Supplementary Information

Electrical Fatigue Behavior of NBT-BT-*x*KNN Ferroelectrics: Effect of Phase Transformations and Oxygen Vacancies

Xi Shi[1], Nitish Kumar[1] and Mark Hoffman[1,2]

1. School of Materials Science and Engineering, UNSW Sydney, Australia

2. Faculty of Engineering, UNSW Sydney, Australia

S1. Typical Strain loops

The figure S1 (a-c) show the typical strain loops (S(E)) of NBT-6BT at room temperature (NR), NBT-6BT-3KNN at 40 °C (ER) and NBT-12BT at room temperature (FE) measured at 10 Hz at maximum electric field of 50 kV/cm. It can be seen that NBT-12BT exhibits typical butterfly shape of S(E) loop with significantly large negative strain. NBT-6BT shows similar behavior, transitions to a ferroelectric phase at the first cycle of field application. Whereas NBT-BT-3KNN in the ER temperature regime shows as "sprout shape" with almost no negative strain, and accordingly larger usable strain than NBT-6BT and NBT-12BT.

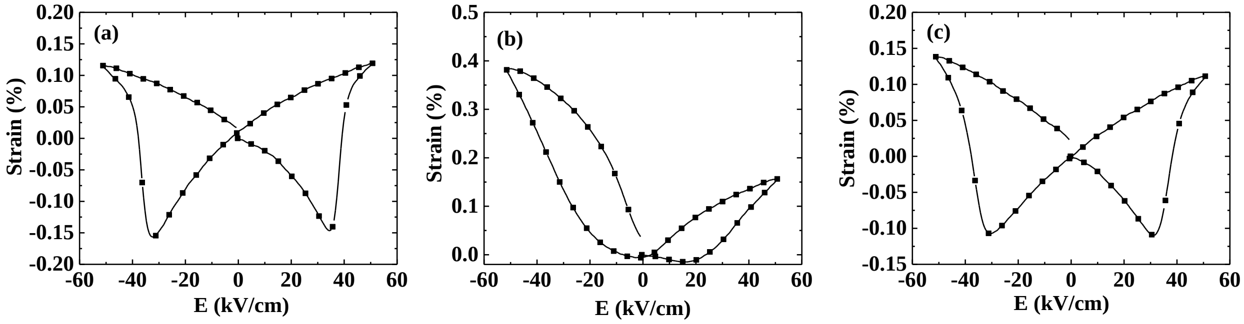

Fig. S1 S(E) loops of NBT-6BT (at room temperature), NBT-BT-3KNN (at 40 °C), NBT-12BT (at room temperature).

S2. Unipolar fatigue of NBT-12BT and NBT-BT-3KNN

Figure S2 shows the P(E) loops before and after unipolar fatigue of NBT-12BT and NBT-BT-3KNN at room temperature at 10 Hz till 10^5 cycles ((a) and (c)), as well as the corresponding evolution of P_{max} with cycles ((b) and (d)). Similar to NBT-6BT, unipolar fatigue for these two compositions also exhibit much less degradation in comparison with bipolar fatigue under the same measurement conditions.

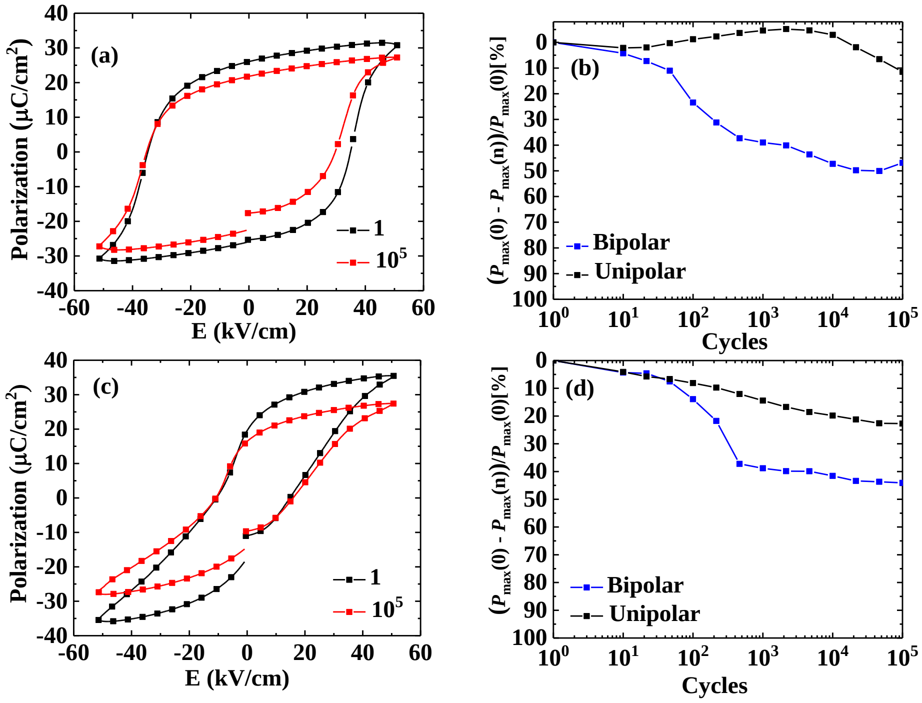

Fig. S2 (a),(c) P(E) loops for unipolar fatigue at room temperature and (b), (d) Comparison of evolution of P_{max} for unipolar and bipolar fatigue experiments for NBT-12BT, NBT-BT-3KNN respectively.

S3. Polarization loops of NBT-BT-3KNN and NBT-12BT with cycles

The evolution of P(E) loops as the function of cycles at 10Hz at maximum field of 50 kV/cm till 10^5 cycles of NBT-BT-3KNN at 40 °C as well as NBT-12BT at room temperature are shown in Fig. 3. The results are consistent with the Fig.1 (e) and (f) in the paper, where NBT-BT-3KNN, an ER, shows almost no degradation in the first 10^3

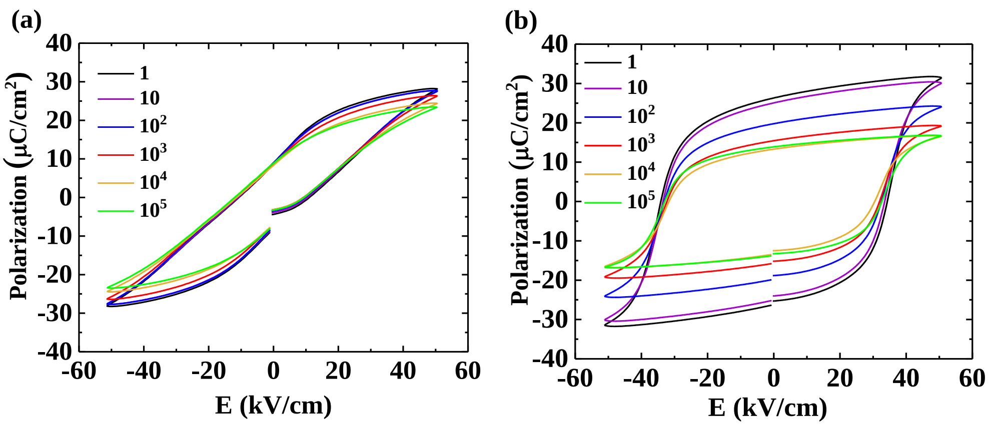

Fig. S3 Evolution of P(E) loops of NBT-BT-3KNN (at 40 °C) and NBT-12BT (at room temperature) fatigued at 50 kV/cm up to 10^5 cycles.

cycles, while NBT-12BT, an FE, displays remarkable fatigue during first 10^2 cycles.

S4. Low field fatigue of NBT-6BT

Figure S4 shows P(E) loops of an NBT-6BT sample before and after fatigue at 10 Hz at maximum field of 19 kV/cm for 10^5 cycles as well as the that after annealing treatment at 500 °C for 4 hours. It is shown that NBT-6BT still experienced significant fatigue at sub-coercive field level, while the full recovery after annealing treatment indicates that only domain wall movement is the dominant contributing factor for fatigue.

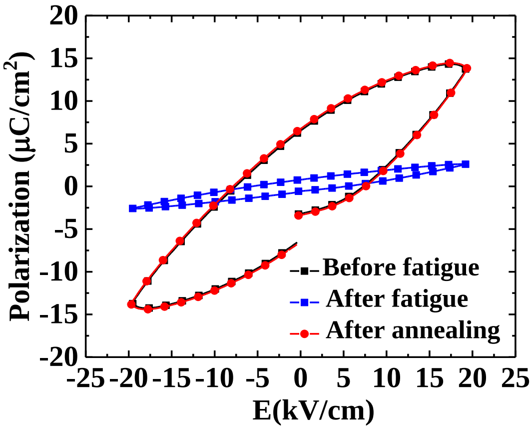

Fig. S4 P(E) loops of NBT-6BT before, after fatigue at 19 kV/cm till 10^5 cycles and after annealing treatment.