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**KRAMERS–KRONIG RELATIONS – SUPPLEMENTARY
 TECHNIQUE TO THE TIME-DOMAIN SPECTROSCOPY**

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We have proposed a new method for obtaining the Kramers–Kronig relations based on the analysis of the earlier proposed solution of the wave equation for a dispersion medium [2]. The use of this solution for ultra-wideband (femtosecond) optical signals allowed us to propose a simultaneous measurement method for both $n(\omega)$ and $k(\omega)$. The latter is a further development of the time-domain spectroscopy method.

Keywords: Kramers–Kronig relations, femtosecond pulse, time-domain spectroscopy technique.

1. Introduction and Short History

In [3], Kronig obtained a relation for the refractive index and absorption coefficient. One year later, Kramers made his famous report [4]. It states that the real and imaginary parts of the complex refractive index $\nu(\omega) = n(\omega) + i\kappa(\omega)$ connected to one another. The basic requirement to get the Kramers–Kronig (KK) relations is the analyticity of a dielectric susceptibility (DS) in the upper half-plane for the adsorbing medium. There are some problems of the calculation of integrals within the Cauchy Principal Value [1].

We propose another method for obtaining analogous relations on the basis of the analysis of the solution of the wave equation in a dispersive medium using the principle of causality.

In addition, as it was stated by C.D. McDonald and K.E. Oughstun in [5], the “Hilbert transform pair relationship is only approximately satisfied”. More exactly, this is the case by experimentally measured data. So, we propose a direct (spectral) method of finding both refractive index $n(\omega)$ and absorption coefficient $\kappa(\omega)$. The basis for this is the analysis of the spectra of ultra-wideband pulse signals for samples of

different sizes (lengths). This proposal is a further development of the terahertz time-domain spectroscopy method [6] over the optical wavelength range, which is supported by recent advances in femtosecond pulse registration technique [7].

2. Theory

We use the formula that describes the propagation of an optical pulse in a medium with a dispersion obtained [2]

$$E(z, t) = \left(\frac{2}{\pi}\right)^{1/2} \int_0^\infty \exp[-\kappa(\omega)\omega z/c] \cos[\omega t] \times \\ \times \left\{ E_{OC}(\omega) \cos\left[n(\omega)\frac{\omega}{C}z\right] - \right. \\ \left. - E_{OS}(\omega)\omega \sin\left[n(\omega)\frac{\omega}{C}z\right] \right\} d\omega, \tag{1}$$

where

$$E_{OC}(\omega) = \left(\frac{2}{\pi}\right)^{1/2} \int_0^\infty E(0, t) \cos[\omega t] dt$$

– Fourier’s cos-transform,

$$E_{OS}(\omega) = \left(\frac{2}{\pi}\right)^{1/2} \int_0^\infty E(0, t) \sin[\omega t] dt$$

– Fourier’s sin-transform, $E(0, t)[U_t(t) - U_t(t - T)]$ – temporal pulse shape at the entrance to the sample, $U_t(t)$ – single step function, T – pulse duration, $\kappa(\omega)$, $n(\omega)$ – absorption coefficient and refractive index, accordingly.

Limiting the pulse duration (T) in time is natural for physical objects.

In the early work of A. Sommerfeld, L. Brillouin [8], and in our work [2], the existence of the so-called “forerunner” for an optical ultrashort pulse was shown by direct calculations. Its characteristic feature is the propagation at the speed of light in vacuum practically irrespective to the nature of the medium dispersion.

We write down the wave equation at the exit of the medium and substitute solution (1) in it

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = 0. \quad (2)$$

After substitution (1) to (2), one can get

$$\int_0^\infty \omega^2 \cos[\omega t] \exp\left[-\frac{\kappa\omega z}{c}\right] \left\{ \cos\left[n\frac{\omega}{c}z\right] [(\kappa^2 - n^2 + 1) \times \right. \\ \left. \times E_{OC} + 2\kappa n E_{OS}] + \sin\left[n\frac{\omega}{c}z\right] [(-\kappa^2 + 2\kappa n)E_{OC} + \right. \\ \left. + (n^2 - 1)E_{OS}] \right\} d\omega = 0. \quad (3)$$

We simplify Eq. (3) at the time

$$t = \frac{z}{c}, \quad \omega^{\max} z/c \rightarrow 0.$$

Then one can get the integral equation

$$\int_0^{\omega^{\max}} \omega^2 [(\kappa^2(\omega) - n^2(\omega) + 1)E_{OC} + \\ + 2\kappa(\omega)n(\omega)E_{OS}] d\omega = 0. \quad (4)$$

Below, we consider two variants of the probing of the sample: 1) a monochromatic signal and 2) a broad-band signal.

3. Variant I

First, we write the input signal in the form

$$E(0, t) = A \cos \omega_0 t + B \sin \omega_0 t. \quad (5)$$

Then we write the formulas for the Fourier cos- and sin-transforms of expression (3)

$$E_{OC}(\omega) = A \left(\frac{\pi}{2}\right)^{1/2} \delta(\omega - \omega_0) + B \left(\frac{2}{\pi}\right)^{1/2} \frac{\omega_0}{-\omega^2 + \omega_0^2}, \quad (6)$$

$$E_{OS}(\omega) = A \left(\frac{2}{\pi}\right)^{1/2} \frac{\omega}{\omega^2 - \omega_0^2} + B \left(\frac{\pi}{2}\right)^{1/2} \delta(\omega - \omega_0). \quad (7)$$

After substituting (6) and (7) into (4) and separating the signals, we obtain a system of two integral equations

$$(\kappa^2(\omega) - n^2(\omega))_{\omega_0} + 1 = -\frac{2}{\pi} \int_0^{\omega^{\max}} \left(\frac{\omega}{\omega_0}\right)^2 \frac{2\kappa n \omega}{\omega^2 - \omega_0^2} d\omega, \quad (8)$$

$$(2n(\omega)\kappa(\omega))_{\omega_0} =$$

$$= \frac{2}{\pi} \int_0^{\omega^{\max}} \left(\frac{\omega}{\omega_0}\right)^2 (\kappa^2 - n^2 + 1) \frac{\omega_0}{\omega^2 - \omega_0^2} d\omega. \quad (9)$$

For narrow-band spectral lines, one can neglect the smooth dependence on $(\omega/\omega_0)^2$ and write the well-known relations

$$(n^2(\omega) - \kappa^2(\omega))_{\omega_0} - 1 = \frac{2}{\pi} \int_0^{\omega^{\max}} \frac{2\kappa n \omega}{\omega^2 - \omega_0^2} d\omega, \quad (10)$$

$$(2n(\omega)\kappa(\omega))_{\omega_0} = -\frac{2\omega_0}{\pi} \int_0^{\omega^{\max}} \frac{n^2 - \kappa^2 - 1}{\omega^2 - \omega_0^2} d\omega. \quad (11)$$

One can calculate integrals in (10) and (11) in the sense of the Cauchy principal value. Formulas (10) and (11) are analytically valid for the well-known Drude–Lorentz model of the substance (see Ref. [9]).

4. Variant II

We will proceed from the general formula for the optical ultra-wideband pulse, which propagates in a medium with dispersion (1). The corresponding spectrum can be recorded as

$$FE(z, \omega) = \exp\left[-\frac{(\kappa(\omega)\omega z}{c}\right] \left\{ E_{OC}(\omega) \cos\left[n(\omega)\frac{\omega}{c}z\right] - \right. \\ \left. - E_{OS}(\omega) \sin\left[n(\omega)\frac{\omega}{c}z\right] \right\}. \quad (12)$$

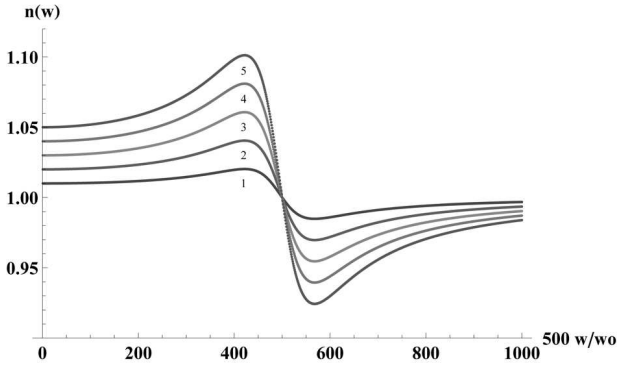


Fig. 1. Dispersion of the refractive index $n(\omega)$: 1 - $Q^2 = 0.02$; 2 - $Q^2 = 0.04$; 3 - $Q^2 = 0.06$; 4 - $Q^2 = 0.08$; 5 - $Q^2 = 0.10$; $\nu^2 = 1/12$

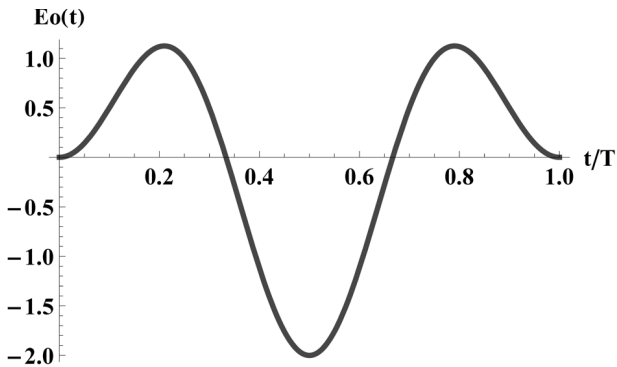


Fig. 2. Shape of a femtosecond pulse $E_0(t)$ of the zero order

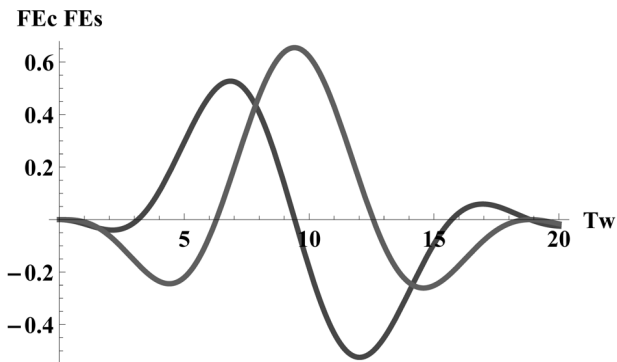


Fig. 3. Fourier cos- and sin-transforms of a pulse $E_0(t)$: $T\omega_0 = 8$. T - pulse duration, $T\omega = T\omega$

To exclude one of the two unknown functions, namely $k(\omega)$, let us write formula (12) for two sample lengths: z and $2z$. Then we divide the square of the

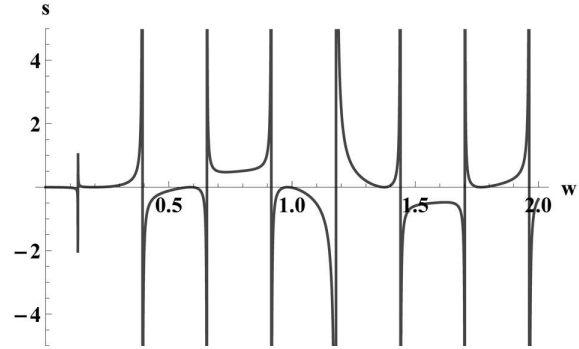


Fig. 4. Spectral coefficient $S(\omega)$: $Q^2 = 0.0001$, $\nu^2 = \frac{1}{12}$, $\frac{\omega_0 z}{c} = 5$, $T\omega_0 = 8$

first spectrum by the second one and get

$$\frac{[FE(z, \omega)]^2}{FE(2z, \omega)} = s(\omega) = \frac{[E_{OC}(\omega) \cos [n(\omega) \frac{\omega}{c} z] - E_{OS}(\omega) \sin [n(\omega) \frac{\omega}{c} z]]^2}{E_{OC}(\omega) \cos [2n(\omega) \frac{\omega}{c} z] - E_{OS}(\omega) \sin [2n(\omega) \frac{\omega}{c} z]} \quad (13)$$

Thus, we have obtained Eq. (13), in which the unknown function $n(\omega)$ enters in the implicit form. Here, $s(\omega)$ is the experimental spectral coefficient.

Next, we will perform a simulation of the solution of Eq. (13) for the classical oscillator model of matter

$$n(\omega) = 1 + \frac{1}{2} Q^2 \frac{1 - W^2}{(1 - W^2)^2 + W^2 \nu^2}, \quad (14)$$

where $Q = \frac{\omega_p}{\omega_0}$, $\omega_p^2 = 4\pi N e^2 / m$, $\omega_p \ll \omega_0$, $W = \frac{\omega}{\omega_0}$, $\nu = \frac{\nu}{\omega_0}$.

Dependences (14) for three concentrations (parameter Q) are shown in Fig. 1.

The probing femtosecond optical pulse can be represented as a zero-order elementary wave packet [10]

$$E_0(0, t) = \cos \left[\frac{2\pi t}{T} \right] - \cos \left[\frac{4\pi t}{T} \right], \quad (15)$$

where $t = t\omega_0$,

$$T = T\omega_0.$$

The shape of the pulse and the corresponding Fourier cos- and sin-transforms are shown in Figs. 2 and 3. We have

$$E_{OC}(\omega) = -\frac{12\pi^2(\omega T) \sin[T\omega]}{64\pi^4 - 20\pi^2(\omega T)^2 + (\omega T)^4}, \quad (16)$$

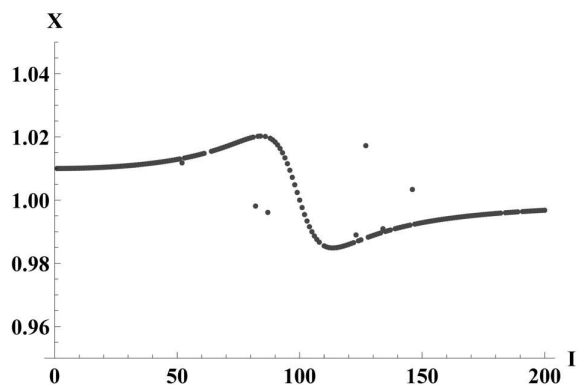


Fig. 5. Numerical calculation of the refractive index $X = n(\omega)$: $Q^2 = 0.02$, $\nu^2 = \frac{1}{12}$, $z = 5$, $T\omega_0 = 8$, $I = 100\omega/\omega_0$

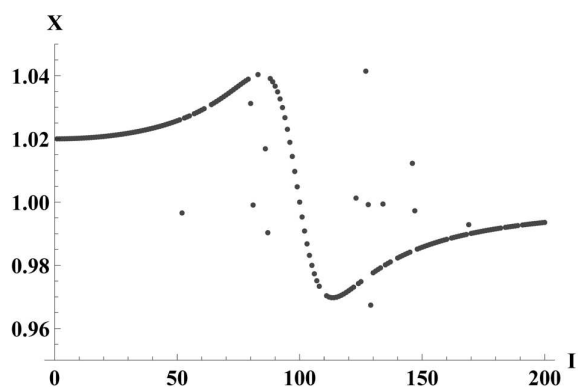


Fig. 6. Numerical calculation of the refractive index $X = n(\omega)$: $Q^2 = 0.04$, $\nu^2 = \frac{1}{12}$, $z = 5$, $T\omega_0 = 8$, $I = 100\omega/\omega_0$

$$E_{OS}(\omega) = -\frac{12\pi^2(-1 + \cos[T\omega])}{64\pi^4 - 20\pi^2(\omega T)^2 + (\omega T)^4}. \quad (17)$$

Let us calculate the spectral coefficient $S(\omega)$ with formulae (13), (14), (16), and (17): $Q^2 = 0.0001$, $\nu^2 = \frac{1}{12}$, $\frac{\omega_0 z}{c} = 5$, $T\omega_0 = 8$. The result is shown in Fig. 4. The spectral dependence $S(\omega)$ is rather complex. Therefore, we cannot refine the simple numerical solution of Eqs. (13).

The results of numerical calculations for the $n(\omega)$ dependence are shown in Figs. 5 and 6.

The increase of the concentration by two times [$Q^2 = 0.02$ (Fig. 5), $Q^2 = 0.04$ (Fig. 6)] leads to the appearance of the instability of solutions at some frequencies.

A refinement of the calculation algorithm, in particular, the optimization of the choice of the initial calculation point, improves the stability of the solution. In Fig. 7, we show a solution for $Q^2 = 0.1$.

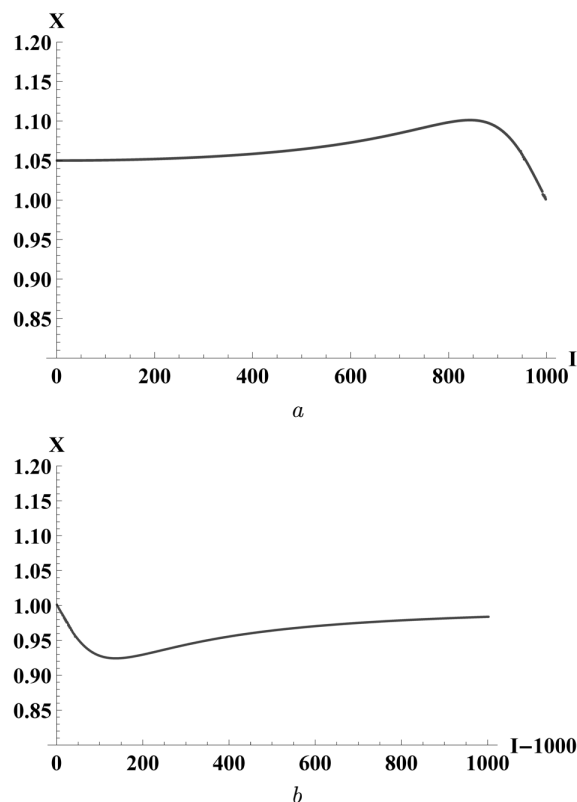


Fig. 7. Numerical calculation of the refractive index $X = n(\omega)$: $Q^2 = 0.1$, $\nu^2 = \frac{1}{12}$, $z = 10$, $T\omega_0 = 8$; (a) $0 \leq \omega \leq 999$, (b) $999 \leq \omega \leq 2000$ (compare with the data in Fig. 1, curve 5) $I = 1000\omega/\omega_0$

The dispersion curves in Fig. 7, *a*, *b* fit the data in Fig. 1 (curve 5). This is due to the improvement of the calculation algorithm. Namely, we use the value $n(\omega)$ calculated on the previous step as the “initial calculation point”.

5. Conclusions

1. The fundamental ground for obtaining the K–K relations is the principle of causality. This is not surprising, since they are written for monochromatic (infinite in time) signals.

2. The analysis of the exact solution of the wave equation for pulse signals propagating in a dispersion medium does not require the application of the principle of causality. It is automatically contained in the solution for physically realizable electromagnetic pulses.

3. The analysis of the spectra of ultra-wideband signals at the output of two samples of different lengths allows us to find the dispersion of both $n(\omega)$ and $\kappa(\omega)$.

4. In the optical range, the algorithm for numerical calculations of $n(\omega)$ and $\kappa(\omega)$ is complicated, because, as usual, $L \gg 2\pi c/\omega$, L – sample length.

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1. M.I. Kozak, V.N. Zhikharev, P.P. Paga, V.Yu. Loya. Kramers–Kronig relations: Validations via calculation technique. *IJISET* **4** (12), 152 (2017).
2. V.S. Ovechko, V.P. Myhashko. Spectral particularities of femtosecond optical pulse propagating in dispersive medium. *Ukr. J. Phys.* **63** (6), 478 (2018).
3. R. de L Kronig. On the theory of dispersion of X-rays. *J. Opt. Soc. Am. & Rev. Sci. Instr.* **12**, 547 (1926).
4. H.A. Kramers. La diffusion de la lumiere par les atoms. *Atti del Congresso Internazionale dei Fisici, Como* **2**, 545 (1927).
5. C.D. McDonald, K.E. Oughstun. Dispersion relation analysis of epsilon (omega) data for H₂O. In: *Proc. IEEE AP-S Int. Symposium, & USNC/URSI National Radio Science Meeting*, paper 1877 (2016).

6. W. Withayachumnancul, M. Naftaly. Fundamentals of measurement in terahertz time-domain spectroscopy. *J. Infrared, Millimeter, & Terahertz Waves.* **35** (8), 610 (2014).
7. W. Cho, S. In Hwang, C. Hee Nam, M.B. Bionta, P. Lassonde, B.E. Schmidt, H. Ibrakim, F. Legare, K. Taec Kim. Temporal characterization of femtosecond laser pulses using tunneling ionization in the UV, visible, & mid-IR ranges. *Sci. Rep.* **9**, 16067 (2019).
8. L. Brillouin. *Wave propagation and Group Velocity* (Academic Press, 1960).
9. T.S. Moss. *Optical Properties of Semiconductors* (Butterworths Scientific Publications, 1959).
10. V.S. Ovechko. Femtosecond optics – optics of the elementary wave packets. *J. Opt. Soc. Am. B* **29**, 799 (2012).

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СПІВВІДНОШЕННЯ
КРАМЕРСА–КРОНІГА – ДОПОВНЮЮЧА
“TIME-DOMAIN” СПЕКТРОСКОПІЮ МЕТОДИКА

Резюме

Запропоновано новий метод одержання співвідношень Крамерса–Кроніга, що ґрунтується на аналізі отриманого раніше розв’язку хвильового рівняння для фемтосекундних імпульсів у диспергуючому середовищі. Вказаний розв’язок використаний нами для подальшого розвитку методики “time-domain” спектроскопії для вимірювання обох матеріальних параметрів речовини $n(\omega)$ і $\kappa(\omega)$ одночасно.