

# The Stochastic Approach in Analysis—the Structures with Vacancies and Impurities

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**Abstract** The stochastic approach to the problem of deformed bulk structures is a unique possibility for obtaining a so–so real picture of physical processes in these structures. The chain of identical atoms with random distances between neighbours as well as the chain with random masses at equal distances is analysed. These analyses were generalized to complex chains, which are spatially and mass deformed. The approximations used were incommensurability; one and more rough, continual approximation. The probabilities are of exponential type. It means that the error in estimation on the basis of stochastic method can go to 100% maximally.

**Keywords** Stochastic · Spatial deformation · Mass deformation · Convolution of distribution densities

## 1 Introduction

This part is dedicated to bulk structures with impurities and vacancies [1, 2]. It is known that purely clean crystals do not exist—some percent of structure defect is always present. The

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influence of structural defects (vacancies and impurities) to behaviour of the system can be exactly treated only in presence of one or two defects [3–5]. In the case of three defects, we have a three-body problem [6, 7]. In all other cases, the stochastical treatment is a unique way for the estimate of behaviour of broken symmetry bulk. In other words, the probabilistic treatment is unique chance [8, 9].

We shall treat stochastically two types of one-dimensional chains. The first type is the chain with identical atoms (molecules) with different distances between neighbour atoms. In practice, it is chain with vacancies. The second one is the chain with identical distances between atoms, but with different masses. It is, in fact, a system with impurities. Even when if it is possible, the results obtained for chains will be generalized to three-dimensional structures.

The results for space distribution of defects (chain with vacancies) and for mass distribution will be generalized by means of distributions convolution [10]. The distribution obtained by convolution as a resulting one, would serve for estimating the chain in which the vacancies as well as impurities are present.

### 2 The Chain with Vacancies. The Spatial Distribution Density

We shall consider the chain in which the distance between  $N_{1D}$  atoms is  $a_1$  between  $N_{2D}$  atoms the distance is  $a_2$ , etc. We assume that there are  $n$  different distances between atoms. The total number of atoms  $N_D = \sum_{s=1}^n N_{sD} = \text{const.}$  [11] and that the length of the chain  $L = \sum_{s=1}^n a_s N_{sD} = \text{const.}$  is conserved.

The statistical probability of the described system is given by

$$P = \frac{N_D!}{\prod_{s=1}^n N_{sD}!} \approx \frac{N_D^{N_D}}{\prod_{s=1}^n N_{sD}^{N_{sD}}}. \tag{1}$$

Analogously to the standard Boltzmann’s approach [12], the most probable distribution of atoms over distances is obtained by equating the variation of the function

$$\Phi = \ln P - \alpha_D N_D - \beta L = N_D \ln N_D - \sum_{s=1}^n (N_{sD} \ln N_{sD} + \alpha_D N_{sD} + \beta a_s N_{sD}) \tag{2}$$

with zero. Undetermined Lagrange coefficients [13] are denoted with  $\alpha$  and  $\beta$ .

Since  $\delta\Phi = -\sum_{s=1}^n (\ln N_{sD} + 1 + \alpha_D + \beta a_s) \delta N_{sD}$ , the equation determining the most probable distribution is

$$\ln N_{sD} + 1 + \alpha + \beta a_s = 0 \tag{3}$$

where from it follows

$$N_{sD} = e^{-(\alpha_D+1)} e^{-\beta a_s}. \tag{4}$$

Substituting (4) in  $\sum_{s=1}^n N_{sD} = N_D$ , we obtain:

$$e^{-(\alpha_D+1)} = \frac{N_D}{\sum_{s=1}^n e^{-\beta a_s}}. \tag{5}$$

Combining (4) and (5), we find the probability that the distance between  $N_{sD}$  neighbour atoms is  $a_s$ :

$$W_s = \frac{N_{sD}}{N_D} = \frac{e^{-\beta a_s}}{\sum_{s=1}^n e^{-\beta a_s}}. \tag{6}$$

The probability  $W_s$  given in the formula (6) is not convenient for further analysis of the chain since neither  $a_s$  nor  $\beta$  are known. For this reason, we introduce incommensurate approximation:

$$a_s = sa_0 \tag{7}$$

where  $a_0$  is minimal distance between neighbour atoms (it is clear from (7) that  $a_1 = a_0$ ). In the case of vacancies, approximation (7) is relatively close to a real situation, but in the case of random impurities this approximation is more rough. Since real distances are  $(s + \rho)a_0$ , the approximations are:  $(s + \rho)a_0 \approx (s + 1)a_0$  if  $0.5 < \rho < 1$  and  $(s + \rho)a_0 \approx sa_0$  if  $0 < \rho < 0.5$ .

Putting (7) into (6), we obtain the approximate expression for probabilities:

$$W_s = \frac{N_{sD}}{N_D} = \frac{e^{-\beta a_0 s}}{\sum_{s=1}^n e^{-\beta a_0 s}}. \tag{8}$$

This formula will be the basic for further analyses.

The first necessary step of the analysis is determining Lagrange multiplier  $\beta$ . It can be achieved by combining of (8) and the conservation law:

$$L = \sum_{s=1}^n sa_0 N_{sD}. \tag{9}$$

Including  $N_{sD}$  from (8) into (9), we obtain

$$\frac{L}{N_D a_0} = \frac{\sum_{s=1}^n s e^{-\beta a_0 s}}{\sum_{s=1}^n e^{-\beta a_0 s}} = -\frac{d}{d(\beta a_0)} \ln \sum_{s=1}^n e^{-\beta a_0 s} = -\frac{d}{d(\beta a_0)} \ln \left( e^{-\beta a_0} \frac{1 - e^{-\beta a_0 n}}{1 - e^{-\beta a_0}} \right). \tag{10}$$

In order to simplify equality (9), we shall take approximately:  $e^{-\beta a_0 n} \approx 0$ . After that (9) reduces to:

$$\frac{L}{N_D a_0} = \frac{d}{d(\beta a_0)} \ln(e^{\beta a_0} - 1) = \frac{1}{1 - e^{-\beta a_0}}. \tag{11}$$

From (11), it follows

$$e^{-\beta a_0} = 1 - \frac{N_D a_0}{L} \tag{12}$$

and further

$$-\beta a_0 = \ln \left( 1 - \frac{N_D a_0}{L} \right). \tag{13}$$

The next approximation is  $\ln(1 - \frac{N_D a_0}{L}) \approx -\frac{N_D a_0}{L}$  and it gives the estimate for  $\beta$ :

$$\beta = \frac{N_D}{L}. \tag{14}$$

In the described way, we estimated Lagrange multiplier  $\beta$  through fundamental characteristics of the chain, i.e., through number of its atoms  $N$  and through its length  $L$ . Naturally, it must be kept in mind that expression for  $\beta$  is approximate.

It is of interest to find the average value of distance  $a_s$  since this average values is the lattice constant of an equivalent chain with identical distances between atoms. This average value is given by:

$$\langle a_s \rangle = \sum_{s=1}^n a_s W_s = \frac{\sum_{s=1}^n a_0 s e^{-\beta s a_0}}{\sum_{s=1}^n e^{-\beta a_0 s}} = -\frac{d}{d\beta} \ln \sum_{s=1}^n e^{-\beta a_0 s} = -\frac{d}{d\beta} \ln \frac{1 - e^{-n\beta a_0}}{e^{-\beta a_0} - 1} \tag{15}$$

where from it follows:

$$\langle a_s \rangle = a_{\text{eq}} = a_0 \left( \frac{e^{\frac{\beta a_0}{2}}}{2 \sinh \frac{\beta a_0}{2}} - \frac{n}{e^{n\beta a_0} - 1} \right), \quad \beta = \frac{N_D}{L}. \tag{16}$$

The generalisation of exposed approach to three-dimensional structure gives the following result:

$$W_{\text{TOT}} = \frac{e^{-\beta_x a_0 s - \beta_y b_0 s - \beta_z c_0 s}}{\sum_{s=1}^{n_x} e^{-\beta_x a_0 s} \sum_{s=1}^{n_y} e^{-\beta_y a_0 s} \sum_{s=1}^{n_z} e^{-\beta_z a_0 s}} \tag{17}$$

where  $\beta_x = \frac{N_{xD}}{L_x}$ ,  $\beta_y = \frac{N_{yD}}{L_y}$ ,  $\beta_z = \frac{N_{zD}}{L_z}$  and  $a_0, b_0$  and  $c_0$  are the minimal distances between neighbours in  $x, y$  and  $z$  directions, respectively.

### 3 The Chain with Impurities Having Different Masses. The Mass Distribution Density

We shall now consider the chain with  $N_M$  equidistant atoms having different masses  $m_1, m_2, m_m$ . It will be assumed that mass  $m_1$  have  $N_{1M}$  atoms, mass  $m_2$  have  $N_{2M}$  atoms and so on. In this case, the number of atoms  $N$  as well as the mass  $M$  of the chain are conserved [14, 15]. It means that conservation laws are:

$$\sum_{s=1}^m N_{sM} = N_M \tag{18}$$

and

$$\sum_{s=1}^m m_s N_{sM} = M. \tag{19}$$

The statistical probability is given by:

$$P = \frac{N_M!}{\prod_{s=1}^m N_{sM}!} = \frac{N_M^{N_M}}{\prod_{s=1}^m N_{sM}^{N_{sM}}}. \tag{20}$$

As in previous case, the most probable distribution is obtained by equating of variation of the function

$$\Psi = \ln P - \alpha_M N_M - \gamma M = N_M \ln N_M - \sum_{s=1}^m (N_{sM} \ln N_{sM} + \alpha_M N_{sM} + \gamma m_s N_{sM}) \tag{21}$$

with zero.

We shall not repeat evaluations from previous section because they are practically the same.

The probability that  $N_s$  atoms have the mass  $m_s$  is given by:

$$\Omega_s = \frac{e^{-\gamma m_s}}{\sum_{s=1}^m e^{-\gamma m_s}} \tag{22}$$

Using the incommensurate approximation for masses:

$$m_s = sm_0 \tag{23}$$

where  $m_0$  is the minimal mass, we obtain the approximate expression for probability  $\Omega_s$

$$\Omega_s = \frac{N_s M}{N_M} = \frac{e^{-\gamma m_0 s}}{\sum_{s=1}^m e^{-\gamma m_0 s}} \tag{24}$$

Combining (19) and (24) and using the same approximations as in Sect. 2, we obtain the estimate for Lagrangian multiplier  $\gamma$ :

$$\gamma = \frac{N_M}{M} \tag{25}$$

The average value of the mass, which represents the mass of equivalent chain having identical masses, is given by:

$$\langle m_s \rangle = m_{eq} = m_0 \left( \frac{e^{\frac{\gamma m_0}{2}}}{2 \sinh \frac{\gamma m_0}{2}} - \frac{m}{e^{\gamma m_0} - 1} \right), \quad \gamma = \frac{N_M}{M} \tag{26}$$

The expression for three-dimensional structure with different masses is given by:

$$\Omega_{TOT} = \frac{e^{-\gamma_x m_{x0} s - \gamma_y m_{y0} s - \gamma_z m_{z0} s}}{\sum_{s=1}^{m_x} e^{-\gamma_x m_{x0} s} \sum_{s=1}^{m_y} e^{-\gamma_y m_{y0} s} \sum_{s=1}^{m_z} e^{-\gamma_z m_{z0} s}} \tag{27}$$

where  $\gamma_x = \frac{N_x M}{M_x}$ ,  $\gamma_y = \frac{N_y M}{M_y}$ ,  $\gamma_z = \frac{N_z M}{M_z}$  and  $m_{x0}$ ,  $m_{y0}$  and  $m_{z0}$  are the minimal masses in  $x$ ,  $y$  and  $z$  directions, respectively.

### 4 The Chain with Vacancies and Different Mass Impurities

The main goal of our considerations is the distribution function of the chain with different masses and with different distances between masses. This chain will be considered as a structure obtained by imbuing of the chains described in Sects. 2 and 3.

The distribution densities of this complex system can be obtained by means of convolution of distribution densities  $W_s$  and  $\Omega_s$  [16–18].

In accordance with mathematical statistics rules, the resulting spatial distribution density represents the sum of kernels  $W_{s-\mu}$  over all probabilities  $\Omega_\mu$ , i.e.

$$R_s = N_R \sum_{\mu=1}^s W_{s-\mu} \Omega_\mu \tag{28}$$

while the resulting mass distribution density represents the sum of kernels  $\Omega_{s-\mu}$ , i.e.

$$\Phi_s = N_\Phi \sum_{\mu=1}^s \Omega_{s-\mu} W_\mu. \tag{29}$$

In (28) and (29), normalising constants are denoted with  $N_R$  and  $N_\Phi$ , respectively.

We shall find the spatial distribution density  $R_s$ , first. Combining (8) and (24), we obtain:

$$R_s = N_R \sum_{\mu=1}^s e^{-(s-\mu)x} e^{-\mu y} \tag{30}$$

where

$$x = \beta a_0 \tag{31}$$

and

$$y = \gamma m_0. \tag{32}$$

After simple calculation, we obtain (from (30)) the normalised spatial distribution density (spatial probability) in the following form:

$$R_s = \frac{(e^x - 1)(e^y - 1)}{(e^y - 1)(1 - e^{-nx}) - (e^x - 1)(1 - e^{-my})} (e^{-xs} - e^{-ys}). \tag{33}$$

By combining (8) and (24) with (29) we obtain:

$$\Phi_s = N_\Phi \sum_{\mu=1}^s e^{-(s-\mu)y} e^{-\mu x} \tag{34}$$

where from follows the formula for mass distribution density (mass probability):

$$\Phi_s = \frac{(e^x - 1)(e^y - 1)}{(e^x - 1)(1 - e^{-my}) - (e^y - 1)(1 - e^{-nx})} (e^{-ys} - e^{-xs}). \tag{35}$$

By means of the formula (33), we shall determine average distance between atoms in complex chains with different distance. It will be a lattice constant of the chain, which is equivalent to described complex chain with different distances and different masses. This average value is given by:

$$\bar{a}_s = \sum_{s=1}^n a_s R_s = a_0 \frac{(e^x - 1)(e^y - 1)}{(e^y - 1)(1 - e^{-nx}) - (e^x - 1)(1 - e^{-my})} \left( \sum_{s=1}^n s e^{-xs} - \sum_{s=1}^n s e^{-ys} \right). \tag{36}$$

After calculations of sums in (36), we obtain the final expression in the form:

$$\begin{aligned} \bar{a}_s &= a_0 \frac{(e^x - 1)(e^y - 1)}{(e^y - 1)(1 - e^{-nx}) - (e^x - 1)(1 - e^{-my})} \\ &\times \left( \frac{1 - e^{-xn}}{4 \sinh^2 \frac{x}{2}} - \frac{1 - e^{-yn}}{4 \sinh^2 \frac{y}{2}} + \frac{ne^{-yn}}{e^y - 1} - \frac{ne^{-xn}}{e^x - 1} \right). \end{aligned} \tag{37}$$

The calculation of the average value  $\overline{a_s^{-1}}$  is of practical interest. If in complex chains, instead of neutral atoms are located ions, then between them are acting Coulomb interactions, which are proportional to reciprocal distance. The determination of  $\overline{a_s^{-1}}$  enables us to find the equivalent value of Coulomb interactions between neighbour ions.

The average value  $\overline{a_s^{-1}}$  is given by:

$$\begin{aligned} \overline{a_s^{-1}} &= \sum_{s=1}^n a_s^{-1} R_s = \frac{N_R}{a_0} \sum_{s=1}^n \left( \frac{1}{s} e^{-xs} - \frac{1}{s} e^{-ys} \right) \\ &= \frac{N_R}{a_0} \left( \int dy \sum_{s=1}^n e^{-ys} - \int dx \sum_{s=1}^n e^{-xs} \right) = \frac{N_R}{a_0} \left( \int dy \frac{1 - e^{-ny}}{e^y - 1} - \int dx \frac{1 - e^{-nx}}{e^x - 1} \right), \\ N_R &= \frac{(e^x - 1)(e^y - 1)}{(e^y - 1)(1 - e^{-nx}) - (e^x - 1)(1 - e^{-my})}. \end{aligned} \tag{38}$$

The integrals figuring in (38) can be solved in the following way. The integral

$$J = \int d\xi \frac{1 - e^{-n\xi}}{e^\xi - 1} \tag{39}$$

after substitution  $\xi = \ln t$  becomes

$$J = \int \frac{dt}{t} \frac{1 - t^{-n}}{t - 1} = \int \frac{dt}{t(t - 1)} - \int \frac{dt}{t^{n+1}(t - 1)}. \tag{40}$$

The function  $\frac{1}{t^{n+1}(t-1)}$  can be expressed as follows:

$$\frac{1}{t^{n+1}(t - 1)} = \frac{1}{t - 1} - \frac{1}{t} - \sum_{s=2}^{n+1} \frac{1}{t^s} \tag{41}$$

so that (40) becomes:

$$J = \int dt \sum_{s=2}^{n+1} t^{-s} = - \sum_{s=2}^{n+1} \frac{1}{s-1} t^{1-s} = - \sum_{v=1}^{n+1} \frac{1}{v} t^{-v}. \tag{42}$$

Taking into account that  $t = e^\xi$  we finally have:

$$J = \sum_{v=1}^{n+1} \frac{1}{v} e^{-v\xi}. \tag{43}$$

By means of (43), we obtain from (38):

$$\overline{a_s^{-1}} = \frac{1}{a_0} \frac{(e^x - 1)(e^y - 1)}{(e^y - 1)(1 - e^{-nx}) - (e^x - 1)(1 - e^{-my})} \sum_{v=1}^n \frac{e^{-vx} - e^{-vy}}{v}. \tag{44}$$

Reciprocal mass appears in phonon theories. The kinetic energy of system of oscillators, for example, is given as  $\frac{p^2}{2m}$ , where  $p$  is momentum and  $m$  is mass of atom (molecule). It is

interesting, therefore, to find the expression for reciprocal equivalent mass of the complex chain. It can be obtained by the formula

$$\begin{aligned} \overline{m_s^{-1}} &= \sum_{s=1}^n m_s^{-1} R_\Phi = \frac{N_\Phi}{m_0} \sum_{s=1}^m \left( \frac{1}{s} e^{-ys} - \frac{1}{s} e^{-xs} \right) \\ &= \frac{N_\Phi}{m_0} \left( \int dx \sum_{s=1}^m e^{-xs} - \int dy \sum_{s=1}^m e^{-ys} \right) = \frac{N_\Phi}{m_0} \left( \int dx \frac{1 - e^{-mx}}{e^x - 1} - \int dy \frac{1 - e^{-my}}{e^y - 1} \right), \\ N_\Phi &= \frac{(e^x - 1)(e^y - 1)}{(e^x - 1)(1 - e^{-my}) - (e^y - 1)(1 - e^{-mx})}. \end{aligned} \tag{45}$$

By the same calculation procedure as in previous case, we obtain the expression for equivalent reciprocal mass:

$$\overline{m_s^{-1}} = \frac{1}{m_0} \frac{(e^x - 1)(e^y - 1)}{(e^x - 1)(1 - e^{-my}) - (e^y - 1)(1 - e^{-nx})} \sum_{v=1}^n \frac{e^{-vy} - e^{-vx}}{v}. \tag{46}$$

The result obtained in Sects. 2 and 3, especially the formulae (8), (24), (33) and (35), give the possibility to find what ideal structures can represent the corresponding deformed ones. These representations cannot pretend to reproduce behaviour of deformed structure exactly. They rather give estimates of behaviour of deformed structure. The obtained probabilities are of exponential type and the standard deviations for this type of distributions are of the order of expectable (average) values. Since standard deviation represents the measure of deflection from expectable value, in the worst case, the error can achieve 100%. Independently, the stochastic method exposed here is a unique possibility for obtaining a fairly well real picture of physical processes of deformed structure. It should be pointed out that having equivalent ideal bulk structure, we can analyse the nanostructure (films or nano-rods) cut off from equivalent ideal bulk. These give the possibility to estimate behaviour of corresponding deformed nanostructures.

### 5 Continual Approximation

In Sects. 2 and 3, incommensurate approximation was used and it gives the above quoted results.

Here we shall use more rough approximation, i.e. we shall go over to continual approximation.

In this approximation

$$a_s \rightarrow \xi, \quad \sum_{s=1}^n \rightarrow \int_0^\infty d\xi, \quad m_s \rightarrow \eta, \quad \sum_{s=1}^m \rightarrow \int_0^\infty d\eta. \tag{47}$$

The calculations in continual approximation (47) are simple, and therefore, we shall quote only final results of some important formulas from Sects. 2 and 3. These final results are:

$$W(\xi) = \beta e^{-\beta\xi}, \tag{48}$$

$$\langle \xi \rangle = \int_0^\infty d\xi \xi e^{-\beta\xi} = \frac{1}{\beta}, \tag{49}$$



$$\Omega(\eta) = \gamma e^{-\gamma\eta}, \tag{50}$$

$$\langle \eta \rangle = \int_0^\infty d\eta \eta e^{-\gamma\eta} = \frac{1}{\gamma}. \tag{51}$$

For the complex lattice with spatial, as well as mass distribution the convolution distribution densities are

$$R(\xi) = \frac{\beta\gamma}{\beta - \gamma} (e^{-\gamma\xi} - e^{-\beta\xi}) \tag{52}$$

and

$$\Phi(\eta) = \frac{\beta\gamma}{\gamma - \beta} (e^{-\beta\xi} - e^{-\gamma\xi}). \tag{53}$$

Average distances are given by:

$$\bar{\xi} = \int_0^\infty d\xi \xi R(\xi) = \frac{\beta + \gamma}{\beta\gamma} \tag{54}$$

while for averages masses we have the formula

$$\bar{\eta} = \int_0^\infty d\eta \eta \Phi(\eta) = \frac{\beta + \gamma}{\beta\gamma}. \tag{55}$$

Average reciprocal distances are:

$$\overline{\xi^{-1}} = \int_0^\infty d\xi \xi^{-1} R(\xi) = \frac{\beta\gamma}{\beta - \gamma} \ln \frac{\beta}{\gamma} \tag{56}$$

while average reciprocal masses are given by:

$$\overline{\eta^{-1}} = \int_0^\infty d\eta \eta^{-1} \Phi(\eta) = \frac{\beta\gamma}{\gamma - \beta} \ln \frac{\gamma}{\beta} = \overline{\xi^{-1}}. \tag{57}$$

It is interesting to note that average distances and average masses are equal for the complex lattice. The same is valid for reciprocal distances and reciprocal masses.

The results obtained are less real than the corresponding results obtained in Sects. 2 and 3, where random variables were discrete ones, but they can serve for quick estimation of processes in deformed structures.

### Conclusion

The ideas as well as obtained results of stochastical approach of spatially deformed and mass deformed bulk structures can be summarized as follows:

1. The stochastical approach is a unique possibility to obtain a so–so real picture of behaving spatially and mass deformed structures.
2. The obtained distribution densities (probabilities) serve for translation of deformed structures into equivalent ideal structures. Thin films of nanorods can be cut off from equivalent ideal structures. The results of analysis cut off from ideal nanostructures give the possibility for estimation of the processes in the corresponding films and nanorods with defects. In Sects. 2 and 3, incommensurate approximation was used, while in Sect. 4, it is used as a more rough continual approximation.

3. Stochastic method is applied to the chain of identical atoms with random distances as well as to the chain with identical distances between atoms with random masses. The investigations were generalized to the complex lattices, which are spatially and mass deformed. The probabilities for spatially and mass deformed lattice are obtained by means of a convolution method.
4. All mentioned probabilities are of exponential type and this means that maximal error in application of obtained formulas can be 100%.
5. Quick estimation was used for continual approximation.

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