Modelling of pyroelectric response in inhomogeneous ferroelectric-semiconductor films

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Abstract. We have modified the Landau-Khalatnikov approach and shown that the pyroelectric response of inhomogeneous ferroelectric-semiconductor films can be described using six coupled equations for six order parameters: average displacement, its mean-square fluctuation and correlation with charge defect density fluctuations, average pyroelectric coefficient, its fluctuation and correlation with charge defect density fluctuations. Coupled equations demonstrate the inhomogeneous reversal of pyroelectric response in contrast to the equations of the Landau-Khalatnikov type, which describe the homogeneous reversal with the sharp pyroelectric coefficient peak near the thermodynamic coercive field value. Within the framework of our model, the pyroelectric hysteresis loop becomes much smoother, thinner and lower as well as the pyroelectric coefficient peaks near the coercive field completely disappear with increase in disordering caused by defects. This effect is similar to the well-known "square to slim transition" of the ferroelectric hysteresis loops in relaxor ferroelectrics. Also the increase of defect concentration leads to the drastic decrease of the coercive field typical for disordered ferroelectrics. Usually pyroelectric hysteresis loops of doped and inhomogeneous ferroelectrics have typical smooth shape without any pyroelectric coefficient peaks, and the coercive field values much lower than the thermodynamic one. Therefore, our approach qualitatively explains available experimental results. A rather well quantitative agreement between our modelling and typical Pb(Zr,Ti)O3 film pyroelectric and ferroelectric loops has been obtained.

Keywords: ferroelectric-semiconductor, charged defect, coercive field, pyroelectric hysteresis loop, coupled equations.

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1. Introduction

The main peculiarity of ferroelectric materials is hysteresis of their dielectric permittivity \( \varepsilon \), spontaneous displacement \( D \) and pyroelectric coefficient \( \gamma \) over electric field \( E_0(t) \) applied to the sample [1]. Dielectric \( \varepsilon(E_0) \), ferroelectric \( D(E_0) \) and pyroelectric \( \gamma(E_0) \) hysteresis loops in an inhomogeneous ferroelectric-semiconductor film have several characteristic features depicted in Fig. 1a.

Nowadays such typical ferroelectric-semiconductors as BaTiO3 ceramics, Pb(Zr,Ti)O3 solid solutions slightly doped with Fe, Cr or La, Nb, Nd, Ce etc., their films, multilayers and heterostructures are widely used in actuators, electro-optic, piezoelectric, pyroelectric sensors and memory elements [2-4]. However, the task how to create ferroelectric material with pre-determined dielectric and/or pyroelectric properties is solved mainly empirically. The correct theoretical consideration and modelling of related problems seems rather useful both for science and applications. It could answer fundamental questions about the nature of lattice-defects correlations, possible self-organization in the system and help to tailor new ferroelectric-semiconductor materials, save time and expenses.
Fig. 1. Dielectric \( \varepsilon(E_0) \), ferroelectric \( D(E_0) \) and pyroelectric \( \gamma(E_0) \) hysteresis loops. Different plots correspond to the data obtained for a semiconductor-ferroelectric film (a), the Landau-Khalatnikov model (b) and our coupled equations (c) for a bulk sample \( (\omega_1 << \omega_2 \) are two frequencies of applied electric field). Note that the Landau-Khalatnikov thermodynamic coercive field \( E_c \) is at least several times greater than the experimentally observed values \( E_{C1} \) and \( E_{C2} \) (sometimes the loops observed in the films with the thickness from 50 nm to 5 \( \mu \)m are shifted on the value \( E_i = E_{C2} - E_{C1} \), the shift being time-dependent).

Conventional phenomenological approaches \([5, 6]\) with material parameters obtained from first-principle calculations \([7, 8]\) can be successfully applied to the pure ferroelectrics materials \([9, 10]\), but they give a significantly incomplete picture of the dielectric, ferroelectric and pyroelectric hysteresis in the doped or inhomogeneous ferroelectrics-semiconductors (compare Fig. 1b and a).

In particular, the pioneer Landau-Khalatnikov approach evolved for the single domain pure ferroelectrics-dielectrics \([11, 12]\) describes homogeneous polarization reversal but presents neither domain pinning nor domain nucleation and domain movement. The calculated values of the thermodynamic coercive field \([1]\) are significantly larger than its experimental values much lower than the thermodynamic ones. Therefore, the modification of the Landau-Khalatnikov approach for inhomogeneous ferroelectrics-semiconductors seems necessary \([18]\).

In our recent papers \([19-23]\), we have considered the displacement fluctuations and extrinsic conductivity caused by charged defects and thus have modified the Landau-Khalatnikov approach for the inhomogeneous ferroelectrics-semiconductors. The derived system of three coupled equations gives the correct description of dielectric and ferroelectric hysteresis loop shapes and experimentally observed coercive field values (see Fig. 1c). In this paper, we develop the proposed model \([20]\) for pyroelectric response and demonstrate that the pyroelectric hysteresis loops of ferroelectrics-semiconductor films with charged defects can be successfully described using six coupled equations.

2. Coupled equations

Let us consider \( n \)-type ferroelectric-semiconductor with sluggish randomly distributed defects. The charge density of defects \( \rho_s(r) \) is characterized by the positive average value \( \rho_S \) and random spatial fluctuations \( \Delta \rho_s(r) \), i.e. \( \rho_s(r) = \rho_S + \delta \rho_s(r) \). Hereinafter the dash designates the averaging over the sample volume. The charged defect distribution is quasi-homogeneous. The average distance between defects is \( d \). Movable screening clouds \( \delta n(r,t) \) with the Debye screening radius \( R_D \) surround each charged center, so the free carriers charge density \( n(r,t) = \bar{n} + \delta n(r,t) \) is characterized by the negative average value \( \bar{n} \) and modulation \( \delta n(r,t) \).

Screening clouds are deformed in the external field \( E_0 \), and the system "defect center \( \delta \rho_s + \) screening cloud \( \delta n \)" causes displacement fluctuations \( \delta D(r,t) \) in accordance with Maxwell’s equations \( \text{div} \vec{D} = 4\pi n + \rho_s \), (see Fig. 2 in our paper), Fig. 1 in \([20]\) and Fig. 3.2 in \([5]\).

In our papers \([20-22]\), we have modified the classical Landau-Khalatnikov equation \( \Gamma \delta D/\delta t + aD + \beta \delta D^2/\delta r^2 = E_z \) for bulk samples and obtain the system of coupled equations for the average displacement \( \vec{D} \), its mean-square fluctuation \( \overline{\delta D^2} \) and correlation with charge defects density fluctuations \( \overline{\delta D \delta \rho_s} \). The average pyroelectric coefficient \( \bar{\gamma} \) can be easily obtained by the differentiation of the coupled system \([20]\) with respect to the temperature \( T \). We suppose that only the coefficient \( \alpha = -\alpha_T (T - T_c) \) and screening radius \( R_D \approx \sqrt{k_B T/4\pi n e e} \) essentially depend on temperature in the usual form \( \partial \alpha/\partial T = \alpha_T \) \([1]\).
\[ \frac{\partial D}{\partial t} = \frac{k_B}{4\pi\varepsilon e} [24, 25]. \] In this way, we obtained six coupled equations for the average displacement \( \overline{D} \), its mean-square fluctuation \( \overline{D^2} \) and correlation with charge density \( \rho \), pyroelectric coefficient \( \gamma = \overline{D}/\overline{D T} \), its deviation \( \overline{\gamma} = \overline{D^2}/\overline{D T} \) and correlation with charge defect density fluctuations \( \overline{\gamma \rho \delta} = \overline{D \delta \rho / \overline{D T}} \) (see Appendix A):

\[ \Gamma \frac{\partial \overline{D}}{\partial t} + \left( \alpha + 3 \beta \overline{D^2} \right) \overline{D} + \beta \overline{D^3} = E_0(t) + E_i(l, t), \]  

(1)

\[ \frac{\Gamma_R}{2} \frac{\partial \overline{D^2}}{\partial t} + \left( \alpha_R + 3 \beta_R \overline{D^2} \right) \overline{D^2} + \beta \overline{D^3} = E_0(t) \left( \frac{\partial \overline{D \rho}}{n} - \partial \varepsilon \right) + \frac{4\pi k_B T}{\mu e} \overline{\delta \rho \delta}, \]  

(2)

\[ \Gamma_R \frac{\partial \overline{D \rho}}{\partial t} + \left( \alpha_R + 3 \beta_R \overline{D^2} + \beta \overline{D^3} \right) \overline{D \rho} = \]  

(3)

\[ \Gamma_R \frac{\partial \overline{\gamma}}{\partial t} + \left( \alpha + 3 \beta \overline{D^2} + 3 \beta \overline{D^3} \right) \overline{\gamma} = \alpha_T + 3 \beta \overline{\gamma} \overline{D}, \]  

(4)

\[ \Gamma_R \frac{\partial \overline{\gamma \rho \delta}}{\partial t} + \left( \alpha_R + 2 \beta_R \overline{D^2} + 3 \beta \overline{D^3} \right) \overline{\gamma \rho \delta} = \]  

(5)

The built-in electric field \( E_i(l, t) = \frac{4\pi Z}{l} (\delta n + \delta \rho), \) and \( E_t \) is time-dependent, its amplitude is proportional to the space charge fluctuations \( \delta n + \delta \rho \). Also, \( E_t \) diffuses paraelectric-ferroelectric phase transition, in particular, it shifts and smears dielectric permittivity maximum. Thus, the ferroelectric film thickness decrease leads to the degradation of their ferroelectric and dielectric properties [18] up to the appearance of relaxor features [28].

Bratkovsky and Levanyuk [10] predicted the existence of built-in field in a finite ferroelectric film within the framework of phenomenological consideration. The field had a rather general nature. Our statistical approach confirms their assumption and gives the possible expression of the field existing in the inhomogeneous ferroelectric-semiconductor film. The inner electric field induced by the film-substrate misfit strain was calculated in our recent paper [29]. Misfit-induced field could be the main reason of the loop shift only in the ultrathin strained films with the thickness less than 50 nm.

The renormalization \( \Gamma_R = \Gamma + \tau_m \) of the Khalatnikov kinetic coefficient in (2), (3), (5), (6) is connected with the contribution of free carrier relaxation with the characteristic time \( \tau_m = 1/4\pi\mu e \) (\( m \) is the n-carrier mobility). Usually, the time \( \tau_m \sim 10^{-2} \text{ s} \) is much smaller than the initial coefficient \( \Gamma \approx 10^{-3} \text{ s} \) for the typical impurity concentration (0.01–1)% and room temperatures [5]. Thus \( \Gamma \approx \Gamma_R \).

The renormalization of coefficients \( \alpha_R = \alpha + (\chi + k_B T/4\pi\mu e)/d^2 \) and \( \alpha_{RT} = \alpha_T + k_B/4\pi\mu e d^2 \) in (2), (3), (5), (6) is connected with the contribution of correlation and screening effects [21].
The coefficient \( \alpha = -\alpha_T (T_C - T) \) is negative in the perfect ferroelectric phase without random defects (\( \delta p_s^2 = 0 \)). For the partially disordered ferroelectric with charged defects (\( \delta p_s^2 > 0 \)), the coefficient \( \alpha_R \) is positive and \( \alpha_R \gg |\beta|, \alpha_R \gg \alpha_T \). For example, for Pb(Zr,Ti)O₃ solid solution \( \alpha \sim -(0.4 \ldots 2) \times 10^{-2} \) [3], the gradient term \( \chi = 5 \times 10^{-16} \text{cm}^2 \), the screening radius \( R_D \sim [10^{-6} \ldots 10^{-4}] \text{cm} \) [5], the average distance between defects \( d \sim [10^{-6} \ldots 10^{-4}] \text{cm} \) and thus \( \alpha_R \sim 1 \ldots 10^2 \). So, the ratio \( \xi = -\alpha_R / \alpha = \alpha_T \) is greater than 100.

The sources of fluctuations in (2), (5) originate from the correlation between diffusion current and display cement fluctuations: 
\[
-\overrightarrow{\partial D} \cdot \partial \rho_s / \overrightarrow{\partial z} \approx \delta \overrightarrow{\partial \rho_s} / \overrightarrow{\partial z} \sim \delta \overrightarrow{\rho_s} (\overrightarrow{\partial \rho_s} + \partial \overrightarrow{n})
\]
[20]. The dimensionless amplitude of these correlations \( g = 4 \eta k_B T_n \left| \overrightarrow{\partial D_s} \right| \) varies in the range from \( 10^2 \) to \( 10^3 \) for Pb(Zr,Ti)O₃ \( (D_s = \sqrt{\alpha / \beta}) \) is spontaneous displacement of the perfect material with \( \delta p_s^2 = 0 \). The positive correlator \( R^2 (t) = -\delta \overrightarrow{\rho_s} \partial \delta \overrightarrow{n} / \overrightarrow{n}^2 \) was calculated in [20] at small external field amplitude and low frequency. In a general case, the correlator \( R^2 (t) \) is time-dependent, but varies in the range (0; 1) because its amplitude is proportional to the charged defects disordering \( \overrightarrow{\partial \rho_s^2} / \overrightarrow{\rho_s^2} \) under the condition of the prevailing extrinsic conductivity \( \overrightarrow{n} = \overrightarrow{\rho_s} \) [20].

Hereinafter, we discuss only the system pyroelectric response near the equilibrium states. The equilibrium solution of (1)-(6) corresponds to the quasi-static external field changing, e.g., for the harmonic applied field \( E_0(t) = E_0 \sin(\omega t) \) the ratio \( \Gamma(\omega / a) \) should be smaller than unity. At low frequencies \( \omega \ll -\alpha / \Gamma \), the initial conditions do not play significant role in the ferroelectric and pyroelectric hysteresis loops shape. The system (1)-(6) quasi-equilibrium behavior is described by the dimensionless built-in field amplitude \( E_m = E_0 / E_C \) and the frequency \( \omega = -\Gamma \omega / a \) as well as by the aforementioned parameters \( \xi = R^2 (w), g \) and temperature \( T / T_C \) \( (E_C = -\alpha \sqrt{\beta / \alpha}) \) is proportional to the thermodynamic coercive field of the perfect material with \( \delta p_s^2 = 0 \). Keeping in mind that the carrier fluctuations \( \partial \delta (t) \) are time-dependent, their frequency spectrum \( \partial \delta (w) \) determines the frequency dispersion of parameters \( R^2 (w) \sim \partial \delta (w), E_m (w) \sim (\partial n + \partial \rho_s) \). It is appeared that, under the conditions \( w < 1, g \gg 1 \) and \( \xi \gg 1 \), the scaling parameter \( g R^2 / \xi \) determines the system behavior.

Fig. 3a demonstrates the typical changes of pyroelectric hysteresis loop caused by the increase of charged defect disordering (note, that \( g R^2 / \xi \sim \delta \overrightarrow{\rho_s^2} / \overrightarrow{\rho_s^2} \)).

Figs 3b,d demonstrate that the disorder parameter \( \delta \overrightarrow{n} \approx -\partial D^2 \) exists in all the region of pyroelectric hysteresis and reaches its maximum value near the coercive field, where \( D \rightarrow 0 \) and \( \overrightarrow{n} \rightarrow 0 \). Let us underline, that in contrast to the Landau-Khalatnikov equations describing homogeneous reversal with \( \overrightarrow{n} \approx 2 \) near the coercive field, our coupled equations demonstrate inhomogeneous ferroelectric and pyroelectric response reversal [20]. Namely, when the external field reaches the coercive value the sample splits into the oppositely polarized regions, so that it is non-polarized as a whole. Every polarized region causes the pyroelectric coefficient fluctuations \( \delta \overrightarrow{n} \).

It is clear from Figs 3a,c that the increase of the \( g R^2 / \xi \) value leads to the essential decrease and smearing of pyroelectric coefficient peaks near coercive field and to the decrease of the coercive field value (compare the Landau-Khalatnikov loops \( (R^2 = 0) \) with dashed curves \( (R^2 \geq 0.2) \)). At \( R^2 \geq 0.5 \), the pyroelectric coefficient peaks near coercive field completely disappears and typical pyroelectric hysteresis loop looks like the ferroelectric one. At \( R^2 \geq 0.8 \), the coercive field is much smaller than its thermodynamic value at \( R^2 = 0 \). Thus, the pyroelectric loop becomes sloped, much thinner and little lower with the increase in charged defect disordering \( \delta \overrightarrow{\rho_s^2} \). This effect is similar to the well-known "square to slim transition" of the ferroelectric hysteresis loops in relaxor ferroelectrics [14].

It seems that the hysteresis loop width could only decrease when the donor concentration increases because the width decreases with \( g R^2 / \xi \) increasing. However, the parameter \( g R^2 / \xi \) decreases when acceptors (e.g., intrinsic vacancies or traps) compensate donors. The loop width increases under \( g R^2 / \xi \) decreasing. Such compensation is possible in “hard” Pb(Zr,Ti)O₃ (PZT) ceramics where donor impurities do not change the loops width [3].

Let us underline that we do not know any experiment, in which pyroelectric coefficient peaks near coercive field were observed. Moreover, usually pyroelectric hysteresis loops in doped ferroelectrics have typical “slim” shape with coercive field values much lower than the thermodynamic one [30, 31]. So, our
approach qualitatively explains available experimental results. The quantitative comparison of our numerical modelling with typical PZT pyroelectric loops is presented in the next section.

3. Comparison with experimental results and discussion

Dopants, as well as numerous unavoidable oxygen O\(^2\)\(^-\) vacancies, can play a role of randomly distributed charged defects in soft PZT. Really, it is known that the low level (0.001 – 1.0 %) donor additives La, Nb, Nd or Ce soften PZT dielectric and pyroelectric properties at room temperature. Ferroelectric and pyroelectric hysteresis loops have got relatively high $\gamma$ and $D$ remnant values, but reveal low coercive fields in soft PZT [3]. Usually pyroelectric hysteresis loops of PZT are rather thin and sloped even at the low frequencies $\omega \sim (0.1...10)$ Hz [3], any pyroelectric coefficient maximum near the coercive field is absent [30, 31].

Investigated Pt-PZT-Pt/Ti-SiO\(_2\)/Si structures with oriented PZT layer were manufactured by radio frequency magnetron sputtering in the system under the conditions described previously [17]. The sputtering target obtained by uniaxial cold pressing includes the mixture of PbO, TiO\(_2\) and ZrO\(_2\) in a stoichiometric composition. The structure includes the top 150 nm Pt electrode, 1.9 $\mu$m layer of oriented PZT, bottom Pt/Ti bilayer (150 nm of Pt, 10 nm of Ti) deposited onto the oxidized (350 nm of SiO\(_2\)) (100) $n$-type Si 350 $\mu$m substrate.

For PZT – Si substrate structure, it is necessary to design the bottom electrode, which possesses not only a stable and sufficiently high electrical conductivity but also simultaneously prevents the interfacial reactions between the electrode, PZT and SiO\(_2\) components in PZT film and Si substrate surroundings at rather high temperatures. The layer of Ti plays an important role in limiting the diffusion of Ti in Pt/Ti intermediate bilayer through Pt layer into the PZT layer and directly into the SiO\(_2\) layer, as well as in correction of poor adhesion of Pt layer. The annealing treatment of the Pt/TiO\(_x\)/SiO\(_2\)/Si substrate structure just before PZT deposition was performed for substrate stabilization, and post-annealing treatment of PZT film was performed to crystallize the

Fig. 3. Hysteresis loops of pyroelectric coefficient $\gamma(E)$ and its deviation $\delta\gamma(E)$ for different $R^2$ values. Other parameters: $g = 100$, $\xi = 100$, $T/T_C = 0.45$, $E_i = 0$, $w = 0.1$ (plots (a, b) for the bulk sample) and $E_m = \pm 0.1R$, $w = 0.05$ (plots (c,b) for the film).
film in the polar perovskite phase. The top Pt-electrode has 1 mm² area.

The pyroelectric response of the PZT films was registered by means of dynamic pyroelectric measurements (see [30] for details). During these measurements, the quasi-static voltage \( V \) varied in the range \((-11 \text{ V}, +11 \text{ V})\) at the low frequency \( \omega \sim 0.01 \text{ Hz} \), the temperature \( T \) changes near the room one with the frequency about 20 Hz. Pyroelectric hysteresis loops for \( U_{\pi 1}(V) \sim \tilde{\gamma} \) and \( U_{\pi 2}(V) \sim \tilde{\gamma}/\tilde{\varepsilon} \) were reconstructed from pyroelectric response amplitude \( U_{\pi 1,2}(V) \) and phase \( \varphi_{\pi 1,2}(V) \). Experimental loops and our calculations are presented in the Fig. 4.

It is clear from the figures that our model both qualitatively and quantitatively describes pyroelectric hysteresis loops in thick soft PZT films. Our modelling of ferroelectric and dielectric hysteresis loops was performed earlier (see e.g. [20]). Ferroelectric and dielectric hysteresis loops measured for the same PZT films by using the conventional Sawyer-Tower method and impedance analyzer are presented in Fig. 5. Note that a film capacity is proportional to the dielectric permittivity \( \tilde{\varepsilon} \) (see equations for \( \tilde{\varepsilon} \) in Appendix B).

It is clear from Figs 4-5 that ferroelectric loop is slightly asymmetrical, i.e., the built-in electric field \( E_i \neq 0 \), whereas the pyroelectric and dielectric loops measured at the lower frequency look almost symmetrical. This means that the built-in field is dynamic in the aforementioned experiments: its value is noticeable at higher external field frequency \( w = 0.15 \) and negligibly small at the low frequency \( w = 0.05 \). Note that sometimes even stabilized loop (i.e., obtained after many circles) is asymmetrical [29, 31]. Also, we obtained that disordering \( R^2(w) - \partial \rho_C/\partial \tilde{\gamma}(w) \) decreases with external field frequency increase, namely, it varies from 0.6 down to 0.4 under the frequency change from 0.05 up to 0.15 (see Figs 4, 5). This result could be explained allowing for the fact that the defect-carrier dipole coupling influences on the switching mechanism [8], but dipole correlations “sluggish defect \( \delta \rho \) + mobile fluctuation \( \delta \tilde{\gamma} \)” weaken with the frequency increase.

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![Fig. 4. Pyroelectric response of 1.9 μm-thick PZT(46/54):Nb film. Pyroelectric hysteresis loops (right plots for \( U_{\pi 1} \sim \tilde{\gamma} \) and \( U_{\pi 2} \sim \tilde{\gamma}/\tilde{\varepsilon} \) ) have been reconstructed from pyroelectric response amplitude and phase (left plots for \( U_{\pi 1,2} \) and \( \varphi_{\pi 1,2} \) ). Squares are experimental data measured by Bravina et al. [30], solid curves are our calculation with the fitting parameters \( w = 0.1 \), \( R^2 = 0.5 \), \( g = 100 \), \( \xi = 100 \), \( E_m = -0.03 \).](image-url)
Thus the modelling based on the coupled equations (1)-(6) gives the realistic coercive field values and typical pyroelectric hysteresis loop shape, in contrast to the Landau-Khalatnikov approach that describes the homogeneous pyroelectric response reversal. Taking into account that the inhomogeneous reversal of spontaneous polarization and pyroelectric response occurs in the doped or inhomogeneous ferroelectrics-semiconductors, the proposed coupled equations can be more relevant for the phenomenological description of their polar and pyroelectric properties than the models based on the Landau-Khalatnikov phenomenology.

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**Appendix A**

The expressions for correlations $\overline{\delta D \delta E_z}$, $\overline{\delta \rho_s \delta E_z}$ and $\overline{\delta^2 \delta D \delta E_z}$ were derived in [20] for the bulk sample. For a finite film with the thickness $l$ these acquire the form:

$$
\overline{\delta D \delta E_z} = \chi \left( \overline{\delta D \delta z} \right)_{x,y} = 4 \pi \chi \left( \overline{\delta \Sigma + \delta \rho_s} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2}
$$

$$
\overline{\delta \rho_s \delta E_z} = -\frac{1}{8 \pi \mu \overline{\delta D}} E_0 \left( \frac{\delta \rho \delta D}{\delta z} \right)_{x,y} - \frac{\kappa}{\mu l} \left( \overline{\delta \rho_s \delta \rho_s} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2}
$$

$$
\overline{\delta^2 \delta D \delta E_z} = -\frac{1}{8 \pi \mu \overline{\delta D}} E_0 \left( \frac{\delta \rho \delta D}{\delta z} \right)_{x,y} - \frac{\kappa}{\mu l} \left( \overline{\delta \rho_s \delta \rho_s} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2} + \frac{\kappa}{4 \pi \mu \overline{\delta D}} \left( \overline{\delta \rho_s \delta D} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2}
$$

If the charged defect distribution is quasi-homogeneous, expressions (B.1-3) can be simplified, namely

$$
\frac{4 \pi E_0}{l} \left( \overline{\delta \Sigma + \delta \rho_s} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2} = 4 \pi \chi \left( \overline{\delta \Sigma} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2},
$$

$$
\frac{\kappa}{\mu l} \left( \overline{\delta \rho_s \delta \rho_s} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2} = 0,
$$

$$
\frac{\kappa}{2 \pi \mu \overline{\delta D}} \left( \overline{\delta \rho_s^2} \right)_{x,y} \left| x \right|_{l/2}^{+1/2} \left| y \right|_{l/2}^{-1/2} = 0.
$$

The constant $z_0$ can be determined from the boundary conditions for $\overline{\delta D}_{x,y}$ at $z = 0$. Under the condition $z_0 = \pm l/2$, the built-in field deviation $\overline{\delta E_i}$ is absent.

**Appendix B**

The system determining the dielectric permittivity $\overline{\delta E} = \overline{\delta D}/\overline{\delta E_0}$, its deviation $\overline{\delta \epsilon} = \overline{\delta \delta D} \overline{\delta E_0}$ and correlation $\overline{\delta \epsilon \delta \rho_s} = \overline{\delta \delta D \delta \rho_s}/\overline{\delta E_0}$ has the form:

$$
\Gamma\overline{\delta E} = \left( \alpha + 3 \beta \overline{\delta \rho_s} \right) \overline{\delta D} = 1 - 3 \beta \overline{\delta \rho_s} \overline{\delta D}, \quad (B.1)
$$

$$
\Gamma_R \overline{\delta \delta \rho_s} = \left( \alpha_R + 2 \beta R \overline{\delta \rho_s} \right) \overline{\delta D} = \alpha_R \overline{\delta \rho_s} \overline{\delta D}, \quad (B.2)
$$

$$
\Gamma_R \overline{\delta \delta \rho_s} = \left( \alpha_R + \beta \overline{\delta \rho_s} \right) \overline{\delta \rho_s} = \alpha_R \overline{\delta \rho_s} \overline{\delta \rho_s}, \quad (B.3)
$$

The constant $z_0$ can be determined from the boundary conditions for $\overline{\delta D}_{x,y}$ at $z = 0$. Under the condition $z_0 = \pm l/2$, the built-in field deviation $\overline{\delta E_i}$ is absent.