Optica Applicata, Vol. XI, No. 4, 1981

Kramers-Kronig analysis of the optical constants of Zn_3As_2 and Zn_3P_2*

II. Substractive KK method and some improvement of Leveque method

KRZYSZTOF JEZIERSKI, JAN MISIEWICZ, JANUSZ WNUK, JANUSZ M. PAWLIKOWSKI

Institute of Physics, Technical University of Wrocław, Wybrzeże Wyspiańskiego 27, 50–370 Wrocław, Poland.

The present paper consists of five parts and is devoted mainly to the results of optical constants examination in Zn_3As_2 and Zn_3P_2 which are compounds of II_3–V_2 type with broad energy gap. The analysis of the up to now state-of-affairs so far as the application of the Kramers-Kronig analysis method (KK) to the complex coefficient of reflection is concerned has indicated that the research should be developed in three additional directions described in the first parts of this series. In the part I a critical analysis of the calculation methods used till now and based on dispersion relations due to Kramers and Kronig has been presented. Also the methods of calculation of optical constants not employing the KK integral have been discussed.

In the parts II and III some suggestions of effective improvement of the methods of calculations based on KK integral are given. In the present (II) part of this paper the methods of calculation, known as the Ahrenkiel and Leveque methods, have been described in detail. It has been suggested that these methods be combined to increase the rate of the self-consistent calculations. Additionally, in both the parts the results of calculations for GaAs (a relatively well known semiconductors) are reported which were employed to test the improved methods of calculation proposed in this paper.

The analysis of errors and accuracies for these methods is widely presented in the part IV. Finally, the part V contains the results of calculations of optical constants for Zn_3As_2 and Zn_3P_2, carried out by using the improved methods of calculation and basing on our own measurements of optical properties of these semiconductors.

1. Introduction

Well known Kramers-Kronig relations allow to calculate either the real or imaginary part of the complex function if the other part of this function is known in the whole domain. These relations have been successfully used in the solid state spectroscopy to calculate the optical constants. It turned out, for instance, that the knowledge of the reflection spectrum for given substance enabled to obtain the spectra for the other optical constants.

Actually, there exist many methods of calculation reported in literature [1]. The present part contains the description of two chosen methods for calculation of optical constants in semiconductors on the base of the Kramers-Kronig analysis [1]. These are: Ahrenkiel method, the so-called SKK method [2], and Leveque method [3]. These methods, especially

---

* Work sponsored by Technical University of Wrocław under contract 14/81, 210173.

the first one, will be described from the practical viewpoint in order to facilitate their usage to those researchers who have no experience of this kind, but who (may be just after having read this paper) would consider them to be worth using. We will give also some suggestion of combining both the methods into a new one offering the advantages of the component methods and causing, in particular, more rapid convergence of the self-consistent calculation based on Leveque method, the measurement of the reflection coefficient from the low energy side being completed.

The results of calculations carried out with the SKK method for GaAs indicate the right choice of the method. Without using any parameters so tedious in matching in other methods (for instance, the extrapolation parameters of the reflection coefficient outside the measurement region), the results obtained are consistent with the results of measurements that may be performed in certain spectral regions.

2. Description of the substractive Kramers-Kronig method (SKK method)

As it is well-known, in order to apply the Kramers-Kronig relation to calculations of optical constants on the base of the reflection spectrum, it is necessary to know the reflection coefficient $R(E)$ in the whole spectral range. Hence the attempts to extrapolate the reflection to the regions available to the measurements become important. Let us assume that the measurements have been carried out in the energy interval (E_a, E_b). The typical calculation procedure of Kramers-Kronig (KK) integral

$$\Theta(E) = \frac{E}{\pi}\int\limits_0^\infty \frac{\ln R(x)}{E^2 - x^2}\,dx \tag{1}$$

consists in splitting this integral into three parts [1]:

$$\Theta(E) = \Theta_1(E)+\Theta_2(E)+\Theta_3(E). \tag{2}$$

Usually the purpose is to estimate correctly the contribution of $\Theta_1(E)$ and $\Theta_3(E)$ to the total phase of the wave reflection – hence the right extrapolation procedure is needed. On the other hand, such a method is sensitive to the type of extrapolation which makes the results uncertain.

However, it is possible to act in a different way, i.e. to reduce the influence of the unmeasured part of the spectrum on the calculation of optical constants in semiconductors in the region of known values of the reflection coefficient, since there exists a strong connection between this effect and the convergence rate of KK integral. This rate, as it was pointed out by AHRENKIEL [2], may be significantly increased by simple transformation of the KK integral at the expense of the knowledge of the phase of the wave reflected at certain point E_0, called the expansion point, and contained in the interval (E_a, E_b). Thus, let us assume that the total phase of the wave reflected at the point $E_0 \in (E_a, E_b)$ is known. The phase at this point will be equal to

$$\Theta(E_0) = \frac{E_0}{\pi} P \int_0^\infty \frac{\ln R(x)}{E_0^2 - x^2} dx. \tag{3}$$

Hence, we may write that

$$\frac{\Theta(E)}{E} - \frac{\Theta(E_0)}{E_0} = \frac{1}{\pi} P \int_0^\infty \frac{\ln R(x)}{E^2 - x^2} dx - \frac{1}{\pi} P \int_0^\infty \frac{\ln R(x)}{E_0^2 - x^2}$$

$$= \frac{E_0^2 - E^2}{\pi} P \int_0^\infty \frac{\ln R(x)}{(E_0^2 - x^2)(E^2 - x^2)} dx. \tag{4}$$

In other words

$$\Theta_{\mathrm{SKK}}(E) = E\left[\frac{1}{\pi} P \int_0^\infty \frac{(E_0^2 - E^2)\ln R(E)}{(E_0^2 - x^2)(E^2 - x^2)} dx + \frac{\Theta(E_0)}{E_0}\right], \tag{5}$$

where the symbol $P\int$ denotes the principal value of the integral. This is the so-called substractive Kramers-Kronig method (SKK method). As it may be seen, the convergence of the SKK integral is much quicker than that of KK integral, since here we have the convergence of $1/E^4$ type, while in the previous method it was of $1/E^2$ type.

Due to a quick convergence the problem of suitable choice of extrapolation practically disappears. For the full correctness of the results it suffices to assume that in the intervals $(0, E_a)$ and (E_b, ∞) the reflection coefficient is constant and equal to $R(E_a)$ and $R(E_b)$, respectively.

As already mentioned, in order to determine the optical constants in semiconductor by using the SKK method it is necessary to know $\Theta(E_0)$. The best solution is to calculate $\Theta(E_0)$ from the Fresnel equation, basing, for instance, on measured coefficients of absorption and refraction at this point. Unfortunately, such data are rarely available, since E_0 lies usually in the region of basic absorption. The ellipsometric measurements of reflection, helpful in such cases, also disappoint in practice, because the measuring procedure is usually very tedious. Thus, if we have no results of the said type another method should be used. In the case of semiconductor there exists the so-called transparency region below the energy gap E_g. In this region the total phase of the wave reflected should be equal to zero, $\Theta(E') = 0, E' \in (0, E_g)$, in other words

$$\frac{\Theta(E_0)}{E_0} = -\frac{1}{\pi} P \int_0^\infty \frac{(E_0^2 - E'^2)\ln R(x)}{(E_0^2 - x^2)(E'^2 - x^2)} dx, \; E' \in (0, E_g). \tag{6}$$

Thus, it suffices to calculate the above integral within the transparency region in order to obtain $\Theta(E_0)$.

The calculations according to SKK method may be splitted into three stages:

- calculation of the integral from the eq. (5) within the whole energy interval,
- calculation of $\Theta(E_0)$,
- calculation of the whole phase from the eq. (5), [2].

3. Determination of $\Theta(E)$

Let the integral from eq. (5) be denoted by I. The equation takes then the form

$$\Theta(E) = E\left[I + \frac{\Theta(E_0)}{E_0}\right]. \tag{7}$$

The calculation of integral I is equivalent to the determination of $\Theta(E) = EI$ under assumprion that $\Theta(E_0) = 0$. Having calculated the integral $I(E)$ in the whole range of the optical tonstants in order to determine $\Theta(E_0)$, it is possible to exploit the existence of the transpacency region, in which $\Theta(E') = 0$, $E' \in (0, E_g)$, then

$$\Theta(E_0) = -E_0 I(E'), E' < E_g, \tag{8}$$

The point E', at which $\Theta(E_0)$ is calculated, must be suitably chosen and somewhat less than E_g, while the values $I(E')E$ should lie on the straight line in the transparency region or, in other words, the values of $I(E)$ in this region should be the same (in reality at least close to one another). Finally

$$\Theta(E) = E[I(E) - I(E')]. \tag{9}$$

AHRENKIEL [2] treated somewhat sketchily the problem of the selection of the point E (the authors of the work [4] do not mention it at all). Probably he chose E' so that the integral $I(E')$ be of minimum value and, consequently, the negative values of $\Theta(E)$ in the transparency region be avoided.

Another procedure is also possible, namely, we may calculate the phase in the expansion point as an average value of integrals $I(E')$ at several points belonging to the transparence regions and assume the rms deviation between these values as the measure of reliability of calculation of $\Theta(E_0)$:

$$\Theta(E_0) = -E_0 \frac{1}{m} \sum_{i=m}^{m} I(E_i),\ E_i \in (0, E_g), \tag{10}$$

$$\sqrt{\sigma^2 [\Theta(E_0)]} = \left\{ \frac{1}{m(m-1)} \sum_{i=1}^{m} [\Theta(E_0) + E_0 I(E_i]^2 \right\}^{1/2}. \tag{11}$$

This method may generate slightly negative valus of $\Theta(E)$ at some points of the transparency region (which is pretty often in the KK method, though it follows from another reason) and therefore, the best thing to do is to join the two methods. If the value $\sqrt{\sigma^2}$ is small $\Theta(E_0)$ has to be accepted from the minimizing procedure applied ot $I(E')$. If $\sqrt{\sigma}$ is high such procedure leads to increasing the value of $\Theta(E_0)$ and, in consequence, to lowering the values of optical constans. Thus, it is better to agree upon the slightly nonphysical assumptions in the region of semiconductor transparency, all the more so that the measurements allowing to determine the optical constants are possible in this region.

4. Numerical calculations

In order to estimate $\Theta(E)$ numerically, it is necessary to derive the respective analytical expression determining the change in the phase angle during reflection on the base of the reflection coefficient spectrum known in the measurement interval (E_a, E_b). This task may be reduced to determining the analytical form of the integral from the eq. (5).

Outside the measurement region it is assumed that

$$R(E) = \begin{cases} R(E_a), & E \in (0, E_a) \\ R(E_b), & E \in (E_b, \infty). \end{cases} \tag{12}$$

The integral (5) calculated within the limits $(0, E_a)$ for the arbitrary E takes the form

$$\frac{1}{\pi} P \int\limits_{0}^{E_a} \frac{(E_0^2-E^2)\ln R(E_a)}{(E_0^2-x^2)(E^2-x^2)}\, dx = \frac{\ln R(E_a)}{2\pi}\left[\frac{1}{E}\ln\left|\frac{E+E_a}{E-E_a}\right| + \frac{1}{E_0}\ln\left|\frac{E_0-E_a}{E_0+E_a}\right|\right]. \tag{13}$$

While in the limits (E_b, ∞):

$$\frac{1}{\pi} P \int\limits_{b}^{\infty} \frac{(E_0^2-E^2)\ln R(E_b)}{(E_0^2-x^2)(E^2-x^2)}\, dx = \frac{\ln R(E_b)}{2\pi}\left[\frac{1}{E}\ln\left|\frac{E_b-E}{E_b+E}\right| + \frac{1}{E_0}\ln\left|\frac{E_b+E_0}{E_b-E_0}\right|\right]. \tag{14}$$

It has been assumed that the expansion point is contained in the interval (E_a, E_b), in accordance with the conclusions from the previous Section.

Within the limits (E_a, E_b), when approximating $\ln R(E)$ by the fragments of parabolas, the integral from the eq. (5) is reduced to the form

$$\begin{aligned} \frac{1}{\pi} P \int\limits_{E_a}^{E_b} \frac{(E_0^2-E^2)\ln R(x)}{(E_0^2-x^2)(E^2-x^2)}\, dx = \frac{1}{2\pi} \sum_{i=2,4\ldots}^{N-1} \Bigg\{ a_i \Bigg[E \ln\left|\frac{(E-E_{i-1})(E+E_{i+1})}{(E+E_{i-1})(E-E_{i+1})}\right| \\ + E_0 \ln\left|\frac{E_0-E_{i-1})(E_0+E_{i+1})}{(E_0+E_{i-1})(E_0-E_{i+1})}\right|\Bigg] + b_i \ln\left|\frac{(E^2-E_{i-1}^2)(E_0^2-E_{i+1}^2)}{(E_0^2-E_{i-1}^2)(E^2-E_{i+1}^2)}\right| \\ + c_i \left[\frac{1}{E}\ln\left|\frac{(E-E_{i-1})(E+E_{i+1})}{(E+E_{i-1}(E-E_{i+1})}\right| - \frac{1}{E_0}\ln\left|\frac{(E_0-E_{i-1})(E_0+E_{i+1})}{(E_0+E_{i-1})(E_0-E_{i+1})}\right|\right]\Bigg\}, \end{aligned} \tag{15}$$

where a_i, b_i, c_i are coefficients of parabola in the interval $(E_{i-1}, E_{i+1}) \in (E_a, E_b)$ calculated rom the system of equations

$$\begin{aligned} a_i E_{i-1}^2 + b_i E_{i-1} + c_i &= \ln R(E_{i-1}), \\ a_i E_i^2 + b_i E_i + c_i &= \ln R(E_i), \\ a_i E_{i+1}^2 + b_i E_{i+1} + c_i &= \ln R(E_{i+1}). \end{aligned} \tag{16}$$

Energies E_i, $i = 1, 2, \ldots, N$ are the measurement points of the coefficient of reflection $R(E)$:

$$E_a = E_1 < E_2 < \ldots < E_i < \ldots < E_N = E_b.$$

If the division of the interval (E_a, E_b) is constant, of difference $\Delta = E_i - E_{i-1}$, the coefficients a_i, b_i, c_i are expressed by the equations:

$$
\begin{aligned}
a_i &= -\frac{1}{2\Delta^2}\ln\frac{R^2(E_i)}{R(E_{i-1})R(E_{i+1})}, \\
b_i &= -2E_i a_i - \frac{1}{2\Delta}\ln\frac{R(E_{i-1})}{R(E_{i+1})}, \\
c_i &= \ln R(E_i) - E_i^2 a_i - E_i b_i .
\end{aligned}
\tag{17}
$$

Finally, the analytic form in the eq. (5) is obtained by summing the right hand sides of eqs. (13), (14) and (15).

In order to determine the optical constants in semiconductor by using the SKK method a programme for ODRA 1305 computer has been written in Fortran IV. This programme has been tested for the relatively well known semiconductor compound, GaAs, basing on the measurements of both reflection coefficient in the region 0–25 eV, and the dielectric function $\hat{\varepsilon}(E)$ reported in [5].

15. Optical constants of GaAs

In figure 1 the real and imaginary parts of the complex dielectric permittivity $\hat{\varepsilon}(E)$ are shown for GaAs calculated on the base of the SKK method. The circles mark the results of calculations made by **Philipp** and **Ehrenreich** [5] and the triangles in the enlarged part of the graph denote the experimental data from the work [5]. As may be seen, our calculations are well consistent with the results reported in [5] and with the experimental data. The coefficient of absorption calculated by using the same method is also in good agreement with the measurement results in the interval 1.5–2.7 eV, reported in [6].

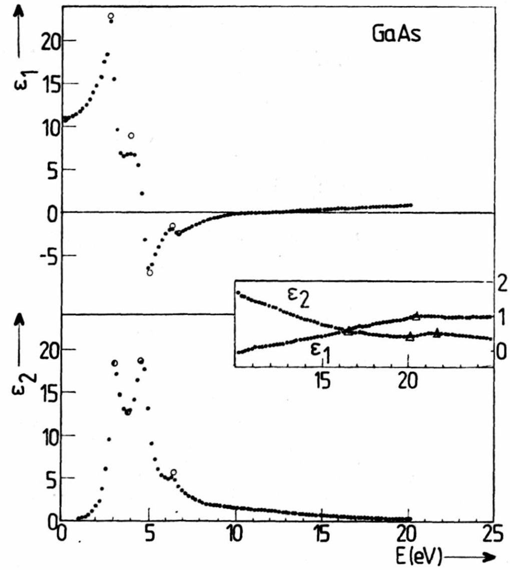

Fig. 1. Real (ε_1) and imaginary (ε_2) parts of the complex dielectric permittivity for GaAs calculated by the SKK method

By using the summation [7]

$$n_{\mathrm{eff}}(E) = \frac{m_0}{2\pi^2 N_1 e^2 \hbar^2} \int_0^E x\varepsilon_2(x)\,dx, \tag{18}$$

$$\varepsilon_{0,\mathrm{eff}}(E) = 1 + \frac{2}{\pi} \int_0^E \frac{1}{x}\varepsilon_2(x)\,dx, \tag{19}$$

where N_1 is the concentration of atoms in the crystal, and m_0 denotes the electron mass. Next, the effective number of electrons per atom, n_{eff}, which take part in the optical transitions as well as the effective static dielectric permittivity, $\varepsilon_{0,\mathrm{eff}}(E)$ have been calculated. The results are presented in figs. 2 and 3, where the effective values n_{eff} and $\varepsilon_{0,\mathrm{eff}}$ calculated earlier in the work [5] are also marked. In accordance with the theoretical predictions n_{eff} reaches the value 4 after having exhausted the possibilities of valence electron transitions (for energies of about 20 eV) and next increases due to the transitions of electrons from the shell *d* in Ga atoms.

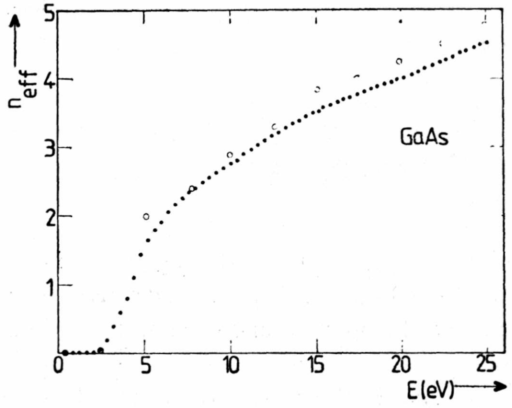

Fig. 2. Effective number of electrons n_{eff} per 1 atom for GaAs calculated from the spectrum of $\varepsilon_2(E)$. The circles denote the results of calculations according to [5]

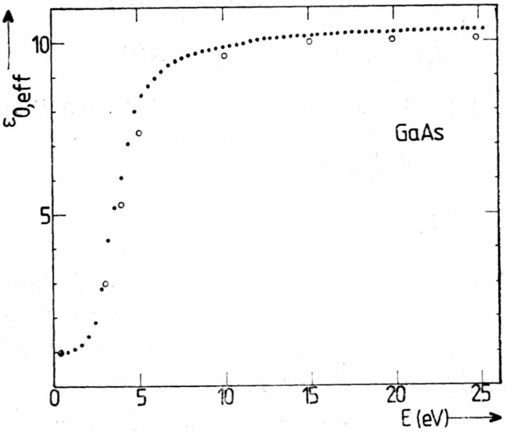

Fig. 3. Effective static dielectric permittivity $\varepsilon_{0,\mathrm{eff}}$, for GaAs calculated from the spectrum of $\varepsilon_2(E)$. The circles denote the results of calculations according to [5]

In order to determine $\Theta(E_0)$ from the condition $\Theta(E) = 0$, $E < E_g$, the method of average value discussed in Section 3 was used (see formula (10)), for several points in the transparency region. The method of minimum (formula (11)) gave here worse results.

6. Application of SKK method to Leveque method

The self-consistent Leveque method allowing to complete the spectra of reflection coefficients from the low energy side has been shortly discussed in [1]. In order to join it with the SKK method some of its elements should be considered more broadly. In [3] LEVEQUE took account of the relations:

$$k(E)=\frac{2\sqrt{R(E)}\sin\Theta(E)}{1+R(E)-2\sqrt{R(E)}\cos\Theta(E)}, \tag{20}$$

$$R(E)=\frac{[n(E)-1]^2+k^2(E)}{[n(E)+1]^2+k^2(E)}, \tag{21}$$

$$\alpha(E)=\frac{2E}{\hbar c}\,k(E), \tag{22}$$

$$n(E)=1+\frac{2}{\pi}P\int\limits_0^\infty\frac{k(x)x}{x^2-E^2}\,dx, \tag{23}$$

$$\Theta(E)=\frac{E}{\pi}P\int\limits_0^\infty\frac{\ln R(E)}{E^2-x^2}\,dx \tag{24}$$

which allow to determine $R(E)$, if $\alpha(E)$ is known, and vice versa:

$$\begin{aligned}&R(E)\overset{24}{\rightarrow}\Theta(E)\overset{20}{\rightarrow}k(E)\overset{22}{\rightarrow}\alpha(E),\\&\alpha(E)\overset{22}{\rightarrow}k(E)\overset{23}{\rightarrow}n(E)\overset{21}{\rightarrow}R(E).\end{aligned} \tag{25}$$

Let $[T]$ be the integral operator such that

$$\begin{aligned}&\alpha(E)=[T]R(E),\\&R(E)=[T]^{-1}\alpha(E).\end{aligned} \tag{26}$$

If the measurements of reflection and absorption in some partly overlaping energy intervals (fig. 4) are available, then we may determine such a set of curves R and α, defined within the whole energy interval (E_a', E_b), which would be self-consistent and close to the experimental data in the respective intervals.

Let $[P]$ be an operator modifying an arbitrary curve $R(E)$ or $\alpha(E)$ to make it identical with the experimental value in the measurement range. Now, the method of defining the functions R and α consists in starting form the curve $R(E)$ with any classical extrapolation and next applying the operators $[T]$, $[T]^{-1}$ and $[P]$ according to the scheme presented in fig. 5.

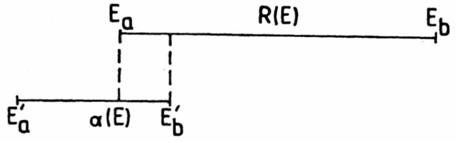

Fig. 4. Examplified overlapping of experimental data for $R(E)$ and $\alpha(E)$, see text

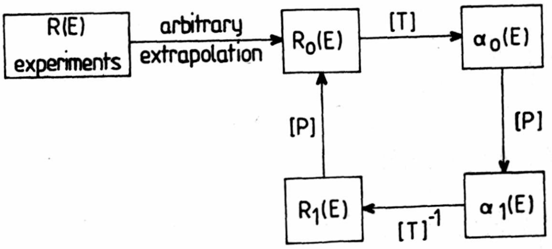

Fig. 5. Scheme of cyclic calculations of the functions R and α (for explanation see text)

If the calculations are convergent (which theoretically is the problem of good initial experimental data for R and α) the obtained set of two functions R and α is fully consistent with the experiment [3].

Our partial suggestion is that instead of relations (23) and (24) suitable relations with the SKK integrals should be applied to the cycle calculations. For given energy E_0 we have from the formula (23):

$$n(E_0) = 1+\frac{2}{\pi}P\int_0^{\infty}\frac{k(x)x}{x^2-E_0}\,dx. \tag{27}$$

By substracting respectively the relations (23) and (27) from one another we obtain

$$n(E)-n(E_0) = \frac{2}{\pi}\left[P\int_0^{\infty}\frac{k(x)x}{x^2-E^2}\,dx-P\int_0^{\infty}\frac{k(x)x}{x^2-E_0^2}\,dx\right]. \tag{28}$$

Hence

$$n(E) = n(E_0)-\frac{2}{\pi}P\int_0^{\infty}\frac{(E_0^2-E^2)k(x)x}{(E_0^2-x^2)(E^2-x^2)}\,dx. \tag{29}$$

The formula (24) is assumed in the form (5) derived earlier. The introduced replacements (23) → (29), and (24) → (5) leave the property (25) unaffected.

The values of the phase change $\Theta(E)$ in the reflected wave may be calculated from the formula (5) in the way given in Sections 3 and 4, while the refractive index $n(E)$ from the formula (29) is estimated in an analogical way from eq. (29). In the regions $(0, E_a')$ and (E_b', ∞) we use the straight line extrapolation summing $n(E) = n(E_a')$ and $n(E) = n(E_b')$, respectively, and obtain for the integral from eq. (29) the relations

$$\frac{2}{\pi}P\int_0^{E_a'}\frac{(E_0^2-E^2)k(E_a')x}{(E_0^2-x^2)(E^2-x^2)}\,dx = \frac{k(E_a')}{\pi}\ln\left|\frac{(E_0^2-E_a'^2)E}{(E^2-E_a'^2)E_0^2}\right|, \tag{30}$$

$$\frac{2}{\pi}P\int_{E_b}^{\infty}\frac{(E_0^2-E^2)k(E_b')x}{(E_0^2-x^2)(E^2-x^2)}\,dx = \frac{k(E_b')}{\pi}\ln\left|\frac{E^2-E_b^2}{E_0^2-E_b'^2}\right|. \tag{31}$$

The integral (29) in the limits (E_a, E_b) is calculated, for instance, by using the parabola method applying the approximation

$$k(E)E = a_iE^2+b_iE+c_i \tag{32}$$

in the respective intervals (E_{i-1}, E_{i+1}), $i = 2, 4, \ldots, N-1$, $E_i \in (E_a', E_b')$ and calculating the parabola coefficients from the following set of equations

$$\begin{aligned} &a_iE_{i+1}^2+b_iE_{i-1}+c_i = E_{i-1}k(E_{i-1}), \\ &a_iE_i^2+b_iE_i+c_i = E_ik(E_i), \\ &a_iE_{i+2}^2+b_iE_{i+2}+c_i = E_{i+1}k(E_{i+1}). \end{aligned} \tag{33}$$

The calculations of optical constants may now be made as follows: As a first step the phase change $\Theta(E)$ of the reflected wavefront is calculated by using the SKK method on the base of the reflection spectrum in the region (E_a, E_b) with the extrapolation by straight line in the regions $(0, E_a)$ and (E_b, ∞) as in the Section 2.4. Obviously, we assume that the region of transparency is induced in the interval $(0, E_a)$, since just for these reasons the Leveque method is introduced to determine $R(E)$ in this region. The properties of $\Theta(E)$ calculated in this way become the first approximation.

From the whole calculation interval of $\Theta(E)$ only one value is needed, i.e. the first value of the phase change in the expansion point during cyclic calculations. The scheme of cyclic calculations is similar to that used by Leveque; the difference concerns the additional complication caused by the necessity of knowing the phase change and the refractive index at the expansion points. The expansion point E_{01} for the eq. (5) is chosen slightly above E_a, while the point E_{02}, for the eq. (29) slightly below E_b', to obtain the highest accuracy of SKK integral calculations at the vicinity of the contact of the measurement regions $R(E)$ and $a(E)$. The values of $\Theta(E_{01})$, except for the first one, and the values of $n(E_{02})$ are calculated from the values of $k(E_{01})$ and $R(E_{02})$ estimated by half a cycle earlier, respectively, as well as the experimental data of $R(E_{01})$ and $k(E_{02})$, respectively, using the relations (20) and (21). The scheme of the cyclic calculations is shown in fig. 6. Below the symbols of the physical quantities the energy regions are given for these quantities known from experiment (exp) or calculated (num) and also the extrapolation by straight line outside those regions is marked (—). Such simple extrapolation may be applied only when SKK method is used. The broken line denotes a single act of supplying the input data.

It may be expected that due to the properties of SKK methods the values of optical constants obtained in the successive stages of calculations should be closer to the true values than those estimated by the KK method (spectral regions of both $a(E)$, and $R(E)$ are narrow, especially for $a(E)$ and the advantages offered by the SKK methods become evident) which resulted in quicker convergence of the cyclic calculations. However, there is a danger that the application of the SKK method will increase even more the requirements concerning the accuracy of measurements, which are already high enough in the Leveque method.

After having achieved the convergence in the cyclic calculations the values of $R(E)$ self-consistent with the measurements of $a(E)$ are obtained in the region (E_a', E_b'). When having a broadened spectral region for $R(E)$ at disposal the optical constants of the examined semiconductors can be calculated by applying again the SKK method.

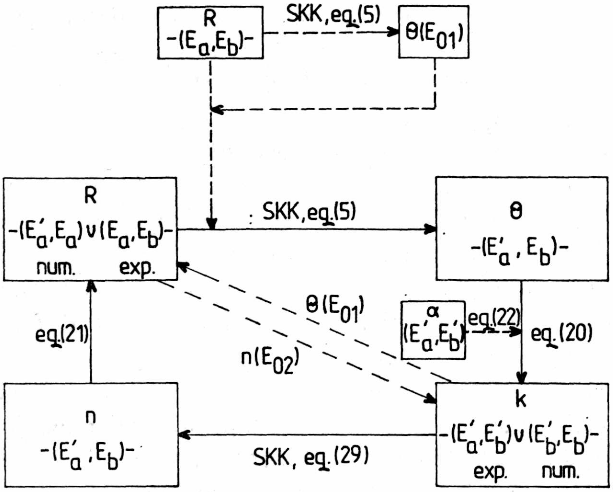

Fig. 6. Scheme of cyclic calculations in the improved Leveque method (see text)

7. Conclusions

Various comparative tests made for the KK and SKK methods (in [2,4], for instance) have confirmed higher correctness of the results offered by the SKK method than that obtained by using the KK method for relatively narrow measurement ranges (E_a, E_b). The idea of those tests was to narrow the region of reflection coefficient measurements exploited in calculations of optical constants and to examine the influence of the resulting contraction upon the shape of optical functions. The error in determining the optical constants occurring due to neglected part of the reflection spectrum is several times less for the SKK method than that for the KK method in the examined measurement range. The SKK method gives some useful results (of error less than 10%) even for significant contraction (above 50%) of the measurement region. Here, the region of the spectrum $R(E)$ of strong structure has been assumed to be 100%.

All the advantages offered by SKK method in comparison with the classical KK analysis become evident when only a narrow range of known values of the reflection coefficient (or order of few eV) is available, since the error of this method depends upon the squares of distances of E from E_0. For this reason the energy E_0 may be chosen in the central part of the considered energy interval [2]. By the way, the error occurring due to positioning of E_0 may be reduced by calculating several times the optical constants for several values of E_0 distributed uniformly in the calculation interval and next by fitting the results. Besides, the greater is the measurement region the less number of elements of the $R(E)$ structure ignored in the calculations and it is just the error due to neglecting these elements which is of concern.

Writing that the SKK method improves the correctness of the results for low energies and worsens it for high energies [3]. Leveque had probably in mind the fact that the advantages of this method appear evidently in the energy range corresponding to the range of the reflection coefficient measurements. Such measurements are, however, usually available in the region of low energies. The calculations performed for GaAs in the present work confirm fully the applicability of the SKK method to the calculations of optical constants in semiconductors also in the broader energy range.

Summing up, the SKK method offers many advantages like relatively low degree of calculation complexity and simultaneously high correctness of the results as well as a possibility of making the calculations as soon as the measurement results in a narrow measurement region become available. The application of this method is restricted only by the necessity of knowing the values of the phase changes Θ at the single point E_0. This difficulty (when the results of other measurements are unavailable) can be, however, avoided if the spectrum $R(E)$ is known in the region of transparency.

The condition that the values of phase changes should be known may be easily satisfied by the other method discussed in this part of our paper, i.e., the Leveque method, however, et the expense of certain complication of the calculations. In spite of this, it should be noticed that the advantages following from the application of these methods (in our opinion, especially of the first one) highly exceed the eventual calculation difficulties.

References

[1] Jezierski K., Misiewicz J., Wnuk J., Pawlikowski J. M., Part I of this work, Optica Applicata XI (1981), 571.

[2] Ahrenkiel R. K., J. Opt. Soc. Am. **61** (1971), 1651.

[3] Leveque G., J. Phys. **C10** (1977), 4877.

[4] Sobolev V. V., et all., *Rastchoty opticheskich funkcii poluprovodnikov po sootnosheniyam Kramersa-Kroniga*. Ed. AN Moldavskoy SSR, Shtyinca, Kishynev 1976.

[5] Phillip H. R., Ehrenreich H., Phys. Rev. **129** (1963), 1550.

[6] Struge M. D., Phys. Rev. **127** (1962), 768.

[7] Altarelli M., et all., Phys. Rev. **B6** (1972), 4502.

Received January 25, 1981

Определение оптических констант полупроводников Zn_3As_2 и Zn_3P_2 с помощью соотношения Крамерса-Кронига

Часть II. Дифференциальный метод Камерса-Кронига и совершенствование метода Левека

Работа состоит из пяти частей и посвящена главным образом результатам исследований оптических констант Zn_3As_2 и Zn_3P_2 двух соединений типа II_3–V_2 с большой энергетической щелью. Результатом анализа существующего до настоящего времени состояния в области применения метода анализа Крамерса-Кронига (КК) для комплексного коэффициента отражения оказалась необходимость развития работ в дополнительных направлениях, описанных в трёх первых частях. В I части представлен критический анализ применяемых до настоящего времени методов расчёта, основанных на дисперсионных соотномения Крамерса-Кронига. Обсуждены также методы расчёта оптических компонент, в которых не используется интеграл КК.

Во II и III частях описаны предложения эффективных усовершенствований методов расчёта, основанных на применении интелграла КК. В настоящей (II) части работы подробно описаны методы расчёта, известные как методы Аренкеля, а также Левэка. Предложено также обьединение обоих методов, направленное на увеличение скорости самосогласований расчётов. Дополнительно, в обоих частях представлены результаты расчётов для GaAs, который послужил (в качестве по-

лупроводника со сравнительно хорошо известными оптическими свойствами) для проверки улучшенных нами методов расчёта.

Анализ ошибок и точности методов расчётов очень подробно описан в IV части. Пятая часть содержит результаты расчётов оптических констант Zn_3As_2 и Zn_3P_2, произведённых с помощью усовершенствованных методов расчётов, а также на основе наших результатов измерений оптических свойств этих полупроводников.