June 13, 1950 **H. EKSTEIN ET AL** 2,511,151

X-RAY APPARATUS AND METHOD

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Fig. 1

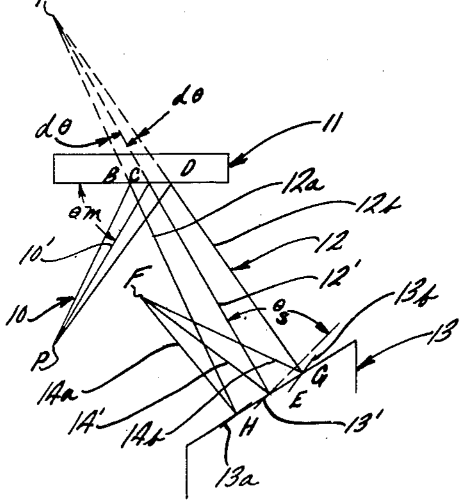


Fig. 3

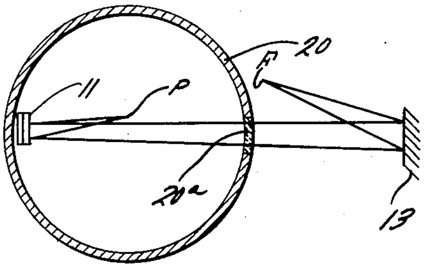


Fig. 2

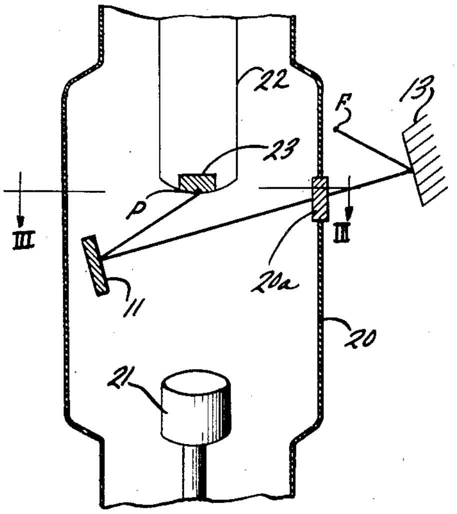
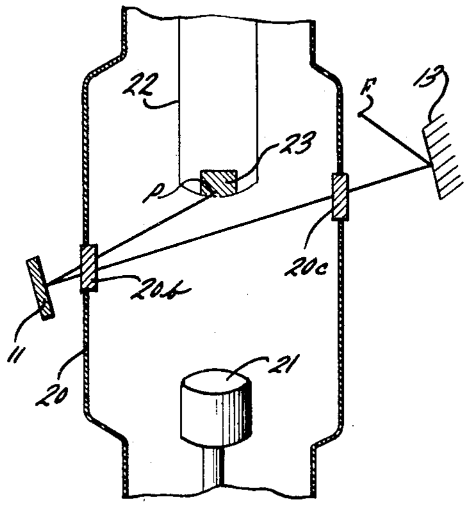


Fig. 4



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2,511,151

X-RAY APPARATUS AND METHOD

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This invention relates to an improved method and apparatus for facilitating the precision determination of lattice parameters, and particularly to a method and apparatus for effecting the focusing of a beam of X-rays upon an X-ray sensitive indicating medium after diffraction of such beam by a sample to be analyzed.

The desirability of producing a focusing of X-rays to facilitate X-ray analysis of crystalline structure is in itself not a new concept; however, all previous methods utilized were based on geometrical focusing in which supposedly monochromatic X-rays are brought to a focus. However, at large angles of diffraction, which are particularly desirable in precision lattice determinations, such as in phase studies, stress measurements, etc., the diffraction line becomes inevitably broad because of the finite spectral width of the characteristic radiation and because of the diffraction mechanism.

In contrast, the methods and apparatus constituting this invention are based upon the concept of producing a focusing of X-rays of different wavelengths. Hence, the methods and apparatus of this invention permit the determination of lattice parameters with improved accuracy by making the Debye-Scherrer line sharper than is possible by any method heretofore known.

The deficiencies of the presently known methods will be clearly apparent from the following analysis. The limiting factor of the precision possible by conventional focusing methods is the finite spectral width of the characteristic X-ray line. A strictly parallel and monochromatic beam, when striking a crystal aggregate of sufficiently large grains, will give rise to a diffracted beam of negligible angular width. As it is not possible to produce a characteristic X-ray line of a single wave-length, and since such characteristic X-ray line consists of a finite band of wavelengths, it follows that under the usual conditions the resulting diffracted beam cannot give rise to a line any narrower than that corresponding to the spectral width of the initial beam.

It is true that other factors, the geometric conditions and the small size of crystal grains, usually cause a broadening of the line in excess of that due to the spectral impurity of the incident radiation; but at large Bragg angles θ of the diffraction, where the Bragg angle θ has the largest sensitivity to changes in lattice parameters, the width caused by this special impurity is predominant when the geometric arrangements are as refined as currently feasible.

In other words, no further refinement of the geometry of the system can produce a line substantially narrower than those currently obtained.

Accordingly, the methods and apparatus embodied in this invention provide a decided improvement in the focusing of X-rays through the attainment of an indicating line substantially narrower than those currently obtained, by eliminating the line broadening effects caused by the spectral impurity. The methods and apparatus of this invention contemplate the utilization of a diverging beam of X-rays including a band of wavelengths and a portion of the rays of each distinct wavelength are caused to pass through a predetermined focal point after diffraction by a polycrystalline sample being analyzed. If an X-ray sensitive film or other recording instrument is placed at such focal point, the resulting line indication will be very narrow and hence adaptable for precision X-ray analysis purposes.

Accordingly, it is an object of this invention to provide an improved method and apparatus for X-ray analysis of crystal structure, and particularly a method and apparatus for producing a sharper focus of X-rays diffracted by a polycrystalline sample than has heretofore been possible of accomplishment.

Another object of this invention is the provision of an improved method and apparatus for focusing of X-rays which does not require a monochromatic beam of X-rays for its successful operation but which will produce a sharp focus of a beam of X-rays of a finite band of wavelengths.

Still another object of this invention is to provide a method and apparatus for focusing of an X-ray beam deflected by a polycrystalline sample wherein large Bragg angles of incidence of the X-rays upon the polycrystalline sample may be employed without reducing the sharpness of the focus achieved.

A particular object of this invention is to provide an improved X-ray tube envelope construction particularly adapted for producing a beam of X-rays of such characteristics that the diffraction of such beam by a polycrystalline sample located exteriorly of the tube envelope will automatically produce a focusing of the diffracted beam, and particularly a shielding envelope construction which may be conveniently adjusted to produce such focusing for any one of a plurality of distinct bands of X-ray wavelengths.

The specific nature of this invention, as well as other objects and advantages thereof, will become apparent to those skilled in the art from the following detailed description taken in conjunction with the annexed sheet of drawings, which, by way of preferred example only, illustrates two embodiments of this invention.

On the drawings:

Figure 1 is a schematic view of an X-ray focusing system illustrating the fundamental concept employed in accordance with this invention to obtain focusing of an X-ray beam after diffraction by a polycrystalline sample;

Figure 2 is a schematic sectional view illustrating one arrangement of an X-ray tube and shielding jacket for producing X-ray focusing in accordance with this invention;

Figure 3 is a transverse sectional view taken on the plane III—III of Figure 2; and

Figure 4 is a schematic sectional view illustrating a modified construction of an X-ray tube and jacket for producing focusing of X-rays by the principles of this invention.

As shown on the drawings:

Referring to Figure 1, let it be assumed that at the point P a source of X-rays is located which produces a diverging original beam of X-rays **10**. The beam **10** preferably comprises a finite band of wavelengths and this condition will be recognized by those skilled in the art as particularly easy to fulfill inasmuch as that is the type of radiation commonly achieved from any particular point on the target of a conventional X-ray tube. In any event, let it be assumed that the central ray **10′** of the diverging beam **10** has a wavelength λ.

Now, in accordance with this invention, the divergent, multi-wavelength beam **10** is permitted to strike a plain crystal face such as that provided by a monochromator **11**. It will be recognized that a diffracted or reflected beam **12** will thereupon be produced. However, the beam **12** will be divergent and will have the further characteristic that every ray in a particular angular portion of the beam **12** will have only one wavelength because it has been diffracted under an angle of diffraction different from any other ray. This phenomenon is readily apparent from consideration of the fundamental law of X-ray diffraction which is commonly set forth in the following equation:

$$\lambda=2d_m \sin \theta \qquad (1)$$

where λ is a particular wavelength diffracted, d_m is the atomic spacing in the diffracting material and θ is the glancing angle of the radiation of wavelength λ with respect to the crystal producing the diffraction. The angle θ is commonly referred to as the Bragg angle.

The diffracted beam **12** produced by the monochromator **11** is therefore formed of rays whose angular position with respect to the central ray **12′** of wavelength λ is a function of the difference in wavelength of each of the particular rays and the central ray **12′**. Thus the ray **12a** at one side of the beam **12** is angularly spaced from the central ray **12′** by the angle $d\theta$, and the wavelength of such ray **12a** may be λ plus $d\lambda$. The ray **12b** at the other edge of the beam is angularly spaced from the central ray **12′** of wavelength λ by an angle of minus $d\theta$, and the wavelength of this ray is λ minus $d\lambda$.

The beam **12** is then permitted to strike the surface of a polycrystalline sample **13** which is to be analyzed. With such a spatial distribution of the various wavelengths forming the diffracted ray **12**, it is obvious that each ray of the beam **12** will be diffracted by the polycrystalline sample only if it meets a crystal grain suitably oriented, this orientation being different for each incident ray. The various oriented crystals for the required diffraction of the central ray **12′** as well as the edge rays **12a** and **12b** are respectively indicated by the lines **13′**, **13a** and **13b**. When such diffraction occurs, the resulting diffracted rays, respectively represented by **14′**, **14a** and **14b**, will no longer be divergent but will be convergent, and under certain circumstances such diffracted rays can be made to intersect approximately at a point F. Therefore, if an X-ray sensitive film or other recording or indicating instrument is placed at the point F, then the line indication produced on such film will be very sharp, and precise determinations may be made of the lattice parameters of the polycrystalline sample **13**.

Geometrical analysis will indicate not only the conditions under which such focusing of the beam diffracted by the polycrystalline sample will occur at the point F but also the location of the point F with respect to the point source P and the monochromator **11**. The geometrical analysis proceeds as follows:

Still utilizing Figure 1, let P′ represent the apparent source of the X-ray beam **12** as viewed from the polycrystalline sample **13**.

Let d_m be the atomic spacing in the monochromator **11**; θ_m be the Bragg angle for the central ray **10′** diffracted by the monochromator **11**; d_s be the atomic spacing in the polycrystalline sample **13**; and θ_s be the Bragg angle for the central ray **12′** of wavelength λ diffracted by the polycrystalline sample **13**.

Then by Bragg's law, the diffraction of the central ray of wavelength λ which occurs by the monochromator **11** may be represented by the following equation:

$$\lambda=2d_m \sin \theta_m \qquad (2)$$

Likewise, the diffraction of the same ray by the polycrystalline sample **13** at the point E may be represented by:

$$\lambda=2d_s \sin \theta_s \qquad (3)$$

Considering a ray of slightly different wavelength and diffraction angle, i. e., $\lambda+d\lambda$ and $\theta+d\theta$, we obtain by differentiating (2) and (3) and setting equals against equals

$$d_m \cos \theta_m d\theta_m = d_s \cos \theta_s d\theta_s$$

or

$$d\theta_s=\frac{d_m \cos \theta_m}{d_s \cos \theta_s} d\theta_m \qquad (4)$$

By geometric analysis, the angle

$$HFE=2d\theta_s+d\theta_m \qquad (5)$$

Let the distance from the point source P to the sample **13** measured along the path of the central ray of wavelength λ (which, of course, is the distance P′E) equal L, and the distance from point E on sample **13** to focus point F equal f.

Then from the geometry of Figure 1,

$$-\frac{f(\text{angle } HFE)}{\cos 2\theta_s}=L d\theta_m \qquad (6)$$

and by Equations 4 and 5

$$\frac{L}{f}=\left(1+\frac{2d_m \cos \theta_m}{d_s \cos \theta_s}\right)\left(-\frac{1}{\cos 2\theta_s}\right) \qquad (7)$$

Since

$$\frac{d_m}{d_s}=\frac{\sin \theta_s}{\sin \theta_m}$$

(7) resolves to

$$\frac{L}{f}=\left(1+2\frac{\tan \theta_s}{\tan \theta_m}\right)\left(-\frac{1}{\cos 2\theta_s}\right) \qquad (8)$$

or

$$f=\frac{-L \cos 2\theta_s}{\left(1+2\frac{\tan \theta_s}{\tan \theta_m}\right)} \qquad (9)$$

It has therefore been demonstrated that the location of the focusing point F as measured by the distance along the central ray of lavelength λ from the polycrystalline sample **13** may be definitely computed for any selected wavelength and Bragg angle relationship of the X-ray beam with respect to the monochromator **11** and the polycrystalline sample **13**.

It should be particularly noted that the most desirable dimensional relationships are obtained at Bragg angle approaching 90°.

For values of θ_s approaching 90° and $\theta_s = \theta_m$, it will be observed that the ratio of the total distance between the point source P and the polycrystalline sample **13** traversed by the central ray to the distance of the focusing point F from the sample **13** as measured along the path traversed by the central ray is equal to 3. Hence very practical dimensional relationship can be obtained at Bragg angles approaching 90°, for if the distance from the sample **13** to the focal point F is selected as 5 cm., then the distance L traversed by the central ray need only be 15 cm., which is an entirely practical arrangement. For lower Bragg angles θ_m it may be readily observed that the ratio of L/f increases rapidly and hence results in less practical spatial positioning of the source of X-rays P with respect to the monochromator **11** and the sample **13**.

Referring now to Figures 2 and 3, there is shown schematically an apparatus for conveniently utilizing the X-ray focusing methods heretofore disclosed. Thus a shielding jacket **20** is provided in surrounding relationship to a focusing X-ray cathode **21** and target anode **22**. That portion of the surface of target anode **22** upon which the high velocity electrons impinge may, if desired, have a removable target **23** mounted thereon in conventional fashion. Thus, by selection of the material of the removable target **23**, it is possible to obtain a variety of discrete bands of wavelengths of the resulting X-rays. The monochromator **11** is then mounted in any convenient fashion within the shielding jacket **20** and in such position as to receive a beam of X-rays emitted from a point or line P of the target surface at a large Bragg angle of incidence.

The electrons are brought to a focus on the target point P by use of any suitably sharp focusing arrangement. The shape of the focus on the point P may be in the form of a point or line, or any other suitably shaped sharp focus. The electron source is within the cathode **21**. Apparatus is provided to permit the angular position of the monochromator **11** to be adjusted with respect to the target **23**, and such adjustment permits the apparatus to be conveniently adapted to employ any one of a large variety of wavelength bands of X-rays.

The shielding jacket **20** is provided with a window portion **20***a* capable of transmitting X-rays therethrough which is disposed opposite the monochromator **11** and in selected spatial arrangement therewith so that all diffracted beams from the monochromator **11** will pass through the window **20***a* to the exterior of the jacket **20**. The polycrystalline sample **13** to be analyzed is disposed exteriorly of the jacket **20** in the path of the diffracted beam from the monochromator **11**. A focusing of the rays diffracted by the polycrystalline sample **13** will then be obtained at a point, such as F, exteriorly of the jacket **20** and hence permits conventional X-ray recording and/or indicating equipment (not shown) to be positioned at this point.

In the modified arrangement shown in Figure 4, the shielding jacket **20** is provided with two X-rays transmitting windows **20***b* and **20***c*, respectively. The monochromator **11** is then positioned to the exterior of the jacket **20** and adjacent the X-ray transmitting window **20***b* so that an original beam of X-rays produced from a point P on the target **23** will pass through the window **20***b* and impinge upon the monochromator **11**. The diffracted beam then passes through the window **20***b* in the reverse direction and traverses the interior of jacket **20** to pass through the second X-ray transmitting window **20***c* and then strike the polycrystalline sample **13** again disposed on the exterior of the jacket **20**. The diffracted beam from the sample **13** may then be brought to a focal point F exteriorly of the jacket **20**.

It should be particularly noted that in both embodiments of the preferred forms of apparatus for effecting the focusing of X-rays in accordance with this invention, Bragg angles of diffraction approaching 90° are utilized both in the diffraction by the monochromator **11** and by the polycrystalline sample **13**, and further, the focusing of the X-rays is accomplished without interference of the various diffracted beams with each other. Any other beams originating on the target anode will likewise produce very little interference effects.

It is therefore apparent that the method and apparatus of this invention provides an unusually simple yet highly precise method of accomplishing the focusing of an X-ray beam including a distinct band of wavelengths and, as a result, the accuracy of X-ray analysis has been substantially improved without requiring apparatus of any greater expense or complexity than that conventionally employed in the known methods to produce inferior results.

It will, of course, be understood that various details of construction and application may be modified through a wide range without departing from the principles of this invention, and it is, therefore, not the purpose to limit the patent granted hereon otherwise than necessitated by the scope of the appended claims.

We claim as our invention:

1. The method of X-ray analysis which comprises producing a polychromatic beam of X-rays diverging from a substantially point source and having a central ray of wavelength λ, disposing a flat face of a crystal in the path of said beam to produce a reflected beam, whereby the reflected beam is angularly divergent from said central ray but has a frequency distribution proportional to the angular separation from said central ray, disposing a polycrystalline sample in the path of said reflected beam of X-rays, thereby producing a second reflected beam, and locating an X-ray sensitive indicating medium in the path of said second reflected beam and at a predetermined distance from said sample measured along the path of said central ray of wavelength λ of said second reflected beam, said predetermined distance being equal to

$$\frac{-L\cos 2\theta_s}{1+2\dfrac{\tan\theta_s}{\tan\theta_m}}$$

where L equals the sum of the distances from the source of the X-rays to the polycrystalline sample measured along the path of said central ray of wavelength λ, θ_m is the Bragg angle for said central ray of wavelength λ diffracted by the crystal, and θ_s is the Bragg angle for said central ray of wavelength λ diffracted by the polycrystalline sample.

2. The method of X-ray analysis which comprises producing a polychromatic beam of X-rays from a point source having a central ray of wavelength λ, placing a flat face of a crystal in the path of the beam, locating a sample to be analyzed in the path of the diffracted beam, and positioning an X-ray sensitive indicating medium in the path of the X-ray beam diffracted by the sample at a distance from the sample equal to

$$\frac{-L\cos 2\theta_s}{\left(1+2\frac{\tan\theta_s}{\tan\theta_m}\right)}$$

where L equals the sum of the distances from the source of the X-rays to the sample measured along the path of said central ray of wavelength λ, θ_m is the Bragg angle for the central X-ray wavelength λ diffracted by the crystal, and θ_s is the Bragg angle for the central X-ray of wavelength λ diffracted by the sample.

3. The method of focusing a polychromatic beam of X-rays having a central ray of wavelength λ which beam is reflected from a polycrystalline sample which comprises arranging the flat face of a crystal monochromator intermediately between the source of the polychromatic X-ray beam and the sample so that the polychromatic X-ray beam incident upon the sample is initially diffracted by said monochromator, and positioning the sample with respect to the monochromator so that the ratio of the total distance traversed by the central ray of wavelength λ from the source to the sample to the distance from the sample to the desired focal point measured along the path of the central ray of wavelength λ equals

$$\left(1+2\frac{\tan\theta_s}{\tan\theta_m}\right)\left(-\frac{1}{\cos 2\theta_s}\right)$$

where θ_m is the Bragg angle for the central ray of wavelength λ reflected by the monochromator, and θ_s is the Bragg angle for the central ray of wavelength λ reflected by the sample.

4. The method of focusing a polychromatic beam of X-rays reflected from a polycrystalline sample and having a central ray of wavelength λ which comprises arranging a flat face of a crystal immediately between the source of the polychromatic beam of X-rays and the sample so that the X-ray beam impinging on the sample is initially reflected by said crystal at a Bragg angle approaching 90°, and positioning the sample with respect to the said crystal and the source of the X-rays so that the ratio of the total distance traversed by the central ray of wavelength λ from the source to the sample to the distance from the sample measured along the path of the central ray of wavelength λ at which focusing of the beam is desired, equals

$$\left(1+2\frac{\tan\theta_s}{\tan\theta_m}\right)\left(-\frac{1}{\cos 2\theta_s}\right)$$

where θ_m is the Bragg angle for the central ray of wavelength λ reflected by the crystal, and θ_s is the Bragg angle for the central ray of wavelength λ reflected by the polycrystalline sample.

5. Apparatus for X-ray analysis comprising a source of diverging, polychromatic X-rays producing a beam having a central ray of wavelength λ, a flat faced, crystal monochromator, means for adjustably positioning said crystal monochromator in the path of said beam to produce a diverging reflected polychromatic beam, means for positioning a polycrystalline sample in the path of said diverging reflected beam, thereby producing a converging reflected beam of X-rays, and an X-ray sensitive indicating medium disposed in the path of said second reflected beam at a predetermined distance from said sample measured along the path of said central ray or wavelength λ equal to

$$\frac{-L\cos 2\theta_s}{1+2\frac{\tan\theta_s}{\tan\theta_m}}$$

where L equals the sum of the distances from the source of X-rays to the sample measured along the path of the central ray of wavelength λ, θ_m is the Bragg angle for the central ray of wavelength λ reflected by the monochromator, and θ_s is the Bragg angle for the central ray of wavelength λ reflected by the polycrystalline sample.

6. Apparatus for X-ray analysis comprising a tube envelope enclosing a source of a beam of polychromatic X-rays, said envelope having a first X-ray transmitting window therein permitting a beam of X-rays to pass through said envelope, a monochromator disposed in the path of said polychromatic beam and arranged to produce a first reflected beam of polychromatic X-rays directed through said first window back into said envelope, said envelope having a second X-ray transmitting window disposed in the path of said first reflected beam, means for positioning a polycrystalline sample exteriorly of said envelope and in the path of said first reflected beam, thereby producing a second reflected beam, and an X-ray sensitive recording medium located in the path of said second reflected medium at a predetermined distance from said sample corresponding to the location of a focal point of said second reflected beam.

7. Apparatus for X-ray analysis comprising a tube envelope enclosing a source of a beam of polychromatic X-rays having a central ray of wavelength λ, said envelope having a first X-ray transmitting window therein permitting said polychromatic beam of X-rays to pass through said envelope, a monochromator disposed in the path of said beam and arranged to produce a first reflected polychromatic beam of X-rays directed through said first window into said envelope, said envelope having a second X-ray transmitting window disposed in the path of said first reflected beam, means for positioning a polycrystalline sample exteriorly of said envelope and in the path of said first reflected beam, thereby producing a second reflected beam, and an X-ray sensitive recording medium located in the path of said second reflected medium at a distance from said sample measured along the path of said central ray of wavelength λ equal to

$$\frac{-L\cos 2\theta_s}{1+2\frac{\tan\theta_s}{\tan\theta_m}}$$

where L equals the sum of the distances from the source of the X-rays to the sample and is measured along the path of the central ray of wavelength λ, where θ_m is the Bragg angle for the central ray of wavelength λ reflected by the monochromator and θ_s is the Bragg angle for the central ray of wavelength λ reflected by the polycrystalline sample.

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