Supplementary Material

**Enhanced tunneling electroresistance effect in composite ferroelectric tunnel junctions with asymmetric electrodes**

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At the beginning, we assume both ferroelectric polarizations for 24 Å-thick BTO films in the composite FTJs are taken as 18 µC/cm2 from Ref. 31 where no STO layer exists (namely Pt/BTO/SRO on STO substrate). Considering the epitaxial strain in BTO relative to lattice mismatch among the electrodes and thin film layers, the ferroelectric polarization might be different in these FTJs. TABLE SI shows the lattice constants of bulk materials involved. It is likely that inserting an ultrathin STO layer between Pt/BTO or BTO/SRO can make BTO more compressive due to the smallest lattice constant of STO. That’s to say, 24 Å-thick BTO films in our Pt/STO/BTO/SRO and Pt/BTO/STO/SRO FTJs could have a large polarization than that in Pt/BTO/SRO structure with the same ferroelectric thickness. However, it is hard to differentiate the strength of ferroelectricity of BTO thin films between these two composite FTJs in the above way.

**TABLE SI** Lattice constants of bulk materials involved in FTJs

|  |  |  |
| --- | --- | --- |
| material | lattice constant (Å) | reference |
| Pt | 3.92(cubic) | 34 |
| BTO | 3.99(in plane) | 35 |
| SRO | 3.93(pseudo-cubic) | 36 |
| STO | 3.87(cubic) | 3, 6 |



**FIG. S1** TER (a) and asymmetric factor (b) as functions of the thickness of STO for Pt/STO/BTO/SRO (red) and Pt/BTO/STO/SRO (other colors) FTJs.

We expect the performance of Pt/STO/BTO/SRO FTJ is still better even for a smaller ferroelectric polarization in it. Thus, in the following calculations, the polarization of 24 Å-thick BTO films in Pt/STO/BTO/SRO is kept to 18 µC/cm2 while that in Pt/BTO/STO/SRO is assumed to a large value. FIG. SI(a) shows the dielectric-thickness dependence of TER ratio of Pt/STO/BTO/SRO FTJ (P=18 µC/cm2, red line) and of Pt/BTO/STO/SRO FTJ for different polarization strengths (lines with other colors). It is seen that the Pt/STO/BTO/SRO still preponderates as the polarization of BTO in Pt/BTO/STO/SRO FTJ has the bulk value of 26 µC/cm2. Even if the polarization of BTO reaches a huge value of 31.5 µC/cm2 that was reported for Co/ BTO (12 u.c.)/SRO FTJ on STO substrate,[37] the TER in Pt/STO/BTO/SRO is also larger by one order of magnitude, as can be seen in the figure. FIG. S1(b) shows the corresponding  curves. The enhanced TER in Pt/BTO/STO/SRO FTJ with ferroelectric polarization (FIG. S1(a)) could be explained by the increasing asymmetric factor. However, this increasing asymmetric factor is still smaller than that of Pt/STO/BTO/SRO FTJ, which further proves that larger is the asymmetric factor, the larger is the TER ratio. It is the large asymmetry factor for Pt/STO/BTO/SRO FTJ that is responsible for the giant TER effect. As the size effect of ferroelectricity is considered to better predict the performance of Ti/BTO/STO/SRO FTJs in Ref. 18, we use data from Ref. 31. The polarization strengths for 2 u.c.-,4 u.c.-, 6 u.c.- and 8 u.c.-thick BTO films are taken as 5, 11, 18 and 21 µC/cm2, respectively. At zero biased voltage, the calculated average barrier height of the composite barrier (for BTO polarized left and  for BTO polarized right respectively) and the corresponding asymmetric factor  for different BTO thicknesses  are listed in TABLE SII. It is seen that  and TER have the same changing trend and a larger  corresponds to a larger TER value. This result further confirms the correlation between TER and asymmetric factor.

**TABLE SII** Calculated average barrier height and asymmetric factor  as well as experimental TER values for FTJs with different BTO thicknesses at zero biased voltage (see Ref. 18 supplementary material)

|  |  |  |
| --- | --- | --- |
| (u.c.) | (e V)  (e V) | TER (%) |
| 2 | 0.6409 0.5591 | 1.1463 400 |
| 4 | 0.6787 0.5213 | 1.3020 7500 |
| 6 | 0.6824 0.5176 | 1.3184 36000 |
| 8 | 0.6678 0.5392 | 1.2385 7100 |

To further confirm the correlation between TER and asymmetric factor, the effects of the values of the model parameters including the dielectric constants of BTO and STO as well as the screening length of SRO on TER and asymmetric factor were investigated. The results are shown in FIG. S2. Still, TER and the corresponding asymmetric factor show the similar changing trends. This trend can be more clearly seen in FIG. S3 where STO has the same thickness as BTO, 24 Å. Owing to a larger asymmetric factor, Pt/STO/BTO/SRO FTJ has a larger TER than Pt/BTO/STO/SRO FTJ. These results suggest that the tight correspondence between TER and asymmetric factor could be extended to other systems.

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**FIG. S2** TER as a function of STO thickness for different dielectric constant of BTO (a), different dielectric constant of STO (b), and different screening length of SRO (c) for Pt/BTO/STO/SRO FTJs. TER as a function of STO thickness for different dielectric constant of BTO (d), different dielectric constant of STO (e), and different screening length of SRO (f) for Pt/STO/BTO/SRO FTJs. Insets show the corresponding asymmetric factors. = 2.4 nm, *P*=18 µC/cm2, =0.45 Å.

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**FIG. S3** TER as functions of the dielectric constant of BTO (a), dielectric constant of STO (c), and screening length of SRO (e) for Pt/BTO/STO/SRO and Pt/STO/BTO/SRO FTJs. (b), (d), (f) show the corresponding asymmetric factors, respectively. = 2.4 nm, *P*=18 µC/cm2, =0.45 Å.

**References**

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