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METHODS FOR SOLVING PROBLEMS ON THE DETERMINATION OF OPTICAL CONSTANTS FROM THE ABSORPTION AND REFLECTION SPECTRA BY THE KRAMERS – KRONIG ANALYSIS

The historical aspect of the origin of the Kramers-Kronig dispersion relations is considered. The theoretical basis of the Kramers-Kronig analysis is shown, namely, the dielectric susceptibility function is a fundamental function of the linear response, since it is obtained from the motion equation of the oscillator in an external field in the microscopic electromagnetic Lorentz's theory. Two aspects of the application of the Kramers-Kronig relationships and related ratios in spectroscopy are shown – the determination of the optical characteristics of the absorbing medium from the absorption (transmission) spectra and reflection spectra. A new approach to the application of the Kramers-Kronig relations is proposed, to some extent, – a verification of known reference data on the optical constants of well-studied materials, crystals, glasses, and other.

Keywords: Kramers-Kronig relations, Robinson relation, transmission spectra, absorption spectra, reflection spectra

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 Fig. 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 Fig. 2, there is very rare.

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 relations (KKR). They state that the real and imaginary parts of the dielectric susceptibility $\zeta(\omega) = \xi(\omega) + i\eta(\omega)$ are related by integral equations:

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

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

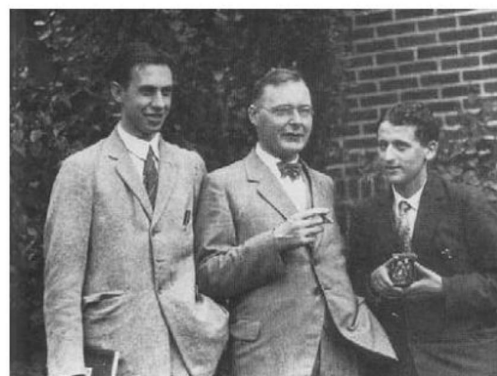


Fig. 1. George Uhlenbeck (left) and Samuel Goudsmit (right), with Hendrik Kramers who first noticed a twofold degeneracy in the solutions to the Schrödinger equation with spin: the Kramers degeneracy. All three were students of Paul Ehrenfest in Leiden.

where \wp means that the integral is in the sense of Cauchy's principal value, ω is a circular frequency.

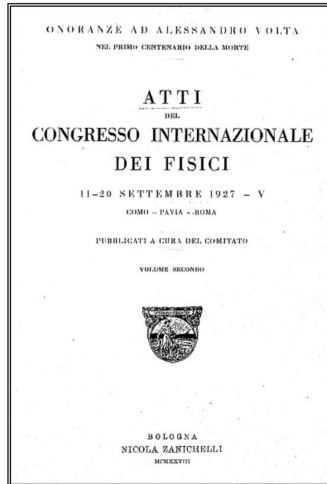


Fig. 2. The title page of the Materials book of the International Congress on Physics in Como, Pavia, and Rome in 1927.

To this pair of equations the other pair is equivalent, which follows from the parity of the function ξ , $\xi(-\omega) = \xi(\omega)$, and the oddness of the function η , $\eta(-\omega) = -\eta(\omega)$, :

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

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

Formula (1) Kramers obtained two years earlier and reported to the Royal Academy in Copenhagen. It was also received in the equivalent form 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].

We note that the Kramers-Kronig relations are obtained in the sequence, first (1) – (2), and then (3) – (4), and not vice versa, as is often stated in the literature. This is clear, because Kramers was fascinated by the quantum theory of dispersion, where the polarization function has the form of type in (1) – (2) [5 – 7].

There are several approaches to the proof of the KKR, and the relations themselves are discussed in hundreds of articles, therefore in

this paper there is no way at least superficially to analyze them. For the reader who is interested in the subject matter, the author recommends [8 – 18] and the literature in them.

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

$$n(\omega) = 1 + \frac{c}{2\pi^2} \wp \int_0^{\infty} \frac{\alpha(\omega') d\omega'}{\omega'^2 - \omega^2}, \quad (5)$$

where $\alpha = 4\pi k/\lambda = 2\omega k/c$ is the absorption coefficient, k is the extinction coefficient, c is the speed of light in vacuum. Thus, he applies the Kramers procedure to a complex index of refraction, more precisely to $(n - 1) - ik$:

$$n(\omega) - 1 = \frac{2}{\pi} \wp \int_0^{\infty} \frac{\omega' k(\omega') d\omega'}{\omega'^2 - \omega^2}. \quad (6)$$

The relation (6) is not as good as (1). It is good for the X-ray region where Kronig was investigated, or for gas, when the real part of refractive index is close to 1.

The Kramers relations (1) - (2) and (3) - (4) are absolutely exact and legitimate, since the dielectric susceptibility $\zeta(\omega)$ is a linear response function and the fundamental causality principle is applied to it [13, 14, 16 – 19]. This applies in full, naturally, to the permittivity $\varepsilon(\omega) = \zeta(\omega) + 1$. However, for functions related to $\zeta(\omega)$ or $\varepsilon(\omega)$, i.e. elementary functions of them, the application of the Kramers-Kronig analysis requires additional discussions. First of all, the refractive index is a function $\sqrt{\varepsilon(\omega)}$. Kramers himself, summing up, emphasized that it would be interesting to check whether his and Kronig's relations work for media that are denser than gas. The Kronig relation (6) is also satisfied well because only in the X-ray region of the spectrum the refractive index is only slightly greater than 1.

A new period in the history of the Kramers-Kronig analysis began with the onset of an intensive study of the properties of semiconductors in the early 1950s, namely, their optical properties. Then a new problem arose: the determination of the optical constants

of solids, mainly crystals, from their fundamental reflection spectra. Beginning with the works of Avery, Robinson and Price [20 – 22], the formulation of the problem was clear. It is necessary to apply the Kramers-Kronig analysis to the function

$$\ln r = \ln |r| + i\theta, \quad (7)$$

that is, to the logarithm of the complex reflection coefficient $r = |r| e^{i\theta}$.

Applying the Kramers-Kronig analysis to the function (7), we obtain equations:

$$\ln |r(\omega)| = -\frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\theta(\omega') d\omega'}{\omega' - \omega}, \quad (8)$$

$$\theta(\omega) = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\ln |r(\omega')| d\omega'}{\omega' - \omega}. \quad (9)$$

We shall call equations (8) – (9) logarithmic Kramers – Kronig relations.

Since, with the experiment we have a reflection spectrum $|r(\omega)|^2$, then applying the second KK relation (2) or (4) to (7), we get the phase θ . The determination of the optical constants is then carried out according to the formulae:

$$n = \frac{1 - |r|^2}{1 + |r|^2 - 2|r| \cos \theta}, \quad (10)$$

$$k = \frac{-2|r| \sin \theta}{1 + |r|^2 - 2|r| \cos \theta}. \quad (11)$$

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 [23], and then in [24] showed how to calculate this phase. For this Jahoda used the theory of networks in the Bode idea [25, 26]. According to Bode and Jahoda

$$\theta(\omega) = \frac{1}{\pi} \int_0^{\infty} \ln \left| \frac{\omega' + \omega}{\omega' - \omega} \right| \frac{d \ln |r|}{d\omega'} d\omega'. \quad (12)$$

This relationship for the phase in the form of a formula (12) is densely included in many articles, books and textbooks [27 – 46]. 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 (13) was suggested by Plaskett and Schatz [47, 48]. According to [47],

$$\theta(\omega) = -\frac{2\omega}{\pi} \int_0^{\infty} \frac{\ln |r(\omega')| d\omega'}{\omega'^2 - \omega^2} + \left[\pi - 2 \arctan \frac{\beta}{\omega} \right]. \quad (13)$$

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

Both approaches, by Jahoda, and by Plaskett and Shatz, are very highly theoretical. At least, we do not see works in which it is transparently demonstrated how from the reflection spectrum we can get the phase, what is the accuracy of the method, etc. As a few exceptions, we can point to articles that discuss the practical application of these methods and attempts to implement them numerically [49 – 58].

The aim of this article is to show that calculations based on Kramers-Kronig analysis are not difficult to produce. However, it is very important to take into account the reliability of the initial data, primarily the width of the measuring range, the accuracy of the measurements and the measurement step.

2 Theory, Methods, and Calculations

The motion equation of a charge (electron) in an external periodic field has the form [59]:

$$m\ddot{\mathbf{r}} - \gamma\dot{\mathbf{r}} + \kappa\mathbf{r} = E_0 e^{i\omega t}, \quad (14)$$

$$\gamma = \frac{2e^2}{3c^3}, \quad (15)$$

where e , m are charge and mass of an electron, respectively, c is the light speed in vacuum, κ is a quasielastic constant, ω is external wave frequency. Its solution contains a complex

amplitude through which we calculate the dielectric susceptibility. Kramers, in the original, writes as follows:

$$\zeta(\omega) = \frac{e^2}{m} \frac{1}{\omega_0^2 - \omega^2 + i\delta}, \quad (16)$$

where

$$\delta = \frac{2e^2\omega^3}{3mc^3}, \quad (17)$$

ω_0 is eigenfrequency of an electron. The equation (14) is "exact," the first term expresses Newton's second law, the third is Hooke's law, and the second term is necessary to fulfill the law of conservation of energy. 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:

$$m\ddot{\mathbf{r}} + m\Gamma\dot{\mathbf{r}} + \kappa\mathbf{r} = E_0 e^{i\omega t}, \quad (18)$$

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

$$\zeta(\omega) = \frac{A}{\omega_0^2 - \omega^2 + i\Gamma\omega}. \quad (19)$$

Now we'll show that relations (3) - (4) are satisfied with any preassigned accuracy, if the final interval of integration $[-a, a]$ is increased, and the step of the integration is decreased. To do this, we replace the integral by a discrete operator, for example, according to the formula of trapeziums

$$\int_a^b f(x) dx = \Delta x \{ f(a)/2 + f(a + \Delta x) + f(a + 2\Delta x) + \dots + f(a + [N-2]\Delta x) + f(b)/2 \}, \quad (20)$$

where Δx is the integration step, N is the number of points of integration. The points that invert the integral into infinity, we simply miss. It turns out that any attempts to improve the

accuracy of calculations by mathematical methods in our case are completely inappropriate. The accuracy is determined solely by the accuracy of the experiment.

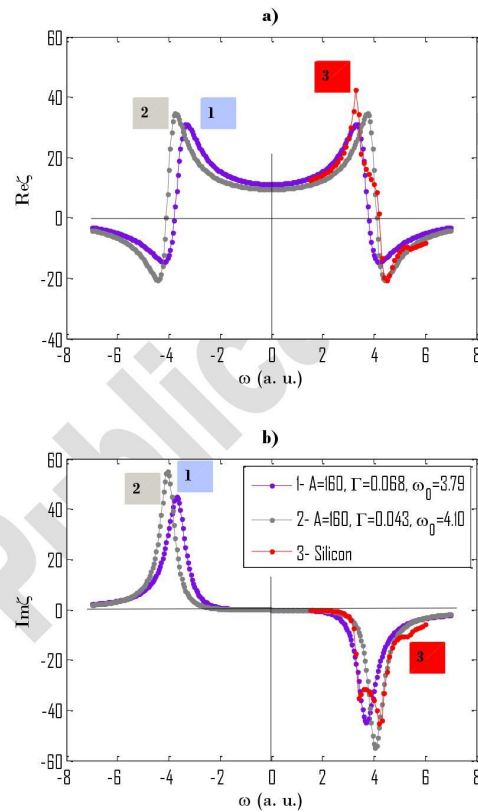


Fig. 3. The general form of the model function of dielectric susceptibility according to formula (19), its real part (a), and the imaginary part (b).

Fig. 3 shows the general form of the model function of the dielectric susceptibility according to equation (19). The parameters A , Γ and ω_0 are chosen in such a way as to approximate the model function to the real function for silicon. We took the data for silicon from a remarkable paper [44].

Applying the Kramers-Kronig transform to the function of the dielectric susceptibility constructed in this way according to formula (19), we must obtain the same function. As the numerical experiment shows, we can achieve arbitrarily high accuracy. It is only important choose a sufficiently wide interval of integration and a sufficiently small step of

integration. In Fig. 4 shows the result of the calculation in the range from -25 to 25 (the units of measurement are arbitrary, they correspond to eV in the experiment). The integration step is 0.1, since we adhere to the data of [44].

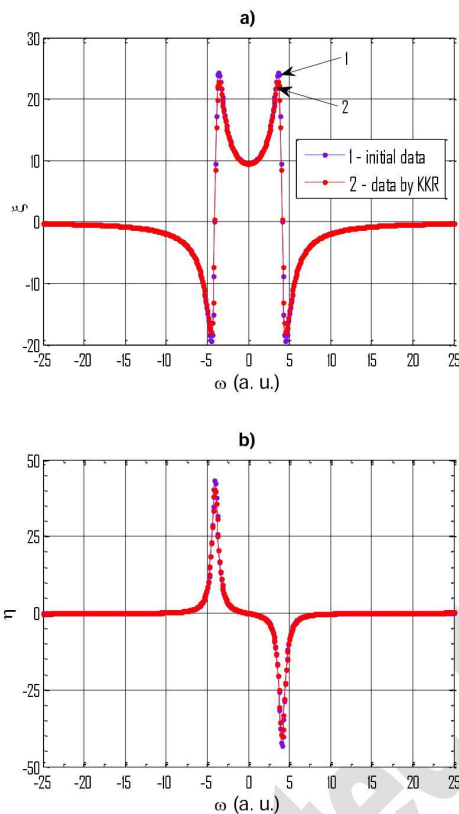


Fig. 4. Plots of the dielectric susceptibility, its real part (a), and the imaginary part (b): the initial data (1) and the Kramers-Kronig transform according to formulas (3) - (4) with a frequency step of 0.1 (2).

Now, we formulate two tasks:

- I) determine the refractive index from extinction coefficient data. As is known, these data can be obtained from transmission (absorption) spectra;
- II) determine the refractive index and extinction coefficient from the reflection spectra data.

As we see, the Kramers-Kronig transformation is performed well theoretically. In practice, a much narrower range of measurements is encountered. Following the same work 1, we will apply the Kramers-Kronig transformation for silicon in the range from 1.5 to 6 eV. Now

we must solve problem 1) directly. To do this, we take the data for silicon from the same article [44]. The problem is that in this work the data is given in a range that is not enough for us.

In the case where the range of application of the KKR is not wide enough, the result is different from the actual function sought. In this regard, there is a need to introduce one or more control points, determined by some other method. In this case, the integral in formulas (3) or (4) should be divided into three parts:

$$X(\omega) = I_0 \pm \frac{2}{\pi} \oint_a^b \frac{\Omega Y(\omega') d\omega'}{\omega'^2 - \omega^2} + I_\infty, \quad (21)$$

where

$$I_0 = \pm \frac{2}{\pi} \oint_0^a \frac{\Omega Y(\omega') d\omega'}{\omega'^2 - \omega^2}, \quad I_\infty = \pm \frac{2}{\pi} \oint_b^\infty \frac{\Omega Y(\omega') d\omega'}{\omega'^2 - \omega^2}.$$

The value of $\Omega = \{\omega, \omega'\}$ is chosen according to the sign before the integral $\{+, -\}$. It turns out that $I_0 + I_\infty$ depends weakly on ω , so we can put a constant for the given value. We select its value in such a way that the function $X(\omega)$ passes through the control point. Note that the control point should not be selected in the "tail" of the integration interval (the "tail" is about 0.5 eV). For silicon, for example, we can use the refractive index and the extinction coefficient as a reference point, which were obtained by ellipsometry at a wavelength of 0.6328 μm (1.96 eV) [Wang].

Thus, applying this approach and using the Kronig formula (5), (6), we solve the problem I). An alternative solution is also possible in the general approach of Kramers. To do this, we will have to apply the method of solving the non-linear integral equation. A method of successive approximations is useful. We choose some zero approximation for the refractive index (even a constant, $n_0 = \text{const}$, see Fig.5) and form a function of the dielectric susceptibility

$$\xi_0(\omega) + i\eta_0(\omega) = [n_0(\omega) - ik(\omega)]^2 - 1. \quad (22)$$

After applying the KKR, we get a new value, and, accordingly, the refractive index $n_1(\omega)$, already as a function

$$n_1(\omega) - ik(\omega) = \sqrt{\xi_1(\omega) + i\eta_1(\omega) + 1},$$

ie.

$$n_1(\omega) = \text{Re} \sqrt{\xi_1(\omega) + i\eta_1(\omega) + 1}. \quad (23)$$

Repeating the procedure after a few iterations yields the result. It depends, of course, on the choice of the zeroth approximation. Therefore, we can, as such, choose the refractive index, which gives the Kronig formula (6). In Fig. 5 shows the result for silicon based on the experimental data of the same work [44].

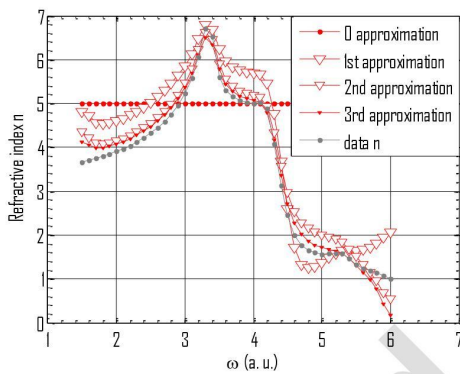


Fig. 5. The result of calculations of the refractive index for silicon by the method of successive approximations.

The solution of problem II) is connected with considerably greater difficulties than problems I), which was already shown in the introduction. Here we can not just put $I_0 + I_\infty = const$. We again consider the example of silicon from [44]. According to the data in the energy range [1.5, 6.0] eV n and k we calculate the complex reflection coefficient

$$r(\omega) = |r| e^{i\theta} = \frac{\sqrt{\epsilon} - 1}{\sqrt{\epsilon} + 1} = \frac{(n - ik) - 1}{(n - ik) + 1}. \quad (24)$$

and $\ln|r|$. Applying the relation (), we calculate the phase $\theta(\omega)$. From formula (24) it is clear that

$$n - ik = \frac{1 + r}{1 - r}, \quad (25)$$

those

$$n - ik = \text{Re} \frac{1 + r}{1 - r} + i \text{Im} \frac{1 + r}{1 - r}, \quad (26)$$

whence, as we see, it is possible to obtain an inverse transformation for finding n and k . The last relation is obviously equivalent to the relations (10) – (11).

Fig. 6 shows graphically the result of applying the Kramers-Kronig logarithmic transformation for two variants. In the first case, the transformation is performed for the range, as in [44], [1.5, 6.0] eV (data 1).

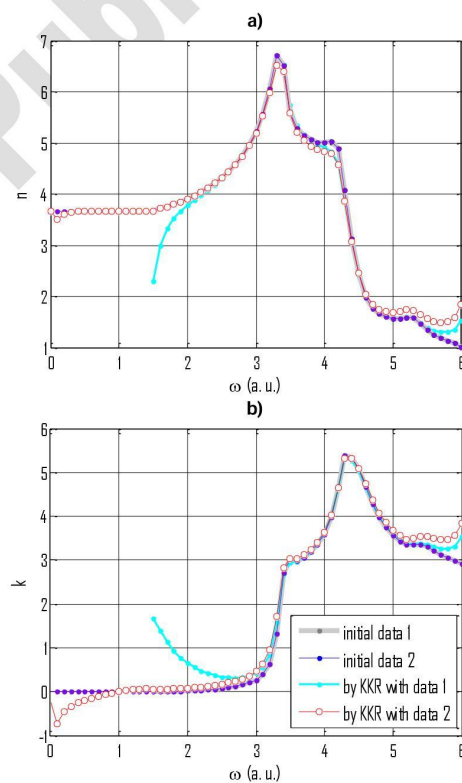
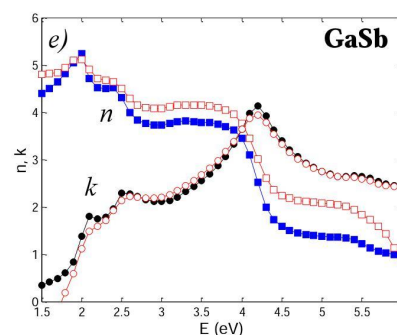
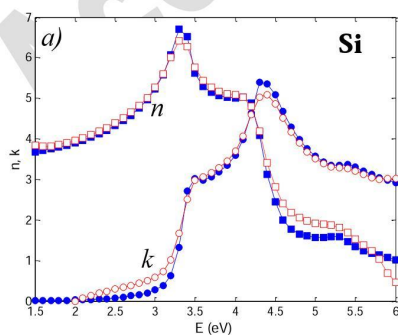
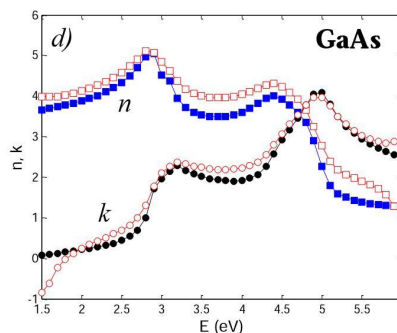
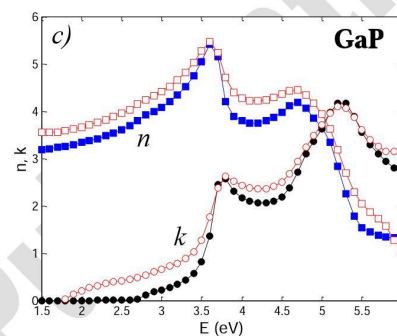
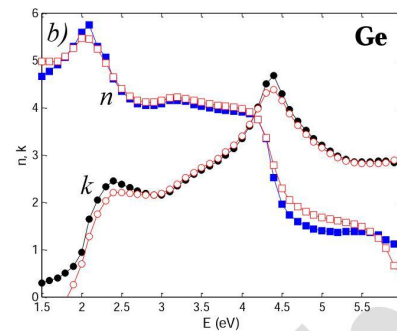


Fig 6. Plots of the refractive index (a) and extinction coefficient (b) of Silicon, obtained as a result of applying the Kramers-Kronig logarithmic transformation; data 1 - range from 1.5 to 6.0 eV, data 2 - range from 0 to 6.0 eV.

As can be seen from Fig. 6, the accuracy is lost on the "tails" [1.5, 2] and [5.5, 6.0]. To improve the result, we put in the range [0, 1.5] $\ln|r|$ everywhere constant and equal to $\ln|r(1.5)|$ (data 2). As you can see, now in the low-energy "tail" we received an excellent agreement. It would be desirable to note, but how not to note here, putting inaccurate data at one end of the range, we lost accuracy at the opposite end (see fig. 6). Apparently, this is a general property of the integral dependence of functions. Consideration of this aspect leads to next topic, which we consider in the next section.

3 Discussion and Result

As was said at the beginning, the optical constants are determined by various methods, depending on the degree of absorption. The Kramers-Kronig dispersion relations are integral relations, and they are satisfied the more accurately, if the broader the spectral region. It follows that only the Kramers-Kronig relations are precisely the exact test that will show whether the data are reliable or not, or to what extent they are reliable. In this context, we applied the Kramers-Kronig analysis to well-studied crystals (Fig. 7). In article [44], numerical data are presented for the canonical crystals of silicon and germanium, and for crystals of group A^{III}B^V. The obvious fact is that, for reliable data, the discrepancy between the initial data and the data as a result of the Kramers – Kronig transformation should be observed only in the "tails" of the spectrum. As we see this is not always the case.



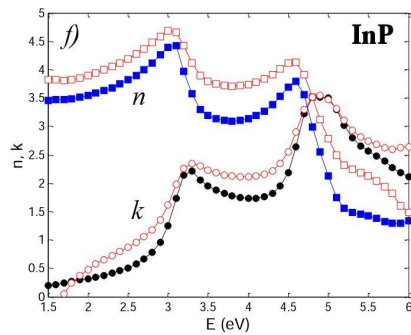


Fig. 7. Graphs of dependence of the refractive index and the extinction coefficient on the quantum energy for a) Silicon, b) Germanium, and crystals of group $A^{III}B^V$: c) Gallium Phosphide, d) Gallium Arsenide, e) Gallium Antimonide, and f) Indium Phosphide. Closed marker – reference data, open marker – values after conversion using the Kramers-Kronig relations.

And finally, we can not get around one more problem. Often it happens that without the numerical data of the experiment, we would like to use the literary data, which are shown in a graphic form. Then, it would seem, you can digitize the desired graphic data. For example, it is done in the preparation of reference publications [32]. In such cases, the Kramers-Kronig analysis is possible in exceptional cases, when greater accuracy of scanning and digitization can be ensured. We will set an example for demonstrating a measure of loss of accuracy. We took the work [60], scanned and digitized the data for the fundamental reflection spectrum of As_2Se_3 glass. The result of the application of the Kramers-Kronig analysis is shown in Fig. Of course, in many practical cases it is very important to have at least rough estimates of the optical parameters of the material.

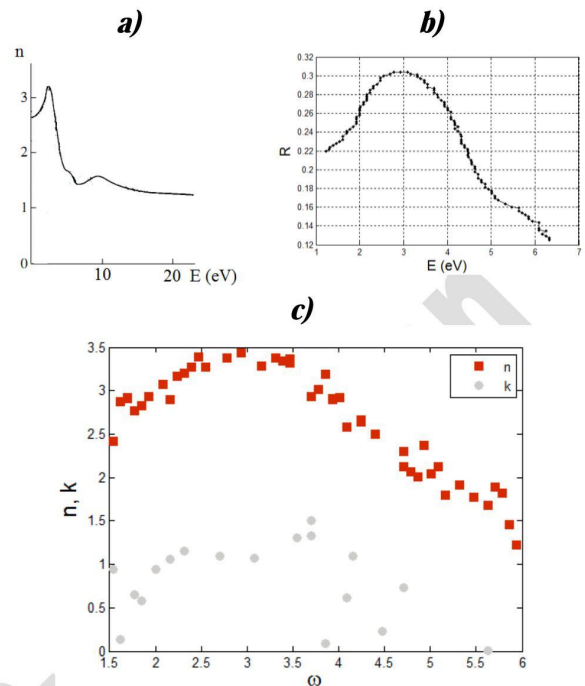


Fig. 8. Graphical demonstration of calculations of the refractive index and extinction coefficient using the Kramers-Kronig logarithmic relation with fuzzy data for the glass As_2Se_3 reflection coefficient: a) the refractive index of the glass from Ref. [60]; b) the plot of the reflectivity obtained by scanning the graphic data from Ref. [60]; c) point plots of the refractive index and extinction coefficient obtained as a result of the application of the KKR.

4 Conclusions

The main conclusion of this paper is that the application of the Kramers-Kronig relations to determine the optical parameters of media from the absorption and reflection spectra can be realized using a simple computational procedure. However, the most important requirements for ensuring the reliability of obtaining these analyzes is to ensure the reliability of the experimental data and the width of the frequency range of these data.

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