

## Spontaneous polarization and piezoelectric constants of III-V nitrides

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The spontaneous polarization, dynamical Born charges, and piezoelectric constants of the III-V nitrides AlN, GaN, and InN are studied *ab initio* using the Berry-phase approach to polarization in solids. The piezoelectric constants are found to be up to ten times larger than in conventional III-V and II-VI semiconductor compounds, and comparable to those of ZnO. Further properties at variance with those of conventional III-V compounds are the sign of the piezoelectric constants (positive as in II-VI compounds) and the very large spontaneous polarization. [S0163-1829(97)51740-1]

In this paper we report an *ab initio* study of the spontaneous polarization, piezoelectric constants, and dynamical charges of the III-V nitride semiconductors AlN, GaN, and InN.<sup>1</sup> This class of polarization-related properties is of obvious importance for the study of nitride-based piezodevices<sup>2</sup> and multilayer structures. In particular, knowledge of these properties allows an insightful treatment of the polarization (and ensuing electric fields) in strained and polarized nitride junctions and superlattices under any strain condition, as discussed elsewhere.<sup>3</sup> From the present study, one of the first applications of the modern theory of polarization in solids<sup>4,5</sup> to real and “difficult” materials of technological interest, the nitrides emerge as highly unusual III-V materials, resembling II-VI oxides and in some respects ferroelectric perovskites. The results we report here are of special interest in view of the scarcity of the data (both experimental and theoretical) available at present for the nitrides.

In the absence of external fields, the total macroscopic polarization  $\mathbf{P}$  of a solid is the sum of the spontaneous polarization  $\mathbf{P}^{\text{eq}}$  in the equilibrium structure, and of the strain-induced or piezoelectric polarization  $\delta\mathbf{P}$ . In the linear regime, the piezoelectric polarization is related to the strain  $\epsilon$  by

$$\delta P_i = \sum_j e_{ij} \epsilon_j, \quad (1)$$

which defines the components of the piezoelectric tensor  $e_{ij}$  (Voigt notation is used).

The natural structure of the III-V nitrides is wurtzite, a hexagonal crystal structure defined by the edge length  $a$  of the basal hexagon, the height  $c$  of the hexagonal prism, and an internal parameter  $u$  defined as the anion-cation bond length along the (0001) axis in units of  $c$ . Wurtzite is the structure with highest symmetry compatible with the existence of spontaneous polarization,<sup>6,7</sup> and the piezoelectric tensor of wurtzite has three nonvanishing independent components. Therefore, the polarization in these materials system will both a spontaneous and a piezoelectric component.

In this work we shall restrict ourselves to polarizations along the (0001) axis; this is the direction along which both

standard bulk materials and nitride superlattices are grown. The spontaneous polarization along the  $z$  axis is  $\mathbf{P}^{\text{eq}} = P^{\text{eq}} \hat{\mathbf{z}}$ , and the piezoelectric polarization is simply expressed via the piezoelectric coefficients  $e_{33}$  and  $e_{13}$  as

$$\delta P_3 = e_{33} \epsilon_3 + e_{31} (\epsilon_1 + \epsilon_2), \quad (2)$$

where  $a_0$  and  $c_0$  are the equilibrium values of the lattice parameters,  $\epsilon_3 = (c - c_0)/c_0$  is the strain along the  $c$  axis, and the in-plane strain  $\epsilon_1 = \epsilon_2 = (a - a_0)/a_0$  is assumed to be isotropic. The third independent component of the piezoelectric tensor,  $e_{15}$ , is related to the polarization induced by a shear strain, and will not be considered in this work.

To linear order, the change in polarization can also be expressed as

$$\delta P_3 = \frac{\partial P_3}{\partial a} (a - a_0) + \frac{\partial P_3}{\partial c} (c - c_0) + \frac{\partial P_3}{\partial u} (u - u_0), \quad (3)$$

where  $a_0$ ,  $c_0$ , and  $u_0$  are the equilibrium lattice constants and internal parameter. (Note that the parameters  $a$ ,  $c$ , and  $u$  are not independent, because of the condition of vanishing forces along the axis direction.) Once the polarization derivatives in Eq. (3) are known, the macroscopic piezoelectric tensor coefficients can be expressed as

$$e_{33} = c_0 \frac{\partial P_3}{\partial c} + \frac{4ec_0}{\sqrt{3}a_0^2} Z^* \frac{du}{dc} \quad (4)$$

and

$$e_{31} = \frac{a_0}{2} \frac{\partial P_3}{\partial a} + \frac{2e}{\sqrt{3}a_0} Z^* \frac{du}{da}, \quad (5)$$

where

$$Z^* = \frac{\sqrt{3}a_0^2}{4e} \frac{\partial P_3}{\partial u} \equiv \mathcal{Z}_3^{(T)} \quad (6)$$

is the axial component of the Born, or transverse, dynamical charge tensor  $\mathcal{Z}^{(T)}$ . In the last equation above, it is implicit that the vector connecting the cation with the anion has

modulus  $uc$  and points in the direction of the  $c$  axis. The first term in Eqs. (4) and (5) is called the clamped-ion term, and it represents the effect of the strain on the electronic structure. The second term quantifies the effects of *internal* strain on the polarization. The derivatives of  $u$  with respect to  $c$  and  $a$  in Eqs. (4) and (5) are related to the strain derivatives of  $u$  through  $c_0 du/dc = du/d\epsilon_3$  and  $a_0 du/da = 2 du/d\epsilon_1$ .

We have calculated the polarization within the Berry phase approach of Ref. 4. The method's output is the difference in polarization among two states of a system, provided they can be connected by an adiabatic transformation which leaves the system insulating. The difference of electronic polarization  $\Delta\mathbf{P}_e$  among two systems is then related to the geometric quantum phase by

$$\Delta\mathbf{P}_e = \mathbf{P}_e(\lambda_2) - \mathbf{P}_e(\lambda_1), \quad (7)$$

where

$$\mathbf{P}_e(\lambda) = -\frac{2e}{(2\pi)^3} \int_{\text{BZ}} d\mathbf{k} \frac{\partial}{\partial \mathbf{k}'} \phi^{(\lambda)}(\mathbf{k}, \mathbf{k}') \Big|_{\mathbf{k}'=\mathbf{k}}, \quad (8)$$

the integration domain in momentum space is the reciprocal unit cell,  $\lambda$  characterizes the adiabatic transformation, and  $\phi^{(\lambda)}$  is the geometric quantum phase

$$\phi^{(\lambda)}(\mathbf{k}, \mathbf{k}') = \text{Im} \{ \ln [ \det \langle u_m^{(\lambda)}(\mathbf{k}) | u_n^{(\lambda)}(\mathbf{k}') \rangle ] \} \quad (9)$$

of the occupied crystal Bloch states  $u_n^{(\lambda)}(\mathbf{k})$ . The definition of the geometric quantum phase introduces an arbitrary constant in the value of the polarization so that the latter is well defined only modulo  $e\mathbf{R}/\Omega$ , with  $\mathbf{R}$  a real-space lattice vector. This is not a difficulty in practice, as one is generally interested in polarization changes such that  $|\Delta\mathbf{P}| \ll |e\mathbf{R}/\Omega|$ , so that no ambiguity can arise.

The adiabatic transformation labeled by  $\lambda$  is trivially identified as far as the strains  $\epsilon_1$ ,  $\epsilon_2$ , and  $\epsilon_3$  are concerned, so that the strain derivatives of the polarization within the wurtzite structure are easily calculated. On the other hand, the *absolute* value of the spontaneous polarization of the wurtzite structure [state  $\lambda_2$  in Eq. (7)] must be calculated as a difference relative to a structure having zero polarization, and functioning as reference state  $\lambda_1$ , since only polarization differences can be meaningfully calculated within the modern theory:<sup>4,5</sup> zinc blende is the natural choice for this purpose. One need not actually devise a gap-preserving adiabatic transformation turning zinc blende into wurtzite (a quite nontrivial task): the interface theorem of Ref. 8 proves that Eq. (7) can be used directly provided that an insulating interface can be built connecting the two different structures. As this is found to be the case<sup>3</sup> for the systems of relevance here, the absolute polarization of wurtzite nitrides can be easily computed (see also Ref. 7).

All present first-principles calculations are based on density-functional theory<sup>9</sup> in the local density approximation for the exchange-correlation energy functional, for which we adopt the Ceperley-Alder<sup>10</sup> form as parametrized by Perdew and Zunger.<sup>11</sup> The wave functions are expanded in a plane-wave basis set up to an energy cutoff of 25 Ry. Ultrasoft pseudopotentials<sup>12</sup> have been employed for all the elements involved in the calculations. In particular, the Ga and In pseudopotentials are constructed so as to include explicitly

TABLE I. Structural parameters for AlN, GaN, and InN.

	$a_0$ (bohr)	$c_0/a_0$	$u_0$
AlN	5.814	1.6190	0.380
GaN	6.040	1.6336	0.376
InN	6.660	1.6270	0.377

the semicore  $d$  electrons in the valence, resulting in a very accurate description of the electronic and structural properties.

All calculations for the wurtzite structure used a mesh of 12 Chadi-Cohen special points for the Brillouin-zone sampling.<sup>13</sup> The  $k$ -space integration for computing the polarization was instead done on a (4,4,10) Monkhorst-Pack<sup>14</sup> mesh; this amounts to 16  $\mathbf{k}_\perp$  points times 10  $\mathbf{k}_\parallel$  points. Refining the mesh to 360 points [the (6,6,10) mesh] gave no appreciable variation of the calculated values. The partial derivatives of the polarization appearing in Eq. (3) are easily evaluated numerically through the polarization change induced by typically a  $\pm 2\%$  change in the crystal parameters  $a$  and  $c$  (with  $u$  kept fixed, i.e., at clamped ions) and in the internal parameter  $u$ , around their equilibrium values.

Since we are interested in the spontaneous polarization, a careful determination of the equilibrium structure parameters is needed. The three independent structural parameters were obtained by polynomial interpolation of the total-energy values calculated over a grid of values of  $a$  and  $c$ . The internal parameter  $u$  was optimized at each  $(a,c)$  grid point following the Hellmann-Feynman forces. The results, summarized in Table I, are very close to those reported in our previous work.<sup>15</sup>

In Table II we report the spontaneous polarization, dynamical charges, and piezoelectric coefficients (and parts thereof) for the III-V nitrides. Also included are the same quantities calculated for ZnO and BeO (with the Zn  $3d$  states included in the valence). Our results for these materials are in good agreement with the available experimental values<sup>16</sup> for the effective charges ( $-2.10$  for ZnO,  $-1.85$  for BeO) and the  $e_{33}$  constant ( $\approx 1.0$ – $1.2$  C/m<sup>2</sup> for ZnO,  $\approx 0.09$  C/m<sup>2</sup> for BeO), showing some slight overall improvement over the Berry phase full-potential linearized augmented plane-wave calculations of Ref. 16, also listed in the table. To facilitate further comparison with other III-V and II-VI systems, in

TABLE II. Calculated spontaneous polarization (in units of C/m<sup>2</sup>), Born effective charges (in units of  $e$ ), and piezoelectric constants (in units of C/m<sup>2</sup>) for III-V wurtzite nitrides and II-VI wurtzite oxides. Theoretical results for ZnO and BeO are included.

	$P^{\text{eq}}$	$Z^*$	$du/d\epsilon_3$	$e_{33}$	$e_{31}$	$e_{33}^{(0)}$	$e_{31}^{(0)}$
AlN	-0.081	-2.70	-0.18	1.46	-0.60	-0.47	0.36
GaN	-0.029	-2.72	-0.16	0.73	-0.49	-0.84	0.45
InN	-0.032	-3.02	-0.20	0.97	-0.57	-0.88	0.45
ZnO	-0.057	-2.11	-0.21	0.89	-0.51	-0.66	0.38
BeO	-0.045	-1.85	-0.06	0.02	-0.02	-0.60	0.35
ZnO <sup>a</sup>	-0.05	-2.05	-0.25	1.21	-0.51	-0.58	0.37
BeO <sup>a</sup>	-0.05	-1.72	-0.09	0.50	—	-0.29	—

<sup>a</sup>Reference 16

TABLE III. Piezoelectric constants (in units of  $C/m^2$ ) of several zinc-blende compounds, calculated via linear response theory in Refs. 17 (III-V), 18 (CdTe), and 19 (other II-VI).

	$e_{33}$	$e_{31}$		$e_{33}$	$e_{31}$
CdTe	0.03	-0.01	AlAs	-0.01	0.01
ZnS	0.10	-0.05	GaAs	-0.12	0.06
ZnSe	0.04	-0.02	InAs	-0.03	0.01
AlP	0.04	-0.02	AlSb	-0.04	0.02
GaP	-0.07	0.03	GaSb	-0.12	0.06
InP	0.04	-0.02	InSb	-0.06	0.03

Table III we collect the piezoelectric constants for a number of III-V and II-VI compounds from Refs. 17, 18, and 19, calculated by density functional linear-response theory.<sup>20</sup>

To compare zinc-blende and wurtzite structures in Tables II and III, we transformed the zinc-blende piezoelectric tensor by rotating to a coordinate system with  $z$  along a (111) direction. Thus,  $e_{33} = 2e_{14}/\sqrt{3}$  and  $e_{31} = -e_{14}/\sqrt{3}$ , with  $e_{14}$  the piezoelectric constant of the zinc-blende material referred to the cubic axes. This direct comparison is meaningful since (1) the possible deviations from the ideal wurtzite structure do not influence significantly the polarization derivatives, and (2) it is known that in polytypical materials the values of the piezoelectric constants in the two competing structures agree to within a few percent (modulo the appropriate coordinate transformation).<sup>16</sup>

The III-V nitrides exhibit rather striking and unusual properties. Overall they resemble II-VI oxides, and ZnO in particular, and appear to be very different from conventional III-V semiconductors. Several features of the data in Tables II and III confirm this statement.

(1) The absolute value of the piezoelectric constants is up to about ten times larger than in conventional III-V and II-VI compounds. In particular, both the constants  $e_{33}$  and  $e_{31}$  of AlN are larger than those of ZnO, and are therefore the largest known so far among the tetrahedrally bonded semiconductors.

(2) The spontaneous polarization, i.e., the polarization at zero strain, is very large in the nitrides. That of AlN, in particular, is the largest of our set of semiconductors, and only about 3–5 times smaller than in typical ferroelectric perovskites.<sup>21</sup> Indeed, the piezoelectric constants themselves are only about three times smaller than in those ferroelectrics.<sup>22</sup>

(3) The piezoelectric constants have the same sign as in II-VI compounds, and opposite to III-V compounds. While in normal III-V compounds the clamped-ion term is larger in absolute value than the internal-strain ionic contribution, in the nitrides the latter prevails due to the larger  $Z^*$ . Compared to normal III-V compounds, this sign inversion is a qualitative difference of obvious practical relevance.<sup>23</sup>

(4) The nitrides follow qualitatively a well-defined III-V trend: the piezoelectric constants increase in magnitude as a function of the anion chemical identity as one moves upwards within period V, i.e., from Sb to N, because the ionic contribution tends to prevail over the electronic “clamped-ion” term as the anion becomes lighter (note that a similar trend is also obeyed by Zn II-VI compounds). The nitrides are an extreme case of this trend, and their piezoelectric re-

sponse is by far larger than that of all other III-V compounds (see Table III), and of opposite sign.

(5) The Born effective charge in the nitrides is within 10% of the nominal ionic charge, i.e., the ratio  $Z^*/Z_{\text{ion}}$  is about unity. The same ratio is at most 0.7 in III-V compounds. In this context, it is appropriate to mention that the effective charges have no rigorous relation to nominal ionicity (although they may be a reasonable qualitative measure of ionicity in many binary compounds, suggesting trends analogous to those given by, e.g., the charge asymmetry  $g$  parameter of Garcia and Cohen.<sup>24</sup>) In particular,  $Z^*$  needs not be smaller than or equal to the nominal ionic charge except in a purely ionic picture; conversely, whereas  $Z^* < Z_{\text{ion}}$  may be interpreted as “smaller ionicity” in some sense,  $Z^* > Z_{\text{ion}}$  is not meaningful in an ionic picture. Effective charges much larger than the nominal ionic charge are common in perovskites<sup>21</sup> and (to a lesser extent) even in alkaline-earth oxides.<sup>25</sup> This behavior is due to covalency and correlation contributions. We find (see Table II)  $Z^* > Z_{\text{ion}}$  for InN, but with a deviation of  $\sim 1\%$ , i.e., the effective charge is again very close to nominal ionicity.

Because of the sensitive dependence of the polarization on the structural parameters, there are some quantitative differences in spontaneous polarization for the three nitrides. For instance, the increasing nonideality of the structure going from GaN to InN to AlN ( $u$  gets longer and  $c/a$  gets shorter), corresponds to an increase of the spontaneous polarization. To check this, we calculated the spontaneous polarization of AlN using the ideal structural parameters  $u_{\text{id}} = 0.375$  and  $c_{\text{id}} = \sqrt{8/3} a$ , and obtained a spontaneous polarization equal to  $-0.033 C/m^2$ , very similar to GaN and InN, and a factor of 2.5 smaller than the spontaneous polarization in the actual equilibrium structure. The piezoelectric constants (i.e., polarization derivatives) are instead expected to vary mildly.

Additional III-V-like trends to be observed are that in III-V compounds the effective charge has similar values for AlAs and GaAs, but a rather larger value for InAs.<sup>17</sup> This trend is clearly obeyed by the nitrides as well, indeed in a more clearcut way than in most other III-V compounds. Another interesting trend is the connection between the chemical nature of the cation and the piezoelectric constant. The piezoelectric constant for Al and In III-V compounds was found to be<sup>17</sup> consistently larger than the corresponding value for Ga compounds. This is also true of the nitrides, where the deviations are again larger. Since our calculations include the semicore  $d$  states in the valence for both In and Ga, this trend is not an artifact of the neglect of semicore states as hypothesized in Ref. 17.

While it is known that treating the semicore  $d$  states as valence is necessary to obtain good structural properties for GaN and InN,<sup>15,26</sup> neither the spontaneous polarization nor the effective charges are influenced strongly. Using a Ga pseudopotential in which the  $3d$  states had been frozen in the core, we recalculated the spontaneous polarization and effective charges of GaN using the structural parameters given in Table I: the results are  $P^{\text{eq}} = -0.031 C/m^2$  and  $Z^* = 2.73$ , showing that the direct influence of the  $3d$  states on the final polarization value is minor. This was to be expected since the  $d$  bands in GaN are quite flat over most of the Brillouin

zone, so that the overlap matrix [Eq. (9)] is nearly the identity for those states, and its contribution to the Berry phase is small.

Experimental data to compare our predictions with are very scarce. No data are available for the spontaneous polarization, a quite elusive quantity in this respect; our calculated values will be a useful input for device modeling.<sup>2,3</sup> Also in the case of the piezoelectric constants, our predicted data should be of use to future experimental work. The only experimental data available<sup>27</sup> to our knowledge are for AlN,  $e_{33}^{\text{expt}} = 1.55 \text{ C/m}^2$ , and  $e_{31}^{\text{expt}} = -0.58 \text{ C/m}^2$ . Our predicted values of 1.46 and  $-0.60 \text{ C/m}^2$  agree with experiment to within  $\sim 5\%$ .

In summary, we calculated the spontaneous polarization, dynamical charges, and piezoelectric constants of III-V nitrides by means of the Berry phase approach. The results

show that III-V nitrides resemble II-VI compounds in terms of the sign of the spontaneous polarization and of the piezoelectric constants; the latter constants are much larger in absolute value than those of all III-V and II-VI compounds, except for ZnO. The values of spontaneous polarization are comparable to or larger than those of II-VI compounds; AlN has the largest value reported so far for any binary compound, and only a factor of 3–4 smaller than in typical perovskites. Some remnant of normal III-V-like behavior survives, embodied in the trends of the piezoelectric constants and effective charges when the chemical identity of the anion or cation changes.

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- <sup>1</sup>F. A. Ponce and D. P. Bour, *Nature (London)* **386**, 351 (1997).
- <sup>2</sup>A. Bykhovski, B. Gelmont, M. Shur, and A. Khan, *J. Appl. Phys.* **77**, 1616 (1995).
- <sup>3</sup>F. Bernardini, V. Fiorentini, and D. Vanderbilt, in *III-V Nitrides*, edited by F. A. Ponce, T. D. Moustakas, I. Asaki, and B. A. Monemar, MRS Symposia Proceedings, No. 449 (Materials Research Society, Pittsburgh, 1997), p. 923.
- <sup>4</sup>R. D. King-Smith and D. Vanderbilt, *Phys. Rev. B* **47**, 1651 (1993).
- <sup>5</sup>R. Resta, *Rev. Mod. Phys.* **66**, 899 (1994).
- <sup>6</sup>J. F. Nye, *Physical Properties of Crystals* (Oxford University Press, Oxford, 1985).
- <sup>7</sup>See M. Posternak, A. Baldereschi, A. Catellani, and R. Resta, *Phys. Rev. Lett.* **64**, 1777 (1990).
- <sup>8</sup>D. Vanderbilt and R. D. King-Smith, *Phys. Rev. B* **48**, 4442 (1993).
- <sup>9</sup>R. Dreizler and E. K. U. Gross, *Density-Functional Theory* (Springer, New York, 1990).
- <sup>10</sup>D. M. Ceperley and B. J. Alder, *Phys. Rev. Lett.* **45**, 556 (1980).
- <sup>11</sup>J. Perdew and A. Zunger, *Phys. Rev. B* **23**, 5048 (1981).
- <sup>12</sup>D. Vanderbilt, *Phys. Rev. B* **41**, 7892 (1990).
- <sup>13</sup>D. J. Chadi and M. L. Cohen, *Phys. Rev. B* **8**, 5747 (1973).
- <sup>14</sup>H. J. Monkhorst and J. D. Pack, *Phys. Rev. B* **13**, 5188 (1976).
- <sup>15</sup>A. Satta, V. Fiorentini, A. Bosin, F. Meloni, and D. Vanderbilt, in *Gallium Nitride and Related Compounds*, edited by R. D. Dupuis, J. A. Edmond, F. Ponce, and S. Nakamura, MRS Symposia Proceedings, No. 395 (Materials Research Society, Pittsburgh, 1996), p. 515.
- <sup>16</sup>A. Dal Corso, M. Posternak, R. Resta, and A. Baldereschi, *Phys. Rev. B* **50**, 10 715 (1994).
- <sup>17</sup>S. de Gironcoli, S. Baroni, and R. Resta, *Phys. Rev. Lett.* **62**, 2853 (1989).
- <sup>18</sup>A. Dal Corso, R. Resta, and S. Baroni, *Phys. Rev. B* **47**, 16252 (1993).
- <sup>19</sup>S. de Gironcoli, S. Baroni, and R. Resta, *Ferroelectrics* **111**, 19 (1990).
- <sup>20</sup>S. Baroni, P. Giannozzi, and A. Testa, *Phys. Rev. Lett.* **58**, 1861 (1987).
- <sup>21</sup>W. Zhong, R. D. King-Smith, and D. Vanderbilt, *Phys. Rev. Lett.* **72**, 3618 (1994); R. Resta, M. Posternak, and A. Baldereschi, *ibid.* **70**, 1010 (1993).
- <sup>22</sup>K. M. Rabe and U. W. Waghmare, *Phys. Rev. B* **55**, 6161 (1997).
- <sup>23</sup>AlP and InP indeed have (Ref. 17) a positive, but very small,  $e_{14}$ , which is again related to larger-than-average  $Z^*$ .
- <sup>24</sup>A. Garcia and M. L. Cohen, *Phys. Rev. B* **47**, 4215 (1993); **47**, 4221 (1993).
- <sup>25</sup>M. Posternak, R. Resta, H. Krakauer, and A. Baldereschi, *Phys. Rev. B* **55**, 15 983 (1997).
- <sup>26</sup>See, e.g., V. Fiorentini, M. Methfessel, and M. Scheffler, *Phys. Rev. B* **47**, 13 353 (1993); V. Fiorentini, A. Satta, D. Vanderbilt, S. Massidda, and F. Meloni, in *The Physics of Semiconductors*, edited by D. J. Lockwood (World Scientific, Singapore, 1995), p. 137.
- <sup>27</sup>J. G. Gualtieri, J. A. Kosinski, and A. Ballato, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **UFFC-41**, 53 (1994).