends on only one optical constant, and the final result is independent of the plasma frequency.

III. INEQUALITY FOR THE REFLECTANCE

If we know that the optical constant satisfies

$$0 \leqslant \mathscr{O}(\omega) \leqslant \mathscr{A} \tag{24}$$

for the interval $[0,\infty)$, and we consider a weight function $W(\omega)$ which is decreasing on the same interval, then

$[\mathscr{A} - \mathscr{O}(\omega)][W(\omega) - W(\omega')]$ is positive for ω on the interval $[0,\omega']$

and

$\mathscr{O}(\omega)[W(\omega') - W(\omega)]$ is positive for ω on the interval $[\omega',\infty)$.

Hence,

$$\int_0^{\omega'} [\mathscr{A} - \mathscr{O}(\omega)][W(\omega) - W(\omega')]\,d\omega + \int_{\omega'}^\infty \mathscr{O}(\omega)[W(\omega') - W(\omega)]\,d\omega \geqslant 0. \tag{25}$$

Equation (25) can be rewritten as

$$F(\omega') \equiv \mathscr{A}\int_0^{\omega'} W(\omega)\,d\omega + W(\omega')\left[\int_0^\infty \mathscr{O}(\omega)\,d\omega - \mathscr{A}\omega'\right] \geqslant \int_0^\infty \mathscr{O}(\omega)W(\omega)\,d\omega. \tag{26}$$

The optimum upper bound is determined from the minimum of $F(\omega')$. Hence,

$$\frac{\partial F(\omega')}{\partial \omega'} = \left[\int_0^\infty \mathscr{O}(\omega)\,d\omega - \omega\mathscr{A}\right]\frac{\partial W(\omega')}{\partial \omega'} = 0 \tag{27}$$

and therefore

$$\omega' = (1/\mathscr{A})\int_0^\infty \mathscr{O}(\omega)\,d\omega. \tag{28}$$

Equation (26) may be written in the following form:

$$\int_0^\infty \mathscr{O}(\omega)W(\omega)\,d\omega \leqslant \mathscr{A}\int_0^{\omega'} W(\omega)\,d\omega \tag{29}$$

with ω' given by Eq. (28). Equation (29) is a variant of Steffensen's inequality,[9] and the above derivation is related to a procedure of Apéry.[10] Equation (29) would be potentially very useful if the appropriate upper bounds on the various optical constants were available. Unfortunately, except for one important case, such upper bounds are unknown.

Consider the special case of Eq. (29) for the reflectance $R(\omega)$,

$$\mathscr{O}(\omega) \equiv R(\omega), \tag{30}$$

$$\mathscr{A} = 1, \tag{31}$$

$$\omega' = \int_0^\infty R(\omega)\,d\omega, \tag{32}$$

and choose a convenient weight function

$$W(\omega) = e^{-\omega/\omega_0}, \tag{33}$$

where ω_0 is some arbitrary frequency. Combining Eqs. (29)–(33) yields

$$(1/\omega_0)\int_0^\infty R(\omega)e^{-\omega/\omega_0}d\omega \leqslant 1 - e^{-\omega'/\omega_0}. \tag{34}$$

This is clearly a refinement on the obvious inequality

$$(1/\omega_0)\int_0^\infty e^{-\omega/\omega_0}R(\omega)\,d\omega < 1 \tag{35}$$

and will be a substantial improvement when $(1/\omega_0)\int_0^\infty R(\omega)\,d\omega$ is much less than 1.

IV. DISCUSSION

The procedures discussed in Sec. II can be applied to the dispersive and absorptive parts of the generalized dielectric constant, to yield exactly analogous inequalities to those reported for the real and imaginary parts of the generalized refractive index. The approach also applies to most of the optical constants if appropriate equations analogous to Eqs. (7) and (10) can be established.

It is to be stressed that the reported inequalities are necessary constraints only. In those cases where experimental data lead to a violation of one of the inequalities, then that data is to be regarded as inaccurate. Nothing can be said about the accuracy of the data when the inequalities are satisfied, since offsetting inaccuracies may still exist in the experimental data.

For some of the inequalities derived herein, we have not made use of the precise asymptotic behavior of the optical constant under investigation, although it is implicitly assumed that it is such that all integrals under discussion converge. This contrasts with the situation for the derivation of some exact sum rules, where the asymptotic behavior must be precisely known.

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[1]W. M. Saslow, Phys. Lett. A **33**, 157 (1970).
[2]M. Altarelli, D. L. Dexter, H. M. Nussenzveig, and D. Y. Smith, Phys. Rev. B **6**, 4502 (1972).
[3]A. Villani and A. H. Zimerman, Phys. Lett. A **44**, 295 (1973).
[4]A. Villani and A. H. Zimerman, Phys. Rev. B **8**, 3914 (1973).
[5]M. Altarelli and D. Y. Smith, Phys. Rev. B **9**, 1290 (1974).
[6]F. W. King, J. Math. Phys. **17**, 1509 (1976).
[7]F. W. King, J. Chem. Phys. **71**, 4726 (1979) and references therein.
[8]E. F. Beckenbach and R. Bellman, *Inequalities* (Springer-Verlag, Berlin, 1971).
[9]D. S. Mitrinovic, *Analytic Inequalities* (Springer-Verlag, Berlin, 1970).
[10]R. Apéry, Atti Q. Cong. Un. Mat. Ital. (Roma 1953), **2**, 3 (1951).