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THERMOELECTRIC HEATING AND COOLING IN SEMICONDUCTOR STRUCTURES: NONEQUILIBRIUM CHARGE CARRIERS. (Review)

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The paper is dedicated to the analysis of thermoelectric cooling phenomena in semiconductors containing potential barriers at the p-n-junction interface. The formulation of an adequate self-consistent theoretical model describing the effect is presented. The role of the recombination rate as a new source of heat in linear approximation of the electric current is discussed, leading to a reformulation of the heat balance equations. The presented recombination of the nonequilibrium carriers always depends on the temperature heterogeneity connected with thermoelectric cooling. Therefore, the nonequilibrium carrier concentrations do not disappear even at very short life times. The importance of redistribution of nonequilibrium charge carriers which has been ignored in most publications on this subject is also shown. The conventional theory of thermoelectric cooling, not taking into account the influence of the nonequilibrium charge carriers, is shown to be inadequate. Besides, when the recombination rate decreases, cooling changes to heating.

Key words: Joule effect, Peltier effect, Thomson effect, Seebeck effect.

Introduction

Traditionally, researchers considered thermoelectric cooling (heating) in terms of the availability of sinks or sources of heat in a heterogeneous system through which the electric current flows [1]. However, in 2 – 4 it has been shown that the thermodynamic process of cooling (heating) can be explained by considering the Le Chatelier-Braun principle [5]. To summarize the content of 2 and 4, the change in the drift heat flux $q_{dr} = \Pi j$ (where Π is the Peltier coefficient and j the electric current density) in a heterogeneous system causes a thermodiffusion heat flux $q_{diff} = -\kappa \nabla T$ (where κ is the thermal conductivity and T the temperature) compensating this change. Due to this thermodiffusion heat flux, temperature heterogeneity arises that cools (heats) the system depending on the electric current direction and material properties. When the temperature in the system is below the equilibrium temperature, we have the effect of thermoelectric cooling, and when the temperature is above the equilibrium temperature, we have the thermoelectric heating effect.

A *p-n* structure is used to make the thermoelectric refrigerator, [1, 6 – 9] because of thermoelectric drift fluxes directed (at a corresponding direction of a current from the *n*-region to *p*-region) from the interface toward the edge in both layers of the *p-n* structure that strengthens the cooling phenomenon [2]. Traditionally studies of the Peltier effect do not consider the nonequilibrium charge carriers [1 – 4, 6 – 9], so that only majority charge carriers and their electric current are taken into account in the expressions for heat fluxes in *n*- and *p*-regions, in spite of the fact that the current of minority charge carriers near the *p-n* junction has the same order of magnitude as the current of

majority charge carriers [10]. In this way, the thermal generation and extraction of minority charge carriers must take place near the interface to allow the flow of electric current [10]. As a consequence, nonequilibrium charge carriers will arise. In Ref. 11 some aspects of this problem were discussed. On the other hand, the effect of nonequilibrium charge carriers on the generation of thermoelectromotive force (EMF) (Seebeck effect) has been studied in detail [12, 13]. In this work, an alternative approach to the physics of thermoelectric cooling in *p-n* junctions is presented [14-16].

Nonequilibrium Charge Carriers Effect On Thermoelectric Cooling (Heating)

In the linear approximation with respect to the electric current, the heat balance equation is given by [2], [17]:

$$\nabla \cdot q = \varepsilon_g R, \tag{1}$$

where $q = q_{dr} + q_{diff}$ is the heat flux, R is the recombination rate, ε_g is the band gap. In Section III we will show that, in linear approximation, for both strong and weak recombination rates the right hand side of Eq. (1) becomes null.

The expression for q_{dr} in bipolar semiconductors is as follows:

$$q_{dr} = P_n j_n + P_p j_p \tag{2}$$

Here q_{dr} are the drift heat fluxes in *n*- and *p*-type semiconductors, $j_{n,p}$ and $P_{n,p}$ are the electron and hole electrical currents and the Peltier coefficients.

The Peltier coefficients in nondegenerated semiconductors are defined by the following expressions [7]:

$$\Pi_{n,p} = \mp \frac{1}{e}\left(\left[r_{n,p} + 5/2 \right] T - \mu_{n,p} \right) \tag{3}$$

where $\mu_{n,p}$ are the Fermi quasi-levels for electrons and holes in *n*- and *p*-regions, *e* is the hole charge, and $r_{n,p}$ are the exponents in the momentum relaxation times [18]. It should be borne in mind that the absolute value of the Peltier coefficient of minority carriers can be much in excess of that of majority carriers.

Since the Fermi quasi-levels depend on the concentration of majority and minority charge carries, the Peltier coefficients will depend on the coordinate x (in the space-charge layer near the *p-n* junction ($-r_D^n < x < r_D^p$, where $r_D^{n,p}$ is the Debye radius in the *n*- and *p*-region)) even in linear approximation of Eq. (2) with respect to the current, due to the coordinate dependence of the equilibrium concentrations near the *p-n* junction.

The expression for diffusion heat fluxes $q_{diff}^{n,p}$ is as follows:

$$q_{diff} = -\left(\kappa_n + \kappa_p + \kappa_{ph} \right) \nabla T , \tag{4}$$

where κ_n, κ_p, and κ_{ph} are the electron, hole, and phonon heat conductivities in *n*- and *p*-regions.

Since $\kappa_{n,p} << \kappa_{ph}$ in nondegenerated semiconductors then Eq. (4) reduces to:

$$q_{diff}^{n,p} = -\kappa_{ph}^{n,p} \nabla T, \tag{5}$$

Taking into account the considerations given above, the heat balance Eq. (1) can be rewritten as follows:

$$-\kappa_{ph} \Delta T + \Pi_n \nabla \cdot j_n + \Pi_p \nabla \cdot j_p + j_n \nabla \Pi_n + j_p \nabla \Pi_p = \varepsilon_g R \tag{6}$$

Since the current densities can be calculated as [19]:

$$\begin{aligned} j_n &= -\sigma_n\left(\nabla\phi - \nabla\mu_n/e + \alpha_n\nabla T\right), \\ j_p &= -\sigma_p\left(\nabla\phi + \nabla\mu_p/e + \alpha_p\nabla T\right), \end{aligned} \tag{7}$$

then the calculation of μ_n, μ_p, and ϕ is required. Here $\sigma_{n,p}$ are the electric conductivities, $\alpha_{n,p}$ the Seebeck coefficients ($\Pi_{n,p} = \alpha_{n,p}T$), and ϕ the electric potential.

The macroscopic description of the transport of nonequilibrium charge carriers is performed using the continuity equations for the electron and hole current densities and the Poisson equation [20]:

$$\nabla \cdot j_n = eR, \tag{8a}$$

$$\nabla \cdot j_p = -eR, \tag{8b}$$

$$\Delta\phi = 4\pi\frac{\rho}{\varepsilon} \tag{9}$$

where ρ are the space charges, ε the permittivities, and R the recombination rates in n- and p-regions.

The recombination rate in the presence of temperature gradient was obtained in [20], [21]:

$$R = \tau^{-1}\left[\left(n(x) - n_0\right) + A\left(p(x) - p_0\right) + \beta\left(T(x) - T_0\right)\right], \tag{10}$$

where n and p are the concentrations of electrons and holes, n_0 and p_0 the equilibrium concentrations of electrons and holes, and T_0 the equilibrium temperature. Expressions for τ, A, and β can be found in [20], [21]. It must be emphasized that τ, A and β only depend on the semiconductor properties. It is important to note that τ varies in inverse proportion to the capture coefficients, whereas A and β are finite at any magnitude of the capture coefficients. Let us emphasize that the chemical potentials of electrons and holes ($\mu_{n,p}$) and their concentrations (n and p) are connected by simple algebraic expressions [13]. Let us note that $\mu_n + \mu_p = -\varepsilon_g$ at equilibrium. Hence the system of four equations (7) – (9) describes the behaviour of four unknown functions δn, δp, $\delta\phi$, and δT (or $\delta\mu_n$, $\delta\mu_p$, $\delta\phi$, and δT), where $\delta n = n - n_0$, $\delta p = p - p_0$, $\delta\phi = \phi - \phi_0$, $\delta T = T - T_0$, $\delta\mu_{n,p} = \mu_{n,p} - \mu^0_{n,p}$ (T_0, $\mu^0_{n,p}$, and ϕ_0 are the temperature, chemical potentials, and electric potential of the p-n structure at equilibrium respectively). Nonequilibrium temperatures appear in our linear problem from Peltier effect [2-3]. In linear approximation, $\delta n = (n_0/T_0)\delta\mu_n$, $\delta p = (p_0/T_0)\delta\mu_p$ [13].

The charge density in Eq. (9) may be written as $\rho = \rho_0 + \delta\rho$,where ρ_0 and $\delta\rho$ are the equilibrium and non equilibrium densities of charge that are composed of electrons, holes and the carriers (both electrons and holes) captured on impurity levels, therefore [14, 23]

$$\delta\rho = e(B\delta p - C\delta n + D\delta T) \tag{11}$$

An approximation commonly used to solve the system of equations (8 – 9) in p-n junctions is the assumption of quasineutrality out of the space charge region [24]. The use of the quasineutral approximation is acceptable if the quasi-neutral region length and the minority carrier diffusion lengths are both larger than the Debye length. In this case [13, 23] instead of the Poisson equation we have (see Eq. (11)):

$$\delta\rho = 0 \tag{12}$$

Eq. (7) may be rewritten as:

$$R = R_n = R_p = 1/\tau[E\delta n + F\delta T] \tag{13}$$

Boundary Conditions

The system of Eqs. (1) and (8-9) which defines the thermoelectric cooling must be complemented with the appropriate boundary conditions, which describe the electric currents, the heat flux, and the electric potential through the interface. A very important question is the choice of the boundary conditions used when solving the carrier-transport equations. It should be noted that the expressions commonly used are valid only for semiconductor devices operating in open-circuit conditions (see, for instance Ref. 10). Since in normal operation a current flows at the terminals, the widespread use of boundary conditions for open-circuit conditions is incorrect. For closed-circuit conditions, a different set of boundary conditions needs to be derived. This problem has only been addressed in the last few years [19, 22].

Let us assume that in the *y*- and *z*- direction the *p-n* junction is adiabatically isolated. Then the boundary conditions in the remaining direction (i.e. the *p-n* junction interface is orthogonal to the *x*-axis and assuming that the interface is located at $x = 0$, the n region located between $x = -l_n$ and $x = 0$, the p region between $x = 0$ and $x = l_p$) are given below.

Assuming that an ideal metal-semiconductor contact is placed at $x = -l_n$ we can write the following boundary conditions for the excess of temperature and carrier densities (hereafter a superscript *n* or *p* in a magnitude refers to the *n* or *p* region respectively):

$$\delta T^n(-l_n) = 0 \tag{14}$$

$$\delta n(-l_n) = 0 \tag{15}$$

$$\delta\phi(-l_n) = 0 \tag{16}$$

These boundary conditions are justified because of the high value of the thermal conductivity of metals and intense recombination at the metal-semiconductor interface. Similar boundary conditions may be written at the metal-semiconductor interface at $x = l_p$:

$$\delta T^p(l_p) = 0 \tag{17}$$

$$\delta p(l_p) = 0 \tag{18}$$

$$\delta\phi(l_p) = -V \tag{19}$$

where V is the applied voltage. These boundary conditions assume that the semiconductor is at the equilibrium in $x = -l_n$ and in $x = l_p$, in other situation, the electric potential (i.e. Eqs. (16) and (19) cannot be rigorously defined [25].

At the *p-n* junction interface we can introduce three additional boundary conditions [19], [22]:

$$\begin{aligned}
&\delta\phi^n(0) - \frac{\delta\mu_n^n(0)}{e} - \frac{1}{e}\frac{\partial\mu_{n_0}^n}{\partial T}\delta T^n(0) = \\
&\delta\phi^p(0) - \frac{\delta\mu_n^p(0)}{e} - \frac{1}{e}\frac{\partial\mu_{n_0}^p}{\partial T}\delta T^p(0) \\
&\delta\phi^n(0) + \frac{\delta\mu_p^n(0)}{e} - \frac{1}{e}\frac{\partial\mu_{n_0}^n}{\partial T}\delta T^n(0) = \\
&\delta\phi^p(0) + \frac{\delta\mu_p^p(0)}{e} - \frac{1}{e}\frac{\partial\mu_{n_0}^p}{\partial T}\delta T^p(0)
\end{aligned} \tag{20}$$

$$Q^n(0) = Q^p(0) \tag{21}$$

$$j_n^n(0) = j_n^p(0) \tag{22}$$

$$\delta T^n(0) = \delta T^p(0) \tag{23}$$

These boundary conditions are obtained, assuming respectively continuity of the electrochemical potential at the interface that both heat and electrical conductivities are very large at the junction, and absence of surface recombination. In fact, because the p-n interface is inside the depletion region, this assumption is not realistic and boundary conditions with finite conductivities need to be used [19], in this work we use Eq. (20) – (23) for the sake of simplicity. Finally, it should be noted that in the regions where quasi-neutrality condition holds $\delta\rho = 0$ and the Poisson equation becomes algebraic adopting the form $\delta n = -A'\delta p - B'\delta T$ where A' and B' are constants.

Simplification of the Model of Thermoelectric Cooling in two Limiting Cases

In this Section we will analyse the thermoelectric cooling in a p-n junction in the two limiting cases: strong and weak recombination.

A. Weak Recombination

Let us now consider that the volume recombination is weak. In this case, the conditions $l_D >> l_{n,p} >> r_D$ are fulfilled, which means that the weak recombination is correct for thin film *p-n* structures. Formally $R = 0$ when $\tau \to \infty$. Under this condition the right-hand side of Eq. (1) becomes trivially null and Eq. (1) along with Eqs. (4) and (5) transforms into:

$$\Delta T = 0 \tag{24}$$

and Eqs. (8) into:

$$\nabla \cdot j_{n,p} = 0. \tag{25}$$

From Eqs. (25) it follows that $j_{n,p}$ are not dependent on the coordinates and $j_n^n + j_p^n = j_n^p + j_p^p = j_0$, where j_0 is the whole of current through *p-n* structure. From the boundary conditions for currents [19] it follows that $j_n^n + j_n^p = j_0^n$, $j_p^n + j_p^p = j_0^p$ $(j_0^n + j_0^p = j_0)$.

It is not difficult to understand that the concentrations of the nonequilibrium carriers (δn and δp) are maximal in this case.

It may seem that the calculation of the thermoelectric cooling does not require the use of Eqs. (25) in the absence of recombination because there are no other unknown functions in Eq. (24). So, it would look like that the thermoelectric cooling does not depend on the nonequilibrium carrier concentrations. However, the boundary conditions to Eq. (24) must be formulated for heat fluxes (Eqs. (2) and (5)). The drift heat fluxes depend on the current of majority and minority carriers (Eq. (2)). The latter essentially depends on the distribution of nonequilibrium carrier concentration because of the terms $(\nabla\mu_{n,p})/e$. Therefore there are no reasons to assume a priori that $j_n^n >> j_p^n$ and $j_p^p >> j_n^p$.

The problem is reduced to the calculation of currents in the electrical circuit composed of two circuits connected in parallel. One of them is composed of two *n*-type semiconductors connected in series with the concentrations n^n and n^p, whereas the other is composed of two *p*-type semiconductors connected in series with the concentrations p^n and p^p. At a chosen direction of current (from *n*- to *p*-region) heating instead of cooling takes place at weak recombination [2, 11].

With respect to what was said above, let us notice that the classic theory for current-voltage characteristic through the *p-n* junction [26] obeys the following expression:

$$j_0 = j_s\left(\exp\left(\frac{eV}{T}\right)-1\right), \tag{26}$$

where the saturation current (j_s) varies in direct proportion to the capture coefficients. It follows from Eq. (26) that the current j_0 through the *p-n* junction is equal to zero when the recombination is absent under any voltage.

It means that the model (Eq. (26)) is not correct when the recombination is weak. At the same time, Eq. (25) (together with Eq. (24)) will give the correct expression for current j_0 (at least under weak voltage). The main result is that the temperature deviation from equilibrium at the junction may be obtained:

$$\begin{aligned}
\delta T^n(0) = {} & j_0 l_n\left[\left(\Pi_n^p+\Pi_p^n\right)^2\left(\left(\Pi_n^p\right)^2\frac{l_p}{\sigma_p^p}+\left(\Pi_p^n\right)^2\frac{l_n}{\sigma_n^n}\right)\right]+\\
& +j_0 l_n H\left[\left(\Pi_n^p\right)^2\left(\frac{l_p}{\sigma_n^p}+\frac{l_n}{\sigma_p^n}\right)\left(\frac{l_n}{\sigma_p^n}+\frac{l_p}{\sigma_p^p}\right)\right]+\\
& +j_0 l_n H\left[\left(\Pi_p^n\right)^2\left(\frac{l_p}{\sigma_n^p}+\frac{l_n}{\sigma_p^n}\right)\left(\frac{l_p}{\sigma_n^p}+\frac{l_p}{\sigma_n^n}\right)\right]+\\
& +j_0 l_n H\left[\left(\Pi_p^n+\Pi_n^p\right)^2\left(\frac{l_p}{\sigma_n^p}+\frac{l_n}{\sigma_p^n}\right)\frac{l_n l_p}{\sigma_n^p\sigma_p^n}\right]
\end{aligned} \tag{27}$$

The expression of H is:

$$H = \frac{T_0}{l_n l_p}\left(\kappa_{ph}^n l_p + \kappa_{ph}^p l_n\right) \tag{28}$$

From this expression it follows that a positive current will generate heat instead of cooling at the junction in strong contrast with the conventional results.

B. Strong Recombination

Let us assume that the recombination is very strong. The physical meaning is that $l_{n,p} >> l_D \to 0 >> r_D \to 0$. From a mathematical point of view we have $\tau \to 0$ in Eq. (10).

Since the recombination rate (R) cannot be infinite at $\tau \to 0$, then from Eq. (10) it follows that:

$$\delta n = -A\delta p - \beta\delta T. \tag{29}$$

At the same time, the magnitude R is finite but not defined. Adding Eqs. (8a) and (8b) we have:

$$\nabla\cdot\left(j_n^n + j_p^n\right) = 0,\ \ \nabla\cdot\left(j_n^p + j_p^p\right) = 0. \tag{30}$$

It is important to emphasize that the nonequilibrium charge carrier concentrations (δn and δp) are not equal to zero in the considered approximation. Therefore, there is no reason to state that $j_n^n >> j_p^n$ and $j_p^p >> j_n^p$.

Therefore, the volume Eq. (1) transforms again into:

$$\Delta T = 0. \tag{31}$$

Once again, just like in the weak recombination case, the right-hand side of Eq. (1) also becomes zero, but for very different physical reasons. But, as in the case of weak recombination, the heat flux depends on the nonequilibrium carrier concentrations. The latter are defined by Eqs. (29), (30), and (31) with the corresponding boundary conditions.

In the previous case, it was noted that Eq. (26) is not correct when the recombination is weak enough. Also, it is not difficult to understand that Eq. (26) is not correct in the case of strong recombination. It follows from the expression for j_s that $j_s \to \infty$ when $\tau \to 0$ at any applied voltage V. The last statement is not correct from the physical point of view. The method described above allows the calculation of the current-voltage characteristic of the *p-n* junction in the case of the strong recombination in the linear regime with respect to the applied voltage V.

The temperature deviation at the junction has been analytically obtained:

$$\delta T^n(0) \propto j_0 l_n \left[\sigma_n^n \sigma_p^p \left(\Pi_n^n - \Pi_p^p \right) + \sigma_p^n \sigma_p^p \Pi_p^n - \sigma_n^n \sigma_n^p \Pi_n^p \right] \tag{32}$$

This expression clearly differs from that commonly used:

$$\delta T^n(0) \propto j_0 \sigma_n^n \sigma_p^p \left(\Pi_n^n - \Pi_p^p \right) l_n \tag{33}$$

The differences are not only in magnitude but also in sign. Contrary to Eq. (33), that for the positive values of j_0 only predicts increases in temperature with j_0, Eq. (32) predicts that a *p-n* junction under the same bias conditions (positive values of j_0) may be heated or cooled depending on the values of the Peltier coefficients and electrical conductivities in the *p-n* junction. Moreover, Eq. (32) clearly shows the paramount importance of the nonequilibrium carriers at both sides of the junction (neglected in Eq. (33)) since they control the sign of $\delta T^n(0)$.

Finally, let us emphasize that only when the following two criteria are simultaneously met:

$$\frac{\Pi_n^n}{\Pi_n^p} \gg \frac{\sigma_n^p}{\sigma_p^p} \quad \frac{\Pi_p^p}{\Pi_p^n} \gg \frac{\sigma_p^n}{\sigma_n^n} \tag{34}$$

Eq. (32) reduces to Eq. (33).

When is the Conventional Theory of the Thermoelectric Cooling Correct?

The equations used to describe the thermoelectric cooling in conventional theory are [1 – 4, 6 – 9] Eq. (31) and

$$q_n = -\kappa^n{}_{ph} \nabla T + \Pi^n{}_n j^n{}_n \tag{35}$$

$$q_p = -\kappa^p{}_{ph} \nabla T + \Pi^p{}_p j^p{}_p \tag{36}$$

and it is assumed that $j^n{}_n = j^p{}_p = j_0$ and do not depend on the coordinates. Also $\Pi^n{}_n$ and $\Pi^p{}_p$ are assumed to be constant in space.

However, the question "when is the appointed model correct?" is not discussed in any of the references cited above (and it is absent from all the works concerned with the problem of the thermoelectric cooling). Only in Ref. 11 was the previous question considered from physical (but not mathematical) point of view. Therefore, the following question arises: "can Eqs. (31), (35), and (36) be correct?" and if the answer is "yes", then "under what conditions?"

As it was shown in the previous section, the heat balance equation has the form of Eq. (31) in the two limit cases (strong and weak recombination) if the quasineutrality condition is assumed. Eqs.

(35), (36) do not apply to the weak and intermediate recombination cases. Therefore, we focus our attention only on the strong recombination case. We observe that Eqs. (2) and (5) can be reduced to Eqs. (35)-(36), if the following conditions are fulfilled:

$$j^n{}_n >> j^n{}_p, j^p{}_p >> j^p{}_n \tag{37}$$

These conditions are satisfied only when δn, $\delta p \to 0$. However, as it can be seen from Eqs. (12) and (29), δn, $\delta p \to 0$ only when $\beta \to 0$. From physical point of view it would mean that the charge carrier concentrations do not depend on local temperature $T(x)$. However, this situation will never occur.

Therefore, the thermoelectric cooling described by Eqs.(31), (35) and (36) in the conventional theory is not correct.

Finally, let us emphasize that one of the main assumptions in this work is that the lateral surfaces of the $p-n$ structure are thermally isolated (adiabatic isolation, adiabatic Peltier effect [2]). In this situation the $p-n$ structure, which is being investigated, does not have energy interaction with the surroundings, thus the Peltier effect shows itself "clearly".

However, in practice, there is another situation of great interest, namely the ideal thermal interaction of the *p-n* structure with the surroundings (isothermal Peltier effect [1, 9]). It is clear that $\nabla T = 0$ inside of the structure under these conditions. In this case, the problem of thermoelectric cooling is reduced to the calculation of the amount of heat, absorbed (released) by the *p-n* structure from (to) the surroundings, in order to satisfy the condition imposed by $\nabla T = 0$. From our point of view, such problem formulation is too artificial.

Finding the heat flux in the conventional theory is easy, from Eqs. (35) – (36), and imposing $\nabla T = 0$, we directly have

$$q_{ext} = (\Pi^n{}_n - \Pi^p{}_p) j_0 \tag{38}$$

In the model presented in this article, the calculation of q_{ext} demands the solution of the system of Eqs. (8), (12), and (31) with $\nabla T = 0$ and the corresponding boundary conditions.

For the isothermal Peltier effect the condition $\tau \to 0$ (strong recombination) assures the disappearance of the nonequilibrium charge carrier concentrations $\delta n = \delta p = 0$ and the conversion to conventional theory.

Conclusions

It was shown that the theory of thermoelectric cooling (adiabatic Peltier effect) can not be created without taking into account the existence of nonequilibrium electron and hole concentrations. The presented recombination of the nonequilibrium carrier always depends on the temperature heterogeneity connected with the thermoelectric cooling. Therefore, the nonequilibrium carrier concentrations do not disappear even at very short life times. The simplifications related to the quasineutrality approximation for both weak and strong recombination were analyzed. In the present work it is demonstrated that the Peltier effect strongly depends on the recombination rate. In particular, it is shown that the sign of the Peltier effect changes with the value of the recombination rate.

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