Self-Assembly of Ultrathin PVDF on Monolayer Graphene Oxide

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Introduction:

Ferroelectric random access memories (FeRAM) are a class of nonvolatile memories that employ ferroelectric (FE) thin films as active materials. Depending on the direction of the polarization (up and down) in FE materials, they represent binary memory states "0" or "1"^[1]. Obtaining ultrathin (sub 50 nm) flexible FE films is a prevalent challenge. Organic ferroelectric materials such as Poly (vinylidene fluoride) or PVDF with its co-polymers based on trifluoroethylene (TrFE) are highly preferred due to its flexibility and easy fabrication. Among these, PVDF-TrFE based flexible polymers are known to have excellent FE performance^[2]. To fabricate sub 50 nm films with high ferroelectricity, 2D graphene based polar nanomaterials were utilized as they can interact with the polymer to enhance the FE behavior^[3].

Experimental:

In the present study, we employed monolayer (~1 nm thick) graphene oxide (GO) for crystallization of 40 nm PVDF-TrFE film by the self-assembly approach. We expect that GO provides high orientation to PVDF chains in a specific direction resulting in excellent ferroelectric nature. Also, oxygen-functional groups in GO give high ferroelectric domain stability to PVDF (hydrogen bonds and columbic interactions). Scanning probe microscopy based techniques can allow us to read and write information at the nanoscale for the realization of miniaturized FE memory^[4]. We use piezoresponse force microscopy (PFM) to probe the piezoresponse and FE domain dynamics in the FE films.

Results:

A large piezoresponse of 145.75 ± 20.91 pm/V was observed in the ultrathin PVDF FE film which is 10 times higher than the conventional perovskite based FE films. Nanometer size FE domains were achieved through switching the FE polarization by applying pulses of -10 V (V_{write}) of varying time periods to the PFM cantilever (PFM images are shown in Fig. 1.). The FE domains remained stable over 1500 h with no volatilization revealing excellent domain stability. As these ultrathin FE films reveal



Fig. 1. FE domain dynamics in 40 nm PVDF-TrFE FE film. (a) PFM amplitude (b) PFM phase images of artificially fabricated FE domains by applying pulses of V_{write} with varying time.

highly dense memory, these materials will be explored further for applications in FeRAM.

References:

- [1] J. F. Scott, C. A. de Araujo, *Science* **1989**, *246*, 1400.
- [2] P. Taylor, T. Furukawa, *Phase Transitions* **1989**, *18*, 143.
- [3] P. Viswanath, K. K. H. De Silva, M. Yoshimura, Jpn. J. Appl. Phys. 2020, 59, SN1006.
- [4] D. Zhang, D. Sando, P. Sharma, X. Cheng, F. Ji, V. Govinden, M. Weyland, V. Nagarajan, J. Seidel, *Nat. Commun.* 2020, 11, 5.