

The Kramers-Kronig Relations: Validation via Calculation Technique

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Abstract

The historical aspect of the discovery of the Kramers-Kronig dispersion relations is considered. A calculational technique is shown for investigating the validity of the Kramers-Kronig relations. The dielectric susceptibility function is constructed as a classical linear response function in the framework of two equations of charge motion of the microscopic Lorentz theory. It is shown that the Kramers-Kronig relations can be satisfied in the presence of poles in both the upper and lower half-planes simultaneously. It is shown that the accuracy of fulfilling the KK's relations depends on the removal of the pole of the lower half-plane from the real axis. To determine the optical parameters of the absorbing medium from the reflection spectra, it is necessary to perform a conformal transformation. An example of the application of this approach to the reflection spectrum of silicon is shown.

Keywords: *Kramers-Kronig relations, Optical parameters, Reflection spectrum, Jahoda formula*

1 Introduction

September this year marked the 90th anniversary of the opening of the International Congress on Physics in Italy (Como, Pavia, and Rome). At the famous Congress among many grandees of physics was and prof. Kramers (photo in Figure 1). Kramers made a report “Diffusion of light by atoms” [1], which in the future was predetermined to be as reference in the thousands scientific articles. The edition [1], the title page of which is in Figure 2, there is very rare¹.

So, in his report, Kramers finally formulated the dispersion relations, which we have been using for nearly a century with the name of the Kramers-Kronig (KK) relations. They state that the real and imaginary parts of the dielectric susceptibility (DS) $\zeta(\omega) = \Re(\zeta) + i\Im(\zeta) \equiv \xi(\omega) + i\eta(\omega)$ are related by integral equations:

$$\xi(\omega) = -\frac{2}{\pi} \wp \int_0^{\infty} \frac{\omega' \eta(\omega') d\omega'}{\omega'^2 - \omega^2}, \quad (1)$$

$$\eta(\omega) = \frac{2}{\pi} \wp \int_0^{\infty} \frac{\omega \xi(\omega') d\omega'}{\omega'^2 - \omega^2}, \quad (2)$$

where \wp means that the integral is in the sense of Cauchy's principal value, ω is a circular frequency. To this pair of equations the other pair is equivalent, which follows from the parity of the function $\xi(\omega)$, $\xi(-\omega) = \xi(\omega)$, and the oddness of the function $\eta(\omega)$, $\eta(-\omega) = -\eta(\omega)$,

$$\xi(\omega) = -\frac{1}{\pi} \wp \int_{-\infty}^{\infty} \frac{\eta(\omega') d\omega'}{\omega' - \omega}, \quad (3)$$

$$\eta(\omega) = \frac{1}{\pi} \wp \int_{-\infty}^{\infty} \frac{\xi(\omega') d\omega'}{\omega' - \omega}. \quad (4)$$

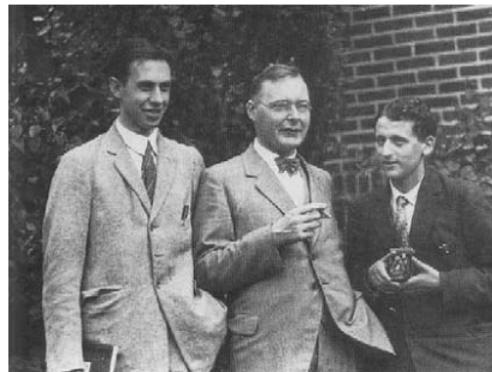


Figure 1. Hendrik Kramers, George Uhlenbeck (left), and Samuel Goudsmit (right).

Kramers derived these formulas two years earlier and reported to the Royal Academy in Copenhagen. A partially similar result were obtained at this time by Kronig [2], and by Kallman and Mark [3]. This is emphasized in the article of Kramers [1]. The formula (2) was first obtained by Kramers, Kronig mentions this much more later in the article [4].

¹ We will refer the reader to the site of the Lorentz Institute at the University of Leiden https://www.lorentz.leidenuniv.nl/IL-publications/sources/Kramers_27.pdf



Figure.2. The title page of the Materials book of the International Congress on Physics in Como, Pavia, and Rome in 1927.

We note that the Kramers-Kronig relations are obtained in the sequence, first (1) – (2), and then (3) – (4), and not vice versa [1, 5], as is often stated in the literature. This is due to the fact that Kramers used the Sellmeier formula [1],

$$\zeta = \sum_k \frac{e^2}{m} \frac{f_k}{\omega_k^2 - \omega^2},$$

which he attracted both to classical and to quantum dispersion theory [6, 7].

If to be consistent, it should be noted that Kronig derived a different relation. He obtained a relation for the refractive index [2]

$$n(\omega) = 1 + \frac{c}{2\pi^2} \oint_0^\infty \frac{\alpha(\omega') d\omega'}{\omega'^2 - \omega^2}, \quad (5)$$

where $\alpha = 4\pi\kappa/\lambda = 2\omega\kappa/c$ is the absorption coefficient, κ is the extinction coefficient, c is the speed of light in vacuum.

The Kramers relations (1) – (2) and (3) – (4) are absolutely exact and legitimate, since the dielectric susceptibility $\zeta(\omega)$ is a fundamental material response function. This applies in full, naturally, to the permittivity $\epsilon(\omega) = \zeta(\omega) + 1$. However, the function of the complex refractive index $v(\omega) = n(\omega) + ik(\omega)$ ($-k = \kappa$) is not such, and the Kramers-Kronig relations for it (more precisely, for $v(\omega) - 1$) are valid [8]. This is confirmed by practice.

Following Landau's assertion [9], the only essential property of the dielectric permittivity function (dielectric susceptibility) that is used in the derivation of the Kramers-Kronig formulas is the absence of singular points in the upper half-plane². There are known proofs of the

Kramers-Kronig relations [10 – 19], which became textbook ones, in which, in fact, the requirement of analyticity of a DS function in one of the half-planes is mandatory. We intend to clarify this issue using the calculation technique, investigating the function DS for the presence of singular points, poles. This is the first. The second question, which we will be dealing with, is how the KK relations are fulfilled, depending on the kind of function of application.

2 Theory and Calculations

Our approach is simple, to describe the reaction of the medium to an external electromagnetic field, that is, to construct function $\zeta(\omega)$ we use the microscopic electromagnetic Lorentz's theory. According to the microtheory, the equation of motion of a charge (electron) in an external periodic field has the form [20]:

$$m\ddot{\mathbf{r}} + \frac{2e^2}{3c^3} \ddot{\mathbf{r}} + k\mathbf{r} = eE_0 e^{i\omega t}, \quad (6)$$

where e , m are charge and mass of an electron, respectively, c is the light speed in vacuum, ω is external wave frequency. The first term in (14) expresses Newton's second law, the third is Hooke's law, so the coefficient k is called a quasielastic constant. This equation is exact in the sense that the second term in it expresses the law of conservation of energy. Its solution contains a complex amplitude through which we calculate the dielectric susceptibility:

$$\zeta(\omega) = \frac{A}{\omega_0^2 - \omega^2 + i\Gamma\omega^3}, \quad (7)$$

where

$$\Gamma = \frac{2e^2}{3mc^3}, \quad (8)$$

$\omega_0^2 = k/m$ is eigenfrequency of an electron, $A \sim (e^2/m)E_0$. Kramers in the original used exactly this expression.

In practice, an approximate equation is used, which, as it turns out, describes dispersion very well. It is obtained by replacing the third derivative with the first derivative under the condition of almost periodic motions:

$$\ddot{\mathbf{r}} + \gamma\dot{\mathbf{r}} + \frac{k}{m} \mathbf{r} = \frac{e}{m} E_0 e^{i\omega t}, \quad (9)$$

where $k/m = \omega_0^2$, and

$$\zeta(\omega) = \frac{A}{\omega_0^2 - \omega^2 + i\gamma\omega}. \quad (10)$$

² Landau considered the amplifying medium, therefore, singular points are located in the lower half-plane. We next consider the absorbing medium, in this case the essentially singular points are located in the upper half-plane.

In another method [21], an expression is found that unites our two cases (7) and (10): $\zeta \sim 1/[\omega_0^2 - \omega^2 + i(\gamma\omega + \tau\omega^3)]$. Its use is superior only in convenience.

The function $\zeta(\omega)$ is a complex-valued function of a real variable ω . In order to investigate its properties, it is advisable to consider it as a function of the complex variable $z = \omega + i\omega_{im} \rightarrow \omega$. This is accepted in mathematical analysis, and this technique is used in all known proofs of the Kramers-Kronig relations. As a result, formula (7) turns into formula

$$\zeta(\omega) = \frac{A}{\omega_0^2 - z^2 + i\Gamma z^3}, \tag{11}$$

where $z = \omega + i\omega_{im}$.

So, we will find the poles for the functions represented by formulas (7) and (10). Clearly, we must find the zeros of the denominator. The first case, let the denominator in the unfolded form will equate to zero:

$$[\Gamma\omega_{im}^3 - 3\Gamma\omega^2\omega_{im} + \omega_{im}^2 - \omega^2 + \omega_0^2] + i\omega[\Gamma\omega^2 - 3\Gamma\omega_{im}^2 - 2\omega_{im}] = 0. \tag{12}$$

The condition for the fulfillment of relation (12) is the zero values of the real and imaginary parts of this relation, which in turn is equivalent to a system of two equations of two unknowns ω and ω_{im} . The values of these unknowns determine the poles. The first of them is easy to notice at once, with ω equal to zero the imaginary part vanishes, and the real part vanishes when the cubic equation is valid:

$$\Gamma\omega_{im}^3 + \omega_{im}^2 + \omega_0^2 = 0. \tag{13}$$

As is easy to see, this equation has a real solution, which can only be negative. Thus, we have a pole in the lower half-plane.

We find two more poles by putting from the imaginary part

$$\omega = \pm \sqrt{3\omega_{im}^2 + \frac{2}{\Gamma}\omega_{im}} \tag{14}$$

into the real part and equating it to zero:

$$\omega_{im}^3 + \frac{1}{\Gamma}\omega_{im}^2 + \frac{1}{4\Gamma^2}\omega_{im} - \frac{1}{8\Gamma}\omega_0^2 = 0. \tag{15}$$

This cubic equation has one real and two imaginary roots. Using the real root and formula (13), we obtain two more symmetric poles in the upper half-plane.

In the case of formula (10), it is easy to show that the function $\zeta(\omega)$ has two poles in the upper half-plane. A pair of poles can be of two types. First, $\omega \neq 0$:

$$\omega_{im} = \frac{\gamma}{2}, \quad \omega = \pm \sqrt{\omega_0^2 - \frac{\gamma^2}{4}}. \tag{16}$$

The second, $\omega = 0$:

$$\omega_{im} = \frac{\gamma}{2} \pm \sqrt{\frac{\gamma^2}{4} - \omega_0^2}. \tag{17}$$

Under condition $\gamma^2/4 = \omega_0^2$, the two poles degenerate into one pole in the upper half-plane on the imaginary axis: $\omega = 0, \omega_{im} = \gamma/2$.

Thus, when the function $\zeta(\omega)$ is described by the formula (10), it has no singularities in the lower half-plane. The KK's relations will be satisfied for any reasonable values of the parameters ω_0, A , and γ . In this connection, let us return to the formula (7) and analyze how the KK's relations are satisfied in the presence of poles in both the upper and the lower half-planes.

In order for function $\zeta(\omega)$ to satisfy KK's relations, we set the parameters in formulas (7) and (10) so that function $\zeta(\omega)$ approximately approximates the dielectric susceptibility of silicon. The data for silicon are obtained from remarkable work [22], where the dielectric permittivity for silicon, germanium, and crystals of class $A^{III}B^V$ in the range [1.5, 6.0] with a step of 0.1 eV is given. DS function for silicon and for model functions of DS according to formula (7) and formula (10) are shown in Figure 3.

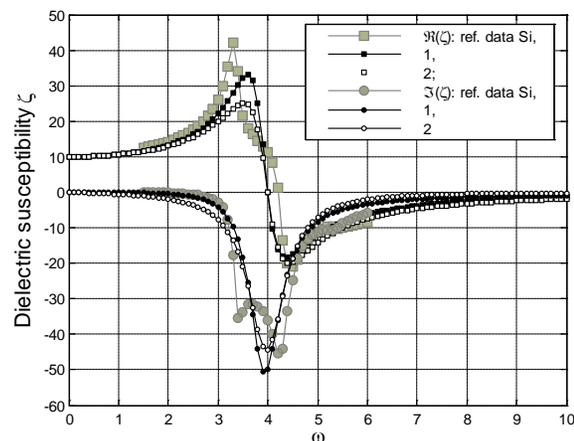


Figure 3. The general form of the DS model functions according to formula (7) (1 - A = 160, $\omega_0 = 4.0, \Gamma = 0.05$), and formula (10) (2 - A = 160, $\omega_0 = 4.0, \gamma = 0.9$).

In order to verify how the KK's relations for the function under investigation are fulfilled, we must first of all choose the method of calculating integrals in the sense of the Cauchy principal value, which are used everywhere in the Kramers-Kronig analysis. To do this, the discrete integration operator in the form of the formula of trapeziums is quite useful:

$$\int_a^b f(x)dx = \Delta x \{ f(a)/2 + f(a + \Delta x) + f(a + 2\Delta x) + \dots + f(a + [N - 2]\Delta x) + f(b)/2 \}, \quad (18)$$

where Δx is the integration step, N is the number of points of integration. The points that invert the integral to infinity, we simply omit.

The calculation result for the formula (7) in range [-30, 30] with step 0.1 is shown in Figure 4. (Here and further in this paper, the units of frequency ω are not indicated, since in view of the specificity of the KK's relations, any scale factor for ω under the integral sign will be reduced, and the ω will be dimensionless. In our case, we should consider ω as energy in eV).

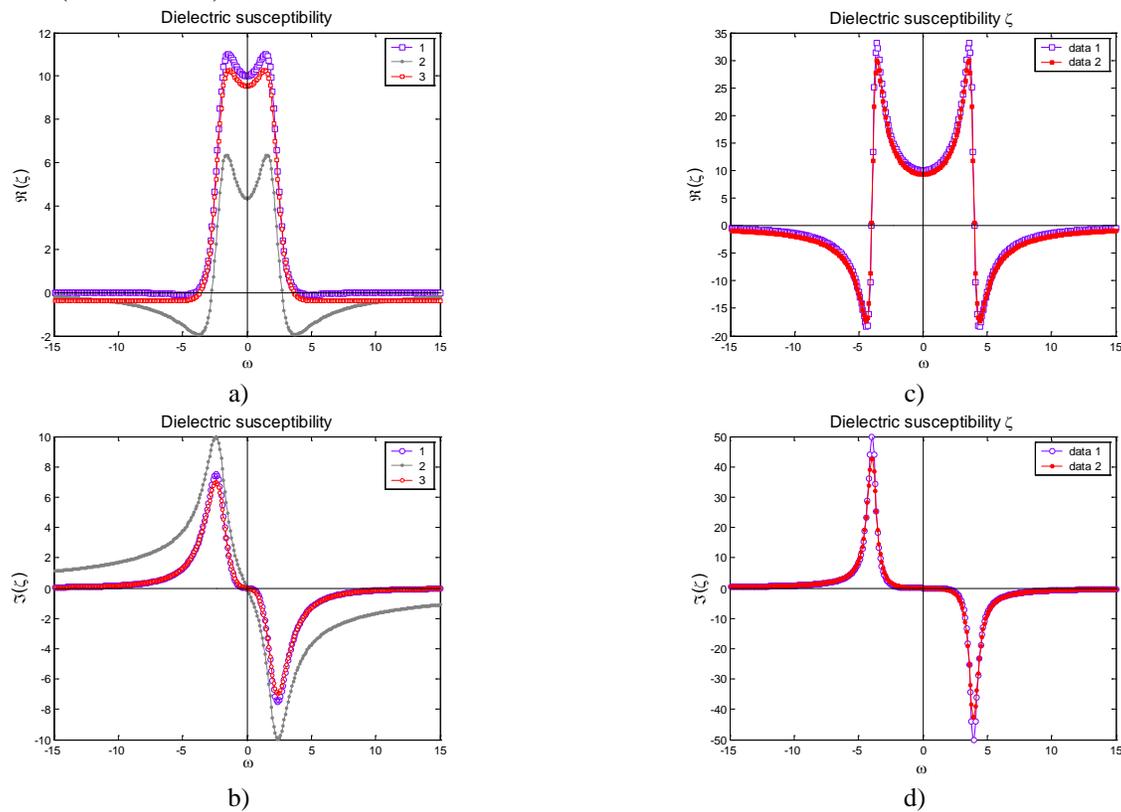


Figure 4. Plots of the DS model function by formula (7) in two principal cases: a), b) – $A = 160, \omega_0 = 4.0, \Gamma = 1$: the KK relations are not satisfied; c), d) – $A = 160, \omega_0 = 4.0, \Gamma = 0.05$: the KK relations are satisfied. For graphs a) – b), data 1 is the initial form of the function, data 2 is the form of the function after the first transform, and data 3 is the form of the function after the second transform by KK's relations; for graphs c) – d), data 1 is initial, data 2 is after the first and second transformation by KK's relations.

In the first case (Figure 4, a, b), we moved away from the values of the parameters according to Figure 3 and put $\Gamma = 1$. As a result, as we see, the KK's relations are not satisfied. However, we went further and applied the transformation once more to the result obtained. We came to an unexpected situation – the relations are being fulfilled. In the case when the parameters correspond to the values from Figure 3, as expected, the relations are satisfied (Figure 4, c, d). Moreover, the results of the first and second transformations coincide perfectly. Thus, some difference for the graphs of the initial function and the function after the first transformation is not a consequence, as it would seem, of the error of calculations, but a consequence of the properties of the function itself, which is to be transformed. We return to this important discussion, but first we will clarify the difference between the analytic properties of the function $\zeta(\omega)$ in the two cases considered. To do this, we construct contour graphs of lines of the same level for the complex function $\zeta(z)$ according to formula (11), see Figure 5. On these graphs, the poles of the function can clearly be seen, the positions of which can be calculated according to formulas (13) – (15).

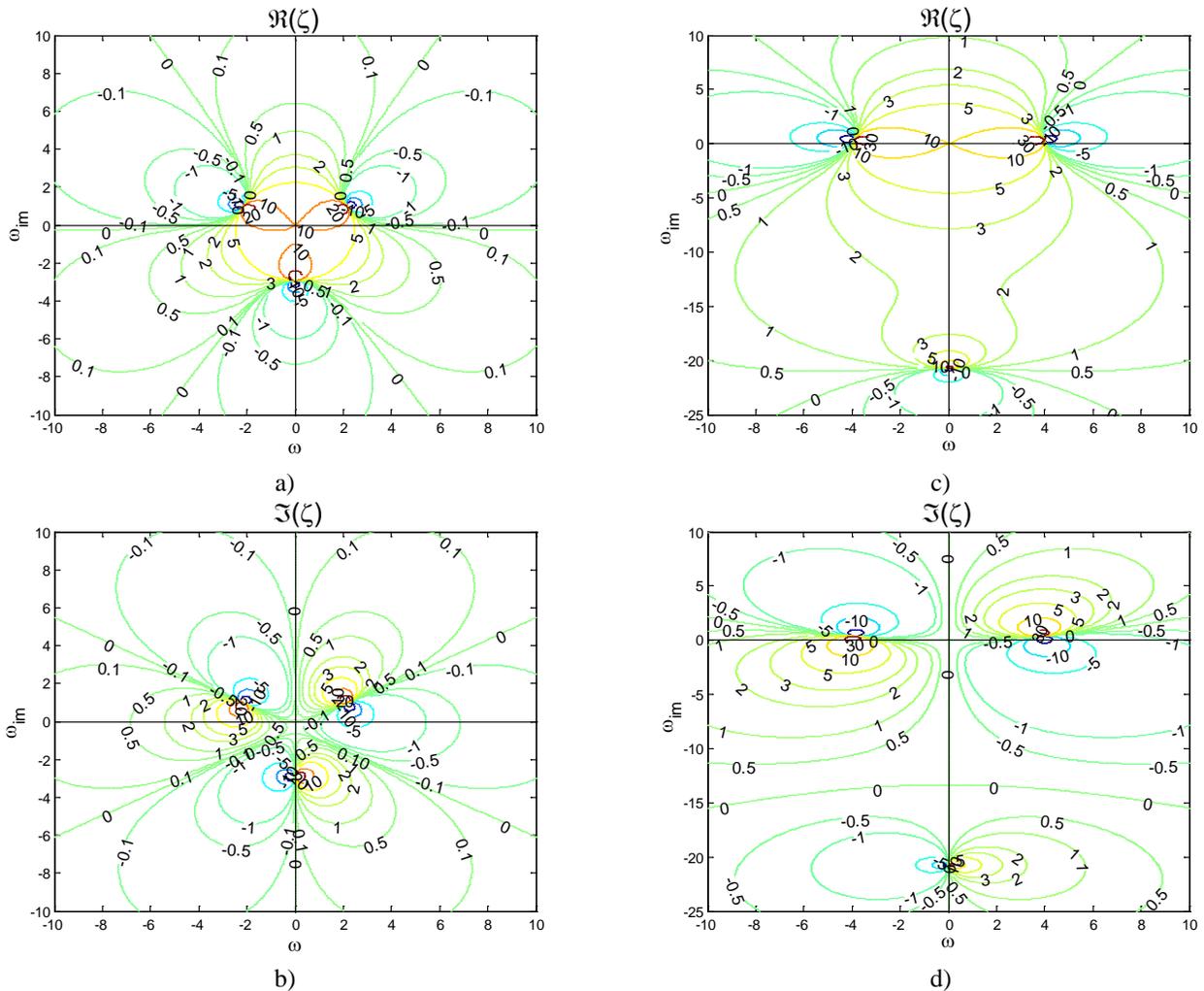


Figure 5. Contour plots of the model DS function $\zeta(z)$ by formula (11), $z = \omega + i\omega_{im}$, in the complex plane z for the cases shown in Figure 4: a), b) – $A = 160$, $\omega_0 = 4.0$, $\Gamma = 1$; c), d) – $A = 160$, $\omega_0 = 4.0$, $\Gamma = 0.05$; a), c), and b), d) are real and imaginary parts, respectively.

The Figure 5 shows for the dielectric susceptibility function two poles in the upper half-plane, of which we have heard so much, but have not seen; and one pole in the lower half-plane, of which we have not heard and seen. If the parameter Γ is sufficiently reduced, the pole on the imaginary axis will shift to side $-\infty$, and the Kramers-Kronig relations will be satisfied. We came to an important conclusion. From the condition that in the upper

half-plane the function has poles, and in the lower half-plane it is analytic (holomorphic), it follows that the Kramers-Kronig relations hold for it. However, from the condition that the Kramers-Kronig relations hold for a function it does not follow that the function is analytic only in one of the half-planes.

Now we turn, we can say, to the second part of our work, and consider the application of the KK relations to

functions that do not have the form of a classical response function. Considering the dielectric susceptibility function, we can intuitively assume that the response function must have properties. It must be a complex-valued function of the real argument. Its real and imaginary parts must vanish at infinity $\pm\infty$. Its real part is an even function, and the imaginary part is an odd function. Its real part changes sign on the positive (negative) axis, and the imaginary part does not change. Other functions, in addition to function DS, have these properties. For example, first of all, the functions associated with the dielectric permittivity $\epsilon(\omega) - 1$, with the complex refractive index $\nu(\omega) - 1$, and the reflection coefficient

$$r(\omega) = \frac{1 - \sqrt{\epsilon(\omega)}}{1 + \sqrt{\epsilon(\omega)}} \quad (19)$$

The consideration of the reflection coefficient has only theoretical value, in practice the use of the reflection coefficient has no significance. Neither the real nor the imaginary part can be measured separately by experiment. Then, as the logarithm of the reflection coefficient and the KK analysis applied to it, has great importance for optics and spectroscopy. This value is so large in the optics of semiconductors that we must make a small digression and touch upon this problem.

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With the beginning of the 50's semiconductor optics developed rapidly. The determination of the optical constants from the reflection and absorption spectra was the first task. In the pioneering works of Avery, Robinson and Price [23 – 25] the idea of determining optical constants from the spectra of the fundamental reflection of optical crystals was put forward. The formulation of the problem was clear, it is necessary to apply the Kramers-Kronig analysis to the function

$$\ln r = \ln |r| + i\theta, \quad (20)$$

that is, to the logarithm of the complex reflection coefficient $r = |r|e^{i\theta}$. Applying the Kramers-Kronig analysis by formulae (1) and (2) to the function (7), we obtain equations:

$$\ln|r(\omega)| = -\frac{2}{\pi} \oint_0^\infty \frac{\omega'\theta(\omega')d\omega'}{\omega'^2 - \omega^2}, \quad (21)$$

$$\theta(\omega) = \frac{2\omega}{\pi} \oint_0^\infty \frac{\ln|r(\omega')|d\omega'}{\omega'^2 - \omega^2}. \quad (22)$$

Since from the experiment we have a reflection spectrum $|r(\omega)|^2$, then applying the second relation (9), we obtain the

phase angle $\theta(\omega)$ of the reflection coefficient, and hence the optical constants – real refractive index n and extinction coefficient κ :

$$n = \frac{1 - |r|^2}{1 + |r|^2 - 2|r|\cos\theta}, \quad (23)$$

$$\kappa = \frac{-2|r|\sin\theta}{1 + |r|^2 - 2|r|\cos\theta}. \quad (24)$$

However, things did not go as good as they wanted. Several years later Jahoda found the key to the cherished casket. He, first in his dissertation [26], and then in [27] showed how to calculate this phase. For this Jahoda used the theory of networks in the Bode idea [28]. According to Bode and Jahoda

$$\theta(\omega) = \frac{1}{\pi} \int_0^\infty \ln \left| \frac{\omega' + \omega}{\omega' - \omega} \right| \frac{d \ln |r|}{d\omega} d\omega'. \quad (25)$$

This relationship for the phase in the form of a formula (12) is densely included in many articles, books and textbooks [29 – 53]. We note that an investigation of the optical properties of silicon, germanium, and some other classical semiconductor crystals was carried out with the help of this formula.

Another approach to solving equation (9) was suggested by Plaskett and Schatz [54 – 56]. According to [54],

$$\theta(\omega) = -\frac{2\omega}{\pi} \oint_0^\infty \frac{\ln|r(\omega')|d\omega'}{\omega'^2 - \omega^2} + \left[\pi - 2 \arctan \frac{\beta}{\omega} \right]. \quad (26)$$

Here, β is the imaginary part of the frequency, which can tend to 0 or ∞ .

3 Example

Let's consider an example of silicon from the already cited work [22]. According to the reflectivity data in the range [1.5, 6.0] eV, we construct the function $\ln|r(\omega)|$ in the range [-6.0, 6.0], defining it on the interval (-1.5, 1.5) using model data. We apply to this function the KK transformation according to formula:

$$\theta(\omega) = \frac{1}{\pi} \oint_{-\infty}^\infty \frac{\ln|r(\omega')|d\omega'}{\omega' - \omega}.$$

As a result, we obtain curve 2 in Figure 7. To determine the angle of rotation, we must use several control points in

which the dielectric constant (refractive index and extinction coefficient) is reliably known, measured by some other method (the prism method in the transparency region, or from the absorption spectrum in the region of moderate absorption). The figure shows that this angle is equal to the sum of the slope angles modules of the lines, which are the linear trend of the given and the determined curves. In Figure 6 curve 1 is calculated from the total data of silicon, and curve 3 is determined by the rotation of curve 2. As can be seen, the angle of rotation is $-(\arctan|0.084| + \arctan|-0.06|)$. The obtained refractive index for silicon is shown in Figure 7. Note that we intentionally did not put model data in the range $(6.0, \infty)$. As a result, as we see, there are "loose" tails appear on the edge of the interval, there is no accuracy. This underconversion in KK transform can be eliminated by expanding the measuring range of reflectivity and continuing its model data.

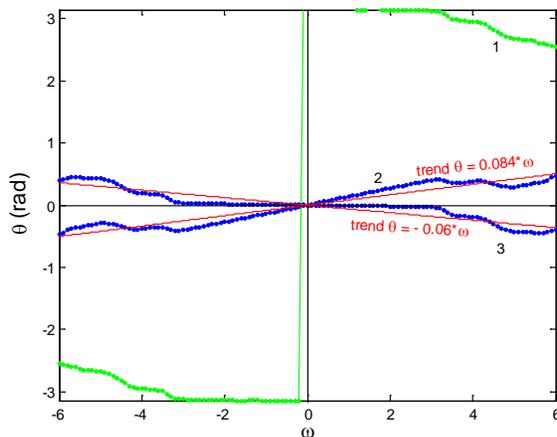


Figure 6. To the determination of the phase function for silicon.

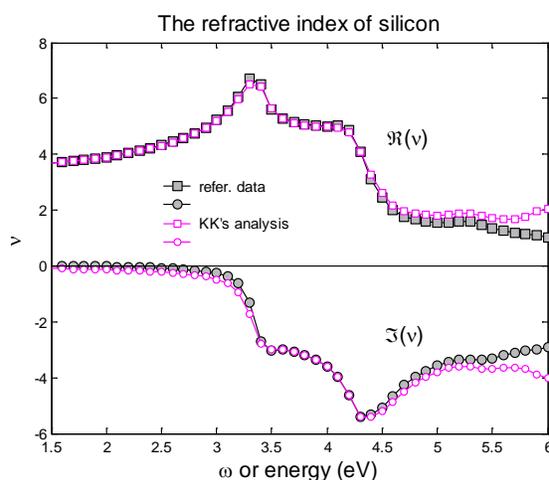


Figure 7. The refractive index of silicon is determined from the reflection spectrum.

4 Conclusion

The current state of materials science is such that it is technologically possible to create new types of media - nanostructured media, composite materials, in general the so-called metamaterial media. This leads to new statements of problems for electrodynamics and optics of condensed matter. In this respect, the Kramers-Kronig analysis will continue to play a large role in connection with its fundamental origin. We see the prospects as follows. On the one hand, the application of KK relations makes it possible to verify the values of optical characteristics for well-studied media, such as semiconductor crystals, for example. On the other hand, it is necessary to develop the KK method itself in the theoretical sense. This article has been, to a greater extent, devoted to this problem.

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