Scale-Free Ferroelectricity induced by atomic-scale electric dipoles in HfO_2 may enable ultimately high-density computer memories

Scientists are constantly improvising on nanoelectronic devices that can help develop better computer memories. The structure of an inorganic compound called hafnium oxide that scientists have explored offers immense possibilities for creating robust yet reversibly switchable electric dipoles for advancing nanoelectronics devices which can enable high-density computer memories.

A collaboration between Prof Umesh Waghmare of Jawaharlal Nehru Centre for Advanced Scientific Research (JNCASR), Bengaluru an autonomous institute of the Department of Science & Technology (DST), Government of India and Prof Jun Hee Lee's group from S Korea uncovers Natural Crystal Structural Architecture of Hafnium (IV) oxide (HfO_2), also known as hafnia, that may enable realistic, ultimately high-density computer memories using controllable atomic-scale electric dipoles. The work was published in the peer-reviewed journal '*Science*' on July 2, 2020.

Ferroelectric materials which are electric analogues of magnets exhibit spontaneously ordered electric dipoles, whose direction can be reversed by applying an electric field in opposite direction and are used to store binary information. In contrast to magnets, however, existence of electric monopoles or charges makes these dipoles rather sensitive to surrounding, thus limiting the miniaturization of ferroelectrics-based devices. Development of ultra-high-density ferroelectric memories critically depends on the stability of the dipolar structure at small sizes, and the dynamics of domain walls (walls that separate domains with distinct dipolar orientations).

Secondly, commercial ferroelectric materials are typically perovskite oxides, which are oxides of a metal and a transition metal (ABO_3 in which A is metal and B a transition metal). In addition to the challenge of intricate fabrication processes of growing nano-scale structures of ABO_3 oxides, the toxicity of A cation (often Pb) necessitates development of high-performance lead-free ferroelectric materials that are can be readily synthesized and fabricated at different scales.

In their work reported in Science Magazine on July 2, 2020, Prof Lee and Prof Waghmare have used computer simulations of quantum electronic states in crystals to uncover scale-free ferroelectricity (in which stability of a dipolar domain does not depend on its width or length-scale) in HfO_2, a material that is an important part of metal oxide semiconductor (CMOS) devices in use today. They have shown that orthorhombic crystal of HfO_2 has a structural architecture that provides a lateral array of 2-dimensional dipolar sheets separated by non-polar sheets. The dipolar sheets act like natural domain walls and sheaths separating adjacent dipolar domains (sheets), protecting them from detrimental effects of surroundings making it more stable and allowing the devices to be as small as one wants. The scale-free or stable nature essentially means that these ferroelectric dipolar sheets in HfO_2 are robust and individually switchable, supporting stable domains of all widths (length-scale), starting from about 0.25 nm. With consequent negligible effect of surrounding, they can be used for information storage at ultimately high density.

Their work also provides a fundamental understanding of the origin of scale-free ferroelectricity in HfO2. While polar displacements of cations are primarily responsible for electric dipoles in ABO_3 ferroelectrics, they demonstrate that equal partnership between polar and antipolar displacements of anions give rise to the array of alternating 2-D sheets of in-plane dipoles and sheets of dead (no dipoles at all) layer. Such a mechanism is possible because the polar and antipolar phonons in HfO_2 have the same energy (technically called as belonging to flat bands). A flat band of phonon waves essentially means that their group velocity vanishes. Such flatness of phonon bands is thus responsible for sluggish dynamics of dipolar domain walls observed in HfO_2. In addition, they show that domain walls in HfO2 are very special: their thickness and energy cost are vanishingly small.

As HfO_2 is used as a dielectric material in charge-storing capacitors in semiconductor electronic devices, ferroelectric HfO_2 if integrated into semiconductor chips, its ferroelectric properties add tremendously to its functionality. Professor Lee had spent a year working on ferroelectric superlattices in Waghmare's group at JNCASR during his doctoral studies at Seoul National University in 2007-8. He refreshed the collaborative work with a stimulus from recent observations in HfO_2 with technological significance and carried out the study under the umbrella of DST-supported India-Korea Joint Network Centre in Computational Materials Science with support by a JC Bose National Fellowship of SERB-DST.

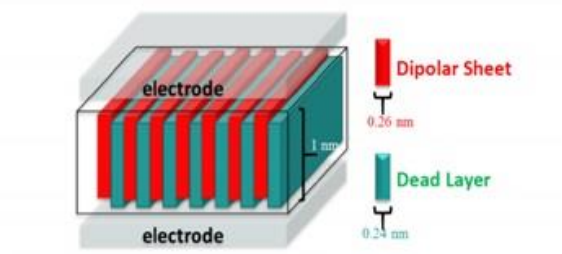

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Figure Caption: Red regions denote atomically thin dipolar sheet-like domains, and green regions are the dead, nonpolar layers with vanishing electric dipoles.

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