Development of the Electron Microscope in Japan

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Abstract:

This article is about how Japan has attained today's top position in electron microscope manufacturing. My observation is that it is simply because of

(1) cooperative, and at the same time, competitive, alliances among universities and industries for R&D, and
(2) from industry's side, continuous and determined efforts to reach the theoretical limit, disregarding the short term efficiency.

Since the first electron microscope, showing higher resolution than the optical microscope, was constructed by E. Ruska and B. von Borries at Siemens more than a half century ago, and due to its high resolution and analytical capabilities, the electron microscope has become the essential research tool for the biological and material sciences.

During the same time, as is well known, Japan became its largest manufacturer country, and perhaps less well known, also its largest user country in the world.

According to our estimation, in 1988 alone, more than 2200 units of the electron microscope were produced in the world, excluding Communist countries: about 500 units of transmission microscope and about 1700 units of scanning microscope. In this estimation, small microscopes such as simple scanning and the biggest ones such as 1000kV microscopes, are both counted as units. Of these numbers, about 70-80% were manufactured, and 40% installed, respectively, in Japan.

Why and how could Japan reach such a position? Though there are many products in which Japan occupies the leading position in manufacturing today, the high share of the world's electron microscope market is quite unique compared to other sophisticated scientific instruments.

In this paper I would like to analyze briefly the background of what happened in this half century, by using mostly firsthand facts, due to my privilege of being one of the founders of JEOL Co. Ltd.

1) Japan was an early starter, though not an inventor.

According to the record, as soon as the report of E. Ruska and B. von Borries[1] arrived in Japan, it had a strong impact on the Japanese scientific community. I was too young at that time, and therefore, I was not a member of that circle. However, from my point of view, it is unbelievable that such an ambiguous photograph of coli bacterium created such enthusiasm in Japan, which at that time was embittered and poverty-stricken by the war.

The 37th subcommittee was created quickly the next year, 1939, in the Japan Society for the Promotion of Science. This subcommittee consisted of 15 members from universities, national research institutes and research centers of big industries such as Hitachi and Toshiba, etc. The research project of this subcommittee was to study and construct the electron microscope and to promote its application in three years with a budget of 80,000 Yen. Taking into consideration the inflation rate, this is roughly equivalent to one million of today's dollars. The effect of this encouragement was really astonishing. When the 2nd World War ended in 1945, nearly 20 electron microscopes were working throughout Japan. The level of these instruments and their applications were far behind compared to Germany's and the U.S.'s. However, the basic knowledge was there which was necessary to appreciate the abundance of information which was pouring in by much world literature.

It is very interesting to see that the example of a cooperative R&D program existed even at that time.

When the original JEOL company was created in 1946 for the sole purpose of producing the electron microscope, we did not know of the existence of this official organization. However, JEOL was certainly supported during this most difficult time by the continuing enthusiastic atmosphere of the Japanese community.

2) Though we started from an inferior position, we have continued to reach toward the highest theoretical limit of resolution, even when we could not find any real application for it.

When the war ended in 1945, I believe that Siemens' microscope had already reached a resolution of 10-20Å. I was able to witness this fact with my own eyes when I was fortunate enough to attend the 3rd International Conference of Electron Microscopy in London in 1954. The first introductory report was presented by B. von Borries, who was one of the inventors of the electron microscope, but, unfortunately, he died a few years later of a head tumor. Fig. 1 and Fig. 2[2] are the copy of his first presentation. It is interesting that 12 manufacturers existed in the world at that time, including 4 Japanese, one of which was ours, Japan Electron Optics. As a matter of fact, the name of JEOL comes from our original name, J̲apan E̲lectron O̲ptics L̲aboratory Co., Ltd.

The point I would like to mention is that only Siemens' Elmiskop showed a resolution of about 10Å, and no others. And this was clearly demonstrated by the crystal lattice photograph of phthalocyanine taken and published by Menter[3] which was shortly taken with Siemens Elmiskop 1. This was the opening of the new era of the high resolution microscope and I would like to follow the development of the resolution up to today, using this same phthalocyanine. The molecular structure of the phthalocyanine is shown in Fig. 3. In this special case, the center position is Cu and sixteen marginal CH bonds are replaced with C-CL bonds. When this material is vacuum condensed on the cleavage surface of sylvin single crystal, the epitaxial growth gives rise to thin crystallites just as shown in Fig. 4, which are suitable for electron microscope observation. The first photograph, Fig. 5, shows the above mentioned materials, which is similar to that published by Menter, though we took it several years later. This reveals the line structure of 13Å separation, but no details concerning the atom configuration. Next Fig. 6 does show the diamond-shaped molecular image though still does not reveal the atom structure.

Fig. 7 begins to show the atom structure and Fig. 8 clearly illustrates the position of Cu at the center and 16 CL atoms. Photographs from Fig. 5 to Fig. 8 were obtained by N. Uyeda, T. Kobayashi, and E. Suito(4), together with JEOL's people. The time span from Fig. 5 to Fig. 8 was nearly 30 years, and the resolution of the electron microscope which took Fig. 8 was 1.4Å. This means that the improvement of the resolution from 10Å - 1.4Å needed 30 years of continuous effort.

Unfortunately, we don't see the same kind of effort from the front runners' group of 1954. This is somehow quite understandable and reasonable because of the difficulty of making thin film material and because of the wider use of the electron microscope for biological applications, higher resolution than 10Å was considered unnecessary for many years. But when the time finally came for direct atom observation, which should be the ultimate goal for the electron microscope, the most ready instrument was Japanese.

I heard that when Sir Hillary was asked why he climbed Mt. Everest, his answer was because it was there. The same philosophy could also apply to the world of science.

A few examples of the Atom image are a single gold crystal in 110 orientation containing a stacking fault shown in Fig. 9 and YBa Cu high Tc Super Conductor containing the local defect shown in Fig. 10.

3) <u>The concept of the analytical electron microscope was established much earlier in Japan than anywhere else.</u>

Because of the influence from the electron diffraction group which had been traditionally very powerful in Japan even at the early stage, the electron microscope was considered to be an extension of the diffraction instrument, as the optics theory implied. Many researchers joined from the electron diffraction field. For them the information obtained with the electron microscope was complementary to diffraction's, and this was the prime reason for their interest in electron microscopy. So naturally, all techniques utilized in the diffraction field were to be transplanted into the electron microscope.

It was not easy for the manufacturers, because the demand for the specimen stage was quite different in both cases and these analytical capabilities in many cases contradicted the conditions needed to obtain high resolution.

However, we had to accept the market need and just had to try. Now we say very often that, here in Japan, we have to accept the principle of "the Customer is god." It seems that even in the electron microscope this philosophy pushed us to modify the instrument accordingly, and inevitably this was much more complicated mechanically, however, it eventually resulted in final success. It should be noted that this happened even under the conditions of:

a) biological market, which did not require any complications besides high resolution and low contamination, has always been much larger that the material market;

b) thinning bulk material, good enough for the transmission electron microscope was not possible until a later day.

Fig. 11 shows the advertisement of our microscope made in the French Journal in 1954. It said besides high resolution, the capabilities of transmission as well as of reflection, together with diffraction and in-situ specimen heating up to 1,000°C. Compared to Fig. 12, which was the Siemens' advertisement of the same time, both were already very different instruments.

Normally the electron microscope means the transmission microscope. In that case then, as I said repeatedly, the bulk material could be studied only by replica and not the material itself, evidently the diffraction of which was not obtainable. However, in the book another type such as reflection method was also mentioned, though not very commonly utilized. I thought perhaps the combination of this method with routinely used reflection diffraction might be very useful for material research and started to market the first commercial instrument, as shown earlier in Fig. 11. Fig. 13 shows an example of the result obtained with this instrument(5). We could have a surface image (a) with diffraction (c) which looks more interesting than the replica image (b) of the same sample. However, this was not successful at that time because the reflection method required another difficult specimen preparation. Practically, the angle of incidence has to be very small, less than 8 degrees, meaning that the surface of the studied sample must be flat by atom scale to obtain useful results, which was out of reach at that time.

Long after, this method was beautifully used for the study of a single crystal of silicon by K. Yagi(6), an example of which is shown in Fig. 14. The specimen is Si(111)-7x7. One step corresponds to one atom layer of 3.1Å and a screw dislocation is pointed to by an arrow. It is quite true that new ideas become really useful only at a much later time.

Fig. 15 is a series of photographs of heating experiments taken by N. Takahashi et al.(7). The specimen is the evaporated film of 50-50 Al-Cu alloy of a bout 500Å thickness. Together with diffraction, we could follow the phase transformation of this material, though we could not fully analyze the image contrast. These were the trials to accommodate electron microscope and electron diffraction in one instrument. The real further step for the electron microscope to be an analytical instrument was realized by the successful combination of scanning capability in the transmission electron microscope.

The modern electron microscope has a very strong objective lens, so that the specimen is deep inside the magnetic field, and the upper half of the objective lens before the specimen acts as a strong condenser lens, as illustrated in Fig. 16. By combining with the condenser lens sitting above the objective lens, one can control the size of the illumination from a minimum of 10Å to the whole area of the specimen. With the addition of a scanning coil vertical to the beam axis, one can scan the finest beam across the specimen. Now the only remaining problem for realizing the scanning image is how to collect the secondary electrons created from the sample. Fortunately, the energy of the secondary electron is so small that they are confined along the axis spiralling upward with 100% efficiency due to the strong magnetic force which can be collected by the photomulitplier tube by easily adding the proper potential. This device was patented by us[8] and we started to commercialize the analytical electron microscope, the schematic diagram of which is shown in Fig. 17. Around the specimen, sitting at the center, there are detectors for the secondary electron image (SEI), backscattered image (BEI), two detectors for the energy dispersive X-ray spectrometers, high angle EDS and ultra thin window EDS, and at the bottom, a fluorescent screen for the transmission electron microscope image (TEM image) and several types of diffraction, such as selected area diffraction (SAD), micro beam diffraction (MBD) and convergent beam diffraction (CBD), and scanning transmission electron microscope (STEM) and electron energy loss spectrometer (EELS). TV monitor is also applicable. Some applications are shown in the following figures. Fig. 18 is three photographs of copper oxide powder; (1) secondary electron image, (2) transmission image and (3) replica with transmission. Fig. 19 is the EDS image together with the TEM. The above is the Fe distribution along the line scan and the lower shows the Fe distribution image. Fig. 20 is the EELS image of Boron Nitride.

All these capabilities together with computer image processing, are the modern electron microscope, which we have created step by step in these 30 years.

4) Close cooperation network has been established with worldwide top scientists.

In order to develop the instrument by the philosophy of "Market is the God," we have to know the market. In this respect we have had constant good relations with many electron microscopists throughout the world, to whom I express my sincere appreciation. Some I have referred to in this paper, and though I cannot mention all of them, just to show that we are operating internationally, I would like to mention some who are non-Japanese and who personally gave me useful advice and opportunities in the new world: Prof. B. von Borries, Prof. J. Trillat, Prof. G. Dupouy, Sir. P.B. Hirsh, Prof. J.M. Cowley, and Prof. G. Thomas.

5) Conclusion

In these past 50 years, the electron microscope has been metamorphosed to the very complete analytical instrument. The first instrument we manufactured is shown in Fig. 21 and one of the latest, 1,000 kV electron microscope, in Fig. 22. The difference is really unbelievable. Now, how about the future. Though we have nearly reached the limit of resolution, about 1.0Å, there are still many demands being presented for the application of electron beam, either as an analytical tool or as a fine manufacturing machine. I personally believe that there will be no time yet to be able to rest on past accomplishments.

Reference

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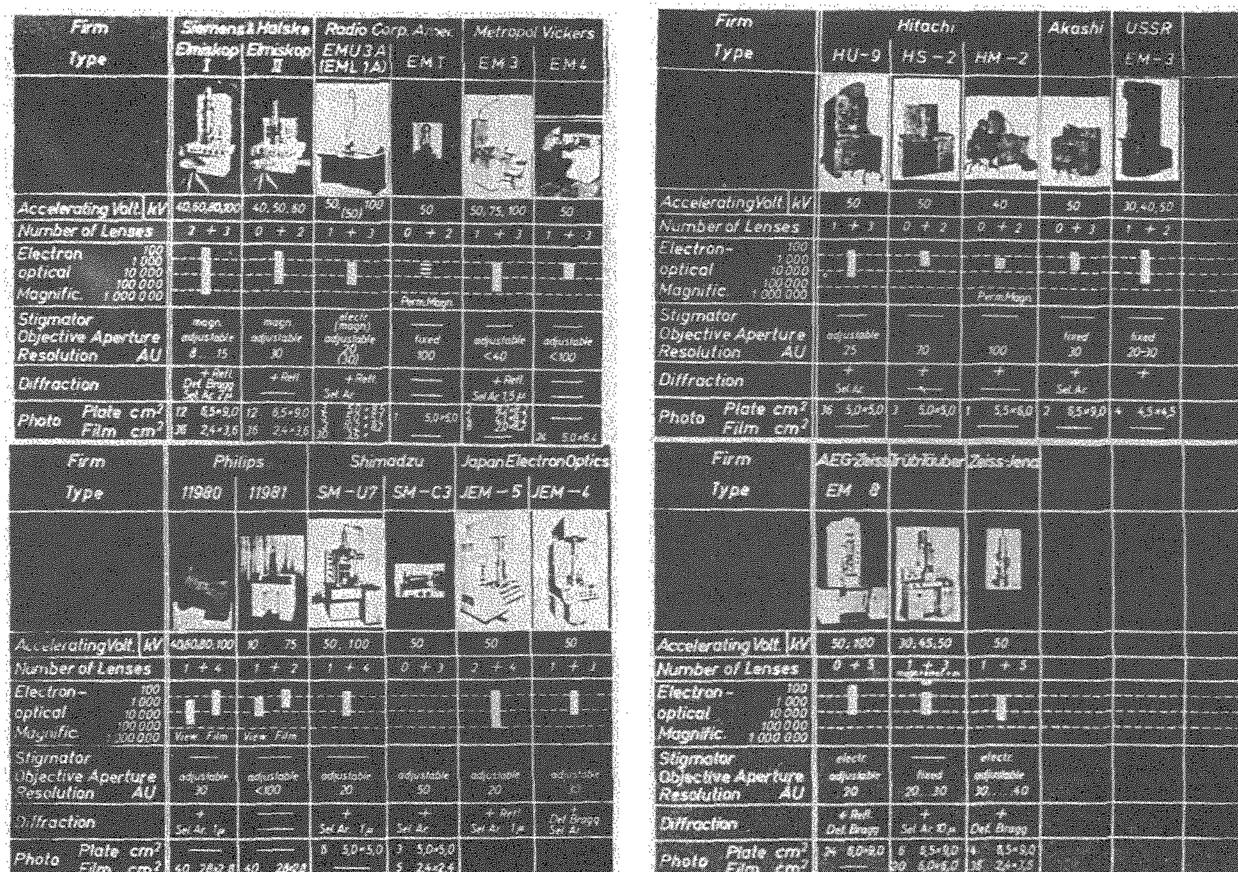


Firm	Siemens & Halske		Radio Corp. Amer.		Metropol Vickers	
Type	Elmiskop I	Elmiskop II	EMU 3 A (EML 1 A)	EMT	EM 3	EM 4
Accelerating Volt. kV	40, 60, 80, 100	40, 50, 80	50 (50) 100	50	50, 75, 100	50
Number of Lenses	2 + 3	0 + 2	1 + 3	0 + 2	1 + 3	1 + 3
Electron-optical Magnific. (100 / 1 000 / 10 000 / 100 000 / 1 000 000)				Perm. Magn.		
Stigmator	magn.	magn.	electr. (magn.)	—	—	—
Objective Aperture	adjustable	adjustable	adjustable	fixed	adjustable	adjustable
Resolution AU	8 ... 15	30	20 (50)	100	<40	<100
Diffraction	+ Refl. Del. Bragg Sel. Ar. 2μ	+ Refl. —	+ Refl. Sel. Ar.	—	+ Refl. Sel. Ar. 1.5μ	—
Photo Plate cm²	12 6.5×9.0	12 6.5×9.0	[illegible]	1 5.0×5.0	[illegible]	—
Photo Film cm²	36 2.4×3.6	36 2.4×3.6	36 [illegible]	—	[illegible]	24 5.0×6.4

Firm	Hitachi			Akashi	USSR	
Type	HU-9	HS-2	HM-2		EM-3	
Accelerating Volt. kV	50	50	40	50	30, 40, 50	
Number of Lenses	1 + 3	0 + 2	0 + 2	0 + 3	1 + 2	
Electron-optical Magnific. (100 / 1 000 / 10 000 / 100 000 / 1 000 000)			Perm. Magn.			
Stigmator	—	—	—	—	—	
Objective Aperture	adjustable			fixed	fixed	
Resolution AU	25	70	100	30	20-30	
Diffraction	+ Sel. Ar.	+ —	+ —	+ Sel. Ar.	+	
Photo Plate cm²	36 5.0×5.0	3 5.0×5.0	1 5.5×8.0	2 6.5×9.0	4 4.5×4.5	
Photo Film cm²	—	—	—	—	—	

Firm	Philips		Shimadzu		Japan Electron Optics	
Type	11980	11981	SM-U7	SM-C3	JEM-5	JEM-4
Accelerating Volt. kV	40, 60, 80, 100	10 75	50, 100	50	50	50
Number of Lenses	1 + 4	1 + 2	1 + 4	0 + 3	2 + 4	1 + 3
Electron-optical Magnific. (100 / 1 000 / 10 000 / 100 000 / 1 000 000)	View Film	View Film				
Stigmator	—	—	—			
Objective Aperture	adjustable	adjustable	adjustable	adjustable	adjustable	adjustable
Resolution AU	30	<100	20	50	20	[illegible]
Diffraction	+ Sel. Ar. 1μ	—	+ Sel. Ar. 1μ	+ Sel. Ar.	+ Refl. Sel. Ar. 1μ	+ Del. Bragg Sel. Ar.
Photo Plate cm²	—	—	3 5.0×5.0	3 5.0×5.0		
Photo Film cm²	40 2.4×2.8	40 2.4×2.8	—	5 2.4×2.4		

Firm	AEG-Zeiss	Trüb Täuber	Zeiss-Jena			
Type	EM 8					
Accelerating Volt. kV	50; 100	30, 45, 50	50			
Number of Lenses	0 + 5	1 + 3	1 + 5			
Electron-optical Magnific. (100 / 1 000 / 10 000 / 100 000 / 1 000 000)						
Stigmator	electr.	—	electr.			
Objective Aperture	adjustable	fixed	adjustable			
Resolution AU	20	20 ... 30	30 ... 40			
Diffraction	+ Refl. Del. Bragg	+ Sel. Ar. 10μ	+ Del. Bragg			
Photo Plate cm²	24 6.0×9.0	6 6.5×9.0	4 6.5×9.0			
Photo Film cm²	—	30 6.0×6.0	35 2.4×3.5			

Fig. 1 and Fig 2 From the introductory paper of B. von Borries at the International Conference in London, 1954.

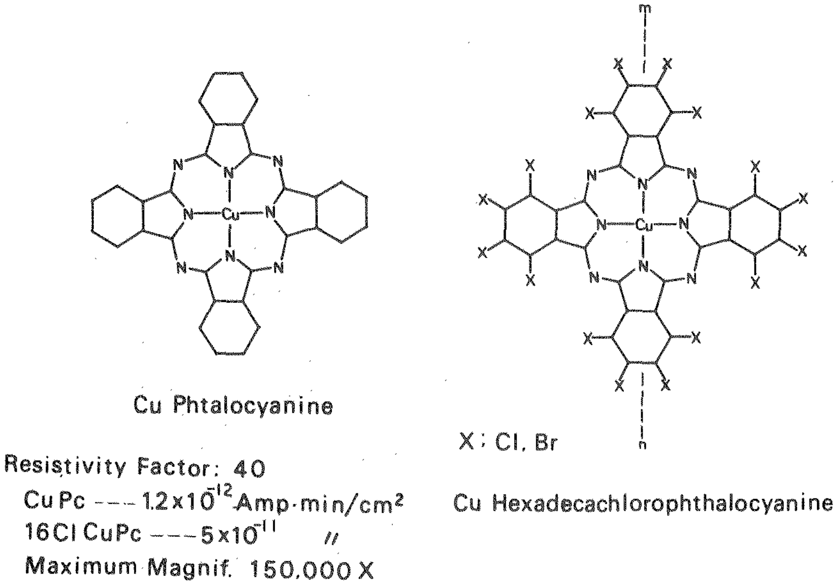


Fig. 3 Molecular structure of Cu-Phthalocyanine. From Fig. 3 to Fig. 8, courtesy of N. Ueda.

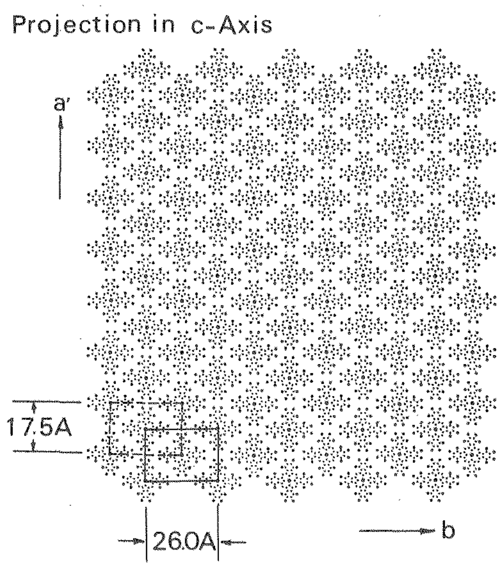


Fig. 4 Crystallized phthalocyanine observed in the electron microscope.

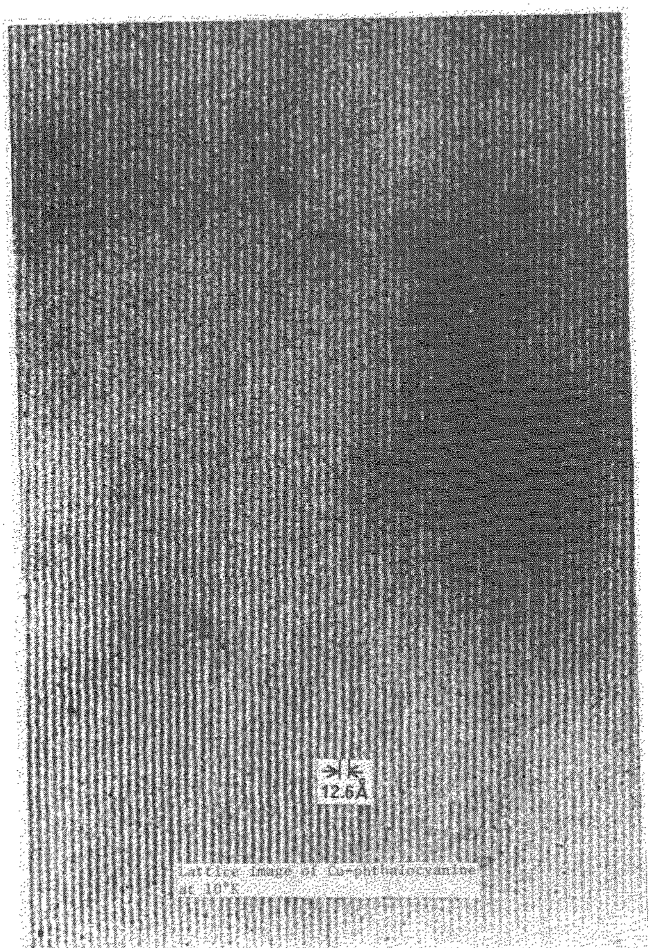


Fig. 5 Cu-phthalocyanine in the 1950s

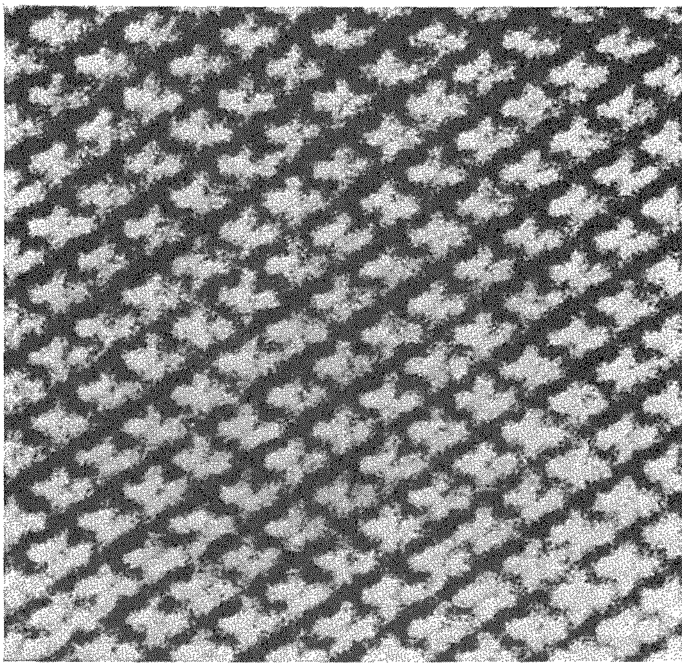


Fig. 6 Cu-phthalocyanine in the 1960s

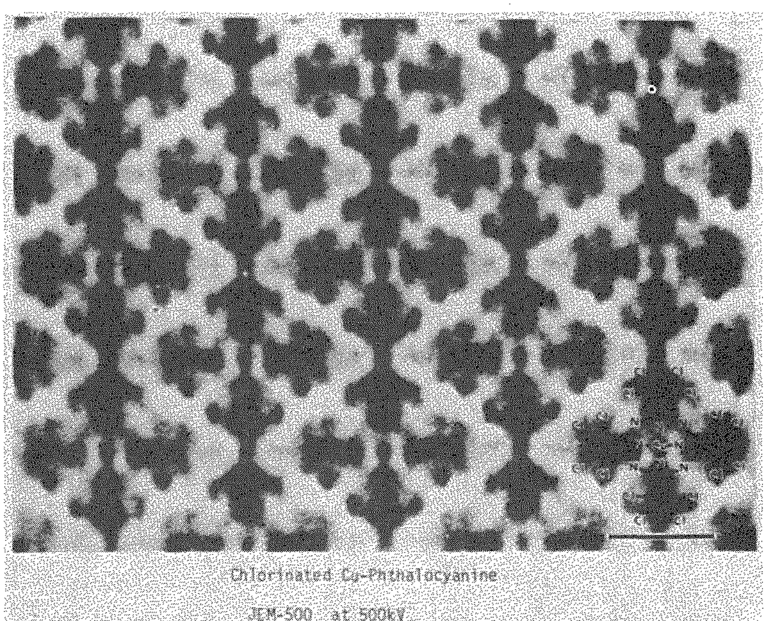


Fig. 7 Cu-phthalocyanine in the 1970s

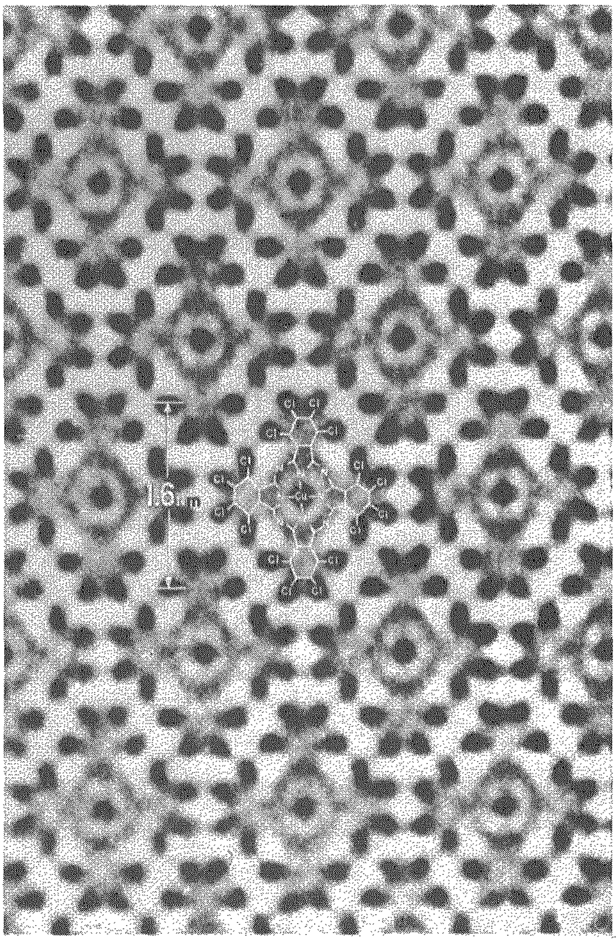


Fig. 8 Cu-phthalocyanine in the 1980s

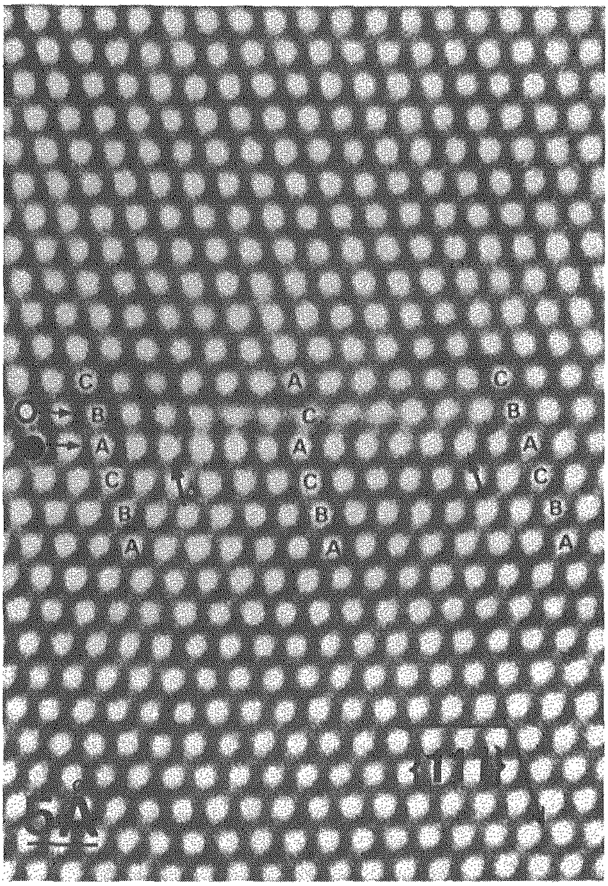


Fig. 9 Single gold crystal containing stacking fault, courtesy of H. Hashimoto.

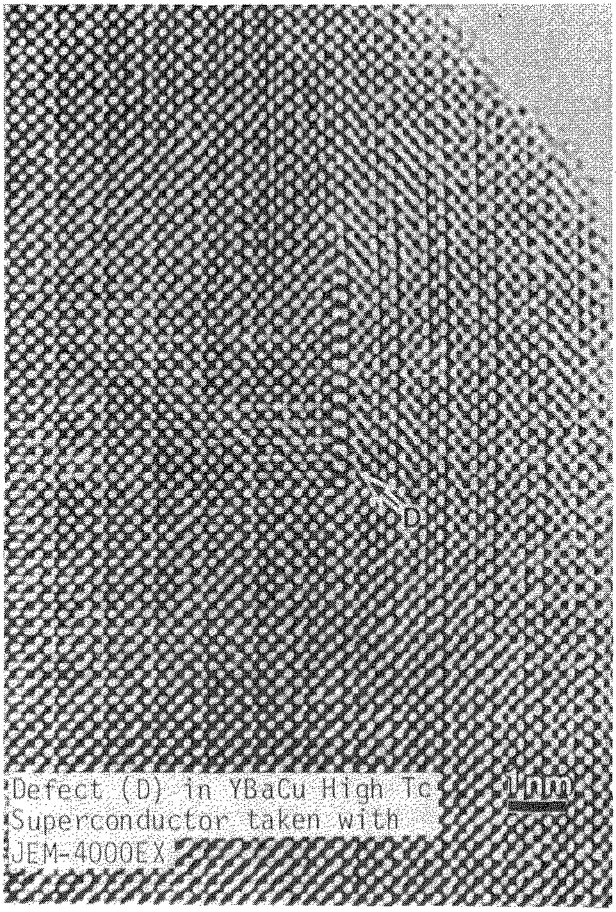


Fig. 10 YBaCu high Tc Super conductor, courtesy of K. Hiraga.

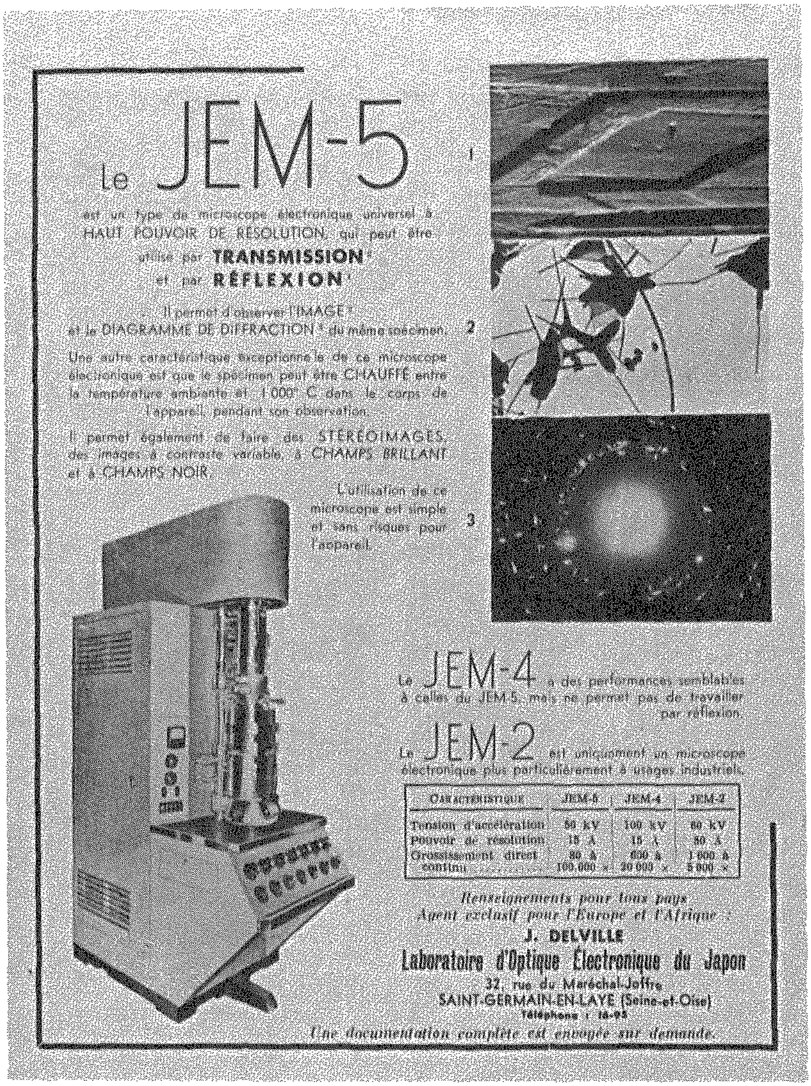


Fig. 11 Advertisement of JEOL in 1954

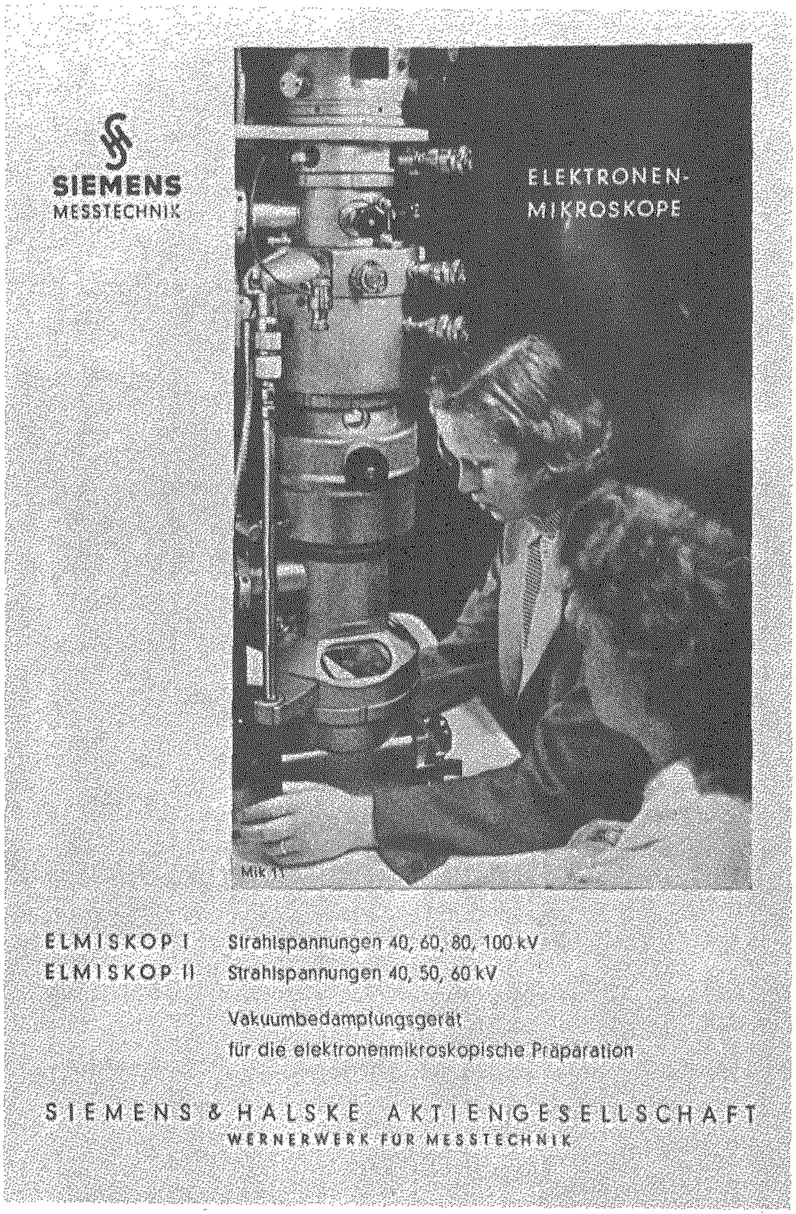


Fig. 12 Advertisement of Siemens in 1955

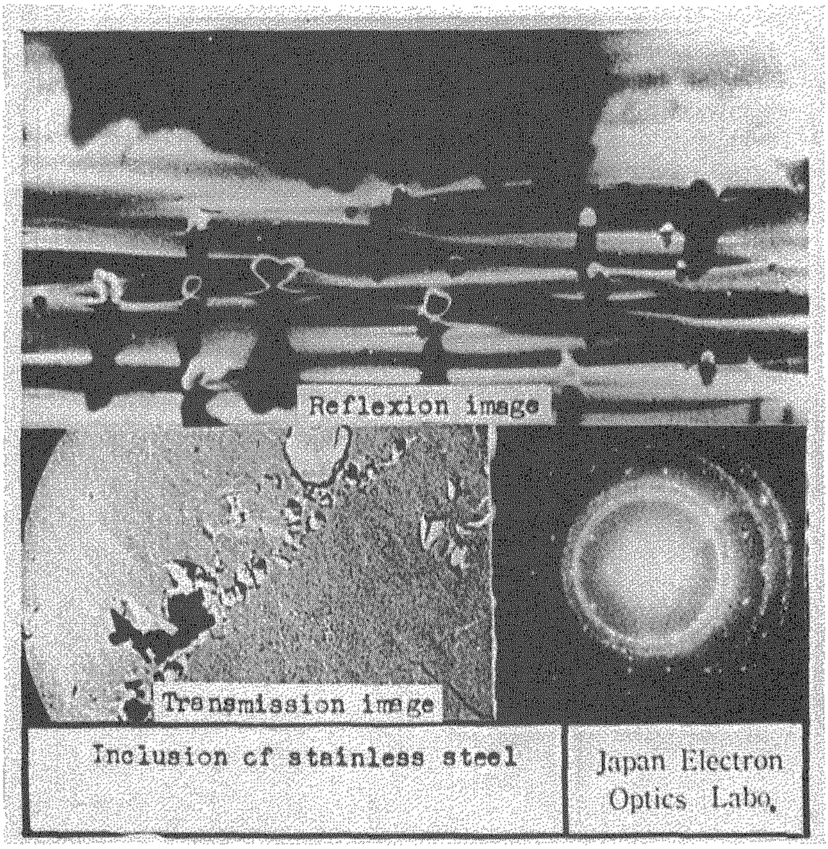


Fig. 13 Reflection microscopy compared to replica method.

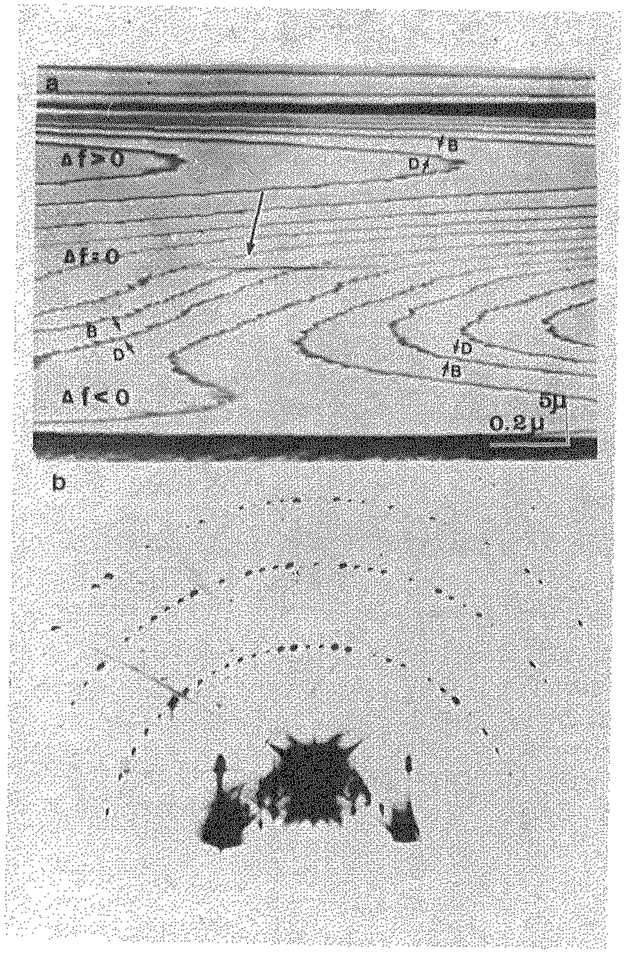
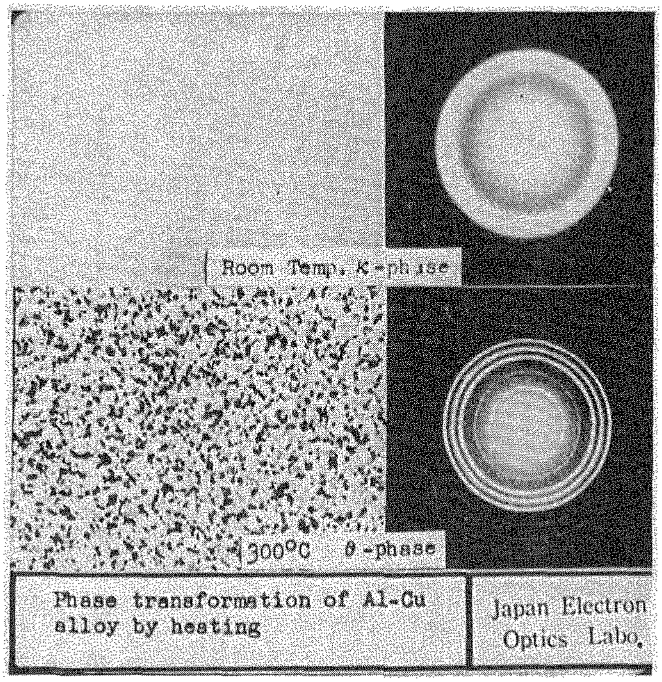
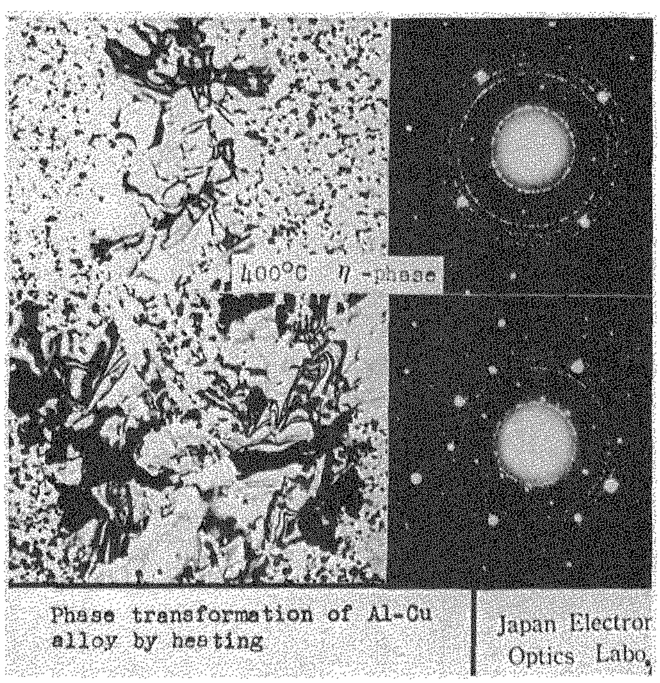


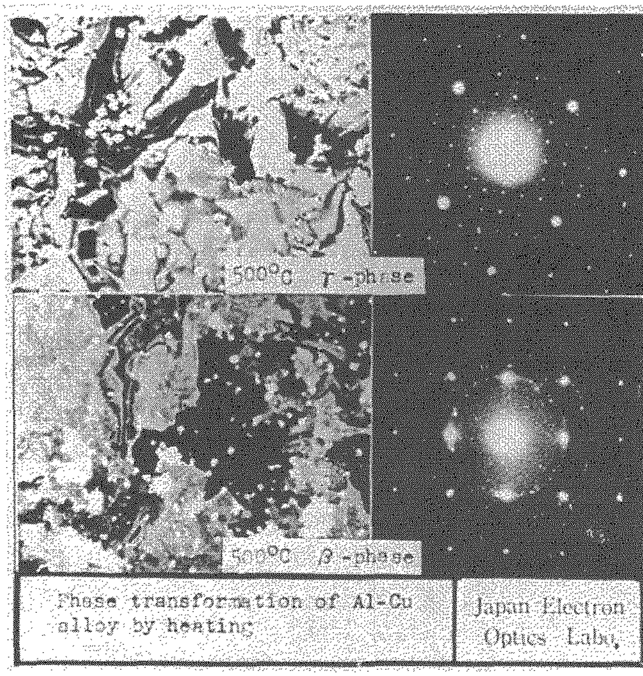
Fig. 14 Surface of Si observed by reflection method, courtesy of K. Yagi.


Room Temp. κ-phase
300°C θ-phase
Phase transformation of Al-Cu alloy by heating
Japan Electron Optics Labo.

(a)

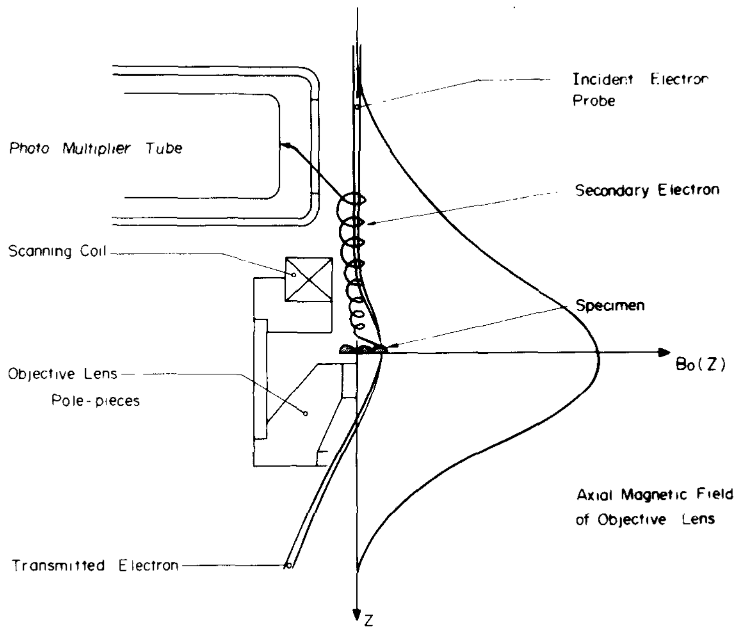

400°C η-phase
Phase transformation of Al-Cu alloy by heating
Japan Electron Optics Labo.

(b)



(c)

Fig. 15 (a) (b) (c) 50-50 Al-Cu alloy phase transformation courtesy of N. Takahashi.



Schematic Illustration of Secondary Electron

Fig. 16 Schematic illustration of secondary electrons' behavior in the objective lens.

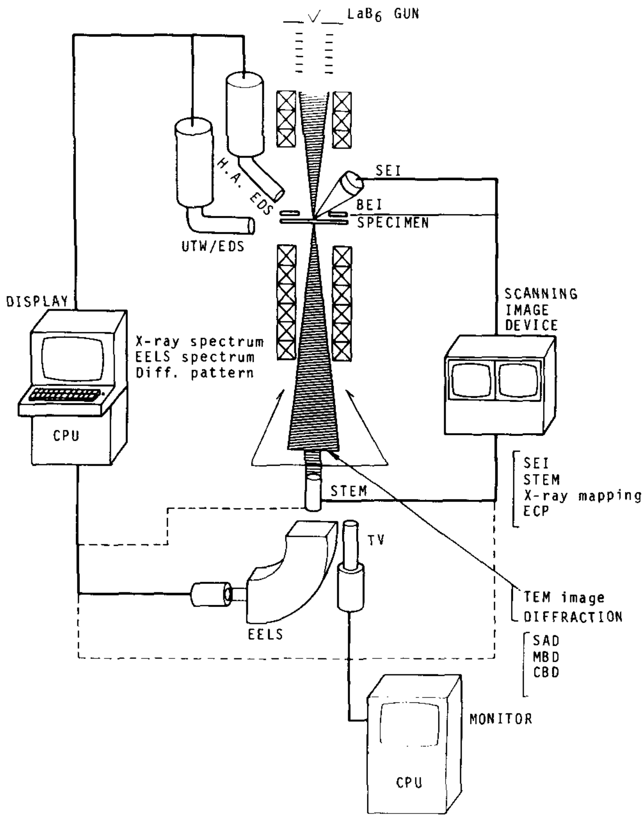


Fig. 17 Schematic illustration of analytical microscope.

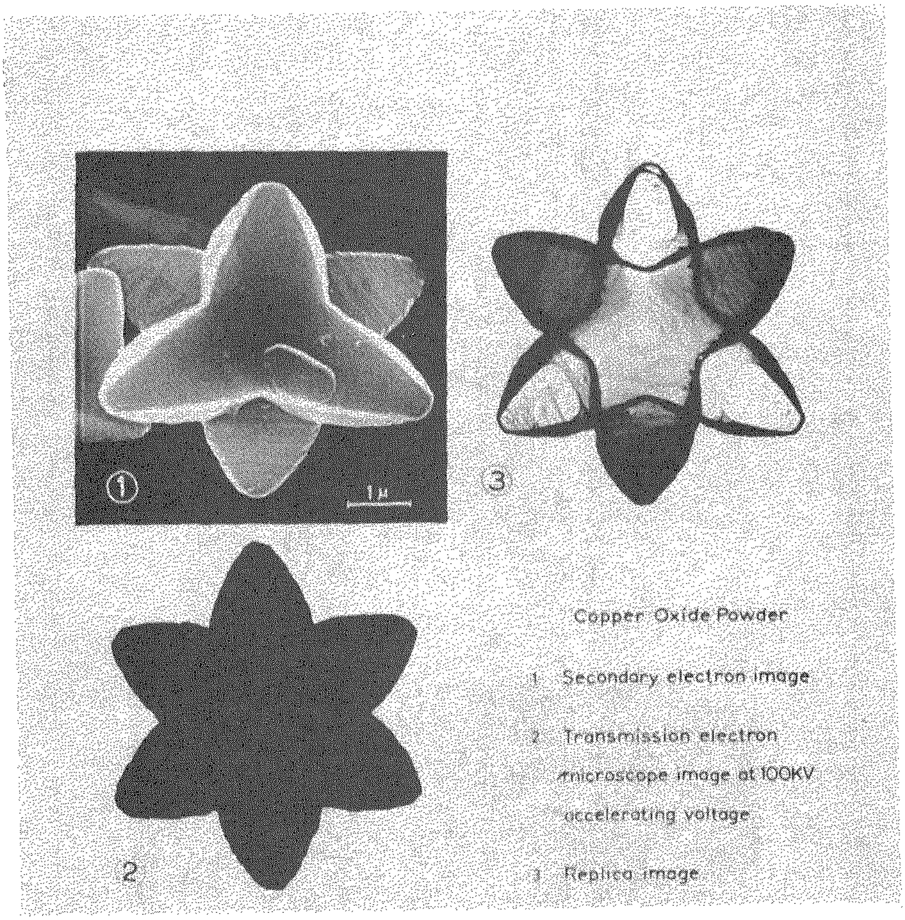


Fig. 18 Comparison between TEM and SEM.

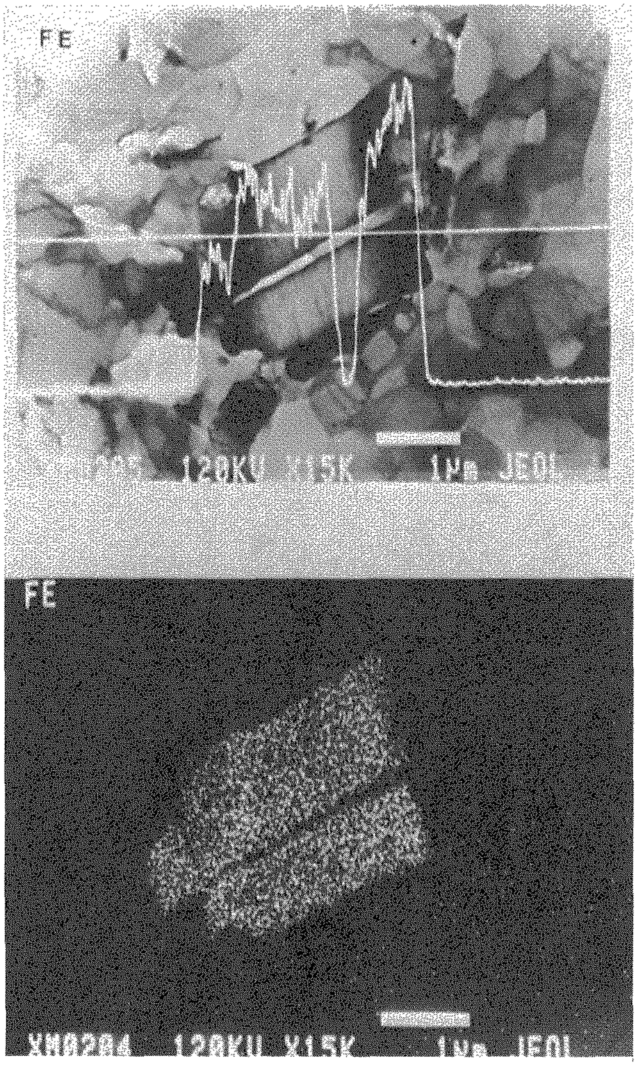


Fig. 19 Energy Dispersive X-ray spectrscopy together with electron microscope image.

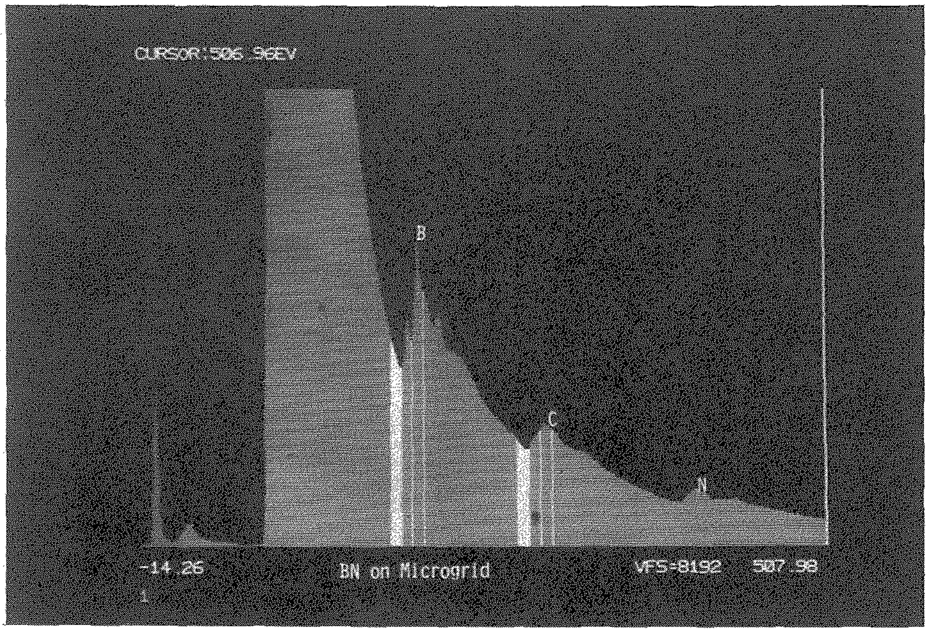


Fig. 20 One example of electron energy loss spectroscopy.

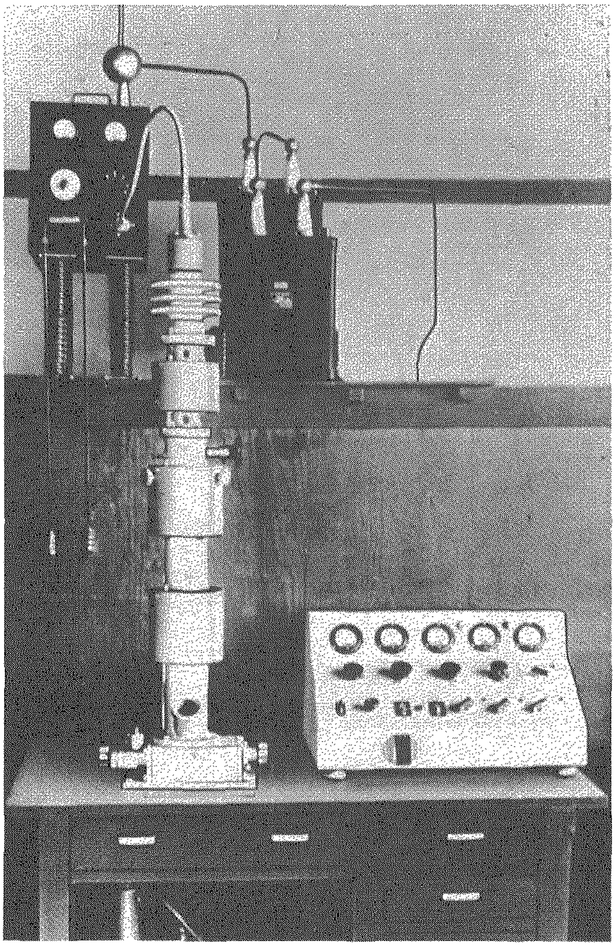


Fig. 21 The first electron micrsocope manufactured by us in 1946.

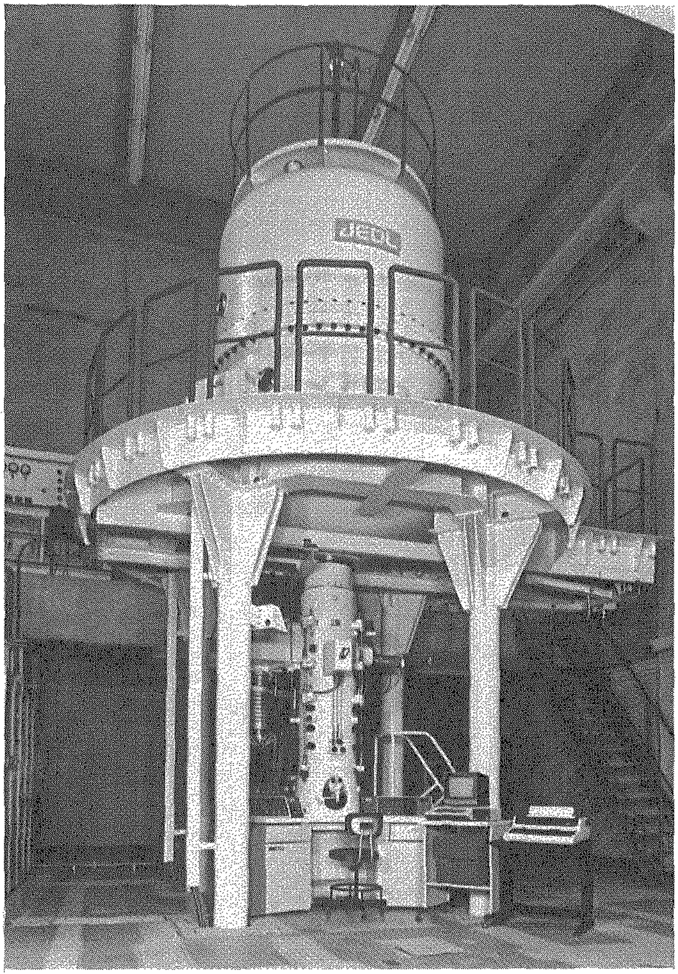


Fig. 22 The latest microscope of 1,000 kV accelerating voltage.