LOCAL ELECTROMECHANICAL RESPONSE OF IONIC SEMICONDUCTOR THIN FILMS

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We report on the calculations of a local electromechanical response of ionic semiconductor thin films induced by local changes of the concentration of ions (*stoichiometric contribution)* and free electrons and holes (*electron-phonon interaction via the deformation potential*). Dynamic strain-voltage hysteresis loops are obtained for an ionic semiconductor thin film with mobile acceptors (donors) and holes (electrons).

In case of ion-blocking electrodes, changes in the hole (electron) concentration make a dominant contribution to the dependence of the mechanical displacement of the film surface on the voltage applied to the probe, which is directly registered by scanning probe microscopy (SPM) methods. Thus, the displacement of the ionic semiconductor surface can provide an important information on local changes of the charge state of acceptors (donors) and electron-phonon correlations via the deformation potential.

1. Introduction

Progress in information technologies depends on high-rate and high-capacity devices with energy-independent memory. At the present time, the intense investigations of various alternatives to modern memory units are aimed at the obtaining of more powerful and functional systems [1]. One of the promising concepts is that of resistive memory [2] based on semiconductors with mixed conduction types, for example, the ion-electron one [3, 4]. In thin films used in resistive memory cells, of principal importance is the electromigration and the diffusion of ions.

The theory of the effect of diffusion and electromigration on the mechanical properties of ionic semiconductors is not only of fundamental interest. It is also very important for the optimization of their electrophysical properties useful for many applications [1]. That is why, of high interest are resistive materials such as correlated oxides $La_{1-x}Sr_xCo_yFe_{1-y}O_{3-\delta}$, $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$, $La_{1-x}Sr_xCoO_{3-\delta}$, and $Sr(Fe_xTi_{1-x})O_{3-\delta}$ having specific electrophysical characteristics [5–7].

Measurements of local mechanical displacements of ionic-film surfaces can provide an important information on the diffusion and the migration of electrons and ions (acceptors, donors), local oxidation levels, electron-phonon interaction via the deformation potential [8] and even Jahn–Teller deformations in films of correlated oxides [9–11].

A local electromechanical response (local deformation of the ionic film surface) is caused by the diffusion and the electromigration of charge carriers (ions, vacancies, electrons, and holes) under the action of the nonuniform electric field of a scanning probe. Local displacements of the film surface are registered by the SPM equipment (Fig. 1).

In the case where a probe electrode and a substrate are ion-blocking, a change of the electron and hole concentrations by means of the electron-phonon coupling results in a mechanical displacement of the film surface induced by the electric voltage applied to the probe [12, 13]. The coupling constant is proportional to the deformation potential, which can be intensified, in turn, by Jahn–Teller local deformations existing in correlated oxides. This allows one to link the electromechanical response with the local deformation potential of correlated oxides [12, 13].

2. Motivation of the Work

A comprehensive analytic theory of electromigration and diffusion of charge carriers in ionic semiconductors and their thin films represents a complicated, incompletely solved problem [1–4, 14]. In particular, the volt-ampere characteristics were analyzed mainly numerically and only in the framework of the Boltzmann approximation for the chemical potential and/or the linear Debye screening theory in the approximation of constant conductivity independent of the particle concentration [3, 4, 15–18]. These approximations and assumptions are invalid in the regions of space-charge accumulation usually appearing close to surfaces and interfaces of ionic semiconductor films [19–21]. Considering ionics, one cannot neglect the electromigration of ions, as well as the

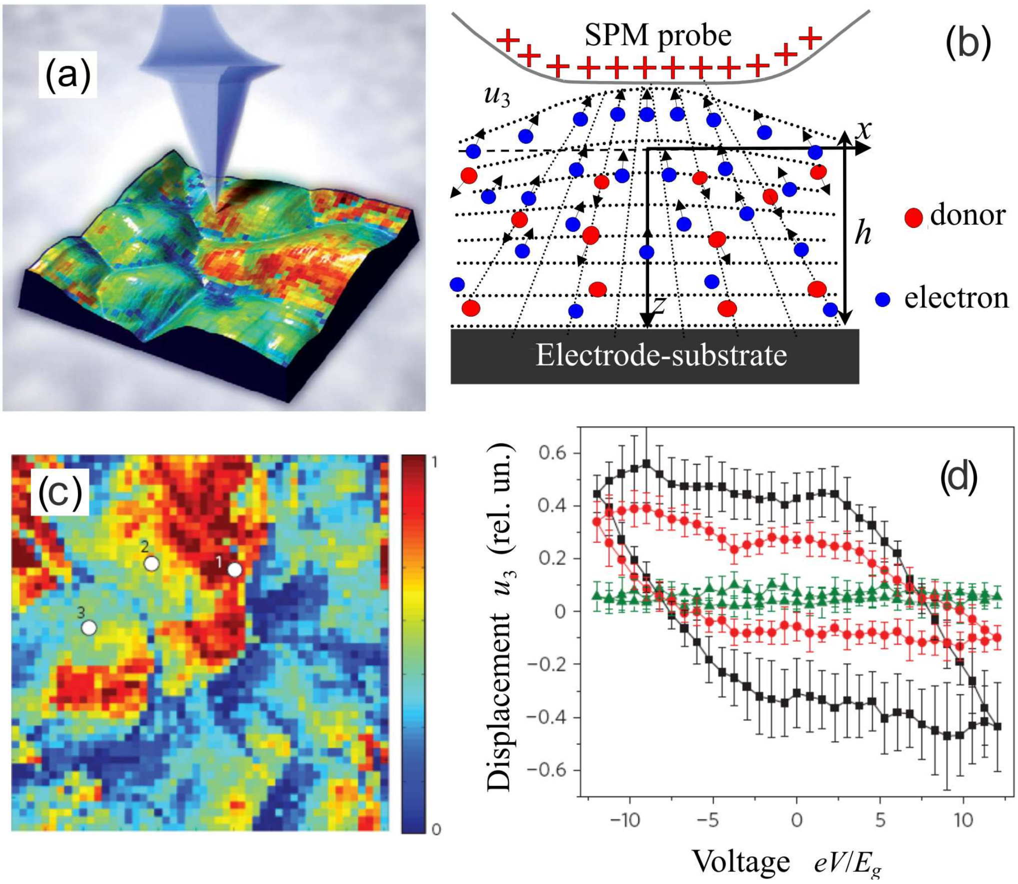

Fig. 1. (*a*) Diagram of measuring the local electromechanical response of an ionic with the help of a scanning probe [11], (*b*) [13]. Experimental results for a LiCoO$_2$ film: (*c*) amplitude maps of the SPM local electromechanical response and (*d*) hysteresis loops of the local electromechanical response of an ionic measured at various points of the map (*c*) [11]

mobility of acceptors (donors) and the intrinsic width of the impurity band [21].

The deformation of the film surface accompanying the ion electromigration and/or recharging is usually neglected in theoretical considerations [12, 13]. The deformation effect is very important for the operation of modern ion materials used for energy accumulation, as well as in memory units. However, the majority of studies devoted to this problem ignore the space charge layers formed during the ion diffusion and the origination of electric fields, whereas the main efforts are concentrated on the consideration of nonplanar geometry, which makes the problem very complex.

Based on the performed literature review, the following conclusions can be made:

1. Volt-ampere characteristics of ionic semiconductors and their thin films were theoretically analyzed mainly numerically and only in the framework of the common Boltzmann approximation in the linear drift-diffusion theory.

2. The local electromechanical response of ionic semiconductors measured by SPM methods has no theoretical description.

These facts have motivated our theoretical investigations. This work reports on the calculations of the local electromechanical response of an ionic semiconductor film with the energy state density typical of correlated oxides with mobile acceptors, donors, electrons, and holes. The obtained results were compared with those calculated in the Boltzmann approximation.

The aim of the work was to perform analytical and numerical calculations of the local electromechanical response of an ionic semiconductor film with the specified state density with mobile acceptors, donors, electrons, and holes and to compare the obtained results with those calculated in the Boltzmann approximation.

3. Problem Statement and Solution Techniques

The theoretical calculations were carried out in the framework of the classical theory of semiconductors, electrodynamics, and continuum mechanics.

The Maxwell equations for the electric potential φ of an ionic semiconductor film with mobile ions (acceptors or donors), electrons, and holes

$$\Delta\varphi(\mathbf{r},t) =$$

$$= -\frac{e}{\varepsilon_0\varepsilon}\left(N_d^+(\mathbf{r},t) - N_a^-(\mathbf{r},t) + p(\mathbf{r},t) - n(\mathbf{r},t)\right) \qquad (1)$$

were solved in the quasistatic approximation. Here, e denotes the elementary charge, ε is the static dielectric susceptibility of an ionic semiconductor, ε_0 is the universal dielectric constant, $n(\zeta) = \int_{-\infty}^{\infty} d\varepsilon\, g_n(\varepsilon)\, f\left(\varepsilon - \zeta + \Xi_{ij}^C u_{ij}\right)$ is the electron concentration in the conduction band, and $p(\zeta) = \int_{-\infty}^{\infty} d\varepsilon\, g_p(\varepsilon)\, f\left(\zeta - \Xi_{ij}^V u_{ij} - \varepsilon\right)$ is the hole concentration in the valence band with $\Xi_{ij}^{C,V}$ standing for the deformation potential of electrons and holes, respectively [22, 23]. In addition, $N_d^+(\zeta) = \int_{-\infty}^{\infty} d\varepsilon\, g_d(\varepsilon)\, f(\zeta - \varepsilon)$ is the concentration of ionized donors and $N_a^-(\zeta) = \int_{-\infty}^{\infty} d\varepsilon\, g_a(\varepsilon)\, f(\varepsilon - \zeta)$ is the concentration of negative acceptors, where the function $f(x) = \frac{1}{1+\exp(x/k_\mathrm{B}T)}$ is the Fermi–Dirac distribution function. The electrochemical potential $\zeta(z) = \mu + e\varphi(z)$ determines the equilibrium concentration of charge carriers as a function of the distance $\mathbf{r}$ to the film surface.

The equilibrium energy state density of charge carriers $g(\varepsilon, E_m, \delta E_m)$ was specified in the form of localized distributions with the width δE_m and different exponents $k > 1$, which is characteristic of strongly doped semiconductors [5]:

$$g(\varepsilon, E_m, \delta E_m) = g_m \exp\left(-\frac{|E_m - \varepsilon|^k}{\delta E_m^k}\right), \qquad (2)$$

where g_m is a constant, the indices $m = a, d, n, p$ mark acceptors, donors, electrons, and holes, respectively, and $k > 1$. For comparison and determination of the degree of degeneracy of electrons or holes, we also used the Boltzmann approximation, $g(\varepsilon, E_m, \delta E_m) \sim g_m \exp\left(-\frac{E_m - \varepsilon}{k_\mathrm{B}T}\right)$, instead of Eq. (2).

The boundary conditions for Eq. (1) corresponded to the geometry and properties of the electrodes:

$$\varphi(\rho, z = 0, t) = V_0(\rho)\exp(i\omega t), \quad \omega(z = h, \rho) = 0, \qquad (3)$$

where h is the film thickness, and $V_0(\rho)\exp(i\omega t)$ is the periodic potential distribution created by the scanning probe (see Fig. 2).

The kinetics of charge carriers was calculated from the continuity equations without regard for the processes of generation-recombination of “hot” carriers:

$$\frac{\partial N_d^+}{\partial t} + \frac{1}{e}\mathrm{div}\,\mathbf{J}_d = 0, \quad -\frac{\partial N_a^-}{\partial t} + \frac{1}{e}\mathrm{div}\,\mathbf{J}_a = 0, \qquad (4a)$$

$$\frac{\partial p}{\partial t} + \frac{1}{e}\mathrm{div}\,\mathbf{J}_p = 0, \quad -\frac{\partial n}{\partial t} + \frac{1}{e}\mathrm{div}\,\mathbf{J}_n = 0. \qquad (4b)$$

The ion and electron currents were supposed to be proportional to the gradients of their electrochemical potentials $\zeta_{a,d}(\mathbf{r})$ and Fermi quasilevels $\zeta_{p,n}(\mathbf{r})$:

$$\mathbf{J}_p = -e\eta_p p\,\mathrm{grad}\zeta_p,$$

$$-\zeta_p(\mathbf{r}) \approx +e\varphi(\mathbf{r}) + k_\mathrm{B}T\ln\left(\frac{p(\mathbf{r})}{p_0}\right), \qquad (5a)$$

$$\mathbf{J}_n = e\eta_n n\,\mathrm{grad}\zeta_n,$$

$$\zeta_n(\mathbf{r}) \approx -e\varphi(\mathbf{r}) + k_\mathrm{B}T\ln\left(\frac{n(\mathbf{r})}{n_0}\right). \qquad (5b)$$

$$\mathbf{J}_a = e\eta_a N_a^-\,\mathrm{grad}\zeta_a,$$

$$\zeta_a(\mathbf{r}) = -e\varphi(\mathbf{r}) - k_\mathrm{B}T\ln\left(\frac{N_a - N_a^-(\mathbf{r})}{N_a^-(\mathbf{r})}\right), \qquad (5c)$$

$$\mathbf{J}_d = -e\eta_d N_d^+\,\mathrm{grad}\zeta_d,$$

$$-\zeta_d(\mathbf{r}) = e\varphi(\mathbf{r}) - k_\mathrm{B}T\ln\left(\frac{N_d - N_d^+(\mathbf{r})}{N_d^+(\mathbf{r})}\right), \qquad (5d)$$

where η_m is the carrier mobility. The signs of approximate equality in Eqs. (5a) and (5b) correspond to the Boltzmann approximation. The approximation of the linear drift-diffusion theory was considered separately. The boundary conditions for the currents on the film surface were considered in two limit cases of the Chang–Jaffe conditions [24] for electrodes being blocking and conducting for various charge carriers. We mainly considered electrodes that were blocking for ions (or vacancies), i.e.

$$J_{d,a}^z(\rho, z = 0, t) = 0, \quad J_{a,d}^z(\rho, z = h, t) = 0,$$

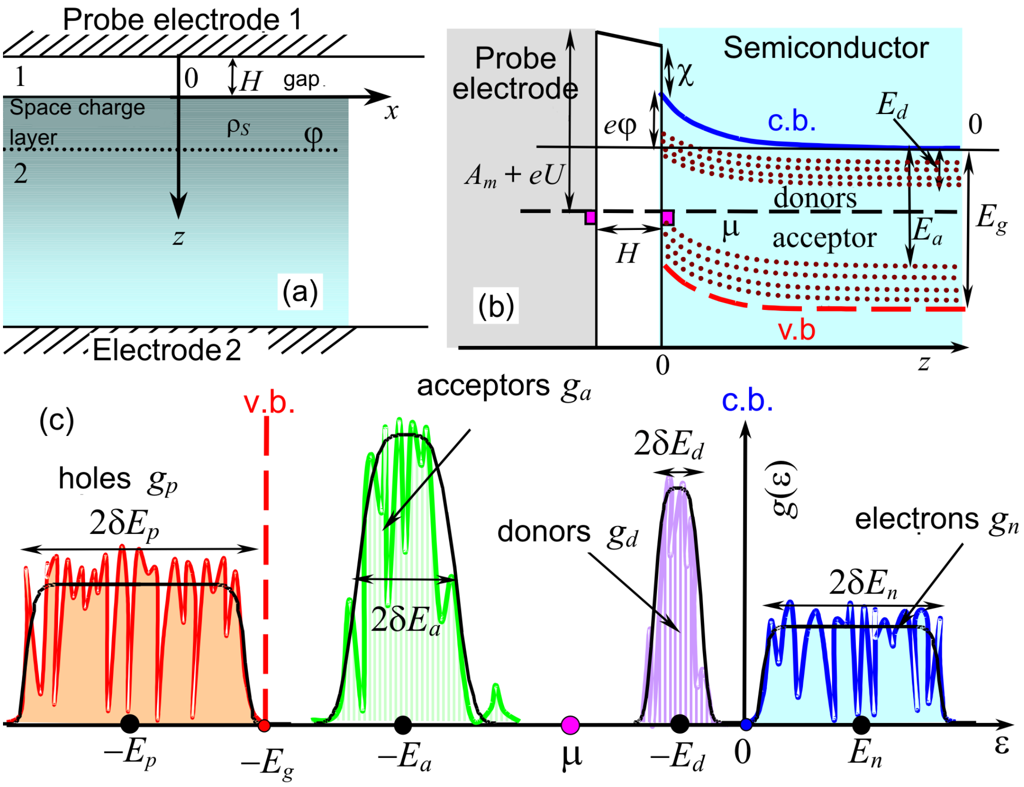

Fig. 2. (*a*) Diagram of the studied asymmetric heterostructure "probe electrode/gap/ionic semiconductor film". (*b*) Band structure near the surface $z = 0$: A_m is the work function difference, U is the electric voltage applied to the probe tip (upper electrode) at $z = -H$, φ is the electric potential, χ is the electron affinity in the semiconductor, and μ is the chemical potential level. (*c*) Localized state densities $g(\varepsilon)$ for holes, acceptors, donors, and electrons. Filled regions with uneven edges mark the typical state density calculated from the density functional theory. Exponential approximations (2) are shown by smooth lines

$$0 \le \rho \le \infty, \qquad (6)$$

though ohmic for electrons (or holes), i.e. the total charge was equal to zero: $\rho_S(0, h) = 0$.

The mechanical deformation of the film u_{ij} was calculated from the linear Lamé equation:

$$c_{ijkl}\frac{\partial^2 u_k}{\partial x_j \partial x_l} = -\frac{\partial}{\partial x_j}\times$$

$$\times\begin{pmatrix} -\beta_{ij}^{a}\left(N_a^-(\mathbf{r}) - N_{a0}^-\right) - \beta_{ij}^{d}\left(N_d^+(\mathbf{r}) - N_{d0}^+\right) + \\ +\Xi_{ij}^{C}\left(n(\mathbf{r}) - n_0\right) + \Xi_{ij}^{V}\left(p(\mathbf{r}) - p_0\right) \end{pmatrix}. \qquad (7)$$

where c_{ij} is the elastic-stiffness tensor and $\beta_{ij}^{d,a}$ are the Vegard expansion tensors [25–27] for ions (donors, acceptors) or vacancies. Electrons and holes make a contribution to the mechanical deformation of the film proportionally to the deformation potential $\Xi_{ij}^{C,V}$.

The mechanical boundary conditions depend on the elastic properties of the substrate and the upper electrode (probe or gap). In particular, normal strains at the probe/film contact are negligibly small, and the displacement at the contact with the substrate is absent:

$$\sigma_{3i}(x_1, x_2, z = 0) = 0, \quad u_i(x_1, x_2, z = h) = 0. \qquad (8)$$

In the approximation of separation of the electromechanical coupling, the deviations $\delta N_d^+(\mathbf{r}) = \left(N_d^+(\mathbf{r}) - N_{d0}^+\right)$, $\delta N_a^-(\mathbf{r}) = \left(N_a^-(\mathbf{r}) - N_{a0}^-\right)$, $\delta n(\mathbf{r}) = \left(n(\mathbf{r}) - n_0\right)$, and $\delta p(\mathbf{r}) = \left(p(\mathbf{r}) - p_0\right)$ of the concentrations of donors $N_d^+(\mathbf{r})$, acceptors $N_a^-(\mathbf{r})$, electrons, and holes from the equilibrium values N_{d0}^+, N_{a0}^-, n_0, and p_0 are determined from the solution of Eqs. (1)–(4) without regard for the elastic subsystem. In this way, the approximate solution of the Lamé equation (6) was obtained by the Green's function method [12]:

$$u_3(z = 0, \rho, \omega) \approx \frac{1}{c_{33}}\int_0^\infty dk J_0(k\rho)\, k\times$$

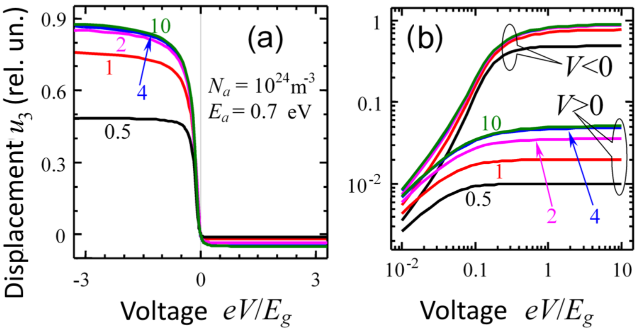

Fig. 3. Normalized static mechanical displacements of the surface u_3 calculated for various film thicknesses $h/R_S = 0.5$, 1, 2, 4, and 10 (see numbers near curves). The activation energy of acceptors $E_a = 0.7$ eV, the acceptor concentration $N_a = 10^{24}$ m^{-3}, the donor concentration $N_d = 10^{20}$ m^{-3}, the screening radius $R_S = 3.0$ nm, and $\mu = -0.80$ eV

$$\times\begin{pmatrix} \Xi_{33}^{C}\int\limits_{0}^{\infty}\exp\left(-k\,z\right)\delta n\left(k,z,\omega\right)dz+ \\ +\Xi_{33}^{V}\int\limits_{0}^{\infty}\exp\left(-k\,z\right)\delta p\left(k,z,\omega\right)dz \\ -\beta_{33}^{d}\int\limits_{0}^{\infty}\exp\left(-k\,z\right)\delta N_{d}^{+}\left(k,z,\omega\right)dz- \\ -\beta_{33}^{a}\int\limits_{0}^{\infty}\exp\left(-k\,z\right)\delta N_{a}^{-}\left(k,z,\omega\right)dz \end{pmatrix}, \qquad (9)$$

where $\delta N_d^+(k,z,\omega)$, $\delta n(k,z,\omega)$ are the Fourier spectra, $k^2 = k_x^2 + k_y^2$, and $J_0(x)$ is the zero-order Bessel function.

It follows from Eq. (9) that, in the case of the one-dimensional geometry ($k = 0$) and ion-blocking electrodes with the conservation of the ion charge, the mechanical displacement of the film surface due to the electric voltage applied to the probe is caused only by changes in the hole and electron concentrations. This displacement can be directly registered by SPM methods.

The numerical stationary solutions of the nonlinear equations (1)–(8) were obtained with the help of a special Matlab program.

4. Results and Their Discussion

The static local electromechanical response is depicted in Fig. 3. Graph (b) represents graph (a) on the log-log scale. The displacement graph is strongly asymmetric with respect to the change in the voltage sign ("rectification" of the electromechanical response by two orders of magnitude), which is an analog of the diode effect.

The dynamic local electromechanical response calculated in the linear approximation is shown in Fig. 4. In this approximation, loops have the elliptic form and are degenerated into a straight line in the static case of zero frequency. Hysteresis loops appear due to a delay of the carrier phase with respect to the change in the voltage one.

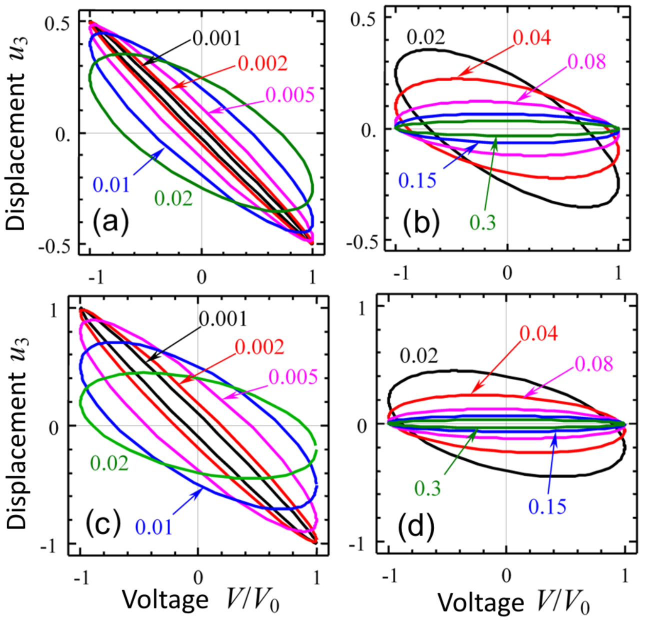

Fig. 4. Hysteresis loops of the linear local electromechanical response: the real part of the surface displacement u_3 as a function of the real part of the applied voltage at various frequencies $w\,\tau_M$ dimensionless for the Maxwellian charge relaxation time τ_M (numbers near curves) calculated for a gap with the width $\tilde{H}/R_S = 0$ (a, b) and 1 (c, d). The film thickness $h/R_S = 100$, the boundary conditions: $J_\omega^c(0) = 0$, $\rho_\omega(h) = 0$ (gap at the surface, ohmic contact with the upper electrode)

The typical nonlinear dynamic strain-voltage hysteresis loops in an ionic semiconductor thin film with mobile acceptors and holes are given in Figs. 5 and 6. Graphs (a, c, e) are plotted with the use of localized DOS for ions and electrons, while graphs (b, d, f) are constructed in the Boltzmann approximation.

Some types of hysteresis with pronounced memory windows (hysteresis loop width) and double loop are observed experimentally in correlated oxides and materials with resistive memory. The theoretically predicted strain-voltage hysteresis loops of the piezoelectric type that have the butterfly shape must be confirmed experimentally by SPM methods.

It is worth noting that independent measurements of the capacity-voltage characteristics (i.e. the dependence of the total electric charge on the applied voltage) and their comparison with the electromechanical (i.e. voltage-deformation) ones allow one to estimate the corresponding constant of the deformation poten-

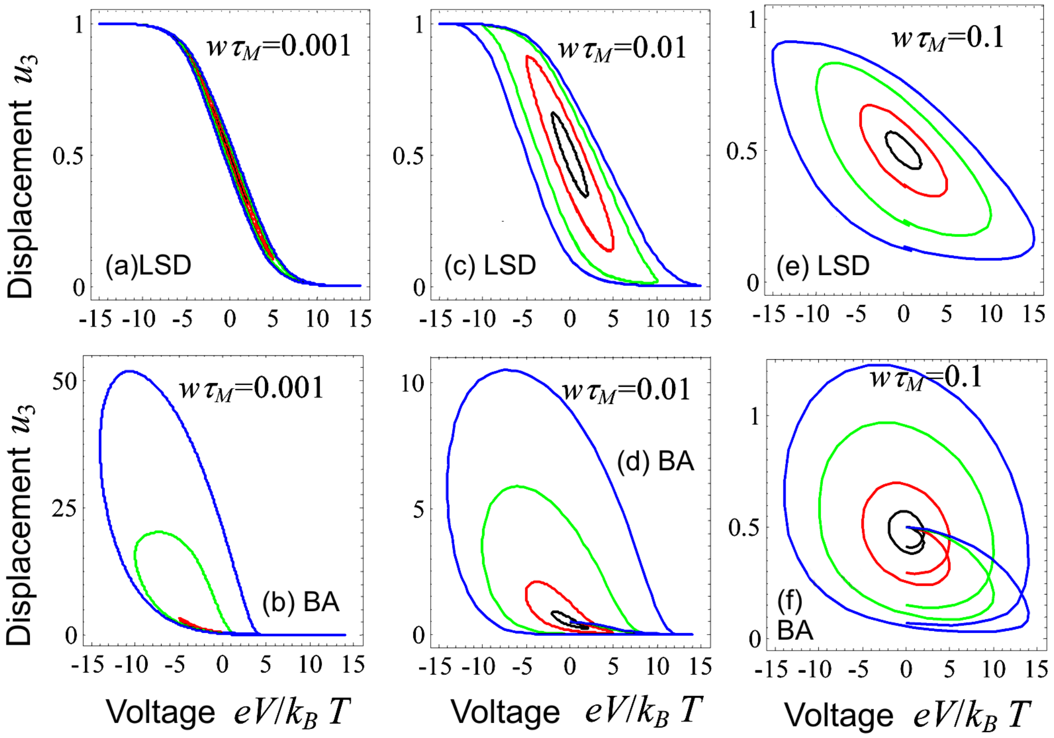

Fig. 5. Nonlinear local electromechanical response calculated for various frequencies: $w\tau_M = 0.001$ (*a, b*), $w\tau_M = 0.01$ (*c, d*), and $w\tau_M = 0.1$ (*e, f*). Different loops correspond to different values of the maximum voltage $V = 2$, 5, 10, and 15 (in units of $k_{\rm B}T/e$). Graphs (*a, c, e*) are plotted with the use of localized state densities (LSD), graphs (*b, d, f*) – in the Boltzmann approximation (BA). LSD parameters: $k = 4$, $\delta E_a/k_{\rm B}T = 2$, $\delta E_p/k_{\rm B}T = 20$, $N_a = N_p$; the film thickness $h/R_S = 5$, the acceptor and hole mobilities relate as $\eta_a/\eta_p = 0.1$. Asymmetric boundary conditions are imposed: the lower electrode is hole-conducting $\rho_S(h) = 0$, the upper one is blocking $J_c^p(0) = 0$ (ρ is the charge density, J is the current)

tial that is sometimes poorly known. For this purpose, one can use the following relation between the displacement of the film surface ($\delta u_3(z = 0,\omega)$), the components of the tensor Ξ^V_{ij}, and the hole concentration δp in a film with mobile acceptors: $\delta u_3(z = 0,\omega) = \left(\frac{s_{12}(\Xi^V_{11}+\Xi^V_{22})}{s_{11}+s_{12}} - \Xi^V_{33}\right)\int_0^h dz\delta p(z,\omega)$. The electromechanical response of a film with mobile ionized donors and electrons can be analyzed in a similar way.

5. Conclusions

The work is devoted to the calculation of the static and dynamic local electromechanical response of ionic semiconductor thin films. Using the well-localized state density and the Boltzmann approximation, we have analyzed how variations in the concentrations of ions, holes, and electrons result in the mechanical displacement of the film surface due to the electric voltage applied to the probe, which is directly registered by SPM methods. It is shown that, with increase in the maximum voltage amplitude, the Boltzmann approximation becomes invalid for the description of a local electromechanical response of films, and one should use the localized state density to calculate the Fermi quasilevels, electric currents, and electromechanical response.

The results were derived within the linear Debye screening theory (the corresponding hysteresis loops of the electromechanical response have the elliptic form) and beyond it (nonlinear approximation). We have obtained a large variety of nonlinear static and dynamic strain-voltage hysteresis loops in an ionic semiconductor thin film with mobile acceptors (donors) and holes (electrons).

If a probe electrode and a substrate are blocking for ions (acceptors or donors), a change in the hole (electron) concentration via the electron-phonon coupling results in a mechanical displacement of the film surface measured by SPM methods. The magnitude of the coupling is proportional to the deformation potential that can be intensified, in turn, by Jahn–Teller local deformations

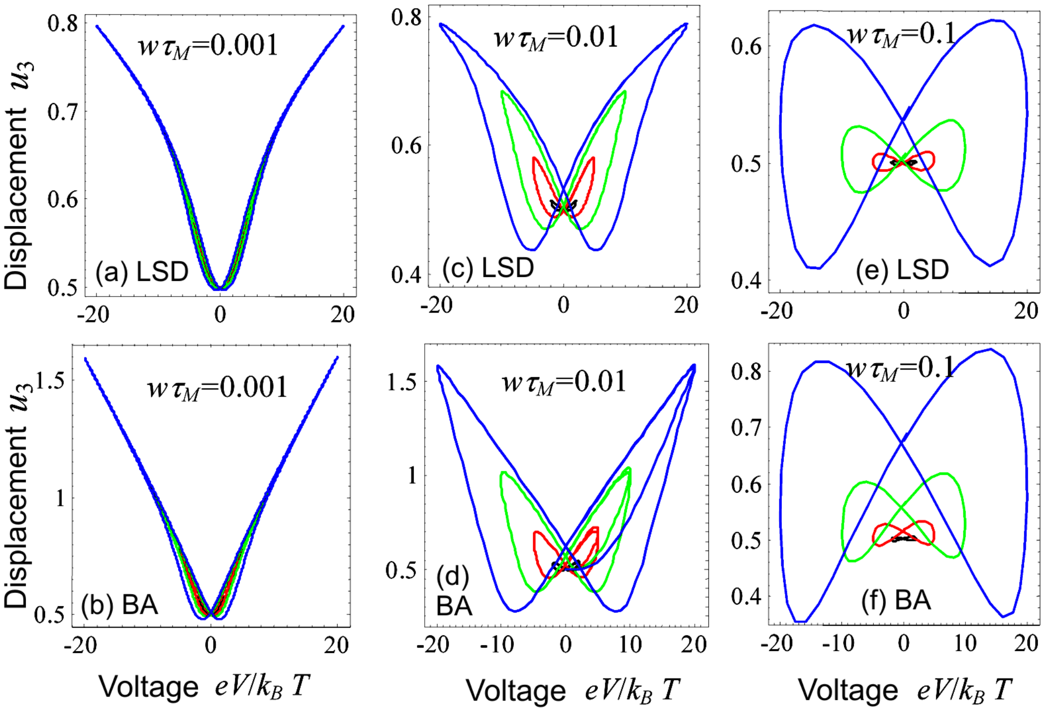

Fig. 6. Nonlinear local electromechanical response calculated for various frequencies: $w\tau_M = 0.01$ (*a, b*), $w\tau_M = 0.03$ (*c, d*), and $w\tau_M = 0.1$ (*e, f*). Different loops correspond to different values of the maximum voltage $V_0 = 2$, 5, 10, and 20 (in units of k_BT/e). Graphs (*a, c, e*) are plotted with the use of localized state densities (LSD), graphs (*b, d, f*) – in the Boltzmann approximation (BA). LSD parameters (2): $k = 4$, $\delta E_a/k_BT = 2$, $\delta E_p/k_BT = 20$, $N_a = N_p$; the film thickness $h/R_S = 5$, the mobility ratio $\eta_a/\eta_p = 0.1$. Symmetric boundary conditions for the charge densities are imposed: $\rho_S(0) = \rho_S(h) = 0$

existing in correlated oxides such as $La_{1-x}Sr_xMnO_3$ and $La_{1-x}Sr_xCoO_3$. This allows one to link the electromechanical response with the local deformation potential of correlated-oxide ionics.

Thus, it is proved that the studies of a displacement of the ionic-semiconductor surface can provide the important information about local changes of the charge state of acceptors (donors), as well as about electron-phonon correlations via the deformation potential.

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ЛОКАЛЬНИЙ ЕЛЕКТРОМЕХАНІЧНИЙ ВІДГУК ТОНКИХ ПЛІВОК НАПІВПРОВІДНИКІВ-ІОНІКІВ

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Резюме

Проведено розрахунки локального електромеханічного відгуку тонких плівок напівпровідників-іоніків, викликаного локальними змінами концентрації іонів (*стехіометричний внесок*) та вільних електронів і дірок (*електрон-фононна взаємодія внаслідок деформаційного потенціалу*). Отримано динамічні деформаційно-вольтові петлі гістерезису в тонкій плівці іоніка-напівпровідника з рухливими акцепторами (донорами) і дірками (електронами).

У випадку “блокуючих” електродів, які не пропускають іони, зміни концентрації дірок (електронів) вносять основний внесок у залежність механічного зміщення поверхні плівки від електричної напруги, прикладеної до зонда, що безпосередньо реєструється методами скануючої зондової мікроскопії (СЗМ). Таким чином, СЗМ переміщення поверхні іоніка-напівпровідника може надати важливу інформацію про локальні зміни зарядового стану акцепторів (донорів) та електрон-фононні кореляції через деформаційний потенціал.