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Designing Multifunctionality via Assembling Dissimilar Materials: Epitaxial AlN/ScN Superlattices

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Abstract

First-principles calculations are performed to investigate the effect of epitaxial strain on energetic, structural, electrical, electronic and optical properties of 1×1 AlN/ScN superlattices. This system is predicted to adopt four different strain regions exhibiting different properties, including optimization of various physical responses such as piezoelectricity, electro-optic and elasto-optic coefficients, and elasticity. Varying the strain between these four different regions also allows the creation of an electrical polarization in a nominally paraelectric material, as a result of a softening of the lowest optical mode, and even the control of its magnitude up to a giant value. Furthermore, it results in an electronic band gap that can not only change its nature (direct *versus* indirect), but also cover a wide range of the electromagnetic spectrum from the blue, through the violet and near ultraviolet, to the middle ultraviolet. These findings thus point out the potential of assembling two different materials inside the same heterostructure to design multifunctionality and striking phenomena.

Having multifunctional materials for which several properties can be simultaneously controlled or optimized is desired [1]. One possible idea for designing such materials is to bring together within the same heterostructure two compounds that have separately different structural phases, and play with an external control knob to go from one of these phases to another via transitions. One example to test this idea is to (i) combine AlN (or GaN or InN) with ScN to form superlattices, since AlN, GaN and InN ground states are wurtzite [2, 3] while ScN has been predicted to exhibit a (meta)stable layered phase in its hexagonal form [4–6]; and (ii) choose the control knob to be the epitaxial strain, since it is known to be an effective tool to enhance properties and generate novel phenomena [7–13]. For instance, epitaxial strain can turn the nominally paraelectric SrTiO₃ into a ferroelectric [8, 9], as well as induce phase transitions and large piezoelectric, dielectric, elasto-optic and electro-optic responses in many systems (see, e.g., Refs. [7, 11–13]).

Moreover, GaN/ScN and InN/ScN superlattices have been predicted to experience under hydrostatic pressure a rare phenomenon [14], that is a so-called *isostructural* phase transition [15–17]. This further emphasizes the potential for ScN-based superlattices to have striking features, allowing for the modification and/or optimization of various properties as a result of the phase transitions [18]. Some (Ga,Sc)N and (In,Sc)N ordered alloys have also been predicted to exhibit symmetry-breaking transitions between different phases when under epitaxial strain, accompanied by a large piezoelectricity [19]. On the other hand, we are not aware of systematic investigation of AlN/ScN superlattices, despite the fact that a large piezoelectric coefficient d_{33} and ferroelectric switching have both been observed in *disordered* Sc_xAl_{1-x}N alloys [20, 21] as a result of the transformation towards a polar structure of the aforementioned predicted peculiar layered phase of pure ScN.

The aim of this Letter is to investigate the effect of epitaxial strain on the properties of AlN/ScN superlattices. As we will see, many different physical properties can be altered and optimized, partly because of the strain-driven occurrence of a phase transition.

This work focuses on properties of epitaxial 1×1 AlN/ScN superlattices as a function of the in-plane lattice constant (results for epitaxial 1×3 AlN/ScN superlattices are also provided in the Supplemental Material (SM) [22]). In the following, $\eta_{\text{in}} = (a - a_{\text{eq}})/a_{\text{eq}}$ characterizes the magnitude of the epitaxial strain, with the zero of strain corresponding to the in-plane lattice constant, a_{eq} , of the equilibrium structure (i.e., the one yielding the lowest total energy). We also limited the calculations for strains ranging between -6% and $+6\%$.

We perform first-principle calculations (see the SM [22]) to predict (i) structural parameters, with our present results being estimated to underestimate the lattice parameters of 1×1 AlN/ScN

by about 0.78%; (ii) electronic band gaps, with our data being estimated to underestimate the experimental values by about 1.58 eV in the studied 1×1 AlN/ScN superlattices; (iii) electrical polarization; (iv) the S_{33}^{epi} elastic compliance element; (v) e_{33} piezoelectric stress coefficients; (vi) d_{33}^{epi} piezoelectric strain coefficients; and (vii) elasto-optic coefficients, p_{ij} (in Voigt notation).

We also predicted electro-optic (EO) coefficients via density functional perturbation theory, which is based on the linear response of the optical dielectric tensor induced by a static (or low frequency) electric field \mathcal{E}_k [25, 26]. Practically, the linear EO (Pockels) effect is described by the third-rank EO tensor r_{ijk} ,

$$\Delta(\varepsilon^{-1})_{ij} = \sum_{k=1}^3 r_{ijk} \mathcal{E}_k, \quad (1)$$

where $(\varepsilon^{-1})_{ij}$ is the inverse of the optical dielectric tensor (approximated by the electronic dielectric tensor). Two different types of EO coefficients should be distinguished [13, 25, 26], namely the clamped (strain-free) *versus* the unclamped (stress-free for σ_{33} but clamped in-plane) ones. The clamped EO tensor, r_{ijk}^{η} , can be expressed as a sum of two contributions: (1) a bare electronic part, r_{ijk}^{el} , which depends on the nonlinear optical dielectric susceptibility $\chi_{ijk}^{(2)}$ (which defines the second-order change of the induced-polarization with respect to \mathcal{E}_k); and (2) an ionic contribution, r_{ijk}^{ion} , which depends on the first-order change of the linear dielectric susceptibility due to atomic displacements. This ionic contribution is related to the Raman susceptibility α_{ij}^m of mode m and polarity p_k^m of a transverse optical mode of frequency ω_m . One thus has

$$r_{ijk}^{\eta} = r_{ijk}^{\text{el}} + r_{ijk}^{\text{ion}} = \frac{-8\pi}{n_i^2 n_j^2} \chi_{ijk}^{(2)} - \frac{4\pi}{n_i^2 n_j^2 \sqrt{\Omega_0}} \sum_m \frac{\alpha_{ij}^m p_k^m}{\omega_m^2}, \quad (2)$$

where n_i and n_j are the principal refractive indices, and Ω_0 is the cell volume.

Regarding the unclamped EO coefficients r_{ijk}^{σ} , they can be expressed as

$$r_{ijk}^{\sigma} = r_{ijk}^{\eta} + \sum_{\alpha, \beta} p_{ij\alpha\beta} d_{\alpha\beta k}^{\text{epi}}, \quad (3)$$

where $p_{ij\alpha\beta}$ are elasto-optic constants and $d_{\alpha\beta k}^{\text{epi}}$ are piezoelectric strain coefficients. In other words, unclamped EO coefficients consist of the sum of the EO clamped coefficients and of a second term involving the products between elasto-optic constants and the piezoelectric strain coefficients.

Let us now present some of our results for the 1×1 AlN/ScN superlattices as a function of strain. Figure 1(a) reports the total energy, with the zero of energy corresponding to the ground state. Figures 1(b)–1(c) display the c/a axial ratio and internal parameters (u_{AlN} and u_{ScN}), (see SM [22] for the definition of these internal parameters). Figure 1(d) shows the out-of-plane polarization P_z .

Figure 1(e) reports predictions about the vibrational properties consisting of phonon frequencies at the Γ point. Finally, Fig. 1(f) shows various electronic band gaps, such as the difference in energy between the conduction band minimum and valence band maximum (which we will refer to as the electronic band gap), but also direct band gaps at the Γ , H and K points of the first Brillouin zone.

Figure 1(a) reveals that 1×1 AlN/ScN exhibits two minima in the total energy-*versus*-strain curve, that occur at 0% and -5% strains. They respectively correspond to a hexagonal-derived structure [to be denoted as *h*-derived, and shown in Fig. 2(a)] and a wurtzite-derived phase [to be coined as *w*-derived, and displayed in Fig. 2(d)].

The behaviors of the properties reported in Figs. 1(b)–1(f) allow the determination of four strain regions. Region I pertains to strain ranging between $+6\%$ and $\sim -0.7\%$ and is characterized by an axial ratio that is relatively close to that of the layered hexagonal *h* phase of ScN (1.21). P_z is null, consistent with the paraelectric nature of this *h* phase, and the u_{AlN} and u_{ScN} parameters are fixed to 0.5 by symmetry. We can thus classify Region I as *h*-derived. The phase in Region I has the (paraelectric) $P\bar{6}m2$ space group (note that the *h* phase in pure ScN has a $P6_3/mmc$ space group) [37]. In Region I, the LDA-calculated direct band gap at Γ [see blue square in Fig. 1(f)] varies from 1.0 to 1.92 eV. Correcting the LDA underestimation as indicated above should then lead to a variation from about 2.58 to 3.5 eV, which will then correspond to colors covering blue, violet and ultraviolet, which is promising for some applications (e.g. blue-violet laser and ultraviolet sensor devices) [38–41]. The nature of the electronic band gap [see red circle in Fig. 1(f)] changes from direct at Γ , for strains between $+6\%$ and $+1.5\%$, to indirect (Γ -to-K) between $+1\%$ and -0.7% . Figure 1(e) also reveals that the lowest $A_2''^{(1)}$ phonon at the Γ point continuously softens towards a zero frequency mode when approaching the transition from Region I to Region II, after which it becomes an $A_1^{(1)}$ mode of the *w*-derived structure.

Such softening leads to the appearance of a polar Region II of $P3m1$ symmetry in which a polarization appears, extending from $\eta_{\text{in}} \sim -0.7\%$ to -1.8% . The softening of the polar mode has two origins: (1) Al prefers to form four-fold coordinated Al-N covalent bonds by displacing towards one of the out-of-plane N_3 ions, which is in line with the fact that AlN has a polar wurtzite ground state structure; (2) There is a steric effect related to the in-plane Sc-N bonds (see SM [22]).

In Region II, c/a has a different slope with respect to Region I, while the internal parameters now decrease when the compressive strain strengthens in magnitude. Region II is associated with an intermediate structure that is close to the *h*-derived structures [see Fig. 2(b)]. In this Region II, the LDA electronic band gap is indirect (Γ -to-K) and varies from 1.67 to 2.24 eV (the corrected-

with-respect-to-LDA value varies from 3.25 to 3.82 eV, lying in the near ultraviolet region), and the direct band gap at the K point linearly and significantly increases when increasing the magnitude of the compressive strain.

The left border of Region II at $\sim -1.8\%$, marking the passage to another Region to be called Region III (that also has a $P3m1$ space group), is marked by several features. For instance, the direct band gap at the K point stops being sensitive to strain and an *anticrossing* between the two lowest phonon modes of A_1 symmetry, that are denoted as $A_1^{(1)}$ and $A_1^{(2)}$ in Fig. 1(e), occurs there. Details of such anticrossings are further provided in the SM [22]. The axial ratio in Region III also adopts a linear behavior with strain that is slightly different from that of Region II, until c/a deviates from linearity at about -3% . Such a deviation marks the left border of Region III and the beginning of Region IV [42]. The structure associated with Region III in the range of about -1.8% to -3% is another intermediate phase that is polar but now close to the wurtzite-derived structures [see an example in Fig. 2(c)]. In terms of electronic structure, the band gap becomes direct again at Γ in Region III and varies from 2.24 to 2.7 eV according to LDA (the corrected value varies from 3.82 to 4.28 eV, which is associated with the electromagnetic spectrum from near ultraviolet to middle ultraviolet).

Finally, Region IV occurs for η_{in} smaller than -3% and corresponds to wurtzite-derived structures, as evidenced by a c/a axial ratio being close to 1.6 for large compressive η_{in} . Like Regions II and III, Region IV adopts the $P3m1$ space group. The right border of Region IV is marked by a direct band gap at the H-point possessing a kink, and is mostly characterized by different behaviors of the direct band gaps for compressive strains above 3% : the one at Γ increases, while the one at H decreases, and the one at the K-point is nearly insensitive to η_{in} . As a result, the electronic band gap is direct at the Γ point from $\sim -3\%$ to -4.5% and then direct again but at the H point from $\sim -5\%$ to -6% , with corrected values ranging between 4.28 to 4.8 eV (which is associated with middle ultraviolet) and then from 4.88 to 4.78 eV (which also corresponds to middle ultraviolet). Note that ultraviolet sensor devices are important for space communications, bio-medical instrumentation and high temperature plasma research, etc. [43].

It is remarkable that Figs. 1-2 reveal so many striking features: (1) the continuous evolution of the structure from a h -derived to a w -derived phase; (2) an out-of-plane polarization increasing from 0 to 1.07 C/m^2 , when passing through Region II to Region IV, with this value of 1.07 C/m^2 being even bigger than the polarization of the tetragonal phase of PbTiO_3 films (0.87 C/m^2) [11]; and (3) the control of the nature (direct *versus* indirect) and value of the electronic band gap,

covering the blue, violet, and near and middle ultraviolet parts of the electromagnetic spectrum. The Born effective charges also show some specific evolutions from Regions I to IV (see SM [22]).

It is also important to know that our theoretical in-plane lattice constant corresponding to $\eta_{\text{in}} = 0\%$ is 3.507 Å in the 1×1 AlN/ScN superlattices, which, according to the aforementioned expected underestimation of 0.78% of the lattice parameters, should correspond to a value of 3.534 Å. The substrates should thus correspond to the following strains: Sc (3.309 Å, that is -6.4%), ZnO (3.2496 Å, that is -8%), Hf (3.18 Å, that is -10%), and wurtzite InN (3.544 Å, that is 0.3%) [2, 44]. Similar to the strategy of Ref. [45], one can envision growing 1×1 AlN/ScN superlattices with different thicknesses on such substrates to allow the strain to partially relax by different amounts, which will then result in covering all the strain ranges associated with Regions I to IV. Note also that the w -derived structure of 1×1 AlN/ScN can be seen as a (meta)stable polymorph of the h -derived ground state of this system, implying that it should be practical to reach large compressive strains with respect to the h -derived ground state – consistent with the fact that strain of this order of magnitude has been experimentally found in BiFeO₃ films because of the existence of a metastable T phase in addition to the R ground state [46–48].

Let us now reveal the impact of these different regions on responses, such as EO and elasto-optic coefficients, piezoelectricity, and elastic responses (as reported in Fig. 3). For that, let us first recall that the EO tensor in the $P\bar{6}m2$ space group has one independent nonzero coefficient r_{22} , while it has four independent elements in Voigt notation in the $P3m1$ (C_{3v}) symmetry: r_{13} , r_{22} , r_{33} and r_{42} . Figures 3(a) and 3(b) display the clamped and unclamped EO coefficients, respectively, as a function of the misfit strain. For the clamped EO coefficients, we indeed obtain $r_{13}^{\eta} = r_{33}^{\eta} = r_{42}^{\eta} = 0$ and very small values of r_{22}^{η} in Region I. At the boundary between Regions I and II, large values of the clamped EO coefficient r_{33}^{η} are predicted, as a result of the aforementioned softening of the $A_2''^{(1)}$ and $A_1^{(1)}$ modes (see the second term of Eq. (2) indicating that small frequencies should give rise to large clamped coefficients, and the decomposition analysis in the SM [22]). Strikingly, such large values near this critical strain region are larger than the experimental values in LiNbO₃ and tetragonal BaTiO₃ [49–51]. Also note that none of the clamped r_{ij}^{η} exhibit any significant change when crossing the boundaries between Regions II, III and IV.

For the case of unclamped EO coefficients, we just show r_{13}^{σ} and r_{33}^{σ} in Fig. 3(b), because, unlike for r_{22}^{σ} and r_{42}^{σ} , they can be significantly enhanced from their clamped values in Region II. For instance, at $\eta_{\text{in}} \sim -0.7\%$, unclamped r_{33}^{σ} and r_{13}^{σ} also show a very large value. Note that r_{33}^{σ} is strongly dependent on the strain with a narrow peak, which implies that a small change of

epitaxial strain can make a large difference in the EO coefficients. Note also that, experimentally, the epitaxial strain can evolve with temperature if the superlattice and substrate have different thermal expansion coefficients. Figures 3(c) and 3(d) show that the d_{33}^{epi} , p_{13} and p_{33} coefficients are maximal and really large in magnitude at this boundary, explaining why the EO coefficients are enhanced in the unclamped case with respect to those of the clamped situation for strains close to -0.7% , as fully consistent with Eq. (3). In particular, d_{33}^{epi} shows a large value at $\eta_{\text{in}} \sim -0.7\%$, which is many times larger than the one found in scandium aluminum nitride alloy thin films [20]. Similarly, a large value of elasto-optic coefficients p_{33} is predicted at the boundary between Regions I and II, with this value being larger than the ones found in most trigonal, tetragonal and hexagonal crystals [52]. The piezoelectric stress coefficient e_{33} of Fig. 3(e) adopts a strong peak at this phase boundary between Regions I and II (as a result of the softening of $A_2''^{(1)}$ and $A_1^{(1)}$ modes and the $P\bar{6}m2$ -to- $P3m1$ transition), and the elastic compliance coefficient S_{33}^{epi} of Fig. 3(f) has a maximum within Region III, which corresponds to the saddle point of the total energy peak in Fig. 1(a). Another interesting feature is that p_{13} has an extremum at the boundary between Regions III and IV. On the other hand, r_{33}^{σ} and r_{13}^{σ} behave rather smoothly in Regions II, III and IV, because of the continuous behavior of their corresponding clamped values as well as the product between the $p_{ij\alpha\beta}$ elasto-optic constants and $d_{\alpha\beta k}^{\text{epi}}$ piezoelectric strain coefficients – in line with Eq. (3).

In summary, based on first-principles calculations, we predict the existence of four strain-induced regions in 1×1 AlN/ScN superlattices, that are accompanied by an optimization and/or control of several properties as well as by striking features. In particular, the continuous evolution from the h -derived phase of Region I to an intermediate state characterizing Region II is driven by a softening of the the lowest $A_2''^{(1)}$ and $A_1^{(1)}$ mode and a phase transition, which naturally lead to very large piezoelectric, electro-optic and elasto-optic responses as well as the appearance of an electrical polarization. Moreover, Region III possesses the maximal value of the S_{33}^{epi} elastic compliance, and exhibits anticrossing between two phonon modes of identical A_1 symmetry on its right border. A subtle Region IV is further identified, based on anomalies in electronic band structure and the lowest magnitude of the p_{13} elasto-optic coefficient. It is also remarkable that the strain evolution from -6% to $+6\%$ allows (i) a systematic control of the magnitude of the polarization from a null to rather large value of 1.07 C/m^2 ; and (ii) the electronic band gap to cover a large range of the electromagnetic spectrum including blue, violet, near and middle ultraviolet. The SM [22] further demonstrates that many of these spectacular effects still hold when investigating epitaxial 1×3 AlN/ScN superlattices, not only with the discovery of the largest electro-optic

r_{33}^{σ} and elasto-optic p_{33} coefficients ever predicted or measured, but also cover a wider range of the electromagnetic spectrum from the yellow color to middle ultraviolet (note that r_{33}^{σ} and p_{33} are also larger than that of the 1×1 AlN/ScN superlattices around the boundary between Regions I and II, for any relative strain with respect to the one defining such boundary). We thus hope that this Letter will encourage the experimental investigation of superlattices made from rather dissimilar materials in the search for optimization and control of properties.

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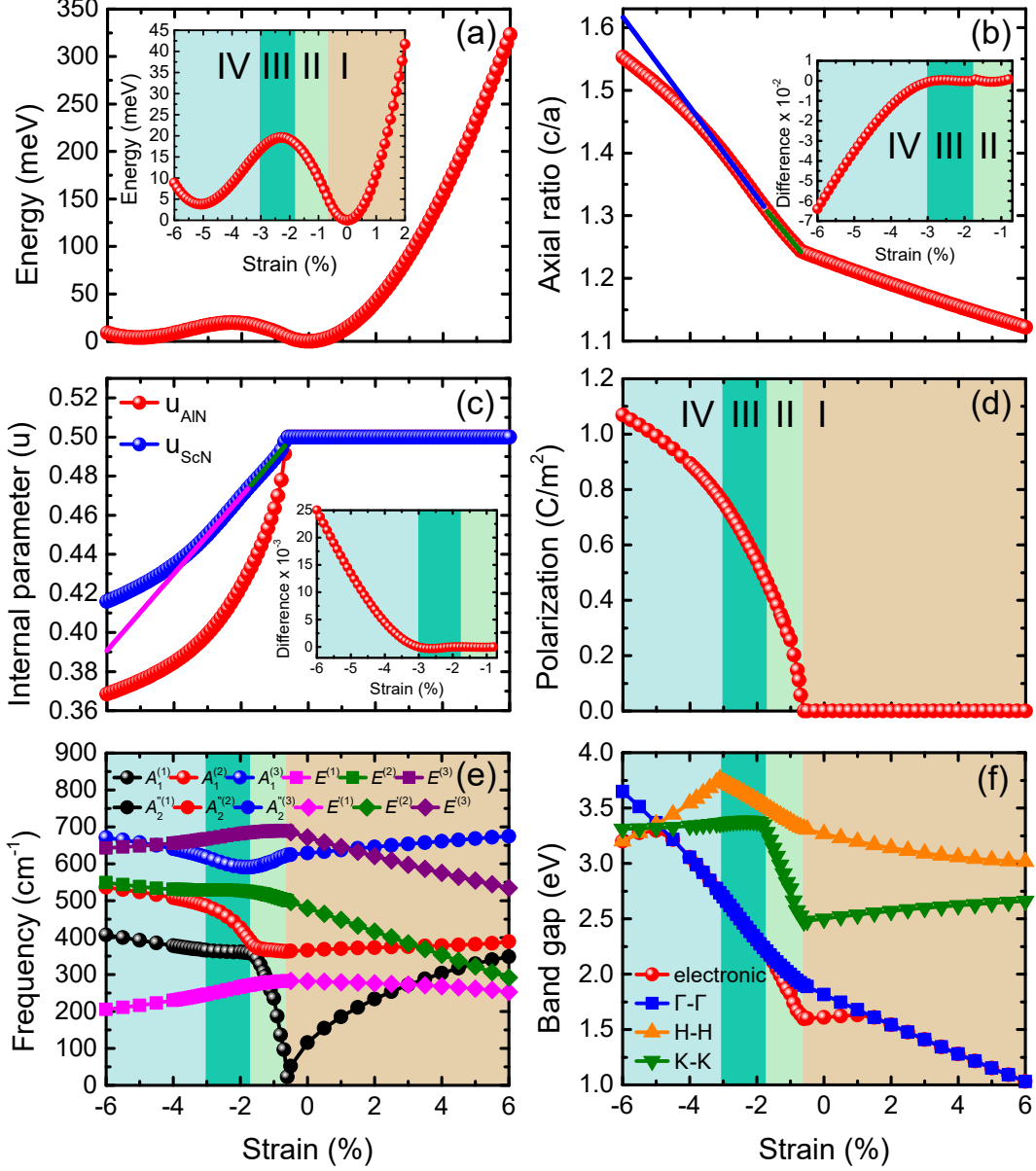


FIG. 1. Structural properties of 1×1 AlN/ScN as a function of strain: (a) total energy (the inset zooms in the data for strains between -6% and $+2\%$); (b) axial ratio c/a [the inset shows the difference between the c/a data and two straight lines that pass through Regions II (green line) and III (blue line)]; (c) internal parameters [the inset shows the difference between the u_{ScN} data and two straight lines that pass through Regions II (green line) and III (magenta line)]; (d) polarization P_z ; (e) phonon spectrum at the Γ point of the first Brillouin zone; (f) the electronic band gap and the direct band gaps at Γ , H and K.

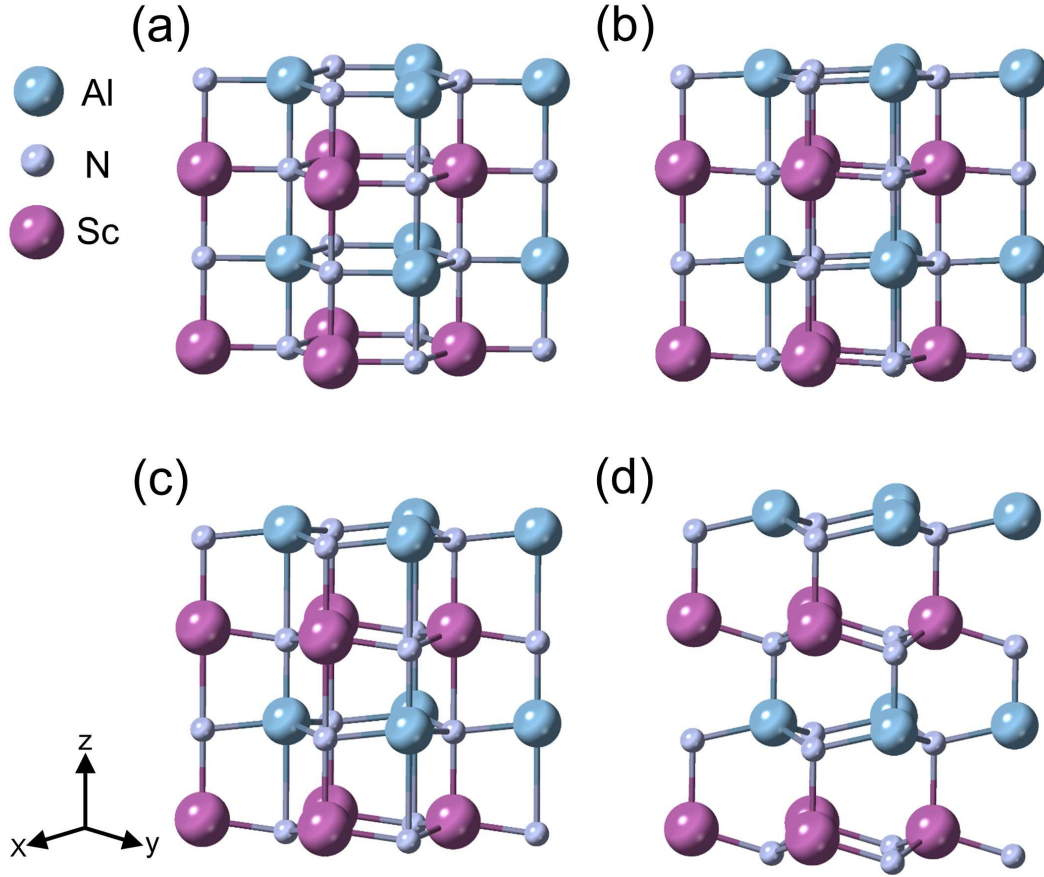


FIG. 2. Crystal structures of the studied 1×1 AlN/ScN superlattices. Panel (a) depicts the hexagonal-derived structure (for the strain of 0%) of Region I. Panel (b) shows an intermediate phase for $\eta_{\text{in}} = -1\%$ in Region II. Panel (c) displays an intermediate phase of Region III for $\eta_{\text{in}} = -2\%$. Panel (d) shows the wurtzite-derived structure of Region IV for $\eta_{\text{in}} = -5\%$.

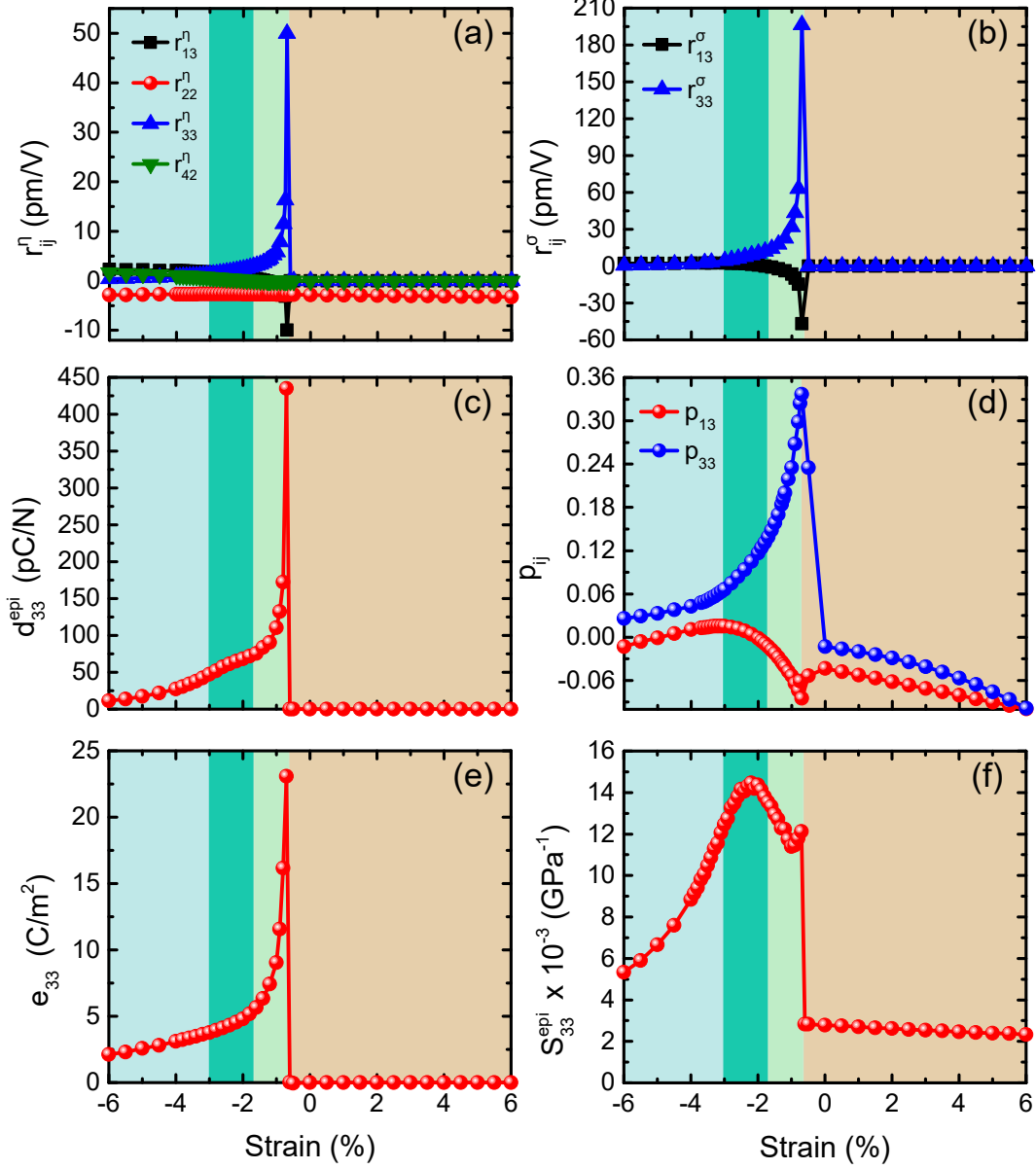


FIG. 3. Evolution with strain of the (a) clamped EO coefficients; (b) unclamped EO coefficients; (c) piezoelectric strain coefficients d_{33}^{epi} ; (d) elasto-optic coefficients p_{13} and p_{33} ; (e) piezoelectric stress coefficients e_{33} ; and (f) elastic compliance constants S_{33}^{epi} in the studied 1×1 AlN/ScN superlattices.