HF5000 Field-emission Transmission Electron Microscope: High Spatial Resolution and Analytical Capabilities for Material Science

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1. Overview

In recent years, the field of highly functional materials and devices—including not only research and development, but also quality control and other areas—has begun to demand instruments capable of making simple and stable observations, with improved analytical performance, on length scales ranging from nanometers to atomic sizes. In an effort to meet the wide-ranging material analysis needs of this growing field, we have developed the HF5000 field-emission transmission electron microscope (TEM), an instrument equipped with a cold field-emission electron gun (CFEG)—with an accelerating voltage of 200 kV—that is designed to achieve high spatial resolution while offering sample tilting and advanced analytical capabilities (Figure 1).

Guided by the mission of achieving high sensitivity analyses and high-resolution observation in a 200 kV TEM platform at the sub-angstrom (Å) level, the HF5000 offers a STEM spatial resolution of 78 pm, high sample tilt angles, and large-solid-angle energy dispersive X-ray spectroscopy (EDX) all with a single pole piece.

In addition to retaining key features of previous instruments—such as the automatic correction function, the symmetry-dual Silicon Drift Detector (SDD), and the ability to capture spherical-aberration-corrected atomically resolved SE images thanks to the Hitachi-designed spherical aberration corrector of the “HD-2700” scanning transmission electron microscope (STEM)—the HF5000 aggregates and fuses core technologies perfected in the development of Hitachi’s HF series of TEMs.

As a high-resolution, high-sensitivity TEM/STEM instrument supporting an expanded range of observation and analysis techniques, the HF5000 is not only a powerful tool for high-end expert operators, but also offers improved ease-of-use features designed to make the instrument accessible to a wide range of users.

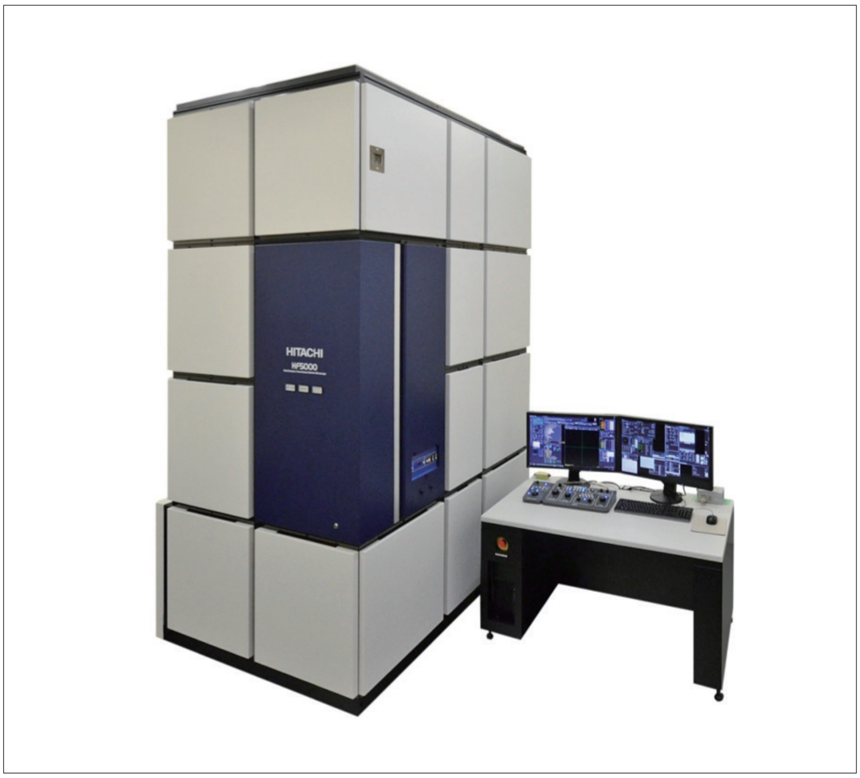

Fig. 1 Hitachi’s HF5000 Field-emission Transmission Electron Microscope

2. Overview and Specifications of the HF5000

The HF5000 TEM retains the key strengths of previous-generation TEM models while offering improvements over basic units in several areas, including the electron gun, the electron-optics system, and the mechanical structure and electrical stability of the instrument. These have been revisited and redesigned to improve analytical performance in the atomic-scale regime and expand the instrument's applicability to a wider variety of samples. The microscope column has been enclosed in an instrument cover, reducing the sensitivity of the structure to temperature variations and other external influences. In addition, the fluorescent screen binocular loupe in previous-generation models has been replaced by a charge-coupled device (CCD) camera for observation; this allows the instrument to be operated in bright laboratories, or via remote control from outside the laboratory in which the TEM is installed. The controls necessary to calibrate the instrument are aggregated into a single control panel, and the press of a single button configures the detector, the camera, the aperture position, and the lens settings as appropriate for STEM or TEM observations—allowing users to make observations without even being aware of which detector, camera, or apertures is in use.

Table 1 lists key specifications of the HF5000. In addition to the 200 kV accelerating voltage, an available option enables the instrument to support low accelerating voltages, allowing observation of samples susceptible to electron-beam damage. Switching accelerating voltages requires approximately three (3) minutes. At an accelerating voltage of 200 kV, the TEM resolution (lattice) is 0.102 nm, while the STEM resolution is 78 pm. The field-of-view size at the smallest STEM magnification corresponds to 1.1 mm × 1.1 mm; wide-area observations are possible when searching for the appropriate field of view or recording the position of an observation.

Table 1: Key specifications of the HF5000 transmission electron microscope

Accelerating voltages	60*, 200 kV
Electron gun	Cold field-emission electron gun
Cs corrector	STEM (Probe) corrector
STEM resolution	78 pm
STEM magnification	High Mag ×300 - ×4,000,000 Low Mag ×20 - ×260
TEM resolution	0.102 nm (lattice)
TEM magnification Camera length	High Mag ×3,000 - ×1,500,000 Low Mag ×100 - ×3,000 Diff 0.1 - 1.5 m
Sample tilt angle (Hitachi double tilt specimen holder*)	α ±25°, β ±35°
EDX detector *	100 mm^2 silicon drift detector (SDD)
Single	1.01 sr
Dual	2.02 sr
Image observation modes	
TEM	TEM imaging, diffraction patterns
STEM	SE, DF, BF-STEM

Asterisks indicate available options. Values quoted for resolution and magnification are for accelerating voltage 200 kV.

Table 2 lists the camera/detector combinations that may be used with the HF5000. The cameras used for capturing and recording TEM images and diffraction patterns each have their own advantages. To offer HF5000 users maximal freedom in customizing their use of the instrument for specific applications, we have equipped the instrument with two Hitachi cameras plus a 35 mm camera port for optional use; moreover, devices from other manufacturers—such as EELS detectors, CCD or CMOS cameras for high-resolution observations—can be installed in the space beneath the lens column.

Meanwhile, the instrument is equipped with three STEM detectors: a bright-field STEM (BF-STEM) detector and an annular dark-field STEM (DF-STEM) detector, both mounted below the sample, and an Everhart-Thornley (E-T) secondary-electron (SE) detector mounted on the upper side of the sample above the objective lens. All three signals may be recorded and saved simultaneously, both as static images or as movies. When capturing additional STEM images there is no need to re-scan and re-store a view position that has already been observed. Thus, for samples susceptible to electron-beam damage—for which beam scans are a precious resource—it is possible to acquire and compare various types of data from the sample with just a single scan. The ability to capture STEM images and SE images—which are sensitive to surface information—simultaneously, at high speed, is also useful for finding the optimal field of view for the analysis in question. Figure 2 shows examples of simultaneously captured high-resolution STEM images of a sample consisting of gold nanoparticles, approximately 20 nm in diameter, evaporated on a thin carbon film. An advantage of this measurement is that multiple types of information are obtained at the same time: the DF-STEM image indicates atomic-number-dependent Z-contrast (the technique is commonly known as high-angle annular dark field (HAADF-STEM), as it is particularly useful for detecting high-angle scattering, but in this report we refer to it simply as DF-STEM), the BF-STEM image indicates phase contrast, and the SE image reflects the three-dimensional surface structure of the sample. The ability to observe atomic resolution SE images as shown in Figure 2 in real time is among the most powerful features of the HF5000[1-8].

Table 2: Camera/detector combinations available on the HF5000 transmission electron microscope

TEM/STEM	Camera/Detector	Primary uses and key features
TEM	35 mm camera port camera (*1)	Diffraction patterns, wide field of view
	Hitachi-made screen CCD camera (*2)	Searching for field of view, diffraction patterns, calibration
	Hitachi-made standard CCD camera (*2)	TEM observation, aberration correction
	High-resolution recording camera (*1)	High-resolution TEM image/movie capture
	Electron beam energy loss spectrometer (EELS) (*1)	Energy-filter images, spectrum imaging
STEM	Secondary-electron detector (SE)	Observation of surface information
	Annular dark-field STEM detector (DF)	Atomic-number-dependent Z-contrast images of samples
	Bright-field STEM detector (BF)	Phase-contrast images
	EDX detector	Element spectral analysis, mapping

(*1) Optional
(*2) Hitachi-made camera

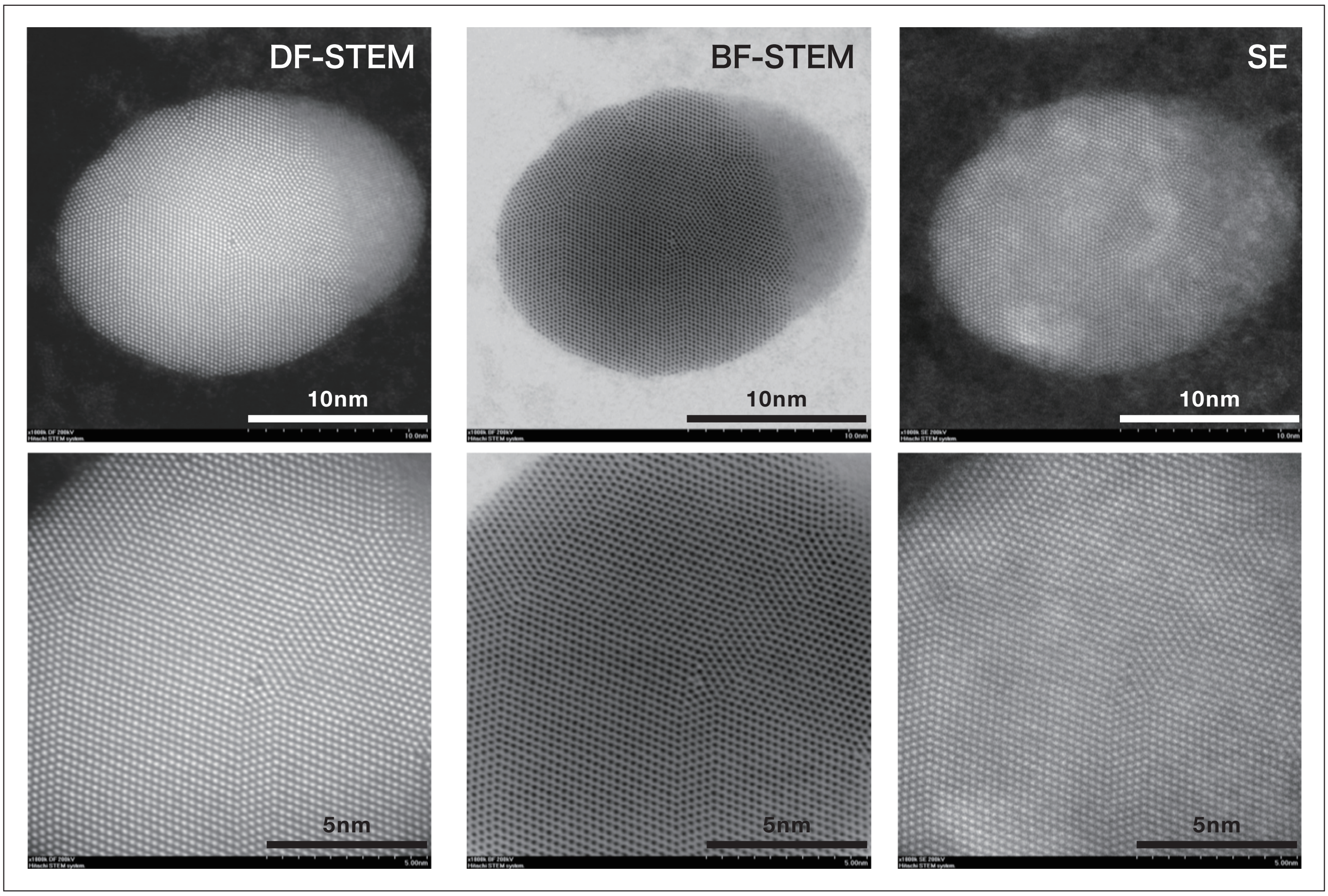

Fig. 2 Illustrative high-resolution STEM images of a gold nanoparticle evaporated onto a thin carbon film, observed at an accelerating voltage of 200 kV with STEM and SE detector signals acquired simultaneously. The lower images were obtained at a higher magnification.

In the following section we will present some key technical features of the HF5000.

3. Key Development Goals

3-1. Achieving high-resolution observation, high-sensitivity analysis, and high sample-tilt angles—all in a single objective lens design

Realizing this combination of capabilities—an increasingly essential requirement for studies in the field of materials analysis—imposed several challenges on the development of the HF5000: to improve resolution capability, increase the sensitivity of the EDX detector, and ensure the largest possible sample tilt angle, the optical design parameters of the objective lens pole piece and the position of the EDX detector were optimized and the structure of the sample stage was redesigned (Figure 3). In addition, several strategies may be used to heighten the X-ray detection sensitivity for EDX analysis: increasing the solid angle, installing multiple detectors, and using windowless detectors[9-10]. The HF5000 allows two windowless SDDs of detector area 100 mm^2 (Oxford Instruments X-Max^N 100 TLE) to be mounted facing each other in a left-right symmetric configuration with respect to the sample. The signals obtained from the two detectors can be combined. Moreover, a single detector can be used to correct the degradation in sensitivity caused by the shadow cast by the sample holder when the sample is tilted, reducing the effect of detector sensitivity loss with sample tilt angle. This has the advantage of allowing the orientation of the sample to be prioritized while restricting the effect of sensitivity loss to a minimum. Figure 4 plots the intensities of the signals obtained from the two EDX detectors, and their sum, for Ni-Kα lines obtained from a NiO_x thin-film sample at tilt angles of 0° (no tilt) and 10° (in the direction of detector 1). For the 0° case, the intensities of the signals from the two detectors are equal and the total count is 23,000. On the other hand, for a sample tilt of 10° in the direction of detector 1, the counts reported by detector

1 increase, while those from detector 2 decrease, yielding approximately 22,000 total counts. This indicates that the combined count of the two detectors remains roughly equal independent of sample tilt angle, demonstrating that this detector configuration exhibits little variation with sample tilt angle.

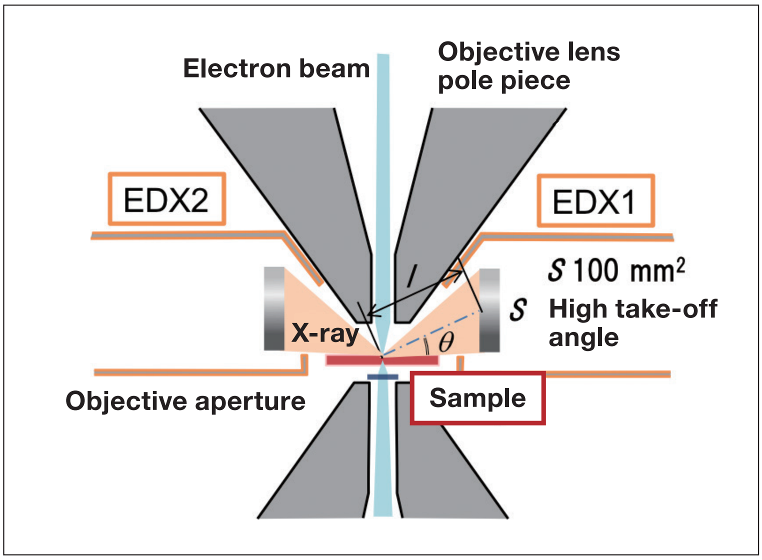

Fig. 3 Schematic of objective lens unit

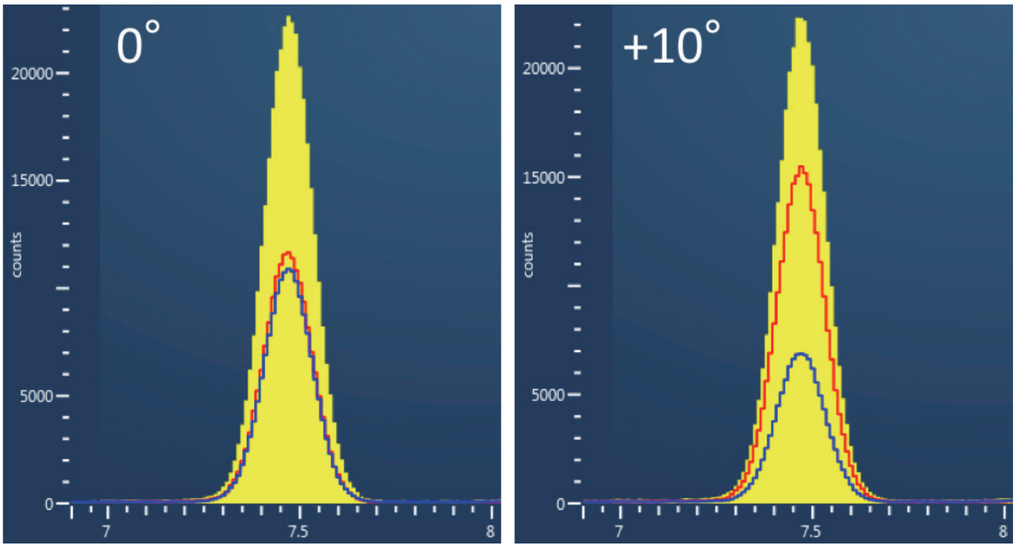

Fig. 4 Dependence of Ni-Kα lines on sample tilt (Left: sample tilt 0°; Right: 10°)
Red: detector 1 counts. Blue: detector 2 counts. Yellow: Combined counts. Sample: NiO_x thin film.

Another requirement for high-sensitivity EDX analysis is the ability to irradiate samples with large probe currents. Figure 5 compares DF-STEM images of various samples acquired by the HF5000 under typical STEM irradiation conditions (irradiation modes). The electron beam is focused on the sample by the electron lens, which plays the role of a convex lens; however, in practice, spherical aberration gives rise to broadening at the imaging surface. Spherical aberration is one factor obstructing the achievement of high resolution in electron microscopy. The function of aberration corrector acts similar to that of concave lenses; it has the effect of dispersing the electron beam along the near axis. By incorporating an aberration corrector into the instrument, the size of the electron beam on the sample is compressed to sub-angstrom sizes; moreover, focusing the electron beam at larger angles makes it possible to obtain larger probe currents. The HF5000 offers an ultra-high-resolution (UHR) mode as well as modes (such as normal mode or EDX mode) in which large probe currents around 1 nA are available. The optical configuration required for these irradiation modes is preset within the HF5000 control software and can be selected as necessary for the application in question.

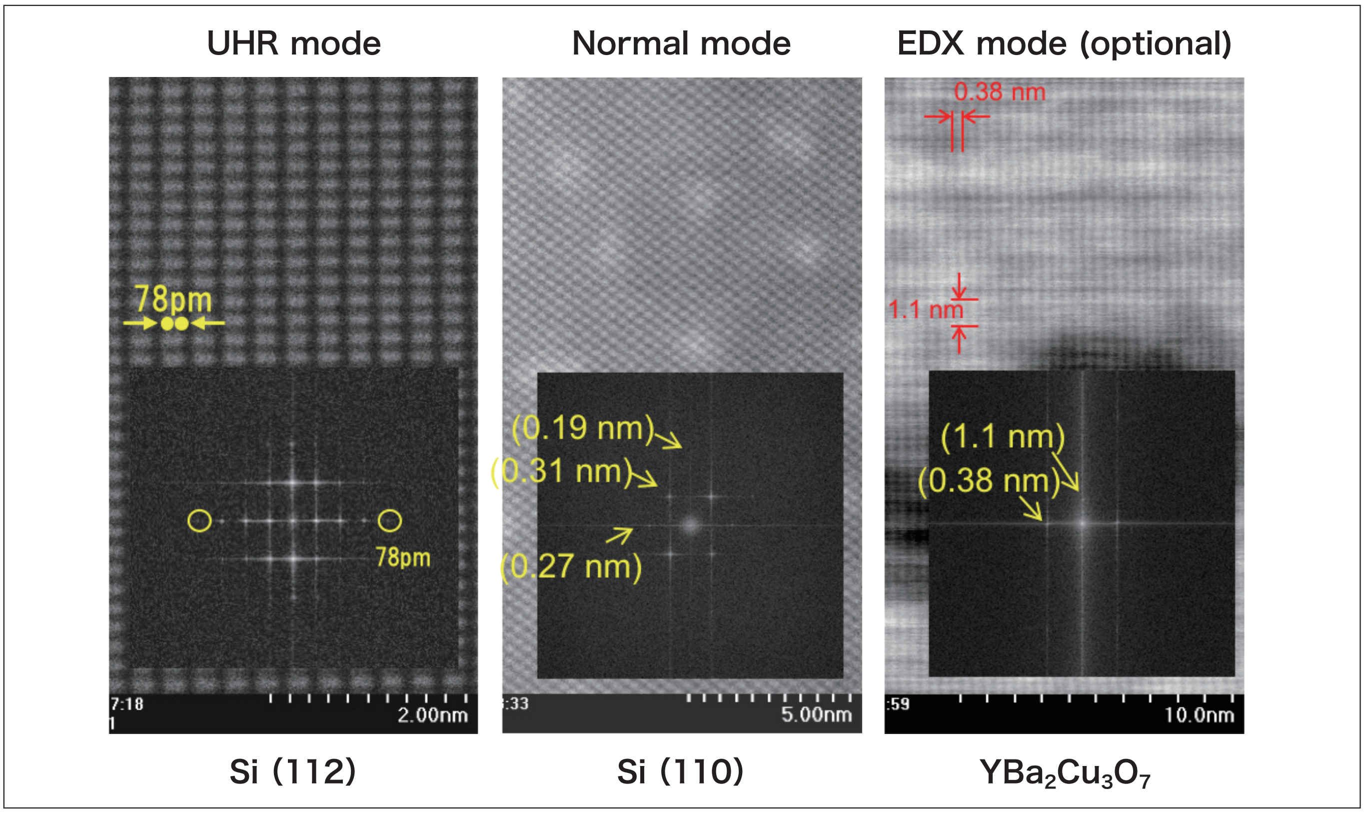

Fig. 5 Relationship between STEM illumination modes and observable images
(Probe currents satisfy the ranking UHR<Normal<EDX; probe diameters satisfy UHR<Normal<EDX)

3-2. Achieving the instrumental stability required for atomic-scale analysis

Enabling stable observation and analysis at atomic scales requires not only improving the performance of the electron optics system, but also developing solutions to several other significant challenges: electron-beam stability in the electron microscope, greater electrical and mechanical stability, and reduction of external disturbances and other factors affecting performance.

Regarding the cold field-emission electron gun used by the HF5000, existing electron guns offer high brightness and outstanding interference properties, but reducing FE-tip noise due to temporal fluctuations in probe current and stabilizing the beam current over long periods of time have proven challenging. To address these issues and provide an electron beam with long-term stability and reduced fluctuation in probe current, we have improved the design of the instrument to improve the vacuum pressure in the vicinity of the electron-gun anode[11]. In addition, we have improved the stability of various components whose electrical properties determine the spatial resolution of electron microscopes—including the high-voltage power supply, the lens system, and the deflector power supply—and have made structural changes to the instrument to reduce the effect of external disturbances on the electron microscope column to the greatest possible extent. Mechanically, the effect of external disturbances is reduced by the addition of a cover for the microscope column, and the redesigned unit reflects computational optimizations to suppress vibration sources and mechanical transfer function. In addition to introducing a vacuum system to reduce contamination, we have taken steps to maintain stability through major changes in electron-optics conditions—such as changes in accelerating voltage—and the instrument is replete with technical innovations to facilitate observation and analysis.

3-3. Automatic adjustment functionality and improved ease of use, for the Cs corrector

Although Cs correctors are extremely useful for STEM, optimal performance cannot be achieved unless the residual aberration that arises when correcting spherical aberration is eliminated. It is not impossible for users to perform this adjustment while looking at the image, but doing this successfully requires an extraordinary level of expertise. The HF5000 incorporates the Cs corrector and automatic aberration-correction functionality designed by Hitachi and included in the Hitachi HD-2700 STEM, allowing correction of aberrations using amorphous samples. Figure 6 illustrates the conceptual flow of this process; when aberration-correction mode is initiated and the start button is pressed from the control window, the system automatically performs correction up to third-order aberration, facilitating high-resolution observations.

Although many settings are pre-configured in the form of presets—including settings for the irradiation system, which determine beam conditions for TEM and STEM, the lens conditions and aperture setting of the imaging system, which determine the magnification and detection angle, and the deflector condition—the instrument also allows "free lens control", in which expert users themselves freely adjust and configure settings to create arbitrary observational configurations. The Personal Data Set feature allows these specialized sets of lens conditions to be saved and restored, facilitating repeat evaluation at later times. In addition, the optional remote-operation feature allows the instrument to be controlled remotely using a PC monitor and a cloned representation of the control panel, allowing the TEM and the operating rooms to be physically separated. In this case, analytical instruments from other manufacturers may also be used via remote control.

Fig. 6 Automated adjustment using Hitachi's Cs corrector

4. Application Data

In this section we present illustrative data—including both images and analytical data—from real-world applications that illustrate the various features of the HF5000 discussed thus far, including the improved performance of the electron-optics system, the improved stability, the Cs corrector, and the instrument's improved ease of use.

Figure 7 shows a DF-STEM image and atomic resolution EDX maps of a thin-film sample of single-crystal $SrTiO_3$, acquired at an accelerating voltage of 200 kV. The DF-STEM image, Sr atoms appear as bright spots due to its high atomic number (Z=38), while slightly darker Ti atoms (Z=22) are visible in atomic column positions. On the other hand, the light element O (Z=8) cannot be seen in this image. In this case, we acquired signals from two simultaneous detectors. Because we are using a windowless detector, the positions of the oxygen columns may also be identified. An advantage of this measurement is that the two EDX detectors are positioned to face each other, so the effect of sample tilt on crystal orientation is minimal.

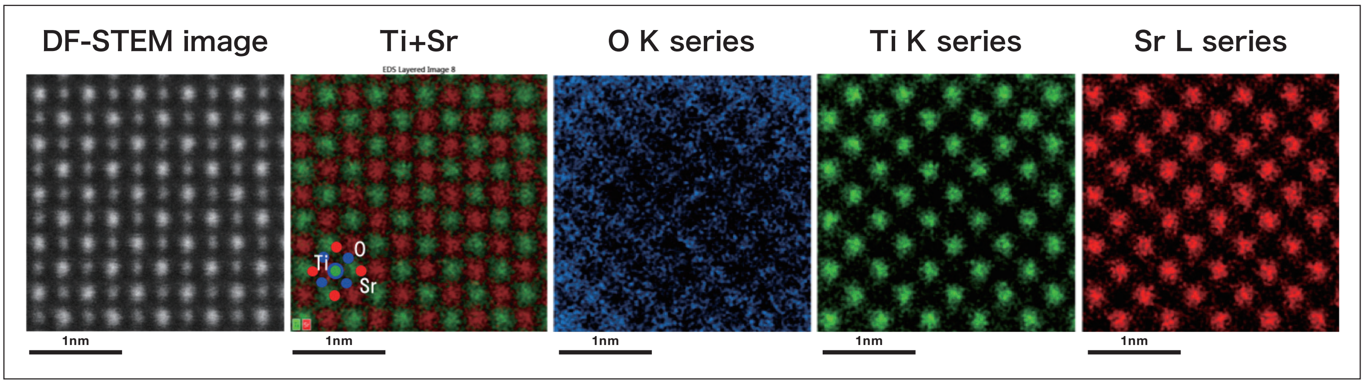

Fig. 7　DF-STEM image, and results of atomic resolution EDX mapping, for a $SrTiO_3$ sample

The HF5000 offers an optional Annular Bright Field (ABF) aperture for observing atomic columns of light elements in STEM images. When used in place of the BF-STEM aperture—which is mounted immediately above the BF-STEM detector and has a retractable mechanism—this allows the detector angle to be configured optimally for observation of light elements, allowing acquisition of "ABF-STEM" images.

In characterizing activity states of metallic catalyst particles, a comparison of SE and STEM images yields important information on the overall structure of the material composition of the sample and the 3-dimensional structure of material surfaces. Figure 8 shows simultaneously acquired atomic resolution DF-STEM and SE images of an Au nanoparticle, approximately 5 nm in size, adhering to a CeO_2 supporting material. The DF-STEM image provides information on the material composition of the sample in the form of atomic-number-dependent contrast. The SE image allows acquisition of topographical information reflecting surface states, such as the way in which Au adheres to the supporter.

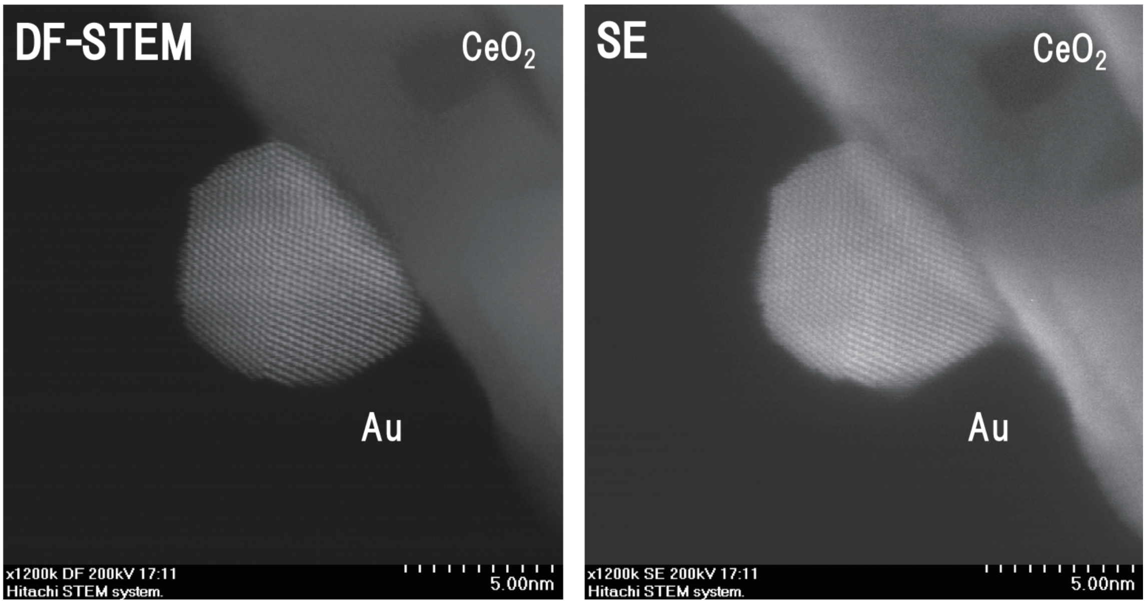

Fig. 8　High-resolution DF-STEM and SE images of Au nanoparticle catalyst supported by CeO_2 (Haruta Catalyst)

When observing samples susceptible to electron-beam damage, it is possible to avoid damaging the sample by taking steps such as reducing the dose of electron-beam irradiation or reducing the accelerating voltage to reduce the energy of the irradiating beam. Reducing the accelerating voltage has the added benefit of enhancing contrast in TEM images. Figure 9 shows a high-resolution TEM image of the vicinity of the substrate portion of a silicon device, captured at an accelerating voltage of 60 kV (an optional feature available for the HF5000); the observation yields a high-contrast lattice-resolved image.

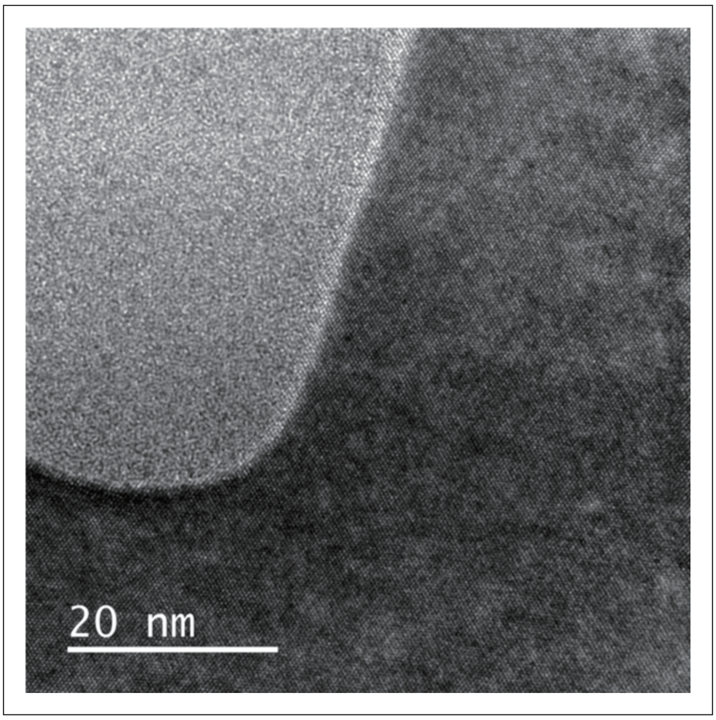

Fig. 9 High-resolution TEM image of a silicon device captured at accelerating voltage 60 kV

5. Conclusions

In this report, we introduced the technological capabilities of Hitachi's HF5000 field-emission TEM and presented case studies illustrating applications of the instrument, focusing primarily for STEM observations. This instrument also offers a full suite of optional features that allow it to be used as an analytical electron microscope in a wide range of applications. The HF5000 renews Hitachi's commitment to providing instruments with sub-angstrom spatial resolution and high-performance analytical capabilities—in combination with an ever-expanding range of observational and analytical techniques—as tools to facilitate characterization of highly functional materials and devices, for use not only by high-end expert operators but by a wide range of user communities.

References

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Abbreviations

※1 TEM: Transmission Electron Microscope
※2 STEM: Scanning Transmission Electron Microscope
※3 EDX: Energy Dispersive X-ray Spectroscopy
※4 SE: Secondary Electron
※5 SDD: Silicon Drift Detector
※6 CCD: Charge-Coupled Device
※7 CMOS: Complementary MOS
※8 EELS: Electron Energy Loss Spectroscopy
※9 Cs: Spherical aberration coefficient

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