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Development of an operando laser-based photoemission electron microscope capable of characterizing ferroelectric properties

Established a new method for observing and evaluating ferroelectric properties of next-generation memory material, such as hafnia-based ferroelectrics

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Summary

- **Established an evaluation method based on directly observing devices in operation to evaluate their ferroelectric properties, which is essential for applying hafnia-based material, one of the most promising candidates for next-generation memory material.**

- **This is the first report on ferroelectric material characterization using an operando laser-based photoemission electron microscope (laser-PEEM) system with a Sawyer-Tower (ST) circuit.**

In 2011, a German research group reported that a thin film of hafnium dioxide (HfO_2), one of the most promising candidates for next-generation memory material, show ferroelectricity, and a research team led by Hirokazu Fujiwara at the Institute for Solid State Physics in the University of Tokyo has been working on the research and development of observation techniques to characterize its properties. In particular, they have been developing an observation system using operando laser-based photoemission electron microscope (laser-PEEM), which enables the direct observation of characteristic modulations. In this study, a Sawyer-Tower (ST) circuit was implemented in a laser-PEEM to evaluate the characteristics of ferroelectric devices during operation, and the world's first ferroelectric property evaluation was successfully demonstrated. This technology contributes to establishing observation and evaluation techniques that are useful for clarifying the mechanism of characteristic modulations, which is indispensable for the practical application of hafnia-based ferroelectrics.

Details

Hafnia-based ferroelectrics, a new material for next-generation memory

A German research group reported the ferroelectricity of a thin film of hafnium dioxide (HfO_2) in 2011; the film thickness is about 10 nm and has attracted much attention. Thus, research and development of "hafnia-based ferroelectrics" (*1) have been active as a new material for next-generation memories.

Applying voltage to ferroelectric material produces macroscopic "polarization", which remains even when the voltage is reduced to zero. Ferroelectric random-access memory (FeRAM) is an application of this physical storage media feature. Among ferroelectric memories, "perovskite-type oxide ferroelectrics," which use inorganic oxides with perovskite structures (*2) are currently the most used for practical applications. They are widely used in transportation IC cards owing to their low-voltage operation and high-speed response.

Due to features such as low-voltage drive and high-speed response performance, ferroelectric memory is expected to be a technology that will contribute to lower power consumption and greater computing efficiency through artificial intelligence if it can replace DRAM (*3), the main memory of computers. However, perovskite-type oxide ferroelectrics in practical use have the disadvantage of not being able to retain their ferroelectricity when miniaturized. Thus, it is not easy to apply them to large-scale integrated circuits, such as smartphones, where miniaturization is progressing. However, the new Hafnia-based ferroelectric material can exhibit stable ferroelectricity even in thin films of 10 nm or less, and if applied, it can replace the main memory in computers.

However, hafnia-based ferroelectrics have disadvantages, such as durability. By 10^5 –10^{10} times of write/erase operations, after the polarization is modulated, eventually it causes dielectric breakdown. When this breakdown occurs, the memory function is no longer retained. DRAM used as a working memory must withstand information rewriting 10^{15} times or more; however, how the amount of ferroelectricity polarization changes owing to repeated write/erase operations (information rewriting), leading to dielectric breakdowns, remains unresolved from a physical standpoint.

Therefore, fundamental physical studies are required to elucidate the mechanism of characteristic modulations to improve the performance of hafnia-based ferroelectrics. Hirokazu Fujiwara and his research team at the University of Tokyo implemented a "Sawyer-Tower (ST) circuit" (*5) in a laser-based photoemission electron microscope (laser-PEEM*4) and performed characterization of the ferroelectric devices in operation. This technology contributes to establishing observation and evaluation techniques helpful in elucidating the mechanisms of characteristic modulations in hafnia-based ferroelectrics.

Enables the advanced observation of devices and catalysts in operation

In order to investigate the mechanism of characteristic modulations in hafnia-based ferroelectrics, it is important to directly observe how the characteristics are modulated in an operating device, or perform 'operando observation'. Our research team has achieved this observation by utilizing 'operando laser-PEEM,' says Fujiwara.

The word "operando" means "working" or "operating" in Latin, and operando observation is a technique for directly observing a device or catalyst in operation.

Microscopes are used for the direct observation of devices. Optical microscopes have excellent physical properties and depth of detection (several micrometers) but low resolution (~500 nm). Electron microscopes have moderate observation of physical properties and high resolution (>1 nm) but have a problem with the detection depth (<10 nm). Fujiwara et al. used PEEM, which combines optical and electron microscopy.

PEEM uses an imaging technique to magnify and image electrons emitted by the photoelectric effect (a phenomenon in which electrons are ejected from the material when light is exposed on the material surface) caused by irradiating the material with an ultraviolet (UV) laser. Unlike other electron microscopes, this technique is sensitive to the electronic and chemical states of the material. Furthermore, it has the advantage of nondestructive observation of films embedded under electrodes and other materials, making it ideal for observing ferroelectric material.

Conventional PEEMs have a resolution of only 20–50 nm, making them unsuitable for research on semiconductor devices in which advanced microfabrication is applied. However, the cutting-edge PEEM at the Institute for Solid State Physics in the University of Tokyo, which uses a deep-ultraviolet continuous-wave (CW) laser (*6) as its light source, can achieve the world's highest resolution 2.6 nm, thereby enabling research on semiconductor devices.

The group of Fujiwara et al. utilized this operando laser-PEEM to report operando observations of resistance change memory in 2018 and the dielectric breakdown process observations of hafnia-based ferroelectric capacitors in 2022.

Operando observation under AC voltage, the key to ferroelectricity evaluation, is realized.

“Previous studies used operando laser-PEEM to characterize physical properties based on DC voltage. In this study, we developed the AC-voltage-based system. The novelty is that we can now quantitatively evaluate the amount of device polarization, which is important in ferroelectricity evaluation based on capacitance and inductance.,” says Fujiwara.

Although the electrical resistance can be mainly evaluated under DC voltage, the actual amount of ferroelectric polarization cannot be fully evaluated only from the electrical resistance. Fujiwara's research group implemented the "Sawyer-Tower (ST) circuit" in an operando laser-PEEM system. It succeeded for the first time in the world in evaluating electrical polarization while applying 1 MHz-class cycling stress (voltage application that reverses polarization many times) by AC –voltage-application system (Figure).

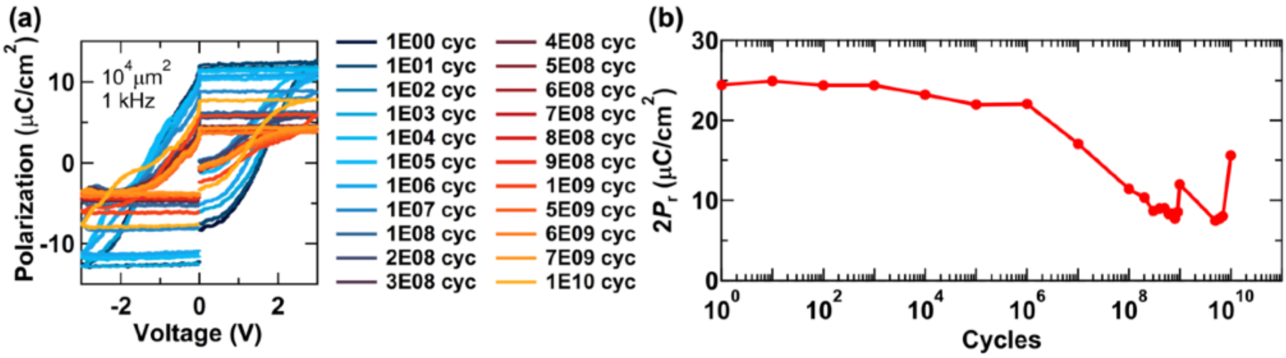
“The strengths of the observation and evaluation methods we have established thus are that they enable us to get closer to the mechanisms of characteristic modulations that lead to the dielectric breakdown of ferroelectric material. In the future, we would like to elucidate the chemical changes that cause characteristic modulations in hafnia-based ferroelectric capacitors, particularly decrease of polarization. In the future, I would like also to expand my research into the mechanisms of characteristic modulations in various ferroelectric memory devices and contribute to their practical applications.” says, Fujiwara.

The research group will continue its research to characterize "ferroelectric field effect transistors (FeFETs)" and "ferroelectric tunnel junctions (FTJs)," which are expected to be used as non-volatile memories in the future.

Annotation

*1 **Hafnia-based ferroelectrics**: Inorganic compounds with a basic composition of hafnium oxide (HfO_2) are known for their ferroelectricity in thin films with a thickness of approximately 10 nm.
*2 **Perovskite structures**: A type of crystal structure is found in inorganic compounds with an ABX3 composition. $BaTiO_3$ and $PbTiO_3$, which exhibit ferroelectricity, also exhibit perovskite structures.
*3 **Dynamic random-access memory (DRAM):** A type of read/write semiconductor memory, used in many modern PCs as main memory.
*4 **Laser-PEEM /** Laser-based photoemission electron microscope: An electron projection microscope that performs imaging by magnifying and imaging the electrons (photoelectrons) emitted by the photoelectric effect from the object under observation when irradiated with light having energy greater than or equal to the work function.
5 **Sawyer-Tower (ST) circuit**: A circuit that generates electric field hysteresis, which is necessary to evaluate ferroelectricity in ferroelectric materials
6 **Deep ultraviolet continuous wave (CW) laser:** A laser light source using deep ultraviolet light. Because there is no instantaneous photoelectron excitation as in conventional pulsed light sources, "Coulomb repulsion" between excited photoelectrons can be suppressed and high resolution can be obtained.

Figures



Field cycling characteristics of a 100 µm x 100 µm capacitor with 10 nm thick $Hf_{0.5}Zr_{0.5}ZO_2$ were observed without applying high voltage between the sample and the objective lens. (a) P-V hysteresis loop obtained by the PUND method; (b) extracted changes in $2P_r$ due to electric field cycling.