Supporting Information

Tunable Tunneling Electro-resistance in Ferroelectric Tunnel Junctions by Mechanical Loads

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Analysis of temperature effect on polarization

Based on the expression of Landau-Devonshire Helmholtz free energy of a bulk ferroelectric, the thermodynamic potential of strained ferroelectric capacitors under the mean field theory is given by Pertsev[1], Tagantsev[2], and Zheng[3]. According to the work of Pertsev et al.,[1] the polarization of symmetrical nanaoscale strained junction in the P^4 approximation can be written as

$$P_s^2 = \frac{1}{2\left[a_{11}^{\eta} - q_{11}^2/\left(2c_{11}\right)\right]}\left[\frac{T_C - T}{2\varepsilon_0 C_0} - 2\left(\frac{q_{11}c_{12}}{c_{11}} - q_{12}\right)\eta_m - \frac{1}{2\left(\varepsilon_0 + c_i h\right)}\right] \quad \text{(S1)}$$

where a_{11}^{η} is the dielectric stiffness coefficients at constant misfit strain η_m, q_{11} is the electrostrictive constants, c_{11} and c_{12} are the film elastic stiffnesses, c_i is the total interfacial capacitance, and T_C and C_0 are the Curie-Weiss temperature and constant of the bulk ferroelectrics.

For a ferroelectric junction with specific compressive strain η_m and thickness *h*, we can get the following relationship

$$P_s^2 \propto C^*\left(T_C^* - T\right) \quad \text{(S2)}$$

where C^* is constant for a film with given compressive strain and thickness, and $T_C^* = \frac{T_C}{2\varepsilon_0 C_0} - 4\left(\frac{q_{11}c_{12}}{c_{11}} - q_{12}\right)\eta_m \varepsilon_0 C_0 - \frac{\varepsilon_0 C_0}{\varepsilon_0 + c_i h}$ is the Curie temperature of the strained ferroelectric film in FTJ.

From energy rescaling method, the Curie temperatures of the m=2, 3, 4 unit cells FTJ under -2.5% strain are estimated to be 470K, 990K, and 1200K, respectively, and their respective zero temperature polarizations obtained from DFT calculations are $90\,\mu C/cm^2$, $92\,\mu C/cm^2$, and $95\,\mu C/cm^2$. Using these values and relationship

(S2), the room temperature (300K) polarizations of the above strained junctions can be speculated to be 54 $\mu C / cm^2$, 76 $\mu C / cm^2$, and 82 $\mu C / cm^2$, respectively.

Example of the GPR calculation in m=2 unit cells FTJ

As shown in Figure 3(a) in the paper, the electron conductance of the m=2 unit cells FTJ is $G_\eta = 2.06\times10^{-3}\ e^2/h$ under a compressive strain of -3.5%, and the relative value of the unstrained FTJ is $G = 4.8\times10^{-2}\ e^2/h$. Using Eq (4) GPR=$\frac{R_\eta - R_0}{R_0}$, and note that the resistance is related to the conductance by relationship $R=\frac{1}{G}$, the calculated GPR ratio under a compressive strain of -3.5% is 2230%.

Influence of defects on the electronic transport properties of FTJ

After the introduction of Oxygen vacancies (V_O) or Nitrogen doping (N_D) defects, the ferroelectric crystal structures are deformed by the defect induced electric field. It is noted that the concentration of the defects is 50% in the TiO terminal interface. Due to the different charge state in the V_O^{2+} and N_D^{3-} defects, the directions of the defect induced electric field are different in these defects containing FTJ. As a result, the polarizations produced by V_O^{2+} and N_D^{3-} defects field are antiparallel in the unstrained FTJ, as shown in Figure S1.

The calculated resistances of the above V_O^{2+} and N_D^{3-} defects containing FTJ under compressive strains of -2.5% and 0% are given in Table S1. It is clear that the deleterious influence of V_O^{2+} defects on the GPR ratio is smaller than those have N_D^{3-}

defect. However, even with the appearance of defects in these FTJ, the GPR ration is still very large, indicating promising future in realistic applications.

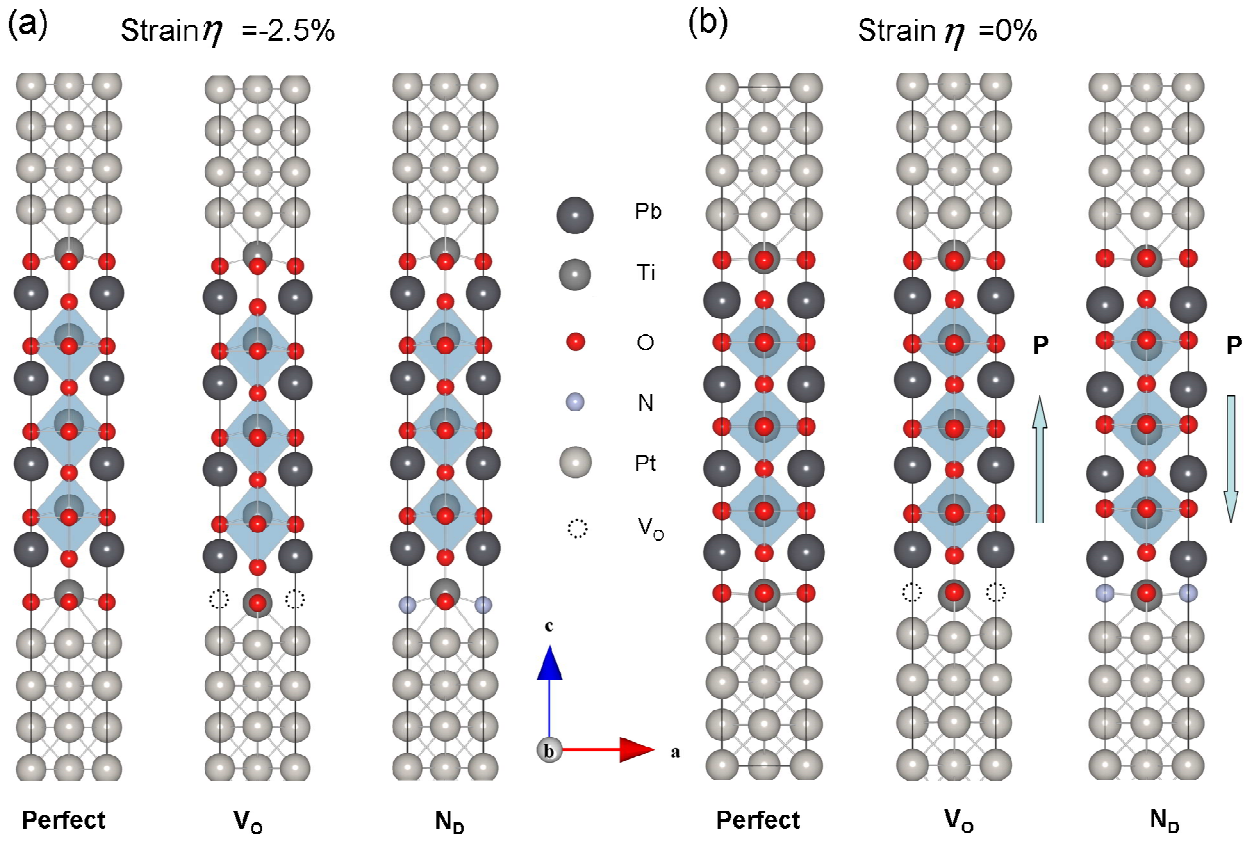

Figure S1. The relaxed Atomic structures of the Pt/PTO/Pt strained ferroelectric tunneling junctions (FTJ). Various perfect and defect structures are presented in (a) the FTJ with a compressive strain of -2.5%, and (b) the unstrained FTJ.

Effects of strain on the bandgap and Schottky barrier height of $Pt/PbTiO_3/Pt$ FTJ

Since the transport properties are dominated by the electronic structure of the FTJ, it would be interesting to see how the applied strain influence the band gap and the Sschottky barrier for electrons, *i.e.*, the Fermi energy positions with respect to the

minimum of the conduction band (MCB). It is clear from Figure S2 that band gap of the central $PbTiO_3$ unit cell becomes larger as the applied strain increase, and a similar trend is obtained for the Schottky barrier. As it is well known that DFT calculations always underestimate the band gap of insulator, the typical LDA values (1.6-1.8eV) are smaller than the experimental value of 3.4eV. By applying a Hubbard-U correction to Ti *3d* states, the band gaps of the $PbTiO_3$ are open up. From the more reasonable LDA+U values, it is found that the strain dependence of band gap and Schottky barrier has the same trend as those obtained from LDA calculations. Furthermore, the Schottky barrier height in the ferroelectric phase ($\eta < -1.1\%$ for m=4 FTJ) increase more dramatically than those in paraelectric phase.

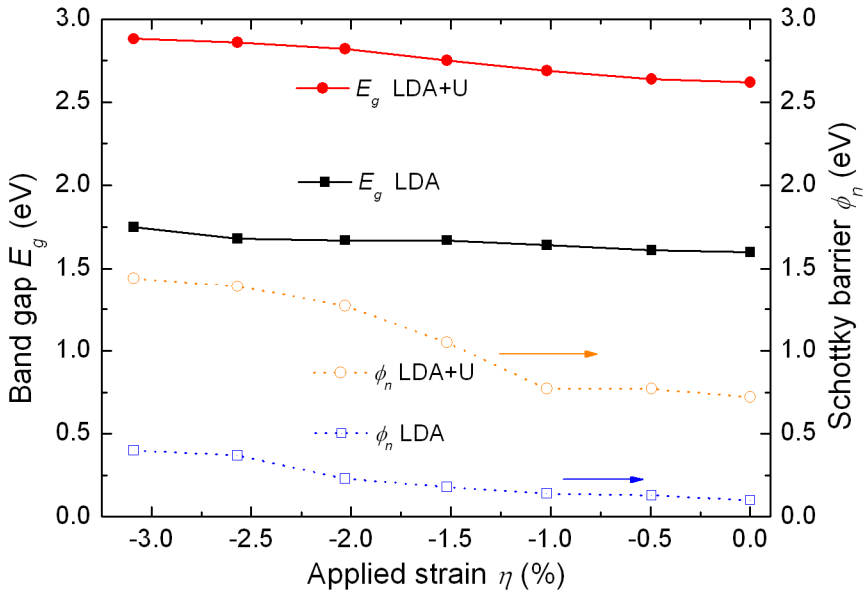

Figure S2. Band gap (E_g) of the central $PbTiO_3$ layer in the m = 4 unit cell FTJ as a function of different applied strain η. Their corresponding Schottky barrier height (ϕ_n) are also given. The LDA+U (where U=6eV) method is used to get the reasonable band gap and Schottky barrier height.

TABLE S1. Calculated conductance (G) for m=4 unit cells strained FTJ, the GPR ratio are obtained under a compressive strain of -2.5%.

FTJ Structure / Strain / G	Perfect FTJ	V_O FTJ	N_D FTJ
Unstrained	7.73×10^{-3}	3.43×10^{-3}	1.73×10^{-3}
With -2.5% compressive strain	2.84×10^{-4}	1.42×10^{-4}	1.43×10^{-4}
GPR ratio	2620%	2310%	1100%

The unit of conductance is e^2/h.

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