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Converse-piezoelectric effect on current-voltage characteristics of symmetric ferroelectric tunnel junctions

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Current-voltage characteristics of symmetric ferroelectric tunnel junction were studied in an ultrathin ferroelectric barrier with the consideration of the thickness and strain dependent converse-piezoelectric effect. With proper boundary conditions, large piezoelectric strain can be achieved in the ferroelectric barrier when its thickness is in the vicinity of the critical thickness. Because of the exponential relationship between the tunneling current and the effective film thickness, tunneling current can be greatly enhanced by the external electric field and substrate induced strain. © *2012 American Institute of Physics*. [doi:10.1063/1.3673600]

INTRODUCTION

Advances in nanoscale growth and characterization techniques over the past decade have led to the preparation of high-quality ultrathin ferroelectric films with thickness of only a few nanometers, which give us the possibility of observing direct quantum-mechanical tunneling and the giant change of electroresistance through a ferroelectric barrier.[1–4] Typically, ferroelectric tunnel junction (FTJ) is formed by an insulating ultrathin ferroelectric barrier sandwiched between two metal electrodes. With the interplay of ferroelectricity and electron tunneling, the polarized barrier may have a profound effect on the conductance of FTJs, which may have potential applications in tunable memories and sensitive nano devices.[5–7]

For direct electron tunneling, the current across the barrier is described by the density of states in the electrodes, the tunneling matrix element and the thermal occupation probability of the states. According to the formula given by Simmons[8] with thermal occupation probability simplified by Winsal–Kramer-Brillouin approximation, the current of typical tunnel junctions depends exponentially on the barrier thickness, the square root of the effective mass, and the square root of the barrier height. Therefore, even a small change in any of these parameters will have a significant effect on the tunneling current.[9] Compared with common non-polar barrier dielectricities, spontaneously polarized materials add more freedom as well as new physical richness to the tunnel junction.

For an electric controllable ferroelectric barrier, the thickness of the ferroelectric film should be thin enough to achieve large tunneling current, but the thickness cannot go down below the critical thickness so that the polarization can still exist to give us the possibility of electric control of the polarization. With the tremendous improvements in nano scale fabrication and characterization techniques, high quality ultrathin ferroelectric films can be fabricated. A critical thickness of only 1.2 nm for $PbTiO_3$ film was detected by synchrotron radiation with clean interface and no chemical disorder.[10] The observed critical thickness is coincident with the prediction of the density-functional theory, where the reduction of the critical thickness and the enhancement of the ferroelectric phase were attributed to the ionic relaxations in the metal-oxide electrodes[11] even with a strong depolarization field in nanoscale films.[12]

When the barrier thickness is very close to the critical thickness of the ferroelectric film, dielectric and piezoelectric properties will have a sharp increase in the vicinity of the critical point under a DC electric field, generating a large piezoelectric strain. Although this piezoelectric strain may be less stable, it will result in a significant effect on the effective electron mass, conduction-band edge and the effective thickness of the barrier.[13] The piezoelectric coefficient of ultrathin ferroelectric films with thickness of 5-30 nm has been theoretically and experimentally investigated.[14] The piezoelectric coefficient along the normal direction of the film surface was largely degraded except a slight increase when the thickness of the film was under 12 nm. Garcia *et al.*[2] observed a large out-of plane piezo response phase contrast but only obtained a rather rough estimate of the piezoelectric coefficient of $d_{33} = 2$-5 pC/N. with the consideration of the large piezoelectric effect on the tunnel current in ferroelectric tunnel junctions, more details of the piezoelectric effect need to be worked out.

In this study, the polarization and piezoelectric properties of a highly strained ferroelectric thin film with the thickness below 5 nm were investigated based on a phenomenological theory. Using the theoretical value of the piezoelectric coefficient near the critical thickness, the current-voltage characteristic was theoretically analyzed with the consideration of the converse-piezoelectric effect under different mechanical boundary conditions.

PHENOMENOLOGICAL THEORY

Our study focused on a special FTJ composted of two symmetric conducting $SrRuO_3$ electrode layers separated by a single domain ultrathin $PbTiO_3$ layer with the thickness h. The $PbTiO_3$ thin film, with an equivalent cubic cell lattice

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parameter of 3.9633 Å, can be epitaxially grown on the $SrTiO_3$ (with lattice parameter $a = 3.905$ Å) or $NdGaO_3$ (with lattice parameter $a = 3.86$ Å) substrates.[15] The thick substrate can be considered as rigid, thus, the in-plane dimensions of the $PbTiO_3$ layer can be totally constrained by the lattice mismatch strain induced by the substrate. Under compressive substrate stress, the polarization is usually formed along the direction perpendicular to the interface of the film and substrate. Therefore, we consider the case with the polarization and electric field along the film normal direction. When the lateral dimensions are constrained by the substrate, the field-induced strain is also along the film normal direction. The free energy function was constructed by using the spontaneous polarization as the order parameter with the consideration of collective effects of depolarization field, interface charges and strain.

Because the electrodes in the electrode-ferroelectric-electrode heterostructure are not perfect conductors, the depolarization field still exists due to the incomplete screening of bond charges of the polarization near the surfaces even in short-circuit situation, which can largely suppress the polarization if the film is very thin. The electrostatic potential distribution, therefore, is highly related with the magnitude and direction of the polarization of the ultrathin barrier and the depolarization field. We should note that the ionic offset in a few atomic layers of the electrodes adjacent to the ferroelectric film might affect the screening due to the polarization-dependent atomic orbital hybridizations at the interface.[11] In this work, we simplified this problem by using an equilibrium result of the total energy potential with simultaneous consideration of the exact electrostatic potential and the polarization under the incompletely screened depolarization field.

Assuming that the compensation charges existed in the electrodes are $\pm q_e$, an internal depolarizing field E_{dep} in the FE film is given by $E_{dep} = -(P - q_e)/\varepsilon_b$. In the absence of external electric field, the compensation charges in the electrodes can be solved from the short-circuit boundary condition as $q_e = Ph\varepsilon_b/(l_{s1}\varepsilon_{e1} + l_{s2}\varepsilon_{e2} + h\varepsilon_b)$, where $\varepsilon_{e1}, \varepsilon_{e2}$ and l_{s1}, l_{s2} are the dielectric constants and screening lengths in the electrodes, respectively. Thus, the depolarization field related with the film thickness is finally derived as[16,17]

$$E_{dep} = -\frac{1}{\varepsilon_b}\left(1 - \frac{h\varepsilon_e}{l_{s1}\varepsilon_{b1} + l_{s2}\varepsilon_{b2} + h\varepsilon_e}\right)P = -\frac{1}{\varepsilon_b}(1-\theta)P. \tag{1}$$

Since the two electrodes are the same, there is no built-in electric field induced by different work function steps at the two ferroelectric-electrode interfaces. Recent results showed that the built-in field can reduce the critical size and even made the critical size vanished in special cases.[18]

According to the work of Tagantsev *et al.*,[19] the surface energy of the ferroelectric film in nanoscale heterostructures can be written as a Taylor series expansion with the two lowest terms given by $\Phi_S = (\xi_1 - \xi_2)P + (\eta_1 + \eta_2)P^2/2$, where ξ_1, η_1 and ξ_2, η_2 are expansion coefficients of the two surfaces, respectively. If the two electrodes are the same, the surface energy can be simplified as $\Phi_S = \eta P^2$.

For a ferroelectric film with two symmetric electrodes under a non-zero electric field, the thermodynamic potential can be obtained by solving the differential equation relation $dg/dE = -D$,[16] where E is the total electric field composed of external and internal fields, i.e., the external electric field E_{ext} and the depolarization field E_{dep}. With the consideration of external strain, electric field, and the surface energy, the total energy can be rewritten as

$$\Delta\Phi = \left[\frac{\alpha^*}{2}P^2 + \frac{\beta^*}{4}P^4 + \frac{\gamma}{6}P^6 + (s_{11} + s_{12})^{-1}u_m^2 - E_{ext}P\right]h, \tag{2}$$

with the renormalized coefficients $\alpha^*$and β^*given by

$$\alpha^* = \alpha_0(T - T_{c0}) - \frac{4Q_{12}u_m}{s_{11} + s_{12}} + \frac{1}{\varepsilon_b}(1 - \theta^2) + \frac{2\eta}{h}, \tag{3a}$$

$$\beta^* = \beta + \frac{4Q_{12}^2}{s_{11} + s_{12}}, \tag{3b}$$

where α_0, β, and γ are the expansion coefficients of the Landau free energy, T is the temperature, T_{c0} is the Curie temperature of the counterpart bulk material, Q_{12} is the electrostrictive coefficient, s_{ij} is the elastic compliance tensor, and u_m is the total external strain imposed on the ferroelectric film by the substrate. The first, second, third, and fourth terms of α^* are related with thermal, strain, depolarization and surface, respectively.

Without external applied electric field, the total electric field E is simply the depolarization field E_{dep}. The variation of the total free energy with respect to the polarization can be written as

$$\frac{-\delta\Phi}{\delta P} = h(\alpha^* P + \beta^* P^3 + \gamma P^5) = 0. \tag{4}$$

The stable polarization under zero electric field can be obtained from the above equation. The low-voltage dielectric constant along the normal direction of film-substrate interface is given by

$$\varepsilon_{33}^* = \varepsilon_0^{-1}\left(\frac{\partial^2\Phi}{\partial P^2}\right)^{-1} = \varepsilon_0^{-1}(\alpha^* P + 3\beta^* P^2 + 5\gamma P^4)^{-1}. \tag{5}$$

The variation of polarization and dielectric constant with respect to the external field will be slightly different, which may involve nonlinear piezoelectric responses.[20] To simplify the calculations, we still use a linear piezoelectric constitutive equation to derive the piezoelectric response of a constrained thin film under a small external electric field. With the boundary conditions of uniform in-plane strain $S_1 = S_2 = u_m$ and vertical stress $T_3 = 0$, the strain in the vertical direction can be derived from the piezoelectric constitutive equations as

$$S_3 = \frac{2s_{13}u_m}{s_{11} + s_{12}} + \left(Q_{11} - \frac{2s_{13}Q_{12}}{s_{11} + s_{12}}\right)(P + P^E)^2, \tag{6}$$

where P^E is the induced polarization by the external field. The first term is the strain induced by the external initial strain. Without considering nonlinear effects, the piezoelectric coefficient induced by the external electric field can be approximated as

$$d_{33}^{*}\Big|_{E_{ext}=0} = \frac{dS_3}{dE} = 2\left(Q_{11} - \frac{2s_{13}Q_{12}}{s_{11}+s_{12}}\right)P\varepsilon_0\varepsilon_{33}^{*}, \qquad (7)$$

with $\partial(P+P^E)/\partial E|_{E_{ext}=0} = \varepsilon_0\varepsilon_{33}^{*}$, ε_{33}^{*} and ε_0 are the relative dielectric constant at zero field and the vacuum dielectric permittivity, respectively.

With the application of an electric field along the thickness direction of the film, piezoelectric strain $\Delta S = d_{33}^{*}V/h$ can be induced. Since the converse piezoelectric strain can modify the effective barrier thickness, electron mass, and position of the conduction band edge, these characteristics will also be related to the polarization. The modified effective thickness, electron mass, and the conduction band potential can be expressed as $h^{*} = h(\Delta S + 1)$, $m^{*} = m_0(1+\mu_{33}\Delta S)$, and $\phi_c = \phi_c^0 + \kappa_3\Delta S$, respectively. Here, ϕ_c^0 is the minimum of conduction band potential in a constrained barrier at $V=0$, κ_3 is the relevant deformation potential of the conduction band, m_0 is the effective electron mass at $V=0$, and μ_{33} is a coefficient describing the increase of mass with respect to strain.[13] The corresponding total energy ϕ can be written as $\phi = \phi_c + \hbar^2 k_3^2/2m^{*}$, from which the wave number normal to the barrier plane can be derived as $-k_3^2 = 2m^{*}(\phi_c - \phi)\hbar^{-2}$ by using a one-band model.[13]

When an electron wave with wave vector k_3 encounters a potential step higher than its energy, there is a penetration probability if the barrier thickness is thin enough. Under external voltage, the penetration probability, which is also known as the transmission coefficient $T(\phi)$, can be simplified by using the WKB approximation,

$$\begin{aligned} T(\phi) &= \exp\left(-2\int_0^{h^{*}}\sqrt{-k_3^2}\,dx_3\right) \\ &= \exp\left(\frac{4h^{*}\sqrt{2m^{*}}}{3\hbar eV}\left[(\phi_c - eV - \phi)^{3/2} - (\phi_c - \phi)^{3/2}\right]\right). \end{aligned} \qquad (8)$$

Substituting the piezoelectric effected thickness, electron mass, and the conduction band potential by using $h^{*} = h(\Delta S+1)$, $m^{*} = m_0(1+\mu_{33}\Delta S)$, and $\phi_c = E_c^0 + \kappa_3\Delta S$, respectively, the integral expression for the current density $J = I/A$ at $T = 0$ K can be approximated as[13]

$$J = \frac{m_e e}{2\pi^2\hbar^3}\left[eV\int_0^{E_F - eV} T(\phi)d\phi + \int_{E_F - eV}^{E_F}(E_F - \phi)T(\phi)d\phi\right], \qquad (9)$$

where E_F is the Fermi level of the electrodes and m_e is the free electron mass. The voltage dependent current density J through a piezoelectric film can be calculated by using the approximate series expansions of $J = C_1V + C_2V^2 + C_3V^3 \ldots$.[13] The first linear coefficient is given by

$$C_1 = \frac{\Theta}{m_0 h^2}(\Xi + 1)e^{-\Xi}, \qquad (10)$$

where $\Theta = m_0 e^2/8\pi^2\hbar$ and $\Xi = (2h/\hbar)\sqrt{2m_0\phi_0}$; $\phi_0 = E_c^0 - E_F$ is the barrier height at $V = 0$. C_1 is unaffected by the piezoelectric effect, but highly dependent on the intrinsic properties of the barrier and electrons, i.e., the thickness of the barrier, the mass of the electron, and the barrier height under zero field. The second square coefficient can be written as

$$C_2 = -\frac{4\Theta}{t_0\hbar^2}d_{33}^{*}\left[\phi_0\left(1+\frac{2}{\Xi}+\frac{2}{\Xi^2}\right)(2+\mu_{33}) + \frac{\kappa_3}{h}\right]e^{-\Xi}. \qquad (11)$$

Since it is linearly proportional to the piezoelectric coefficient d_{33}^{*}, large piezoelectric coefficient will cause significant change in the tunnel current. The third cubic coefficient can be simplified as

$$C_3 = \frac{\Theta}{t_0^2\hbar^4\Xi^2}\left(\frac{4m_0e^2h^4\Xi}{3} + A\phi_0\hbar^2 d_{33}^{*2}\right)e^{-\Xi} + B, \qquad (12)$$

with $A = 8(3+2\mu_{33}+\mu_{33}^2)(1+\Xi+\frac{1}{2}\Xi^2) + \Xi^3(2+\mu_{33}+\frac{\kappa_3}{\phi_0})^2$, $B = \frac{e^2\Theta\Xi^2}{4\hbar^2\phi_0}\mathrm{Ei}(-\Xi)$. $\mathrm{Ei}(z)$ is the exponential integral function with expression $\mathrm{Ei}(z) = -\int_{-z}^{\infty}[\exp(-x)/x]dx$ for $z > 0$. As shown in Eq.(12), the cubic term has a quadratic relation with the piezoelectric coefficient, hence, the piezoelectric effect will greatly increase the contribution of this cubic term to the tunneling current.

RESULTS AND DISCUSSIONS

The parameters (listed in Table I) used for $PbTiO_3$ and electrode $SrRuO_3$ were taken from Refs. 13, 16, and 21. The calculations were carried out at zero temperature in order to compare with the results of Ref. 13. The film thickness of less than 5 nm was used to study the thickness-dependent physical characteristics of the barrier. External substrate strains of −0.032, −0.026, and −0.022 were chosen to demonstrate strong strain dependence of the piezoelectric effect.

Since the thickness of the barrier should be above the critical phase transition thickness, we first studied the critical thickness by finding where the paraelectric to ferroelectric phase transition occurs in the solution of Eq. (4). Because external strain can greatly decrease the critical thickness, strain engineering is a key method to make high quality ultrathin ferroelectric films ferroelectric. Our results showed that the polarization at zero temperature can be maintained down to the thickness of about 1.2 nm with the strain of −0.032 and the thickness becomes thicker when the constraint of the

TABLE I. Parameters of the $PbTiO_3$ and $SrRuO_3$.

$PbTiO_3$	$\alpha = 7.6\times(T-752)\times10^5$ $C^{-2}m^2N$, $\beta = -2.9\times10^8$ $C^{-4}m^6N$, $\gamma = 1.56\times10^9$ $C^{-6}m^{10}N$, $Q_{11} = 0.089$ $C^{-2}m^4$, $Q_{12} = -0.026$ $C^{-2}m^4$, $s_{11} = 8.0\times10^{-12}$ m^2N^{-1}, $s_{12} = -2.7\times10^{-12}$ m^2N^{-1}, $s_{13} = -2.7\times10^{-12}$ m^2N^{-1}, $\varepsilon_b = 90\varepsilon_0$; $\phi_0 = 0.5$ eV, $\kappa_3 = -4.5$ eV, and $\mu_{33} = 10$, $m_0 = 0.2m_e$.
$SrRuO_3$	$\varepsilon_{e1} = \varepsilon_{e2} \approx 8.45\varepsilon_0$, $\varepsilon_0\eta \approx 0.01\times10^{-10}$ m, $l_{s1} = l_{s2} \approx 0.6\times10^{-10}$ m; $m_e = 9.1094\times10^{-31}$ kg, $\varepsilon_0 = 8.85\times10^{-12}$ F/m.

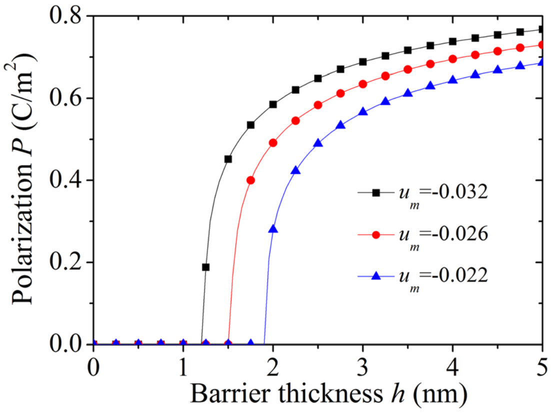

FIG. 1. (Color online) Thickness-dependence of polarization and critical characteristics under external strains of −0.032, −0.026, and −0.022. Critical thickness is defined at which the polarization disappears.

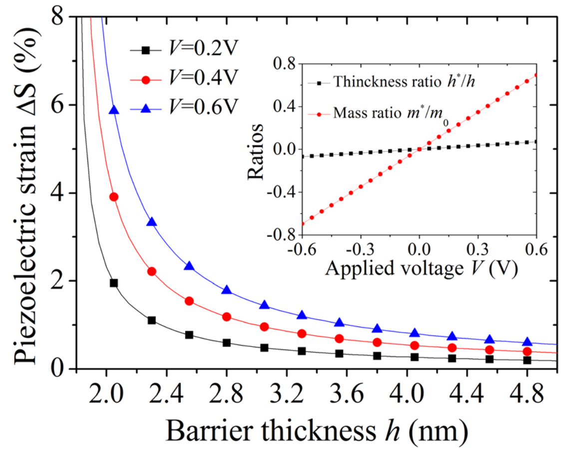

FIG. 3. (Color online) Change of the piezoelectric strain with film thickness under different applied voltage. Insert is the thickness ratio h^*/h and the mass ratio m^*/m_0 vs applied voltage for the barrier with initial thickness of 2 nm and external strains of −0.022.

substrate is weaker (Fig. 1). One could use compressive strain to counterbalance the detrimental influence of thickness reduction and preserve ferroelectricity at very low thickness. It is important to keep in mind that the properties of the film might be quite unique and less stable in the vicinity of the critical point.

As given in Eq. (7), the effective piezoelectric coefficient is very sensitive to the dielectric coefficient. Due to the singularity of the dielectric coefficient at the critical point, there is a sharp increase of the dielectric coefficient in the vicinity of the critical thickness, resulting in a strong enhancement of the piezoelectric coefficient (Fig. 2). Due to the modification by the piezoelectric strain, the effective thickness of the barrier with large piezoelectric coefficient will have noticeable change under external electric field. Our results indicated that a large piezoelectric coefficient might be achieved right above the critical thickness, even though it is less unstable. Due to the complex interplay of the piezoelectricity, thickness and the external strain, it is not always true that larger compressive strain will produce better properties for the barrier for a given thickness. Of course, the compressive strain at least should be large enough to sustain the ferroelectric state. A proper external strain applied to the film with a particular thickness could produce a relatively large and stable piezoelectric coefficient.

With the application of an electric field, the induced piezoelectric strain might be very high for a film with thickness right above the critical thickness, but decreases sharply with the increase of the film thickness as shown in Fig. 3. One can see that the strain can reach almost 6% in a film with the thickness of 2.1 nm under an external voltage of 0.6 V and an external strain of −0.022. In fact, a strain of about 6% can reduce the thickness of a film from 2.1 nm to 1.97 nm, which will produce significant enhancement of tunneling current because of the exponential relationship between the tunneling current and the film thickness. Large change also happens to the effective electron mass, the square of which can reach about 20% for a barrier with thickness of 2 nm and electron mass of 0.2 m_e as shown in the insert of Fig. 3.

The current-voltage characteristics in a symmetric FTJ are shown in Fig. 4. As expected, the tunneling current quickly increases with the decrease of the barrier thickness. As given in Fig. 2, the piezoelectric coefficient in the film with the thickness of 2.8 nm and external strain of −0.032 is about 50 pC/N, similar with the chosen piezoelectric constant in Ref. 13. However, the corresponding tunneling current is calculated much lower than other samples (Fig. 4).

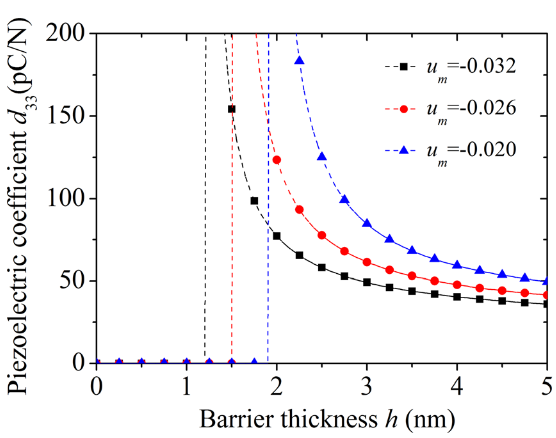

FIG. 2. (Color online) Thickness-dependent piezoelectric coefficients under various external strains.

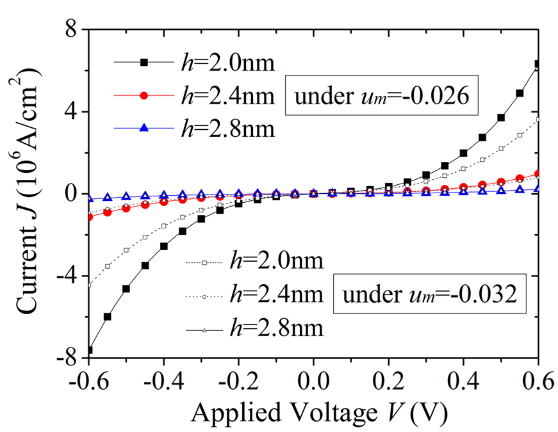

FIG. 4. (Color online) Current-voltage characteristic of ferroelectric tunnel junction with initial barrier thickness of 2.0 nm, 2.4 nm, and 2.8 nm under external strains of −0.026 and −0.032.

Since the barrier thickness is very close to the critical thickness, the piezoelectric properties and the corresponding effect on the tunneling current should be carefully considered. For example, film with a compressive strain of -0.026 will have larger piezoelectric coefficients than those with compressive strain of -0.032, resulting a larger enhanced tunneling current with smaller external substrate strain.

CONCLUSIONS

Piezoelectric and ferroelectric properties of ultrathin barrier were investigated theoretically with the consideration of external strain, depolarization, and surface effect. Critical thickness of the $PbTiO_3$ film is about 1.2 nm with an application of external strain of -0.032. The piezoelectric coefficient increases when the thickness of the film is right above the critical thickness with a smaller compress strain. An appropriate external strain was suggested to give a relatively larger piezoelectric effect and to maintain the properties more stable. With the inverse piezoelectric effect, great enhancement of tunneling current could be achieved due to the exponential relationship of the tunneling current with the thickness and the effective electron mass.

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