# On the relation of the Meyer-Neldel rule and the Urbach rule

(FROM POINT OF VIEW OF THE BARRIER-CLUSTER MODEL)

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The connection between the Meyer-Neldel rule (MNR) and Urbach rule (UR) is briefly discussed in this paper from the viewpoint of the barrier-cluster model of disordered semiconductors. We assume that a significant role in this connection has to be ascribed to monoenergetical phonons produced in the process of non-radiative recombination of the electron-holes (*e-h*) pairs.

## 1. Introduction

Up to now there is no generally accepted model explaining the physical essence of the Meyer-Neldel rule [1-12]. One meets a similar situation in interpretations of the Urbach rule [13-22]. The author's intent to explain a connection between two unexplained phenomena may seem to be strange for the readers. The autors standpoint is based on his preceding works, where the barrier-cluster model has been proposed and where also the upper mentioned phenomena were discussed and explained in the frame of this model [23-31]. The first considerations above Urbach rule has been published in [23, 24] and subsequently also an explanation of the Meyer-Neldel rule in [27, 29, 30]. From that point of view, the search for a relation between both the rules seems to be clear, we believe.

## 2. The Urbach rule

The Urbach rule is empirical, describing the absorption of light in exponential tails of optical absorption spectra. It expresses the dependence of the coefficient of the optical absorption α on the photon energy *hf* and the temperature *T*. Mathematicaly it can be expressed by the relation

$$\alpha = C \exp( \gamma \frac{hf - E}{kT})  \tag{1a}$$

or equivalently as

$$\ln \alpha = \left( \gamma \frac{hf - E}{kT} \right) + const  \tag{1b}$$

Thus, lnα (*hf*) is a straight line. Here γ, *E* are parameters and *k* is the Boltzmann constant. This relation holds for relatively high temperatures. At low temperatures, the absorption is given by a relation similar to (1a,b), but the real temperature *T* has to be replaced by a constant parameter $T_o$. This point was not explained satisfactorily up to now [1, 13-22].

### 2.1 Explanation of the Urbach rule

The Urbach rule in high temperature region has been explained by the barrier-cluster model assuming that an electron during the optical transition from the conduction into valence band can absorb both the energy of a photon and of a phonon [23, 24]. Then the energy increase of the electron is equal to

$$hf + W_{phon}$$

where $W_{phon}$ is the energy gained by the electron from lattice vibrations. The photon energy is given by the frequency of light. The phonon energy has a statistical character.

If the photon energy $hf < 2W$ is less than the width of the forbidden gap of a non-crystalline semiconductor then the absorption of light cannot happen. But, the absorption can come into being if the missing energy $2W - hf$ needed for the transition is supplied by a phonon. This is the energy, which an electron needs in order to equalize its energy with the width of the forbidden gap.

The probability of occurrence of phonons with an energy higher than or equal to $2W - hf$ is proportional, at temperature *T*, to the expression

$$p = C \exp(- \frac{2W - hf}{2kT}) = C \exp\left( \frac{hf - 2W}{2kT} \right)  \tag{2}$$

The probability of optical absorption (with the participation of phonons) is proportional to the same

expression. The absorption coefficient α in an energy region of photons less than the width of the forbidden gap can be determined as

$$\alpha = C \exp(\frac{hf - 2W}{2kT}) \qquad (3a)$$

where $C$ is a constant. Thus

$$\ln \alpha = \left( \frac{hf - 2W}{2kT} \right) + const \qquad (3b)$$

This relation represents a straight line (exponential tail of optical absorption), with a slope decreasing with increasing the temperature $T$. This behaviour corresponds to the one known with chalcogenide glasses (if $E = 2W$).

However, it has become known from experiments that the situation changes with the decrease of the temperature. The slope of the straight lines given by (3b) continues being unchanged with temperature at low temperatures. Only a parallel shift to the lower absorption of "exponential tails" is observed.

The relation (3a) is no longer valid. At lowering the temperature of a non-crystalline semiconductor, the parameter $T$ in relation (3a) no longer represents a temperature. It seems as if the temperature were freezed at a certain value of $T_0$. The essence of this phenomenon is not reliably explained till now [1], but we will try to clarify it partially. Let us note that the parameter γ is about 0,4 – 0,55 [32-34] in the chalcogenide glasses. When comparing (1a) and (3a), we see that the barrier-cluster model yields the constant γ = 0,5. This may be considered as a good agreement in the research of non-crystalline semiconductors.

From experiments it follows that the parameter $E$ in (1a,b) has a value, which corresponds to the width of the forbidden band. This is in good agreement with our barrier-cluster model.

## 3. The Meyer-Neldel rule

The MNR has been observed for the first time with examining the electric conductivity of non-crystalline semiconductors [3]. Semiconductors are characterized by their temperature dependence of the electrical conductivity [32-34]. Most semiconductors exhibit an exponential temperature dependence of the conductivity,

$$\sigma = \sigma_0 \exp(-\frac{W}{kT}) \qquad (4)$$

where $\sigma_o$ is a constant and $W$ is an activation energy.

For many classes of materials, especially organic semi-insulators, chalcogenide glasses, amorphous silicon, experimental evidence suggests that a correlation exists between the activation energy and the pre-exponential factor of the following form [1-12]

$$\ln \sigma_0 = bW + \ln \sigma_{00} \qquad (5a)$$

where $b$ and $\sigma_{oo}$ are constant. This relation can be written as

$$\sigma_0 = \sigma_{00} \exp(\frac{W}{kT_0}) \qquad (5b)$$

here $b = 1/kT_o$. The relation (5b) gives the dependence of the pre-factor $\sigma_o$ on the activation energy $W$ and represents the Meyer-Neldel empirical rule. Equation (5b) is often refereed to as the MN rule or the compensation rule. Constant $\sigma_{oo}$ is often called the Meyer-Nedel pre-exponential factor and $kT_o$ the MN characteristic energy.

This rule holds in disordered materials when $W$ is varied by doping, by surface absorption, light soaking or by preparing films under different conditions. This rule has also been observed in liquid semiconductors and fullerens. The validity of the MN rule has also been reported in the case of chalcogenide glasses. In the case of these glasses this rule is observed by variation of $W$ when the composition of the glassy alloys is changed in a specific glassy system. Electrical conductivity in dark is measured as a function of temperature for this purpose.

### 3.1 An explanation of the MNR

In the following we will present some notes on one possible explanation of MNR, which was published in the works [27, 29, 30]. In this model it is assumed, that activation energy of a semiconductor influences the recombination process of current carriers. An increase of activation energy $W$ of a non-crystalline semiconductor decreases probability of carrier recombination. This process necessary influences equilibrium concentration of conduction electrons (carriers) and subsequently electric conductivity of the semiconductor. As a result we obtain the relation identical with that one representing empirical Meyer-Neldel rule.

### 3.2 Dependence of conductivity on activation energy

A transition of an electron from conduction band to valence band in a non-crystalline semiconductor proceeds in our case predominantly by production of phonons. The total energy of produced phonons will correspond to that one released in electron transition. Further we shall assume that in a substance under consideration dominates phonon production, which average energy is ΔE. It means that phonon production of other phonons is negligible. At the transition of an electron from conduction band to valence band gained energy $2W$ is used in production of $N$ phonons each of them has energy ΔE so that $2W = N\Delta E$ or

$$N = 2W/\Delta E \qquad (6)$$

Let $w_1$ be the probability of production of one phonon

which energy is equal to $\Delta E$. Probability $w_N$ of production of $N$ phonons of equal energy $\Delta E$ due to the electron – lattice interaction will be

$$w_N = (w_1)^N \tag{7a}$$

If we write down probability $w_1$ as

$$w_1 = \exp(-\varepsilon_1) \tag{8}$$

where $\varepsilon_1$ is a positive value, then probability $w_N$ one can write with respect to (6,7a) as

$$w_N = \exp(-N\varepsilon_1) = \exp(-\varepsilon_1 2W/\Delta E) = \exp(-bW) \tag{7b}$$

where the constant $b$ is given by

$$b = 2\,\varepsilon_1/\Delta E \tag{9}$$

Relation (8) gives at the same time the probability of recombination; it means the transition probability of an electron from conduction to valence band. That one is proportional to the probability of production of $N$ phonons and also proportional to the $\exp(-bW)$. With an increase of activation energy probability of recombination according to (8) exponentially decreases.

### 3.3 Equilibrium concentration of conduction electrons

In equilibrium (or steady) state it holds

$$\left(\frac{dn}{dt}\right)_{gen} = \left(\frac{dn}{dt}\right)_{recom} \tag{10}$$

A number of free electrons generated in a unit time is given as

$$\left(\frac{dn}{dt}\right)_{gen} = C_1 \exp\left(-\frac{W}{kT}\right) \tag{11}$$

where $C_1$ is a constant.

For a recombination process we suppose (in accordance with the relation (8)) that the relation

$$\left(\frac{dn}{dt}\right)_{recom} = n C_2 \exp\left(-bW\right) \tag{12}$$

is valid. Here $C_2$ is a constant. The number of recombinations in a unit of time is proportional to the number $n$ as well as to the probability (8) of production of $n$ phonons at the electron transition. In equilibrium state is valid $R = G$, so that

$$C_1 \exp(-W/kT) = n.C_2 \exp(-bW) \tag{13}$$

From that relation follows for equilibrium concentration $n$ of free carriers

$$n = C_{00}\exp(bW)\exp(-W/kT) \tag{14}$$

where $C_{00}$ is determined by the constants $C_1$ and $C_2$.

### 3.4 Electric conductivity

It is known that electric conductivity $\sigma$ is proportional to $n$: $\sigma \approx n$. If mobility of carriers is independend on activation energy $W$ (or that dependence is negligible) one can write with respect to (14)

$$\sigma = \sigma_{00}\exp(bW)\exp(-W/kT) \tag{15}$$

respectively

$$\sigma = \sigma_o(W)\exp(-W/kT) \tag{16}$$

where $\sigma_{00}$ is a constant and

$$\sigma_o(W) = \sigma_{oo}\exp(bW) \tag{17}$$

If we put $b = 1/kT_{MN}$, the relation (15) takes the form which is identical with dependence (4)

$$\sigma = \sigma_{00}\exp\left(\frac{W}{kT_{MN}}\right)\exp\left(-\frac{W}{kT}\right) \tag{18}$$

The relation (17) acquires the form

$$\sigma_0 = \sigma_{00}\exp\left(\frac{W}{kT_{MN}}\right) \tag{19}$$

which expresses the conventional Meyer-Neldel rule (3).

## 4. On the relation of the Meyer-Neldel rule and the Urbach rule

At explanation of Meyer-Neldel rule based on the barrier-cluster model we used conception that a non-radiate recombination transition of an electron from a conduction band to a valence band in a non-crystalline semiconductor is connected with production of series monoenergetical phonons. Further we shall speculate about that how these monoenergetical phonons influence an optical absorption of the non-crystalline substance. Our next consideration shall lead to explanation of a mechanism of origin exponential tails of optical absorption that is to explanation of the Urbach empirical rule. We show this way that there is a connection between Meyer – Neldel rule and Urbach rule.

It is known, that exponential tails of the optical absorption reach deep in a forbidden band of the width $2W$. If $hf < 2W$, energy of a photon alone is not sufficient for an optical transition of an electron to the conduction band. The missing energy $2W - hf$ phonons can offer to electron not only thermal ones (controlling by Maxwell-Boltzmann distribution function) as it was described upper, but also monoenergetical ones. The energy $\Delta E$ of one monoenergetical phonon is small, usually much smaller than is deficit of energy $2W - hf$ ($\Delta E << 2W - hf$). An optical transition demand the electron has accept needed energy $N\Delta E$ from more ($N$) monoenergetical

phonons so as the condition

$$N\Delta E \geq 2W - hf \quad (20)$$

was fulfilled.

Let $w_1$ denotes probability that an electron at optical absorption under upper described conditions accepts one monoenergetical phonon of energy $\Delta E$. Probability of absorption $N$ monoenergetical phonons then will be $w_N = w_1^N$. We shall write down $w_1$ as

$$w_1 = \exp(-s) \quad (21)$$

where $s > 0$ (because $w_1 < 1$). Minimal number $N$ obtained monoenergetical phonons then will be

$$N = (2W - hf)/\Delta E,$$

so that

$$w_N = \exp\left(-s\frac{2W - hf}{\Delta E}\right) = \exp\left[-b_2(2W - hf)\right] \quad (22)$$

kde $b_2 = s/\Delta E$. It holds

$$w_N = \exp\left(-\frac{2W - hf}{U}\right) \quad (23)$$

where $U = 1/b_2$ is for a given substance a constant parameter. Just probability $w_N$ will determine level of an optical absorption related to monoenergetical phonons. This component of absorption will be proportional to the value of $w_N$ an so proportional to the expression
$\exp(-(2W - hf)/U)$.

Absorption will consist of two components – besides the component given by the relation (3a) there will be present the component proportional to the probability $w_N$. Resulting coefficient of absorption $\alpha$ can be written as

$$\alpha = C_1' \exp\left(-\frac{2W - hf}{2kT}\right) + C_2' \exp\left(-\frac{2W - hf}{U}\right) \quad (24)$$

Here $C_1'$ a $C_2'$ are pre-exponential factors.

The gained relation is the generalization of the relation (3a). The meaning of the first term was explained before. The second term represents the contribution of monoenergetical phonons to the absorption. The upper introduced relation represents a spreading of the relation (3a) to the case when absorption is also affected by monoenergetical phonons participating in a non-crystalline substance.

In the next consideration we stress that monoenergetical phonons can be created by two different mechanisms. The first one will be thermal excitation. The second one is related to the absorption of light. The parameter $C_2'$ consists generally of two components

$$C_2' = C_{2t} + C_{2o} \quad (25)$$

The first component corresponds to the thermal production of monoenergetical phonons and the second one – to the optical production monoenergetical phonons. Pre-exponential factors $C_{2t}$, $C_{2o}$ are in some measure depending on the temperature and energy of the photons. The parameter $C_{2o}$ evidently depends also on impacting light flux on a sample. The relation (3a) one can write as

$$\alpha = C_1' \exp\left(\frac{hf - 2W}{2kT}\right) + (C_{2t} + C_{2o}) \exp\left(\frac{hf - 2W}{U}\right) (26)$$

We assume that dependences of $C_{2t}(T, hf)$, $C_{2o}(T, hf, \Phi)$ are not so strong to be change exponential character of particular terms in the relation (24). In this relation appear exponential terms explicitly.

One can expect that at sufficiently high temperature the first term on the right of relation (24) dominates. This is in agreement with experiments. This term represents optical absorption at higher temperature when the slope of linear dependences $\ln\alpha(hf)$ changes with change of the temperature. This corresponds to the Urbach`s rule for region of higher temperatures.

With lowering temperature successively begins dominate the second term on the right of the relation (24). In such a case the slope of the straight lines of $\ln\alpha(hf)$ practically becomes unchanged. The possible change of parameters $C_{2t}$ a $C_{2o}$ in the process was ignored.

In the case of very small illumination $C_{2t} \gg C_{2o}$, then absorption is described by the relation

$$\alpha_{2t} = C_{2t} \exp\left(\frac{hf - 2W}{U}\right) \quad (27)$$

The weak level of illumination under these conditions in fact does not affect the value of absorption coefficient.

In general one can express the absorption coefficient as follows

$$\alpha = \alpha_1 + \alpha_{2t} + \alpha_{2o} \quad (28)$$

where

$$\alpha_1 = C_1' \exp\left(\frac{hf - 2W}{2kT}\right) \quad (29)$$

$$\alpha_{2t} = C_{2t} \exp\left(\frac{hf - 2W}{U}\right) \quad (30)$$

$$\alpha_{2o} = C_{2o} \exp\left(\frac{hf - 2W}{U}\right) \quad (31)$$

At low illumination is $\alpha = \alpha_1 + \alpha_{2t}$. At low illumination and high temperatures the term $\alpha_1$ dominates. At higher values of the flux of illumination the absorption coefficient will depend also on the level of lighting.

Now, the question is, when in the relation (24) the second term $\alpha_{2\,t}$ begins to dominate; or when the slope of the straight lines $\ln(hf)$ stay unchanged at lowering temperature? (Only, small parallel shifts of the tails are observed in direction to the lower absorptions.)

At lowering temperature apparently not only the term $\alpha_1$ but also $\alpha_{2\,t}$ (to some measure) are decreasing. At lowering temperature the production of thermal mono-energetical phonons decreases and therefore value of the parameter $C_{2\,t}$ decreases too. There is not doubt about it. But, the other factor plays the role. It is process of thermalization of mono-energetical phonons. At higher temperatures these phonons are intensive scattered – thermalized. At lower temperatures this process is less effective and subsequently the lifetime of mono-energetical phonons is longer. Moreover the decrease of the parameters $C_{2\,t}$ is relatively small and subsequently the term $\alpha_{2\,t}$ begins to dominate. Of course, this problem calls for more detail analysis (This analysis will be certainly not simple).

At intensive lighting of a substance at lower temperatures can dominate in the relation (24) the term $\alpha_{2o}$. The value of that term depends on the level of lighting (on the light flux). The absorption coefficient is also a function of light intensity.

**Notice:** In crystalline semiconductors (e.g. of the type CdSe) production of mono-energetical phonons is very small so that the term $\alpha_{2\,t}$ begins to prevail (the term $\alpha_1 \ll \alpha_{2\,t}$) at much lower temperature than in non-crystalline semiconductors. In semiconductors in which production of mono-energetical phonons is negligible, observable absorption is determined by the term $\alpha_1$ only. Urbach's exponential tails can appear in semiconductors in which the conditions for absorption exist under energetical participation of phonons. This is the case of substances in which the validity of MNR is observed. In such substances probably transition appears through Urbach's border - that is the transition from changing slope of straight lines of $\ln(hf)$ (by changing temperature) to parallel thermal shifting of the tails $\ln(hf)$.

## 5. Open problems and questions

The physics of the disordered materials is extremely wide. The publications [32] and [33] belong to the first, which offer the complex of view on this problem. The actual questions are solved and discussed in [34-39].

One can oppose to the upper introduced considerations. Author is aware that described model of relation between MNR and Urbach`s rule possesses intuitive background. More detailed theoretical substantiation would be needful. This article is intended to inspire theoretical physicists involved in the field of condensed mater physics to solve that problem. On the other side, quantitative theoretical explanation of the behavior of exponential tails in the long term resists to the theoretical explanation. The present time no standpoint is known for promising theory. This is what the author's motivated to submit and publish this intuitive theory.

### 5.1 Interpretation of another connection

In frame of the barrier-cluster model it is possible to explain other effects observed in experiment, such as the temperature dependence of photoconductivity of chalcogenide glasses $(As_2S_3)$ in the low temperature region.

As an example, we can note a plot of the photoconductivity dependence on the temperature $T$ showed in the figure published in Ref. [1, 22]. One can see that the photoconductivity decreases with decreasing the temperature, but at a certain low enough temperature, this decrease of the photoconductivity is stopped. By further lowering the temperature, the photoconductivity becomes actually constant. This behaviour of the chalcogenide glass can be explained by the use of barrier-cluster model.

In the same figure in [1, 22] another graph is seen, presenting a dependence of the photoluminescence on the temperature. Photoluminescence increases with the temperature decrease. At a certain temperature this increase stops and a further lowering of the temperature actually doesn't change the photoluminescence. This behaviour of the photoluminescence can also be explained by the barrier-cluster model. The "temperature fault" appears at the temperature at which the behaviour of both the phenomena (photoconductivity and photoluminescence) exibits changes. This temperature is actually the same for both the phenomena. We do not believe that this is accidenta. Analogous dependences were described in [18-19].

### 5.2 Further remarks

According to the barrier-cluster model [23-31], the free electrons catalyze a non-radiant recombination of bound *e-h* pairs. The more non-radiant recombinations, the less radiant ones are present, and vice versa. Any decrease of free electrons (charge carriers) causes an increase of the photoluminescence. On the other hand, when the concentration of free electronss enhances, the photoluminescence becomes oppressed. Whenever the minimum of the conductivity is achieved, the luminescence will to attain its maximum value. This relationship bettween the photoconductivity and photoluminescence is well observable in the figure presented by Tanaka [1, 22]. The primary reason of way the optical quantities become saturated at sufficiently low temperatures is the production of monoenergetic phonons arisen with the optical absorption associated with non-radiant recombinations of *e-h* pairs. Thus, the existence of monoenergetic phonons enables to clarify not only the essence of the MNR, but also the Urbach rule, and even other connections between optical phenomena.

### 6. Conclusion

An explanation of the relation of the Meyer-Neldel rule to the Urbach rule has been given in this paper. These two phenomena – as we believe - are connected via

monoenergetical phonons offering the possibility to explain the MNR rule. These monoenergetical phonons influence the optical absorption. The monoenergetical phonons are also created in a substance at photoabsorption. Exploiting a similar approach, it is possible to explain observed dependences concerning photoconductivity and photoluminescence in the low temperature region.

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