COMPENSATION EFFECT IN THE AMORPHOUS HYDROGENATED SILICON FROM POINT OF VIEW OF THE BARRIER-CLUSTER MODEL

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ABSTRACT

Although hydrogenated amorphous silicon has its important practical application there still is not any generally accepted model explaining all the physical phenomena on-going in this matter. The aim of this paper is to familiarize the reader with a model allowing to explain the empirically observed compensation effect (Meyer-Neldel rule – MNR). This effect reflects the correlation between activation energy and pre-exponential factor in relation to the activation dependence of electrical conductivity of semiconductors, including a-Si:H. The model assumes that the recombination of carriers is conditioned by emission of series of monoenergetic phonons. The number of emitted phonons is dependent on the activation energy of disordered semiconductor, which influences the probability of recombination and thus the concentration of free electrons. Consideration of the relationship between compensation effect and Urbach rule is also presented.

Keywords: *hydrogenated amorphous silicon, compensation effect, Meyer-Neldel rule, pre-exponential factor, hopping MNR*

1. INTRODUCTION

Amorphous silicon, and its more useful alloy form, hydrogenated amorphous silicon (a-Si:H), has been the subject of investigation for over 30 years [1-3]. Research into a-Si began in England [4], but the major development, the demonstration of doping and the subsequent implementation of devices was made in Scotland by Spear and LeComber [4-7].

A-Si:H is a low-cost, efficient material which is used extensively for electronic devices. The advantages of a-Si:H are particularly evident when considering the photovoltaic application. The a-Si:H has a large optical absorption coefficient, the energy gap can be modulated to allow optimum conversion efficiency for sunlight. It can be alloyed with other elements (carbon, germanium) to create multijunction structures with increased energy conversion efficiency for sunlight.

The intrinsic disorder in amorphous silicon (a-Si) creates broken bonds, which negatively impact the electronic properties of materials. The main role of hydrogen in amorphous silicon is the passivation of the Si dangling bonds to restore a proper energy gap and the semiconducting properties, thus enabling extensive application of a-Si:H in the microelectronics and the photovoltaic industry. However, several questions are still unanswered.

The first report of hydrogenated amorphous silicon solar cells in 1976 [8] sparked worldwide interest as a promising economically viable source of renewable energy. The materials used in the a-Si:H based solar cells are in fact silicon hydrogen alloys which typically contain about 5 to 20 % of hydrogen.

Serious problem of solar cells based on a-Si:H is so called Staebler-Wronski effect [9] associated with light induced degradation of the material. Continuous progress has been made over the years not only in improving the initial properties of a-Si:H based materials but also in reducing their light induced degradation.

Hydrogenated amorphous silicon is still the subject of intense research. Dark conductivity in a-Si:H can be described by two main processes. The first is the standard extended states conduction process. The second conduction process is referred to as variable-range hopping (VRH) conduction. Great attention is paid to the system a-Si / C: H, respectively a-C:H.

2. MEYER-NELDEL RULE AND a-Si:H

Semiconductors are usually characterized by the temperature dependence of their electrical conductivity, σ

$$\sigma = \sigma_0 \exp\left(-\frac{W}{kT}\right), \tag{1}$$

where σ_0 is a constant and W – activation energy [10]. Activation dependence (1) corresponds to the first of these mechanisms. For many classes of materials, especially amorphous hydrogenated silicon, chalcogenide glasses, and organic semi-insulators, experimental evidence suggests that a correlation exists between the activation energies and pre-exponential factors in the following form [11-15]

$$\sigma_0 = \sigma_{00} \exp(bW) \tag{2a}$$

or

$$\ln \sigma_0 = \ln \sigma_{00} + bW, \tag{2b}$$

where b and σ_{00} are constant. This relation can be written as

$$\sigma_0 = \sigma_{00} \exp\left(\frac{W}{kT_{MN}}\right), \quad \sigma_0 = \sigma_{00} \exp\left(\frac{W}{E_{MN}}\right). \tag{3}$$

Here $b = 1/kT_{MN} = 1/E_{MN}$. The value $E_{MN} = kT_{MN}$ is the MN-energy and T_{MN} the MN-temperature. Equation (2) is often referred to as the compensation rule, or Meyer-

Neldel empirical rule (MNR). Constant σ_{00} is often called the Meyer-Nedel pre-exponential factor and $E_{MN} = kT_{MN}$ the Meyer-Neldel characteristic energy.

For electric conductivity of above mentioned group of substances holds (1,3)

$$\sigma = \sigma_{00} \exp\left(\frac{W}{kT_{MN}}\right) \exp\left(-\frac{W}{kT}\right) . \tag{4}$$

An a-Si:H doped with different impurities, which affect the activation energy, shows such dependence (Fig. 1). Amorphous hydrogenated carbon a-C:H and systems of a-Si / C:H have similar behaviour.

MN rule holds in disordered materials when W varies by doping, by surface absorption, light soaking or by preparing films under different conditions. This rule has also been observed for 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 on changing the composition of the glassy alloys in a specific glassy system.
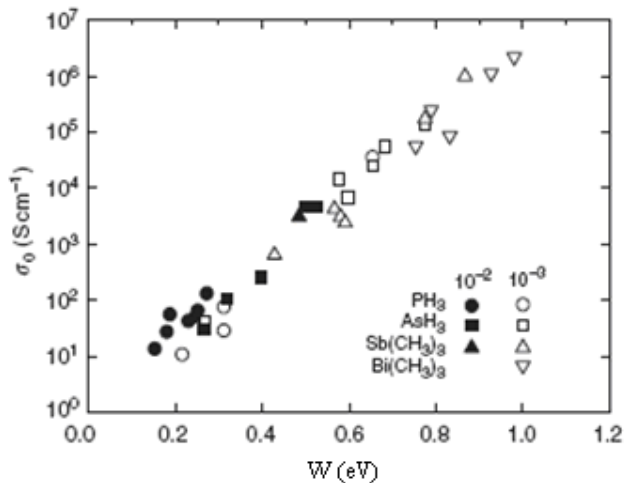

Fig. 1 The pre-exponential factor σ_0 plotted as a function of activation energy W for a-Si:H doped with various dopants. The fitting of the experimental data (rel.3) produces $E_{MN} = 67$ meV and $\sigma_{00} \approx 1$Scm^{-1} [16]

3. 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 [11,12]. In our barrier-cluster 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 the 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 compensation effect - Meyer-Neldel rule.

3.1. 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

$$N = 2W/\Delta E . \tag{5}$$

Let w_1 be the probability of production of one phonon which energy is equal to ΔE. Probability w_N of production of N phonons of equal energy ΔE due to the electron – lattice interaction will be

$$w_N = (w_1)^N . \tag{6}$$

If we write down probability w_1 as

$$w_1 = \exp(-\varepsilon_1), \tag{7}$$

where ε_1 is a positive value, then probability w_N one can write with respect to (6,7) as

$$w_N = \exp(-N\varepsilon_1) = \exp(-\varepsilon_1 2W/\Delta E) = \exp(-bW) , \tag{8}$$

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.

3.2 Equilibrium concentration of conduction electrons

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

$$G = \left(\frac{dn}{dt}\right)_{recom} = C_1 \exp\left(-\frac{W}{kT}\right) , \tag{10}$$

where C_1 is a constant. For a recombination process we suppose (in accordance with the relation (8)) that the relation

$$R = \left(\frac{dn}{dt}\right)_{recom} = nC_2 \exp\left(-bW\right) \tag{11}$$

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

$$C_1 \exp(-W/kT) = n.C_2 \exp(-bW) . \tag{12}$$

From that relation follows for equilibrium concentration n of free carriers

$$n = C_{00} \exp(bW) \exp(-W/kT), \qquad (13)$$

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

3.3. Electric conductivity

It is known that electric conductivity σ is proportional to n: $\sigma \approx n$. If mobility of carriers is independent on activation energy W (or that dependence is negligible) and if we put $b = 1/kT_{MN}$, one can write with respect to (13)

$$\sigma = \sigma_{00} \exp \left(\frac{W}{kT_{MN}} \right) \exp \left(-\frac{W}{kT} \right) \qquad (14)$$

and for the pre-exponential factor

$$\sigma_0 = \sigma_{00} \exp \left(\frac{W}{kT_{MN}} \right), \qquad (15)$$

which expresses the conventional compensation effect – or Meyer-Neldel rule (3).

4. HOPPING CONDUCTION AND "HOPPING MNR"

At low enough temperatures, variable-range hopping (VRH) in the localized states is expected to become predominant as the number of free carriers in the band decreases. Assuming a constant density of states $N(E_F)$ within kT of the Fermi level, Mott [7] obtained for VRH,

$$\sigma = \sigma_0 \exp \left(- \left(\frac{T_H}{T} \right)^{1/4} \right). \qquad (16)$$

Here, σ_0 and T_H are functions of $N(E_F)$. Eq. (16) is widely known as Mott's $T^{-1/4}$ law of VRH.

It was empirically found [17] that many semiconductors including a-Si:H, in the area of hopping conduction follow a new MNR - hopping MNR, namely

$$\sigma_0 = \sigma_{00} \exp \left(\left(\frac{T_H}{T_{HMN}} \right)^{1/4} \right), \qquad (17)$$

where, in analogy with $1/E_{MN}$ of the conventional MNR, we have defined $(1/T_{HMN})^{1/4}$ as the slope of the straight line (3), in the hopping MNR plot.

In the previous we have presented a possible explanation for the conventional MNR (2, 3). Concerning hopping MNR, there is no available explanation. In this paper we would like to show some formal parallel concerning two empirical relationships. These relationships are indeed quite different, corresponding to the two different mechanisms of electrical conductivity, but anyhow there is some similarity. Let us look at the equations

$$\sigma = \sigma_{00} \exp \left(\frac{W}{E_{MN}} \right) \exp \left(-\frac{W}{kT} \right), \qquad (18)$$

$$\sigma_0 = \sigma_{00} \exp \left(\left(\frac{T_H}{T_{HMN}} \right)^{1/4} \right) \exp \left(-\left(\frac{T_H}{T} \right)^{1/4} \right). \qquad (19)$$

Comparing both relations (18,19) we see that the role of activation energy W in the second relation was taken by variable T_H and E_{MN} was replaced by variable T_{HMN}. Exponential expressions on the right sides of both equations represent the character of temperature dependence of electrical conductivity at different conduction mechanisms. Exponential expressions in terms of pre-exponential factors (first from left) are in analogical way "linked" to an exponential expressions figuring after them.

The analogy of two relations offers the idea that both types of MNR (conventional MNR and hopping MNR) are aroused by some analogous mechanism. By the hopping conductivity – as it is commonly presented in literature – it is about processes taking place close to the Fermi level E_F. In this case, activation energy is not involved into the process. On the other hand, it is generally accepted that electron hopping cannot take place without the participation of phonons and the phonons mean energy. Therefore, some authors are talking about the "hopping energy", respectively the level at which hopping takes place. But so far there is no uniform view on this issue.

Theoretically, the value T_H in relation (19) could be related with the ability of the grid to absorb e.g. impulse of an electron after hopping-skipping to a new position at the free level in the mobility gap. We have in mind the probability of such absorption. The question is: will this "impulse absorption" go on in certain doses as it is by the absorption of energy $2W$ by conventional MNR? Is it not impulse, respectively quaziimpulse hidden by variable T_H? Variable T_{HMN} might be then connected with the grid ability to absorb change of the impulse and thus actually sending compensation phonons. These are however only our questions.

Notes: Physical properties of disordered semiconductors are the topic of the works [18-25]. The explanation of some electrical and optical properties of disordered semiconductors based on barrier-cluster model was published in [11,12,26-30]. In the paper [26] was published "Band model of diffusion", that allows to explain compensation effect in the diffusion and diffusion-related processes, which include catalysis, crystallization.

5. MNR AND OPTICAL ABSORPTION

In this paragraph we will be interested in the optical absorption in the area of exponential tail, where Urbach's rule holds.

5.1. The Urbach's rule

The Urbach's rule is an empirical rule, describing the absorption of light in exponential tails of optical absorption spectra [5-7,23]. It expresses the dependence of the coefficient of the optical absorption α on the photon energy hf and the temperature T. It can be expressed mathematically by the relation

$$\alpha = C \exp(\gamma \frac{hf - E}{kT}), \tag{20}$$

or equivalently as

$$\ln \alpha = \left(\gamma \frac{hf - E}{kT} \right) + const. \tag{21}$$

Thus, $\ln \alpha(hf)$ 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 (20, 21) but the real temperature T has to be replaced by a constant parameter T^*. This point was not explained satisfactorily up to now [5, 18, 23].

6. AN ATEMPT TO EXPLAIN THE URBACH'S RULE

The Urbach's rule can be explained on the base of 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 [27-30]. 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 photon energy $hf < 2W$ is less than the width of the forbidden gap 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

$$p = C \exp(-\frac{2W - hf}{2kT}). \tag{22}$$

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

$$\alpha = C \exp(\frac{hf - 2W}{2kT}) = C \exp(\gamma \frac{hf - 2W}{kT}) \tag{23}$$

where $\gamma = \frac{1}{2}$, C is a constant. Thus

$$\ln \alpha = \left(\frac{hf - 2W}{2kT} \right) + const. \tag{24}$$

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 disordered semiconductors (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 (24) continues being unchanged with temperature at low temperatures. Only a parallel shift to the lower absorption of "exponential tails" is observed.

The relation (23) is no longer valid. At lowering the temperature of a non-crystalline semiconductor, the parameter T in relation (23) no longer represents temperature. It seems as if the temperature was freezed at a certain value of T^*. 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 (experiments) about $0.4 - 0.55$ [6,7] in the chalcogenide glasses. When comparing (20) and (23), we see that the (barrier-cluster model) yields the constant $\gamma = 0.5$. This may be considered as a good agreement in the research of non-crystalline semiconductors.

Remarks: We cannot agree with the idea that the exponential tails are caused by the distribution of density of states in the mobility gap. Of course, the possible density of states can to some extent influence exponential tails, but we do not consider it the primary cause of such tails. We consider the participation of phonons in absorption the primary cause of tail formation. By this it is easy to explain "universality" of tails - exponential character and also practically the same slope in a rich variety of semiconducting materials. Something like this cannot be provided by the density of states of various and varied materials. In our interpretation, the distribution density of states is a secondary factor. This is in agreement with the characteristics of a-Si, respectively a-Si:H, in which the standard exponential tails appear just after the elimination of free bonds, thus after the hydrogenation. Thus the tails appear after the forbidden zone purification.

6.1. MNR and Urbach's rule – consideration and questions

The basic idea is subsequent. As it has been said above, the energy released at non-radiative recombination of *e-h* pairs is transformed into the energy of mono-energetic phonons. These phonons create a phonon field in a semiconductor. At higher temperatures this additional phonon field is relatively "weak" and that is why its effect does not appear significant – thermal phonons dominate. At higher temperatures, the relation (23) holds, where T denotes the temperature. The slope of a straight line $\ln \alpha(hf)$ decreases with the increase of the temperature.

At lower values of kT, it will be necessary to accept the substitution

$$kT + kT^* = E_U,$$

instead of kT. The second term, kT^*, represents effective energy of mono-energetic phonons. It will be valid that

$$\alpha = C \exp(\gamma \frac{hf - 2W}{E_U}) = C \exp(\gamma \frac{hf - 2W}{kT + kT*}). \tag{25}$$

By lowering the temperature, the first term kT becomes small and the second term $kT^* = E^*_U$ begins to dominate. By a subsequent cooling, the relation achieves the limiting form

$$\alpha = C \exp(\gamma \frac{hf - 2W}{E^*_U}) . \qquad (26)$$

A result is that the slope of the exponential tails (in the straight line representation) by lowering temperatures will gradually become smaller. It looks as if at very low temperatures the tail of optical absorption is shifted almost parallel to the lower absorption. The limiting straight line will be the one, which corresponds to the exponential function (26). The question remains: Can the considered effect be the major cause of Urbach's rule in the region of low temperatures?

Remarks: The described effect of monoenergetic phonons (produced in non radiative recombination), the optical absorption is in principle certainly possible, and real. An open question is the extent of such an effect. It is possible that under normal conditions, the effect is negligible and that it occurs noticeably only at sufficiently low temperatures and high light intensity. We also cannot exclude certain "resonance effect" by absorption of monoenergetic phonons that was produced before by considered system alone. Questions remain.

7. CONCLUSIONS

Electrical conductivity in a-Si:H can be described by two main processes. The first is the standard extended states conduction process. The second conduction process is referred to as variable-range hopping (VRH) conduction. Validity of the conventional Meyer-Neldel rule in non-crystalline semiconductors is in presented paper explained assuming that recombination transition of an electron from conduction band to valence band is conditioned by emission of a number of monoenergetical phonons. Probability of such complex process is determined by number of emitted phonons. This probability exponentially decreases with increasing number of phonons and consequently with the width of the forbidden band. The probability of recombination influences equilibrium concentration of free electrons in conduction band and subsequently electric conductivity. In such a way we have finally obtained the relation which is equivalent to compensation effect, or the Meyer-Neldel rule. By lower temperatures in disordered semiconductors the hopping transport dominates. In this type of electrical conductivity specific "hopping MNR" is experimentally observed.

It is shown in the paper that monoenergetic phonons can influence optical absorption. This enables explaining of Urbach´s rule in low temperature area.

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