Studying interface charge distribution in HfO2 and Al2O3 based nanocapacitors by operando electron holography

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Background

Among dielectric materials, HfO2 and Al2O3 are widely studied for micro and nanoelectronic applications. They present very interesting properties for several types of devices such as a higher dielectric constant compared to SiO2, high thermal stability and a compatibility with complementary metal oxide semiconductor (CMOS) technologies. For instance, resistive random-access memory (RRAM) devices [1, 2] or memristor devices [3] containing high-κ HfO2 and Al2O3 dielectrics are promising candidates in neuromorphic computing. However, the exact location of the charge stored in such devices is subject of debate and hence the mechanisms for charge trapping remain unclear. Particularly, the influence of interface charges has rarely been addressed because of the lack of suitable characterization techniques.

Whilst dielectric properties can be readily measured macroscopically using electrical methods, the task is difficult at the nanoscale. What is missing is a local and direct measurement of the electrical potential distribution within such devices upon biasing. Indeed, the phase of the electron hologram can be directly related to the electrostatic potential encountered by the fast electron along its trajectory. Recently, we were able to perform operando electron holography experiments in the same region under different biases and to successfully map the electric potential across a working SiO2-based MOS nanocapacitor with unprecedented sensitivity [4]. We observed an unexpected charging of the dielectric at both interfaces with the electrodes, over a distance extending up to few nanometers, a value much larger than the structural or chemical width of each interface [4]. In this study, we aim to take one step further to quantify the trapped charge at several interfaces, including insulator-insulator interfaces and metal-insulator interfaces, for which we have designed two thin tri-layer nanocapacitors: TiN/HfO2/Al2O3/HfO2/TiN (T-1) and TiN/Al2O3/HfO2/Al2O3/TiN (T-2) as shown in Fig. 1(a) & (b).

Methods

The tri-layer nanocapacitors were fabricated by atomic layer deposition (ALD). Each HfO2 or Al2O3 layer has a nominal thickness of 20 nm. TiN layers were deposited by sputtering before and after the insulating layer to serve as top and bottom electrodes during electrical biasing. In situ biasing TEM experiments necessitate a specific and complex sample preparation that minimizes preparation artifacts whilst ensuring the electrical functionality of the specimen itself. Specimens were prepared by FIB (FEI Helios 600i), on a Hummingbird chip-based biasing holder. Holography experiments were caried out using a Hitachi HF-3300 (I2TEM) microscope operating at 300 kV in Lorentz mode enabling a large field of view with a spatial resolution of 0.5 nm. Holograms were acquired during long exposure time using dynamical automation for compensating instabilities under different applied bias [5]. Finite element method (FEM) modelling was performed using COMSOL software to interpret the phase profiles quantitatively, including factors such as specimen geometry, preparation artefacts and stray fields around the sample device.

Results

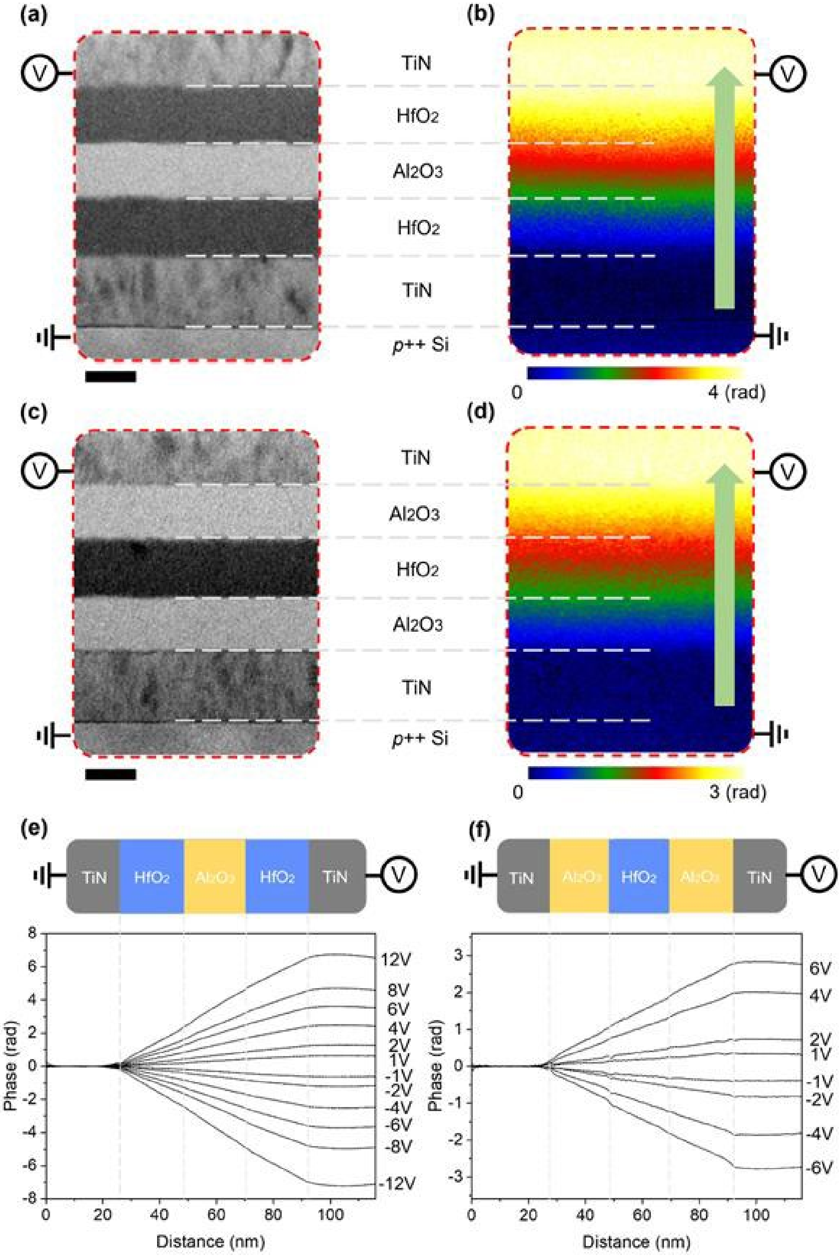
Figures (a) and (b) show the studied regions for T-1 and T-2, respectively while the corresponding phase maps extracted from holograms acquired under a bias of 6V are given in Figure (b) and (d), respectively. The phase map has been corrected by the reference hologram at 0 V, by which the experimental artifacts were removed and the remaining signal can be attributed to the applied electrical biasing. The artifacts comprise the possible variable lamella thickness, damage layers, diffraction contrast, and electron-beam-induced charging. The amplitude of the measured phase profiles across the tri-layer insulators (Figures (e) and (f)) extracted from phase images rise with increased bias, and appear symmetric when switching the sign of the bias, for example, 6 V and -6 V. However, the potential distributions as well as the measured electrical fields are quite different from those expected theoretically. Considering a relative permittivity of 7.4 for Al2O3 and 18 for HfO2, both measured by C-V tests at 100 kHz for their macro devices, the electric field should be 2.44 times higher in Al2O3 than HfO2, which consequently should result in a 2.44 times higher potential drop in Al2O3. In contrast, the experimental phase profiles highlight a homogenous electric field in all the layers for both T-1 and T-2 due to the appearance of charged layers at insulator-insulator interfaces as well as metal-insulator interfaces. Combined with extensive FEM simulations, we successfully quantified the charge densities of each interface for both capacitors as a function of the applied bias responsible of the homogenous electric field. These charges densities linearly vary with the applied bias and present a thickness between 3 and 5 nm width. Scanning TEM electron energy loss spectroscopy (STEM-EELS) measurements have been carried out to understand the origin of these charged interfaces.

Figure. (a) Device structure for T-1; (b) phase map of projected electric potential obtained by electron holography for T-2; (c) Device structure of T-2; (d) phase map of T-2. Here, (b) and (d) correspond to the same regions as (a) and (c) for T-1 and T-2, respectively. Scale bars are 20 nm in (a) and (c). (e) and (f) are the phase profiles extracted from the green arrows as a function of applied biasing for T-1 and T-2, respectively.

Conclusion

We used operando electron holography to evidence and quantify charges occurring at the different interfaces in tri-layer HfO2/Al2O3/HfO2 and Al2O3/HfO2/Al2O3 nanocapacitors with TiN electrodes. An equivalent uniform electric field distribution throughout the whole dielectric stack in HfO2 and Al2O3 is observed even though HfO2 has a much larger dielectric permittivity than Al2O3. The electric field distribution originates from the presence of charges trapped at each interface. It is revealed that there is a linear relationship between the surface charge density and the applied bias, and, at each bias, the interfaces between HfO2 and Al2O3 have an equivalent trapped charge density. With unprecedented sensitivity and spatial resolution, the information gained with operando electron holography on the location of the trapped charges and on their densities provide unique insights on the functioning of nanocapacitors. Further work could be devoted to the interface charging behaviour in ferroelectric-dielectric devices.

Graphic:



Keywords:

Electric field mapping, electron holography

Reference:

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