Brief Reports

Brief Reports are accounts of completed research which, while meeting the usual **Physical Review** *standards of scientific quality, do not warrant regular articles. A Brief Report may be no longer than four printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Kramers-Kronig relations in optical data inversion

K.-E. Peiponen and E. M. Vartiainen
Väisälä Laboratory, Department of Physics, University of Joensuu, P.O. Box 111, SF-80100 Joensuu, Finland
(Received 21 March 1991)

Some remarks are made on the use of Kramers-Kronig relations in optical data inversion. It is shown that symmetry relations imposed on the optical constant should be taken into account when modeling the tails of the absorption and extinction curves.

I. INTRODUCTION

Dispersion relations are widely used in physics to obtain indirectly, via calculations, some of the properties of a system with the aid of measured data. In optical physics, data inversion is usually based on the use of Hilbert transforms,[1,2] which can be derived by means of the theory of complex-variable contour integration. Dispersion relations have found applications in quantum optics to describe analytic signals,[3] linear optical constants[4,5] in the case of linearly-polarized-light interactions, and also circularly-polarized-light interactions.[6] Recently, a method was presented describing nonlinear susceptibilities with several complex-angular-frequency variables, basically using Hilbert-transform-type integrals.[7]

An alternative method for inverting the data of linear optical constants has been introduced by King.[8,9] His method is based on the use of conjugate Fourier series and the symmetry properties of linear optical constants. This method has been found effective in the analysis of the frequency-dependent refractive-index change of F color centers in mixed alkali-halide crystals.[10]

A basically similar method to that of King has been applied in the study of the real and imaginary parts of nonlinear susceptibilities with the aid of the squared modulus of the susceptibility.[11] This kind of formalism has been applicable in the interpretation of nonlinear Raman susceptibilities.[12,13] In addition to the normal data inversion, the dispersion relations have been exploited to derive sum rules for the linear optical constants[14–20] and nonlinear susceptibilities.[7,11,21,22]

In this paper we consider some basic mathematical and physical requirements that are important in connection with Kramers-Kronig relations,[4] which are well established relations in optical data inversion. We show that some theoretical line-shape models, appropriate in the description of some properties of a system, may give erroneous results in connection with Kramers-Kronig relations.

II. DISPERSION RELATIONS

The Hilbert transforms for a function f, $f(z)=u(z)+iv(z)$, which is assumed to be an analytic function of a complex variable $z=x+iy$, are as follows:[1]

$$u(x')=\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{v(x)}{x-x'}dx\ ,\qquad v(x')=-\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{u(x)}{x-x'}dx\ , \tag{1}$$

where x' is a pole on the real axis, x is a real variable, and P denotes the Cauchy principal value. If we consider only the mathematical properties of the function f, the class of functions that fulfill the Hilbert transforms is rather wide. There is a relatively weak assumption about the asymptotical behavior of f: $f(z)\sim|z|^{-\delta}$, $\delta>0$, as $|z|\to\infty$. Note that there are no symmetry properties imposed on f.

In linear optics the dispersion relations of Eq. (1) are given using the energy, angular frequency, or wavelength of the incident light as a variable. There is one physical restriction on the variable: it must be positive. Because of this restriction on measurement, the dispersion relations of Eq. (1) are usually given in a different form, known as Kramers-Kronig relations. The basic idea of providing more applicable relations is based on the symmetry properties, known as crossing relations, of the linear optical constants. The symmetry properties follow from the fact that a real-valued electric field must produce a real-valued polarization of the charges.[4]

If we give the symmetry relations for the complex-angular-frequency-dependent refractive index, $\hat{n}(\omega)=n(\omega)+ik(\omega)$, we can write

$$n(-\omega)=n(\omega)\ ,\qquad k(-\omega)=-k(\omega)\ , \tag{2}$$

i.e., the real refractive index n is even and the extinction coefficient k is an odd function of angular frequency ω,

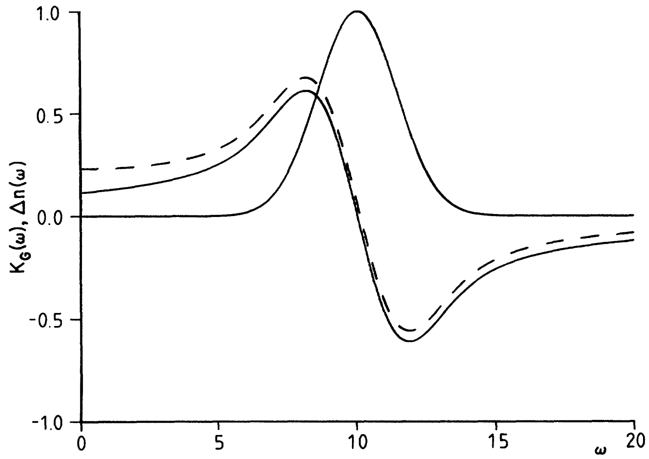

FIG. 1. Gaussian extinction curve ($A=1$, $\omega_0=10$, $W=2$) and corresponding true (solid line) and erroneous (dashed line) dispersion curves.

respectively. If we write the Kramers-Kronig relation for the change of the real refractive index, Δn, in its usual form, we have

$$\Delta n(\omega')=\frac{2}{\pi}\mathbf{P}\int_0^\infty \frac{\omega k(\omega)}{\omega^2-\omega'^2}d\omega \ . \tag{3}$$

However, this relation is valid only if k is an odd function of ω; otherwise, one has to use a relation similar to the first relation of Eq. (1). If we know nothing about the symmetry of the data to be inverted, we may try to decompose the data, using some model, to be given as a sum of even and odd parts. Such a decomposition is valid for any arbitrary function. In such a case we may write the dispersion relation using a physically observable integration interval from zero to infinity. There exists, however, a problem, since we know by measurement the sum function but not the functional behavior of its possible even and odd parts.

As an example, in Fig. 1 we demonstrate how the physical demand of $k(\omega)$ being an odd function may affect the result of a Kramers-Kronig calculation. The extinction coefficient is chosen to be a Gaussian line, which falls off rapidly at its tails. It can be given in the form

$$k_G(\omega)=A\exp\left[-\left[\frac{\omega-\omega_0}{W}\right]^2\right] ,$$

where A is the amplitude, ω_0 is the central frequency, and $2(\ln 2)^{1/2}W$ is the full width at half maximum. We observe that $k_g(-\omega)\neq -k_G(\omega)$. We calculated the refractive index change using the conventional form of Eq. (3) and the exact form in this case as follows:

$$\Delta n(\omega')=\frac{1}{\pi}\mathbf{P}\int_{-\infty}^{\infty}\frac{k_G(\omega)}{\omega-\omega'}d\omega \ . \tag{4}$$

It was observed that when we used Eq. (3) the ratio ω_0/W was crucial to obtain correct values of Δn. If ω_0/W is large, then the integral of Eq. (3) gives a good approximation. On the other hand, if the ratio has a relatively low value, then Eq. (3) gives an erroneous result, as demonstrated in Fig. 1. It is worth noting that the zero dispersion is shifted from the peak position when using Eq. (3) in the present example. The shift increases as ω_0/W decreases.

One also has to take care when using the absorption coefficient in data inversion. According to the well-known relation, the absorption coefficient is given by $\alpha(\omega)=2\omega k(\omega)/c$. From this it follows that the absorption coefficient must be an even function of ω, i.e., $\alpha(-\omega)=\alpha(\omega)$, in order to make use of Eq. (3). As an example we mention that a widely accepted absorption band shape in color-center physics,[23] the Gaussian line shape, is not consistent with the above symmetry requirement.

In many practical cases the tails of the extinction coefficient or absorption coefficient usually have to be approximated somehow beyond the measured data in order to perform the Kramers-Kronig calculations. We recommend that the symmetry relations imposed on the optical constant are taken into account when approximating the tails in order to get reliable approximations for the calculated optical constant.

III. DISCUSSION

In this paper we have drawn attention to the fact that one should be careful when choosing a particular model to describe optical properties in connection with Kramers-Kronig relations. There is a possibility of obtaining erroneous results, which was demonstrated by using a Gaussian line shape for the extinction coefficient. One has to make sure that the measured data are approximated in the low- and high-frequency limits so that they are consistent with symmetry relations imposed on optical constants. Only in such a case may one expect a good approximation of the refractive index change or other optical constant calculated using Kramers-Kronig relations. If the application of sum rules gives strange results, the broken symmetry of the line model should be taken into account.

We calculated the dispersion using also King's model in cases where the extinction has a Gaussian line shape. In this case the dispersion curve is rather well approximated and the location of zero dispersion was always correct. King's model, however, has not attracted as much attention as it may deserve. It is computationally a fast and reliable method for calculating dispersion in the case of a single band. Finally, we wish to emphasize that similar arguments to those above hold also for the interpretation of nonlinear susceptibilities obtained with the aid of Hilbert transforms of several angular-frequency variables.

[1] P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953).
[2] H. M. Nussenzweig, *Causality and Dispersion Relations* (Academic, New York, 1972).
[3] J. R. Klauder and E. C. G. Sudarshan, *Fundamentals of Quantum Optics* (Benjamin, New York, 1968).
[4] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Addison-Wesley, Reading, MA, 1960).
[5] D. Y. Smith, in *Handbook of Optical Constants of Solids,* edited by E. D. Palik (Academic, Orlando, 1985).
[6] D. Y. Smith, in *Theoretical Aspects and New Developments in Magneto-Optics,* edited by J. T. Deversec (Plenum, New York, 1980).
[7] K.-E. Peiponen, Phys. Rev. B **35**, 4116 (1987).
[8] F. W. King, J. Phys. C **10**, 3199 (1977).
[9] F. W. King, J. Opt. Soc. Am. **68**, 994 (1978).
[10] P. Ketolainen, K.-E. Peiponen, and K. Karttunen, Phys. Rev. B **43**, 4492 (1991).
[11] K.-E. Peiponen, Phys. Rev. B **37**, 6463 (1988).
[12] E. M. Vartiainen, K.-E. Peiponen, and T. Tsuboi, J. Opt. Soc. Am. B **7**, 722 (1990).
[13] E. M. Vartiainen and K.-E. Peiponen, in *Nonlinear Optics in Solids,* edited by O. Keller, Springer Series on Wave Phenomena Vol. 9 (Springer-Verlag, Berlin, 1990).
[14] M. Altarelli, D. L. Dexter, H. M. Nussenzweig, and D. Y. Smith, Phys. Rev. B **6**, 4502 (1972).
[15] M. Altarelli and D. Y. Smith, Phys. Rev. B **9**, 1290 (1974).
[16] F. W. King, J. Math. Phys. **17**, 1509 (1976).
[17] F. W. King, J. Chem. Phys. **71**, 4726 (1979).
[18] K.-E. Peiponen, Lett. Nuovo Cimento **44**, 445 (1985).
[19] K.-E. Peiponen, J. Phys. C **20**, 2785 (1987).
[20] D. Y. Smith, Phys. Rev. B **13**, 5303 (1976).
[21] K.-E. Peiponen, J. Phys. C **20**, L285 (1987).
[22] K.-E. Peiponen, E. M. Vartiainen, and T. Tsuboi, Phys. Rev. A **41**, 527 (1990).
[23] D. L. Dexter, Phys. Rev. **111**, 119 (1958).