Abstract—The observation of ferroelectric, ferromagnetic, and ferroelastic phases in thin films of binary oxides attracts the broad interest of scientists and engineers. However, the theoretical consideration of the physical nature of the observed behavior was performed mainly for HfO₂ thin films from the first principles, and in the framework of Landau–Ginzburg–Devonshire (LGD) phenomenological approach with special attention to the role of oxygen vacancies in both cases. Allowing for the generality of the LGD theory, we applied it to the group of binary oxides in this work. The calculations have been performed based on the assumption that oxygen vacancies, as elastic dipoles, can be partially transformed into electric dipoles due to the defect site-induced and/or surface-induced inversion symmetry breaking (via, e.g., piezoelectric effect), and can “migrate” throughout the depth of an ultrathin film. Since many films of binary oxide are ferroelectric and ferromagnetic due to the oxygen vacancies, they can be multiferroics. Performed calculations have shown that thin films of binary oxides can be considered as new multiferroics with physical properties useful for broad spectra of applications in nanoelectronics and nanotechnology. The properties can be controlled by the choice of oxygen vacancy concentration, film thickness, and special technological treatment, such as annealing.

Index Terms—Ferroelectricity, magnetic defect, multiferroicity, oxygen vacancy, thin film.

I. INTRODUCTION

A. Ferromagnetism in Thin Films of Binary Oxides

The investigation of the magnetization of oxide thin films, which are nonmagnetic in bulk, started in 2004 when Venkatesan et al. [1] reported unexpected room-temperature magnetism in HfO₂ thin films on sapphire or silicon substrates. Then, the observation of room-temperature magnetism in oxide thin films, such as HfO₂, TiO₂, SnO₂, and In₂O₃, on various substrates has attracted much attention of the scientific community due to the so-called $d^0$-magnetism [2]–[8].

The magnetic moment was measured with the help of a SQUID magnetometer for the samples oriented perpendicular or parallel to applied magnetic field, where the parallel magnetic moment appeared to be 19% smaller. Extrapolated Curie temperature was much higher than 400 K, and the magnitude of magnetic moment and hysteresis loop characteristics were shown to be significantly dependent on the type of substrate. These investigations, performed in [3] and [4], allowed to pour light on the nature of magnetic defects in thin films of oxides nonmagnetic in bulk, i.e., on the mechanisms of $d^0$ magnetism. Allowing for that all the HfO₂, TiO₂, and In₂O₃ films were colorless, shiny, and highly transparent, the concentration of magnetic impurities can be well below $10^{-2}$ wt.%, and so the high value of magnetization is hard to attribute to any kind of existing impurities. Because of this, it was supposed that the oxygen vacancies at the interface between the film and substrate are the main sources of the magnetization (see [4], [9], where it was shown that oxygen vacancies are magnetic defects in the vicinity of surface). The disappearance of magnetization after annealing in oxygen atmosphere [7] confirmed this supposition.

Another point that is important to emphasize is the strong thickness dependence of the magnetic moment. In fact, 10-nm-thick TiO₂ and HfO₂ films’ saturation magnetization was, respectively, 20 and 15 times larger than that of 200-nm-thick films (see [3] and Refs. therein), i.e., it is approximately inversely proportional to the film thickness. Therefore, the number of magnetic defects (oxygen vacancies) in the film is independent of its thickness. One can directly interpret this as the evidence that the observed magnetization originates from defects localized mostly near the interface between the film and its substrate. Allowing for that the physical reasons of the majority of abovementioned features stayed unexplained up to now, Glinchuk et al. [10] found out that the localization of the oxygen vacancies mostly at the interfaces are the reasons of binary oxide films magnetization.

B. Ferroelectricity in Thin Film of Binary Oxides

Recent observations of ferroelectricity in thin films of TiO₂, HfO₂, and related solid solutions have attracted the attention of scientific community due to fundamental perspective as well as the potential for applications [11]–[14]. From the
application perspective, ferroelectricity in binary oxide thin films has a crucial relevance for performance of ferroelectric memories due to the ease of synthesis and compatibility with Si processing [15]–[19], highly scalable and manufacturable ferroelectric materials of choice for advanced nonvolatile and random access memories [20].

Hoffmann et al. [21] revealed that doping with Gd, which creates oxygen vacancies because of charge compensation, induces the ferroelectric phase in thin films of HfO2, as well as demonstrated the essential role of TaN electrodes due to interface oxidation of the electrodes. Pešić et al. [22] found out that no new defects are generated during the wake-up technological process of the device, but the existing defects including oxygen vacancies redistribute within the device. Different phases, including the ferroelectric phase [23], were revealed in thin films of HfO2 doped with different amounts of Si, but corresponding phonon modes in HfO2 [24] appeared slightly dependent on temperature in the wide temperature range 20–800 K.

Allowing for all the abovementioned facts, vacancy diffusion has been identified as the main cause for the phase transformation and consequent increase of the remanent polarization in the binary oxide films [25]. It is significant that the results of [25] for polarization and dielectric susceptibility hysteresis loops’ shape, remanent polarization, coercive field, as well as the changes of these characteristics taking place along with the increase in film thickness are in agreement with experimental loops measured by Polakowski and Müller [20].

Since the binary oxide film is both ferroelectric and ferromagnetic due to the oxygen vacancies, it can be a multiferroic, and this work explores the issue.

II. THERMODYNAMICS OF FERROELECTRICITY APPEARANCE

Thermodynamics permits us to use an LGD-type free-energy functional for the quantitative consideration of ferroelectricity induced by oxygen vacancies in binary oxides. Gibbs potential density of oxide film with paraelectric nonlinearity [26] has the form as (1a) and (1b), shown at the bottom of this page [27].

In (1b), $P_i$ denotes the components of polarization vector ($i = 1, 2, 3$) and $a_{ij}$ is the elastic stress tensor. The summation is performed over all repeated indices. Equations of state $\partial G/\partial a_{ij} = -u_{ij}$ determine the strains $u_{ij}$. Euler–Lagrangian equations $\partial G/\partial P_i = 0$ determine the polarization components. The surface energy $G_S$ is defined by the oxide-substrate and oxide-electrodes interface chemistry.

The expansion coefficients $a_{ij}$ are positive and typically diagonal, $a_{ij} = \delta_{ij} a_{ii}$ because the ferroelectricity is absent in the absence of vacancies. The coefficient $a_{ii} \equiv 1$ is related to a bulk relative dielectric permittivity tensor $\varepsilon_{ij}$ as $a_{ij} = 1/[\varepsilon_{0}(\varepsilon_{ii} - 1)]$, where $i = 1, 2, 3$; $\varepsilon_{0}$ is a universal dielectric constant and $\varepsilon_{ii} \approx 25$ for HfO2 or $\varepsilon_{ii} \approx 80$ for TiO2 [28]. The nonlinearity $\beta = a_{33}$ can be estimated for HfO2 as $5 \times 10^{10}$ J m$^{-3}$ C$^{-4}$.

The matrix of the gradient coefficients $g_{ijkl}$ is positively defined. $Q_{ijkl}$ is the electrostriction tensor, $s_{ijkl}$ is the elastic compliances tensor, and $F_{ijkl}$ is the fourth-rank tensor of flexoelectric coupling ($F_{ijkl} \sim 10^{-11}$ m$^3$/C). $E_i(r)$ denotes the electric field.

The last term in (1b) includes a Vegard-type concentration-deformation energy, $U_{ij}^{V} = \left[ \delta N_d(\mathbf{r}) \right] \sigma_{ij}$, determined by the random mobile vacancies (charged or electroneutral) with concentration $\delta N_d(\mathbf{r}) \sim \sum_k (\delta(\mathbf{r} - \mathbf{r}_k) - N_{3D})$. The vacancy concentration in the bulk is small enough in thermodynamic equilibrium, namely, $N_{3D} \ll 10^{28}$ m$^{-3}$. The vacancies, or their complexes, tend to accumulate in the vicinity of any inhomogeneities, surfaces, and interfaces [see Fig. 1(a)], since the energy of their formation in such places, as a rule, is much smaller than in the homogeneous bulk [29]–[32]. Keeping in mind that the measurements of ferroelectricity in the majority of binary oxides were performed in symmetrical stacks, e.g., TiO2/HfO2/TiN, in the following, we will consider the symmetric geometry shown in Fig. 1(a) as the most suitable for the comparison of the theory with experiments (see [25]). The symmetric geometry simplifies the theoretical calculations, since in the case the homogeneous stress originated from the lattice mismatch between the thin film, top and bottom electrodes compensates one another throughout the film depth. The vacancies can create sufficiently strong fields in the places of their accumulation, which in turn lead to appearance of new phases in oxides, including polar (ferroelectric) ones. On contrary, the nonpolar state remains in the places where
there are few vacancies. Therefore, the coexistence of polar ferroelectric and nonpolar states can be realized in the films.

The average distance between defect centers $2R$ should be associated with the average size per inclusion. The defect size characteristic $r_0$ is much smaller than $R$, e.g., $r_0 \ll R$, where $r_0$ is the ionic radius $\sim (0.1–1) \ \text{Å}^3$ [see Fig. 1(b)].

A dilatation center with the equal distortion can be considered as a simple elastic model of impurity atom or vacancy, whose own distortion (Vegard strain) is [33], [34]

$$w_{x,y}(r) = w_{y,z}(r) = w_{z,x}(r) = W \delta(r - r_k).$$

Equation (2) is derived under the assumption of isotropic and diagonal Vegard expansion tensor $W_{ij}$, $W_{ij} = W \delta_{ij}$, where $W = \pm (5 - 20) \ \text{Å}^3$ [35] is the volume change in the point of defect localization $r = r_k$. In general case, the structure of Vegard expansion tensor $W_{ij}$ (elastic dipole) is controlled by the symmetry (crystalline or Curie group of symmetry) of the material [35]–[37].

Nonzero components of the elastic displacement, strain and stresses induced by a spherically symmetric elastic point defect (e.g., dilatation center) located in the coordinate origin, $r = 0$, have the form [33], [34]

$$u^W(r) = -\frac{(1 + v)W}{4\pi(1 - v) r^3} \equiv \frac{(1 + v)W}{4\pi(1 - v) r^2} \left(\frac{1}{r} \right),$$

$$u^W_{ii}(r) = -\frac{(1 + v)W}{4\pi(1 - v) \partial x_i r^3} = \frac{(1 + v)W}{4\pi(1 - v) \partial x_i r^2} \left(\frac{i}{r} \right),$$

$$u^W_{ij}(r) = \frac{(1 + v)W}{4\pi(1 - v) \partial x_i \partial x_j r^3} = \frac{(1 + v)W}{4\pi(1 - v) \partial x_i \partial x_j r^2} \left(\frac{i}{r} \right),$$

$$\sigma^W_{ii}(r) = -\frac{G(1 + v)W}{2\pi(1 - v) r^5} \equiv \frac{G(1 + v)W}{2\pi(1 - v) r^4} \left(\frac{1}{r^2} \right),$$

$$\sigma^W_{ij}(r) = -\frac{G(1 + v)W}{2\pi(1 - v) \partial x_i \partial x_j r^5} = -\frac{G(1 + v)W}{2\pi(1 - v) \partial x_i \partial x_j r^4} \left(\frac{i}{r^2} \right).$$

where the radius $r = (x_i^2 + x_j^2 + x_k^2)^{1/2}$ is introduced in (3). Poisson’s ratio is $v = -s_{12}/s_{11}$ for cubic m3m symmetry, $G$ is the shear modulus, and $s_{ij}$ are elastic compliances.

The substitution of elastic fields (3) into the potential (1b) leads to the renormalization of the coefficients $a_{ij} \rightarrow a^R_{ij}$ by the electrostriction coupling with Vegard expansion. For the diagonal component, we obtain

$$\alpha^R_{kk}(r) \approx \alpha^R_{kk} \equiv \frac{1}{\varepsilon_0(\varepsilon_{kk} - 1)} - 2Q_{ijkl} \sum_l \sigma^W_{ij} \left[\partial N_d(r - r_l) \right].$$

The summation is performed over all the sites of vacancies. One can see from (4) that the local polar state, occurring under the condition $\alpha^R_{kk} < 0$, is not excluded in the spatial regions, where the defect and free carrier concentration is high enough.

A. Soft Mode Origin of Ferroelectricity

Using ergodic hypothesis, the averaging and the summation in (4) reduce to the averaging over the film volume. The averaging of the function $2Q_{i3j3}\sigma^W_{ij} \left[\partial N_d(r) \right]$ in (4) reduces to the averaging over the cylindrical volume, $4\pi R^2h$, and for m3m symmetry, we obtain

$$\langle \alpha^R_{ii}(r) \rangle = \frac{1}{\varepsilon_0(\varepsilon_{ii} - 1)} + 2(\varepsilon_{ii} - Q_{ij}) \int_0^h \! dz \int_0^R \! \rho \, dp \times \frac{G(1 + v)W}{2\pi(1 - v) r^3} f(r) \left(\frac{r}{R} \right).$$

Here, $i = 1, 2; 3; \rho = (x_i^2 + x_j^2)^{1/2}$ is the polar radius; $\varepsilon = x_3$; and we assumed $Q_{11} = Q_{22} = Q_{33}$ and $Q_{12} = Q_{13} = Q_{23}$ for the m3m parent symmetry. The film thickness is $h$. The average 2-D concentration of defects is $N_{2D} = 1/4\pi R^2$. The distribution of vacancies depends on the distance from the film surface, and reveals exponential or linear decay, namely, [38]

$$f(r) = \frac{1}{\pi R^2 h} \exp \left(\frac{-r}{\varepsilon_0} \right), \quad 0 < \varepsilon < \varepsilon_0.$$
exist in the sense of mean field theory. A qualitatively similar estimate is valid for TiO$_2$, but here the difference is much smaller. In the following, we will show that the contribution of flexoelectric effect and Vegard stress will lead to the ferroelectricity appearance.

B. Flexochemical Origin of Ferroelectricity

Next, let us estimate the polarization and electric field variations induced by the joint action of Vegard stresses and flexoelectric coupling. Equations of state $\partial G/\partial \sigma_{ij} = -u_{ij}$ give the strains $u_{ij}$ as

$$u_{ij} = s_{ijkl} \sigma_{kl} + \sum_I u_{ijI} [\delta N_0(r - r_i)] - F_{ijkl} \frac{\partial P_l}{\partial \epsilon_{kl}} + Q_{ijkl} P_k P_l. \quad (8)$$

In linear approximation polarization, the variation $\delta P_l$ is induced by defects (2) located at $r = r_m$. The defects induce elastic fields (3) due to the joint action of Vegard stresses and flexoelectric coupling. In particular, the variation $\delta P_l$ can be estimated as $\delta P_l(r) = f_{ijkl}(\delta \epsilon_{jk}^W[\delta N_0(r)]/\partial x_l)$. For $z$-component, this gives a nonzero average

$$\delta P_3(r) \approx \frac{(1 + v)W}{4\pi(1 - v)} \sum_m \left( \tilde{f}_{33} \frac{\partial^3}{\partial z^3} + \tilde{f}_{31} \left( \frac{\partial^3}{\partial x_3 \partial \epsilon_{13}^W} + \frac{\partial^3}{\partial x_3 \partial \epsilon_{23}^W} \right) \right) \left| r - r_m \right|^{-1} \quad (9a)$$

$$\langle \delta P_3(r) \rangle = \frac{(1 + v)W}{2(1 - v)} \left( \tilde{f}_{33} - \tilde{f}_{31} \right) \int_0^h dz f(z) \left( \frac{\partial^3}{\partial z^3} \sqrt{R^2 + z^2} \right) \approx \frac{1 + v}{1 - v} \frac{W}{2\pi R^2 h} \left( \frac{1}{\sqrt{R^2 + z_0^2}} - \frac{1}{R} \right) \quad (9b)$$

where $\tilde{f}_{ijkl}$ is the flexoelectric effect stress tensor related with the tensor $f_{ijkl} \sim (5-10)$ V by expression $\tilde{f}_{ijkl} = (a_{ijk}^W(r)) f_{ijkl} = \varepsilon_0(\varepsilon_{33} - 1) f_{ijkl}$. The value $\tilde{f}_{ijkl} \sim 10^{-8}$C/m$^2$ can be estimated from the Kogan model [40]. Estimates based on (9) gives relatively high polarization $\sim(2-5) \mu$C/cm$^2$ typical for improper ferroelectricity, and the hysteresis loops measured by Nishimura et al. in thin undoped 20-nm HfO$_2$ films correspond to the same order of remanent polarization ($\sim 2.5$ $\mu$C/cm$^2$).

Dependences of static polarization $P_S(T, h, N_S)$ on the applied voltage, calculated at room temperature, fixed $N_S$, and different film thickness $h = (5-25)$ nm, are shown in Fig. 2. The remanent polarization changes from $10 \mu$C/cm$^2$ for $h = 5$ nm to $1 \mu$C/cm$^2$ for $h = 25$ nm, and the loop becomes rather slim and tilted, with an increase in $h$ from 5 to 25 nm (compare loops 1–5 in Fig. 2).

Further increase of the film thickness $h$ does not lead to any significant changes in the loop shape and sizes because for the value of $h$ higher than 25 nm, the loop in itself exhibits dynamic effect.

C. Origin of Multiferroicity

Since the binary oxide films are ferroelectric and ferromagnetic due to similar oxygen vacancies, they can be considered as a new group of multiferroics. Moreover, the first principle calculations and their comparison with experiment had shown [21] that the oxygen vacancies as elastic dipoles can lead to the appearance of structural ferroelastic phases. On the other hand, it is important to mention that the first principal calculations of the magnetization induced by oxygen vacancies are shown in [41] and [42].

Generally speaking, thin films of binary oxides with oxygen vacancies can be considered as the materials with a mixture of electric and elastic dipoles. By definition, oxygen vacancies are known to be elastic dipoles, which transform into electric dipoles in the vicinity of surfaces with surface-induced piezoelectric effect. Indeed, the contribution of elastic dipoles into total energy $E_{tot}$ can be written as $E_{tot} = w_{ij} \eta_{ij}$, where $w_{ij}$ and $u_{ij}$ are elastic dipole and mechanical tension, respectively. This tension due to the piezoelectric effect leads to the appearance of electric field $E_k = u_{ij} \eta_{jk}$, where $\eta_{ijk}$ is the piezoelectric coefficient of the tensor. The contribution of an electric field into energy can be written as its product with electric dipole $d_k$. Substituting $u_{ij} = E_k \eta_{ijk}$ into the abovementioned expression for energy $E_{tot}$, $E_{tot} = w_{ij} E_k \eta_{ijk} = d_k E_k$ is obtained, so that the electric dipole moment of oxygen vacancy $d_k = w_{ij} \eta_{ijk}$ and its elastic dipole moment $w_{ij} = d_k \eta_{ijk}$. It is obvious that the phase diagram of the oxides thin films has to depend on the general concentration of oxygen vacancies to overcome the percolation threshold for the appearance of long-range order. The first principle calculations of the HfO$_2$ thin film phase diagram as a function of oxygen vacancies concentration had shown the appearance of the orthorhombic FE phase, and cubic, tetragonal, monoclinic paraelectric (ferroelastic) phases at room temperature.

Therefore, binary oxide thin films indeed can be considered as a new wide class of multiferroics with the coexistence of ferroelectric, ferroelastic, and magnetic phases, and thus with physical properties useful for broad spectra of applications in modern nanoelectronics and nanotechnology. The choice of necessary properties can be controlled by the concentration of oxygen vacancies, film thickness, and technological treatment.
III. DISCUSSION AND CONCLUSION

The calculations have been performed in the assumption that oxygen vacancies, as elastic dipoles, can be partially transformed into electric dipoles due to the defect site-induced and/or surface-induced inversion symmetry breaking (via, e.g., piezoelectric effect) and can “migrate” throughout the entire depth of an ultrathin film.

The proposed model opens the way for the calculation of multiferroic properties and complex phase diagrams of binary oxide films. To perform the calculations of the phase diagrams, one has to consider polarization and elastic fields induced by oxygen vacancies, being electric and elastic dipoles, respectively, as the long-range-order parameters of ferroelectric and ferroelastic phase transitions. Keeping in mind the existence of magnetization in many binary oxide thin films, it can be considered as the third long-range-order parameter. Therefore, rotomagnetic, magnetoelectric, and magnetoelastic interactions will represent the coupling between magnetization and two other order parameters [43], and the coupling between them will be represented by the Vegard-type interaction.

The developed theory opens the way for the selection of binary oxide films with optimal properties (namely, thickness, vacancies concentration, and annealing time), which can be used in high-performance electronic devices based on highly scalable, Si-compatible, and manufacturable ferroelectric thin films for advanced memories and capacitor industry.

Allowing for generality of the proposed approach, it can be applied to many binary oxides, i.e., both undoped and doped ones. Furthermore, it is applicable to other cases of dipolar impurities, e.g., originated from the trapped charges at the interfaces, etc.

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Anna N. Morozovska photograph and biography not available at the time of publication.

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