Piezoelectric fields in strained heterostructures and superlattices

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(Received 23 January 1992)

The piezoelectric response of strained heterostructures and superlattices is examined. A general crystallographic direction of growth is assumed for a subcritical cubic piezoelectric layer. Piezoelectric fields appear only along (or opposite to) the growth axis, provided all three direction cosines of the latter, relative to the cubic axes, are nonzero. The formulation is based on elasticity theory, and the results are compared with other approaches in the literature. Standard and less standard directions of growth are worked out explicitly.

Strained piezoelectric (PZ) layers in heterostructures (HS) and superlattices (SL) are capable of exhibiting PZ fields.[1] A number of theoretical and experimental works have appeared in recent years which deal with this problem (see, for instance, Refs. 2–4 and 5–11, respectively). Depending on the magnitude of the strains and the PZ constants, the fields may reach significant values, exceeding 10^5 V/cm, provided the concentration of mobile carriers is low enough to keep the fields unscreened.[3,7,9,10] In the presence of PZ fields the overall behavior of the layered system is modified. Changes in the electronic band structure are among the most important consequences, as demonstrated by spectroscopic and nonlinear-optical techniques.[5–10] Furthermore, the PZ fields are expected to affect the degeneracy and frequency of the long-wavelength optical phonons, by analogy to similar effects induced by misfit strains.[12] At present, the PZ fields are determined for a general direction of pseudomorphic growth, assuming cubic PZ materials. The thickness of the structure is less than the critical value. The strain tensor can be treated in the framework of elasticity theory and be considered uniform throughout the volume of the dislocation-free layers. A detailed treatment of the elastic strains and stresses has been presented in Refs. 13–15; only the necessary results will be stated here.

We designate the cubic axes by $\mathbf{x}_1\|[100]$, $\mathbf{x}_2\|[010]$, $\mathbf{x}_3\|[001]$, and the HS or SL axes by $\mathbf{x}'_1\|[l_1m_1n_1]$, $\mathbf{x}'_2\|[l_2m_2n_2]$ (in-plane axes), and $\mathbf{x}'_3\|[l_3m_3n_3]\equiv\mathbf{N}$ (direction of growth), $l_\lambda, m_\lambda, n_\lambda$ being their direction cosines relative to $x_1x_2x_3$. Primed (unprimed) tensor components refer to the primed (unprimed) system, and all Latin (Greek) indices run from 1 (1) to 6 (3). By a_ν and h_ν we designate the bulk lattice constants and the thicknesses of the two constituents, i.e., $\nu=e$ (s) for epilayer (substrate) in HS's $(h_e \ll h_s)$, and $\nu=1$ (2) for layer 1 (layer 2) in SL's. The lattice fractional misfit f is equal to $(a_s/a_e)-1$ in HS's, and $(a_2/a_1)-1$ in SL's. Wherever obvious, the index ν is omitted for simplicity. The reduced tetragonal distortion of either layer is computed directly, using l_3, m_3, n_3 and the corresponding elastic stiffnesses C_{ij},[13]

$$\Delta\tilde{\epsilon}=\Delta\epsilon/\epsilon^{\|}=(\epsilon^{\|}-\epsilon^{\perp})/\epsilon^{\|} =\frac{3B}{\Delta}[C_{44}^2+CC_{44}(1-T_{33})+3C^2(l_3m_3n_3)^2]\ , \tag{1}$$

where we have set $\epsilon'_3\equiv\epsilon_\perp$ (strain component normal to the plane) and $\epsilon'_1=\epsilon'_2\equiv\epsilon^{\|}$ (in-plane isotropic strain component). $B=(C_{11}+2C_{12})/3$ is the bulk modulus, and

$$C=C_{11}-C_{12}-2C_{44}\ , \tag{2a}$$

$$T_{33}=l_3^4+m_3^4+n_3^4\ , \tag{2b}$$

$$\Delta=C_{11}C_{44}^2+(CC_{44}/2)(C_{11}+C_{12})(1-T_{33}) +C^2(C_{11}+2C_{12}+C_{44})(l_3m_3n_3)^2\ . \tag{2c}$$

Within a numerical factor, the common in-plane lattice constant of both constituents in a free-standing, subcritical (coherently grown) SL is[14]

$$a^{\|}=\frac{h_1G_1a_1+h_2G_2a_2}{h_1G_1+h_2G_2}\ , \tag{3}$$

with $a_{\|}=a_s$ for a subcritical (coherently grown) HS. G_ν is an appropriate shear modulus for each material given by[14]

$$G_\nu=3B_\nu(3-\Delta\tilde{\epsilon}_\nu)\ . \tag{4}$$

The in-plane and normal-to-the-plane strain components of either layer are, in this case,

$$\epsilon^{\|}=(a^{\|}/a)-1\ , \tag{5a}$$

$$\epsilon^{\perp}=(1-\Delta\tilde{\epsilon})\epsilon^{\|}\ . \tag{5b}$$

The remaining nonzero strain components ϵ'_4 and ϵ'_5 are proportional to the corresponding $\epsilon^{\|}$ and given by[13]

$$2\epsilon'_{23}=\epsilon'_4=\frac{3BC}{\Delta}[C_{44}T_{34}+C(T_{31}T_{34}-T_{35}T_{36})]\epsilon^{\|} \equiv\tilde{\epsilon}'_4\epsilon^{\|}\ , \tag{6a}$$

$$2\epsilon'_{31}=\epsilon'_5=\frac{3BC}{\Delta}[C_{44}T_{35}+C(T_{32}T_{35}-T_{36}T_{34})]\epsilon^{\parallel}$$
$$\equiv\tilde{\epsilon}'_5\epsilon^{\parallel}\ , \quad (6b)$$

$$2\epsilon'_{12}=\epsilon'_6=0\ , \quad (6c)$$

where the fully symmetric fourth-rank tensor $T_{\lambda\mu\kappa\rho}$ is defined by

$$T_{ij}=T_{ji}=T_{\lambda\mu\kappa\rho}=l_\lambda l_\mu l_\kappa l_\rho+m_\lambda m_\mu m_\kappa m_\rho+n_\lambda n_\mu n_\kappa n_\rho\ . \quad (7)$$

In the primed system of axes, the component array of the strain tensor is $\epsilon'=(\epsilon^{\parallel},\epsilon^{\parallel},\epsilon^{\perp},\epsilon'_4,\epsilon'_5,0)$. In the unprimed system the strain tensor becomes[15]

$$\underline{\epsilon}=\begin{bmatrix}\epsilon_1\\ \epsilon_2\\ \epsilon_3\\ \epsilon_4\\ \epsilon_5\\ \epsilon_6\end{bmatrix}=\epsilon^{\parallel}\begin{bmatrix}1\\1\\1\\0\\0\\0\end{bmatrix}-\frac{3B\epsilon^{\parallel}}{\Delta}\begin{bmatrix}l_3^2(C_{44}+Cm_3^2)(C_{44}+Cn_3^2)\\ m_3^2(C_{44}+Cn_3^2)(C_{44}+Cl_3^2)\\ n_3^2(C_{44}+Cl_3^2)(C_{44}+Cm_3^2)\\ m_3n_3(C_{44}+Cl_3^2)(C_{11}-C_{12}-Cl_3^2)\\ n_3l_3(C_{44}+Cm_3^2)(C_{11}-C_{12}-Cm_3^2)\\ l_3m_3(C_{44}+Cn_3^2)(C_{11}-C_{12}-Cn_3^2)\end{bmatrix}. \quad (8)$$

The reason for writing the strain tensor in both systems is that the computation in a particular situation is easier if one or the other form is used. Having laid the necessary background information, we proceed to the main objective of this work, i.e., the general formulation of the PZ effect. We start with the phenomenological definition of the PZ phenomenon in terms of the strain-induced polarization,[16]

$$P_\lambda=e_{\lambda,\mu\kappa}\epsilon_{\mu\kappa}=e_{\lambda,i}\epsilon_i\ . \quad (9)$$

The PZ constant $e_{\lambda,\mu\kappa}=e_{\lambda,i}$ $(i=1-6)$ is a third-rank tensor with only one independent component e in the cubic PZ classes T and T_d. In $x_1x_2x_3$ it can be written as

$$e_{\lambda,\mu\kappa}=e|\epsilon_{\lambda\mu\kappa}|\ , \quad (10)$$

where $\epsilon_{\lambda\mu\kappa}$ is the antisymmetric unit tensor (i.e., $\epsilon_{123}=-\epsilon_{213}=1$, $\epsilon_{113}=\epsilon_{122}=\epsilon_{333}=0$, etc.). In $x'_1x'_2x'_3$ it can be shown that the PZ tensor takes the form

$$e'_{\lambda,\mu\kappa}=eT_{\lambda\mu\kappa}\ , \quad (11)$$

where

$$T_{\lambda\mu\kappa}=(l_\lambda m_\mu+l_\mu m_\lambda)n_\kappa+\text{c.p.}=2(l_\lambda m_\mu n_\kappa+\text{c.p.})-\epsilon_{\lambda\mu\kappa} \quad (12)$$

is a fully symmetric third-rank tensor analogous to $T_{\lambda\mu\kappa\rho}$ of Eq. (7), and c.p. means cyclic permutation over l,m,n.

From Eqs. (8)–(10) we find

$$\mathbf{P}=\begin{bmatrix}P_1\\P_2\\P_3\end{bmatrix}=e\begin{bmatrix}\epsilon_4\\ \epsilon_5\\ \epsilon_6\end{bmatrix}=-\frac{3Be\epsilon^{\parallel}}{\Delta}\begin{bmatrix}m_3n_3(C_{44}+Cl_3^2)(C_{11}-C_{12}-Cl_3^2)\\ n_3l_3(C_{44}+Cm_3^2)(C_{11}-C_{12}-Cm_3^2)\\ l_3m_3(C_{44}+Cn_3^2)(C_{11}-C_{12}-Cn_3^2)\end{bmatrix}. \quad (13)$$

In $x'_1x'_2x'_3$, on the other hand, the primed components of the polarization are easily reached by writing Eq. (9) directly in the primed system. The results are

$$P'_\lambda=-e\epsilon^{\parallel}(T_{\lambda33}\Delta\tilde{\epsilon}-T_{\lambda23}\tilde{\epsilon}'_4-T_{\lambda13}\tilde{\epsilon}'_5)\ . \quad (14)$$

Alternatively, $\mathbf{P}'$ can be obtained from Eq. (13) by rotation; after some computation, the following compact form for P'_λ is reached in this way:

$$P'_{\substack{1\\2}}=\left|-\frac{3Be\epsilon^{\parallel}}{2\Delta}\right|[(C_{11}-C_{12})C_{44}T_{\substack{133\\233}}-2C^2(l_3m_3n_3)T_{\substack{35\\34}}]\ , \quad (15a)$$

$$P'_3=\left|-\frac{3Be\epsilon^{\parallel}}{\Delta}\right|[(C_{11}-C_{12})^2+2C_{44}^2-C^2T_{33}]\times(l_3m_3n_3)\ . \quad (15b)$$

Then, the in-plane component of $\mathbf{P}$ $(=\mathbf{P}')$ is written as

$$\mathbf{P}_{\parallel}=\mathbf{P}'_{\parallel}=\hat{x}'_1P'_1+\hat{x}'_2P'_2=\mathbf{P}-\mathbf{N}P'_3\ . \quad (16)$$

The magnitude $P_{\parallel}=P'_{\parallel}$ is best obtained from Eqs. (15a),

$$P'_{\parallel}=(P'^2_1+P'^2_2)^{1/2}=\left|-\frac{3Be\epsilon^{\parallel}}{2\Delta}\right|\{2(C_{11}-C_{12})^2C_{44}^2(1-T_{33})-4(l_3m_3n_3)^2[9(C_{11}-C_{12})^2C_{44}^2-C^4(l_3^6+\text{c.p.}-T_{33}^2)+2(C_{11}-C_{12})C_{44}C^2(1-3T_{33})]\}^{1/2}\ , \quad (17)$$

where higher-order identity expressions for the direction cosines have been used.[13] As expected, the components P_λ, P'_3, and $P_\parallel = P'_\parallel$ depend only on l_3, m_3, n_3.

Of the two components of $\mathbf{P}'$, only P'_3 gives rise to a PZ field E'_3, and only $P'_\parallel$ gives rise to a PZ displacement $D'_\parallel$. The electrostatic equations applied to each layer yield[2]

$$E'_3 = -\frac{P'_3}{\epsilon_0 \kappa_s}, \quad E'_\parallel = 0 , \tag{18a}$$

$$D'_3 = 0, \quad \mathbf{D}'_\parallel = \frac{\mathbf{P}'_\parallel}{\epsilon_0} , \tag{18b}$$

where ϵ_0 is the vacuum permittivity and κ_s is the static (relative) dielectric constant of the layer. It has been assumed that no external charges are present and the dielectric constant is uniform throughout the layer. The same applies to the polarization components which exhibit a dependence on x'_3 only at the interfaces, i.e., $\mathbf{P}' = \mathbf{P}'(x'_3)$; this implies $\nabla' \cdot \mathbf{P}'_3 \neq 0$, $\nabla' \times \mathbf{P}'_\parallel \neq 0$, $\nabla' \times \mathbf{P}'_3 = \nabla' \cdot \mathbf{P}'_\parallel = 0$.

Equations (18) combined with Eqs. (15) and (17) provide direct expressions for the PZ field and displacement for an arbitrary direction of growth. The following general rule becomes clear now: PZ fields are induced only for those directions of growth for which all three direction cosines l_3, m_3, and n_3 are nonzero (equivalently, the direction of growth $\mathbf{N}$ must not lie on the mirror planes specified by any two of the cubic axes x_1, x_2, and x_3). Following the terminology often encountered in the literature, we define such directions of growth as *polar axes.* Thus PZ fields are expected to be present for $\mathbf{N}$ along [111], [11$\bar{2}$], [211], [113], etc., but not for [100], [110], [120], etc. The PZ displacement, on the other hand, is zero only for $\mathbf{N}$ along the high-symmetry directions [001] and [111] or their equivalent directions.

We give below results for some standard and less standard directions $\mathbf{N}$:

(i) $\mathbf{N} = [001]$,

$$P'_1 = P'_2 = P'_3 = 0, \quad E'_3 = 0, \quad D'_\parallel = 0 . \tag{19}$$

(ii) $\mathbf{N} = [111]/\sqrt{3}$,

$$P'_1 = P'_2 = 0, \quad D'_\parallel = 0 ,$$
$$P'_3 = -2e\Delta\epsilon/\sqrt{3}, \quad E'_3 = -P'_3/\epsilon_0\kappa_s , \tag{20a}$$

where[13]

$$\Delta\epsilon = \frac{9B\epsilon^\parallel}{C_{11} + 2C_{12} + 4C_{44}} . \tag{20b}$$

(iii) $\mathbf{N} = [110]/\sqrt{2}$,

$$P'_2 = P'_3 = 0, \quad E'_3 = 0, \quad P'_1 = -e\Delta\epsilon, \quad D'_\parallel = P'_1/\epsilon_0 , \tag{21a}$$

where[13] $\mathbf{x}'_1 \parallel [001]$ and

$$\Delta\epsilon = \frac{6B\epsilon^\parallel}{C_{11} + C_{12} + 2C_{44}} . \tag{21b}$$

(iv) $\mathbf{N} = [120]/\sqrt{5}$,

$$P'_2 = P'_3 = 0, \quad E'_3 = 0 , \tag{22a}$$

$$P'_1 = -\frac{10e(C_{11} - C_{12})\Delta\epsilon}{8C_{11} - 8C_{12} + 9C_{44}}, \quad D'_\parallel = P'_1/\epsilon_0 , \tag{22b}$$

where[15] $\mathbf{x}'_1[001]$ and

$$\Delta\epsilon = \frac{3B\epsilon^\parallel(8C_{11} - 8C_{12} + 9C_{44})}{4(C_{11}^2 - C_{12}^2) + C_{44}(17C_{11} - 8C_{12})} . \tag{22c}$$

(v) $\mathbf{N} = [11\bar{2}]/\sqrt{6}$,

$$P'_1 = 0, \quad P'_2 = e\Delta\epsilon/\sqrt{3} , \tag{23a}$$

$$P'_3 = \frac{3e(C_{11} - C_{12})\Delta\epsilon}{\sqrt{6}(C_{11} - C_{12} + C_{44})} , \tag{23b}$$

$$E'_3 = -P'_3/\epsilon_0\kappa_s, \quad D'_\parallel = P'_2/\epsilon_0 , \tag{23c}$$

where[15] $\mathbf{x}'_2 \parallel [111]/\sqrt{3}$ and

$$\Delta\epsilon = \frac{18B\epsilon^\parallel(C_{11} - C_{12} + C_{44})}{3(C_{11} - C_{12} + C_{44})(C_{11} + C_{12} + 2C_{44}) - C^2} . \tag{23d}$$

Apart from an opposite sign for P'_2 and P'_3, the same results hold for grown along $[211]/\sqrt{6}$, with $\mathbf{x}'_2 \parallel [\bar{1}11]/\sqrt{3}$. PZ studies for [211]-grown multiple quantum wells were reported recently.[10]

(vi) $\mathbf{N} = [113]/\sqrt{11}$,

$$P'_1 = \left[-\frac{24\sqrt{2}Be\epsilon^\parallel}{11\sqrt{11}\Delta} \right] [(C_{11} - C_{12})C_{44} + 9C^2/11^2] ,$$
$$P'_2 = 0 , \tag{24a}$$

$$P'_3 = \left[-\frac{9Be\epsilon^\parallel}{11\sqrt{11}\Delta} \right] [(C_{11} - C_{12})^2 + 2C_{44}^2 - 83C^2/11^2] , \tag{24b}$$

$$E'_3 = -P'_3/\epsilon_0\kappa_s, \quad D'_\parallel = P'_1/\epsilon_0 , \tag{24c}$$

where[15] $\mathbf{x}'_1 \parallel [33\bar{2}]/\sqrt{22}$ and

$$\Delta = C_{11}C_{44}^2 + 19CC_{44}(C_{11} + C_{12})/11^2 + 9C^2(C_{11} + 2C_{12} + C_{44})/11^3 . \tag{24d}$$

To summarize, explicit expressions have been derived for the PZ fields and displacements in strained HS's and SL's grown along an arbitrary direction. The layers have been treated as homogeneous and dislocation-free, with no interfacial disorder; we have not considered corrections due to internal displacements [internal strain parameter ζ (Ref. 17)]. In HS's, all present results concern the epilayer, with $\epsilon_e^\parallel = f$ (no effect on the substrate). In SL's, the results concern each layer ν independently, with $\epsilon_\nu^\parallel$ given by Eq. (5a). Often, on the other hand, overcritical HS or SL systems are completely relaxed at the growth temperature T_g, because of misfit dislocations. In such cases, the in-plane strain $\epsilon_\nu^\parallel$ at room temperature T_0 is no longer determined from Eq. (5b), which is valid only for coherently grown subcritical systems. Instead, it is equal to $-(\beta-\beta_\nu)(T_g-T_0)\equiv-\delta\beta\Delta T$, where β_ν is the thermal expansion coefficient for layer ν, and β is a thermal expansion coefficient characterizing the entire system; its value is given by an equation similar to Eq. (3), with $a^\parallel$ and a_ν replaced by β and β_ν, respectively[18] (more precisely, β and β_ν stand for their mean values in the temperature range $\Delta T=T_g-T_0$).

The sign of P'_3 (and E'_3) for a strained layer (i.e., whether it points along **N** or $-\mathbf{N}$), depends on four factors, (i) the polarity (cation A or anion B) of the layer's face nearest to the free surface, (ii) the sign of the PZ constant, (iii) the sign of the in-plane strain, and (iv) the direction of growth, through the sign of the product $(l_3m_3n_3)$, according to Eq. (15b). [It is assumed that the bracket of Eq. (15b) is positive, as is the case for most materials under consideration.] By definition, a positive component P'_3 points from the layer's B face towards its A face. There is a fifth factor, the hydrostatic pressure which, depending on the choice of constituents, may also affect the sign of the piezoelectric field.[18]

It should be emphasized that whereas E'_3 is always in the direction of $\pm\mathbf{N}$, the same is not true for the polarization $\mathbf{P}'$. In fact, according to Eq. (17), in-plane components of $\mathbf{P}'$ are always expected to exist, with the exception of growth along [100] and [111], or equivalent. The in-plane components of $\mathbf{P}'$ may give rise to in-plane PZ fields in low dimensionality systems, i.e., quantum wires and quantum dots. To the best of the author's knowledge, no such phenomena have been reported thus far.

The present approach to the PZ fields in HS's and SL's has been treated in the framework of elasticity theory, whereby the medium is looked upon as an elastic continuum, as far as the boundary conditions for the strain and stress fields are concerned. In this sense, the results differ from those of Ref. 2 where the boundary conditions have been chosen so that the interface atoms have the correct local bonding on stepped surfaces. Such surfaces occur for low-symmetry directions of growth. On the contrary, the results of Ref. 2 coincide with the present ones for the high-symmetry directions [111] and [110].[19] To quantify the differences between the two approaches, we have calculated E'_3 and $P'_\parallel$ for a single layer of GaAs grown along [113]. We have taken $C_{11}=119$, $C_{12}=54$, $C_{44}=59$ in GPa, $e=-0.16$ C/m^2, $\kappa_s=12.9$, and $\epsilon^\parallel=0.01$. Using Eqs. (24), we find $E'_3=-7.3\times10^6$ V/m and $P'_\parallel=1.4\times10^{-3}$ C/m^2. On the other hand, the results of Ref. 2 lead to the same signs and higher values by 60% and 20%, respectively.[19] It would be interesting to compare the numerical results of both approaches against accurate experimental data.[11]

Finally, since the strength of PZ fields may reach substantial values, because of large values of $\epsilon^\parallel$ and/or e, it is necessary in such cases to consider terms in Eq. (9) which are nonlinear in $\epsilon_{\mu\kappa}$. Such nonlinearities can be best incorporated into the present analysis by including electrostrictive strains, i.e., strains which are quadratic in the components of the PZ fields.[16] Nonlinearities in the PZ fields have already been observed.[6]

This work was supported in part by the General Secretariat for Research and Technology, Greece. Useful communications with Dr. C. Mailhiot are acknowledged with appreciation.

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