

# Super High Resolution Imaging with Atomic Resolution Electron Microscope of JEM-ARM300F

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Through the technology evolved in the R005 project and the JEM-ARM200F, we developed an atomic resolution electron microscope of the JEM-ARM300F as a new platform for a super high-resolution instrument. The developed microscope is equipped with an ultra-stable cold field emission gun and spherical aberration correctors for probe and image forming systems. The stability of the microscope in TEM was tested by a lattice fringe of a crystal specimen and Young's fringe for a thick specimen showing beyond  $(50\text{ pm})^{-1}$  spatial information in these images. Ga-Ga dumbbells separated by 63 pm for a GaN [211] specimen was resolved in high angle annular dark field STEM imaging. Sub-50 pm imaging was demonstrated in STEM using a Ge [114] specimen.

## Introduction

Since an aberration correction system has been practically established [1-3], electron microscopes with the system drastically enhanced analysis capability in a scanning transmission electron microscopy (STEM) and structural study in transmission electron microscopy (TEM). During the R005 project (Project leader: Prof. Kunio Takayanagi 2004-2009), Tokyo Institute of Technology and JEOL Ltd. realized resolution of 0.05 nm [4,5], with a developed 300 kV high resolution electron microscope equipped with probe- and image-forming aberration correctors [3]. For an electron source, a cold field emitter was used to achieve high-brightness and narrow energy spread. 63-pm resolution was demonstrated using a GaN crystalline specimen observed from the [211] orientation [6]. Sub-50 pm resolution was demonstrated [7] using 47-pm separated atomic columns of Ge-Ge with Ge[114] specimen [8,9]. Li atomic column was detected with annular bright field (ABF) imaging technique for a sample of  $\text{LiV}_2\text{O}_4$  [10].

For 200 kV microscopes, spherical aberration correctors of CESCOR and CETCOR (CEOS GmbH) were installed into the JEM-2100F and the JEM-2200FS in 2003 [11]. And the JEM-2100F with a STEM aberration corrector imaged atomic structures of segregations at grain boundaries [12]. Next, we developed a 200 kV high-end

microscope for commercial use in 2009; it is the JEM-ARM200F, which is equipped with a spherical aberration corrector for a probe-forming system in the standard configuration. The microscope demonstrated sub-Ångström imaging with Ge and Si specimens observing from the [112] orientation. Light elements were visualized by ABF with a developed STEM detecting system using a bright field aperture [13]. Atomic resolution analysis by electron energy loss spectroscopy (EELS) and energy dispersive spectroscopy (EDS) were also demonstrated.

Refining these technologies cultivated in the R005 project and the JEM-ARM200F, we developed a new atomic resolution microscope of 300 kV JEM-ARM300F. In this paper, the features and the performances of the microscope are introduced.

## Instrumental features

### Appearance of JEM-ARM300F

**Figure 1** shows appearances of the JEM-ARM300F. There are four types of configurations depending on corrector equipment; basic version which has no corrector, STEM corrector version [Fig.1(a)], TEM corrector version, and double corrector version [Fig.1(b)]. The width of an operation table is same as that of the JEM-ARM200F, whereas the JEM-ARM300F is higher than JEM-ARM200F; the height of the STEM-version of the JEM-ARM300F is 3.16 m and that of the double version is 3.44 m. The JEM-ARM300F is usable at a higher accelerating voltage up to 300 kV and better resolution is attainable, as shown later. Then, we name the microscope "GRAND ARM"

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as a nickname, presenting the highest class of the ARM series.

6-SIPs & TMP pumping system and high performance cold field emission gun

To achieve high quality vacuum at the specimen

stage, we employed a differential pumping system composed of six sputter ion pumps (SIP) and a turbo molecular pump (TMP), as shown in Fig. 2(a). The specimen stage is pumped with a 150 L/s SIP, the intermediate lens (IL) and the condenser lens (CL) systems are pumped with two individual 20 L/s SIPs. The pre-evacuation of a specimen holder is performed with the TMP, whereas the TMP is

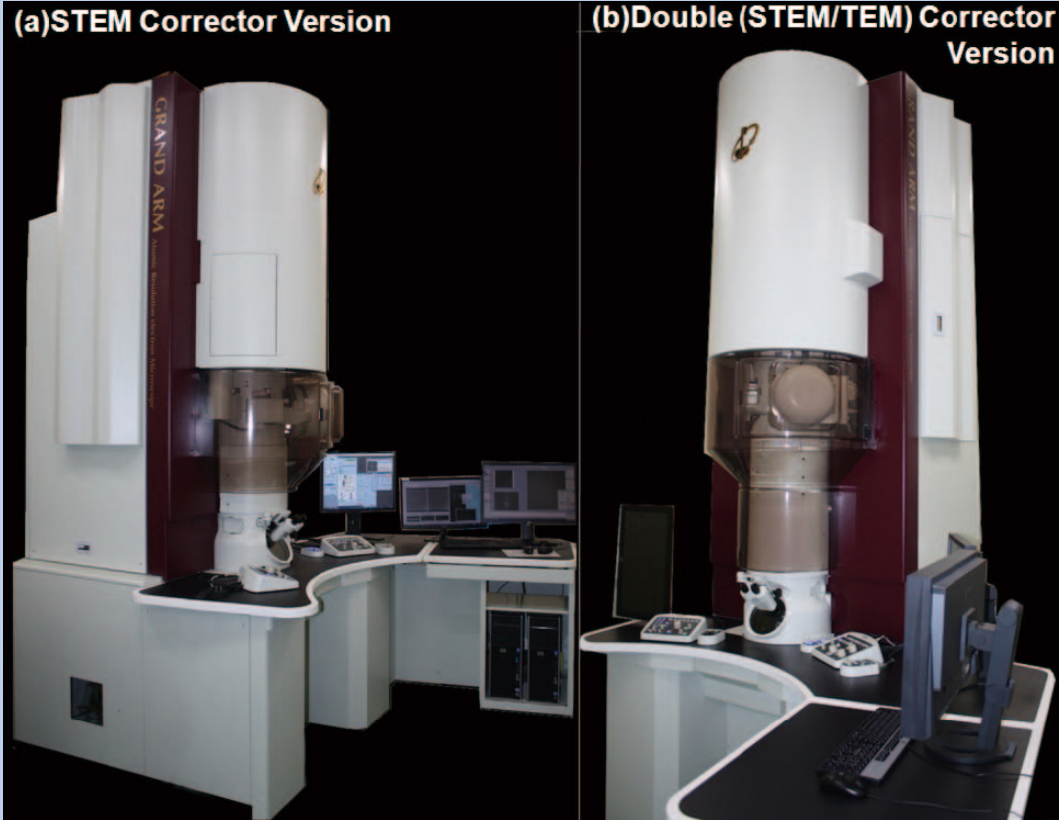


Fig. 1 Appearance of JEM-ARM300F (GRAND ARM).  
(a) STEM corrector version (b) Double corrector version.

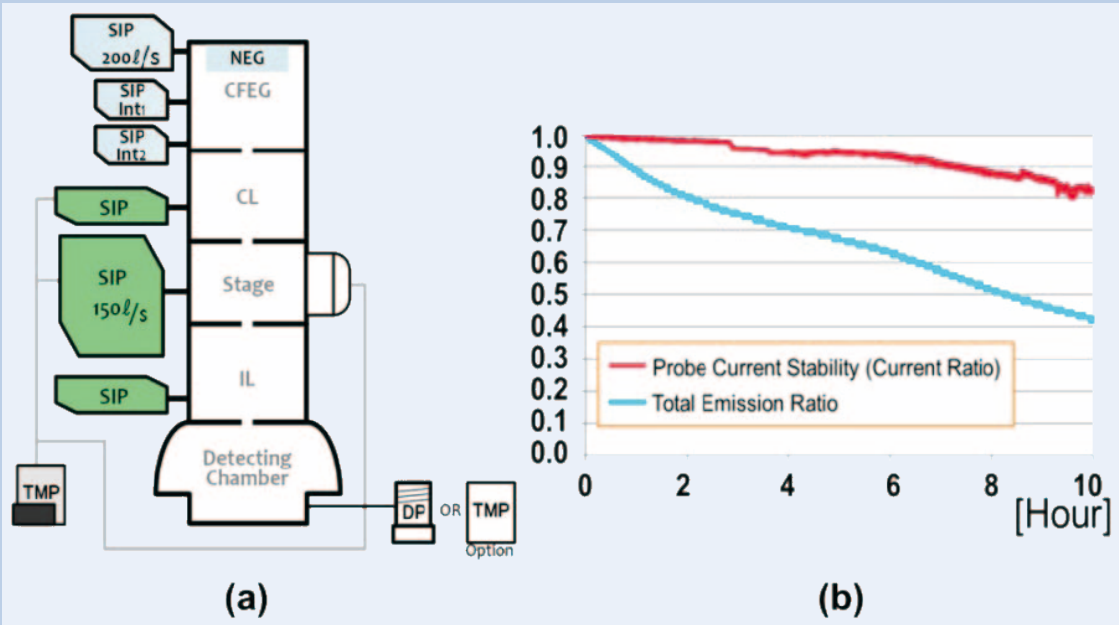


Fig. 2 (a) Schematic of the pumping system in JEM-ARM300F. (b) Emission stability for the cold field emission gun.

stopped during observation. The TMP is also used to pump the column during the baking of column and/or a liquid nitrogen tank for anti-contamination. With this pumping system, a pressure level of  $2\sim 10\times 10^{-6}$  Pa is typically achievable at the stage measured with the ion current of the 150 L/s SIP.

The developed CFEG is evacuated with non evaporable getters (NEG), a 200 L/s SIP (with noble pump) and two 20 L/s SIPs. NEG pumps the gas around the emitter, and the 200 L/s SIP pumps the accelerating tube, and the 20 L/s SIPs pumps the 1<sup>st</sup> and 2<sup>nd</sup> intermediate chambers [14]. The pumping system enables achieving an ultimate current stability of the CFEG. Figure 2(b) shows current stability, after a flashing of the emitter and a setting an emission current to be 10  $\mu$ A. Total time for the procedures of flashing and auto-emission takes approximately 1 min. Owing to an ultra-high vacuum of  $4\sim 10\times 10^{-9}$  Pa measured at the accelerating tube, the probe current keeps 90 % of initial current even after four hours. Thus, the 6-SIPs & TMP pumping system is very effective to keep a specimen stage

vacuum clean and stabilize the emission current from the electron source well.

## Corrector system

The JEM-ARM300F is equipped with an aberration corrector, which was developed in the R005 project [3]. The spherical aberration is compensated by two three-fold astigmatism fields [1] generated in two dodeca-poles (**Fig. 3(a)**). As a further optical innovation, an electron trajectory is expanding toward a specimen in the corrector and largely expanding between the condenser-mini lens and the transfer condenser-mini lens (CM-CMT) or between the objective-mini lens and the transfer objective-mini lens (OM-OMT) [3]. The expanding trajectory enables us to reduce disturbance in above elements resulting that extra chromatic aberration and noise from the corrector can be reduced. We call the system as ETA (Expanding Trajectory Aberration) optical system (Fig. 3(b), (c)).

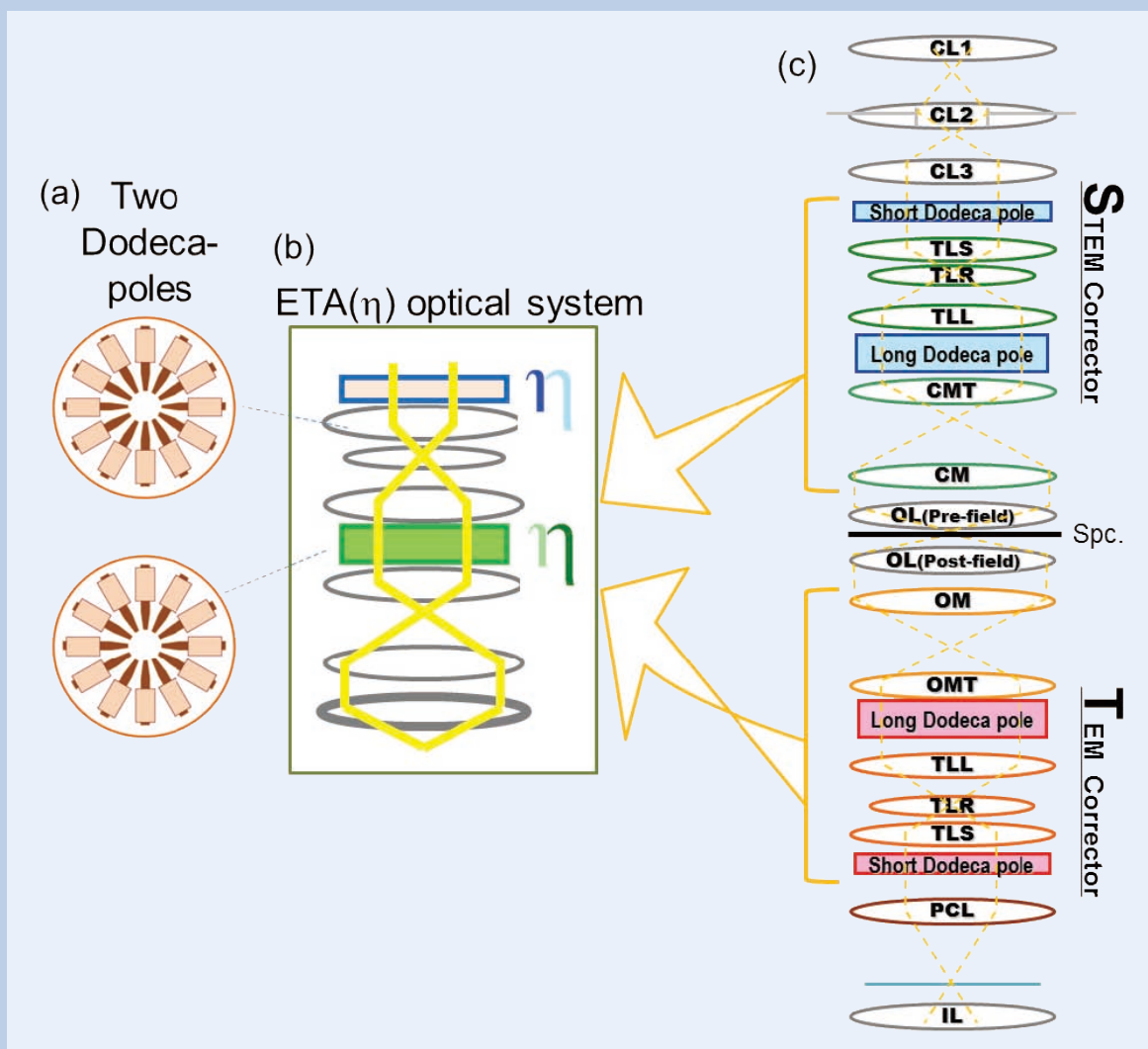


Fig.3 (a) Schematic of dodeca-poles. (b)ETA (Expanding Trajectory Aberration Correction) optical system. (c) Schematic drawings of STEM and TEM ETA correctors.



## Corrector system module

For a corrector control software, we developed the corrector system module (JEOL COSMO: corrector system module), which measures aberrations up to 5<sup>th</sup> order. For tuning of a probe-forming lens system, under and over defocused Ronchigrams are recorded to measure aberrations by the segmented Ronchigram auto-correlation function matrix (SRAM) method [15] (**Fig. 4(a-d)**). For tuning of an image-forming lens system, diffractogram tableau [16] are used for aberration measurement (**Fig. 4(e,f)**).

## Highly stabilized column

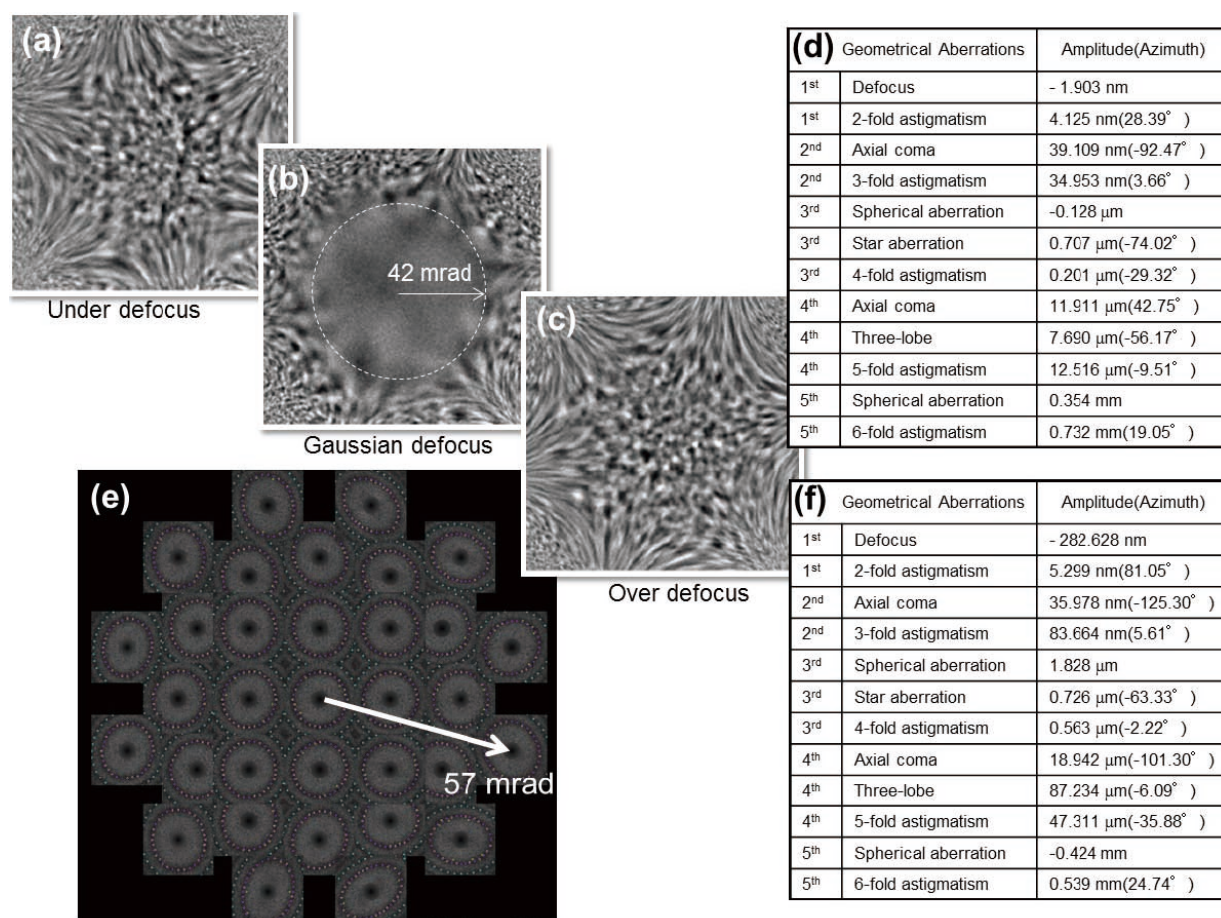
**Figure 5(a)** shows a power spectrum of Fourier transform from the TEM image of a Si [110] specimen at an accelerating voltage of 300 kV, where lattice information beyond 50 pm can be confirmed. Young's fringe test including non-linear term using a thick specimen of gold particles on a carbon film

is shown in **Fig. 5(b)**. Fringes are extended to spatial information better than (50 pm)<sup>-1</sup>. These results indicated that the mechanical and electric stability of the microscope realized the capability of 50 pm resolution.

**Figure 5(c)** is a high-resolution HAADF STEM image of Si [110] at 300 kV with an acquisition time of 10 s and **Fig. 5(d)** is one with an acquisition time of 80 s. A high-resolution image with a long acquisition time in **Fig. 5(d)** shows little distortion, indicating that the scanning system and the stage are highly stabilized against not only high frequency disturbance but also low frequency fluctuation.

## Detecting System

The microscope is equipped with a viewing chamber and a detecting chamber (**Fig. 6(a)** and **(b)**), where four STEM detectors can be attached totally: high-angle annular dark field (HAADF), low-angle annular dark field (LAADF), annular bright field (ABF),



**Fig. 4** (a-c) Under and over defocused Ronchigrams at 300 kV. (d) Measurement values for a probe-forming lens by SRAM at an accelerating voltage of 300 kV. (e) Diffractogram tableau for aberration measurement at 300 kV. (f) Measurement values for the image-forming lens by diffractogram tableau at an accelerating voltage of 300 kV.

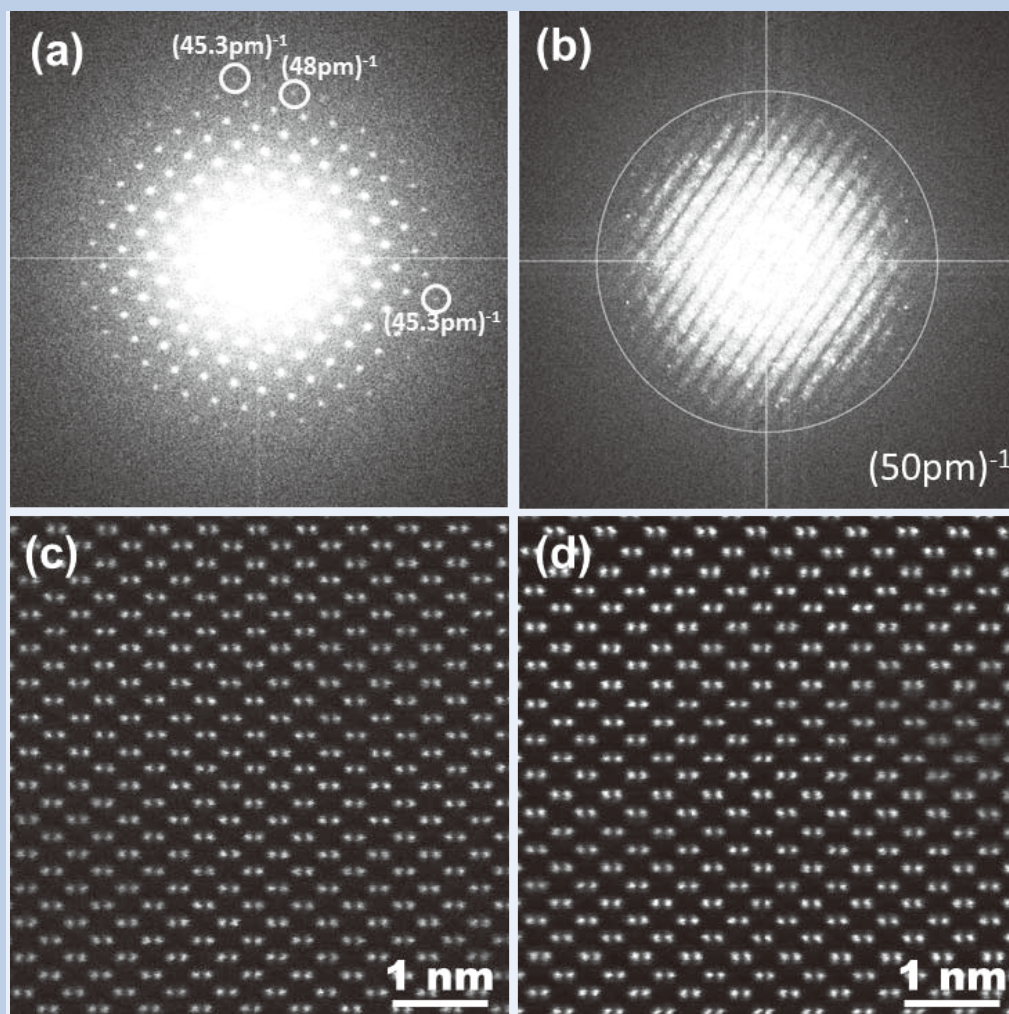


Fig. 5 (a) The power spectrum of Fourier transform from a high-resolution TEM image of Si [110]. (b) Young's fringe test using gold particles on a carbon thick film. (c,d) Si [110] high-resolution HAADF images with an acquisition time of (c) 10 s and (d) 80 s.

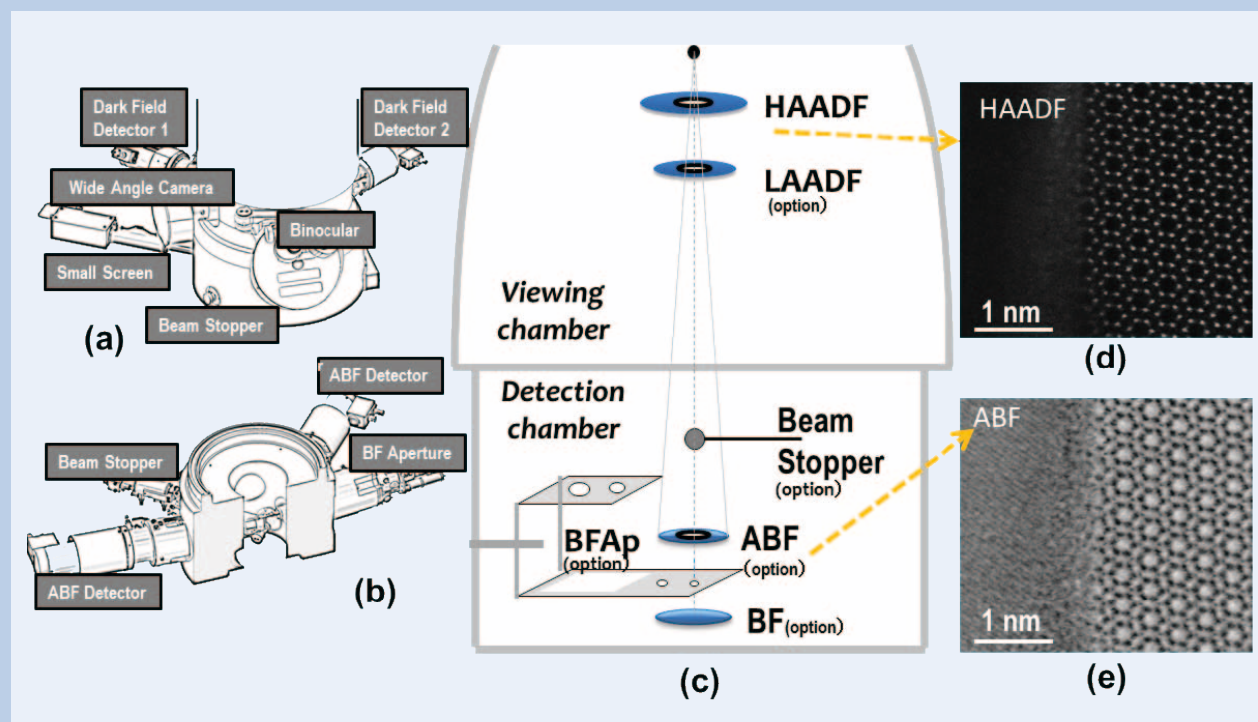


Fig. 6 Schematics of (a) a viewing chamber and (b) a detecting chamber with detectors. (c) Schematic of STEM detector configuration. (d) High-resolution HAADF image of a grain boundary in a  $\beta$ -Si<sub>3</sub>N<sub>4</sub> polycrystalline specimen at 300 kV. (e) High-resolution ABF image.



and bright field (BF) detectors (Fig.6(c)). Four images by these detectors can be obtained, simultaneously. Figure 6(d) and (e) respectively show an example of a HAADF image and a simultaneously acquired ABF image for a specimen of  $\beta$ -Si<sub>3</sub>N<sub>4</sub>. Nitrogen atomic columns are detectable in the image of ABF.

### Wide range of accelerating voltage

The microscope can be operated at wide range accelerating voltages from 80 to 300 kV. **Figure 7**(a-c) show high-resolution HAADF images of a Si [110] crystalline specimen. Observation at 300 kV can image a sub-Ångström structure, as shown later, and 80 kV imaging is useful for soft materials to reduce the specimen damage. At 80 kV, 136 pm imaging is achievable, as shown in Fig. 7 (c). Operation at 160 kV is optionally available (Fig. 7(b)), and 160 kV imaging is useful for a semiconducting material because of high-resolution and relatively small damage compared with higher voltage. Lower voltage operation at 60 kV is optionally available.

### Developed two objective pole pieces and ultra-high resolution imaging

We newly developed two types of pole pieces of an objective lens for the JEM-ARM300F. They are FHP (full high resolution pole piece) for the ultra-high resolution configuration and WGP (wide gap pole piece) for the high resolution analytical configuration. FHP is designed [3] for ultra-high resolution observation with a small chromatic aberration; a chromatic aberration coefficient value of 1.35 mm in STEM/TEM at 300 kV is significantly small. WGP is useful for a high-

sensitive EDS analysis with a large solid angle of the detection, for a thicker special holder owing to a large space inside a pole piece gap, and for a higher tilt angle of a specimen holder compared with FHP.

Ultra-high resolution STEM images were obtained using GaN and Ge crystalline specimen using the developed STEM ETA corrector. 63 pm separation was clearly detected in a raw image (**Fig.8**(a)) and an intensity line profile of HAADF at 300 kV using the FHP pole piece (Fig.8(d)). Spots smaller than (63 pm)<sup>-1</sup> information were detected in the power spectrum in Fig.8 (b).

As a further challenging, a sub-50 pm resolution was imaged in HAADF using the FHP (**Fig. 9**). 47-pm separation was confirmed in the intensity line profile in Figure 9(f) and a spatial information of (47 pm)<sup>-1</sup> was detected in the power spectrum in Fig.9 (d). The microscope of “GRAND ARM” has a capability to image sub-50 pm resolution. The stability must be effective in not only ultra-high-resolution imaging but also robust data acquisition for analysis and structure study.

### Summary

We developed a new atomic resolution microscope of the JEM-ARM300F. The developed cold field emission gun showed ultra-high emission stability due to an ultra-high vacuum around the emitter. Stability of the microscope was confirmed using the power spectrum of TEM images and an atomic-resolution STEM image with a long acquisition time. Ultra-high resolution STEM images were demonstrated, by using the developed corrector and objective lens. The microscope will be new platform for an atomic resolution study.

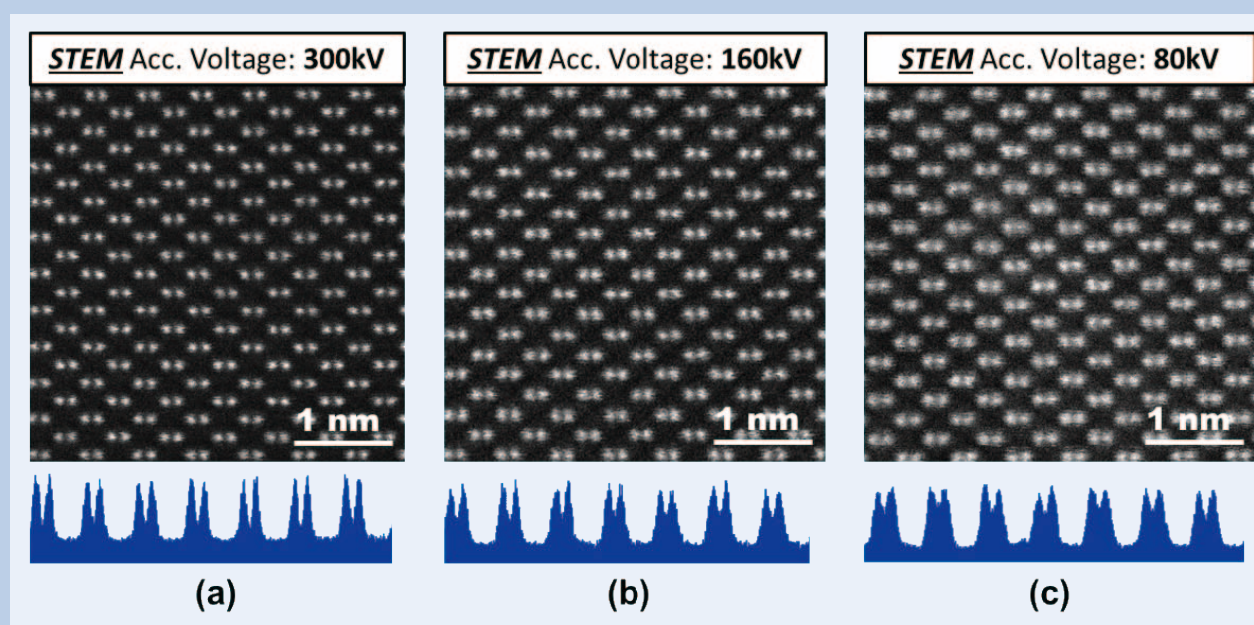


Fig. 7 High resolution HAADF imaging of Si [110] at (a) 300 kV, (b) 160 kV, and (c) 80 kV with their intensity line profiles, as shown at the lower parts.

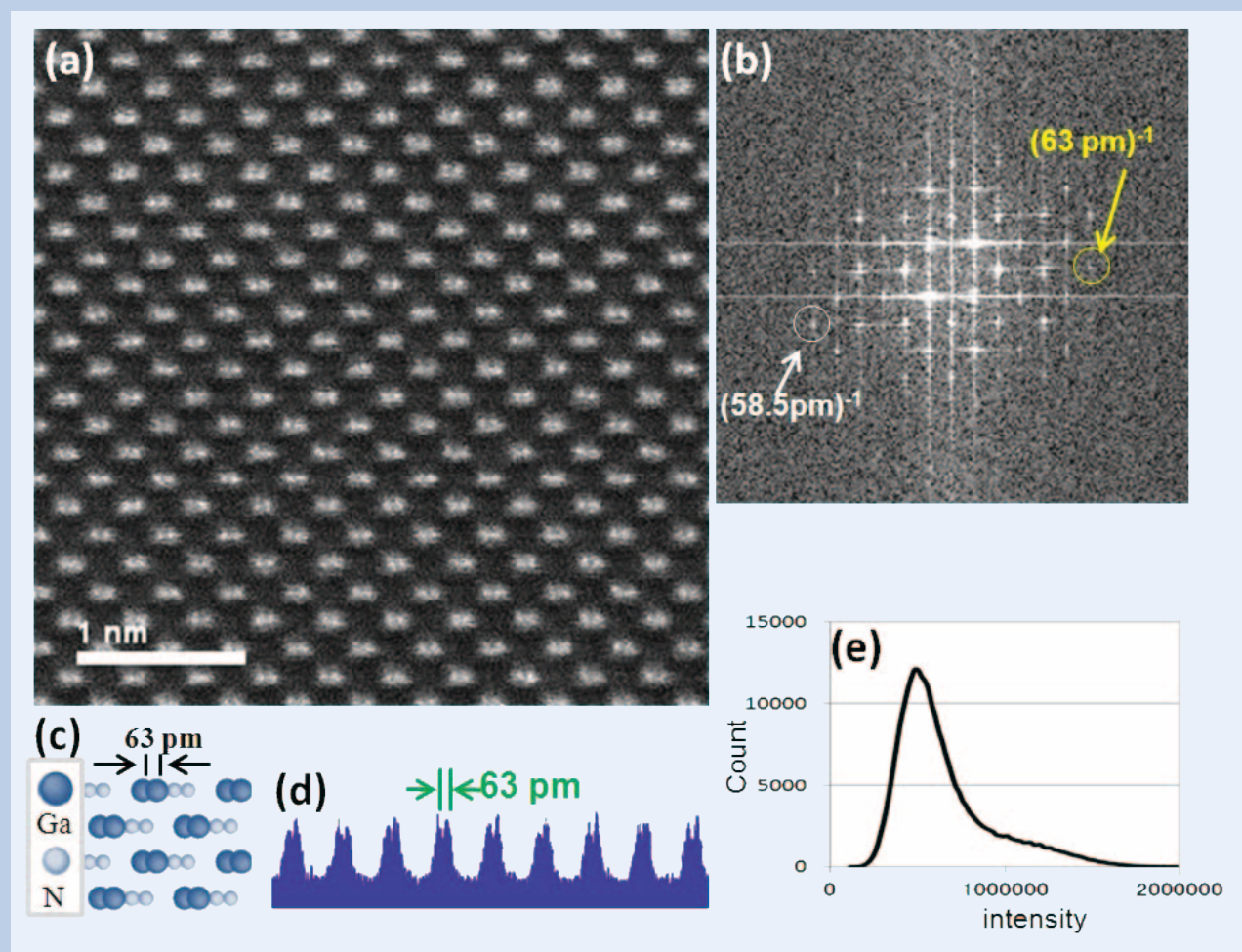


Fig. 8 (a) Raw HAADF image of a GaN [211]. (b) The power spectrum from Fig.8(a). (c) Atomic structure model of a GaN[211]. (d) Intensity line profile of a GaN[211] raw image. (e) Intensity histogram of Fig.8(a).

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