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Ground-state structure of $KNbO_3/KTaO_3$ superlattices: Array of nearly independent ferroelectrically ordered planes

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Phonon spectra of the paraelectric phase, structure and distribution of polarization in the polar ground state, and energies of ferroelectrically and antiferroelectrically ordered phases were calculated for stress-free $(KNbO_3)_1(KTaO_3)_n$ superlattices ($n = 1$–7) from first principles within the density-functional theory. Quasi-two-dimensional ferroelectric state with the polarization localized in the $KNbO_3$ layer was revealed to be the ground state for the superlattices with $n \geq 2$. The interaction between these polarized layers decreases exponentially with increasing the interlayer distance, and the ground-state structure transforms to an array of nearly independent ferroelectrically ordered planes at large n.

1 Introduction Ferroelectric superlattices (SLs) have drawn much attention in recent years. It appeared that many physical properties of these artificial structures, such as the Curie temperature, spontaneous polarization, dielectric constant, nonlinear dielectric and optical susceptibilities, exceed considerably the properties of bulk crystals and thin films of the corresponding solid solutions.

Ferroelectric phenomena in $KTa_{1-x}Nb_xO_3$ solid solutions and $KNbO_3/KTaO_3$ superlattices have been studied for many years. The experiment shows that in solid solutions a polar phase appears at $x > x_c = 0.008$ [1], but its properties in the vicinity of critical concentration x_c are modified by manifestation of quantum effects and by local disorder (the dipolar glass). To explain the peculiar properties of the solid solutions, a number of models has been proposed [2, 3] including a hypothesis about off-centering of Nb atoms in $KTaO_3$ [4–8]. However, as was shown in Ref. [9], the single-well shape of the local potential for Nb atom in the solid solution excludes its off-centering.

Ferroelectric properties of $KNbO_3/KTaO_3$ superlattices have been studied both experimentally [10–15] and theoretically [16–21]. The ferroelectric properties of the SLs grown on $KTaO_3$ substrates were calculated assuming that the polar ground-state structure is tetragonal [19]. The signs of antiferroelectricity in these SLs observed experimentally [14] were confirmed by observation of unstable phonon at X point of the Brillouin zone in first-principles calculations [19]. At the same time, our recent calculations of the ground state for both stress-free and substrate-supported superlattices with $n = 1$ and 3 [20] revealed that the ground-state structure of $KNbO_3/KTaO_3$ superlattices is monoclinic or orthorhombic, and is not tetragonal. Along with the ferroelectric instability, three unstable phonon modes that are not typical neither for $KNbO_3$ nor for $KTaO_3$ were observed at X, Z, and R points of the Brillouin zone in the paraelectric phase of these SLs. In contrast to other instabilities found in ten SLs studied in Ref. [20], the origin of these instabilities was not clear. The calculations [21] of phonon frequencies at the Γ point for the paraelectric phase of $(KNbO_3)_1(KTaO_3)_7$ superlattice with the lattice parameter equal to that of $KTaO_3$ revealed a weak ferroelectric instability of this phase with the polarization parallel to the layer plane. However, the distribution of polarization in the low-symmetry polar phase was not analyzed.

The purpose of this paper was to study the development of the ferroelectric properties in stress-free $(KNbO_3)_1(KTaO_3)_n$ superlattices when increasing their period $L = n + 1$. We show that quasi-two-dimensional (2D) ferroelectric state

with polarization localized in the $KNbO_3$ layer is the ground-state structure for the superlattices with $n \geq 2$ and it transforms to an array of nearly independent ferroelectrically ordered planes at large n.

2 Calculation details The $(KNbO_3)_1(KTaO_3)_n$ superlattices considered in this work are periodic structures grown in [001] direction and consisted of the $KNbO_3$ layer with a thickness of one unit cell and the $KTaO_3$ layer with a thickness of n unit cells ($1 \leq n \leq 7$). The modeling of these structures was performed using supercells with a size of $1 \times 1 \times (n+1)$ unit cells.

The calculations were performed within the first-principles density-functional theory (DFT) with pseudopotentials and a plane-wave basis set as implemented in ABINIT software [22]. The local density approximation (LDA) [23] for the exchange-correlation functional was used. Optimized separable nonlocal pseudopotentials [24] were constructed using the OPIUM software [25]; to improve the transferability of pseudopotentials, the local potential correction [26] was added. Parameters used for construction of pseudopotentials and other details of calculations are given in Ref. [20]. The plane-wave cut-off energy was 40 Ha (1088 eV). The integration over the Brillouin zone was performed on a $8 \times 8 \times 4$ Monkhorst–Pack mesh for SLs with $n = 1$ and 2, and on a $8 \times 8 \times 2$ one for SLs with $n = 3$–7. The relaxation of atomic positions and of the unit cell parameters was stopped when the Hellmann–Feynman forces were below 5×10^{-6} Ha/Bohr (0.25 meV/Å). The phonon spectra were calculated using the density-functional perturbation theory. The spontaneous polarization was calculated using the Berry phase method [27].

The structures of $KNbO_3$ and $KTaO_3$ calculated using this approach agree well with experiment. The calculated lattice parameters for cubic $KNbO_3$ and $KTaO_3$ are 3.983 and 3.937 Å, they differ from the experimental values (4.016 and 3.980 Å) by 0.81% and 1.07%, respectively (small underestimation of the lattice parameters is typical for LDA approximation). The calculated c/a ratio of 1.0197 for tetragonal $KNbO_3$ is close to the experimental value (1.0165). The ground-state structure is rhombohedral ($R3m$) for $KNbO_3$ and cubic ($Pm3m$) for $KTaO_3$, in agreement with experiment. The calculated spontaneous polarization of 0.372 C/m^2 for tetragonal $KNbO_3$ is close to experimental values of 0.37–0.39 C/m^2 [28].

3 Results Calculated frequencies of two lowest A_{2u} and E_u ferroelectric modes at the Γ point of the Brillouin zone in the paraelectric $P4/mmm$ phase of stress-free $(KNbO_3)_1(KTaO_3)_n$ superlattices with $n = 1$–7 are presented in Fig. 1. In SL with $n = 1$ both the modes, which result from unstable TO phonon of the perovskite structure, are unstable. With increasing n, the A_{2u} mode becomes stable, and there remains one unstable E_u phonon mode, whose frequency is weakly dependent on the $KTaO_3$ layer thickness.

When searching for an equilibrium structure, all possible structure distortions were tested. For SLs with $n = 1$, 2, and 3,

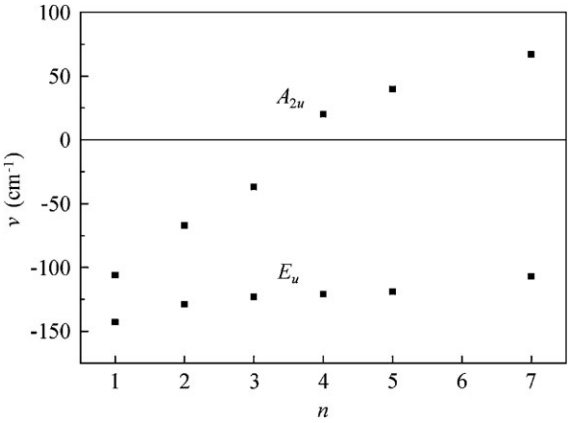

Figure 1 Frequencies of lowest A_{2u} and E_u polar optical modes in the paraelectric $P4/mmm$ phase as a function of the thickness of the $KTaO_3$ layer in $(KNbO_3)_1(KTaO_3)_n$ superlattices. Imaginary frequencies of unstable modes are plotted as negative values.

in which two unstable modes appeared in the paraelectric phase, monoclinic phases with Cm and Pm space groups (polarized along $[xxz]$ and $[x0z]$ directions) as well as tetragonal $P4mm$ and two orthorhombic $Amm2$ and $Pmm2$ phases were considered. For SLs with $n > 3$ only the E_u mode is unstable, and so only orthorhombic $Amm2$ and $Pmm2$ phases polarized along $[xx0]$ and $[x00]$ directions were possible. The calculations revealed that for SL with $n = 1$ the ground-state structure is Cm [20] whereas for other SLs—in spite of the existence of unstable A_{2u} mode in the paraelectric phase of SLs with $n = 2$ and 3—the ground-state structure is $Amm2$.

Calculation of phonon frequencies at high-symmetry points of the Brillouin zone for paraelectric $P4/mmm$ phase of SLs with $n \leq 3$ reveals, in addition to the ferroelectric instability, unstable phonons at Z, X, and R points (Table 1). However, among all possible distorted structures the polar Cm and $Amm2$ phases had the lowest total energy, and all phonons at all points of the Brillouin zone were stable in this phases.

As follows from Table 2, the energy gain resulting from the ferroelectric ordering do not vanish with increasing n. This fact and the existence of unstable E_u phonon in the paraelectric phase give evidence for stability of the ferroelectric state in $KNbO_3$ layers of minimal thickness (one unit cell) located between layers of $KTaO_3$ of arbitrary thickness.

The polarization profiles for the superlattices were calculated using the approximate formula $P_{s\alpha} = (e/\Omega)\sum_i w_i Z^*_{i,\alpha\beta} u_{i\beta}$, where $P_{s\alpha}$ is the α-component of polarization ($\alpha = x, y, z$), Ω is the unit cell volume of the layer under consideration, $u_{i\beta}$ is the β-component of displacement of the ith atom from its position in the paraelectric phase, $Z^*_{i,\alpha\beta}$ is the Born effective charge tensor of this atom,[1] and w_i is the weight equal to 1 for Nb(Ta) and O

[1] Born effective charge tensors obtained for $P4/mmm$ phase of the superlattice with $n = 1$ were used in calculations; the effective charges for SLs with $n = 2$–7 were very close to those used.

Table 1 Frequencies of the unstable phonon modes at high-symmetry points of the Brillouin zone in the *P*4/*mmm* phase of $(KNbO_3)_1(KTaO_3)_n$ superlattices with $n = 1$, 2, and 3.

n	ν (cm^{-1})			
	Γ (0,0,0)	Z (0,0,1/2)	X (1/2,0,0)	R (1/2,0,1/2)
1	143*i*	126*i*	71*i*	61*i*
2	118*i*	114*i*	50*i*	50*i*
3	114*i*	113*i*	46*i*	45*i*

atoms lying in the Nb(Ta)–O layer under consideration, 1/2 for K and O atoms lying in two nearest K–O planes, and 0 for other atoms. The dependence of $|P_s|$ on the layer number in SLs with $n = 3$, 5, and 7 are given in Fig. 2. It is seen that in the region between $KNbO_3$ layers the polarization decays exponentially, with a characteristic length of ~3 Å. This result indicates a strong localization of polarization in the potassium niobate layer and proves the *quasi-two-dimensional character of ferroelectricity* in considered superlattices. The polarization in the $KNbO_3$ layer decreases monotonically from 0.279 to 0.257 C/m^2 with increasing n from 3 to 7 (for comparison, for bulk orthorhombic $KNbO_3$ our calculations give $|P_s| = 0.418\ C/m^2$). The total polarization of the supercells calculated using the above formula differs from the result of correct Berry phase calculations (Table 2) by only 3%.

In order to determine the interaction energy W_{int} between two polarized $KNbO_3$ layers separated by $KTaO_3$ layer, the total energies of $[(KNbO_3)_1(KTaO_3)_n]_2$ superlattices of doubled period with ferroelectric and antiferroelectric ordering of polarization in neighboring $KNbO_3$ layers were calculated. The energy difference between the two ordered structures (per Nb atom) is given in Table 3. As follows from these data, the energy difference decreases exponentially with increasing thickness of $KTaO_3$ layer with a characteristic decay length of 2.9 Å.

The polarization in two-dimensional layer is stable against its spontaneous reversal if $2W_{int} < U$, where W_{int} is the energy of interlayer interaction and U is the height of the potential barrier separating different orientational states of polarization in the layer. The multiplier two in the above formula relates to the worst case in which the considered layer is polarized antiparallel to the polarizations in neighboring layers. In our structures, the easiest way to reorient the polarization is to rotate it in the layer plane, and so the U value is equal to the energy difference between [110]- and [100]-polarized structures. According to our calculations, $U = 1.84$ meV for SL with $n = 2$ and 1.69 meV for SL with $n = 3$. Comparison of U and $2W_{int}$ values (Table 3) shows that the stability criterion is satisfied at $n \geq 2$. From the data presented in Tables 2 and 3 one can see that in superlattices with $n \geq 3$ the interaction energy between neighboring polarized layers is less that 10% of the ordering energy. This gives the reason to consider the ground state in these superlattices as *an array of nearly independent ferroelectrically ordered planes*, with quasi-two-dimensional ferroelectricity realized in each layer.

The results obtained for $(KNbO_3)_1(KTaO_3)_n$ superlattices grown on $KTaO_3$ substrates are very different from those obtained for stress-free superlattices. As follows from Table 4, for $KTaO_3$-supported superlattices the ground-state structure is monoclinic for $n \leq 4$ and orthorhombic for $n = 5$–7 (data for $n = 6$ and 7 are not shown). This result can

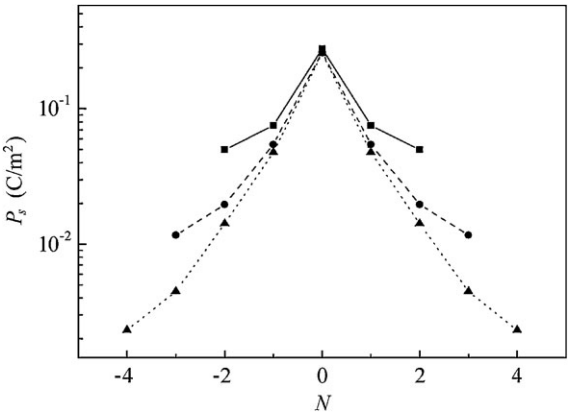

Figure 2 The polarization profiles for $(KNbO_3)_1(KTaO_3)_n$ superlattices with $n = 3$, 5, and 7. The number $N = 0$ corresponds to the $KNbO_3$ layer.

Table 3 The energy difference between ferroelectrically and antiferroelectrically ordered structures in $[(KNbO_3)_1(KTaO_3)_n]_2$ superlattices (per Nb atom).

n	1	2	3
$2W_{int}$ (meV)	4.282	1.089	0.278

Table 2 The total energy difference between paraelectric and ground-state structures, the lattice parameters, and the Berry-phase polarization for the ground-state structure of $(KNbO_3)_1(KTaO_3)_n$ superlattices with different n.

n	1	2	3	4	5	7
ΔE (meV)	11.738	7.532	5.898	5.365	5.090	4.625
$a = b$ (Å)	3.9656	3.9567	3.9508	3.9478	3.9459	3.9437
c (Å)	7.9235	11.8500	15.7908	19.7298	23.6680	31.5436
γ (°)	89.9581	89.9746	89.9866	89.9913	89.9936	89.9958
$\alpha = \beta$ (°)	89.9701	90.0	90.0	90.0	90.0	90.0
$\lvert P_s \rvert$ (C/m^2)	0.2515	0.1192	0.0822	0.0612	0.0486	0.0338

Table 4 The ground-state structure, lattice parameters, the Berry-phase polarization (P_x, P_y, P_z), and the in-plane polarization in the $KNbO_3$ layer (P_x^*, P_y^*) for $(KNbO_3)_1(KTaO_3)_n$ superlattices grown on $KTaO_3$ substrate.

n	1	2	3	4	5
structure	*Cm*	*Cm*	*Cm*	*Cm*	*Amm*2
$a = b$ (Å)	3.9374	3.9374	3.9374	3.9374	3.9374
c (Å)	7.9903	11.9126	15.8358	19.7591	23.6861
$P_x = P_y$ (C/m^2)	0.0730	0.0605	0.0507	0.0442	0.0385
P_z (C/m^2)	0.2546	0.1769	0.1216	0.0710	0.0000
$P_x^* = P_y^*$ (C/m^2)	0.1322	0.1495	0.1585	0.1646	0.1675

be understood as follows. For $KNbO_3$ grown on $KTaO_3$ substrate, the ground-state structure is *Cm*. The uniform P_x-component of polarization does not create macroscopic electric field in SLs, so that the interaction of polarizations in neighboring layers is weak and P_x varies slowly with increasing n. On the contrary, the energy needed to maintain the same electric displacement fields normal to the layers in two materials increases with increasing the thickness of nonpolar layer n, thus weakening the instability of z-polarized unstable phonon. This results in fast decrease of P_z with increasing n.

Another interesting result that follows from Table 4 is an increase of the in-plane polarization in the $KNbO_3$ layer with increasing n. At first sight, this result seems strange because the in-plane lattice parameter in SLs remains unchanged. However, if one takes into account the influence of P_z on the effective charge of the E_u soft mode, the result can be explained. According to our calculations, in strained $KNbO_3$ grown on $KTaO_3$ substrate the effective charge decreased by 7.7% when the structure transformed from paraelectric *P*4/*mmm* to polar *P*4*mm* phase. Thus, when increasing n, the decrease in P_z results in an increase of effective charge of E_u mode. This strengthens the dipole-dipole interaction and increases the instability of the soft mode.

4 Discussion The critical size of ferroelectric particles and critical thickness of ferroelectric thin films, below which the ferroelectricity vanishes, are of principle importance. First studies of ferroelectric nanoparticles indicated that the ferroelectricity vanishes when the particle diameter is about 100 Å. However, the experiments on ultrathin ferroelectric films [29] and theoretical calculations from first principles [30–34] have shown that a non-zero polarization normal to the film surface is retained in films with a thickness of 3–6 unit cells if the depolarizing field is compensated. In the PVDF-TrFE copolymer films the switchable polarization was observed in films with a thickness of 10 Å (two monolayers) [35]. The ferroelectric state with the polarization *parallel* to the layers was predicted for $BaTiO_3$ and $PbTiO_3$ films with a thickness of 3 unit cells [36, 37] and PZT films with a thickness of one unit cell [38]. Unstable ferroelectric mode with the polarization in the film layer was predicted for films of $PbB'_{1/2}B''_{1/2}O_3$ solid solutions with a thickness of one unit cell [39]. In all these cases two-dimensional ferroelectricity was a result of decreasing the physical size of the samples in one direction. In contrast, in this work, quasi-two-dimensional ferroelectricity appears as a result of specific interactions in superlattices and can be observed in structures of arbitrary thickness.

The polarization profiles obtained in this work agree with results of atomistic modeling of $KNbO_3/KTaO_3$ superlattices [16, 17]. According to the data of these papers, the polarization parallel to the layers decreased by 3–4 times when shifting by one unit cell from the heterojunction (in contrast to the behavior of the polarization normal to the layers). Our conclusion about the instability of A_{2u} phonon in the paraelectric phase of SLs with $n \leq 3$ agrees with results of [19] only partially: in the latter paper stable polar phase with *P*4*mm* symmetry was obtained only in SLs with $n = 1$ and 2. The instability of the E_u mode in SL with $n = 7$ agrees with that observed in Ref. [21], but in this work it is much stronger.

However, in spite of qualitative agreement of results of this and previous works, the physical conclusions that we make are very different. First, in all stress-free $KNbO_3/KTaO_3$ superlattices the *P*4*mm* phase is not the ground-state structure, as well as in $KTaO_3$-supported superlattices with $n \geq 5$. The reason for this is a tendency of polarization in SLs to incline towards the layer plane. This tendency was revealed in a number of ferroelectric superlattices [20] and was shown to be a way to reduce the electrostatic and mechanical energies in them.

Second, the existence of stable polarization in $KNbO_3$ layers with a thickness of one unit cell and very inhomogeneous distribution of polarization, which is accompanied by exponential decay of the interaction energy between neighboring layers with increasing the interlayer distance, result in a new, previously unknown feature of the ground-state structure of $(KNbO_3)_1(KTaO_3)_n$ superlattice—the formation of an array of nearly independent ferroelectrically ordered planes in the bulk of the superlattice.

Phonon dispersion curves calculated using the interpolation technique from phonon frequencies at Γ, X, M, Z, R, and A points of the Brillouin zone for *P*4/*mmm* phase of $(KNbO_3)_1(KTaO_3)_n$ superlattices enable to clarify the origin of unstable phonon modes at the boundary of the Brillouin zone in these SLs [19, 20]. The dispersion curves for these modes are shown in Fig. 3. Analysis of the eigenvectors shows that the doubly degenerate unstable mode along Γ−Z direction is polarized along [100] and [010] directions while

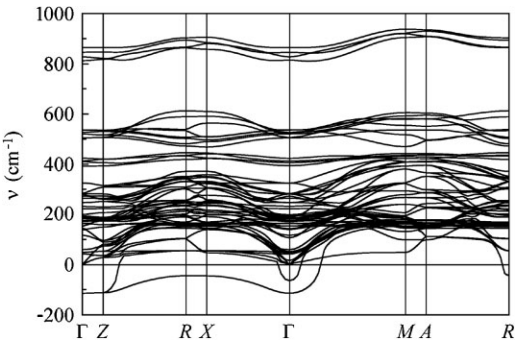

Figure 3 Phonon dispersion curves for paraelectric *P*4/*mmm* phase of $(KNbO_3)_1(KTaO_3)_3$ superlattice.

nondegenerate unstable modes along Z–R, R–X, and X–Γ directions are polarized along [010] direction. The character of atomic displacements for these modes is fully consistent with the ferroelectric instability of . . .–O–Nb–O–. . . chains first observed in $KNbO_3$ [40], but in SLs the chains are formed only in [100] and [010] directions. A nearly zero dispersion of the unstable mode along Γ–Z direction means that the interlayer interaction is weak, and the instability against the formation of ferroelectrically ordered planes does not depend on whether neighboring planes are ordered ferroelectrically or antiferroelectrically. This supports the idea about independence of ferroelectrically ordered planes in SLs with thick $KTaO_3$ layers.

The obtained results demonstrate that in complex systems in some cases the polarization calculated using the Berry phase method cannot be interpreted as a bulk property. The variation in polarization in the unit cell can be so large that the ferroelectric system can acquire the properties of two-dimensional system. To avoid interaction between polarized layers, the polarization should be parallel to the layers (a non-zero normal component results in appearance of electric fields in the structure, and 2D behavior transforms to 3D one). This means that to create arrays of nearly independent ferroelectrically ordered planes in [001]-oriented superlattices, we need SLs with *Pmm*2 or *Amm*2 ground-state structure.

Complex ferroelectric/antiferroelectric behavior has been observed recently in Ruddlesden–Popper (RP) superlattices [41, 42], in which appearance of Goldstone-like rotational mode of the in-plane polarization is possible [43]. In stress-free $BaTiO_3$/SrO superlattices, the in-plane polarization appeared in SLs with the $BaTiO_3$ slab thickness of $n \geq 4$ unit cells; when these SLs were epitaxially grown on $SrTiO_3$ substrates, the polar structure transformed to antiferroelectric (domain-like) structure in which out-of-plane polarization alternated along [100] or [110] directions in each of $BaTiO_3$ slabs. This antiferroelectric structure was studied in more detail in $BaTiO_3$/BaO superlattices grown on $SrTiO_3$ substrate [42]. The instability at X point of the Brillouin zone appeared in structures at a thickness of $BaTiO_3$ slabs of 2–3 unit cells while the zone-center ferroelectric instability appeared only at $n = 4$–5 unit cells. It was supposed that a huge depolarizing field appears in SLs because of the difficulty to polarize BaO layer, and this strongly harden the A_{2u} ferroelectric mode at the Γ point. At the same time, the *X* phonon mode arising from the same ferroelectric instability of . . .–O–Ti–O–. . . chains does not produce macroscopic depolarizing field, and so the influence of the BaO layer on the phonon frequency is weaker. This explains why in RP superlattices the antiferroelectric phase can appear before the ferroelectric one.

The superlattices studied in this work differ markedly from RP ones [41, 42] because in our case the ferroelectric layers are separated by the layers of incipient ferroelectric, which has much higher dielectric constant compared to SrO (BaO). In contrast to the RP superlattices, our SLs do not exhibit antiferrodistortive instability. In addition, the thickness of the $KNbO_3$ layer which is necessary to induce ferroelectricity in our SLs is only one unit cell. To test the influence of the thickness of $KNbO_3$ layer on the appearance of aforementioned antiferroelectric structure, we performed additional calculations for stress-free SLs in which $KNbO_3$ layers with a thickness of 3 and 5 unit cells were separated with a thick (3 unit cells) $KTaO_3$ layer.

The calculations showed that all three $(KNbO_3)_n(KTaO_3)_3$ SLs with $n = 1$, 3, and 5 studied in this work are unstable against the appearance of antiferroelectric structure with out-of-plane displacements of ions described by *X* phonon (*Pmma* space group), but in all cases the energies of corresponding equilibrium structures are higher than the energies of the ferroelectric *Amm*2 phases. This means that antiferroelectric *Pmma* structure is not the ground state of stress-free $KNbO_3/KTaO_3$ superlattice. Moreover, the calculation of phonon frequencies at the Γ point for *Pmma* phase of $(KNbO_3)_1(KTaO_3)_1$ SL showed that this antiferroelectric phase exhibits the *ferroelectric* instability against the appearance of in-plane polarization. Therefore one can conclude that like for RP superlattices, the antiferroelectric phase of $KNbO_3/KTaO_3$ superlattices with out-of-plane displacements of ions is indeed more stable compared to the ferroelectric phase with out-of-plane polarization, but nevertheless the ferroelectric *Amm*2 phase with non-zero in-plane polarization has the lowest energy. In RP superlattices studied so far, the in-plane polarization was suppressed by epitaxial strain from the substrate, and the antiferroelectric phase was the most stable one.

We discuss now possible applications of the arrays of nearly independent ferroelectrically ordered planes. At first sight, the fact that the polarization in $KNbO_3/KTaO_3$ superlattices is oriented in the layer plane can be regarded as not intriguing. So far, when discussing the ferroelectricity in ultrathin films, main attention was paid to films in which the polarization is normal to the film surface as these films are more suitable for different applications. However, the possibility of formation of an array of nearly independent ferroelectrically ordered planes opens new opportunities in design of electronic devices. These structures can be used as a medium for three-dimensional information recording. Accepting that the lateral size of a ferroelectric domain that

has a long-time stability against spontaneous reversal of polarization is 250 Å [44], for the interlayer distance of 16 Å (the period of SL with $n = 3$) the estimated 3D information storage density can be as high as $\sim 10^{18}$ bit/cm^3. This value exceeds the density achieved in modern optical 3D recording media by six orders of magnitude.

One can add that the two-component order parameter, which admits the rotation of polarization by 90° in each of independent $KNbO_3$ layers, offers one more opportunity. In ferroelectric RAM devices with polarization normal to the film surface the leakage currents and conductivity of films are main effects preventing the realization of non-destructive read-out. The systems admitting the rotation of polarization by 90° have a feature that enables to overcome this disadvantage—a non-destructive read-out of polarization in them can be realized using the anisotropy of the dielectric constant in the layer plane. Our calculations predict that in $(KNbO_3)_1(KTaO_3)_n$ superlattice with $n = 2$ the dielectric constants parallel and perpendicular to the polarization are 142 and 188, whereas in the SL with $n = 3$ they are 225 and 281.

5 Conclusions In this work, the density-functional theory was used to calculate the properties of $(KNbO_3)_1(KTaO_3)_n$ superlattices with $n = 1$–7. It was shown that the ferroelectric ordering is retained in $KNbO_3$ layers of minimal thickness (one unit cell) whereas the interaction energy between neighboring polarized layers decreases exponentially with increasing parameter n. Array of nearly independent ferroelectrically ordered planes with the polarization lying in the layer plane was shown to be the ground-state structure for SLs with $n \geq 2$. Using of these structures as a medium for three-dimensional (3D) information recording enables to attain the information storage density of 10^{18} bit/cm^3.

The calculations presented in this work were performed on the laboratory computer cluster (16 cores) and SKIF-MGU "Chebyshev" supercomputer at Moscow State University.

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