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Rose

[11] **3,908,124**

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[54] **PHASE CONTRAST IN HIGH RESOLUTION ELECTRON MICROSCOPY**

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[73] Assignee: **The United States of America as represented by the United States Energy Research and Development Administration**, Washington, D.C.

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[52] **U.S. Cl.** **250/311**; 250/310

[51] **Int. Cl.²** **H01J 37/26**; G01N 23/00

[58] **Field of Search** 250/305, 306, 307, 309, 250/310, 311

[56] **References Cited**

UNITED STATES PATENTS

3,626,184	12/1971	Crewe	250/311
3,857,034	12/1974	Hoppe	250/310 X

Primary Examiner—James W. Lawrence
Assistant Examiner—T. N. Grigsby
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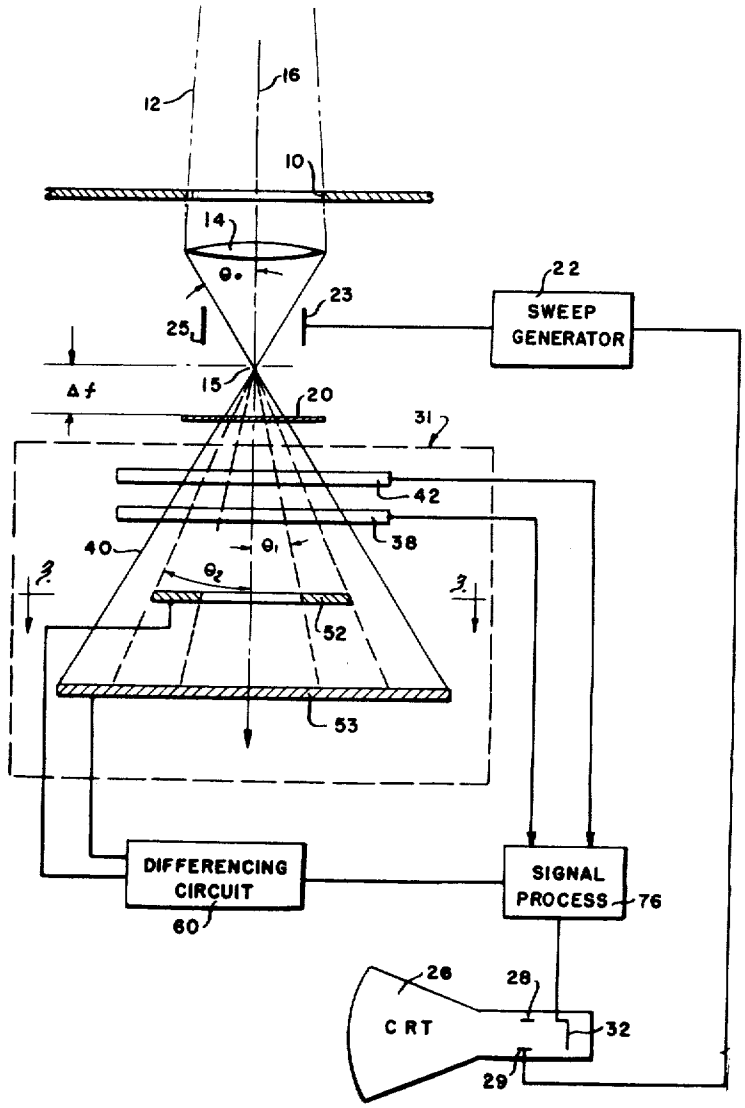
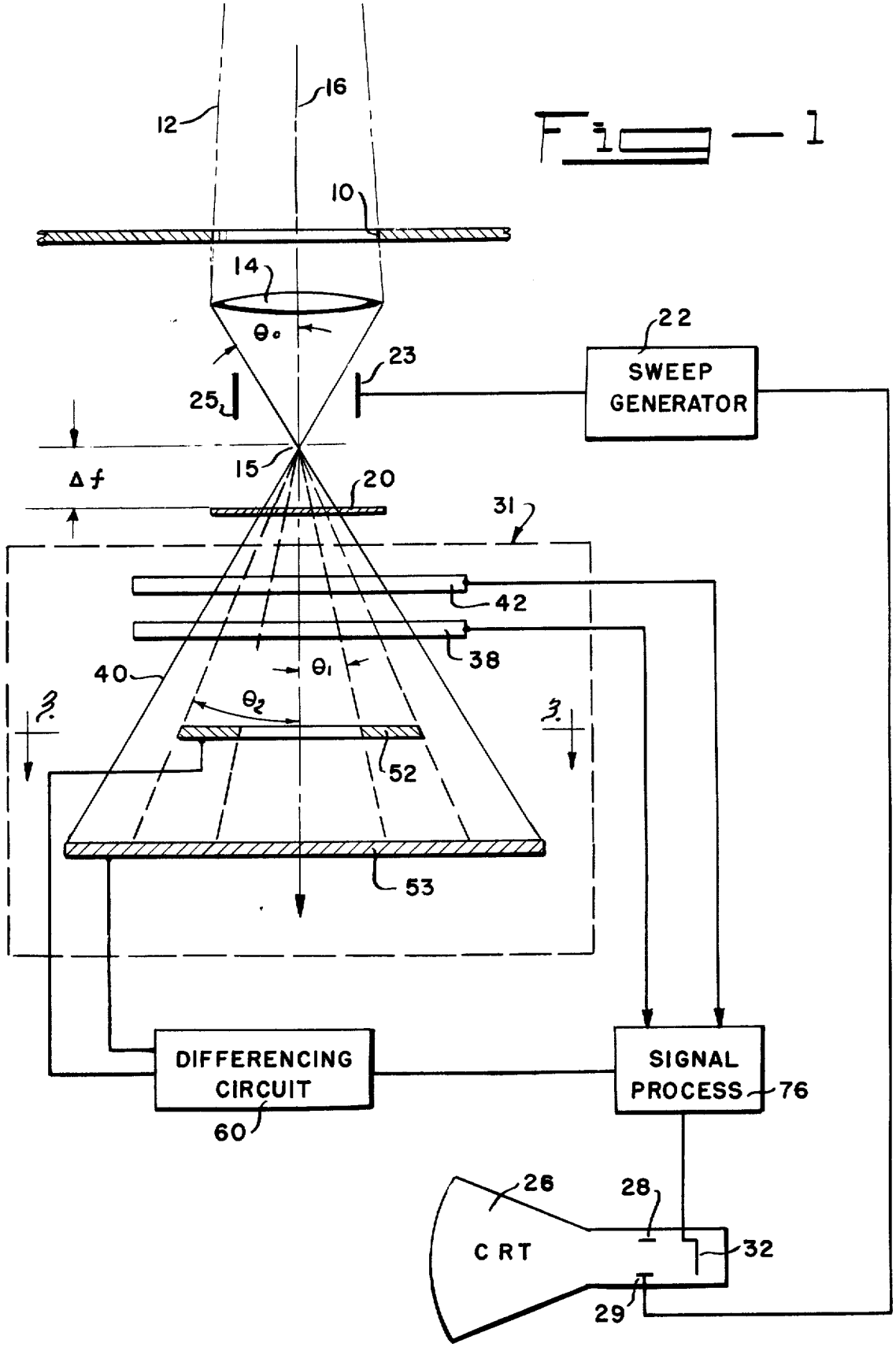
[57] **ABSTRACT**

A device is provided for developing a phase contrast signal for a scanning transmission electron microscope. The lens system of the microscope is operated in a condition of defocus so that predictable alternate concentric regions of high and low electron density exist in the cone of illumination. Two phase detectors are placed beneath the object inside the cone of illumination, with the first detector having the form of a zone plate, each of its rings covering alternate regions of either higher or lower electron density. The second detector is so configured that it covers the regions of electron density not covered by the first detector. Each detector measures the number of electrons incident thereon and the signal developed by the first detector is subtracted from the signal developed by the record detector to provide a phase contrast signal.

8 Claims, 7 Drawing Figures




12
16
10
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θo
22
SWEEP GENERATOR
23
25
Δf
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20
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38
40
θ1
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3
3
52
53
DIFFERENCING CIRCUIT
60
SIGNAL PROCESS
76
26
28
C R T
32
29

Fig — 1

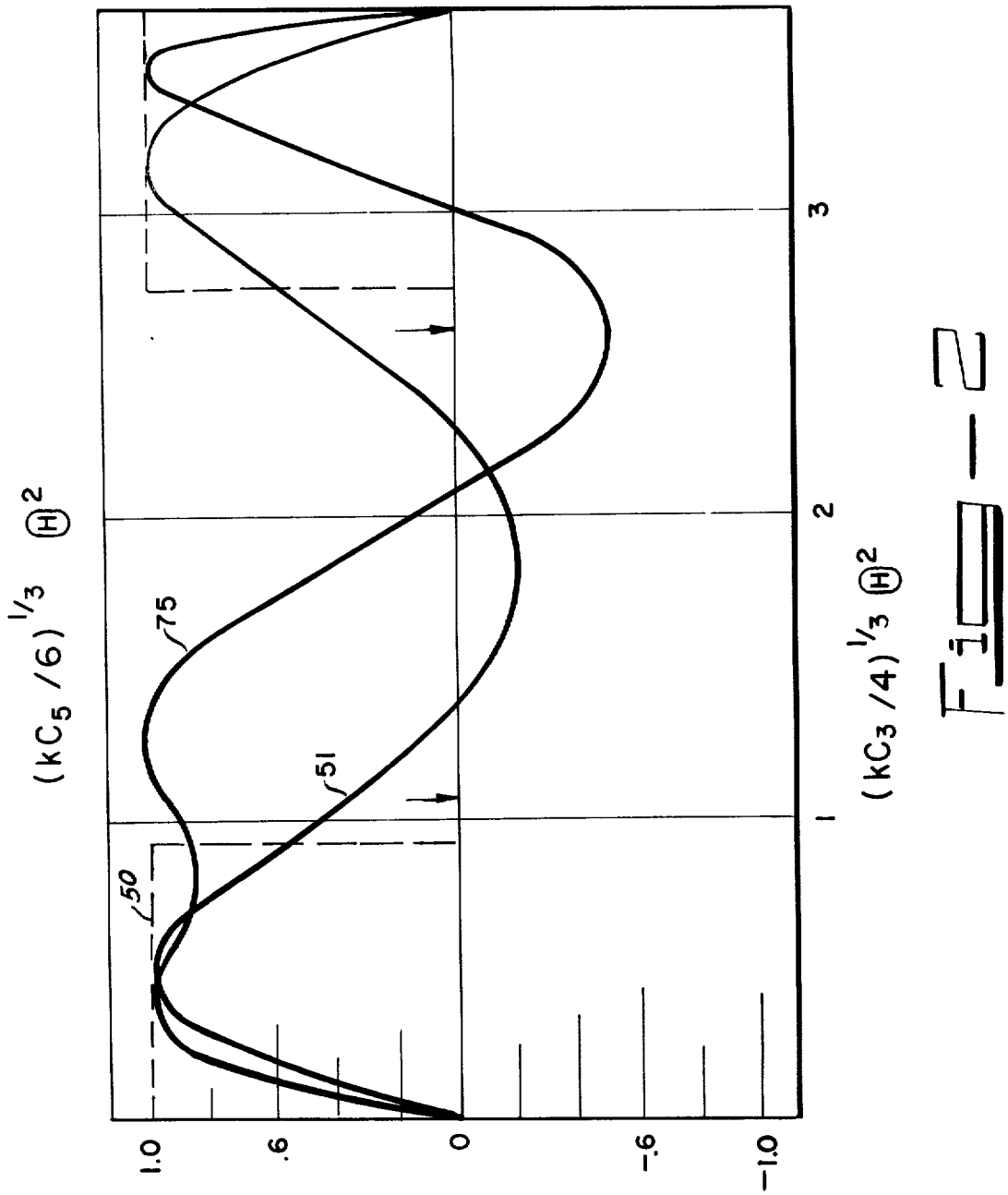
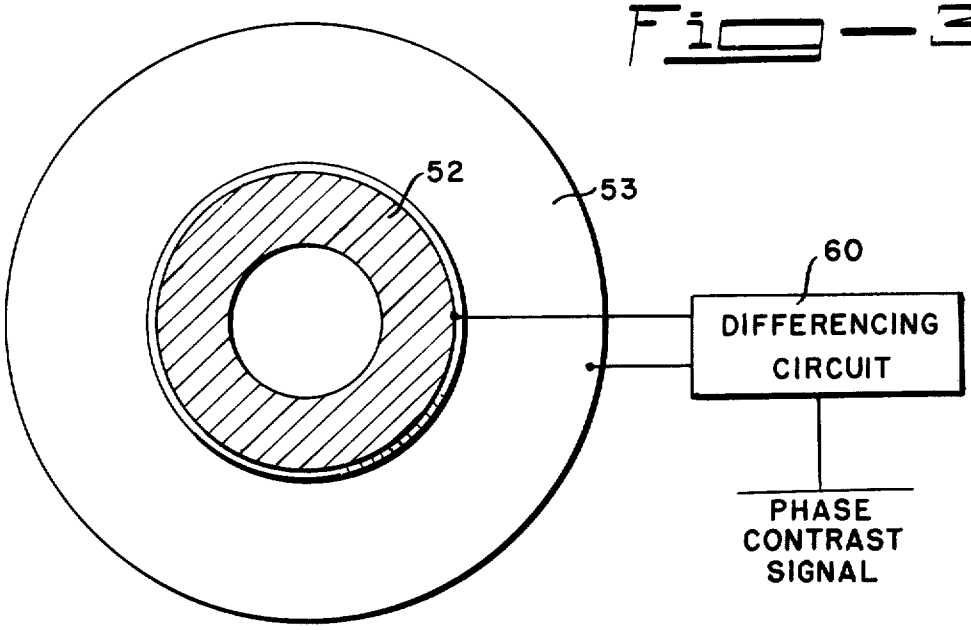


Fig. 2

Fig—3



Fig—4

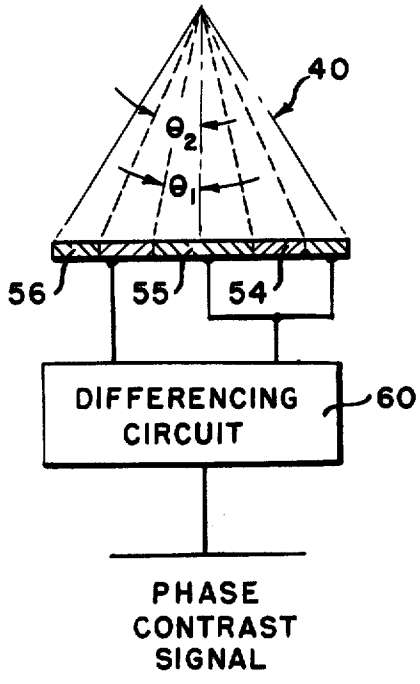


Fig 5

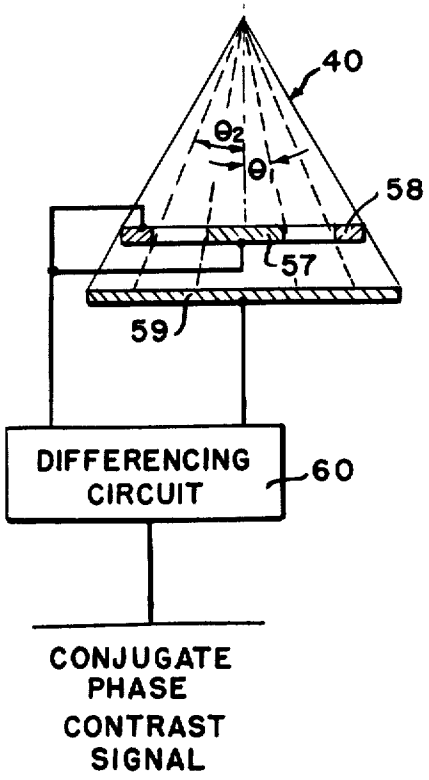
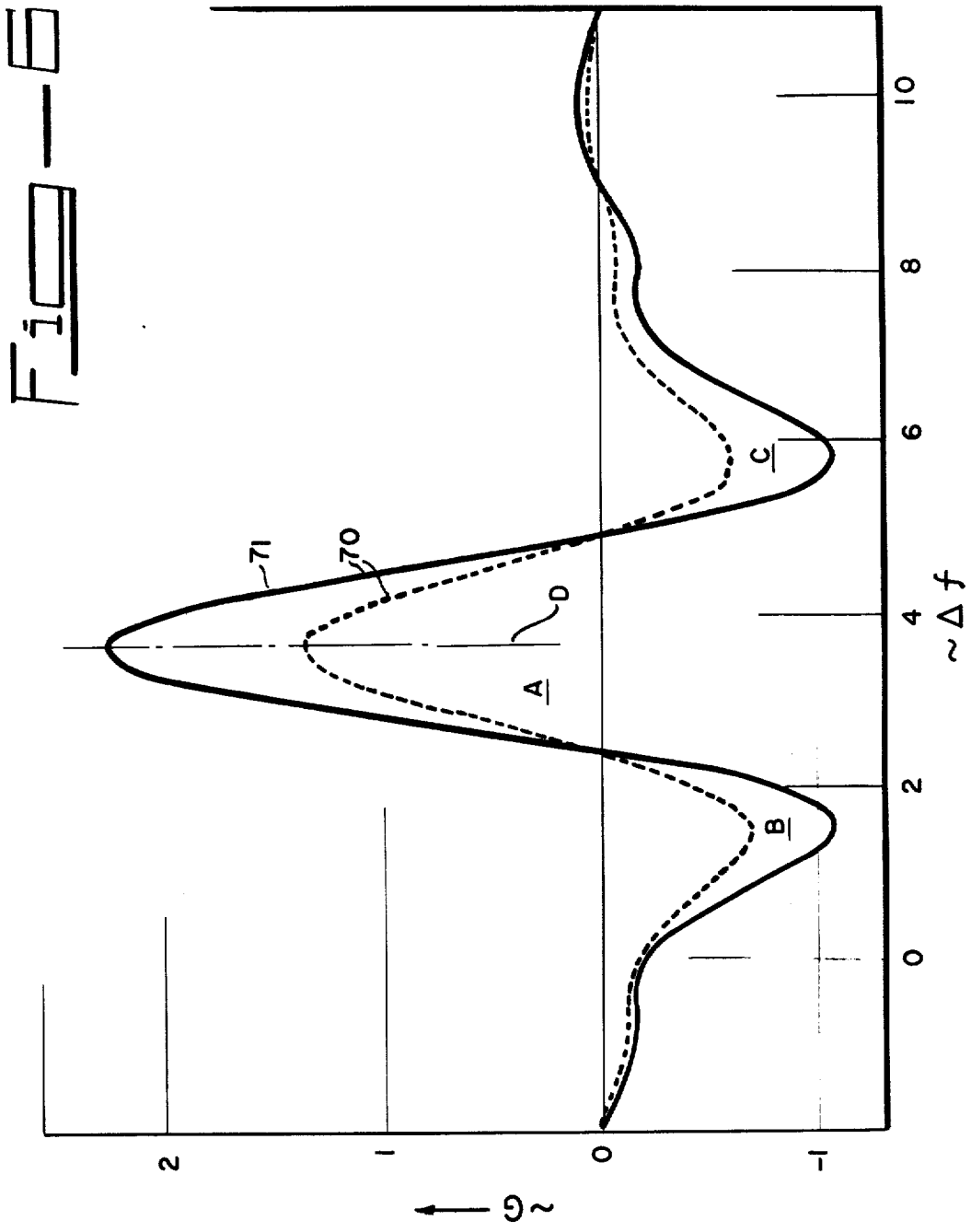


Fig. 5



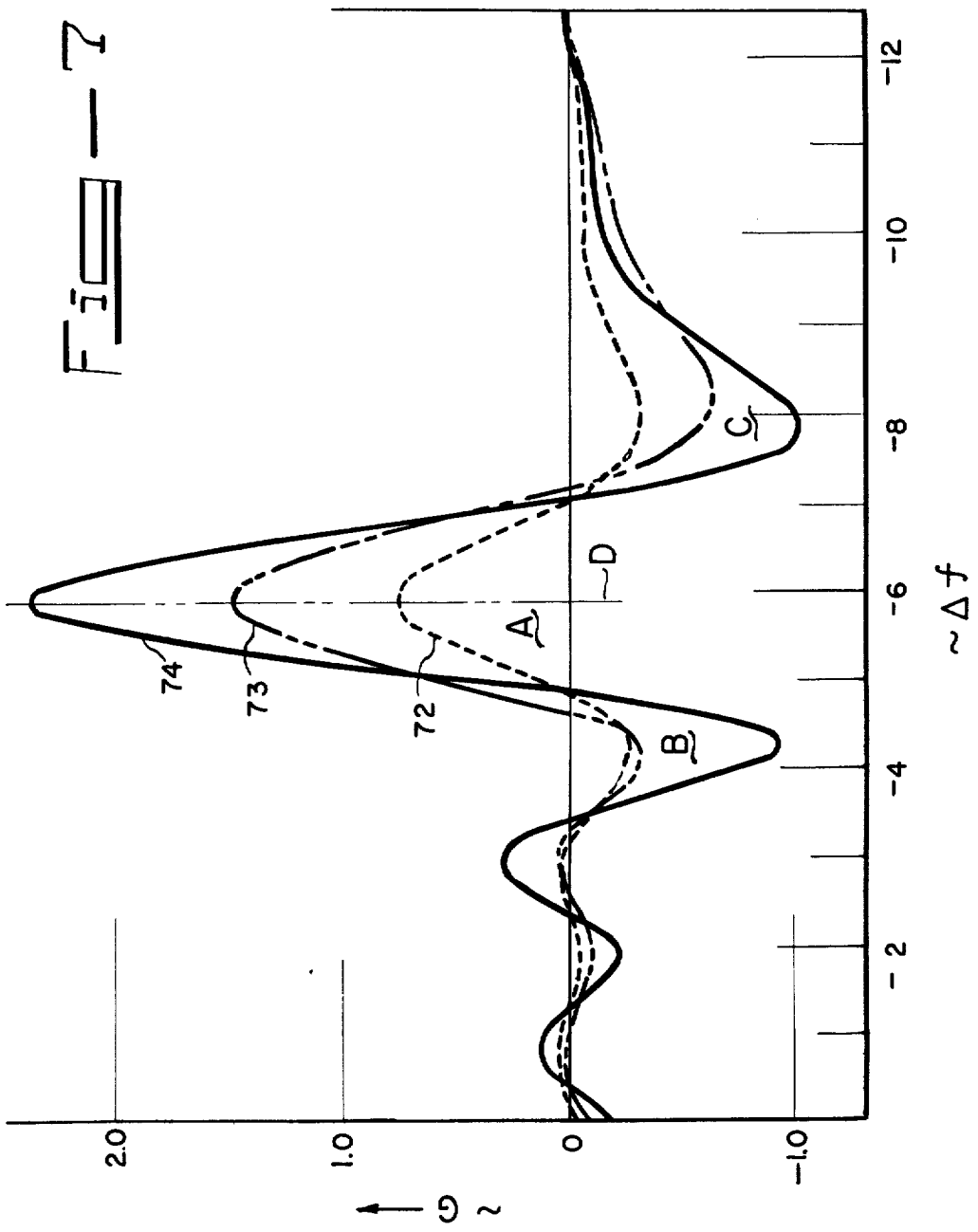


FIG — 7

PHASE CONTRAST IN HIGH RESOLUTION ELECTRON MICROSCOPY

CONTRACTUAL ORIGIN OF THE INVENTION

The invention described herein was made in the course of, or under, a contract with the United States Atomic Energy Commission.

BACKGROUND OF THE INVENTION

The scanning transmission electron microscope has been operated predominantly in the dark field mode using an annular detector to collect scattered electrons falling outside the cone of illumination, as described in U.S. Pat. No. 3,626,184. For resolutions with the limit of resolution $d \gtrsim 2$ Å, the dark field detector yields a high collection efficiency resulting in a short scanning time and reduced dosage of electrons incident on the specimen for each element of the reproduced image. The smaller the dose to produce the image, the lower the radiation damage to the specimen. However, as d is made smaller, more of the elastically scattered electrons remain within the illumination cone, and fewer of them fall onto the dark field detector. Thus, the amount of information obtainable with the dark field detector decreases as resolution increases.

Within the cone of illumination, a phase contrast image results from an interference of unscattered electrons with elastically scattered electrons. Detection of the phase contrast image or interference pattern within the cone of illumination would provide the information relative to elastically scattered electrons within the cone of illumination necessary to allow increased resolution in the scanning electron microscope without an unacceptable increase in dosage.

It is therefore an object of this invention to provide an improved detection system for a scanning electron microscope.

Another object of this invention is to provide a means for obtaining information relative to elastically scattered electrons obtaining information relative to elastically scattered electrons in the cone of illumination.

Another object of this invention is to provide a means for obtaining a phase contrast image within the cone of illumination formed by interference between the unscattered electron wave and the elastically scattered electron wave.

SUMMARY OF THE INVENTION

For the practice of this invention a device is provided for developing in a scanning electron microscope a phase contrast signal representative of the interference within the cone of illumination of electrons elastically scattered by the specimen with unscattered electrons. The microscope is operated in a condition of defocus so that within and concentric with the cone of illumination there will exist alternate regions of constructive and destructive interference between the waves which will correspond to regions of higher and lower electron density. A first detector is placed within and concentric with the cone of illumination and has the form of a zone plate covering alternate regions of higher or lower electron density and is responsive to electrons incident thereon to develop an output signal representative of the incident electrons. A second detector is placed within and concentric with the cone of illumination and is so configured that it detects and develops an output signal representative of those regions of higher or lower electron density undetected by the first detector. The output signals are subtracted from each other to develop a phase contrast signal.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. **1** is a drawing of a scanning transmission electron microscope with the phase contrast detector;

FIG. **2** is a set of curves showing the sine of the phase distribution over the cone of illumination;

FIG. **3** is a sectional view taken along line **3** — **3** of FIG. **1**;

FIG. **4** is a drawing of another embodiment of this invention;

FIG. **5** is a drawing of another embodiment of this invention; and

FIGS. **6** and **7** are curves showing the phase contrast intensity as related to defocus.

DETAILED DESCRIPTION OF THE EMBODIMENT

Referring to FIG. **1** there is shown a portion of the scanning electron microscope, more particularly described in U.S. Pat. No. 3,626,184. The microscope includes an aperture **10** through which an electron beam **12** is projected, an objective lens system **14** which focuses electron beam **12** to as small a spot as possible at the focus point **15** along the optical axis **16** of the lens system **14.** The angle θ_0 which defines the envelope of the focused beam is called the illumination angle. The focused spot is scanned over the area of specimen **20** to be examined in a manner similar to a TV scan. Sweep generator **22** provides scanning voltages to a deflection system in the microscope (represented by deflection plates **23** and **25**). Voltages on the deflection plates act to move the electron beam across the specimen in a desired manner.

Sweep voltages from sweep generator **22** are also applied to the deflection plates of CRT **26** (represented by deflection plates **28** and **29**). The sweep voltages applied to the CRT **26** are in synchronism with the sweep voltages applied to the electron microscope so that the electron beam in the CRT **26** traces a raster on the face of the tube as the specimen **20** is scanned. Detection system **31** placed beneath specimen **20** receives electrons transmitted through the specimen. Signals which are developed by the various types of detectors of the detector system **31** are applied to cathode **32** of CRT **26** to modulate the intensity of the electron beam in CRT **26** according to the electrons received by detector system **31.** The modulated electron beam forms a picture on the face of CRT **26** representative of the specimen being observed.

The electrons which reach the plane of detector **31** comprise three components, those elastically scattered, those inelastically scattered and unscattered electrons. Separation of information relative to inelastically scattered electrons is obtained from spectrometer **38,** more particularly described in U.S. Pat. No. 3,191,028 and that of elastically scattered electrons falling outside the cone of illumination **40** is obtained from dark field detector **42,** more particularly described in U.S. Pat. No. 3,626,184. The device herein disclosed deals with obtaining information relative to elastically scattered electrons falling within the cone of illumination **40.**

Within the cone of illumination, a phase contrast image results from an interference of the electron wave associated with electrons elastically scattered by specimen **20** with the wave associated with the unscattered

electrons. The interference pattern arises because the phase of the scattered wave is shifted with respect to the unscattered wave. Additional phase shift in the cone of illumination will be caused by any spherical aberration of lens system **14** and any defocus in the microscope.

Defocus refers to a distance or defocus length Δf between the specimen **20** and the focus point **15.** If the microscope were in focus, point **15** would generally coincide with specimen **20.** It is well known that phase shifts can be introduced between electrons in the cone of illumination by operating the microscope in an out-of-focus condition, that is with Δf less than 0, which means that focus point **15** is before the specimen as shown in FIG. **1.**

The spherical aberration of a lens system relates to imperfections of the lens system causing failure of all the electrons of the focused beam to converge to the same spot point **15.** It is a variable quantity depending upon the parameters of lens system **14.** Calculated approximations of the distribution of the beam about point **15** associated with spherical aberration in the form of power expansions are well known.

The phase contrast or interference pattern in the cone of illumination will show up most prominently when the total phase shift caused by the specimen, the defocus Δf and the spherical aberration of lens system **14** approaches 0 of π or multiples thereof, which is the condition of maximum constructive or destructive interference. We may approximate the phase shift between elastically scattered and unscattered electrons caused by the specimen as being 90°. A constant phase shift of 0 or π over the entire illumination cone is never possible due to the conservation of the number of electrons. However, concentric hollow cones or regions within the cone of illumination with total phase shift alternately being 0 or π in each cone is allowable. This is illustrated in FIG. **2** by the dashed phase distribution curve **50** which refers to the lower scale of FIG. **2.** Curve **50** is the sin of the ideal total phase shift γ, which is a step function, where in FIG. **2,**

$$k = \frac{2\pi}{\lambda}.$$

c_3 is the third order coefficient ot spherical aberration and Θ is the angle with the optic axis **16.** These hollow cones or regions of constructive and destructive interference will also be hollow cones of higher and lower electron density. It is known that by operating a microscope in an out-of-focus condition, with Δf less than 0, an interference pattern with regions approaching total phase shift of 0 or π can be achieved. For example, consider the distribution of total phase shift when the defocus $\Delta f = -C_1$ for a microscope uncorrected for spherical aberration, that is one with a Seidel order of $n = 3$, where C_1 is the first order coefficient of spherical aberration. The $-\sin$ of the total phase shift γ is shown by curve **51** which refers to the lower scale of FIG. **2.** The distribution of curve **51** provides a reasonable approximation of the ideal distribution of the step function of curve **50.**

To extract information out of the cone of illumination **40** where the sin of the total phase shift approximates the step function as illustrated by curve **51** of FIG. **2,** two-phase contrast detectors are placed beneath specimen **20** within the cone of illumination **40** as shown in FIG. **1** and FIG. **3.** The first detector **52** has the form of a zone plate, each of its m rings generally covering all hollow cones of either construction or destructive interference. For the phase distribution illustrated by curve **51** of FIG. **2** only one such zone plate ring **52** is required, i.e., $m = 1$, to cover the sole region of lower density of this distribution. The second detector consists of a detector in the form of a disc **53** covering the entire illumination cone in the shadow of ring **52** covering those hollow cones not covered by ring **52** which in this embodiment includes the central and outer higher density regions. An alternate embodiment is shown in FIG. **4** where the second detector is coplanar with ring **54** of the zone plate type first detector. Here the second detector is also in the form of a zone plate with a central disc **55** and an outer ring **56.** The signals of the disc **55** and ring **56** are added together to give the same signal as disc **53** of FIG. **1.** A further alternate embodiment is illustrated in FIG. **5** for detecting this phase distribution. Here the second detector is a zone plate with a disc **57** and ring **58** whose signals are added and the first detector is a disc **59** covering the entire illumination cone. In this case, rings **57** and **58** give the signal developed by disc **53** of FIG. **1** and disc **59** gives the signal developed by zone plate **52** of FIG. **1.**

Both detectors are intended to develop output signals corresponding to the number of electrons incident thereon and may be, for example, silicon surface barrier detectors. The signals of these detectors are available simultaneously and can be conveniently combined and applied to the cathode **32** of CRT **26** or only the signal from one detector covering all regions of higher or lower electron density may be used to vary the intensity of each element of the raster scan. Best phase contrast is obtained by subtracting the signal from one detector from the signal from the other detector such as by means of a differencing circuit **60** to form a phase contrast signal. Each scanned element of the specimen varies the phase shift according to its own particular characteristics. Because of the conservation of electrons, the variation in phase shift which causes an increase in electron density in the hollow cones of constructive interference must necessarily also result in a decrease in the density in the hollow cones of destructive interference and visa versa. By subtracting the two signals the effect of each scanned element of the specimen on the resulting phase contrast signal from differencing circuit **60** will be twice as large as the effect on the signal from only one detector. This is because the effect on phase contrast in each region or hollow cone observed by each detector will be opposite in sign. The raster scan of CRT **26** represents a comparison of intensities between each element of the scan so that by doubling the effect of something which varies the comparative intensity one will have made more visibly evident the phase contrast effect.

The phase distribution obtained from a particular microscope lens system operated in an out-of-focus condition is determined by the spherical aberration characteristics of the lens system of the microscope and in the actual value of Δf. The spherical aberration of a lens system is quantitatively described by the interrelated coefficients of spherical aberration denoted by C which are readily obtainable and well known and appear in a power expansion whose coefficients are C_1, C_3, C_5, etc. For a lens system in which no correction for spherical

aberration has been made, i.e., Seidel order $n = 3$, C_3 dominates and all of the outer power terms are negligible. For a microscope corrected for third order aberration, i.e., Seidel order $n = 5$, C_3, C_1 are the dominant coefficients which determine the phase shift caused by the lens system. Generally, one can say that the coefficients of spherical aberration which are dominant in determining optimum phase are C_1, . . . to $C_{2\ \nu+1}$ where ν goes from 0 to $(n-3)/2$, where n is the Seidel order which is limiting resolution. By variation of the lens factors which effect these coefficients the phase distribution is varied. Thus, for an uncorrected microscope, $n = 3$, C_1 is the only free parameter available for control over the phase distribution, and for a microscope corrected for third order aberrations, $n = 5$, C_1 and C_3 are free parameters. By manipulation of the free parameters in the design and operation of lens system **14** by well known means, it is possible to obtain various usable phase distributions to approximate the ideal step function distribution.

The actual values of Δf which provide adequate phase contrast are illustrated in FIG. **6** and FIG. **7** which are curves showing the relative relationship of Δf to the intensity G of the phase contrast signal for an uncorrected microscope ($n = 3$) in FIG. **6** and for a corrected microscope ($n = 5$) in FIG. **7**. As shown by curves **70** and **71** of FIG. **6** and curves **72, 73** and **74** of FIG. **7** which are curves of varying resolutions only three regions A, B and C exist which contribute significantly to phase contrast. Regions B and C are ones of phase contrast opposite in sign from region A. In each case the central region A gives maximum contrast with its maxima point D coinciding with $\Delta f = -C_1$. The region of usable values of Δf has been observed to be about 4 (d^2/λ) on either side of point D for region A and about 8 (d^2/λ) on either side of point D for regions A, B and C where λ is the electron wavelength and d is the limit of resolution. The limit of resolution is a quantity which determines the quality of the microscope. It is determined for an electron microscope by using the well known method of Scherzer. Using Scherzer's method for a microscope providing a phase contrast signal as herein disclosed, d was determined to be, with $n = 3$, $d \approx 0.36(C_3\lambda^3)^{1/4}$ and with $n = 5$, $d \approx 0.31(C_5\lambda^5)^{1/6}$. For a particular microscope with $n = 5$ and with $C_5 \approx 15$ cm, d is about 0.67 A for 100 kv electrons.

Generally, the microscopist is interested in reducing the radiation applied to the specimen. Therefore, given the desired phase distribution from the design of the lens system and from the value of Δf, the best means for determining the optimum position and dimensions for the detectors is to assume an acceptable fixed signal to noise ratio for the phase contrast signal developed by differencing circuit **60** and then minimizing the dose on the specimen. The signal to noise ratio q is obtained by dividing the noise contrast by the mean phase contrast and by well known calculation procedures has been determined to be:

$$(1)\quad q = \frac{4}{3}\ \frac{\alpha}{\beta}\ Z\sqrt{N}\ \ G(O)$$

where α is Sommerfeld's constant, βc is the electron velocity, Z is the atomic number of the atom being imaged, $\sqrt{N}$ is the resulting noise which is identical to the statistical fluctuation in the total number of electrons, N, incident on both detectors, and $G(O)$ is the value at the image point of $G(R)$ which is the intensity distribution of the phase contrast signal as a function of R the effective radius of the image referred to the object plane. $G(R)$ is given by:

$$(2)\quad G(R) = \frac{4}{\pi\theta_a^2 Z} \sum_{\mu=1}^{m} \int_0^{2\pi} \int_0^{\theta} \int_{\Theta_2\mu}^{\Theta_2\mu} J_o(kR|\vec{\theta}-\vec{\Theta}|)\ \ \frac{Z-F}{\vec{\theta}-\vec{\Theta}^2}$$

$$\sin\left[\gamma(\theta) - \gamma(\Theta)\right] \Theta\, d\Theta\theta d\theta d\phi$$

where θ_a is illumination angle, m is the number of rings of the first detector, Θ is the angle with respect to the optic axis defining the cone of constructive or destruction interference covered by each ring of the first detector, F is the atomic form factor, J_o is a Bessel function of zero order, the wavelength, $\lambda = 2\pi/k$, ϕ is the angle subtending the vectors of $\vec{\theta}$—$\vec{\Theta}$ which are perpendicular to the optic axis and $\gamma(\theta) - \gamma(\Theta)$ is the additional phase shift due to spherical aberration and defocus (the phase shift associated with the specimen being assumed to be $\pi/2$) with

$$\gamma(\Theta) = k \sum_{\nu=0}^{\infty} \frac{C_{2\nu+1}}{2\nu+2}\Theta^{2\nu+2}$$

for $2\nu + 1 = n$ the Seidel order, for $\lambda = 2\pi/k$ the wavelength of the electrons. Considering equation (1) for q, it is apparent that for fixed, q, N, the dose, becomes smallest when $G(O)$ reaches its maximum. The problem may be simplified by approximation of the atomic potential by a delta function, i.e.

$$\frac{Z-F}{|\theta-\Theta|^2} = \text{constant} = kZ.$$

As stated previously, phase contrast will be most prominent when the total phase shift approaches 0 to π or multiples thereof. Applying this to the maximization of G, this distribution occurs when half of the incident electrons have an additional phase shift due to spherical aberration and defocus of 0 or π and the other half an additional phase shift of $\pi/2$. Then the phase shift between an arbitrary unscattered electron and a scattered electron will be 0 or $\pi/2$ for certain hollow cones or regions of the cone of illumination and π or $\pi/2$ for the others. The total solid angle of the cones yielding a constructive interference is equal to that of cones with destructive interference. As stated before, the ideal distribution of the phase shift can be approached in practice in an uncorrected microscope with the ideal defocus, $\Delta f = -C_1$ which is the only free parameter available to vary the phase distribution to desired levels. The other free parameters determine the width of the first detector zone rings and do not effect the actual phase shift. To approach the lowest dose for

a real microscope we maximize G with respect to the free parameters θ_o, Θ_σ and $C_{2\ \nu+1}$. The subscript $o\sigma$ runs from 1 to $2m$, m being an integer indicating the number of zone rings of detector **52**. The subscript ν runs from 0 to $(n-3)/2$ where n is the Seidel order of that spherical aberration which cannot be chosen arbitrarily and is limiting the resolution. With the defocus equal to $-C_1$, in an uncorrected microscope then $\Delta f \approx 1.44(C_3\lambda)^{1/2}$. In this case the first phase contrast detector consists of a single ring **52**, i.e., $m = 1$, as shown in FIG. **1**. The optimum angles of this ring are $\theta_1 \approx 0.92(\lambda/C_3)^{1/4}$ radians, $\theta_2 \approx 1.44(\lambda/C_3)^{1/4}$ radians and the optimum illumination angle is $\theta_o \approx 1.71(\lambda/C_3)^{1/4}$ radians. The areas of the second phase contrast detector which do not lie in the shadow of the first detector **52** and which are detected by the second detector's disc **54** are a central disc and an outer ring.

In an microscope corrected for third order spherical aberration, the coefficient C_3 becomes an additional free parameter for effecting the optimum phase contrast distribution. Optimum operating conditions are approached if again we choose $\Delta f \approx -C_1$ so that $C_1 \approx +1.91(C_5\lambda^2)^{1/3}$, $C_3 = -3.2(C_5^2\lambda)^{1/3}$. This is illustrated by curve **75** of FIG. **2** which refers to the upper scale of FIG. **2**. In the upper scale C_5 is the fifth order coefficient of spherical aberration. The optimum configuration of the first detector is the same as in the case of the uncorrected microscope, i.e. $m = 1$. Of course, the optimum illumination and detector angles have changed. These optimized angles are $\theta_1 \approx 1.23(\lambda/C_5)^{1/6}$ radians, $\theta_2 \approx 1.77(\lambda/c_5)^{1/6}$ radians and $\theta_o \approx 1.87(\lambda/C_5)^{1/6}$ radians.

In practice the signal developed by the phase contrast detector can be combined with the dark field detector **42** signal and the signal relating inelastically scattered electrons developed by the spectrometer **38**, by combining the signals together conveniently with signal processor **76** and applying them to cathode **32**. Note that while it is not necessary to filter only the inelastically scattered electrons from the cone of illumination with spectrometer, improved detection is achieved if they are filtered out before phase contrast detection as shown in FIG. **1**, since they would unnecessarily contribute to the intensity measured by the detectors. Spectrometer **38** will bend the cone of illumination to separate the lower energy inelastically scattered electrons, however, since the elastically scattered and unscattered electrons are of the same energy, the bending by spectrometer **38** will not alter the relative distribution of elastically scattered and unscattered electrons in the cone of illumination.

The embodiments of the invention in which an exclusive property or privilege is claimed and defined as follows:

1. In a scanning transmission electron microscope operating in a condition of defocus such that there are within the cone of illumination regions alternately of the quality of higher electron density and of lower electron density, the alternate density regions being caused by the interference within the cone of illumination of electrons elastically scattered by the specimen with electrons unscattered by the specimen, a device for developing a phase contrast signal, comprising:

a first detector positioned within the cone of illumination, and being of such radial configuration with respect to the optical axis that electrons within all alternate regions of the cone of illumination having one of the qualities of density are incident thereon, said first detector being responsive to electrons incident thereon to develop a first output signal representative of the intensity thereof, a second detector positioned within the cone of illumination and being of such configuration that electrons within the cone of illumination not incident upon said first detector are incident upon said second detector, said second detector being responsive to electrons incident thereon to develop a second output signal representative of the intensity thereof, a differencing circuit coupled to said first and second detectors and responsive to said first and second output signals therefrom to subtract said first output signal from said second output signal and to develop a phase contrast signal which is the subtraction of said first output signal from said second output signal, and utilization means coupled to said differencing circuit for utilizing said phase contrast signal.

2. The device of claim **1** wherein said regions within the cone of illumination of alternate electron density are concentric with the cone of illumination, and wherein said first and second detectors are concentric with said cone of illumination.

3. The device of claim **2** wherein the defocus length Δf of the microscope is within $8\ d^2/\lambda$ of $\Delta f = -C_1$ where d is the limit of resolution, λ is the electron wavelength and C_1 is the first order coefficient of spherical aberration.

4. The device of claim **3** wherein $\Delta f = -C_1$.

5. The device of claim **4** wherein the microscope has a Seidel order of $n = 3$ and an angle of illumination of $1.71(\lambda/C_3)^{1/4}$ radians, and wherein electrons transmitted through the specimen and thereby directed between approximately $0.92(\lambda/C_3)^{1/4}$ radians and approximately $1.44(\lambda/C_3)^{1/4}$ radians with respect to the optical axis are incident upon said first detector where C_3 is the third order coefficient of spherical aberration.

6. The device of claim **4** wherein the microscope has a Seidel order of $n = 5$ and an angle of illumination of $1.87(\lambda/C_5)^{1/6}$ and wherein electrons transmitted through the specimen and thereby directed between approximately $1.23(\lambda/C_5)^{1/6}$ radians and approximately $1.77(\lambda/C_5)^{1/6}$ radians with respect to the optical axis are incident upon said first detector where C_5 is the fifth order coefficient of spherical aberration.

7. The device of claim **2** wherein said first detector includes a zone plate having at least one ring and wherein said second detector includes a disc so positioned that said first detector is between said specimen and said second detector.

8. The device of claim **2** wherein said first detector includes a zone plate having at least one ring and wherein said second detector includes a zone plate having at least one ring.

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