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# Addition to the Theory of Liquid Phase Epitaxial Growth I. Distribution of Components in the Liquid Phase and Their Behavior During Growth

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#### Abstract

The subject of the paper is an addition to the theory of liquid phase epitaxial growth. The established theory up till now has treated only the behavior of the components with a positive concentration gradient during liquid phase growth. In the paper it is shown that a part of the components are distributed in the liquid phase with a negative concentration gradient. For these components the liquid phase is always superheated. Despite this, these components participate in the growth process. A possible mechanism for explanation of the participation of these components in the growth process is given. The behavior of the components during growth is discussed. Special attention has been paid to processes carried out within the phase boundary region. The balance equation concerning the sum of the relative changes during growth of the masses and of the concentrations of the components in the liquid phase is derived.

*Key words:* Liquid phase epitaxial growth, distribution of components in the liquid phase, behavior during growth, phase boundary region, balance equation

### 1. Introduction

Despite the significant achievements of gas phase epitaxy methods (MBE, MOCVD, ALE and so on) in the growth of multilayer structures with nanoscale thickness of layers [1-25, 72], the liquid phase epitaxy (LPE) retains its significance for the production of different kinds of devices. During the last few years a considerable progress of LPE methods [59-65, 68-71, 79, 82, 84, 89] has been observed. By Low Temperature LPE (LTLPE) it is now quite possible to grow, with sufficient reproducibility, epilayers with nanoscale thickness (of about 100 Å or less). Taking into account the good electrophysical properties of the LPE layers and the cheapness of the technological equipment, one may conclude that the significance of liquid phase epitaxy in the future for the production of optoelectronics and UHF devices will increase.

The main difference between the various LPE methods is the way supersaturation above the crystallization boundary is created. In the zone melting version, for example, supersaturation arises due to the temperature gradient between the seed crystal and the substrate. In the epitaxy method used for the growth of  $A_3B_5$  and  $A_2B_6$  semiconductors, the supersaturation above the crystallization boundary is created through the cooling of the whole system with a constant cooling rate  $\nu_T = dT/dt$ . In this work the latter version of the LPE will be closely considered, but the conclusions made are valid, with some corrections, for the former as well.

The main aim of the present work is to give a possible explanation of some theoretical problems concerning the LPE method and to present some new correlations of the LPE process, thus opening new possibilities for its improvement.

### 2. Grounds and aims of investigation

The theory and practice of the LPE process was established at the end of the 60s and at the beginning of the 70s [26-59]. Up until now the theoretical concepts for the LPE growth established in this period have undergone very small development [66, 67, 72-76, 80-91] and in general are as follows:

- the crystallization growth rate is limited by the diffusion process in the liquid phase volume of the dilute components the concentration of these components increases always with increasing distance from the phase boundary  $(dC_i^L/dx > 0)$
- the case of one dimensional diffusion is considered
- the mass transfer toward the crystal surface (Fig. 1) creates supersaturation with respect to these components and causes their participation in the growth process.

The solution of the diffusion equation at given boundary conditions (depending on the LPE technique used) gives as a result an expression for the crystal growth rate often confirmed by experiments.

A more detailed investigation of the processes occuring in the liquid phase shows that by this theoretical model a full, thorough and exhaustive explanation of liquid phase epitaxial growth can not be given. Moreover, the crystallization mechanisms (the normal mechanism of growth, the growth by screw dislocations, the growth by two dimensional nucleus) are not discussed in this model and their determination seems to be quite impossible (the crystallization mechanisms of growth and the different techniques of liquid phase epitaxial growth will be considered in a future work).

The molar concentration of an arbitrary i - component in the liquid phase is given by the well known expression:

$$C_i^L = \frac{m_i^L/M_i}{\sum_{j=1}^n m_j^L/M_j}; \qquad \sum_{j=1}^n C_j^L = 1$$
(1)

where  $m_i^L$  is the mass of this component,  $M_i$  is its molar weight and n is the number of the components. This expression is valid: 1) at any moment (t) of the growth process; 2) for an arbitrary value of the co-ordinate (x); 3) for the actual values of the concentrations of the components as well as for their equilibrium values.

Hence, it follows that:

1) For two adjacent points (x) and (x + dx) from the volume of the liquid phase and for an arbitrary moment (t) equation (1) leads inevitably to the conclusion that the sum over all components of the concentration gradients is always zero:

$$\sum_{j=1}^{n} \frac{dC_j^L}{dx} = 0 \tag{2}$$

2) For an arbitrary point of the volume of the liquid phase (x) and for two consecutive moments (t) and (t + dt) equation (1) gives that the sum over all components of the rates of change with time of the concentrations in the liquid phase is also always zero:

$$\sum_{j=1}^{n} \frac{dC_j^L}{dt} = 0 \tag{3}$$

3) Equation (1) can be written for an arbitrary moment (t) and for an arbitrary point from the volume of the liquid phase (x). It can be written once for the equilibrium values of the concentrations of the components  $(C_{i,eq}^L)$  and then for their actual concentrations  $(C_i^L)$ . The difference between these expressions gives a sum over all components of the system - its terms are the deviation of the system from the equilibrium state with respect to all components  $(\Delta C_i^L = C_i^L - C_{i,eq}^L, i \in (1, n))$ . This sum must also always be equal to zero:

$$\sum_{j=1}^{n} (C_j^L - C_{j,eq}^L) = \sum_{j=1}^{n} (\Delta C_j^L) = 0$$
(4)

In the case of only negative terms or in the case of only positive terms a sum can not be equal to zero.



Figure 1. The case of the components distribution with a positive concentration gradient.

Figure 2. The case of the components distribution with a negative concentration gradient.

Equations (2, 3, 4) lead inevitably to the conclusions:

• evaluating equation (2), one may conclude that in the liquid phase the distribution of part of the components is positive  $(dC_i^L/dx > 0, \text{Fig.1})$ , while all other components

are negative  $(dC_i^L/dx < 0, \text{Fig.2}).$ 

The established theory [28, 34-38, 52, 53, 56, 72, 74] of liquid phase epitaxy treats only the components with a positive concentration gradient. This theory can not explain the participation in the growth process of components with the negative concentration gradient - the mass transfer of these components occurs not to the phase boundary region but in the opposite direction - deep into the volume of the liquid phase. Such mass transfer does not lead to the creation of a supersaturation with respect to these components above the crystallization boundary.

The latter components are for example Ga in the LPE process of GaAs and  $Al_xGa_{1-x}As$ , In in the InP LPE process and so on.

- evaluating equation (3), one may conclude that during growth for some components the concentration increases with time  $(dC_i^L/dt > 0)$  and for the rest of them the concentration decreases with time  $(dC_i^L/dt < 0)$
- evaluating equation (4), one may conclude that part of the components participates in the growth process having positive deviation values from the equilibrium ( $\Delta C_i^L > 0$ ) - with respect to these components the liquid phase is supersaturated. This is the classical case discussed in many works. All other components participating in the growth process are with negative values of deviation from equilibrium ( $\Delta C_i^L < 0$ ) with respect to them the liquid phase is not supersaturated, it is rather superheated. The last conclusion contradicts the basics of crystal growth theory.

Upon considering a multicomponent system, the following problems arise:

- 1) Which components during growth are distributed in the liquid phase with positive concentration gradients and which components are distributed with negative ones ?
- 2) For which components does the concentration in the liquid phase during growth increase with time and for which components does it decrease ?
- 3) Which components participate in the growth process with positive values of the deviation from equilibrium and which components participate in the growth with negative values ?
- 4) In which way do the components with a superheated liquid phase participate in the growth process ?

### 3. Assumptions

The case of the two phase system is considered: the n - component solid phase is in contact with the n - component liquid phase from which the layer grows. The following requirements are valid for this system:

- the crystallization process is carried out only on the substrate in the volume of the liquid phase no nucleation takes place;
- the system is isothermal no temperature gradients are available;
- no chemical reactions between the components are observed in the liquid phase;

- no evaporation of the liquid phase takes place.

These are well known assuptions used in many investigations [28, 34-38, 52, 53, 56, 72].

The system defined in this way is practically an isolated system - the mass of the components is constant:  $m_i = m_i^L + m_i^S = const.$ ,  $dm_i^S = -dm_i^L = |dm_i^L|$  where  $m_i^L$  and  $m_i^S$  are the masses of *i* - component in the liquid and in the solid phase respectively and  $dm_i^L$  and  $dm_i^S$  are their changes within the time interval dt caused by growth. During crystallization, the mass  $m_i^S$  always increases with time and the mass  $m_i^L$  always decreases.

The crystallization process is considered in a coordinate system with an Ox axis normal to the crystal surface and the coordinates origin (point O) lies always on the crystal surface. The growth process performed on an oriented substrate is always one-dimensional - along the axes Oy and Oz no concentration gradients are observed and, obviously, no mass transfer is possible.

### 4. Distribution of the components during growth

The well known diffusion equation is considered:

$$D_i \frac{d^2 C_i^L}{dx^2} = \frac{dC_i^L}{dt} \tag{5}$$

For a given concentration  $C_i^L(x,t)$  and by double differentiation with respect to (x), it simply follows that (Fig. 1 and Fig. 2) [75, 77]:

- if the concentration of a given component increases with time  $(dC_i^L/dt > 0)$ , it decreases with the coordinate  $(dC_i^L/dx < 0)$
- if the concentration decreases with time  $(dC_i^L/dt < 0)$ , it increases with the coordinate  $(dC_i^L/dx > 0)$ .

The crystal growth process will now be closely considered. The grown n - component solid phase will be grown from an n - component liquid phase. In the classical case of liquid phase epitaxy, the system is cooled with a constant cooling rate of  $(\nu_T)$  - as a consequence it creates supersaturation for a part of the components and crystallization takes place. For the time (dt) the masses of the (i) - component in the solid phase and in the liquid phase change by the values of  $dm_i^L$  and  $dm_i^S$  respectively, i.e.  $dm_i^S = -dm_i^L$ . During growth the following relations are valid [75-78]:

$$C_{i}^{S} = \frac{dm_{i}^{L}/M_{i}}{\sum_{j=1}^{n} dm_{j}^{L}/M_{j}} = \frac{\dot{m}_{i}^{L}/M_{i}}{\sum_{j=1}^{n} \dot{m}_{j}^{L}/M_{j}}$$
(6)

$$\frac{\dot{C}_i^L}{C_i^L} = \left(\frac{C_i^L}{C_i^S} - 1\right) \frac{\dot{m}_i^L}{m_i^L} \tag{7}$$

where  $\dot{C}_i^L = dC_i^L/dt$ , and  $\dot{m}_i^L = dm_i^L/dt$ .

(Deducing equation (7) it was taken into account that during growth the mass of the components in the liquid phase  $m_i^L$  always decreases with time:  $\dot{m}_i^L = dm_i^l/dt < 0$ . The system will be considered at two consecutive moments of time - the "initial"-moment (t) and the "end"-moment (t + dt). At the initial moment, the system is characterized

by the masses  $m_k^L(t)$ ,  $k \in (1, n)$  and by the temperature T(t) and at the end moment these quantities have the values  $m_k^L(t + dt) = m_k^L(t) - dm_k^L$  and  $T(t + dt) = T(t) - dT = T(t) - \nu_T dt$ , respectively. In these expressions the minus sign (-) already takes into account that the masses in the liquid phase and the temperature always decrease with time during growth and, hence, in all further considerations the quantities  $dm_k^L$ , dT and  $\dot{m}_k^L = dm_k^L/dt$ ,  $\nu_T = dT/dt$  are taken positive, i.e. they participate with their absolute values.)

From equation (7) it follows:

- A) components for which  $C_i^S > C_i^L$  are characterized by a positive concentration gradient  $(dC_i^L/dx > 0)$  and during growth their concentration will decrease with time  $(dC_i^L/dt < 0)$  these components are further denoted by the index "i" and, for simplicity, they will be called "i-components".
- B) components for which  $C_i^S < C_i^L$  are characterized by a negative concentration gradient  $(dC_i^L/dx < 0)$  and during growth their concentration increases with time  $(dC_i^L/dt > 0)$  these components will be further denoted by the index "j" and, for simplicity, they will be called "j-components".
- C) if  $C_i^S = C_i^L$  the concentration does not change with time during growth  $(dC_i^L/dt = 0)$ - this is the case when the growth is performed at the melting point of the compound from the stoichiometric composition of the melt.



In Figure 3 the dependence  $\dot{C}_k^L(C_k^L)$ ,  $k \in (1, n)$  from equation (7) is shown. It is obvious that  $\dot{C}_k^L \to 0$ , when  $C_k^L \to C_k^S$  and when  $C_k^L \to 0$ . Within the interval  $C_k^L \in (0, C_k^S)$ , the quantity  $\dot{C}_k^L$  is always negative  $(\dot{C}_k^L < 0)$  and there is a minimum of  $\dot{C}_k^L$  - this is the region of the "i"-components. If the changes of the ratio  $(\dot{m}_k^L/m_k^L)$  are small  $(\dot{m}_k^L/m_k^L \cong const.)$ , then the minimum will be at the value  $C_k^L = C_k^S/2$ . Significant change of the ratio  $\dot{m}_k^L/m_k^L$  will lead to displacement of the minimum along the Ox-axis. In the region  $C_k^L > C_k^S$ , the

In Figure 3 the dependence

**Figure 3.** The dependence  $\dot{C}_r^L(C_r^L)$  in accordance with equation (7).

quantity  $\dot{C}_k^L$  is always positive and this is the region of the "j"-components.

During growth, the concentration of the *i*-components  $(C_i^S < C_i^L)$  will continuously decrease  $(\dot{C}_i^L < 0)$  moving away from the point  $C_i^S = C_i^L$ . This means that an arbitrary *i*-component will never be a *j*-component. The same is valid also for the *j*-components  $(C_j^S < C_j^L)$  - during growth their concentrations continuously increase  $(\dot{C}_j^L > 0)$  moving away from the value  $C_j^S = C_j^L$  - therefore a *j*-component will also never be an *i*-component. One may conclude that:

- a component initially (at the beginning of growth) being an *i*-component remains an *i*-component during the whole growth process and - a component being initially a *j*-component remains a *j*-component.

Hence, the growth process does not change the character of the components.

### 5. Behavior of the components during growth

Let for time (dt), due to temperature decrease by dT ( $dT = \nu_T dt$ ), the masses  $dm_k^L$  and  $dm_r^L$  of two arbitrary k- and r-components, respectively, be deposited on the substrate building up the epitaxial layer. The molar portions of these components in the grown epilayer will be  $(dm_k^L/M_k)$  and  $(dm_r^L/M_r)$ , respectively. Then the following relation is obvious:

$$\frac{(dm_k^L/M_k)}{(dm_r^L/M_r)} = \frac{g_k}{g_r} \tag{8}$$

where  $g_k$  and  $g_r$  are the indices of the k and r components, respectively, in the stoichiometrical formula of the grown compound -  $g_k$  and  $g_r$  are assumed to be constants  $(g_r = const., r \in (1, n)).$ 

Because k and r are arbitrary components, the masses of all other components could be expressed by the mass of one of them using equation (8). For example, expressed through the mass of k-component (the basic component), the masses of all other components incorporated into the epilayer are:

$$dm_r^L = \frac{g_r M_r}{g_k M_k} dm_k^L ; \qquad r \in (1, n)$$
(8a)

and therefore by summation over all masses  $dm_r^L$ ,  $r \in (1, n)$ , one obtains the mass of the grown epitaxial layer  $dm_o$  and the growth rate  $\nu_o$  of the compound:

$$dm_{o} = \sum_{r=1}^{n} dm_{r}^{L} = \frac{dm_{k}^{L}}{M_{k}g_{k}} \sum_{r=1}^{n} M_{r}g_{r} = \frac{M_{o}}{M_{k}g_{k}} dm_{k}^{L};$$

$$\nu_{o} = \frac{1}{s_{o}\gamma_{o}^{S}} \frac{dm_{o}^{L}}{dt} = \frac{1}{s_{o}\gamma_{o}^{S}} \sum_{r=1}^{n} \frac{dm_{r}^{L}}{dt} = \frac{M_{o}}{s_{o}\gamma_{o}^{S}M_{k}g_{k}} \frac{dm_{k}^{L}}{dt} = \frac{1}{s_{o}\gamma_{k}^{S}} \frac{dm_{k}^{L}}{dt} = \nu_{k}$$
(9)

 $\gamma_o^S$  is the density of the grown compound,  $s_o$  is the area of the grown layer,  $M_o = \sum_{r=1}^n g_r M_r$  is the molar weight of the grown compound.  $\gamma_k^S = (M_k g_k/M_o) \gamma_o^S$  is the density of the k-component in the solid phase - hence,  $\gamma_o^S$  is an additive quantity:  $\gamma_o^S = \sum_{k=1}^n \gamma_k^S$ .

Quantity  $\nu_k$  could be considered as the growth rate of the k-component - this is the rate of the deposition of k-particles onto the substrate and of its insertion into the grown epitaxial layer. In this sense equation (9) postulates that the different components always have the same growth rate, equal to the growth rate of the compound itself.

As mentioned above, the components with a positive concentration gradient  $(dC_i^L/dx > 0)$  (the *i*-component) are always deposited from a supersaturated liquid phase  $(\Delta C_i^L > 0)$  due to the mass transport toward the phase boundary.

With respect to the components with a negative concentration gradient  $(dC_j^L/dx < 0)$ (the *j*-component), the liquid phase is always superheated  $(\Delta C_j^L < 0)$ . This is in good agreement with the fact that their diffusion mass transport is directed deep into the volume of the liquid phase and, in any case, no supersaturation for these components is possible above the phase boundary. The same conclusion could be made considering the supercooling  $(\Delta T = T_{eq} - T > 0)$ and from the deviation of the concentration from its equilibrium value  $(\Delta C_r^L = C_r^L - C_{r.eq}^L)$ , i.e.  $\Delta C_r^L = \Delta T/S_r$  ( $S_r = dT/dC_{r.eq}^L$  is the slope of the liquid surface with respect to the *r*-component,  $r \in (1, n)$ ). Taking into consideration that during growth the supercooling  $(\Delta T)$  is always positive, one may conclude that the quantities  $(\Delta C_r^L)$  and  $(S_r)$  must have the same sign. From the liquidus surface it follows that:

- for the *j*-components  $(C_j^L > C_j^S, dC_j^L/dt > 0, dC_j^L/dx < 0)$ , the slope of the liquidus surface is always negative  $S_j = dT/dC_{j.eq}^L < 0$  and, therefore, the deviation of their concentrations from the equilibrium value is also always negative  $\Delta C_j^L < 0$ , i.e. during growth the liquid phase is always superheated with respect to these components,
- for the *i*-components  $(C_i^L < C_i^S, dC_i^L/dt < 0, dC_i^L/dx > 0)$ , the slope of the liquidus surface is always positive  $S_i = dT/dC_{i.eq}^L > 0$  and, therefore, the deviation of their concentrations from the equilibrium value is always positive too  $\Delta C_i^L > 0$ , i.e. during growth, the liquid phase is always supercooled with respect to these components.

An additional confirmation of the last conclusions is given in the discussion of equation (31).

# 5.1. Non diffusive mass transport of j - components toward the phase boundary

In the case of equilibrium cooling technique close to the phase boundary, the deviation from equilibrium of the liquid phase is very small and, therefore, the actual concentration of the components in the liquid phase could be assumed to be equal to the equilibrium one.



Figure 4. The real situation in the liquid phase in the case of equilibrium cooling technique for i- and for j-components.

In the phase boundary region, the *i*-components will have the minimum value of their concentration in the liquid phase (Figure 4, see [77]) - beginning from the phase boundary, their concentration always increases deep into the liquid phase volume.

In the same region the j - components concentration value will be the maximum one - beginning from this point, their concentration decreases into the liquid phase volume.

Due to the flux  $J_i^L$ , a supersaturation with respect to the *i*-components arises in the phase boundary region - as a consequence, the adsorption of

the consequence, the adsorption of the *i*-particles onto the crystal surface occurs more intensively. Consequently, their concentration decreases in the  $\delta$ -region (the region situated in the liquid phase volume close to the crystal surface - the interface region). The decrease of the concentration  $C_{iI}^L$  (the value of the concentration within the interface region close to the phase boundary [77]) leads inevitably to the increase of the concentration of the *j*-components  $C_{jI}^L$ . The latter concentration becomes greater than its equilibrium value. Hence, in the  $\delta$ -region supersaturation with respect to the *j*-components also arises - the remaining volume of the liquid phase remains superheated [77].

The flux  $J_{jI}^L$  (the value of the flux within the interface region) of the *j*-particles toward the phase boundary (in the physical meaning of the word "flux" as a "directed movement of particles") exists only as a consequence of the supersaturation in the  $\delta$ -region. This is not a diffusion flux because it is not caused by the concentration gradient. It is caused by the created supersaturation and leads to the restoration of the equilibrium state in the phase boundary region.

# 5.2. Epitaxial growth rate

In equation (9) we have concluded that the rate of the insertion of an arbitrary kcomponent into the epitaxial layer is equal to the growth rate of the compound itself. We
will now consider the distribution during growth of an arbitrary *i*-component in the liquid
phase in two consecutive moments (t) and (t + dt) (Fig. 5). At the initial moment the
number of the *i*-particles in the liquid phase in a column above a unit area of the phase
boundary will be:

$$N_i^L(t) = \int_0^{Z_0} n_i^L(x,t) \, dx \tag{10}$$

where  $n_i^L(x,t)$  is the distribution of the *i*-component given in [at/cm<sup>3</sup>],  $Z_0$  is the thickness of the liquid phase above the substrate. As a consequence of cooling (with a rate of  $\nu_T$ ) within the time interval  $\Delta t$ , an epitaxial layer with thickness of  $\Delta H$  is deposited onto the substrate. The *i*-particles are distributed there, with a concentration of  $n_i^S = const.$  $(dn_i^S/dx = 0).$ 

The *i*-particles of quantity  $N_i^L(t)$  (according to equation (10)) will be distributed at the moment (t+dt) between the grown epitaxial layer (their number there will be  $n_i^S \Delta H$ ) and the liquid phase. They will have a distribution in the liquid phase given by the function  $n_i^L(x, t + \Delta t)$ . Consequently, the following expressions are valid:

$$N_{i}^{L}(t) = \int_{0}^{Z_{0}} n_{i}^{L}(x,t) \, dx = n_{i}^{S} \Delta H + \int_{\Delta H}^{Z_{0}} n_{i}^{L}(x,t+\Delta t) \, dx$$
$$n_{i}^{S} \Delta H = \int_{0}^{\Delta H} n_{i}^{L}(x,t) \, dx + \int_{\Delta H}^{Z_{0}} [n_{i}^{L}(x,t) - n_{i}^{L}(x,t+\Delta t)] \, dx \tag{11}$$

The following relations are taken into account:

$$n_{i}^{L}(x,t+\Delta t) = n_{i}^{L}(x,t) + \frac{dn_{i}^{L}}{dt}\Delta t = n_{i}^{L}(x,t) + \dot{n}_{i}^{L}(x,t)\Delta t$$
$$\dot{n}_{i}^{L}(x,t) = \frac{dn_{i}^{L}}{dt} = D_{i}\frac{d^{2}n_{i}^{L}}{dx^{2}} = -\frac{dJ_{i}^{L}}{dx}$$
(12)

$$n_{i}^{L}(x,t) = n_{i.eqo}^{L} + n_{i}^{\prime L}(x,t) \ x = n_{i.eqo}^{L} + \frac{\Delta n_{i1}^{L}}{\Delta H} x$$

where  $n_i^L(x,t) = dn_i^L/dx$ ,  $\Delta n_{i1}^L = n_i^L(\Delta H,t) - n_{i.eqo}^L$ ,  $n_{i.eqo}^L$  is the equilibrium value of the concentration of the *i*-component at the phase boundary (for x = 0).

Let us now consider the case of equilibrium liquid phase epitaxy - the concentration at the phase boundary is always equal to the equilibrium one. We have assumed the thickness  $\Delta H$  of the epitaxial layer to be very small and, therefore, the interval of the first integral  $(0, \Delta H)$  is very small too - within a very small interval the integral function could be approximated by a linear one.

$$n_i^S \Delta H = \left( n_{i.eqo}^L + \frac{\Delta n_{i1}^L}{2} \right) \Delta H - \int_{\Delta H}^{Z_0} \frac{dn_i^L}{dt} dx \Delta t =$$
$$= n_i^S \Delta H = \left( n_{i.eqo}^L + \frac{\Delta n_{i1}^L}{2} \right) \Delta H + \left[ J_i^L(Z_0, t) - J_i^L(\Delta H, t) \right] \Delta t \tag{13}$$

Taking into account that the vector of the growth velocity  $\nu_0 = \Delta H / \Delta t$  is directed in the opposite direction to the vector of the diffusion flux  $J_i^L = -D_i dn_i^L / dx$ , one obtains the following relation for the epitaxial growth rate within the interval  $(t, t + \Delta t)$ :

$$\nu_0 = \frac{\Delta H}{\Delta t} = \frac{J_i^L(\Delta H, t) - J_i^L(Z_0, t)}{n_i^S - n_{i.eqo}^L - \frac{\Delta n_{i1}^L}{2}}$$
(14)

Relation (14) could be simplified, taking into account that the change of the concentration  $\Delta n_{i1}^L$  is usually negligible with respect to the quantities  $n_i^S$  and  $n_{i.eqo}^L$ :

$$\nu_0 = \frac{\Delta H}{\Delta t} = \frac{J_i^L(\Delta H, t) - J_i^L(Z_0, t)}{n_i^S - n_{i.eqo}^L}$$
(15)

Using the same considerations, one may obtain for an arbitrary j-component (Fig. 6):

$$\nu_0 = \frac{\Delta H}{\Delta t} = -\frac{J_j^L(\Delta H, t) - J_j^L(Z_0, t)}{n_j^S - n_{j.eqo}^L} = \frac{J_j^L(\Delta H, t) - J_j^L(Z_0, t)}{n_{j.eqo}^L - n_j^S}$$
(15a)



Figure 5. Behavior of *i*-components during growth.

Figure 6. Behavior of *j*-components during growth.

It is obvious that the epitaxial growth rate depends in a simple way on the parameters of an arbitrary component. Using the distribution of some component in the liquid phase, one may determine each quantity of the last equations and, hence, the epitaxial growth rate.

### 6. Balance equation

Equation (7) shows the significant role of the ratio of the concentration of an arbitrary component in the liquid phase  $(C_k^L)$  to its concentration in the solid phase  $(C_k^S)$  for the behavior of the component during growth. Using equations (1) and (6), this ratio leads to the expression [78]:

$$\frac{C_k^L}{C_k^S}\frac{\dot{m}_k^L}{m_k^L} = \frac{\sum}{\sum}$$
(16)

where  $\sum_{k=1}^{n} = \sum_{k=1}^{n} \left( \frac{\dot{m}_{k}^{L}}{M_{k}} \right)$  and  $\sum_{k=1}^{n} \left( \frac{m_{k}^{L}}{M_{k}} \right)$ . Substituting equation (9) in equation (16) one obtains:

$$\frac{C_k^L}{C_k^S} \frac{\gamma_k^S}{m_k^L} = \frac{\sum}{\nu_0 S_0 \sum} \Rightarrow \frac{C_k^L g_k M_k}{C_k^S m_k^L} = \frac{M_0 \sum}{\nu_0 S_0 \gamma_0^S \sum} \equiv f_0 \tag{17}$$

The ratio on the right side denoted by  $f_0$  is always the same for all components of the liquid phase, i.e.  $M_k$  and the ratio  $g_k/C_k^S = \sum_{r=1}^n g_r$  (taking into account the equality  $C_k^S = g_k/\sum_{r=1}^n g_r$ ) are constants.

This circumstance means that if at the moment (t) the value of the left side is  $f_0$ , then at the moment (t + dt) its value will be  $f_0 + df_0 = f_0(t + dt)$ , determined by the change of the quantities  $C_k^L$  and  $m_k^L$ . During growth, the mass of the components  $m_k^L$  decreases always but their concentrations could decrease (for the *i*-components) or increase (for the *j*-components). Therefore, the following equalities are valid:

$$f_0(t) = \frac{C_k^L(t)g_k M_k}{C_k^S m_k^L(t)};$$

$$f_0(t+dt) = f_0(t) + df_0 = \frac{C_k^L(t) \pm dC_k^L}{m_k^L(t) - dm_k^L} \frac{g_k M_k}{C_k^S}$$
(18)

As a rule the following inequalities are fulfilled:  $dC_k^L \ll C_k^L$  and  $dm_k^L \ll m_k^L$  which allows us to use the approximate formula:  $\frac{1}{1\pm\alpha} \cong 1 \mp \alpha$  (valid for very small values of  $\alpha \ll 1$ ).

### 6.1. Case of positive concentration gradient (i - components)

$$\left(\frac{dC_i^L}{dx} > 0, \ \frac{dC_i^L}{dt} < 0\right)$$

The quantity  $dC_i^L$  from equation (18) in this case is always negative and in a simple way one may obtain the relation:

$$\frac{df_0}{f_0} = \frac{dm_i^L}{m_i^L} - \frac{dC_i^L}{C_i^L}; \qquad \varphi_0(t) = \frac{1}{f_0}\frac{df_0}{dt} = \frac{\dot{m}_i^L}{m_i^L} - \frac{\dot{C}_i^L}{C_i^L}$$
(19)

### 6.2. Case of negative concentration gradient (j - components)

$$\left(\frac{dC_j^L}{dx} < 0, \ \frac{dC_j^L}{dt} > 0\right)$$

In this case the quantity  $C_j^L$  from equation (18) has a positive sign and the corresponding relation is:

$$\frac{df_0}{f_0} = \frac{dm_j^L}{m_j^L} + \frac{dC_j^L}{C_j^L}; \qquad \varphi_0(t) = \frac{1}{f_0}\frac{df_0}{dt} = \frac{\dot{m}_j^L}{m_j^L} + \frac{\dot{C}_j^L}{C_j^L}$$
(20)

By function  $p_k(C_k^L - C_k^S)$  defined in the following way:

$$p_{k} = +1 , \qquad C_{k}^{L} > C_{k}^{S}$$

$$p_{k} = 0 , \qquad C_{k}^{L} = C_{k}^{S}$$

$$p_{k} = -1 , \qquad C_{k}^{L} < C_{k}^{S}$$

$$(21)$$

both equations (19) and (20) could be united.

$$\frac{df_0}{f_0} = \frac{dm_k^L}{m_k^L} + p_k \frac{dC_k^L}{C_k^L}; \qquad \varphi_0(t) = \frac{1}{f_0} \frac{df_0}{dt} = \frac{\dot{m}_k^L}{m_k^L} + p_k \frac{\dot{C}_k^L}{C_k^L}$$
(22)

The relative changes during growth of the mass and of the concentration in the liquid phase for an arbitrary component must be balanced in such a way so that at any moment their sum (in correspondence with equation (22)) is the same for all components. Precisely in this sense, the equation (22) is a balance equation and function  $\varphi_0$  is a characteristic function of the growth process. Its values depend directly or indirectly on the mass of the given component in the liquid phase, on its concentration, on the crystallization mechanisms and so on. Hence, by the values of this function the growth process could be characterized fully enough.

For the growth process, function  $\varphi_0$  is always positive (equation (20)). From equation (19), it follows that the relative change of the mass of the *i*-components is significant because it compensates the increase of function  $\varphi_0$  as well as the relative change of the concentration  $(dC_i^L/C_i^L)$ .

For the case of liquid phase epitaxy of GaAs, the Ga-component is a j-component and the As-component is an i-one. From equations (19) and (20) it follows:

$$\varphi_0 = \frac{\dot{m}_{As}^L}{m_{As}^L} - \frac{\dot{C}_{As}^L}{C_{As}^L} = \frac{\dot{m}_{Ga}^L}{m_{Ga}^L} + \frac{\dot{C}_{Ga}^L}{C_{Ga}^L}$$
(23)

Most significant are the changes of the mass of the *i*-components during growth - the relative change of the mass of As is equal to the sum of three terms:  $(\dot{m}_{Ga}^L/m_{Ga}^L)$ ,  $(\dot{C}_{Ga}^L/C_{Ga}^L)$ ,  $(\dot{C}_{As}^L/C_{As}^L)$ .

### 7. Processes in the phase boundary region

The phase boundary region (the " $\delta$ " - region) not only separates both solid and liquid phases but during growth it is exactly that region where the formation of the grown epitaxial layer is carried out. During crystallization all processes needed for the full adaptation of the liquid phase to the substrate (with respect to the crystal structure and to the composition) are accomplished in this region.

By the influence of the potential field of the crystal substrate the liquid phase region (close to the crystal surface) is ordered to a great degree. Deep into the volume of the liquid phase (far away from the crystal surface) the structure of the matter is typically amorphous. Anisotropy, short range order and so on are observed. Within the limits of the phase boundary region (the " $\delta$ " - region), there are indications of anisotropic properties and of a long range order of the matter - these are the significant characteristics of a crystal structure. This allows us to speak about similarity to the crystal structure of the phase boundary region repeating sufficiently well the structure of the substrate itself.

Hence, with respect to the structure both solid and liquid phases are in the process of approaching each other within the limits of the phase boundary region.

With respect to the composition, however, the difference between both phases increases continuously toward the crystal surface (see the differences  $\left[C_i^S - C_i^L(x=0)\right]$ ,  $\left[C_j^L(x=0) - C_j^S\right]$  in Fig. 4) - at the phase boundary, the liquid phase is enriched significantly with respect to the *j*-components and is strongly depleted with respect to the *i*-components. In this way the diffusing toward the crystal surface *i*-particles get into the liquid phase regions which are more enriched with *j*-particles.



**Figure 7.** Proportionality between the diffusion flux  $dC_i^L/dx$  and the deficit  $[n_i^S - n_i^L(x)]$  for the case of *i*-components.

Therefore, within the  $\delta$  - region there is a significant excess of jparticles corresponding to the difference  $\left[n_j^L(x) - n_j^S\right]$  and, simultaneously, there is a significant deficit of *i*-particles corresponding to the difference  $\left|n_{i}^{S}-n_{i}^{L}(x)\right|$ (Figures 7 and 8). The i-particles corresponding to the flux  $J_i^L =$  $D_i^L dn_i^L / dx$  are incoming continuously into the interface region by diffusion mass transport and simultaneously j-particles are leaving this region also by diffusion mass transport corresponding to the flux  $J_i^L = D_i^L dn_i^L / dx$ .

In order to grow a perfect epitaxial layer (without second phase inclusions and with an exact stoichiometry corresponding to the substrate one), a balance is required to exist between:

- the incoming into the  $\delta$  region by diffusion *i*-particles  $(J_i^L)$  and the free positions for these particles available there (given by the concentration difference  $\left[n_i^S n_i^L(x)\right]$ ) on one side and
- the *j*-particles leaving by diffusion  $(J_j^L)$  the  $\delta$  region and their excess amount there (given by the concentration difference  $\left[n_j^L(x) n_j^S\right]$ ) on the other side.



**Figure 8.** Proportionality between the diffusion flux  $dC_j^L/dx$  and the excess  $[n_j^L(x) - n_j^S]$  for the case of *j*-components.

In Figure 7 the shaded region illustrates the amount of missing *i*-particles in the interface region  $\left[n_i^S - n_i^L(x)\right]$ . In order to form an epitaxial layer with the amount of *i*-components coinciding with that in the substrate the missing amount must be "delivered" into the interface region by diffusion mass transport.

The shaded region in Figure 8 illustrates the excess amount of j-particles in the interface region  $\left[n_j^L(x) - n_j^S\right]$ . For the same reason, this amount must be "removed" from the interface region also by diffusion mass transport.

Hence, a balance is required between the diffusion fluxes of the *i*- and *j*-particles  $D_i^L dn_i^L/dx$  and  $D_j^L dn_j^L/dx$ , on one side, and their corresponding concentration differences  $\left[n_i^S - n_i^L(x)\right]$  and  $\left[n_j^L(x) - n_j^S\right]$ , on the other side. One may postulate:

$$D_{i}^{L}\frac{dn_{i}^{L}}{dx} = K_{i}\left[n_{i}^{S} - n_{i}^{L}(x)\right] ; \qquad -D_{j}^{L}\frac{dn_{j}^{L}}{dx} = K_{j}\left[n_{j}^{L}(x) - n_{j}^{S}\right]$$
(24)

where  $K_i$  and  $K_j$  are coefficients.

The solutions of equations (24) must fulfil the above mentioned requirements: for the *i*-components the concentration gradient must always be positive  $dn_i^L/dx > 0$  and for the *j*-components it must always be negative  $dn_j^L/dx < 0$ . The following expressions satisfy these requirements:

$$n_{i}^{L}(x) = n_{i}^{S} - A_{0i} \exp\left(-\frac{K_{i}}{D_{i}}x\right) ; \qquad n_{j}^{L}(x) = n_{j}^{S} + A_{0j} \exp\left(-\frac{K_{j}}{D_{j}}x\right)$$
(25)

These equations are valid within the interface region (for the values of  $x \in [0, \delta]$ ). For the case of an equilibrium liquid phase epitaxy when the actual concentration at the phase boundary (x = 0) is always equal to the equilibrium one, i.e  $n_i^L(x = 0) = n_{i.eq}^L$  and  $n_j^L(x = 0) = n_{j.eq}^L$ , one obtains the following values for the coefficients  $A_{0i}$  and  $A_{0j}$ :

$$A_{0i} = n_i^S - n_{i.eq}^L(T) ; \qquad A_{0j} = n_{j.eq}^L(T) - n_j^S$$
(26)

The maintenance of the balance between the i- and j- diffusion fluxes, on one side, and the corresponding concentration differences, on the other side, is necessary for growing a perfect epitaxial layer repeating the crystal structure of the substrate as well as its composition. The violation of this balance may lead to significant deviations from the stoichiometric composition of the grown layer as well as to the formation of second phase inclusions.

Hence, for the growth of a perfect epitaxial layer without any deviations from the stoichiometry and without any second phase inclusions, it is necessary to provide such growth conditions so that the incoming by diffusion into the interface region i-particles push out completely the excess j-particles. However, this is a process depending on many factors and, consequently, it could be easily disturbed and hindered.

Now the question arises how deep is the volume of the liquid phase where the processes described above are carried out - in other words the question arises about the dimensions (the depth) of the  $\delta$  - region.

The epitaxial growth is, in fact, an interaction process between two phases - it consists of a separation of a given amount of the liquid phase which builds up the epitaxial layer onto the substrate. The mass separation is inevitably accompanied with an energy exchange corresponding to the activation energy of the phase transition.

In reality, the direct interaction between both phases takes place only within the limits of the interface region. The other volume of the liquid phase (situated far away from the phase boundary) interacts with the substrate indirectly by diffusion and, consequently, with some delay. This delay depends on the distance to the phase boundary and on the velocity of the diffusion mass transport.

The crystal growth process is carried out chain-like. The decrease of the temperature of the system causes a reaction of the deposition of the epitaxial layer onto the substrate. The portions of the ambient are deposited directly from the phase boundary region with depth  $\delta$ . As mentioned above, the liquid phase far away from the crystal surface responds to the given influence with some delay caused by the diffusion.

Using this, one can determine approximately the depth  $\delta$  of the interface region. Considering the epitaxial growth process within a very small time interval  $(\Delta t \to 0)$ , the deposited epitaxial layer of thickness  $\Delta H$  will be formed from a very thin liquid phase region close to the substrate surface. The thickness of this region tends to the thickness of the interface region  $\delta$  for very small values of  $\Delta t$  and  $\Delta H : \Delta t \to 0, \Delta H \to 0$ .

In the rest of the liquid phase volume (for  $x \ge \delta$ ), the distribution of the components remains the same. During the considered time interval  $\Delta t$ , due to the chain-like character of the crystal growth process, the deep regions of the liquid phase, far away from the crystal surface, have no time to follow the deposition of the last portion of epitaxial material.



Figure 9. The situation at the beginning of liquid phase epitaxial growth in the case of *i*-component.

Without sufficient accuracy, the problem could be solved for an arbitrary moment of time. However, for the initial moment of growth  $t \in$  $(0, \Delta t)$ , the accuracy of the solution could be significantly better such consideration will give the initial value of depth  $\delta_0$ . The approach applied above for determination of the growth rate (equations (14) and (15)) will be used.

Fig. 9 shows the situation at the initial moment - the solid and liquid phases are in equilibrium (equilibrium growth supposed). Following the same procedure, one can write the following equation analogous to equation (11):

$$N_{i}^{L}(t=0) = \int_{0}^{Z_{0}} n_{i}^{L}(x,t=0)dx = n_{i}^{S}\Delta H + \int_{\Delta H}^{Z_{0}} n_{i}^{L}(x,t=\Delta t)dx$$
(27)  
$$n_{i}^{S}\Delta H = \int_{0}^{\delta_{0}} n_{i}^{L}(x,t=0)dx - \int_{\Delta H}^{\delta_{0}} n_{i}^{L}(x,t=\Delta t)dx$$

For  $x \ge \delta_0$  both integral functions coincide:  $n_i^L(x, t = 0) \equiv n_i^L(x, t = \Delta t)$  and it is quite sufficient to integrate within the limits  $x \in (\Delta H, \delta_0)$ . The very small time  $(\Delta t \to 0)$ means that both intervals  $x \in (0, \delta_0)$  and  $x \in (\Delta H, \delta_0)$  are also very small. Hence, the function  $n_i^L(x, t = \Delta t)$  within these intervals may be approximated by a linear one.

$$n_i^L(x,t=0) = n_{i.eqo}^L$$

$$n_{i}^{L}(x,t=\Delta t) = n_{i}^{L}(x=\Delta H,t=\Delta t) + \frac{\partial n_{i}^{L}}{\partial x}x = n_{i.eq}^{L} + \frac{\Delta n_{i}^{L}(x,\Delta t)}{\delta_{0} - \Delta H}x =$$
$$= n_{i.eq}^{L} + \frac{n_{i}^{L}(x=\delta_{0},\Delta t) - n_{i}^{L}(x=\Delta H,\Delta t)}{\delta_{0} - \Delta H}x = n_{i.eq}^{L} + \frac{n_{i.eq}^{L} - n_{i.eq}^{L}}{\delta_{0} - \Delta H}x$$
(28)

Substituting into equation (27), by some transformations, one obtains:

$$n_{i}^{S}\Delta H = n_{i.eqo}^{L}\delta_{0} - n_{i.eq}^{L}(\delta_{0} - \Delta H) - \frac{n_{i.eqo}^{L} - n_{i.eq}^{L}}{\delta_{0} - \Delta H} \frac{x^{2}}{2} \Big|_{\Delta H}^{\delta_{0}} = n_{i.eqo}^{L}\delta_{0} - n_{i.eq}^{L}(\delta_{0} - \Delta H) - \frac{n_{i.eqo}^{L} - n_{i.eq}^{L}}{2}(\delta_{0} + \Delta H); \qquad (29)$$
$$\delta_{0} = \frac{2n_{i}^{S} - 3n_{i.eq}^{L} + n_{i.eqo}^{L}}{n_{i.eqo}^{L} - n_{i.eq}^{L}}\Delta H$$

The very small time interval  $(\Delta t \to 0)$  means that  $n_{i.eq}^L \to n_{i.eqo}^L$  and  $n_{i.eqo}^L - n_{i.eq}^L = \frac{\nu_T}{S_i} \Delta t$ . The last expressions simplify substantially (taking into account  $\Delta H/\Delta t = \nu_{gr}$ ) equation (29).



Figure 10. The situation at the beginning of liquid phase epitaxial growth in the case of *j*-component.

$$\delta_0 = 2S_i \frac{n_i^S - n_{i.eqo}^L}{\nu_T} \nu_{gr} \qquad (30)$$

The same expression could be reached for a j-component (Fig. 10).

$$\delta_0 = 2S_j \frac{n_j^S - n_{j.eqo}^L}{\nu_T} \nu_{gr} \qquad (30a)$$

In fact, the calculated quantity  $\delta_0$  gives the depth of the liquid phase, from which the volume of the epitaxial layer is built up. However, for very small times ( $\Delta t \rightarrow 0$ ), this depth coincides with the thickness of the interface region.

The equations (29), (30) and (30a) give an additional correlation for the determination of the interface region

depth  $\delta_0$ . The direct use of these equations gives only the ratio  $\delta_0/\Delta H$ . For the case of liquid phase epitaxial growth of GaAs and for the growth temperature of 1273 K, an approximate calculation gives this ratio as about 1600, for the growth temperature of 1173 K it is about 3500, for 1073 K - 8000 and for 973 K - 21000. This is not enough for the determination of  $\delta_0$  - an additional correlation is needed.

### 8. Discussion

All these considerations allow us to make some general conclusions.

The crystal growth is a strongly correlated process governed by the structure of the substrate and by its composition.

The whole practice has proved that independent of the liquid phase composition (corresponding to the liquidus surface), the interface region passes through the components in proportions preliminarily determined by the stoichiometry of the grown compound. During deposition of GaAs at different temperatures (in the range 600 -  $1000^{\circ}$ C), the composition of the grown solid phase is always constant (50 mol % Ga and 50 mol % As). Despite this the liquid phase composition (close to the liquidus line) undergoes significant changes.

One may conclude that the interface region is like a filter passing through the components of the system in proportions preliminarily given by the solid phase stoichiometry. A possible explanation could be the interface potential field caused by the substrate crystal. Probably, by the potential wells, this field transmits information not only about the structure of the substrate but also about its composition.

The ability of the interface region to be a filter passing through in strict proportions the components during growth, is represented by equations (7), (8), (8a), (9), (17 - 22). Exactly this circumstance unites all these equations.

Equations (30) and (30a) allow us to make conclusions about the depth of the interface region  $\delta_0$ . The difference  $\left(n_i^S - n_{i.eqo}^L\right)$  is a temperature depending quantity - it increases with decrease of the growth temperature. At growth temperatures near the melting point of the compound, it goes to zero. The low growth temperature leads to the low value of the growth velocity. At the same time, the slope of the liquid surface  $S_i$  increases significantly. One can not make only one conclusion with respect to the growth of all possible compounds, but there are grounds to assume that at low growth temperatures the depth of the interface region  $\delta_0$  will increase.

There is reason to assume that the thickness of the interface region  $\delta_0$  is different for different components. This difference could be caused by the different values of their concentrations in the liquid phase and by their different diffusion coefficients. In this case in equations (27 - 30) and (30a) the designation  $\delta_{0i}$  must be used in order to indicate that for the different components, the thickness of the interface is different. Such an assumption, however, will significantly complicate the physics of the crystal growth process.

We have assumed the depth of the interface region  $\delta_0$  to be the same for all components.

It has already been shown [75, 76] that for the process of particle migration on the crystal surface, the different components of the liquid phase must have the same coefficient  $\mu_0$ . This allows us to make the conclusion that the behavior of the components within the interface region is fully different from their behavior in the volume of the liquid phase. The components behavior in the interface region is probably unified despite the different values of their diffusion coefficients and of their concentrations in the liquid phase volume.

The latter allows us to assume that the depth of the interface region is the same for all components. Then from equations (30) and (30a) it leads inevitably that the product

$$S_k(n_k^S - n_{k.eqo}^L) = q_0 > 0 (31)$$

is also the same for all components:  $k \in (1, n)$ . Here one must point out that  $q_0$  depends not on the growth process conditions but on the parameters of the liquidus and solidus surfaces.

 $q_0$  is always a positive quantity  $(q_0 > 0)$ . For the *i*-components, where  $\left(n_i^S - n_{i.eqo}^L\right) > 0$ , it follows that it is identical to  $S_i > 0$ . For *j*-components  $\left(n_j^S - n_{j.eqo}^L\right) < 0$  and one obtains  $S_j < 0$ . This confirms the conclusions made above that the *i*-components are deposited always from a supercooled liquid phase and the *j*-components are deposited always from a superheated liquid phase.

### 9. Conclusion

In the present work the liquid phase multicomponent compound growth is considered. It is shown that part of the components participate in the growth process and are deposited onto the substrate from a supercooled liquid phase. For the rest of the components, the liquid phase is superheated. Despite this, they also participate in the growth process. A possible mechanism is presented explaining the possibility of the components, with respect to which the liquid phase is superheated, also to take part in the growth process.

The behavior of a given component during growth depends on the ratio between its liquid phase concentration and its solid phase one. Through this ratio a characteristic function can be defined depending exactly on the growth conditions. The value of this function during growth, at given temperature, is determined by the sum of the relative changes of the mass and of the concentration in the liquid phase of an arbitrary component.

During crystallization all components are deposited onto the substrate, building up the epitaxial layer in strict correlated proportions. These proprotions do not depend either on the growth temperature or on the liquid phase concentrations of the components. In this way, for the characterization of the growth process, it is quite sufficient to study the behavior of only one component - the basic component. The arbitrary component of the system may be the basic component.

Considering the growth process within a very small time interval, one could determine the depth of that region situated close to the phase boundary, where both phases interact directly by heat and mass exchange. The obtained result allows us to make the conclusion that at high temperature the depth of the interface region must be smaller and this could have substantial importance for the quality of the grown material.

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# Structure - Property Relationship Studies on Chiral Lactic Acid Derivatives

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#### Abstract

In earlier studies the mesophase sequences and ferroelectrics characterizations of the H and HO series of lactic acid derivatives were carried out. The chiral centres were connected either by ether or ester groups to the core. The ester linkage of latic acid preferred the formation of the tilted phase since the ethereal connection rather hindered it. These studies were extended to the compounds, which belong to H, HO, M, MO, DM, DMO series. In this presentation the effect both of lateral methyl substituent and the elongation of the terminal chiral chain by some additional chiral centres will be discussed.

 $Key \ words$ : Ferroelectric liquid crystals, phase sequences, transition temperatures, spontaneous polarization

### 1. Introduction

Since the first high speed electrooptic device based on ferroelectric liquid crystals (FLC) was discovered [1], a great interest has been directed to a search of new FLC substances which would satisfy application demands. High spontaneous polarization ( $P_S$ ) together with low viscosity are important properties which can ensure the high switching speed. We have synthesized new FLC materials (H n/m, M n/m, DM n/m) with high value of  $P_S$  [2, 3]. Chiral groups used in our study were derived from commercially supplied (S)-(-)-ethyllactate which enables a synthesis of a great number of chiral groups differing in the length of the carbon chain (H n/\*\*, HO n/\*\* [4], M n/\*\*, MO n/\*\*, DMO n/\*\*, DMO n/\*\*).



Figure 1. General structural scheme of investigated compounds.

Series	R	$R_1$	$R_2$	$R_3$	X	Y	Table
H n/m	n-alkyl	Η	Η	n-alkyl	OOC	0	2
H n/**	n-alkyl	Н	Η	$S-(2)-Me-Bu^*$	OOC	Ο	2
M n/m	n-alkyl	$CH_3$	Η	n-alkyl	OOC	Ο	3
M n/**	n-alkyl	$CH_3$	Η	S-(2)-Me-Bu*	OOC	Ο	3
MO n/m	n-alkyl	$CH_3O$	Η	n-alkyl	OOC	Ο	4
MO n/**	n-alkyl	$CH_3O$	Η	S-(2)-Me-Bu*	OOC	Ο	4
DM n/m	n-alkyl	$CH_3$	$CH_3$	n-alkyl	OOC	Ο	Fig.2.a-d
BM							5
HO n/m	n-alkyl	Η	Η	n-alkyl	0	COO	6
HO n/**	n-alkyl	Η	Η	$S-(2)-Me-Bu^*$	0	COO	6
DMO n/m	n-alkyl	$CH_3$	$CH_3$	n-alkyl	Ο	COO	7
DMO n/**	n-alkyl	$CH_3$	$CH_3$	$S-(2)-Me-Bu^*$	Ο	COO	7

 Table 1. List of new ferroelectrics materials.

### 2. Experimental

The sequences of phases and the phase transition temperatures were determined from texture observations by an Amplival Pol-U polarizing microscope equipped with a Boetius hot-stage.

Differential Scanning Calorimetry (DSC) measurements were carried out by a Perkin Elmer DSC7 equipment. The spontaneous polarization ( $P_s$ ) values were determined from the P(E) hysteresis loop detected during ferroelectric switching in an a.c. electric field at a frequency of 60 Hz.

#### 3. Results

Phase sequences and transition temperatures of the H n/m and H n/\*\* series are shown in Table 2. All substances exhibit a wide temperature range of SmC\* phase characterised by high  $P_s$  values. Nearly all substances (except six from the H n/m series) exhibit a phase transition from the SmC\* to a low temperature smectic phase, which is denoted by SmN. SmN is an orthogonal phase, dechiralization lines disappear at the phase transition; electrooptic switching is not observable.

The phase sequences of the methyl substituted homologous series (M n/m and M n/\*\*) on cooling are summarized in Table 3. The melting temperatures (m.p.), enthalpies, and the  $P_s$  values are given too. As shown in Table 2. and Table 3., all phase transition temperatures for the M n/m and M n/\*\* are significantly lower compared to the H n/m and H n/\*\* series. This effect is well known and can be explained in terms of the steric influence of the methyl group on molecular packing.

H 12/10

SmN

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•

Comp.			$SmC^*$		$SmA^*$		$N^*$		BP		$I_{SO}$	$\mathbf{P}_{\mathbf{S}}$
H 6/5	$\operatorname{Cr}$	40	٠	127	-		٠	150	-		٠	64
H 6/7	$\operatorname{Cr}$	37	•	124	-		•	143	-		•	57
H 6/8	$\operatorname{Cr}$	40	•	122	-		•	138	•	139	•	62
H $6/9$	$\operatorname{Cr}$	32	•	120	•	126	•	134	•	135	•	57
H 6/10	$\operatorname{Cr}$	40	•	119	•	128	•	132	٠	133	•	45
H $6/12$	$\operatorname{Cr}$	70	•	119	•	129	•	131	•	133	•	43
H 8/**	$\operatorname{SmN}$	64	•	122	-		•	153	-		•	59
H 8/4	$\operatorname{SmN}$	70	•	138	-		•	163	-		•	62
H 8/5	$\operatorname{Cr}$	72	•	134	-		•	145	-		•	91
H 8/7	$\operatorname{Cr}$	63	•	132	-		•	140	-		•	82
H 8/8	$\operatorname{SmN}$	61	•	128	-		•	134	-		•	60
H 9/8	$\operatorname{SmN}$	57	•	129	-		•	130	٠	133	•	95
H 9/9	$\operatorname{SmN}$	67	•	127	-		•	130	•	131	•	88
H 10/**	$\operatorname{SmN}$	72	•	126	-		•	132	•	134	•	77
$H \ 10/5$	$\operatorname{SmN}$	75	•	135	-		•	139	-		•	86
$H \ 10/7$	$\operatorname{SmN}$	69	•	130	-		•	139	-		•	67
$H \ 10/8$	$\operatorname{SmN}$	71	•	133	-		-		-		•	66
H 10/10	$\operatorname{SmN}$	68	•	128	-		-		-		•	61
H 10/12	$\operatorname{SmN}$	75	•	132	-		-		-		•	50
H 12/**	$\operatorname{SmN}$	82	•	142	-		•	157	-		•	32
$H \ 12/4$	$\operatorname{SmN}$	79	•	142	-		•	143	-		•	55
$H \ 12/5$	$\operatorname{SmN}$	69	•	136	-		-		-		•	59
$H \ 12/7$	$\operatorname{SmN}$	81	•	144	-		•	149	-		•	43
$H \ 12/8$	$\operatorname{SmN}$	77	•	136	-		-		-		•	59

Table 2. Phase sequences, transition temperatures (°C) and spontaneous polarization  $P_{\rm S}$  (nC/cm<sup>2</sup>) at the temperature 5°C below the transition to the SmC<sup>\*</sup> phase for the homologous series H n/m, H n/\*\*.

Table 3. Phase sequences, transition temperatures (°C) and spontaneous polarization  $P_{\rm S}$  (nC/cm<sup>2</sup>) at a temperature 5°C below the transition to the SmC<sup>\*</sup> phase for the homologous series M n/m, M n/\*\*.

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Comp.	m.p.	$\operatorname{Cr}$		$\mathrm{SmC}^*$		$N^*$		BP		$I_{\rm SO}$	$\mathbf{P}_{\mathbf{S}}$
M 6/9	63 [28.5]	٠	6	•	77 [2.4]	٠	$105 \ [0.7]$	-		٠	120
M 8/**	46 [15.4]	٠	10	•	$80 \ [3.5]$	•	87	•	$94 \ [0.8]$	•	192
M 8/5	50 [8.3]	٠	10 [25.8]	•	89[3.1]	•		-	104 [1.0]	•	121
M 8/6	65 [23.0]	٠	25	•	80[3.2]	•		-	107 [1.0]	•	180
M 8/10	76[55.0]	•	34[36.5]	•		•	76 [3.6]	-		•	
M 10/**	57[37.4]	•	-10	•	82[3.2]		89	•	94 [1.0]	•	153
M 10/10	75[38.3]	•	25 [35.0]	•	89[4.3]	•	90	•	91 [1.1]	•	83
$M \ 12/6$	44 [21.9]	٠	25[32.4]	•	91 [4.1]	•		-	96[1.1]	•	81
$M \ 12/10$	50 [29.5]	٠	24 [46.1]	•		-		-	87 [6.1]	٠	74

When we compare the properties of the M n/m compounds and their MO n/m analogues, a significant difference can be observed: the SmC\* - Cr phase transitions of the M n/m series are lowered substantially and the transitions to the isotropic state are shifted to slightly higher temperatures (Table 3. and Table 4.). The narrow blue phase (BP), which appears for the M n/m series is not observable for the MO n/m series.

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A comparison of the  $P_S$  values for MO n/m and H n/m (Table 4. and Table 2.) shows that the methoxy group does not influence the  $P_S$  value.

Table 4. Phase sequences, transition temperatures (°C) and spontaneous polarization  $P_{\rm S}$  (nC/cm<sup>2</sup>) at the temperature 5°C below the transition to the SmC\* phase for the homologous series MO n/m, MO n/\*\*.

Comp.			SmC*		SmA		N*		BP	Iso	$\mathbf{P}_{\mathbf{S}}$
MO 8/5	$\operatorname{Cr}$	54	٠	69	•	79	٠	80	-	•	66
MO 8/7	$\operatorname{Cr}$	37	•	82	-		٠	103	-	•	50
MO 8/12	$\operatorname{Cr}$	37	•	65	-		٠	85	-	•	24
MO $10/**$	$\operatorname{Cr}$	55	•	71	•	77			-	•	66
$MO \ 10/5$	$\operatorname{Cr}$	62	•	81	-		-		-	•	57
$MO \ 10/10$	$\operatorname{Cr}$	45	•	89	-		-		-	•	57
$MO \ 10/12$	$\operatorname{Cr}$	61	•	80	-		-		-	•	35
$MO \ 12/5$	$\operatorname{Cr}$	70	•	82	•	85	-		-	•	27
$MO \ 12/7$	$\operatorname{Cr}$	52	•	86	-		-		-	٠	61
$MO \ 12/8$	$\operatorname{Cr}$	66	•	80	•	85	•	87	-	٠	47
MO $12/10$	$\operatorname{Cr}$	49	•	85	-		-			•	76
MO 12/12	$\operatorname{Cr}$	62	•	82	-		-		-	•	66

The data of the DM n/m series [5], measured by DSC, were in good accordance with that of the microscopic observations. The phase diagrams of four homologous series of DM n/m can be seen in Fig.2a-d. Almost every member of the homologous series exhibits monotropic BP, N<sup>\*</sup>, SmC<sup>\*</sup> mesophases, except DM 10/6, which has a very short enantiotropic N<sup>\*</sup> phase (Fig.2c).



Figure 2a-d. Phase diagrams of the homologous series a) DM 7/m, b) DM 8/m, c) DM 10/m, d) DM 12/m .

Binary mixtures of these compounds were prepared and investigated (Table 5). The mixtures (BM1-BM6) were composed of members of the DM n/m series with chiral chains differing from each other by four carbon atoms in length. These mixtures exhibit the enantiotropic SmC<sup>\*</sup> and N<sup>\*</sup> phase. Presumably the intercalated tail-to-tail packing of the molecules stabilizes the ferroelectric phase, making it enantiotropic.

Mixtures	Comp.	Wt[%]	m.p.	$\operatorname{Cr}$	$SmC^*$	$N^*$	Ι
BM1	DM7/6	50	46.5	• (30)	• 51	-	٠
	DM7/10	50					
BM2	DM10/8	60	45	• (38)	• 48	• $53.5$	•
	DM10/12	40					
BM3	DM10/8	46.7	45	• (37)	• 49	• 53	•
	DM10/12	53.3					
BM4	DM12/8	44.1	50	• (37)	• 54	• 55	٠
	DM12/12	45.9					
BM5	DM12/8	43.2	52	• (40)	• 54	-	•
	DM12/12	56.8					
BM6	DM12/8	40	54	• (40)	• 55	-	•
	DM12/12	60					
BM7	DM10/8	33.3	78	$\bullet$ (55)	• (72)	-	•
	M8/10	66.7					
BM8	DM8/10	50	57	• (40)	• $(55.5)$	• 59	٠
	M8/10	50					
BM9	DM7/5	60	51	• (28)	• (36)	• 51	•
	DM12/12	40					
BM10	DM7/5	40	52	• (40)	• (42)	• 52	•
	DM12/12	60					
	M8/10	100	76	• (34)	• 76	-	٠

Table 5. Phase sequences and transition temperatures (°C) for the binary mixtures.

The phase sequences of two series of chiral liquid crystalline substances HO n/m, having an function as connecting group, are shown in Table 6. All compounds exhibit BP,  $N^*$  and SmA\* phases. The ferroelectric SmC\* phase disappears.

**Table 6.** Phase sequences and transition temperatures (°C) for the homologous seriesHO n/m and HO n/\*\*.

Comp.	$\operatorname{Cr}$		$\operatorname{SmB}$		$SmC^*$		SmA*		$N^*$		BP		$I_{SO}$
HO 8/6	٠	32	•	43	-		٠	52	•	95	•	98	٠
HO $8/**$	•	40	•	56	-		٠	66	•	82	٠	85	•
HO $9/6$	•	33	•	42	-		•	53	•	87	•	91	•
HO $10/6$	•	27	•	44	-		•	64	•	92	•	95	•
HO $10/8$	•	39	•	42	-		•	55	•	84	•	87	•
HO $10/10$	•	37	•	40	-		•	61	•	77	•	83	•
HO 10/12	•	36	•	40	-		•	59	•	77	•	83	•
HO 10/**	٠	41	•	53	•	58	٠	65	•	84	•	87	•

Two new chiral homologous series of DMO n/m were synthesized (Fig. 3) and characterised too.



Figure 3. Synthesis of DMO n/m series.

These materials do not possess any liquid crystalline phase (Table 7). Having low melting points, these new materials can be used as chiral additives to decrease the melting points of multicomponent ferroelectric mixtures.

**Table 7.** Melting temperatures (°C) for the homologous series DMO n/m, DMO  $12/^{**}$ and DMO  $12/^{***}$ .

Comp.	m.p.	Ι
DMO 10/6	35-38	•
DMO $10/10$	53 - 54.5	•
DMO $10/12$	44.5 - 46.5	•
DMO $12/10$	52 - 53	•
DMO 12/12	52 - 54	•
DMO 12/**	30-32	•
DMO 12/***	Liquid at -20	

# 4. Conclusion

In the non-substituted series (H n/m), where the chiral center is connected to the core by an ester function, the mesophase formation is enhanced, the ferroelectric SmC<sup>\*</sup> phase has a wide temperature range. This fact is due to the conjugation between the aromatic system and the carbonyl group of the chiral part, since the core of the molecule is enlarged and the in-plane conformation is more favourable.

The methyl or methoxy substituent in position 3 of the phenyl ring (M n/m and MO n/m series), decreases the transition temperatures in comparison to the non-substituted derivatives (H n/m series).

The introduction of two methyl groups into 3,5-positions gives rise to a dramatic change of mesogenic behaviour. The members of the DM n/m series show only monotropic mesophases. The ether linkage of the chiral center hinders the formation of ferroelectric properties (HO n/m), the rigid core of the molecule becomes shorter and it is enough to destroy completely the liquid crystalline behaviour in DMO n/m series.

The introduction of the second chiral center enlarges the molecule, i.e. the elongation of the chiral part, results in a higher value of  $P_s$  (M 10/\*\* and MO 10/\*\*).

These effects suggest that the most powerful compounds are the H n/m, M n/m and MO n/m series which have a wide  $SmC^*$  range and a high value of  $P_s$ .

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# Energy Loss Distributions in Thick Targets

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#### Abstract

A new method for calculating the energy straggling distribution of charged particles after their passage through a thick target has been established. The comparisons of calculated spectra with the experimental alpha-spectra in noble gases show good agreement in the limits of accuracy of Bohr's cross-section formula. The possibility of using the Monte-Carlo simulation of charged particles passage through a medium in testing various cross-section formulae for the projectile energy loss has been discussed.

Key words: Alpha-particles, energy loss, straggling

### 1. Introduction

Energy loss distributions after the passage of charged particles through a target have an asymmetric shape, with a long tail toward higher energy losses. The asymmetry of a peak is more expressed in thin targets than in thick ones. Many authors studied the problems of straggling distributions. We shall mention only a few of them: Landau [1], Vavilov [2], Shulek *et al* [3], Bichsel [4] and Sigmund and Winterbon [5] in thin targets, and Tschalär [6] in thick targets. In our previous paper (Novković *et al* [7]) we derived a simple analytical formula for a peak broadening in thick targets starting from Symon's equations [8]. The formula was verified on experimental alpha spectra in noble gases (Novković *et al* [7]), and, also, in diatomic and polyatomic gases (Milošević *et al* [9]). Since only peak widths were determined, alpha spectra were approximated with the Gaussian. More accurate reproducing of energy straggling distributions requires calculations of more than one parameter of the energy loss distribution functions. The aim of this paper is to establish a method for accurate calculations of charged particle energy distributions.

#### 2. Procedure for calculations of asymmetric distributions

In his known paper Tschalär [6] calculated the energy loss distribution functions of charged particles, by solving Symon's system of differential equations for central moments [8], and applying Johnson's functions [12] as density distribution functions. Symon's system for central moments of the energy straggling distributions reads

$$\frac{d\sigma_n}{dx} = \sum_{l=0}^{\infty} \frac{(-1)^l}{l!} \left[ \sum_{m=1}^n \binom{n}{m} \frac{d^l k_m}{dT^l} \sigma_{n-m+l} - n\sigma_{n-1}\sigma_l \frac{d^l k_1}{dT^l} \right],\tag{1}$$

where  $\sigma_0 = 1$ ,  $\sigma_1 = 0$ , T is the mean energy of the straggling distributions, and

$$k_n = N \int_0^\infty dw \tau^n,\tag{2}$$

where N is the number of atoms per unit mass, and dw is the differential cross-section for a particle energy loss of  $\tau$ . The quantity  $k_1$  is equal to the stopping power. The most frequently used cross-section is Bohr's formula [10]

$$dw = \frac{2\pi z_1^2 z_2 e^4}{mv^2} \frac{1}{\tau^2},\tag{3}$$

where  $z_1e$  and  $z_2e$  are projectile and target charge, respectively, m is the electron mass and v is the projectile velocity. Minimum and maximum energy transfer in a collision are

$$\tau_{\min} = I^2 / \varepsilon T,$$
  
 $\tau_{\max} = \varepsilon T.$ 
(4)

where  $\varepsilon$  is

$$\varepsilon = 4\frac{m}{M} \left(1 + \frac{m}{M}\right)^{-2} \approx 4\frac{m}{M},\tag{5}$$

and M is the projectile mass and I is the mean ionization potential of the stopping medium. Using cross-section formula (3), the expressions for  $k_n$  are obtained [6]

$$k_n = \frac{2N\pi z_1^2 z_2 e^4}{mv^2} \frac{(\varepsilon T)^{n-1}}{n-1}, \qquad n = 2, 3, \dots$$
(6)

and

$$k_1 = \frac{2N\pi z_1^2 z_2 e^4}{mv^2} \ln \frac{\tau_{\max}}{\tau_{\min}} = S(T),$$
(7)

where S(T) is the stopping power. Instead of the above simplified expression for  $k_1$ , empiric or semi-empiric formulae for stopping power can be used. Since

$$\frac{2N\pi z_1^2 z_2 e^4}{mv^2} = S(T) / \ln(\tau_{\max}/\tau_{\min})$$
(8)

expression (6) is changed

$$k_n = \frac{S(T)}{\ln(\tau_{\max}/\tau_{\min})} \frac{(\varepsilon T)^{n-1}}{n-1}, \qquad n = 2, 3, \dots$$
 (9)

The  $n^{th}$  order central moment is not defined in the conventional manner but as [6]:

$$\sigma_n = \int_0^\infty f(T)(\bar{T} - T)^n dT.$$
(10)

Substituting a new variable  $\Delta = T_0 - T$ , where  $T_0$  is an emerging particle energy and  $\Delta$  a particle energy loss, we obtain the conventional formula for the n<sup>th</sup> order central moment:

$$\sigma_n = \int_0^\infty f(\Delta) (\Delta - \bar{\Delta})^n d\Delta.$$
(11)

For the fitting alpha peaks of alpha-spectrometers, Bortels and Colaers [11] successfully applied the following formula

$$f(\Delta) = \frac{A}{t_0} \exp\left(\frac{w^2}{2t_0^2} - \frac{\Delta - \Delta_p}{t_0}\right) \int_{-\infty}^{z} \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{y^2}{2}\right) dy, \qquad (12)$$
$$z = \frac{\Delta - \Delta_p}{w} - \frac{w}{t_0},$$

or

$$f(\Delta) = \frac{A}{t_0} \exp\left(-\frac{w^2}{2t_0^2} - \frac{zw}{t_0}\right) \int_{-\infty}^{z} \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{y^2}{2}\right) dy = \\ = \frac{A}{2t_0} \exp\left(-\frac{w^2}{2t_0^2} - \frac{zw}{t_0}\right) \left\{1 + \operatorname{erfc}\left(\frac{z}{\sqrt{2}}\right)\right\}.$$
(13)

We also used this formula in our calculations since Bortels and Colaers had good results applying it and since the central moment of the formula has closed forms (Appendix B).

In the above expression A, w,  $t_0$  and  $\Delta_p$  are parameters,  $\Delta$  is the charged particle energy loss. The maximum of  $f(\Delta)$  is at the point  $\Delta = \Delta_p$ , therefore  $\Delta_p$  is the most probable energy loss. Constant A is the constant of normalization:

$$\int_{0}^{\infty} f(\Delta) d\Delta \cong \int_{-\infty}^{\infty} f(\Delta) d\Delta = A.$$
(14)

In the above integral the lower limit can be extended from 0 to  $-\infty$  since left of the peak curve  $f(\Delta)$  rapidly decreases to 0. For a normalized spectrum A = 1 (Appendix A).

Mean energy loss  $\overline{\Delta}$ , the second order and third order central moments are (see Appendix A and B)

$$\bar{\Delta} = \Delta_p + t_0, \tag{15}$$

$$\mu_2 = \int_{-\infty}^{+\infty} (\Delta - \bar{\Delta})^2 f(\Delta) d\Delta = t_0^2 + w^2, \qquad (16)$$

$$\mu_3 = \int_{-\infty}^{+\infty} (\Delta - \bar{\Delta})^3 f(\Delta) d\Delta = 2t_0^3, \qquad (17)$$

If the real energy straggling distribution is completely described by function (12), then central moments  $\mu_n$  of f and central moments  $\sigma_n$  of real energy distributions would be equal and all three parameters,  $\Delta_p$ ,  $t_0$  and w could be calculated by expressions (14), (15) and (16). The real energy straggling distributions are not completely determined by functions (12). Therefore, more than three central moments of  $f(\Delta)$  have to be calculated in order to obtain minimum deviation  $f(\Delta)$  from the real energy distribution of charged particles. Parameters  $\Delta_p$ , w, and  $t_0$  should be determined so that  $\mu_n$  minimum deviates from  $\sigma_n$ . The expressions for ten central moments  $\mu_n$  of  $f(\Delta)$  are given in Appendix B. It is evident that the  $n^{th}$  root of  $\mu_n$ 's has the same order of magnitude, hence in order to optimize parameters  $\Delta_p$ ,  $t_0$  and w, we introduce function  $R(t_0, w)$ ,

$$R(t_0, w) = \sum_{n=2}^{N} \left[ 1 - \left( \frac{\mu_n(t_0, w)}{\sigma_n} \right)^{\frac{1}{n}} \right]^2,$$
(18)

where  $\mu_n$  are expressions given in Appendix B, and  $\sigma_n$  are solutions of Symon's system of differential equations (1). Parameters  $t_0$  and w are best determined when  $R(t_0, w)$  has a minimum, i. e. when the following two equations are satisfied

$$\frac{\partial R(t_0, w)}{\partial t_0} = 0$$

$$\frac{\partial R(t_0, w)}{\partial w} = 0.$$
(19)

The number of central moments, N, were varied from 6 to 10 and the obtained results were very close. In (18) ratio  $(\mu_n/\sigma_n)^{1/n}$  is of the same order of magnitude for different n, so all central moments were taken into account in the course of calculation. Usually, instead of expression (18), the ratio  $\sum_{n=2}^{N} \left[1 - \left(\frac{\mu_n(t_0,w)}{\sigma_n}\right)\right]^2$  is used. In this case larger moments have a greater influence on the shape of the peak, since the ratio  $(\mu_n/\sigma_n)$  increases with n.

### 3. Monte-Carlo simulation

Bohr's total cross-section for interaction of a charged particle with a stopping medium is obtained by integration (3) in the limits  $[\tau_{\min}, \tau_{\max}]$ ,

$$w = \int_{\tau \min}^{\tau \max} dw = \frac{4\pi z_1^2 z_2 e^4}{\tau_{\max}} \left( \frac{1}{\tau_{\min}} - \frac{1}{\tau_{\max}} \right) = \frac{S(T)}{N} \frac{1}{\ln(\tau_{\max}/\tau_{\min})} \left( \frac{1}{\tau_{\min}} - \frac{1}{\tau_{\max}} \right) \qquad ,$$
(20)

and the mean path length,  $\lambda_0$ , is

$$\lambda_0 = 1/(Nw) = \frac{\ln(\tau_{\max}/\tau_{\min})}{S(T)} \frac{I^2}{\tau_{\max} - \tau_{\min}}$$
(21)

The distribution function of path lengths is

$$F(\lambda) = 1 - e^{-\lambda/\lambda_0},\tag{22}$$

where  $\lambda$  is a free path length between two successive collisions of a charged particle. Applying the method of inversion function, a free path length between  $(i-1)^{th}$  and  $i^{th}$  collision is

$$\lambda_i = -\lambda_0 \ln(1 - r_i),\tag{23}$$

where  $r_i$  is a pseudo-random number in the range [0, 1). The energy loss distribution density function is

$$\frac{dq}{d\tau} = \frac{1}{w}\frac{dw}{d\tau} = \frac{1}{1/\tau_{\min} - 1/\tau_{\max}}\frac{1}{\tau^2},$$
(24)

which, using the method of inversion function, gives an energy loss in  $i^{th}$  collision,  $\tau_i$ ,

$$\frac{1}{\tau_i} = \frac{1 - r_i}{\tau_{\min}} + \frac{r_i}{\tau_{\max}}.$$
(25)

In the Monte-Carlo simulation of the charged particle passage through a target of thickness d, energy loss is obtained by summing of  $\tau_i$  until the particle exits the target, i. e.,

$$\Delta = \sum_{i} \tau_{i}$$

$$\sum_{i} \lambda_{i} \leq d.$$
(26)

This method is suitable only for thin targets, since  $\lambda_0$  is very small (the order of magnitude of  $\lambda_0$  for alpha-particles in neon amounts to  $10^{-7}$  g/cm<sup>2</sup>).

### 4. Discussion and conclusion

The alpha-spectra of ThC+ThC' single-line (E = 8.7844 MeV) in noble gases have been chosen for an experimental testing of the present method for the calculation of energy straggling distributions. The experimental arrangement and a part of the experimental results have been published earlier (Novković et al. [7]). The alpha straggling spectra in the thin Ne target obtained by the Monte-Carlo simulation, Vavilov's formula [2], and by the present method of calculation are shown in Fig. 1. The curves, evaluated by the Monte-Carlo simulation and Vavilov's formula show excellent agreement, but the spectrum, calculated by the present method, significantly deviates from them. In this case the target is too thin for this method of calculation. The comparison of the experimental spectra with the spectra calculated by the present method or obtained by the Monte-Carlo simulation are given in Figs. 2, 3, 4, 5 and 6. The agreement of the experimental spectra



Figure 1. The comparison of the present method of calculation of charged particles energy loss spectra in the thin neon targe with Vavilov's formula and the Monte-Carlo simulation.

Figure 2. The comparison of the present method of calculation of the energy straggling spectrum in the thick helium target with the experimental spectrum and Monte-Carlo simulation.

with the calculated spectra are reasonably good except for the spectra in He (Fig. 2). Namely, the Monte-Carlo simulation curve is twice narrower than the experimental curve.
This deviation is probably due to the inaccuracy of Bohr's cross-section formula. However, the present method also applied the same formula; hence, we conclude that it is less sensitive to the cross-section energy loss formula than the Monte-Carlo simulation of the passage of the charged particles. This feature of the Monte-Carlo simulation can be used for testing various theoretical cross-section formulae for projectile-stopping medium interaction. Several examples of the calculated spectra by the present method and the experimental spectra after alpha particles passage through thick targets are given in Fig. 7.





**Figure 3.** The comparison of the present method of calculation of the energy straggling spectrum in the thick neon target with the experimental spectrum and Monte-Carlo simulation.

Figure 4. The comparison of the present method of calculation of the energy straggling spectrum in the thick argon target with the experimental spectrum and Monte-Carlo simulation.



0 Experiment Calculated with 4000 ten central moments Monte-Carlo 3000 Xe ∆E=1.20 MeV Counts 2000 1000 0 7.6 7.4 7.7 7.8 7.3 7.5 E [MeV]

Figure 5. The comparison of the present method of calculation of the energy straggling spectrum in the thick krypton target with the experimental spectrum and Monte-Carlo simulation.

Figure 6. The comparison of the present method of calculation of the energy straggling spectrum in the thick xenon target with the experimental spectrum and Monte-Carlo simulation.

All calculations, solving Symon's system of differential equations with ten central moments (1), finding roots of equations (21), deducing the expressions for central moments,  $\mu_n$ , of function  $f(\Delta)$  using recurrence relation (B. 4), have been done by the Mathematica 3.0 program.



Figure 7. The experimental and calculated alpha energy spectra in noble gases.

# 5. Appendix A: The mean energy loss

$$\int_{0}^{\infty} f(\Delta) \ d\Delta \cong \int_{-\infty}^{\infty} f(\Delta) \ d\Delta = w \int_{-\infty}^{\infty} f(\Delta) \ dz =$$

$$= \frac{wA}{t_0\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp\left(-\frac{w^2}{2t_0^2} - \frac{zw}{t_0}\right) \ dz \int_{-\infty}^{z} \exp\left(-\frac{y^2}{2}\right) \ dy =$$

$$= \frac{wA}{t_0\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp\left(-\frac{y^2}{2} - \frac{w^2}{2t_0^2}\right) \ dy \int_{y}^{\infty} \exp\left(-\frac{zw}{t_0}\right) \ dz =$$

$$= \frac{A}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp\left(-\frac{1}{2}\left(y + \frac{w}{t_0}\right)^2\right) \ dy = A.$$
(A.1)

$$\begin{split} \bar{\Delta} &= \int_{0}^{\infty} \Delta f(\Delta) d\Delta \cong \int_{-\infty}^{\infty} \Delta f(\Delta) d\Delta = w \int_{-\infty}^{\infty} \left( wz + \frac{w^2}{t_0} + \Delta_p \right) f(\Delta) dz = \\ &= w^2 \int_{-\infty}^{\infty} z f(\Delta) dz + w \left( \frac{w^2}{t_0} + \Delta_p \right) \int_{-\infty}^{\infty} f(\Delta) dz = \end{split}$$

$$= \frac{w^{2}A}{t_{0}\sqrt{2\pi}} \int_{-\infty}^{\infty} z \exp\left(-\frac{w^{2}}{2t_{0}^{2}} - \frac{zw}{t_{0}}\right) dz \int_{-\infty}^{z} \exp\left(-\frac{y^{2}}{2}\right) dy + A\left(\frac{w^{2}}{t_{0}} + \Delta_{p}\right) = \\ = \frac{w^{2}A}{t_{0}\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp\left(-\frac{y^{2}}{2} - \frac{w^{2}}{2t_{0}^{2}}\right) dy \int_{y}^{\infty} z \exp\left(-\frac{zw}{t_{0}}\right) dz + A\left(\frac{w^{2}}{t_{0}} + \Delta_{p}\right) = \\ = \frac{wA}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \left(y + \frac{t_{0}}{w}\right) \exp\left(-\frac{1}{2}\left(y + \frac{w}{t_{0}}\right)^{2}\right) dy + A\left(\frac{w^{2}}{t_{0}} + \Delta_{p}\right) = \\ = \frac{wA}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \left(y + \frac{w}{t_{0}} - \frac{w}{t_{0}} + \frac{t_{0}}{w}\right) \exp\left(-\frac{1}{2}\left(y + \frac{w}{t_{0}}\right)^{2}\right) dy + A\left(\frac{w^{2}}{t_{0}} + \Delta_{p}\right) = \\ = \frac{wA}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \left(y + \frac{w}{t_{0}}\right) \exp\left(-\frac{1}{2}\left(y + \frac{w}{t_{0}}\right)^{2}\right) dy + \\ + \frac{A(t_{0}^{2} - w^{2})}{t_{0}\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp\left(-\frac{1}{2}\left(y + \frac{w}{t_{0}}\right)^{2}\right) dy + \\ + A\left(\frac{w^{2}}{t_{0}} + \Delta_{p}\right) = A(\Delta_{p} + t_{0})|_{A=1} = \Delta_{p} + t_{0}$$
(A.2)

# 6. Appendix B: The Recurrence formula

The central moment of order n of function  $f(\Delta)$  is defined by the expression

$$\mu_n = \int_{0}^{+\infty} (\Delta - \bar{\Delta})^n f(\Delta) d\Delta \approx \int_{-\infty}^{+\infty} (\Delta - \bar{\Delta})^n f(\Delta) d\Delta.$$
(B.1)

Since

$$z = \frac{\Delta - \Delta_p}{w} - \frac{w}{t_0} = \frac{\Delta - \bar{\Delta}}{w} + \frac{t_0}{w} - \frac{w}{t_0},$$

and

$$dz = \frac{d\Delta}{w}$$

we obtain the following expression for  $\mu_n$ 

$$\mu_n = w^{n+1} \int\limits_{-\infty}^{+\infty} \left( z + \frac{w}{t_0} - \frac{t_0}{w} \right)^n f(\Delta) dz.$$
(B.2)

Derivating the left and right side of the above expression in terms of w, we have

$$\frac{d\mu_n}{dw} = \frac{n+1}{w}\mu_n + nw^{n+1} \int_{-\infty}^{+\infty} \left(z + \frac{w}{t_0} - \frac{t_0}{w}\right)^{n-1} f(\Delta)dz + w^{n+1} \int_{-\infty}^{+\infty} \left(z + \frac{w}{t_0} - \frac{t_0}{w}\right)^n \frac{\partial f}{\partial w}dz = 0$$

$$= \frac{n+1}{w}\mu_{n} + nw\left(\frac{1}{t_{0}} + \frac{t_{0}}{w^{2}}\right)\mu_{n-1} - w^{n+1}\int_{-\infty}^{+\infty} \left(z + \frac{w}{t_{0}} - \frac{t_{0}}{w}\right)^{n}\left(\frac{w}{t_{0}^{2}} + \frac{z}{t_{0}}\right)f(\Delta)dz =$$

$$= \frac{n+1}{w}\mu_{n} + n\mu_{n-1}\left(\frac{w}{t_{0}} - \frac{t_{0}}{w}\right) - \frac{w^{n+1}}{t_{0}} \times$$

$$\times \int_{-\infty}^{+\infty} \left(z + \frac{w}{t_{0}} - \frac{t_{0}}{w}\right)^{n}\left(z + \frac{w}{t_{0}} - \frac{t_{0}}{w} + \frac{t_{0}}{w}\right)f(\Delta)dz =$$

$$= \frac{n+1}{w}\mu_{n} + n\mu_{n-1}\left(\frac{w}{t_{0}} - \frac{t_{0}}{w}\right) - \frac{w^{n+1}}{t_{0}} \times \int_{-\infty}^{+\infty} \left(z + \frac{w}{t_{0}} - \frac{t_{0}}{w}\right)^{n+1}f(\Delta)dz -$$

$$- \frac{w^{n+1}}{t_{0}}\frac{t_{0}}{w}\int_{-\infty}^{+\infty} \left(z + \frac{w}{t_{0}} - \frac{t_{0}}{w}\right)^{n}f(\Delta)dz =$$

$$= \frac{n+1}{w}\mu_{n} + n\mu_{n-1}\left(\frac{w}{t_{0}} - \frac{t_{0}}{w}\right) - \frac{1}{t_{0}w}\mu_{n+1} - \frac{1}{w}\mu_{n} =$$

$$= \frac{n}{w}\mu_{n} + n\mu_{n-1}\left(\frac{w}{t_{0}} - \frac{t_{0}}{w}\right) - \frac{1}{t_{0}w}\mu_{n+1},$$
(B)

$$\mu_{n+1} = nt_0\mu_n + n\mu_{n-1}(w^2 + t_0^2) - t_0w\frac{d\mu_n}{dw}$$
(B.3)

If we introduce a new variable  $\eta = t_0/w$  in the above equation, we have

$$\mu_{n+1} = nt_0\mu_n + n\mu_{n-1}t_0^2\left(1 + \frac{1}{\eta^2}\right) + \eta t_0\frac{d\mu_n}{d\eta}, \qquad n = 2, 3, \dots$$
(B.4)

Since  $\mu_0 = 1$  and  $\mu_1 = 0$ , the following expressions are deduced from recurrence formula (B.4)

$$\begin{split} \mu_2/t_0^2 &= 1 + 1/\eta^2 \\ \mu_3/t_0^3 &= 2 \\ \mu_4/t_0^2 &= 9 + 6/\eta^2 + 3/\eta^4 \\ \mu_5/t_0^5 &= 44 + 20/\eta^2 \\ \mu_6/t_0^6 &= 265 + 135/\eta^2 + 45/\eta^4 + 15/\eta^6 \\ \mu_7/t_0^7 &= 1854 + 924/\eta^2 + 210/\eta^4 \\ \mu_8/t_0^8 &= 7(2119 + 1060/\eta^2 + 270/\eta^4 + 60/\eta^6 + 15/\eta^8) \\ \mu_9/t_0^9 &= 8(16687 + 8343/\eta^2 + 2079/\eta^4 + 315/\eta^6) \\ \mu_{10}/t_0^{10} &= 9(148329 + 74165/\eta^2 + 18550/\eta^4 + 3150/\eta^6 + 525/\eta^8 + 105/\eta^{10}) \end{split}$$

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#### Gamma-Ray Spectroscopy Measurement of Depleted Uranium in Soil

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#### Abstract

Depleted uranium (DU) is a radioactive and chemically toxic substance in which the natural ratio of isotopes  $^{235}$ U and  $^{238}$ U is changed by technological procedure so as to decrease the quantity of  $^{235}$ U. The mountain Plačkovica near Vranje and three other areas in southern Serbia (Vranje, Preševo and Bujanovac) are well known to have been contaminated by DU ammunition since the end of NATO bombing in the summer of 1999. Much of that amount of DU was converted at high temperature into an aerosol, that is, minute insoluble particles of uranium oxide,  $UO_2$  or  $UO_3$ , in a mist or fog [1,2]. The aerosol resists gravity, and is able to travel tens of kilometers in air. Once on the ground, it can be resuspended when the sand is disturbed by motion or wind. Identification of contamination by DU would require specialized equipment for detecting low-level concentrations of DU in soil. This is a complex problem because every soil already has a certain concentration of natural uranium [3]. The research team of the Laboratory of Nuclear Physics at the Faculty of Science possesses both the equipment and long-time experience in measuring low-level concentration of gamma-ray emitting radionuclides in environmental samples. It is shown here that the best analytical method and technique for DU assessment is the determination of the ratio of  $^{238}U/^{226}Ra$  activity because this ratio is significantly altered in the case of samples contaminated by depleted uranium. This is a complicated technique since it requires the detection of lowenergy (below 100 keV) gamma ray of <sup>234</sup>Th, which is the direct short-lived daughter of <sup>238</sup>U. For that purpose we used the ORTEC "GMX" spectrometer which is placed within the low-level iron shield in the Laboratory of Nuclear Physics at the Faculty of Science in Novi Sad. The main advantage of the method is that there is no need for any chemical treatment of the samples. From the measurements of numerous samples in our laboratory we have determined the detection limit of specific activity of depleted uranium in the soil by this method to be about 10 Bq/kg.

Key words: Radioactivity, depleted uranium, low-level gamma-ray spectroscopy

#### 1. Introduction

Uranium, U, is a radioactive element with the atomic number Z = 92. In natural form, uranium is composed of three isotopes <sup>238</sup>U, <sup>235</sup>U and <sup>234</sup>U (see Table 1). <sup>238</sup>U is

the parent of the radioactive decay series in which an equilibrium between the short-lived  $^{234}$ U (member of this chain) and long-lived  $^{238}$ U is established. Therefore, their decay rate is identical and the number of daughter nuclei remains constant relative to the number of parent nuclei.

Isotope	Abundance [%]	Decay	Half-life
$^{238}\mathrm{U}$	$99.2745 \pm 0.0015$	$\alpha$	$(4.468 \pm 0.005) \times 10^9 \text{ y}$
$^{235}\mathrm{U}$	$0.7200 \pm 0.0012$	$\alpha$	$(7.037 \pm 0.011) \times 10^8 \text{ y}$
$^{234}\mathrm{U}$	$0.005\pm0.0005$	$\alpha$	$(2.454 \pm 0.006) \times 10^5 \text{ y}$

Table 1. Isotopes in natural uranium.

 $^{235}$ U is the parent of a second radioactive series. This isotope is susceptible to fission by thermal neutrons in the process of chain-reaction and is of paramount importance for releasing nuclear power. Most technologies require higher  $^{235}$ U content than the natural one. This kind of substance is called enriched uranium. It is used as a fuel in nuclear reactors and in the production of nuclear weapons.

Depleted uranium is a secondary product of isotopic enrichment. It is composed of isotopes  $^{238}$ U,  $^{235}$ U and  $^{234}$ U, but with the concentration of two lighter isotopes 3-4 times lower than in natural uranium ( $^{235}$ U ~ 0.2 %). Specific activity of the depleted uranium is about 60 % of that of natural uranium, mostly due to the lower  $^{234}$ U activity.

Uranium oxide and its aerosols are insoluble in water. Once breathed in, the very small particles of uranium oxide, those that are 2.5 microns or less in diameter, may reside in the lungs for years, slowly passing through the lung tissue into the blood. Uranium oxide dust has a biological half-life in the lungs of about a year. Because of coughing and other involuntary mechanisms by which the body keeps large particles out of the lungs, the larger particles are excreted through the gastro-intestinal tract in feces [4]. The uranium compounds, which enter the body either through the wall of the gastro-intestinal tract or the lungs, can be broken down in the body fluids, and tetravalent uranium is likely to oxidize to the hexavalent form, followed by the formation of uranyl ions. Uranium generally forms complexes with citrate, bicarbonates or protein in plasma, and it can be stored in bone, lymph, liver, kidney or other tissues. Eventually this uranium which is taken internally is excreted through urine. The presence of depleted uranium in urine seven or eight years after exposure is sufficient evidence to substantiate long term internal contamination and tissue storage of this radioactive substance.

Uranium is both a chemical toxic and radioactive hazard. Soluble uranium is regulated because of its chemical toxicity, measured by damage to the kidney and tubules. Uranium is a heavy metal, known to cause uranium nephritis. Because of its slow absorption through the lungs and long retention in body tissues, its primary damage will be due to its radiological damage to internal organs rather than chemical damage to the renal system. Obviously, both types of damage occur simultaneously, therefore it is a matter of judgment which severe damage, radiological or chemical, occurs at the lowest dose level. However, with the lengthening of the time during which the contaminant resides in the body and the low overall dose, the risk of cancer death becomes greater than the risk of significant damage to the renal system.

# 2. Gamma-ray spectrometry of uranium in soil

Since the activities of all nuclides are identical when the radioactive series is in equilibrium, the concentration of the parent nuclide can be determined from any daughter acvtivity. Interruption of equilibrium due to geo-chemical processes may occur by longlived members of series ( $^{230}$ Th and  $^{226}$ Ra in the series of  $^{238}$ U, and  $^{231}$ Pa in the series of  $^{235}$ U).

Emanation of radon as a gas from samples may interrupt equilibrium by nuclides  $^{222}$ Rn and  $^{219}$ Rn. Keeping the samples in hermetically closed vessels during the period of about 10 half- lives (for  $^{226}$ Ra approximately 40 days), this equilibrium may be again established before the measurements are performed. Since the natural isotope ratio of uranium is a constant, the gamma- rays of  $^{235}$ U may be used to measure the amount of  $^{238}$ U. However, a small isotopic abundance of  $^{235}$ U results in low intensity of these lines, which also form doublets with lines from another natural series.

There are some 50 gamma-rays of sufficient intensity emitted in the decay of the whole <sup>238</sup>U series. Figure 1. shows the  $\gamma$ -ray spectrum of the soil from Kalna. However, only 2 percent of the emitted  $\gamma$ -rays originate from the decays which precede <sup>226</sup>Ra, i.e., during the equilibrium with <sup>238</sup>U. Table 2. gives a short list of the most intensive  $\gamma$ -rays emitted by the first <sup>238</sup>U daughters the <sup>234</sup>Th and <sup>234</sup>Pa.

**Table 2.** The most intense  $\gamma$ -rays of first daughter nuclei of <sup>238</sup>U.  $E_{\gamma}$  is the energy of the transition and  $I_{\gamma}$  is a number of emitted  $\gamma$ -rays per disintegration (absolute intensity).

Nucleus	$E_{\gamma} \; [\text{keV}]$	$I_{\gamma}$ [%]
	63.3	3.8
$^{234}$ Th	92.3	2.7
	92.8	2.7
	76.6	0.4
$^{234}$ Pa	98.4	0.2
	1001	0.9

As we see, the most intensive  $\gamma$ -rays in the spectrum of <sup>234</sup>Th are of low energy and are thus not convenient for analysis, considering the problem of efficiency and interference of a great number of lines from other nuclides and characteristic X-rays of the elements from detector surrounding. The line of 1001 keV of <sup>234</sup>Pa is much more amenable to detection, but is of low intensity.

The line of 186 keV is one of the most intense lines of the uranium decay chain. Actually, it is a doublet consisting of two lines: 186.1 keV from the decay of  $^{226}$ Ra and 185.7 keV from the decay of  $^{235}$ U. In the case of equilibrium in the above-mentioned series, these two components contribute equally to the intensity of this instrumentally inseparable doublet. The fraction of the intensity of the 186 keV line originating from  $^{235}$ U, which measures the contents of  $^{235}$ U, or actually  $^{238}$ U, can be obtained by measuring the activity of  $^{226}$ Ra from post-radon analytical lines (in equilibrium).

The decays of post-radon members of the  $^{238}$ U series yield many strong  $\gamma$  transitions susceptible to analytical purpose, for example intense lines of  $^{214}$ Bi: 609.3 keV, 1120.3 keV and 1764.5 keV which do not interfere with lines of other natural series. When radon is in equilibrium, the intensities of these lines give direct information about the concentration of  $^{226}$ Ra.



Figure 1. Gamma-ray spectrum of the soil from Kalna.

Considering these features of radioactive series and the peculiarities of the of gammaray spectrometry technique, essential strategies for measuring the low concentrations of natural uranium and their variations in environmental samples can be developed [5, 6].

First of all, the samples must be kept in hermetically closed vessels for at least 40 days in order to reach the equilibrium due to radon emanation. The relative method is often used for detecting small variations of uranium concentration in samples of similar characteristics (for example, following the accumulation of uranium in soil due to application of fertilizers). By this method, all the samples are measured under the same geometrical conditions and only eventual variations in density of samples give rise to systematic error.

In general, there are three principal methods to estimate the activity of  $^{238}$ U from the complete spectroscopy information realized under the conditions defined above:

- 1. Using all the analytical lines, by assumption of equilibrium in the whole series, yields the statistically best-defined result. Degree of interruption of equilibrium within the limits of measurement uncertainty can be expressed by the statistical treatment of deviations of the activities from the mean value obtained from analytical lines of various radionuclides. It is the only way to express the activity of the whole <sup>238</sup>U series. However, the result of this procedure predominantly depends upon the concentration of <sup>226</sup>Ra in the sample, due to better statistically defined lines of postradon participants in the <sup>238</sup>U chain.
- 2. Determination of the <sup>238</sup>U concentration from the intensity of the 186 keV line. Contribution of <sup>226</sup>Ra to the intensity of this line may be precisely computed from the concentration of <sup>226</sup>Ra as determined from post-radon lines. Eventual variation of radon concentration in the measuring chamber and the resulting wrong estimation of the background as well as imperfect closing of the vessels can give rise to systematic errors. Concentration of <sup>235</sup>U may be calculated from the known concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and the total intensity of 186 keV line, and the concentration of <sup>226</sup>Ra in the sample and <sup>226</sup>Ra in the sample and the concentration of <sup>226</sup>Ra in the sample and the concentrat

 $^{238}\mathrm{U}$  then ceforth, considering the constant isotopic abundance of  $^{235}\mathrm{U}$  in natural uranium.

3. Determination of the concentration of <sup>238</sup>U from the low-energy lines of <sup>234</sup>Th, which is the first daughter of <sup>238</sup>U. Therefore, this provides the most reliable result in spite of the problem of measuring the intensities of low-energy lines. At the energies below 100 keV, the efficiency of detection rapidly decreases and, within lead shieldings, the interference of lead X-rays with the 63.3 keV line is possible. Besides, for relative measurements, different absorption of low energy gamma-rays in the samples of different density could be the source of rather high systematic error (up to some 20 %). However, the computer program OMEGA successfully eliminates most of this error by numerical correction.

According to the presented advantages and disadvantages of these available methods, we can conclude that the combination of the second and the third method provides the most reliable results, considering small variations of  $^{238}$ U concentrations in environmental samples. Uncertainties in the concentration of  $^{238}$ U obtained by this method are estimated to be of the order of 10 %.

# 3. Gamma-ray spectrometry of depleted uranium in soil

The fact that natural uranium is accompanied by its daughter elements, while depleted uranium is separated from the products of its decay, helps distinguish the natural presence of uranium in soil from additional contamination by DU.

When the activity of  $^{238}$ U surpasses its normal activity in the soil (10 Bq/kg 100 Bq/kg) by a factor of ten or higher, gamma-ray spectrometry assessment of depleted uranium is best performed by measuring the low-energy lines of  $^{234}$ Th, which are then sufficiently intense. In that case most of the activity of  $^{238}$ U originates from depleted uranium and the combined method is the most convenient.

The problem becomes more complicated when the activity of  $^{238}$ U from depleted uranium and from natural uranium is comparable. The concentration of natural  $^{238}$ U must be determined only from the activity of  $^{226}$ Ra assuming the radioactive equilibrium between these two isotopes. For the calculating procedure, the empirical factor  $^{238}$ U/ $^{226}$ Ra, which is  $1.3 \pm 0.3$ , must be used [7]. This factor is the mean value of the  $^{238}$ U/ $^{226}$ Ra ratio obtained from more than 100 measurements on soil samples from Vojvodina. Since the isotopic abundance of  $^{235}$ U in depleted uranium is relatively small, determination is only possible by using the post-radon lines, which involves keeping the samples in closed vessels for about 40 days before measurements. The total amount of  $^{238}$ U must be determined from low-energy lines of  $^{234}$ Th, including the above-mentioned problems.

Considering the increased efficiency at low-energies (below 100 keV) the GMX detectors are the most suitable for this purpose.

Activity of  $^{238}$ U from depleted uranium may be obtained as a difference between the activity of  $^{238}$ U and the activity of  $^{226}$ Ra.

#### 4. Results and discussion

When the issue of DU emerged, the experimental advantages of the measuring equipment (GMX type detector with enhanced efficiency below 100 keV, and iron low level shielding) were fully exploited.

Low-level high-resolution  $\gamma$ -ray spectroscopy is a very convenient technique for uranium determination in environmental samples with <sup>238</sup>U activity concentration above 1 Bq/kg. When the issue of DU determination emerged, responding to strong public interest, a method for DU determination was developed.

The radionuclide content of the samples was measured by means of a reversed electrode GMX type HPGe spectrometer made by ORTEC. The nominal efficiency of the detector is 32% and the resolution is 1.9 keV. This detector has a thin lead layer on the outer surface and a beryllium entrance window, thus enabling the detection of  $\gamma$ -rays below 100 keV with high efficiency. The detector was calibrated by means of reference radioactive materials in cylindrical geometry. Matrix effects were taken into account by means of a computer code [8]. The detector was operated inside a 25 cm thick iron shield made from pre II World War cast iron [7]. As compared with lead shields, the iron shields have definite advantages in the low energy region due to the absence of the lead X rays. The typical time of measurement of the samples was 50 ks. The background, which contains the same lines especially post radon lines, is not subtracted.

Characteristic cases of results are shown in Table 3.

SAMPLE	$^{238}$ U [Bq/kg]	$^{226}$ Ra [Bq/kg]	$^{238}\mathrm{U}_D~\mathrm{[Bq/kg]}$	$^{40}$ K [kBq/kg]	$^{232}$ Th [Bq/kg]
LVJ4	$(5.0 \pm 1.3) \cdot 10^3$	$19.8\pm2.1$	$(5.0 \pm 1.3) \cdot 10^3$	$0.53\pm0.04$	$30 \pm 3$
LVJ3	$115\pm15$	$13.2\pm2.3$	$98 \pm 16$	$0.187 \pm 0.022$	$13.5\pm1.1$
LVJ7	$33\pm8$	$22.4\pm2.2$	< 10	$121\pm0.07$	$45\pm3$

Table 3. Activites of characteristic radionuclides in soil.

Specific activities of  $^{238}$ U as calculated from daughters  $^{234}$ Th and  $^{234}$ Pa are given in the second column of Table 3., and the activities of  $^{226}$ Ra calculated from daughters beyond radon:  $^{214}$ Pb,  $^{214}$ Bi and  $^{210}$ Pb are listed in the third column. Calculated values of the activities which may be attributed to contamination by depleted uranium are given in the fourth column. In the fifth and sixth column, the activity of  $^{40}$ K and of the  $^{232}$ Th series are presented. Uncertainties of measurement are given on the 95 % confidence level.

The first sample LVJ4 has a very high activity of  $^{238}$ U which may all be attributed to contamination by depleted uranium and the activity of naturally present uranium may be neglected. The second sample has an activity of  $^{238}$ U which is significantly increased by contamination by depleted uranium. The measurement uncertainty is very small relative to the measurement uncertainty of  $^{226}$ Ra including the uncertainty of the empiric factor of the equilibrium. The third sample is not contaminated and expressed measurement uncertainty equals by order of magnitude the detection limit for  $^{238}$ U contamined by depleted uranium, which is obtained by the above-mentioned method using the equipment described.

## 5. Conclusions

Determination of the activity of depleted uranium in the soil is a complex task, except in the case of very high specific activities. However, determination of medium and low concentrations is needed for the tracing of the migration of radionuclides through the soil and/or if it is necessary to study the effects of decontamination.

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# $0\nu$ Mode in Double Beta Decay of $^{50}\mathrm{Cr}$

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#### Abstract

The intensity of the positron annihilation radiation emitted by a sample of natural chromium trioxide, CrO<sub>3</sub>, containing 4.7 g of <sup>50</sup>Cr, was measured for 30 days. The background was measured with the samples of iron, copper, and no sample at all (air), during a similar measurement time. From the net intensity of the 511 keV line in the HpGe coincidence spectrum, the partial half-life for the positron - electron capture double beta decay of <sup>50</sup>Cr,  $T_{1/2}(\beta^+EC, 2\nu + 0\nu) = 1.3(12) \times 10^{18}$  y at 95 % CL, has been deduced. Due to the ground state spin sequence  $0^+({}^{50}\text{Cr}) \ 6^+({}^{50}\text{Cr}) \ 0^+({}^{50}\text{Ti})$ , the  $2\nu$  mode is much less probable than the  $0\nu$  mode with virtual neutrinos.

Key words: Double beta decay, gamma-spectroscopy

#### 1. Introduction

Two recent claims, the announcement of the SNO collaboration claiming to have proved neutrino oscillations and at the same time to have resolved the solar neutrino problem [1], and the result of the Heidelberg-Moscow experiment claiming to have detected the double beta decay of <sup>76</sup>Ge [2], seem to have finally established the non-zero neutrino mass and greatly stirred the neutrino physics community. Here we will present the results of our measurements of the double beta decay of <sup>50</sup>Cr which either suggests the non-zero result at a confidence level of 95 % or establishes the lower limit for the half-life of this process, one order of magnitude more restrictive than the existing one.

Chromium-50 is one of the four otherwise stable isotopes in the vicinity of the Z, N = 28 magic numbers which are by energy considerations allowed to decay by the simultaneous beta decay of their two protons into the corresponding more stable second neighbor isobars. The four decays in question are:

$${}^{50}_{24}\text{Cr}_{26} \rightarrow {}^{50}_{22}\text{Ti}_{28}, \quad {}^{54}_{26}\text{Fe}_{28} \rightarrow {}^{54}_{24}\text{Cr}_{30}, \quad {}^{58}_{28}\text{Ni}_{30} \rightarrow {}^{58}_{26}\text{Fe}_{32}, \quad {}^{64}_{30}\text{Zn}_{34} \rightarrow {}^{64}_{28}\text{Ni}_{36}$$
(1)

It is seen that the first and last of the cited decays complete the magic number 28 of either neutrons or protons, while the second and the third destroy it. It can thus be expected that due to the structure effects those which complete the magic number are enhanced as compared to those which destroy it. Some evidence to support this point is given in [3].

Out of the four, Nickel-58 has been measured with good sensitivity by Vasilev et. al. [4], while we have recently measured with high sensitivity, and a negative result, the decay of Iron-54 [5], as well as the decay of Zinc-64 [6], this last one with a hint of a positive result even at the 99.7 % CL. The decay of Chromium-50 has been measured by Norman back in 1985 [7], [8], with the sensitivity lower than the other three decays and, minding its possible enhancement similar to that in the case of Zinc-64, we set out to measure it with a sensitivity comparable to that of the other decays.

## 2. Measurements and result

The decay scheme of  ${}^{50}$ Cr is presented in Fig.1. The low Q-value of the decay allows for all types of electron capture - electron capture decays as well as for the electron - capture - positron decays, while the positron - positron decay is forbidden. If the decay is to be measured by positron annihilation radiation only, the partial probability for the positron - electron capture decay will be deduced. It is perhaps also worth commenting that this is probably the optimum range of atomic numbers for this type of decay for study - neither too low, from the view of electron capture, nor too high, from the view of the positron decay. The small phase-space factor for this particular decay may hopefully be at least partially compensated for by some enhancement due to the above mentioned structure effects.



Figure 1. The  $(0\nu + 2\nu)$  decay scheme of <sup>50</sup>Cr.

Since the energy of the first excited state in the daughter nucleus is higher than the Q-value the decay is possible only to its ground state, and since the spin of the intermediate nucleus is very high (6<sup>+</sup>), and the real low-energy neutrinos can not carry high angular momentum while the exchange of a high momentum virtual neutrino between the closely spaced decaying protons is encouraged by the uncertainty principle, the decay would practically be only neutrinoless. The measurement of the emission rate of the positron annihilation radiation in this case thus determines the partial decay constant for the positron - electron capture neutrinoless decay mode only.

The measurement was performed by a technique similar to that used in the case of  $^{64}$ Zn. The 209 g CrO<sub>3</sub> sample containing 4.7 g  $^{50}$ Cr is sandwiched between the  $(3 \times 3)^{\circ}$  NaI(Tl) and 25 % HpGe detector, both placed coaxially in the hole of the  $(9 \times 9)^{\circ}$  NaI(Tl) annular veto detector. The whole detector arrangement is situated on the ground level, within a 1 m<sup>3</sup> shielding chamber made out of the pre WWII iron, with 25 cm thick walls. The use of iron as the background reducing passive shielding has for the present purpose significant advantages over lead shielding. Firstly, the rate of positron production by cosmic-rays at near sea-level is found to depend on the square of the atomic number of the medium divided by its mass number [9], which here gives the advantage of 2.7 as compared to lead. Secondly, as is nicely exemplified by the absence in our single HpGe background spectrum of the neutron induced features which abound in lead shielded detectors [10], the neutron production by cosmic-rays is at an undetectable level, couple orders of magnitude below that within the lead shieldings.

The experimental setup is schematically presented in Fig.2. To minimize accidental coincidences, the two detectors which sandwich the sample are connected in a fast-slow coincidence circuit, while to reject the events generated by the environmental and cosmic radiations, the annular guard detector operates in anticoincidence. The intensity of the 511 keV line in the coincidence-anticoincidence spectrum of the HpGe detector, as analyzed by ADC1, thus measures the number of positrons generated and annihilated within the sample while the contribution of the events generated by radiations coming from the outside are minimized. The coincidence efficiency of the system,  $\varepsilon = R_c/A$ , where  $R_c$  is the coincidence rate and A is the positron activity of the sample, is determined both by the Monte Carlo simulation, which was also used to find the optimum sample thickness, and by direct measurements with the <sup>22</sup>Na calibrated source, and was found to amount to 1.9 %.



Figure 2. The experimental setup for the  ${}^{50}$ Cr decay measurements.

The question of background is of course the most important one. It was determined in three separate measurements: with no material at the sample position, and with samples of iron and copper (of a size equivalent to that of the chromium oxide sample). The overall measurement time was 26 days. The important and rather unexpected result is that no statistically significant difference between these measurements in the intensity of the 511 keV peak has been found. Average background counting rate in the 511 keV line, based on the sum spectrum of the three background measurements, at the 68 % CL is  $97(7) \text{ cc}/10^6$ s. We shall further address the problem of background when discussing our final result.

In Fig.3 the count rates in the region of the 511 keV line for both the chromium sample and the background are presented. Their difference is depicted in the lower part of the figure, with the corresponding net count rate at the 68 % CL of 22(10) cc/10<sup>6</sup>s in the 511 keV line, meaning that we have the non-zero result even at the 95 % CL. If we ascribe this rate to the neutrinoless positron-electron capture decay of <sup>50</sup>Cr we arrive at the partial half-life for this process of  $T_{1/2}(\beta^+ EC, 0\nu) = 1.3(6) \times 10^{18}$  y at the 68 % CL.



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Figure 3. The chromium and the sum background spectrum: solid-Cr, open-background with their difference presented in the lower inset.

**Figure 4.** Same as Figure 3. but with the chromium spectrum shifted one channel to the left.

#### 3. Discussion

The quoted result calls for a number of comments. Some of them deal with the quality of the raw data as presented in Fig.3, while others deal with possible systematic errors in the applied background determination procedure. Closer inspection of our Fig.3 raises some doubts about the stability of the spectrum during the measurements, for the 511 keV line in the chromium spectrum looks somewhat broader and/or shifted in respect of the background line. Also their difference, though the statistics is poor, does not seem centered around 511 keV. To check for the influence of possible spectrum instability on our result, the same spectra are presented in Fig.4, this time with the chromium spectrum shifted one channel to the left, together with their difference at the bottom of the figure. The net intensity in the 511 keV line is now  $20(11) \text{ cc}/10^6$ s, which seems to dismiss the doubts raised above.

When the background determination is concerned, the objection that some other source of positrons different than the double beta decay might exist in the chromium sample, which does not exist in the three background cases, seems well justified. However, it is certainly most reassuring in this respect that significant differences between the positron annihilation rates in the three background measurements were not found. This suggests that the majority of background positrons is created by the radiations arriving from the outside which escape the vetoing procedure (mostly by the muons surviving the 5  $\mu$ s long anticoincidence gate), and that the greater production in the material samples is within statistical errors compensated by the correspondingly greater self-absorption of the annihilation radiation in the samples. The closeness of the atomic numbers of all the samples raises hopes that the same will hold for the chromium sample as well and that the excess is in this case indeed due to the double beta decay of Chromium-50.

One may also be quite confident that other conceivable positron sources do not exist in measurable quantities in the chromium sample. As is well known, positron emitters do not exist in natural radioactivities, except weakly in <sup>40</sup>K, whose 1460 keV gamma-ray, however, does not seem more intense than in the background spectra. The intensity of the omnipresent highest energy 2614.6 keV gamma-ray, which is capable of pair production, is also not found in excess over that in the background spectra. At the same time this means that the troubling 510.8 keV gamma-ray from the same decay, which only weakly, if at all, interfers with the 511 keV line in the coincidence spectra, does not contribute excessively to the intensity of the 511 keV line. All the nuclear reactions possibly induced by the low and high-energy environmental particles which may yield positron emitters are also easily estimated to have negligible contributions. For instance, the most likely reaction to produce positron emitters, the (n, 2n) reaction, is known to have cross sections at most in the barn range for the isotopes we are dealing with [11]. Since, as already discussed, the neutron fluxes are much lower in the iron environment than in the higher Z environments, and neutron signatures are indeed not present in our background spectra, an upper limit for the (n, 2n) reaction inducing neutron flux is at most, according to [10], of the order of  $1 n/m^2$ s. The rate of this reaction would thus be  $\approx 10^{-28} s^{-1}$ , what is at least two orders of magnitude behind the decay rate which we observed.

Our experimental result obtained from positron annihilation is not sensitive to the double decay mode  $(0\nu \text{ or } 2\nu)$ . However, the ground state spin sequence favors the  $0\nu$  mode. Thus we can not exclude the contribution of the  $0\nu$  mode to the measured transition probability.

Future measurements of this decay would have to pay more attention to further reduction of the annihilation radiation background, on top of pushing all the conventional means to the limit, by additionally using the much longer anticoincidence gating times from the guard detector.

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# Application of Graph Theory in the Microscopic Theory of Quantum Wires

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#### Abstract

The subject of the study are the elementary excitations in the rectangular quantum wire with "free" surfaces. We shall study a completely general quadratic harmonic Hamiltonian where the quasiparticle kinematics (Fermi or Bose) is irrelevant due to the use of the single-particle wave-function. We offer a "nonstandard" approach to the solution of the eigenvalue problem within the framework of the graph theory. The eigenvalue problem for the adjacancy matrix of the rectangular lattice graph  $P_{n+1} \times P_{m+1}$  was solved. The explicit formulas for energy levels and the wave-functions of arbitrary elementary excitation are obtained. Some specific analytical expressions are also presented. Quantum wires are perspective candidates for the design of the next generation of integrated circuits, as soon as the nature of the influence of quantum effects is sufficiently well understood.

*Key words:* Quantum wires, lattice graphs, single particle wave-function, eigenvalue problem.

# 1. Introduction

Low-dimensional systems or nanostructures have been the subjects of intensive investigations both from the experimental and theoretical point of view [1-5]. Such systems include so-called two-dimensional systems, or thin films and superlattices, one-dimensional systems, or quantum wires, and zero-dimensional systems, or quantum dots. Quantum size effects in these structures lead to new material properties which promise smaller, faster and lower-power-consuming devices [6].

In recent years quantum wires (QW) have been the subject of numerous theoretical and experimental studies. In such structures the particles (charge carriers) are confined to move along the length of the wire and the motion is quantised in the transverse directions. We propose here to formulate a completely general theory based on the quadratic harmonic Hamiltonian as formulated in terms of Fermi or Bose operators describing arbitrary elementary excitations (quasiparticles).

The model calculations of energy levels and wave-function in a QW have been performed using the envelope function approximation or variational approach [7-13] etc. In contrast to these semi-phenomenological approaches, where the wave-function has been calculated in the continuum approximation, we use the microtheoretical approach and techniques exploited in papers [14-17] dealing with thin films.

Another novelty is that in this communication we apply the graph theory to calculate the energy levels and wave-functions in the case of rectangular QW with free surfaces. This approach proves to be an elegant and simple method for microtheoretical calculations of quantum wires and has not been, to our knowledge, applied to these problems, previously.

#### 2. Hamiltonian of the model and lattice graphs

We consider the quantum wire, denoted by  $W_{N_x,N_y,\infty}$ , which is "cut-out" from a tetragonal bulk structure with lattice constant  $c_x, c_y, c_z$ . Figure 1 displays the geometry of the quantum wire  $W_{2,2,\infty}$ .



**Figure 1.** Rectangular quantum wire  $W_{2,2,\infty}$ .

The general quadratic harmonic Hamiltonian for the quantum wires has the following form [18]:

$$\hat{H} = \sum_{n_x n_y n_z n'_x n'_y n'_z} \hat{O}_{n_x n_y n_z; n'_x n'_y n'_z}$$
(1)

where:

 $\hat{O}_{n_x n_y n_z; n'_x n'_y n'_z} = D_{n_x n_y n_z; n'_x n'_y n'_z} a^+_{n_x n_y n_z} a_{n_x n_y n_z} + V_{n_x n_y n_z; n'_x n'_y n'_z} a^+_{n_x n_y n_z} a_{n'_x n'_y n'_z} a^+_{n_x n_y n_z} a_{n'_x n'_y n'_z} a_{n_x n_y n_z} a_{n_x n_$ 

With regard to the definition of  $W_{N_x,N_y,\infty}$  we assume

$$\begin{split} 0 &\leq n_x \leq N_x \; ; \quad n_x \sim 1 \div 100 \\ 0 &\leq n_y \leq N_y \; ; \quad n_y \sim 1 \div 100 \\ -\frac{N_z}{2} &\leq n_z \leq \frac{N_z}{2} \; ; \quad n_z \sim 10^8 \; . \end{split}$$

 $a^+$  and a are are either Fermi or Bose operators of creation and annihilation of the arbitrary elementary excitations in the Wannier representation; D and V are the matrix elements characterising the self-energy of elementary excitations and transfer from site  $n \equiv (n_x, n_y, n_z)$  to site  $n' \equiv (n'_x, n'_y, n'_z)$ , respectively. Clearly, these matrix elements depend on the considered type of elementary excitations in QW. Here, we accept the nearest neighbors' approximation, so that the Hamiltonian (1) becomes:

$$\hat{H} = \sum_{n_x n_y n_z} \left( \hat{O}_{n_x n_y n_z; n_x + 1 n_y n_z} + \hat{O}_{n_x n_y n_z; n_x - 1 n_y n_z} + \hat{O}_{n_x n_y n_z; n_x n_y + 1 n_z} + \hat{O}_{n_x n_y n_z; n_x n_y - 1 n_z} + \hat{O}_{n_x n_y n_z; n_x n_y n_z + 1} + \hat{O}_{n_x n_y n_z; n_x n_y n_z - 1} \right).$$

$$(2)$$

We shall investigate the system described by the Hamiltonian (2) by calculating the single particle wave-function

$$|\Psi\rangle = \sum_{n_x n_y n_z} A_{n_x n_y n_z} a^+_{n_x n_y n_z} |0\rangle , \quad \sum_{n_x n_y n_z} |A_{n_x n_y n_z}|^2 = 1.$$
(3)

Unknown coefficients  $A_{n_x n_y n_z}$  (amplitudes) are obtained by calculating Heisenberg's equation of motion for the operator  $a_{n_x n_y n_z}$ 

$$(Ea_{n_x n_y n_z} - [a_{n_x n_y n_z}, \hat{H}])|\Psi\rangle = 0, \quad E = \hbar\omega,$$
(4)

assuming simple time-dependence  $a_{n_x n_y n_z}(t) = a_{n_x n_y n_z}(0)e^{-i\omega t}$ .

By substituting Eq. (3) into Eq. (4), we obtain the system of equations for determination of the coefficients  $A_{n_xn_yn_z}$ . It is important to notice that for the single-particle functions, the operator kinematics is not essential, so the system of equations for the coefficients is identical for both Fermi and Bose operators. Since there is a translational symmetry breaking in the directions x and y, we perform the following substitution for the coefficients  $A_{n_xn_yn_z}$  in the system Eqs. (4):

$$A_{n_x n_y n_z} = \phi_{n_x n_y} e^{ik_z n_z c_z}.$$
(5)

The solution of the system is now obtained by solving the following eigenvalue problem:

$$C\mathbf{\Phi} = r\mathbf{\Phi},\tag{6}$$

where the unknown eigenvector  $\mathbf{\Phi}$  is  $[\phi_{00}, \phi_{01}, ..., \phi_{mn}]^T$  and the matrix C is the adjacancy matrix of the rectangular lattice graph  $P_{n+1} \times P_{m+1}$ , Figure 2. Thus, coefficients  $\phi_{n_x n_y}$  in Eq. (5) are components of the eigenvector  $\mathbf{\Phi}$  (for corresponding eigenvalue r) in Eq. (6) which satisfy Eqs. (5) and (3) i.e.:

$$\sum_{n_x n_y} \phi_{n_x n_y}^2 = \frac{1}{N^*} , \quad (N^* = 2\lfloor \frac{N_z}{2} \rfloor + 1 \sim N_z)$$
(7)

since  $\sum_{n_x n_y n_z} |A_{n_x n_y n_z}|^2 = N^* \sum_{n_x n_y} \phi_{n_x n_y}^2 = 1.$ 



**Figure 2.** The rectangular lattice graph  $P_{n+1} \times P_{m+1}$ .

We denote by  $B_{n+1}$  the square matrix of order (n+1):

$$B_{n+1} = \begin{bmatrix} 0 & 1 & 0 & \cdots & 0 & 0 \\ 1 & 0 & 1 & \cdots & 0 & 0 \\ 0 & 1 & 0 & \cdots & 0 & 0 \\ & & & \ddots & & \\ 0 & 0 & 0 & \cdots & 0 & 1 \\ 0 & 0 & 0 & \cdots & 1 & 0 \end{bmatrix}$$
(8)

which represents the adjacancy matrix of the path  $P_{n+1}$  of length n shown in Figure 3.

 $0 \qquad 1 \qquad 2 \qquad \qquad n-1 \qquad n$ 

**Figure 3.** The path  $P_{n+1}$  of lenght n.

The matrix C (of order (n+1)(m+1)) can be written as a square block matrix of order (m+1) of the form

$$C = \begin{bmatrix} B & I & O & \cdots & O & O \\ I & B & I & \cdots & O & O \\ O & I & B & \cdots & O & O \\ & & & \ddots & & \\ O & O & O & \cdots & B & I \\ O & O & O & \cdots & I & B \end{bmatrix}$$
(9)

where B denotes the matrix  $B_{n+1}$ , I and O denote the unit matrix  $I_{n+1}$  of order n+1and null matrix  $O_{n+1}$  of order n+1, respectively.

Eigenvalues r of the equation (6) are in the following way related to the energy levels of the excitations in QW:

$$rV = E - 4D - 2V\cos(c_z k_z). \tag{10}$$

Energy levels and wave-function of the QW will be calculated in the next section.

### 3. Energy levels and wave-functions

Note that the matrix C can be written as:

$$C = B_{m+1} \otimes I_{n+1} + I_{m+1} \otimes B_{n+1}, \tag{11}$$

where  $\otimes$  denotes the Kronecker product of matrices [19], defined by:

$$A_{m_{1}\times m_{2}} \otimes B_{n_{1}\times n_{2}} = \begin{bmatrix} \alpha_{11}B & \alpha_{12}B & \dots & \alpha_{1m_{2}}B \\ \alpha_{21}B & \alpha_{22}B & \dots & \alpha_{2m_{2}}B \\ \ddots & \ddots & \ddots & \ddots \\ \vdots & \ddots & \ddots & \ddots \\ \alpha_{m_{1}1}B & \alpha_{m_{1}2}B & \dots & \alpha_{m_{1}m_{2}}B \end{bmatrix}$$
(12)

where  $\alpha_{ij}$   $(i = 1, ..., m_1; j = 1, ..., m_2)$  denote elements of the matrix  $A_{m_1 \times m_2}$ . Let the matrix  $B_{n+1}$  have eigenvalues  $\lambda_1^{(n+1)}, \ldots, \lambda_{n+1}^{(n+1)}$  with eigenvectors  $b_1^{(n+1)}, \ldots, b_{n+1}^{(n+1)}$ , and let the matrix  $B_{m+1}$  have eigenvalues  $\lambda_1^{(m+1)}, \ldots, \lambda_{m+1}^{(m+1)}$  with eigenvectors  $b_1^{(m+1)}, \ldots, b_{m+1}^{(m+1)}$ . Then  $b_i^{(m+1)} \otimes b_j^{(n+1)}$   $(1 \le i \le m+1, 1 \le j \le n+1)$  is an eigenvector (m+1). of C with eigenvalue  $\lambda_i^{(m+1)} + \lambda_j^{(n+1)}$ , since

$$(B_{m+1} \otimes I_{n+1} + I_{m+1} \otimes B_{n+1})(b_i^{(m+1)} + b_j^{(n+1)}) =$$
  
=  $(B_{m+1}b_i^{(m+1)} \otimes I_{n+1}b_j^{(n+1)}) + (I_{m+1}b_i^{(m+1)} \otimes B_{n+1}b_j^{(n+1)}) =$   
+  $\lambda_i^{(m+1)}(b_i^{(m+1)} \otimes b_j^{(n+1)}) + \lambda_j^{(n+1)}(b_i^{(m+1)} \otimes b_j^{(n+1)}) = (\lambda_i^{(m+1)} + \lambda_j^{(n+1)})(b_i^{(m+1)} \otimes b_j^{(n+1)}) .$ 

It is not difficult to determine the eigenvalues and eigenvectors of  $B_{n+1}$  (and simultaneously of  $B_{m+1}$  by replacing n with m). The eigenvalues of  $B_{n+1}$  are [20]:

$$\lambda_j^{(n+1)} = 2\cos(\frac{\pi j}{n+2}), \ (j = 1, \dots, n+1).$$
(13)

The corresponding eigenvectors are:

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$$b_j^{(n+1)} = [C_n(\lambda_j^{(n+1)}), \dots, C_2(\lambda_j^{(n+1)}), C_1(\lambda_j^{(n+1)}), C_0(\lambda_j^{(n+1)})]^T, \ (j = 1, \dots, n+1), \ (14)$$

where  $C_k(\lambda)$  denotes the k-th member of the sequence of Chebyshev polynomials of the second type [21] defined in the following recursive manner:

$$C_0(\lambda) = 1, \ C_1(\lambda) = \lambda, \ C_k(\lambda) = \lambda C_{k-1}(\lambda) - C_{k-2}(\lambda) \text{ for } k \ge 2.$$
 (15)

For  $|\lambda| < 2$  they are:

$$C_k(\lambda) = \frac{2^{-(k+1)}}{\sqrt{\lambda^2 - 4}} \left[ \left(\lambda + \sqrt{\lambda^2 - 4}\right)^{k+1} - \left(\lambda - \sqrt{\lambda^2 - 4}\right)^{k+1} \right] = \frac{\sin(k+1)\theta}{\sin\theta}$$
(16)

where  $\lambda = 2\cos\theta$ .

Substituting Eqs. (13) and (16) into Eq. (14) we obtain

$$b_{j}^{(n+1)} = \left[\frac{\sin\frac{(n+1)j\pi}{n+2}}{\sin\frac{j\pi}{n+2}}, \frac{\sin\frac{nj\pi}{n+2}}{\sin\frac{j\pi}{n+2}}, \dots, \frac{\sin\frac{2j\pi}{n+2}}{\sin\frac{j\pi}{n+2}}, \frac{\sin\frac{j\pi}{n+2}}{\sin\frac{j\pi}{n+2}}, \frac{\sin\frac{j\pi}{n+2}}{\sin\frac{j\pi}{n+2}}, \right]^{T}.$$
(17)

Taking into account the above mentioned, we obtain (n + 1)(m + 1) eigenvalues (not necessarily different) (6) in the following form  $(m \equiv N_x, n \equiv N_y)$ :

$$r = r^{(ij)} = 2\cos\frac{\pi i}{m+2} + 2\cos\frac{\pi j}{n+2}; \quad i = 1, \dots, m+1; \quad j = 1, \dots, n+1.$$
(18)

Let us denote the coefficients of the vector  $\mathbf{\Phi}^{(\mathbf{ij})}$ , corresponding to  $r^{(ij)}$ , by  $\phi_{st}^{(ij)}$ ,  $s = 0, \ldots, m; t = 0, \ldots, n$  i.e.:

$$\boldsymbol{\Phi} = \boldsymbol{\Phi}^{(ij)} = [\phi_{00}^{(ij)}, \phi_{01}^{(ij)}, \dots, \phi_{0n}^{(ij)}, \phi_{10}^{(ij)}, \phi_{11}^{(ij)}, \dots, \phi_{1n}^{(ij)}, \dots, \phi_{m1}^{(ij)}, \phi_{m2}^{(ij)}, \dots, \phi_{mn}^{(ij)}]^T.$$
(19)

Since  $\mathbf{\Phi}^{(ij)} = (b_i^{(m+1)} \otimes b_j^{(n+1)})$ , then from Eq. (17) we obtain:

$$\phi_{s,t}^{(ij)} = \frac{1}{\sin\frac{i\pi}{m+2} \cdot \sin\frac{j\pi}{n+2}} \sin\frac{(m-s+1)i\pi}{m+2} \cdot \sin\frac{(n-t+1)j\pi}{n+2}, \quad (20)$$

for s = 0, ..., m and t = 0, ..., n.

Note that if the greatest common divisor of n+2 and m+2 is greater than or equal to 3, then there exists more than one common eigenvalue of matrices  $B_{m+1}$  and  $B_{n+1}$ . Then the algebraic multiplicitie of some eigenvalues of the matrix C are greater than one. It means that the dimension of the eigenspace (the set of all eigenvectors) corresponding to an eigenvalue of C can be greater than one.

Thus the coefficients  $\phi_{n_x n_y}$  in Eq. (5) corresponding to an eigenvalue  $r^{(ij)}$  of C, are the coefficients of the vectors from the intersection of the corresponding eigenspace and the (n+1)(m+1)-dimensional sphere with the center in the origin and radius  $\frac{1}{\sqrt{N^*}}$ .

Finally we obtain the energy level of the system (for i and j fixed):

$$E = 4D + 2V(\cos c_x k_x + \cos c_y k_y + \cos c_z k_z), \qquad (21)$$

where:

$$c_x k_x = \frac{\pi i}{m+2}, \ c_y k_y = \frac{j\pi}{n+2}, \ i = 1, \dots, m+1, \ j = 1, \dots, n+1$$

are components of wave vectors along x and y directions in the QW.

For illustration we exhibit two examples for n = 1, m = 2 and n = 1, m = 1 (see Figure 4).



**Figure 4.** The rectangular lattice graphs  $P_2 \times P_3$  and  $P_2 \times P_2$ .

#### a) Example I.

In the first case, the matrix C ( $C = B_3 \otimes I_2 + I_3 \otimes B_2$ ) is the following block-matrix:

$$C = \left[ \begin{array}{ccc} B_2 & I_2 & O_2 \\ I_2 & B_2 & I_2 \\ O_2 & I_2 & B_2 \end{array} \right]$$

where matrices  $B_2$ ,  $I_2$  and  $O_2$  are the following matrices:  $\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$ ,  $\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$  and  $\begin{bmatrix} 0 & 0 \\ 0 & 0 \end{bmatrix}$ , respectively. Eigenvalues of  $B_3$  are  $\lambda_1^{(3)} = \sqrt{2}$ ,  $\lambda_2^{(3)} = 0$ ,  $\lambda_3^{(3)} = -\sqrt{2}$  with corresponding eigenvectors  $b_1^{(3)} = [1, \sqrt{2}, 1]^T$ ,  $b_2^{(3)} = [-1, 0, 1]^T$  and  $b_3^{(3)} = [1, -\sqrt{2}, 1]^T$ , respectively and eigenvalues of  $B_2$  are  $\lambda_1^{(2)} = 1$  i  $\lambda_2^{(2)} = -1$  with corresponding eigenvectors  $b_1^{(2)} = [1, 1]^T$  and  $b_2^{(2)} = [-1, 1]^T$ . Now, eigenvalues of C are:

$$r^{(11)} = \lambda_1^{(3)} + \lambda_1^{(2)} = 1 + \sqrt{2}, \ r^{(12)} = \lambda_1^{(3)} + \lambda_2^{(2)} = -(1 - \sqrt{2}), \ r^{(21)} = \lambda_2^{(3)} + \lambda_1^{(2)} = 1$$
$$r^{(22)} = \lambda_2^{(3)} + \lambda_2^{(2)} = -1, \\ r^{(31)} = \lambda_3^{(3)} + \lambda_1^{(2)} = (1 - \sqrt{2}), \\ r^{(32)} = \lambda_3^{(3)} + \lambda_2^{(2)} = -(1 + \sqrt{2})$$

with corresponding eigenvectors:

$$\Phi^{(11)} = [1, 1, \sqrt{2}, \sqrt{2}, 1, 1]^T, \ \Phi^{(12)} = [-1, 1, -\sqrt{2}, \sqrt{2}, -1, 1]^T, \ \Phi^{(21)} = [-1, -1, 0, 0, 1, 1]^T$$
  
$$\Phi^{(22)} = [1, -1, 0, 0, -1, 1]^T, \\ \Phi^{(31)} = [1, 1, -\sqrt{2}, -\sqrt{2}, 1, 1]^T, \\ \Phi^{(32)} = [-1, 1, \sqrt{2}, -\sqrt{2}, -1, 1]^T.$$
  
The energies of this system (for *i* and *j* fixed) are

$$E^{(11)} = 4D + V[1 + \sqrt{2} + 2\cos(c_z k_z)]; \quad E^{(32)} = 4D - V[1 + \sqrt{2} - 2\cos(c_z k_z)]$$
$$E^{(12)} = 4D - V[1 - \sqrt{2} - 2\cos(c_z k_z)]; \quad E^{(31)} = 4D + V[1 - \sqrt{2} + 2\cos(c_z k_z)]$$
$$E^{(21)} = 4D + V[1 + 2\cos(c_z k_z)]; \quad E^{(22)} = 4D - V[1 - 2\cos(c_z k_z)].$$

# b) Example II.

The case n = 1 and m = 1 was the topic of paper [18]. Since the corresponding eigenproblem was not correctly solved there, we call attention to this case again. The eigenvalues of the matrix  $C = \begin{bmatrix} B_2 & I_2 \\ I_2 & B_2 \end{bmatrix}$  are:  $r^{(11)} = \lambda_1^{(2)} + \lambda_1^{(2)} = 2, \ r^{(12)} = \lambda_1^{(2)} + \lambda_2^{(2)} = 0, \ r^{(21)} = \lambda_2^{(2)} + \lambda_1^{(2)} = 0, \ r^{(22)} = \lambda_2^{(2)} + \lambda_2^{(2)} = -2$  with corresponding eigenvectors:

$$\Phi^{(11)} = [1, 1, 1, 1]^T, \Phi^{(12)} = [-1, 1, -1, 1]^T, \Phi^{(21)} = [-1, -1, 1, 1]^T, \Phi^{(22)} = [1, -1, -1, 1]^T,$$

respectively. Both the algebraic and geometric multiplicities of the eigenvalue zero are 2 and the corresponding eigenspace is the linear combination  $\varphi(0) = \{\lambda \Phi^{(12)} + \mu \Phi^{(21)} \mid \lambda, \mu \in \Re\}$ . The coefficients  $\phi_{n_x n_y}$  in Eq. (5) are coefficients of an arbitrary vector from the intersection  $\varphi(0) \cap S_4$ , where  $S_4$  is the sphere  $\{(x_{00}, x_{01}, x_{10}, x_{11}) \in \Re^4 \mid x_{00}^2 + x_{01}^2 + x_{10}^2 + x_{11}^2 = \frac{1}{N^*}\}$ . In Ref. [18] the authors neglected that the eigenspace for the eigenvalue zero is a two-dimensional subspace of  $\Re^4$  and dealt with the vector  $\frac{1}{2\sqrt{N^*}}[1, 0, 0, -1]^T$  (in our notation) as the unique solution of equation (5) for r = 0 satisfying Eq. (7). Therefore their conclusions are not generally valid.

# 4. Conclusion

We have used in this paper the single-particle wave-function to determine the energy levels of arbitrary elementary excitations described by the quadratic harmonic Hamiltonian in a quantum wire. All our calculations are based on zero-order approximation, when the boundary variations of intermolecular interactions can be neglected, i.e. in the case of free surfaces. The results were obtained by the application of graph theory to solve the corresponding eigenvalue problem. It turned out to be highly efficient, elegant and rather simple to use, once the connection with the particular physical problem is established.

The more general study of QW demands the application of rather sofisticated numerical methods of the physics of low-dimensional systems. The results obtained are valid for an arbitrary number of layers along X and Y directions. We stress that in the case of extremely thin QW we are able to reproduce rather easily, by this approach, all of the results from paper [18]. The examination of many other interesting phenomena which appear in these structures is possible only by the utilization of very cumbersome numerical calculations, so it will be presented separately.

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# Ferrofluid Lubrication of a Porous Bearing with its Slider Having a Convex Pad Surface Considering Slip Velocity

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#### Abstract

We studied here the ferrofluid lubrication of a porous bearing with its slider having a convex pad surface considering slip velocity at the interface of film and porous matrix. Our aim was to study, especially, the effect of slip velocity on the bearing characteristics. Expressions were obtained for dimensionless pressure, load capacity, friction on the slider, coefficient of friction and the position of the centre of pressure. Increase in the slip parameter caused decrease in load capacity, friction and in the coefficient of friction. But, increase in the permeability parameter caused decrease in load capacity and friction and increase in coefficient of friction. Both the parameters could not significantly affect the position of the centre of pressure.

Key words: Ferrofluid, lubrication, slider, convex pad, porous, slip velocity

# 1. Introduction

Puri and Patel [1] analysed a porous slider bearing with a convex pad surface and found that the load capacity, friction on the slider and the coefficient of friction were greater in this case than those of the corresponding bearing with a flat surface [2]. A convex pad surface can be centrally pivoted so that the bearing can operate stably under varying operating conditions and can be used in marine and other applications.

Ferrofluid lubrication of porous slider bearings were considered by Agrawal [3] as well as Ram and Verma[4]. They found that the use of ferrofluid as a lubricant increased the load capacity of such bearings without altering the friction on their sliders. Recently, Shah et al. [5,6] considered the effect of magnetic fluid lubricant on the squeeze films between curved porous rotating circular plates and between porous annular curved plates. All the above investigators assumed that there was no slip at the porous interface. Sparrow et al [7] and Beavers and Joseph [8] showed that such assumptions could not hold at the nominal boundary of a naturally permeable material. In this paper our aim was to study the effects of slip velocity on a porous slider bearing with a convex pad surface lubricated with a ferrofluid.

## 2. Analysis

The bearing shown in Fig.1 consists of a slider with a convex pad surface moving with a uniform velocity U in the x-direction and the stator having a porous matrix of uniform thickness  $H^*$  backed by a solid wall.



Figure 1. Porous slider bearing with a convex pad surface.

The film thickness h is given by the equation

$$h = 4H_c \left(\frac{x^2}{A^2} - \frac{x}{A}\right) + h_2 - (h_2 - h_1)\frac{x}{A}$$
(1)

where  $H_c$  is the central thickness of the convex pad, A is the bearing length,  $h_1$  and  $h_2$  are minimum and maximum film thicknesses.

The eq. governing the film pressure p is [3]

$$\frac{\partial^2 u}{\partial z^2} = \frac{1}{\zeta} \frac{\partial}{\partial x} \left( p - \frac{\mu_0 \bar{\mu} H^2}{2} \right) \tag{2}$$

where u is the x-component of the fluid in the film,  $\zeta$  is the fluid viscosity,  $\mu_0$  is the free space permeability,  $\bar{\mu}$  is the magnetic susceptibility of the fluid particles and H is the magnitude of the external magnetic field

$$\bar{H} = H(\cos\Phi(x, z), 0, \sin\Phi(x, z)), \tag{3}$$

with

$$H^2 = Kx(A - x),\tag{4}$$

where K is a constant chosen to suit the dimensions of both sides of eq.(4) and  $\Phi$  is the inclination of the magnetic field with the x-axis as determined in [3].

Solving eq.(2) under the slip boundary conditions [7]

$$u = U$$
 when  $z = h$ ,  $u = \left(\frac{1}{s}\frac{\partial u}{\partial z}\right)$  when  $z = 0$  (5)

we obtain

$$u = \frac{1+sz}{1+sh}U + \frac{(z+shz+h)(z-h)}{2\zeta(1+sh)}\frac{\partial}{\partial x}\left(p - \frac{1}{2}\mu_0\bar{\mu}H^2\right).$$
 (6)

The integral form of the continuity equation for an infinitely broad slider bearing is

$$\frac{\partial}{\partial x} \int_0^h u \, dz + w_h - w_0 = 0 \tag{7}$$

where w is the axial component of the film fluid velocity and  $w_0$ ,  $w_h$  are its values at z = 0, h respectively.

Since the surface z = h is impermeable,  $w_h = 0$ , and eq.(7) reduces to

$$\frac{\partial}{\partial x} \int_0^h u \, dz = w_0. \tag{8}$$

The velocity components of the fluid velocity in the porous matrix are [3]

$$\bar{u} = -\frac{k}{\zeta} \frac{\partial}{\partial x} \left( P - \frac{1}{2} \mu_0 \bar{\mu} H^2 \right), \quad \bar{w} = -\frac{k}{\zeta} \frac{\partial}{\partial z} \left( P - \frac{1}{2} \mu_0 \bar{\mu} H^2 \right), \tag{9}$$

 ${\cal P}$  being the fluid pressure there.

Using continuity equation, eq.(9) yields

$$\frac{\partial^2}{\partial x^2} \left( P - \frac{1}{2} \mu_0 \bar{\mu} H^2 \right) + \frac{\partial^2}{\partial z^2} \left( P - \frac{1}{2} \mu_0 \bar{\mu} H^2 \right) = 0.$$
(10)

Integrating eq.(10) across the porous matrix, i.e. in the interval  $(-H^*, 0)$ , we obtain

$$\frac{\partial}{\partial z} \left( P - \frac{1}{2} \mu_0 \bar{\mu} H^2 \right) \Big|_{z=0} = -H^* \frac{\partial^2}{\partial x^2} \left( p - \frac{1}{2} \mu_0 \bar{\mu} H^2 \right) , \qquad (11)$$

remembering that the surface  $z = -H^*$  is impermeable and using the Morgan-Cameron approximation [3].

Owing to the continuity of fluid velocity components across the surface z = 0, we have

$$w_0 = \bar{w}_0 = -\frac{k}{\zeta} \frac{\partial}{\partial z} \left( P - \frac{1}{2} \mu_0 \bar{\mu} H^2 \right) \Big|_{z=0}, \qquad (12)$$

using eq.(9).

Using eqs.(6), (11) and (12), eq.(8) yields the Reynolds type equation in this case as

$$\frac{d}{dx}\left[\left\{12kH^* + \frac{h^3(4+sh)}{(1+sh)}\right\}\frac{d}{dx}\left(p - \frac{1}{2}\mu_0\bar{\mu}H^2\right)\right] = 6\zeta U\frac{d}{dx}\left[\frac{h(2+sh)}{1+sh}\right]$$
(13)

where k is the permeability of the fluid and 1/s is the slip parameter.

Introducing the dimensionless quantities

$$X = \frac{x}{A}, \quad \Psi = \frac{kH^*}{h_1^3}, \quad \bar{h} = \frac{h}{h_1}, \quad \bar{s} = sh_1, \quad \bar{p} = \frac{h_1^2 p}{\zeta U A}, \quad \mu^* = \frac{\mu_0 \bar{\mu} K A h_1^2}{\zeta U},$$
$$\bar{H}_c = \frac{H_c}{h_1}, \quad a = \frac{h_2}{h_1} \tag{14}$$

and using eqs.(1), (4) and (13), we have

$$\frac{d}{dX}\left[G\frac{d}{dX}\left\{\bar{p}-\frac{1}{2}\mu^*X(1-X)\right\}\right] = \frac{dE}{dX},\tag{15}$$

where

$$G = 12\Psi + \frac{\bar{h}^3(4 + \bar{s}\bar{h})}{(1 + \bar{s}\bar{h})}, \quad E = \frac{6\bar{h}(2 + \bar{s}\bar{h})}{(1 + \bar{s}\bar{h})}$$
(16)

$$\bar{h} = 4\bar{H}_c(X^2 - X) + a - (a - 1)X \tag{17}$$

## 3. Solutions

Solving eq.(15) under the boundary conditions

$$\bar{p} = 0$$
 when  $X = 0, 1,$  (18)

we obtain the dimensionless film pressure  $\bar{p}$  as

$$\bar{p} = \frac{1}{2}\mu^* X(1-X) + \int_1^X \frac{E-Q}{G} dX$$
(19)

where

$$Q = \frac{\int_{0}^{1} \frac{E}{G} dX}{\int_{0}^{1} \frac{1}{G} dX}$$
(20)

The load capacity W, friction on the slider F, coefficient of friction f and the position of centre of pressure  $\bar{X}$  can be expressed in dimensionless forms as

$$\bar{W} = \frac{h_1^2 W}{\zeta U A^2 B} = \frac{\mu^*}{12} - \int_0^1 X \frac{E - Q}{G} dX$$
(21)

$$\bar{F} = \frac{h_1 F}{\zeta U A B} = \int_0^1 \left[ \frac{\bar{s}}{1 + \bar{s}\bar{h}} + \frac{\bar{h}(2 + \bar{s}\bar{h})(E - Q)}{2G(1 + \bar{s}\bar{h})} \right] dX \tag{22}$$

$$\bar{f} = \frac{Af}{h_1} = \frac{\bar{F}}{\bar{W}} \tag{23}$$

$$Y = \frac{\bar{X}}{A} = \frac{1}{\bar{W}} \left[ \frac{\mu^*}{24} - \frac{1}{2} \int_0^1 X^2 \frac{E - Q}{G} dX \right]$$
(24)

where B is the breadth of the bearing.

# 4. Results and discussion

Expressions for dimensionless pressure  $\bar{p}$ , load capacity  $\bar{W}$ , friction on the slider  $\bar{F}$ , the coefficient of friction  $\bar{f}$  and the position of the centre of pressure Y are given by eqs.(19), (21) to (24). Values of  $\bar{W}$ ,  $\bar{F}$ ,  $\bar{f}$ , Y are computed using Simpson's one third rule with step size 0.1 for various values of the slip parameter  $1/\bar{s}$  and the permeability parameter  $\Psi$  are displayed in Figures 2 - 4 and Table 1 respectively.



Figure 2. Dimensionless load capacity for different values of slip parameter and permeability parameter.



Figure 3. Dimensionless friction for different values of slip parameter and permeability parameter.



Figure 4. Dimensionless coefficient of friction for different values of slip parameter and permeability parameter.

Fig.2 shows that  $\overline{W}$  decreases when  $1/\overline{s}$  or  $\Psi$  increases. It can be seen from Fig.3 that  $\overline{F}$  decreases when  $1/\overline{s}$  or  $\Psi$  increases. From Fig.4 we see that  $\overline{f}$  generally decreases when  $1/\overline{s}$  increases while it increases when  $\Psi$  increases. The position of the centre of pressure Y is not significantly altered by altering  $1/\overline{s}$  or  $\Psi$ , as can be seen from Table 1.

$1/\bar{s} \Psi$	0.0	0.0001	0.001	0.01	0.1
0.1	0.5156	0.5156	0.5154	0.5136	0.5020
0.2	0.5140	0.5140	0.5139	0.5123	0.5023
0.4	0.5128	0.5128	0.5127	0.5115	0.5032
2.0	0.5136	0.5136	0.5135	0.5128	0.5072
$\bar{H}_c = 0.3, a = 2, \mu^* = 0.5$					

**Table 1.** Values of the dimensionless position of the centre of pressure Y for different values of  $1/\bar{s}$  and  $\Psi$ .

The present analysis reduces to the analysis [1] for a conventional lubricant by setting  $1/\bar{s} = \mu^* = 0$  and to the analysis [3] by setting  $1/\bar{s} = \bar{H}_c = 0$ .

The present analysis and analysis [3] agree on the fact that the increase in load capacity is  $\mu^*/12$  and there is no change in friction due to ferrofluid lubricant. However, the present analysis shows that both load capacity and friction decrease due to slip velocity. From Figures 2 and 3, decrease in load capacity is not more significant than that in friction.

#### 5. Conclusions

Existence of slip causes decrease in the load capacity, friction and the coefficient of friction; the position of the centre of pressure is not significantly altered. However, the advantage of the bearing's stability under varying operating conditions remains. Such a bearing can be used in moderate heat environments where low friction is required, inspite of the disadvantage of lower load capacity. The present analysis may help in the design of bearings where naturally permeable material is used in the porous matrix.

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# **Effective Geometries**

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#### Abstract

Realistic test particles in general relativity occupy a non-zero region of spacetime. Therefore, the measured metric differs from the actual one, and so does the corresponding effective geometry. In this paper, a simple procedure is suggested to mimic the realistic averaging effect. It is shown that the resulting effective geometry possesses a number of interesting properties. In particular, the Riemannian spaces can acquire non-vanishing torsion, as a result of this kind of averaging.

Key words: Spacetime geometry, averaging, effective geometries

#### 1. Introduction

Starting with Einstein's general relativity [1-3], spacetime geometry became a wide arena for physical investigations. The basic ingredients which characterize spacetime are its metric  $g_{\mu\nu}$  and the connection  $\Gamma^{\mu}{}_{\nu\lambda}$ . They influence the matter field equations which, depending on the theory adopted, can have a variety of forms. The commonly accepted are the field equations that follow from the equivalence principle of general relativity. In what follows, we shall restrict our considerations to the Riemannian geometry, characterized by the absence of torsion.

Realistic measurements of the spacetime geometry employ matter fields to detect the internal spacetime structure. In general relativity, for example, point particles move along geodesics, and are used to probe spacetime geometry. However, there are no such matter structures as point particles. In reality, the particles occupy a region of spacetime, and consequently, the resulting metric is not the actual metric in a given spacetime point, but instead its average over the region occupied by the particle. In this paper, we shall try to take this fact into account, and obtain the observed effective geometry by averaging the metric over small regions of spacetime. For simplicity, we shall restrict our considerations to spaces of Euclidean signature. The more realistic Minkowski case will be considered elsewhere.

The motivation for this kind of investigation comes from the hope that effective geometries can account for quite a number of observed spacetime properties. For example, the well known dimensional reduction procedure in Kaluza-Klein theories [4-6] is an example of averaging higher-dimensional spacetimes. The metric is averaged over the regions much larger than the size of the extra dimension, and as a consequence, the effective spacetime of different dimension and topology is obtained. There is also a hope that the Minkowski signature is, in fact, the effective property of a properly chosen space of Euclidean signature. If the small scale structure of a space with Euclidean signature defines a privileged direction, the averaging may result in an effective Minkowski space. This is another reason why we decided to restrict our considerations to spaces of Euclidean signature. Finally, the quantum behaviour of a point particle may be a consequence of the small scale fluctuations of flat geometry [7]. The averaged metric would then look as a standard flat space, and the irregular particle trajectory could be interpreted as a quantum effect. There have been attempts in the literature to use the stochastic approach to quantum mechanics [8,9], and the concept of non-local fields of extended particles [10], to give geometric interpretation to quantum physics [11]. In this paper, we shall not attempt to achieve all these results. Instead, we shall investigate a simple averaging model to show that searching for more serious effects is worthy of our attention.

The results obtained in this paper can be summarized as follows: a) the effective geometry has a non-vanishing torsion, no matter that the actual space is torsionless, b) the effective scalar curvature is larger than the actual curvature, and c) the effective volume becomes smaller if R > 0, and larger if R < 0. All these are perturbative results. The drastic changes of space geometry described above can only be obtained by non-perturbative methods, which will be the objective of our subsequent investigations.

The layout of the paper is as follows. In Section 2., we define the notion of the vicinity of a space point, and define a covariant averaging procedure for world scalars. Then, in Section 3., we use the Klein-Gordon equation to define effective metric and connection. Also, the analysis of the obtained results and some examples are given. Section 4. is devoted to concluding remarks.

#### 2. Averaging scalars

Let us first define the vicinity of a point in an *n*-dimensional space of Euclidean signature with metric  $g_{\mu\nu}$ . To do this in a covariant way, we shall make use of the function

$$f_k(x,y) \equiv N(x) e^{-k^2 \ell^2(x,y)}.$$
 (1)

The normalization factor N(x) is chosen to ensure

$$\int d^n y \sqrt{g(y)} f_k(x, y) = 1, \qquad (2)$$

and  $\ell(x, y)$  measures the distance between the points x and y. It is defined as the length of the shortest line (geodesic) connecting these two points. The function  $f_k(x, y)$  will be used as a weight function to suppress the region outside the 1/k vicinity of the point x.

In what follows, the points in the 1/k vicinity of x will be considered indistinguishable. This is the model of space probed by the test particle which is not localized in a point, but instead occupies the region defined by the sphere of radius 1/k. As a consequence, measuring the scalar field  $\varphi$  in the point x will result in  $\overline{\varphi}$  = the average value of  $\varphi$  in the 1/k vicinity of x. The simplest way to define  $\overline{\varphi}$  is

$$\bar{\varphi}(x) = \int d^n y \sqrt{g(y)} f_k(x, y) \varphi(y) \,. \tag{3}$$

This is a covariant definition:  $\bar{\varphi}$  is also a world scalar.

Let us compute this quantity. First, the function  $\varphi(y)$  is covariantly expanded in a power series of the form

$$\varphi(y) = \varphi(x) + \varphi_{,\mu}(x)\ell^{\mu}(x,y) + \frac{1}{2}\varphi_{;\mu\nu}(x)\ell^{\mu}(x,y)\ell^{\nu}(x,y) + \cdots , \qquad (4)$$

where  $\ell^{\mu}(x, y)$  is a vector in x tangent to the geodesic connecting x and y, and having length  $\ell(x, y)$ . The semicolon stands for the covariant derivative with respect to the Levi-Civita connection of the metric  $g_{\mu\nu}$ . Using this expansion in (3), we arrive at

$$\bar{\varphi} = b + b^{\mu}\varphi_{,\,\mu} + \frac{1}{2}b^{\mu\nu}\varphi_{;\,\mu\nu} + \cdots , \qquad (5)$$

where b's are given by

$$b = \varphi(x),$$
  

$$b^{\mu} = \int d^{n}y \sqrt{g(y)} f_{k}(x,y) \ell^{\mu}(x,y),$$
  

$$b^{\mu\nu} = \int d^{n}y \sqrt{g(y)} f_{k}(x,y) \ell^{\mu}(x,y) \ell^{\nu}(x,y), \dots$$
(6)

The coefficients  $b, b^{\mu}, b^{\mu\nu}, \dots$  are world tensors. Using the geodesic equation, we find

$$\ell^{\mu}(x,y) = x^{\mu} - y^{\mu} + \frac{1}{2} \Gamma^{\mu}{}_{\nu\lambda} (x^{\nu} - y^{\nu}) (x^{\lambda} - y^{\lambda}) + \cdots$$
 (7)

The easiest way to compute b's is to make use of the local Euclidean coordinate frame attached to the point x. In this frame,  $g_{\mu\nu}(x) = \delta_{\mu\nu}$ ,  $\Gamma^{\mu}{}_{\nu\lambda}(x) = 0$ , and all the integrals become Gaussian. After we finish calculations in this particular frame, we rewrite the results in a covariant form, and obtain

$$b^{\mu\nu} = \frac{1}{2k^2} g^{\mu\nu} + \mathcal{O}(1/k^4), \quad \text{others} = \mathcal{O}(1/k^4).$$
 (8)

Consequently,

$$\bar{\varphi} = \varphi + \frac{1}{4k^2} \,\Box \varphi + \mathcal{O}(1/k^4) \,. \tag{9}$$

Here,  $\Box$  stands for the covariant d'Alembertian, and  $\mathcal{O}(1/k^4)$  denotes terms of order  $1/k^4$  or higher. In what follows, we shall restrict our considerations to the lowest order in 1/k, aware of the fact that thereby many interesting non-perturbative effects can not be seen.

The effective scalar field  $\bar{\varphi}(x)$  is what we measure in x by the detector having a size of the sphere of radius 1/k. There is, however, another kind of error which is a consequence of the impossibility to distinguish close space points. If we want to measure  $\varphi$  in more than one point, the points can not be closer than 2/k to each other. To obtain a smooth everywhere defined scalar field in a given region, we approximate its experimental values with the simplest polynomial function. For example, the region defined by the sphere of radius 1/k (1/k region) is seen as only one space point. Consequently, in that region, the function  $\bar{\varphi}(x)$  can only be approximated with a constant. Similarly, in a 2/k region, there are two distinguishable points, and the simplest polynomial to approximate  $\bar{\varphi}$  is a linear function. If we want our experimental function to have well defined second derivatives, we
need at least three points, or equivalently, a 3/k region of space. There, we approximate our  $\bar{\varphi}$  with a quadratic polynomial q(x, y):

$$q(x,y) = c(x) + c_{\mu}(x)\ell^{\mu}(x,y) + \frac{1}{2}c_{\mu\nu}(x)\ell^{\mu}(x,y)\ell^{\nu}(x,y).$$
(10)

The coefficients  $c, c_{\mu}$  and  $c_{\mu\nu}$  are determined from the requirement that the functional

$$J(x) \equiv \int d^n y \sqrt{g(y)} \left[\bar{\varphi}(y) - q(x,y)\right]^2 f_{k/3}(x,y) \tag{11}$$

has minimal value. Consequently, we solve the equation  $\delta J = 0$ , or equivalently

$$\int d^{n} y \sqrt{g} \left(\bar{\varphi} - q\right) f_{k/3} = 0,$$

$$\int d^{n} y \sqrt{g} \left(\bar{\varphi} - q\right) \ell^{\mu} f_{k/3} = 0,$$

$$\int d^{n} y \sqrt{g} \left(\bar{\varphi} - q\right) \ell^{\mu} \ell^{\nu} f_{k/3} = 0,$$
(12)

and obtain

$$c = \bar{\varphi} + \mathcal{O}(1/k^{4}),$$

$$c_{\mu} = \bar{\varphi}_{,\mu} + \frac{3}{4k^{2}} \left[ 3 \left( \Box \bar{\varphi} \right)_{,\mu} - 2R_{\mu\rho} \,\bar{\varphi}^{,\rho} \right] + \mathcal{O}(1/k^{4}),$$

$$c_{\mu\nu} = \bar{\varphi}_{;\,\mu\nu} + \frac{9}{4k^{2}} \left[ \left( \Box \bar{\varphi} \right)_{;\,\mu\nu} - \frac{2}{3} \left( R_{\lambda\mu\nu\rho} + g_{\nu\rho}R_{\mu\lambda} + g_{\mu\rho}R_{\nu\lambda} \right) \bar{\varphi}^{;\,\rho\lambda} + \frac{1}{6} \left( R_{\mu\nu;\,\rho} - 3R_{\mu\rho;\,\nu} - 3R_{\nu\rho;\,\mu} \right) \bar{\varphi}^{,\rho} \right] + \mathcal{O}(1/k^{4}).$$
(13)

The coefficients c,  $c_{\mu}$  and  $c_{\mu\nu}$  minimize the functional J, and consequently, determine quadratic polynomial q(x, y) which is the best approximation for what we measure in 3/kvicinity of the point x. Therefore, the average scalar field in 3/k vicinity of the point xwill be defined as

$$\langle \varphi(y) \rangle = q(x,y) \,. \tag{14}$$

This approximation can only be used if we are interested in not higher than second derivatives of  $\langle \varphi \rangle$  in point x. Then, the average measured field and its first two derivatives in x are defined as the value of q(x, y) and its two y-derivatives in y = x. Using (10), we find

$$\begin{array}{ll} \langle \varphi \rangle &= c \,, \\ \langle \varphi_{,\mu} \rangle &= c_{\mu} \,, \\ \langle \varphi_{,\mu\nu} \rangle &= c_{\mu\nu} + \Gamma^{\lambda}{}_{\mu\nu} \, c_{\lambda} \,. \end{array}$$
(15)

Higher derivatives should not be calculated from (14). Instead, we need an approximation by higher order polynomials, in a wider vicinity of the point x. More precisely, to measure n-th derivative, we need (n + 1)/k vicinity of the point where the derivative is taken.

#### 3. Averaging metric

The averaging procedure for the space metric  $g_{\mu\nu}$  can not follow the same rules as in the case of a scalar field. This is because integration of a world tensor over a space region gives a non-tensorial quantity. To define average metric, we shall make use of the fact that geometry is never directly measured, but is, instead, probed by test matter fields, and then deduced from the known field equations.

**Definition**. Suppose we know how to measure a scalar field  $\varphi$ . Assuming that  $\varphi$  satisfies the Klein-Gordon equation in curved space,

$$\left(\Box + m^2\right)\varphi = 0\,,\tag{16}$$

we can use it to directly read the metric. Indeed, in a curved Riemannian space with metric  $g_{\mu\nu}$ , the Klein-Gordon equation reads

$$g^{\mu\nu}\varphi_{,\mu\nu} - \Gamma^{\mu}\varphi_{,\mu} + m^2\varphi = 0, \qquad (17)$$

where  $\Gamma^{\mu} \equiv \Gamma^{\mu}{}_{\nu\lambda}g^{\nu\lambda}$ . Measuring the scalar field  $\varphi$  and its two derivatives, and comparing with (17), we find the metric and the trace connection as the coefficients by  $\varphi_{,\mu\nu}$  and  $\varphi_{,\mu}$ , respectively. Also, we can read the mass of our scalar field.

We know that what we measure is not  $\varphi$ , but instead  $\langle \varphi \rangle$ . The measured values of the scalar field and its two derivatives, as given by (15), do not generally satisfy an equation of the (17) type. It is not difficult, however, to find what equation the average scalar field satisfies. First, we notice that, in the lowest order in 1/k,  $\bar{\varphi}$  obeys the same Klein-Gordon equation as the actual field  $\varphi$ :

$$\left(\Box + m^2\right)\bar{\varphi} = \mathcal{O}(1/k^4).$$
(18)

Then, we find

$$\left( \Box + m^2 \right) \langle \varphi \rangle \equiv g^{\mu\nu} \langle \varphi, \mu\nu \rangle - \Gamma^{\mu} \langle \varphi, \mu \rangle + m^2 \langle \varphi \rangle$$

$$= c^{\mu}{}_{\mu} + m^2 c ,$$

$$(19)$$

which, after using the Klein-Gordon equation (18), and the known values of the coefficients  $c_{\mu\nu}$  and c, leads to

$$\bar{g}^{\mu\nu}\langle\varphi_{,\mu\nu}\rangle - \bar{\Gamma}^{\mu}\langle\varphi_{,\mu}\rangle + \bar{m}^2\langle\varphi\rangle = \mathcal{O}(1/k^4)\,, \tag{20}$$

with

$$\bar{g}^{\mu\nu} = g^{\mu\nu} + \frac{3}{2k^2} R^{\mu\nu} + \mathcal{O}(1/k^4) ,$$
  

$$\bar{\Gamma}^{\mu} = \Gamma^{\mu} + \frac{3}{4k^2} \left( 2\Gamma^{\mu}{}_{\nu\lambda} R^{\nu\lambda} - R^{,\mu} \right) + \mathcal{O}(1/k^4) ,$$
  

$$\bar{m}^2 = m^2 \left( 1 - \frac{9m^2}{4k^2} \right) + \mathcal{O}(1/k^4) .$$
(21)

As we can see, the average scalar field  $\langle \varphi \rangle$  satisfies the Klein-Gordon equation in a curved space with metric  $\bar{g}^{\mu\nu}$ , which differs from the actual metric  $g^{\mu\nu}$ . The new metric  $\bar{g}^{\mu\nu}$  is the average metric we have been looking for. Similarly,  $\bar{\Gamma}^{\mu}$  is the average trace connection. It can be rewritten as

$$\bar{\Gamma}^{\mu} = \bar{g}^{\nu\lambda} \,\bar{\Gamma}^{\mu}{}_{\nu\lambda}\,,\tag{22}$$

where

$$\bar{\Gamma}^{\mu}{}_{\nu\lambda} = \Gamma^{\mu}{}_{\nu\lambda} - \frac{3}{4k^2} \left( \delta^{\mu}{}_{\nu}R_{,\lambda} + A^{\mu}{}_{\nu\lambda} \right) + \mathcal{O}(1/k^4) \,. \tag{23}$$

The average connection  $\bar{\Gamma}^{\mu}{}_{\nu\lambda}$  is determined up to an arbitrary traceless function  $A^{\mu}{}_{\nu\lambda}$ ,

$$\bar{g}^{\nu\lambda}A^{\mu}{}_{\nu\lambda} = \mathcal{O}(1/k^2)\,,\tag{24}$$

which is a consequence of using scalar fields to probe space geometry. If we want to measure all the components of the connection, we need vector or spinor test fields. Finally,  $\bar{m}$  is the average mass of the scalar field. To summarize,

the quantities  $\bar{g}^{\mu\nu}$  and  $\bar{\Gamma}^{\mu}{}_{\nu\lambda}$  are the average values of space metric and connection, and  $\bar{m}$  is the average mass of the test scalar field. In the lowest order in 1/k, they are given by the equations (21) and (23).

Note that the equation (20) determines  $\bar{g}^{\mu\nu}$  and  $\bar{m}$  only up to a multiplicative constant. We fixed it by the requirement that the average and actual metrics coincide in flat geometries.

**Analysis.** Let us see how the effective and actual geometries differ from each other. To do that, we shall first complete the definition of the effective connection  $\bar{\Gamma}^{\mu}{}_{\nu\lambda}$  by appropriately choosing the arbitrary traceless function  $A^{\mu}{}_{\nu\lambda}$ . One should have in mind that this procedure can not give us the real effective connection, since it can only be obtained by performing experiments with vector or spinor test fields. Still, analyzing different choices of  $A^{\mu}{}_{\nu\lambda}$  will tell us which effective geometries are excluded.

The first question concerns the possibility that the effective geometry remains Riemannian. Is there a choice of  $A^{\mu}{}_{\nu\lambda}$  that makes  $\bar{\Gamma}^{\mu}{}_{\nu\lambda}$  a Levi-Civita connection? Equivalently, can an appropriate choice of  $A^{\mu}{}_{\nu\lambda}$  make the effective torsion and non-metricity vanish? The analysis is straightforward, and shows that two distinctive cases can be achieved. First, the effective non-metricity  $(Q_{\mu\nu\lambda} \equiv -g_{\mu\nu;\lambda})$  vanishes, and the torsion  $(T^{\mu}{}_{\nu\lambda} \equiv \Gamma^{\mu}{}_{\nu\lambda} - \Gamma^{\mu}{}_{\lambda\nu})$  remains non-trivial:

$$\bar{Q}_{\mu\nu\lambda} = 0, \qquad \bar{T}^{\mu}{}_{\nu\lambda} = -\frac{3}{4(n-1)k^2} \left(\delta^{\mu}_{\nu}R_{,\lambda} - \delta^{\mu}_{\lambda}R_{,\nu}\right). \tag{25}$$

Second, the effective torsion vanishes, but non-metricity remains non-trivial:

$$\bar{T}^{\mu}{}_{\nu\lambda} = 0, \qquad \bar{Q}_{\mu\nu\lambda} = -\frac{3}{4nk^2} \left( g_{\lambda\mu}R_{,\nu} + g_{\lambda\nu}R_{,\mu} \right). \tag{26}$$

As we can see from (25) and (26), the simultaneous vanishing of both, effective torsion and non-metricity, is possible only if R = const. In general, the effective geometry is not Riemannian. In what follows, we shall always work with  $\bar{Q}_{\mu\nu\lambda} = 0$ , as non-metric geometries can hardly be given physical interpretation. As seen from (25), this means that the general effective geometry accommodates a non-zero torsion. (The basics of torsion dynamics can be found in [12, 13].)

Let us see what happens to the space volume in the process of averaging. Using the formula for  $\bar{g}^{\mu\nu}$  in (21), we find

$$\bar{g}_{\mu\nu} = g_{\mu\nu} - \frac{3}{2k^2} R_{\mu\nu} + \mathcal{O}(1/k^4) \,, \tag{27}$$

wherefrom

$$\bar{g} = g\left(1 - \frac{3}{2k^2}R\right) + \mathcal{O}(1/k^4).$$
(28)

Thus, the effective volume is larger or smaller than actual, depending of whether R > 0 or R < 0. As an example, consider an Einstein space defined by  $R_{\mu\nu} = \lambda g_{\mu\nu}$ ,  $\lambda = \text{const.}$  The averaging procedure then gives

$$\bar{T}^{\mu}{}_{\nu\lambda} = \mathcal{O}(1/k^4), \quad \bar{Q}_{\mu\nu\lambda} = \mathcal{O}(1/k^4), 
\bar{g}_{\mu\nu} = g_{\mu\nu} \left(1 - 3\lambda/2k^2\right) + \mathcal{O}(1/k^4), 
\bar{R}^{\mu}{}_{\nu\lambda\rho} = R^{\mu}{}_{\nu\lambda\rho} + \mathcal{O}(1/k^4).$$
(29)

The effective scalar curvature is then found to be always larger than the actual:

$$\bar{R} = R + \frac{3R^2}{2nk^2} + \mathcal{O}(1/k^4) \,. \tag{30}$$

In the case of a 2-sphere,  $R = -2/r^2$ , and consequently, the effective radius  $\bar{r} > r$ . Thus, the effective sphere is larger than the actual, in agreement with the result (28) concerning space volumes.

The above analysis is strictly perturbative, and holds for small values of 1/k. In other words, only when the test particle is much smaller than the characteristic length of geometry fluctuations, the low order results can be meaningful. For example, averaging the 2-sphere of radius r over regions much smaller than  $4\pi r^2$  gives a larger effective 2-sphere. If, however, the averaging region is larger than  $4\pi r^2$ , the sphere will look as a point to us, and consequently, the effective volume will be zero.

# 4. Concluding remarks

We have shown in this work how realistic measurements define effective geometries. Starting with the idea that matter fields are used to detect internal spacetime structure, we first defined the averaging procedure for world scalars. The average has been taken over small regions of space, thereby mimicking experiments with non-localized test particles. Then, matter field equations have been used to determine the effective metric and connection. Assuming that scalar fields obey the Klein-Gordon equation in a curved geometry, we have shown that their averages do the same. This time, however, the metric and connection that enter the Klein-Gordon equation, were different. They define the effective geometry.

The properties of the effective space have been analyzed only perturbatively. It has been shown that non-zero torsion can appear after the averaging procedure, no matter that the actual space was Riemannian. The effective space volume turns out to be larger or smaller than the actual one, depending on the sign of the scalar curvature. The effective scalar curvature, on the other hand, has been considered in the case of an Einstein space, with the result that averaging increases its value.

The work reported here has been done in the lowest order of the averaging parameter. This means that the averaging area is supposed to be much smaller than the correlation length associated with the fluctuations of geometry. As a consequence, many attractive non-perturbative averaging effects are missed. To obtain Minkowski space by averaging geometry of Euclidean signature, or to explain quantum behaviour as originating from spacetime fluctuations, we need a more complex, non-perturbative averaging procedure.

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# Stark Shift Determination for Two Red Neutral Argon Spectral Lines

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#### Abstract

This paper presents Stark shift measurement results of Ar I 794.818 nm and Ar I 810.369 nm lines. These results are part of a systematic and high precision experimental study of argon spectral line widths and shifts. The measured shifts were compared with available theoretical and other experimental data.

Key words: Stark broadening, Stark shift, spectral line shapes

# 1. Introduction

Stark effect influence on the width and shift of neutral argon spectral lines has been extensively investigated during the last three decades, since argon is a widely available inert atomic gas which produces a variety of favourable conditions for stable discharges. Stark parameters are important for plasma diagnostic purposes of laboratory as well as astrophysical plasmas and also for theory testing and different plasma applications.

Subject of this paper is a Stark shift determination for two red, Ar I 794.818 nm  $(4s'[1/2]_0^0 - 4p'[3/2]_1)$  and Ar I 810.369 nm  $(4s[3/2]_1^0 - 4p[3/2]_1)$ , spectral lines. In the literature there are only two results for shift measurements of the 794.818 nm line [1, 2].

The shifts are measured for electron density interval from  $0.74 \cdot 10^{22} \text{ m}^{-3}$  to  $2.90 \cdot 10^{22} \text{ m}^{-3}$  and temperature interval from 9280 K to 10760 K. Because the two considered lines are very narrow and they have very small shifts for the plasma conditions, we take care about a number of experimental details related to the plasma source, line shape, shift recording, measured data treatment and plasma diagnostics.

# 2. Spectral line broadening and shifting

Broadening mechanisms of spectral lines of atoms and ions in plasma can be divided into three groups:

a) Natural broadening, caused by the finite average lifetime of atoms in the excited state,

- b) Doppler broadening, due to the motion of the emitter atoms with respect to the observation system,
- c) Pressure broadening, caused by the interaction of the emitter with the neutral or charged particles.

Natural broadening is practically always negligible in comparison to other broadening mechanisms. Pressure broadening includes resonance, van der Waals and Stark broadening mechanism. The most important broadening effect for our experimental conditions is the Stark broadening. Doppler and instrumental broadening must always be taken into account. The Stark shift of the spectral line should always be corrected to the van der Waals shift.

As perturbing particles, ions are usually less effective in spectral line broadening than electrons. The exceptions are the lines belonging to one-electron systems and the lines with forbidden components, though in these cases ion broadening is not dominant. Because of this, in considering the isolated line profiles ion broadening is calculated as a small correction to the electron impact broadening. Calculation of the Stark profile, the so-called j(x) profile, in quasi-static approximation [3 - 5], can be written in the form:

$$j_{A,R}(x) = \frac{1}{\pi} \int_0^\infty \frac{W_R(\beta)d\beta}{1 + (x - A^{4/3}\beta^2)^2} \quad , \tag{1}$$

where  $x = (\lambda - \lambda_0 - d_e)/w_e$  is the reduced wavelength,  $\lambda_0$  is the central wavelength of the profile,  $d_e$  and  $w_e$  are the electron impact Stark shift and half-halfwidth respectively,  $W_R(\beta)$  is the distribution of the plasma electric microfield defined in [6], R is the ratio of the mean distance between ions to the Debye radius and A is the static ion broadening parameter (also known as  $\alpha$  see [2, 3]). The total theoretical width,  $w_{th}$ , and the total theoretical shift,  $d_{thp}$ , at the peak of the j(x) profile can be expressed as [2, 3, 7]:

$$w_{th} = 2 \left[ 1 + 1.75 \cdot 10^{-4} N_e^{1/4} A \left( 1 - 0.068 N_e^{1/6} T^{-1/2} \right) \right] \cdot 10^{-16} w_e N_e \tag{2}$$

$$d_{thp} = \left[ d_e \pm 2 \cdot 10^{-4} N_e^{1/4} A w_e \left( 1 - 0.068 N_e^{1/6} T^{-1/2} \right) \right] \cdot 10^{-16} N_e \tag{3}$$

Because of the profile asymmetry, the shift at the halfwidth position  $d_{th1/2}$  has a slightly different value [8]:

$$d_{th1/2} = \left[ d_e \pm 3.2 \cdot 10^{-4} N_e^{1/4} A w_e \left( 1 - 0.068 N_e^{1/6} T^{-1/2} \right) \right] \cdot 10^{-16} N_e \tag{4}$$

Electron half-halfwidth  $w_e$ , shift  $d_e$  and ion broadening parameter A are tabulated in Ref. 2 at  $N_e = 10^{16} \text{ cm}^{-3}$ .  $N_e$  and T are electron density (cm<sup>-3</sup>) and temperature (K) respectively, for which  $w_{th}$  and  $d_{th}$  will be calculated. There are certain restrictions on the applicability of Eqs. (2), (3) and (4) and they are  $R \leq 0.8$  and  $0.05 \leq A \leq 0.5$  [4].

# 3. Experimental apparatus for spectral line profile recording

#### a) Plasma source

As a plasma source we used a wall stabilized electric arc. The arc operates in argon under atmospheric pressure, with a small addition of hydrogen, which is used for diagnostic purposes. Arc consists of six water-cooled discs, 7.1 mm in thickness separated by 0.5 mm Teflon gaskets which serve as insulators and seal off the arc channel from the air. Diameter of the arc channel is 5 mm and its length is 50 mm. Argon is introduced into the plasma through the hollow electrodes at both ends of the arc channel with a flow rate of 3 l/min (Fig. 1). Gas mixture of argon and 4 % hydrogen is introduced into the arc channel through the hole in the copper disc placed next to the cathode with flow rate of 0.1 l/min. Gases are exhausted through the hole in another copper disc placed next to the anode. In order to perform optical alignment of the arc and for side-on plasma spectroscopic observations through the slot between two parts, located in the middle of the arc channel, glass windows are mounted (Fig. 1). The current of 30 A is supplied to the arc from the current stabilized ( $\pm 0.3$  %) power supply.

#### b) Spectral intensity recording

Plasma spectroscopic observations are performed side-on like in experiments [9, 10]. Plasma image is projected using the mirror system  $M_1$  and  $M_2$  onto the entrance slit of the monochromator at the ratio 1:1 (Fig. 1). The choice of the appropriate spherical mirror  $M_2$  (the focal length is 1.5 m) and its position with respect to the plasma source and monochromator ensure total irradiation of the monochromataor dispersion grating. Inverse linear dispersion of the 1 m monochromator with a 1200 grooves/mm grating is 0.833 nm/mm. The monochromator is also equipped with a stepping motor with 36000 step/rev. An He-Ne laser is used for optical alignment of the system plasma source mirror monochromator.



Figure 1. Experimental setup.

The plan-parallel glass plate is located between mirrors  $M_2$  and  $M_3$ . The rotation of this glass plate enabled us to shift the plasma image across the entrance slit of the monochromator. In this way radial scanning of the plasma is performed. The 10 mm diaphragm in front of mirror  $M_2$  and 0.3 mm slot placed in front of the entrance slit, limited light coming from the plasma and enabled observations of thin horizontal slices of the arc plasma.

Radiation coming from the plasma source passes also through the partially transparent (reflecting) mirror  $M_3$ . This enables observation of the radiation from the reference source, used for spectral line shift measurements. In this way by using a chopper, light from the reference source (position B, Fig 1.) or from the arc plasma (position A, Fig 1.) can be detected alternatively by the photomultiplier placed at the exit slit of the monochromator. A low pressure argon Geissler tube is used as a reference source of unshifted argon lines. Lines emitted from this source are also used for the instrumental width measurements. Another He-Ne laser enables optical alignment of the reference source. Both signals are recorded at each wavelength step along the investigated wavelength interval.

Signals from the photomultiplier are led to the digitizing oscilloscope working in an averaging mode, 32 samples over 200 ms for each experimental point. The stepping motor (HP-IB interface), chopper (RS-232 interface) and oscilloscope (HP-IB interface) are controlled by the personal computer which is also used for data acquisition.

# 4. Plasma diagnostics

Radial distribution of electron density  $N_e$  in the range  $(0.74 \ 2.90) \cdot 10^{22} \text{ m}^{-3}$  along the plasma radius was determined from the halfwidth of the Balmer H<sub> $\beta$ </sub> line.  $N_e$  measurement is performed in conjunction with theoretical calculations [11]. Estimated errors of measured electron densities, which do not exceed 9 %, were derived from the error in H<sub> $\beta$ </sub> halfwidth measurements and the typical uncertainty in  $N_e$  determination from H<sub> $\beta$ </sub> profile [12].

Electron temperature  $T_e$  in the range (9280–10750) K is deduced from [13]. Calculations from the plasma composition data, using the procedure described in [14], show that the plasma of the mixture of argon and a small addition of hydrogen (less than 4 %) can be treated as pure argon plasma. Under these conditions, Ref. [5] is used for temperature determination. The error in electron temperature measurements of 3 % is estimated from the uncertainty of  $N_e$  and its influence on  $T_e$  determination.

# 5. Experimental data treatment

In this experiment argon is used at the same time as a working gas and a gas whose spectral lines are considered. In order to avoid absorption effects and distortion of line profiles caused by emission from the electrode region, the spectroscopic observations were made side-on. Set of raw experimental Ar I 794.818 nm line profiles, observed perpendicularly to the arc axis at 16 points along the radius, is given in Fig. 2. Due to the inhomogeneity of the arc radially symmetric plasma, it is necessary to use an Abel inversion procedure to separate contributions from different plasma layers and to obtain the true radial plasma intensity distribution. Here we used an Abel inversion procedure based on the Jacobi polynomials as described in Ref. 15. The Abel inverted profiles of Ar I 794.818 nm line at 12 points along the plasma radius are given in Fig. 3. The profile of the same Ar I line emitted from the reference source is also included in Fig. 3.

The same Abel inversion procedure is also applied to  $H_{\beta}$  line profiles. For further data analysis we used only 12 profiles along the radius. The weak intensity of the last 4  $H_{\beta}$  profiles disabled proper plasma diagnostic procedure.



Figure 2. Set of raw experimental data. r is the arc channel radius.

Figure 3. Set of the profiles after the Abel inversion procedure.

The obtained Abel profiles for both Ar I 794.818 nm and Ar I 810.369 nm line are convolution of all broadening mechanisms. As already mentioned, for our experimental conditions Stark broadening and shifting is dominant while Doppler and van der Waals mechanisms are not completely negligible. Resonance broadening is negligible for these plasma conditions. In this case for both lines Stark halfwidth has a very small value. For example for the Ar I 794.818 nm line and plasma conditions  $N_e = 2.9 \cdot 10^{22} \text{ m}^{-3}$  and T = 10760 K, Stark halfwidth = 0.0299 nm (2), van der Waals halfwidth = 0.0016 nm [3, 8], Doppler halfwidth = 0.009 nm [3], instrumental halfwidth = 0.03 nm (measured). Furthermore, the same values of the Stark and instrumental halfwidth disabled proper deconvolution procedure and Stark halfwidth determination. Ion broadening parameter is also very small, A = 0.022 [4]. So, in this case the considered line profiles are practically symmetric. It means that symmetrical Lorentzian profile can be used for the theoretical description of Stark profile, instead of the j(x) one. Convolution of the Lorentzian profile and Gaussian profile, which contains the Doppler and instrumental profile, gives the so-called Voigt profile [16]. So, for determination of the spectral line center position, symmetrical function can be used, and comparing it to the reference line position, experimental shift can be determined.

# 6. Results and discussion

Shift measured data for the Ar I 794.818 nm line for different electron densities and temperatures along the plasma radius are given in Table 1. The measured shift values  $S_{meas}$  have to be corrected to the shift caused by the van der Waals effect. Van der Waals shift can be estimated as  $S_{vW} = -2/3W_{vW}$ , where  $W_{vW}$  is the van der Waals halfhalfwidth [3]. For calculations of the van der Waals half-halfwidth one can use Refs. [3] and [8]. Data, necessary for these calculations, density of perturbers and corresponding energy level data, are taken from Refs. [13] and [17] respectively. The corresponding poor Stark shift values  $S_S$  are also given in Table 1. Estimated experimental errors for Stark shifts are 1026 % from the highest to lowest electron density.

The obtained results are compared with available theoretical data [4] which are also given in Table 1. Theoretical shift  $S_{Grcalc}$  is calculated from Eq. (4). These values should be corrected to the Debye shielding effect  $(S_{Dscor})$  [4]. Corrected theoretical data  $S_{Gr}$  and

the ratio Stark shift over the theoretical one are given in the last column of the Table 1.

Table 1. The measured shift data for the Ar I 794.818 nm line.  $N_e$  electron density, T electron temperature,  $S_{meas}$  measured shift,  $S_{vW}$  van der Waals shift,  $S_S$  Stark shift,  $S_{Grcalc}$  theoretical shift (4),  $S_{DScor}$  correction to Debye shielding effect [4],  $S_{Gr}$ corrected theoretical shift and  $S_S/S_{Gr}$  Stark shift over the theoretical one.

$N_e$	Т	$S_{meas}$	$S_{vW}$	$S_S$	$S_{Grcalc}$	$S_{DScor}$	$S_{Gr}$	$S_S/S_{Gr}$
$(10^{22} \mathrm{m}^{-3})$	(K)	$(10^{-1} nm)$						
2.90	10760	0.124	-0.0055	0.119	0.1148	-0.0265	0.0883	1.35
2.82	10730	0.118	-0.0055	0.113	0.1116	-0.0254	0.0861	1.31
2.70	10700	0.115	-0.0055	0.109	0.1068	-0.0239	0.0829	1.32
2.46	10550	0.111	-0.0056	0.105	0.0974	-0.0209	0.0765	1.37
2.15	10400	0.092	-0.0057	0.086	0.0849	-0.0172	0.0677	1.27
1.90	10250	0.078	-0.0058	0.072	0.0749	-0.0144	0.0605	1.19
1.60	10050	0.072	-0.0060	0.066	0.0629	-0.0112	0.0517	1.28
1.40	9900	0.059	-0.0061	0.053	0.0550	-0.0093	0.0457	1.16
1.20	9720	0.054	-0.0062	0.048	0.0470	-0.0074	0.0396	1.21
0.98	9520	0.049	-0.0063	0.043	0.0383	-0.0055	0.0328	1.31
0.83	9400	0.037	-0.0064	0.031	0.0324	-0.0043	0.0280	1.11
0.74	9280	0.029	-0.0065	0.023	0.0288	-0.0037	0.0251	0.92

Data, necessary for Debye shielding corrections, are taken from Refs. [3], [4] and [17]. The comparison of measured Stark shifts and theoretical ones for Ar I 794.818 nm line is graphically presented in Fig. 4.

The same above described procedure is applied to the Ar I 810.369 nm line also. Results are presented in Table 2 and Fig. 5.

**Table 2.** The measured shift data for the Ar I 810.369 nm line.  $N_e$  electron density, T electron temperature,  $S_{meas}$  measured shift,  $S_{vW}$  van der Waals shift,  $S_S$  Stark shift,  $S_{Grcalc}$  theoretical shift (4),  $S_{DScor}$  correction to Debye shielding effect [4],  $S_{Gr}$  corrected theoretical shift and  $S_S/S_{Gr}$  Stark shift over the theoretical one.

$N_e$	Т	$S_{meas}$	$S_{vW}$	$S_S$	$S_{Grcalc}$	$S_{DScor}$	$S_{Gr}$	$S_S/S_{Gr}$
$(10^{22} \mathrm{m}^{-3})$	(K)	$(10^{-1} nm)$						
2.90	10760	0.119	-0.0053	0.114	0.1412	-0.0341	0.1070	1.07
2.82	10730	0.119	-0.0053	0.114	0.1372	-0.0328	0.1044	1.09
2.70	10700	0.116	-0.0053	0.111	0.1316	-0.0308	0.1008	1.10
2.46	10550	0.106	-0.0054	0.101	0.1197	-0.0269	0.0928	1.09
2.15	10400	0.09	-0.0055	0.085	0.1044	-0.0222	0.0823	1.03
1.90	10250	0.086	-0.0056	0.081	0.0923	-0.0186	0.0738	1.10
1.60	10050	0.071	-0.0058	0.065	0.0776	-0.0145	0.0631	1.03
1.40	9900	0.065	-0.0059	0.059	0.0677	-0.0119	0.0558	1.06
1.20	9720	0.052	-0.0060	0.046	0.0579	-0.0096	0.0484	1.00
0.98	9520	0.043	-0.0061	0.037	0.0472	-0.0071	0.0400	1.00
0.83	9400	0.041	-0.0062	0.035	0.0398	-0.0056	0.0342	1.02
0.74	9280	0.033	-0.0063	0.027	0.0354	-0.0047	0.0307	1.00

Stark shift results for both Ar I 794.818 nm and Ar I 810.369 nm line show small and similar disagreement with Griem's theoretical results [4]. Average ratio of the measured and theoretical shift for Ar I 794.818 nm line is 1.23, while for Ar I 810.369 nm line the ratio is 1.05. These ratios  $(S_S/S_{Gr} > 1)$  are different from ratios for most blue argon lines considered in Ref. [10]  $(S_S/S_{Gr} < 1)$ .





Figure 4. Comparison of measured and theoretical Stark shifts for Ar I 794.818 nm line.

Figure 5. Comparison of measured and theoretical Stark shifts for the Ar I 810.369 nm line.

In both cases measured shift vs electron density show the proper trend, which implies that there was no systematic error in measurement.

The shift measurements presented in this paper are new reliable data. There is not enough other experimental data available in the literature for any serious comparison with the measured shifts. There are only two experimental results of the Stark shift measurements and only for Ar I 794.818 nm line. In Ref. [1] the authors reported only one shift measurement with  $S_S/S_{Gr} = 1.21$  for  $N_e = 1 \cdot 10^{23}$  m<sup>-3</sup> and T = 13000 K. In Ref. [2] the authors also reported only one shift result with  $S_S/S_{Gr} = 0.60$  for  $N_e = 1 \cdot 10^{23}$  m<sup>-3</sup> and T = 12500 K. For these results see also Refs. [18] and [19].

# 7. Conclusion

Measurement of the Stark broadening parameters is important for precise plasma diagnostics and for various plasma applications. In this work, the values of the Stark shift of Ar I 794.818 nm and Ar I 810.369 nm lines for various electron densities are presented. In order to obtain good and reliable experimental data, we used a stable continuous plasma source and experimental technique, which enabled high precision shift measurements. Particular attention was paid to proper numerical procedures for experimental data treatment.

The obtained results are higher than the theoretical ones and indicate that further improvements of the theory are needed. However, the discrepancy between these and other experimental data shows that new precision measurements should be done in order to clear this discrepancy.

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- 2. E. U. Shirley, *The Theory of Molecules*, p. 399, (The University Press, Cambridge, 1954). for books
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