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High Resolution Electron Spectro-Microscope (HRESM)

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A new 1000 kV electron microscope was installed at Uji campus of Kyoto University in 1988 aiming to reveal atomic structure of organic materials, for example, and to resolve carbon-carbon distance chemically bonded in organic molecules. In addition to its highest resolving power this microscope was designed for elemental analysis in a microscopic area by electron energy loss spectroscopy and also spatial distribution of each element in a specimen. Furthermore, a computer-assisted operation system and a data acquisition system were planned for easy operation of the microscope especially in radiation sensitive specimens. This paper describes the total specifications of this machine.

KEY WORDS: High voltage/ High resolution/ Electron microscope/ Electron energy loss spectroscopy/ Energy filtered imaging/ Computer-assisted operation

Introduction

In 1956, Menter succeeded to observe a crystal lattice image, that is, a set of lattice lines composed of molecular array with a definite interval with an electron microscope.[1] This image was resulted from interference of one or two diffracted electron beams with an undiffracted transmitted beam. His success clearly proved the capability of the electron microscope for observing molecular image or atomic image in a crystal. In 1970 Uyeda et al. in Kyoto University have obtained for the first time two-dimensionally arranging molecular images as a projection of three dimensional thin crystal of copper perchlorophthalocyanine.[2] The image was formed by interference of more than 90 diffracted beams with the central direct beam. Because this image has been formed by many diffracted beams, the method is called "many beams synthesis" or "n-beam method" and has widely spread over the electron microscopists in the field of material science and biological science. On the other hand K. Kobayashi, the emeritus Professor of Kyoto University, has claimed the validity of high voltage electron microscope for a higher resolution since 1953 and a 500 kV electron microscope had been installed at Uji campus of Kyoto University by his group aiming at a high resolution at atomic level in 1973.[3] He aimed to resolve individual molecular stem in polyethylene. Accordingly, the ultimate resolution of the electron microscopy was set at 0.15 nm. However, due to high sensitivity of polyethylene to electron irradiation, his attempt was not achieved. However, using this electron microscope Uyeda et al.[4] improved much the molecular image of $CuPcCl_{16}$ than that they reported in 1970. This material is a thousand times resistant than polyethylene. On the image one can clear-

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ly discriminate chlorine atoms and a copper atom of the copper perchloro-phthalocyanine molecule. However, except these heavy atoms the images of other light elements (carbon and nitrogen) were not well resolved because the practical resolution of this electron microscope was 0.15 nm, which is longer than the atom-atom distances of the light elements in aromatic compounds. Based on this experience we have designed a new high resolution high voltage electron microscope having a resolution better than 0.12 nm and installed it at the Uji campus of Kyoto University in 1989.[5] This new electron microscope is called "High Resolution Electron Spectro-Microscope (HRESM)" because it can analyze energy of electrons passing through a specimen. This report describes the specifications and the practical performance of this electron microscope.

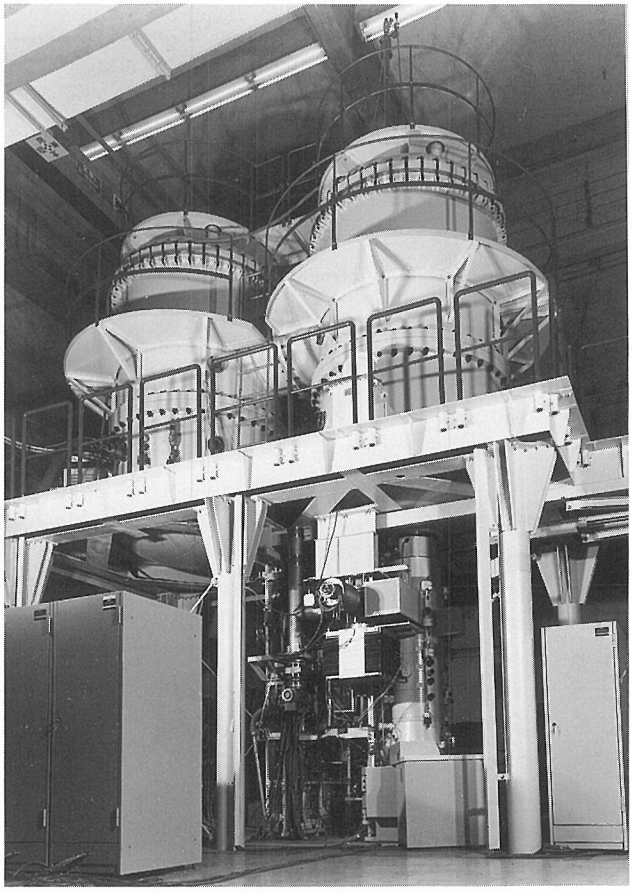

Fig. 1 Total aspect of high resolution electron spectro-microscope (HRESM) in Kyoto University. On the first floor, a large twin-tank system is set up, and on the ground floor, a microscopic system (image formation lenses and image recording systems). Under the ground floor, vibration-proof rubber blocks are arrayed on concrete foundation separated from that of the building.

Apparatus

The total aspect of the electron microscope is shown in Fig. 1. On the ground floor an operation system, image formation lens system and image recording system composed of photo-camera and TV camera are placed, while on the first floor a huge twin-tank is set up with an insulating gas reclaimer in another room. The front tank contains an accelerating tube standing at its center, composed of 25 stages cathodes, and a noise-filter column which rectifies and stabilizes the high tension to suppress the fluctuation of the accelerating voltage less than 1 ppm/min. A Cockcroft Walton type high tension generator and rectifier system are housed in the back tank. To separate the generator and the accelerating tube in two tanks is most important for suppressing the fluctuation in high voltage less than one ppm. In order to avoid the image deterioration due to the mechanical vibrations which come from the ground, the total system weighing about 30 tons, is mounted on a concrete floor of about 170 tons which is supported with 36 rubber blocks arranged on the concrete foundation isolated from the building. Such vibration-proof system has been adopted at our Institute for the previous 500 kV electron microscope and proved the effectiveness of protection from the disturbance.

The lower half of the electron microscope is shown in Fig. 2. At the left a monitor system (M) stands for vacuum, high voltage stability, lens current and stigmators current.

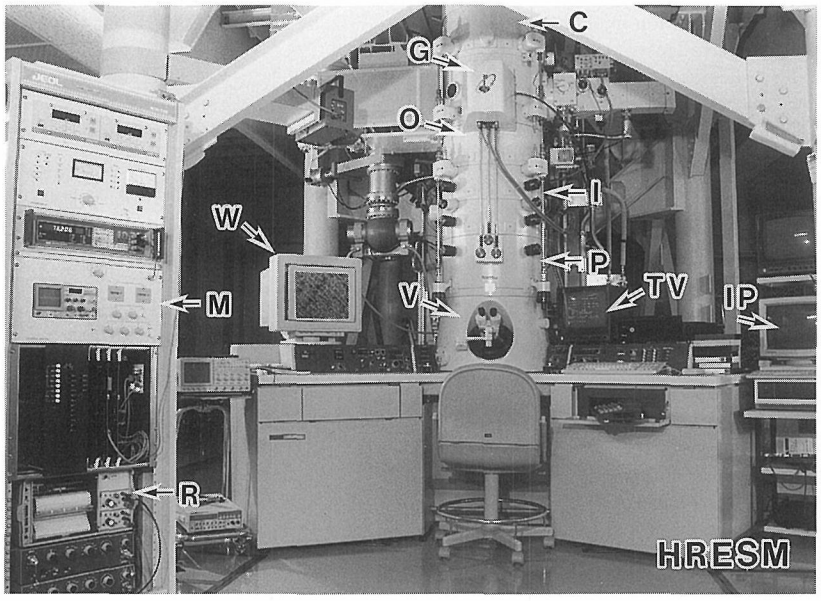

Fig. 2 Lower half of electron microscope. Monitoring system (M), two condenser lenses (C), objective lens (O), intermediate (I) and projection lenses (P). The specimen can be tilted up to 40° in all direction with a goniomter (G). Images are observed on a fluorescent screen in viewing chamber (V) or on a TV monitor (TV), and recorded on photographic films, videotape or imaging plate (IP). Reorder (R) monitors high voltage fluctuation. Sun 4/370 work station (W) also monitors and controlls the state of the EM.

Just below the front tank the magnetic lens system is placed, which is composed of two condenser lenses (C), an objective lens (O), three stages intermediate (I) and a projection (P) lenses. Images or electron diffraction patterns can be observed on a fluorescent screen in the viewing chamber (V) through the binocular or on a TV monitor (TV). The magnification of the electron microscope is changeable in stepwise from 250 to 5 million times on the screen and from 5,000 to 100 million times on the TV screen. The specimen holder is a top-entry type and capable to tilt the specimen at $\pm 40°$ in all directions. Basic specifications of this electron microscope are summarized in Table 1.

Table 1 Specification

High energy electron beam illumination system	JEM-ARM1000
Accelerating voltage (kV)	1000/800/600/500/400
Minimum variable step (V)	250
Stability (at max.Acc.V.)	
Max. fluction voltage	2.0 V/min
Ripple voltage	1.0 Vp.p
Number of acceleration stages	25
Number of generator stages	14
Max. beam current (μA)	25
Condenser lens (CL)	Double CL
Vacuum pump	1000 l/sec SIP
Filament exchange	By airlock
High resolution imaging system	EM-THGZ100
Objective lens	
Polepiece	EM.UHP100
Focal length (mm)	6.0
Spherical aberration coefficient (mm)	1.7
Chromatic aberration coefficient (mm)	3.6
Minimum focus step (nm)	3.0
Max. excitation current (kAT)	42.0
Exciting current stability	1 ppm/min
Theoretical resolution (nm)	0.12
Guaranteed resolution (nm)	0.15
Magnification (steps)	
LOW MAG mode	250-1,200X (8)
MAG mode	1,500-5,000,000X (36)
SA MAG mode	20,000-150,000X (10)
$\mu\mu$ MAG mode	200,000-500,000X (5)
Electron diffraction camera constant (steps)	
SA DIFF mode	1.2-10.0 nm·mm (10)
$\mu\mu$ DIFF mode	0.6-1.0 nm·mm (3)
Specimen chamber	
Number of specimens/load	6
Specimen tile angle	+40°
Specimen movement (X/Y)	+1 mm
(Z)	+3 mm ~ −2 mm

As has become obvious from the many previous works, the resolution of an electron microscope is defined as $\delta = 0.65\,Cs^{1/4}\cdot\lambda^{3/4}$, where Cs is a spherical aberration constant of the objective lens and λ is the wave-length of the electron beam.[6] The resolution limit defined by this equation is called "a Scherzer limit" which represents a point-to-point re-

solution and should be strictly distinguished from a lattice resolution.

The lattice resolution is usually two times higher than the point-to-point resolution but it only represents the mechanical and electrical stabilities of the electron microscope and is meaningless practically in investigation of structure images produced by the coherent interference among many diffracted beams including the undiffracted primary beam. On the basis of a computer simulation, the objective lens was designed to have the spherical and chromatic aberration constants of 1.7 mm and 3.6 mm, respectively, so that the resolution of 0.12 nm can be expected. In order to determine experimentally the value of Cs according to the method proposed by Bourret, we have measured the dependence of image shifts of a small gold particle on the defocus value of the objective lens. The experimental detailed procedure is described in Ref. 7. The results are shown in Fig. 3, from which the Cs-value is obtained as 1.6 ± 0.1 mm. This value promises really the ultimate resolution of 0.12 nm at 1000 kV ($\lambda = 0.00087$ nm) when the fluctuation in the excitation current of the objective lens and the stability of high voltage are sustained to less than 1 ppm/min. The chromatic aberration coefficient was measured to be $C_c = 3.6$ mm by measuring a focus change corresponding to a change of accelerating voltage.

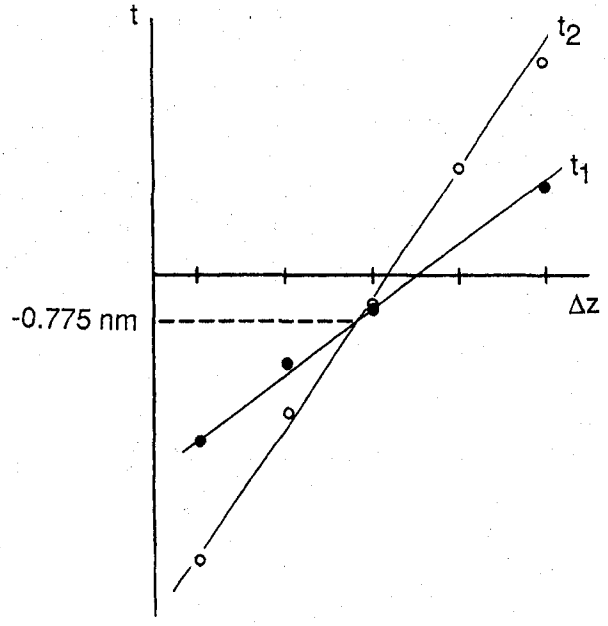

Fig. 3 The Cs was determined through image shifts (t) of a small gold crystal by changing the focus in the vicinity of Scherzer focus. The defocused images due to the 200 (t_1) and 400 (t_2) reflections were used for the measurement.

The fluctuation of accelerating voltage is always monitored on a synchroscope, digital voltmeter or on recording chart on the monitor console standing at the left side of the operation desk. Such an example of monitored voltage fluctuation is shown in Fig. 4, which was measured at the bottom of the resister-condenser (RC) monitor column or the divider column in the accelerator tank. The results confirm the enough stability in high voltage for

obtaining high resolution images.

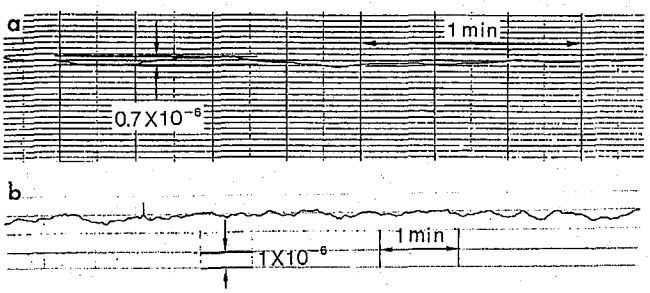

Fig. 4 Stability of high voltage measured under the resister-condenser monitor column(a) and under the devider column(b) in the accelerator tank. Both show enough stability better than 1 ppm/min.

The contrast of a high resolution image of a thin organic specimen is a so-called "phase contrast".[8] The electron wave passing through the specimen suffers a small amount of phase shift due to the interaction with innerpotential of the specimen. The resulting phase difference relative to the unscattered primary wave produces the image contrast reflecting the potential distribution in the specimen. The phase transfer function of the objective lens, which is inherent in each lens and depends on a spherical aberration constant, a defocus value and a wave-length, represents the relationship between the phases of the electron waves on the diffraction plane. As an example, the transfer function at the optimum focus, Scherzer focus, is shown in Fig. 5. At the Scherzer focus the maximum resolution is realized in the true sense of structure image. The Scherzer focus is a focus for which the phase transfer function is close to unity for the widest possible range of the spatial frequency.

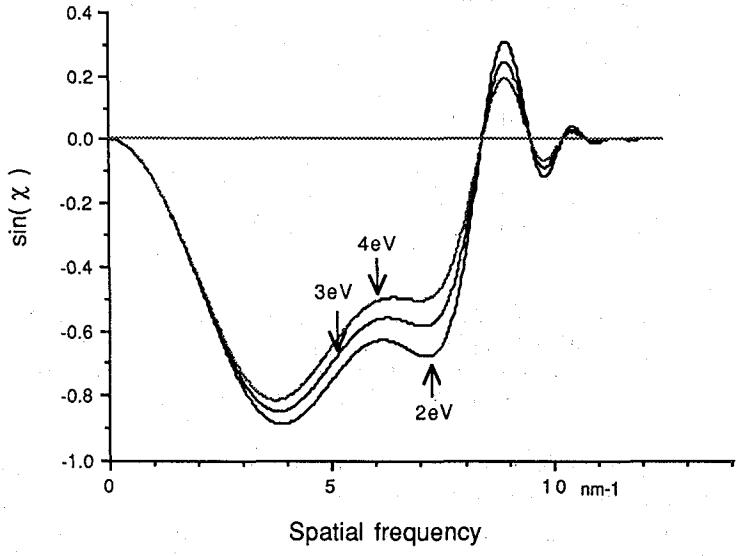

Fig. 5 Transfer function of HRESM, $\sin(\chi)$, with consideration of the total instability of high voltage and current of objective lens.

The function, however, is attenuated strongly at higher spatical frequencies region owing to the instability in lens current and in accelerating voltage both of which result in small spread in defocus values.[9,10] The deterioration in coherency among the diffracted electron waves and the undiffracted wave also attenuates the transfer function in the same way with the electrical instability. Even when all of these factors affecting the attenuation of the transfer function are taken into consideration for calculation of the function, the ultimate resolution of this electron microscope is proved to be 0.12 nm at best as shown in Fig. 5.

This high performance is experimentally proved by a Fourier transform of images of thin amorphous specimen taken with the optimum condition. The Fourier transform can be made by using a computer or by optical method as a diffractogram from the image. In the case of the optical method, laser beam illuminates on a photographic film recording a high resolution image of thin amorphous germanium film so that the laser beam is diffracted and forms a diffractogram on the diffraction plane, which clearly represents the power spectrum of the image as a function of the spatial frequency preserved on the film. The laser diffractogram shown in Fig. 6 (b) was obtained from the image (a) taken with the HRSEM at 1000 kV with a direct magnification of 300,000. It shows clearly that the spatial frequency recorded on the film reaches $(0.12\,\mathrm{nm})^{-1}$ or higher. The diffraction spots found in the diffractogram came from small gold particle deposited on the Ge-film and their lattice spacings were used as a standard to scale the camera length of the diffraction apparatus.

The resolving power of this EM defined by lattice resolution, i.e., line-to-line resolution, was confirmed by observing the crystal lattices. When the small gold particles were observed at a magnification of 500,000 times, lattice spacings of 0.102 nm corresponding to (400) plane of gold crystal were observed.

At the lower part of the microscope column image-recording system is placed. Adding to a usualy electron microscope film, newly developed recording medium, i.e., imaging plate (IP), can also be used with the camera system of the microscope.[11] The imaging plate is said to have a 10,000 times higher sensitivity and a wider dynamic range of four orders of magnitude for electron beam intensity than usual electron microscope films. We have investigated the characteristics of the imaging plate and reported in Refs. 12,13. Only the results are introduced here. The diagram of the system is shown in Fig. 7. The imaging plate is commercially supplied by Fuji-Photo Films Co., type DL-UR_{III}, with a photo-excitable phosphor layer with thickness of 140 μm. The emitted photo-stimulated luminescence light activated by scanning He-Ne laser was read as out-put signal from each 50×50 μm pixel area by the reader over 75 mm $\times$ 100 mm. The emitted light intensity is proportional to the intensity of electron beam irradiated on the plate. A work-station, HP-9000EWS, controls the reading system and data processing.

Though the sensitivity of the imaging plate is proved to be higher by four orders of magnitude than that of the usual electron microscope films, the detection efficiency of the plate decreases with increasing electron energy as the case of photographic films.

The resolving power of a recording medium is another important factor to be investigated. The resolving power was measured by gold-wire method and determined to be about 100 μm at 1000 kV, while those of the electron microscope film was about 25 μm. The modulation transfer function showed the disadvantage of the imaging plate to the electron microscope film. In short, the advantage of the imaging plate is that it provides

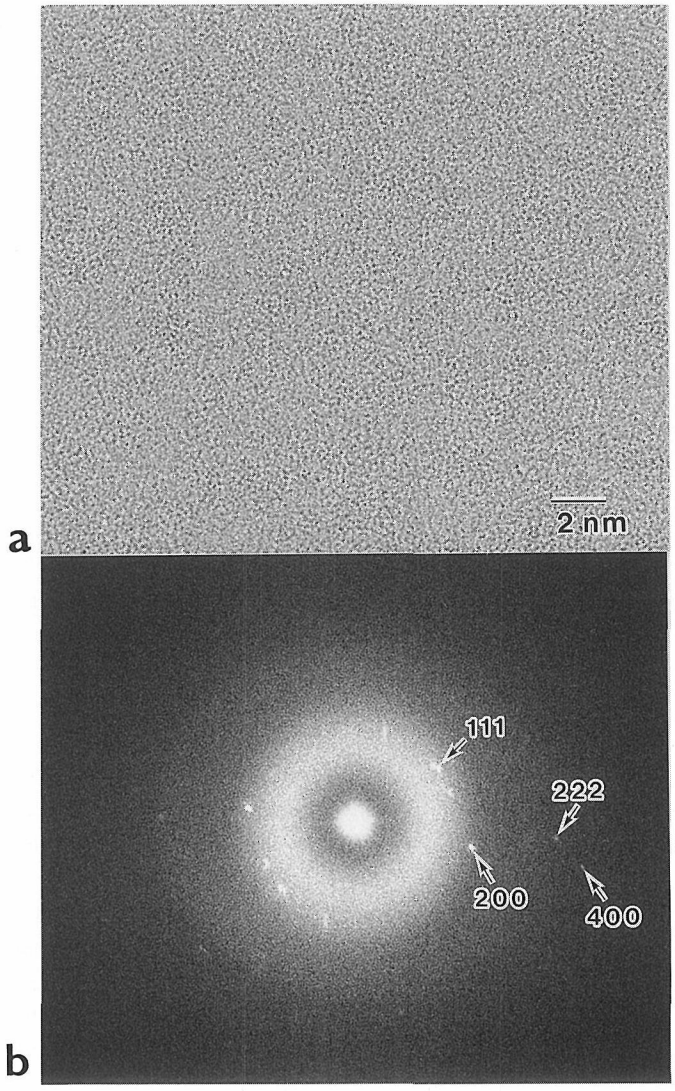

Fig. 6 High resolution image of thin amorphous Ge film taken near the Scherzer focus with the HRESM at 1000kV (a) and its optical diffractogram (b). The first and the second bands of the transfer function are observed in addition to many diffraction spots from small gold particles deposited on the film.

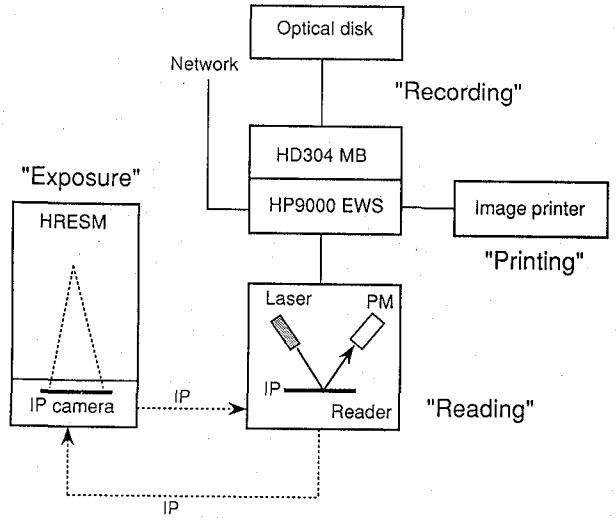

Fig. 7 Imaging plate system. After IP is exposed to electron beam in an electron microscope, photo-stimulated luminescence light activated by laser scanning is read out by photomultiplier (PM) in the reader. The digital data can be stored in a hard disk (HD, 304MB) or in a optical disk. A work station (HP-9000EWS) controls the reading, data processing and image printing systems.

directly digital data of electron beam intensity and responds to the wide range of exposed electron dosage. These characteristics are useful for measuring the electron diffraction intensities and, therefore, are available for crystal structure analysis by electron microscopy combined with the electron diffraction method.[14)]

Under the camera chamber, TV-camera is placed through which one can observe on-line images magnified 20 times larger than those on film or fluorescent screen and can record it on video-tape or on work station as digital data through a video interface board.

The whole system is connected to a super-computer and KUINS with optical fiber (FDDI) or ethernet as shown in Fig. 8 so that the data acquisition and processing are also performed quickly.[15)] Especially the image-data processing is an indispensable process for obtaining a high resolution image of organic or biological specimens because these materials can not tolerate the electron irradiation on specimen required for recording the image at high magnification under a detectable amount of electron dose for the recording system. With less electron dosage the image recorded is noisy and has low signal-to-noise ratio. Such image is interpretable only after the image is well processed. The data acquisition is done through TV-system or imaging plate system. The correction of astigmatisms of the objective lens or of other lens system are also made on-line by using a Fourier-transform of an image, which is only possible with the help with a fast computer system.

The electron spectrometer equipped to the electron microscope can analyze the energy loss of electron passing through the specimen.[16)] An image formation lens system attached to the spectrometer will make it possible to reveal a spatial distribution of each element in the specimen and the system is now under investigation.

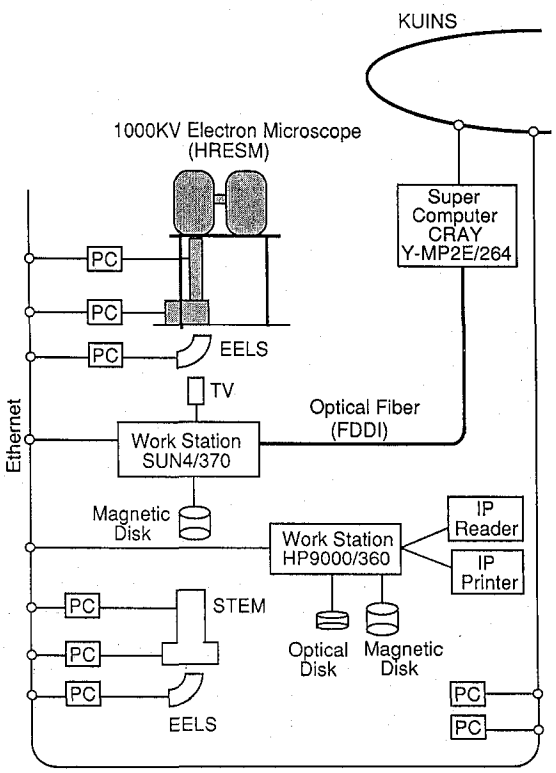

Fig. 8 Whole network system in our laboratory connected with KUINS and the super-computer laboratory in the Institute. Data acquisition can be performed by TV systems or IP system and send to the computer through the FDDI (Fiber optical cables) to Cray Y-MP2E/264 through a work station SUN 4/370.

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