Feb. 23, 1960 S. WEISSMANN 2,926,258

X-RAY REFLECTION MICROSCOPY AND DIFFRACTION APPARATUS AND METHOD

Filed July 9, 1958 5 Sheets-Sheet 1

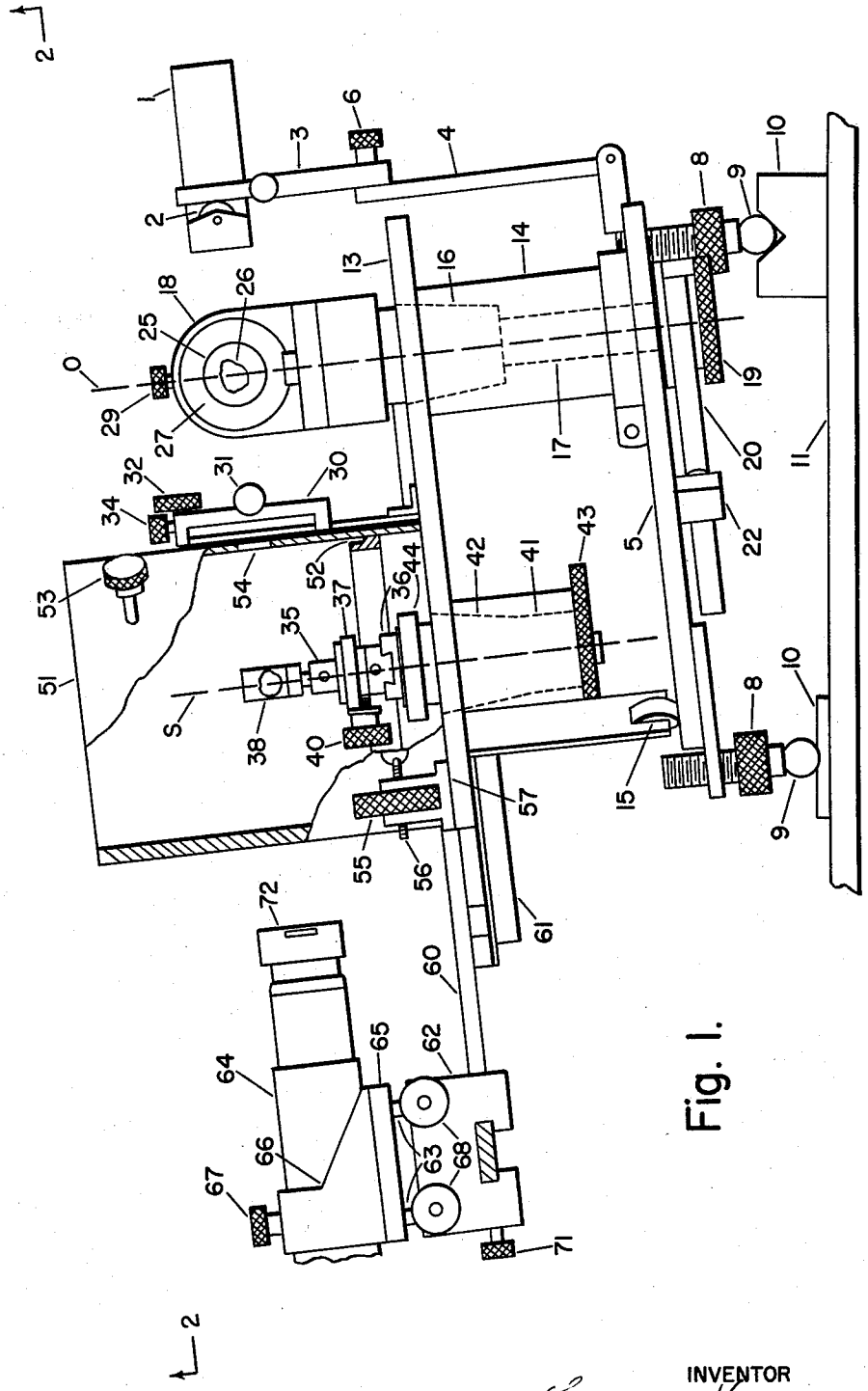


Fig. 1.

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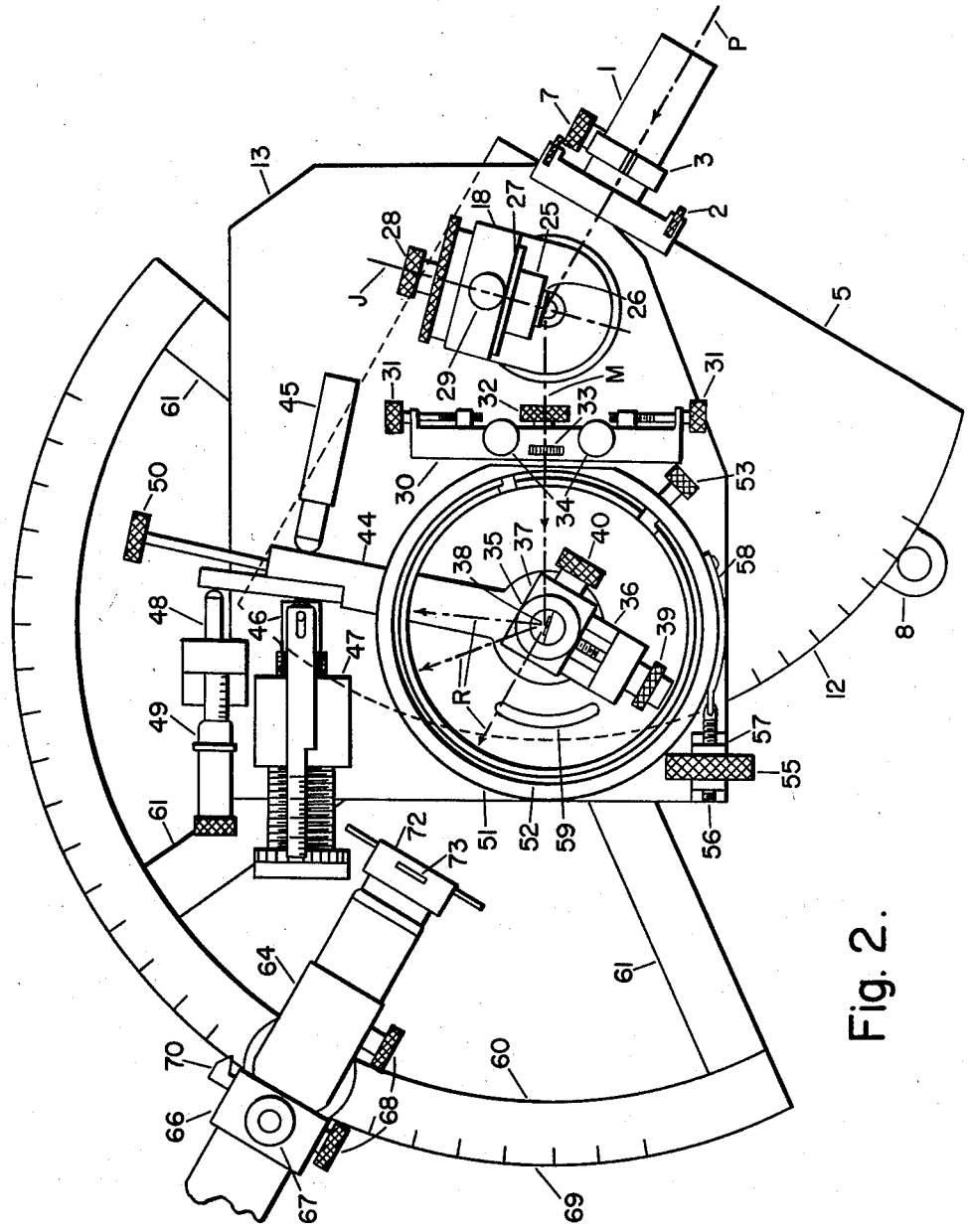


Fig. 2.

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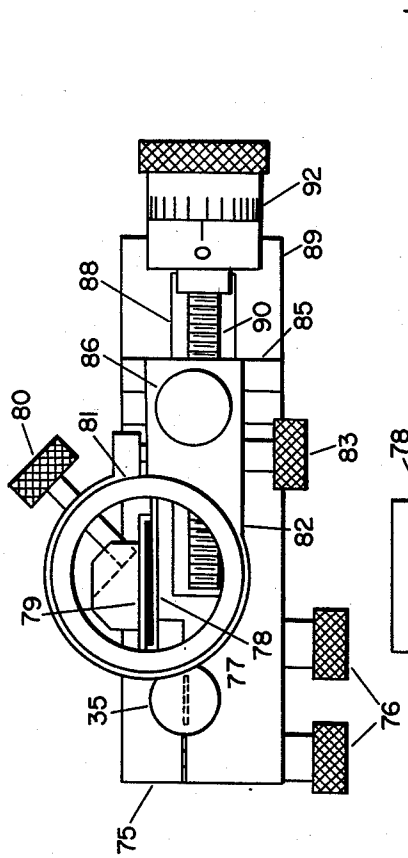


Fig. 4.

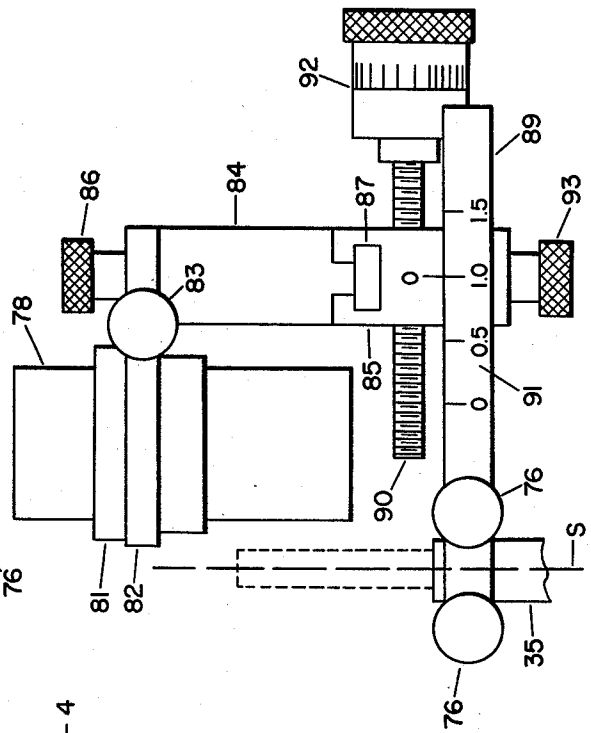


Fig. 3.

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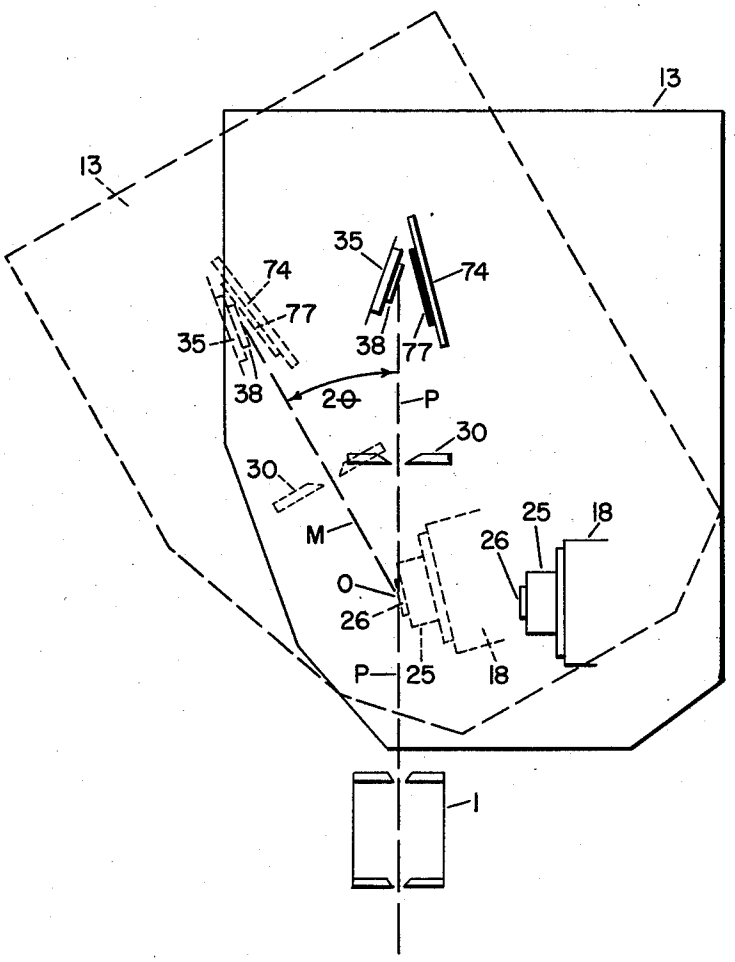


Fig. 5.

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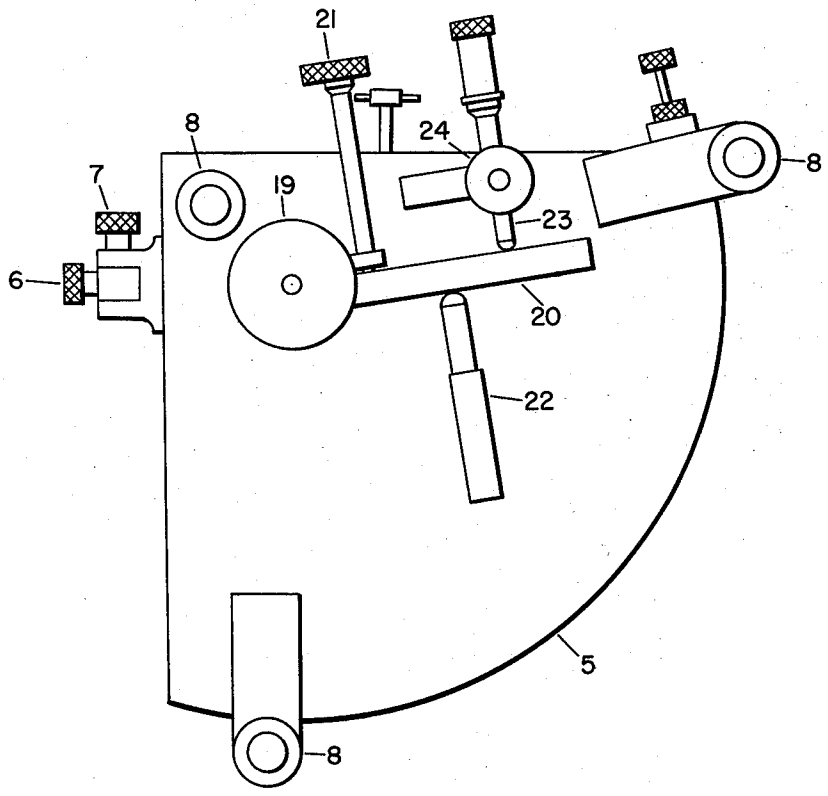


Fig. 6.

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2,926,258

X-RAY REFLECTION MICROSCOPY AND DIFFRACTION APPARATUS AND METHOD

Sigmund Weissmann, Metuchen, N.J.

Application July 9, 1958, Serial No. 747,520

13 Claims. (Cl. 250—53)

The instant invention relates to a novel method by means of which the topographical relation of crystallites of a polycrystalline material as well as the topographical relation of lattice inhomogeneities of single crystals can be analyzed through the successive application of various portions of the spectrum of X-radiation. The invention relates also to a novel apparatus for accomplishing the method. A preferred embodiment of the invention relates to a novel method and apparatus by means of which the crystallites of a polycrystalline material thus analyzed for their topographical relation can be further analyzed, inter alia, for lattice imperfections, structural irregularities, and substructure. More specifically, the invention relates to an improved method and apparatus for recording the topographical characteristics of crystalline materials by X-ray reflection microscopy. The preferred embodiment of my invention combines X-ray reflection microscopy with X-ray diffraction to record topographical characteristics of polycrystalline materials, thereby permitting further analysis of the atomic or molecular structure of such materials.

The investigation of crystalline materials by prior art X-ray reflection microscopy methods usually requires the use of filtered or unfiltered X-radiation and, in some rare cases, the use of crystal monochromatized X-radiation.

If unfiltered or even filtered X-radiation is employed the topographical relation of the crystallites making up the crystalline material may be satisfactorily revealed in some instances, but due to the limitation of the resulting resolution many important structural details of the crystallites will remain undisclosed. The limitation of the resolution is imposed by the spread of the wave-lengths employed and by the resulting divergence of the X-ray beam. If monochromatic radiation is used the resolution is greatly increased, since the probing beam consists essentially of one wave-length component and the beam can be made quite parallel. However, because of the stringent reflecting conditions imposed, only few crystallites or sometimes only portions of the crystallites will reflect and the topographical relation of the crystallites will be greatly suppressed and very frequently made completely unintelligible.

Successive application of various portions of the X-ray spectrum to X-ray reflection microscopy has never been heretofore attempted since, prior to my invention, one could never make certain that the identical crystallites will be in reflecting position if different wave-lengths are used.

The method and apparatus of my invention represent an advance in the art in that there is now provided means to successively apply various portions of the X-ray spectrum to X-ray reflection microscopy insuring a high degree of resolution and topographical correlation and insuring that the identity of the reflecting crystallites is preserved.

Moreover, since my X-ray micrography method can be combined by a tracer method with diffraction analysis,

the identical crystallites investigated by X-ray micrography can be analyzed with respect to their chemical constitution and crystal structure and can also be analyzed with respect to their lattice imperfections, structural irregularities, and substructure, thereby permitting one to obtain not only qualitative but also quantitative information thereof.

The method of my invention as applied to the substructure of metals and alloys discloses the coexistence of various orders of magnitude of substructural entities. The invention permits the quantitative determination of the size, disorientation and lattice misalignment for the various orders of the substructural entities. Furthermore, the nature of small-angle boundaries in metals and alloys may be investigated.

The novel apparatus of my invention provides a useful tool in the study of deformation of metals and alloys since a plurality of crystallites may simultaneously be investigated without removing the natural constraints imposed by the surrounding grains as in prior art methods. Said crystallites may be investigated as if they were isolated single crystals without relinquishing the statistical advantages which the investigation of polycrystalline materials usually offers.

Accordingly it is a principal object of the instant invention to provide a method and apparatus by means of which the topographical characteristics of a crystalline material may be elucidated.

Another object of my invention is the provision of a method and apparatus by means of which the topographical relationship of the crystallites as well as the topographical relationship of the substructural entities of the crystallites may be disclosed.

Another object of the invention is the provision of an apparatus by means of which more details about the texture and topographical relationship of crystallites of a polycrystalline material may be analyzed than by prior art techniques.

Another and important object of the invention is the provision of a method and apparatus by means of which the topographical relationship of the crystallites of a polycrystalline material may be analyzed by X-ray micrography and the crystallites thus analyzed may be further analyzed for lattice imperfections, structural irregularities and substructure by X-ray diffraction techniques.

The subject invention is a result of my discovery that X-ray micrographs of fine-grained or coarse-grained crystalline materials as well as single crystals can be obtained employing successively various portions of the X-ray spectrum to disclose structural details of single crystals and of the individual crystallites of a polycrystalline material while maintaining the identity of said crystallites. I have found, furthermore, that crystallites thus analyzed for the surface texture or topographical relationship can be further analyzed for inclusions of materials of different crystal structure or of different lattice parameters if the images of the reflecting crystallites are traced outwardly at increasing distances from the specimen surface until the images are recorded on the film of a Debye-Scherrer camera, whereby the reflecting crystallites may be identified as to their chemical constitution or crystal structure, or both, by conventional powder diffraction analysis of the Debye-Scherrer lines. An important embodiment of my invention involves the qualitative and quantitative investigation of lattice defects, lattice inhomogeneities and substructure of crystallites making up a polycrystalline test specimen, and is based on the double crystal diffractometer principle, according to which each reflecting crystallite of the specimen surface may independently be regarded to function as the second crystal of a double crystal diffractometer.

A distinct improvement of the instant invention over prior art techniques is due to the detailed disclosure of the topographical relationship of the polycrystalline materials obtained by the successive utilization of various portions of the X-ray spectrum. Thus it is possible to correlate the X-ray micrograph obtained by my method with the corresponding micrograph obtained by conventional optical microscopy. Fine-structural details disclosed by X-ray micrography obtained by my method may also be correlated with micrographs obtained by electron microscopy. Furthermore, through the application of the photographic tracer technique, it is made possible to record directly, by means of an electronic radiation detector, the intensities of spot reflections emanating from polycrystalline specimens, since the radiation detector can be placed precisely at the positions where the spot reflections will occur. Consequently, precise quantitative data can be obtained of lattice imperfections, substructure characteristics, inclusions and precipitates of fine-grained and coarse-grained polycrystalline materials, all correlated to the surface texture of said specimens by X-ray reflection microscopy.

Further features and advantages of my invention will be readily apparent from an inspection of the accompanying drawings and the detailed description of my invention which follows.

In the drawings:

Figure 1 is an elevational view of the embodiment of the apparatus of my invention which includes the X-ray micrography assembly in combination with the X-ray diffraction assembly.

Figure 2 is a detailed plan view of the assembly illustrated in Figure 1 taken along the line 2—2 in Figure 1, with the platform member rotated relative to the base to permit better visualization of the path of the X-ray beams.

Figure 3 is a side elevational view of the photographic film holder assembly for outward tracing of X-ray reflections.

Figure 4 is a plan view taken along the line 4—4 of Figure 3 illustrating the filmholder assembly of Figure 3.

Figure 5 is a fragmentary view of Figure 2 illustrating the relative position of the X-ray beam, monochromatizing crystal and specimen when the platform member is rotated relative to the monochromatizing crystal to permit impingement of crystal monochromatized radiation on the specimen surface.

Figure 6 is a plan view of the lower surface of the base plate member.

The apparatus suitable for recording the topographical characteristics of a crystalline material by X-ray micrography comprises in conjunction with an X-ray tube and means for controlling the X-ray output thereof, first holder means for mounting a monochromatizing crystal, means for rotating the monochromatizing crystal into reflecting position, means for retracting said monochromatizing crystal if the primary beam is to be used as the incident beam, a platform mounted for rotation about an axis coincident with the axis of rotation of the monochromatizing crystal in a manner such as to rotate the platform by twice the Bragg angle of the reflecting planes of the monochromatizing crystal, stationed on said platform and rotatable thereby as a unit means for mounting a crystalline test specimen for impingement by incident X-ray beam, and, in the case of single crystal test specimens, means for bringing said test specimen into Bragg reflection when impinged by the incident X-ray beam, and means for stationing an X-ray film in close contact with the reflecting surface of the test specimen in a manner such as to permit recording of reflection images emanating from the surface of the crystalline material in the form of an X-ray micrograph, whereby X-ray micrographs of the same specimen area are obtained with unfiltered, filtered and crystal monochromatized radiation either singly or in combination.

Pursuant to an embodiment of my invention wherein X-ray micrography is combined with X-ray diffraction for analysis of polycrystalline test specimens the assembly requires in addition to the members above described, preferably, means for rotating the test specimen, a cylindrical camera coaxial with the test specimen or, in the case that the test specimen is rotated, coaxial with the axis of rotation of the test specimen, a photographic film holder for taking X-ray micrographs at the surface of the specimen and for outward tracing of the reflected images at increasing distances from the specimen surface, means for removing said film holder to permit unimpeded analysis of the X-ray diffraction images on a cylindrical film inserted in said cylindrical camera by the Debye-Scherrer method.

In accordance with the preferred embodiment of my invention wherein X-ray micrography is combined with X-ray diffraction for the qualitative and quantitative analysis of polycrystalline test specimens for lattice inhomogeneities and substructure characteristics the assembly requires in addition to the members above described, means for recording the reflection curves (rocking curves) of the individual crystallites of said specimen. This may be achieved by controlled rotation of the test specimen through small angular ranges in discrete angular intervals and controlled rotation of cylindrical film between each of said discrete angular specimen rotation. However, if the crystallites of the test specimen are large enough so that the reflected intensities can be recorded directly by an electronic radiation detector, the apparatus will include, in addition to the X-ray micrography assembly hereinabove described, in combination with film means for recording the reflection curves of the individual crystallites of said specimen, a radiation detector and means for positioning said radiation detector at locations corresponding to the reflection images recorded on said cylindrical film. Preferably, the latter is accomplished by employing an optically transparent insert camera, suitably made of plastic, which aids in the visualization of the location of the reflected images of the crystallites when the inserted exposed film has been developed. Since said insert camera is removable, the reflected intensities of the individual crystallites of said specimen can be directly recorded by said radiation detector when properly positioned by the aforementioned technique. Thus reflection curves (rocking curves) of the individual crystallite are directly obtained when said crystallites are rotated through their angular range of reflection and the intensity variation has been recorded as a function of said rotation.

The method, briefly stated, comprises impinging at a small grazing angle onto the surface of a polycrystalline test specimen divergent X-radiation, preferably unfiltered, utilizing an X-ray tube voltage considerably above that of the excitation potential of the characteristic radiation, recording on film placed close to the surface of said specimen the images of the crystallites of said specimen reflecting said radiation, lowering the tube voltage below the excitation potential of the characteristic radiation thereof, recording on film placed close to the surface of said specimen the images of the crystallites of said specimen reflecting said radiation known in the art as continuous radiation, interposing into the path of the primary X-radiation a monochromatizing crystal in reflecting position, impinging parallel, crystal monochromatized radiation on the same surface of said test specimen, superimposing the images obtained utilizing crystal monochromatized radiation on the images obtained by the primary beam utilizing said continuous radiation, recording on a separate film placed close to the surface of said test specimen the images of the crystallites reflecting parallel, monochromatized X-radiation,

whereby identification of the many crystallites reflecting unfiltered radiation with the few crystallites registered on the film utilizing crystal monochromatized radiation permits disclosure of fine-structural details of said polycrystalline test specimen. Preferably, the method includes the additional steps of tracing out the images recorded with crystal monochromatized radiation at increasing distances from the specimen surface until they are recorded on the Debye-Scherrer lines of a cylindrical camera, whereby the spot reflections on the Debye-Scherrer lines and the crystallites on the specimen surface giving rise to said spot reflections are being correlated. The analysis of said crystallites with regard to the chemical constitution and crystal structure may be carried out by methods well known in the art and correlated to the topographical relation and fine-structural details of said crystallites.

Pursuant to a preferred embodiment of this method wherein X-ray micrography is combined with X-ray diffraction analysis for the qualitative and quantitative analysis of lattice inhomogeneities and substructure characteristics, the method includes the additional steps of obtaining reflection curves (rocking curves) of the individual crystallites by changing the angular position of the specimen and recording the intensity of the reflections of the individual crystallites as a function of specimen rotation either on an X-ray sensitive film or directly by means of an electronic radiation detector. If a film is used as a recording device said angular rotation of the specimen is carried out in discrete angular intervals accompanied by small film shifts between each angular interval of specimen rotation to separate the individual intensity contributions to the reflection curve. If a radiation detector is used as a recording device, said detector is placed by means of said photographic tracer technique at the positions where reflections will occur and said specimen is slowly rotated over a preselected angular range.

It is well known to those skilled in the art the obtained reflection curves of the individual crystallites are related to the lattice imperfections of same and can be analyzed by well known methods.

The method is equally applicable to single test crystals and comprises all the steps outlined above, except that said crystal has to be first put into Bragg reflection to yield an X-ray micrograph and that the reflected recorded images now refer to the substructural entities of said test crystal.

Referring to Figures 2 and 6 numeral **1** designates a collimator through which the primary X-ray beam P passes emerging from an X-ray diffraction tube (not shown). Said collimator **1** is fitted with a slit system which is adjusted by adjustment screws **2** controlling thereby the divergence of the X-ray beam. Said collimator **1** is held by means of a split clamp **3** to the collimator support **4**. Said collimator support **4** is attached to the base plate **5** by means of nuts and bolts. The height and angle of said collimator support **4** with respect to said base plate **5** is adjustable so that the apparatus can be adapted to various commercial diffraction units. Locking screw **6** locks the height and locking screw **7** locks the tilt of said collimator support **4**. It is understood, however, that collimator **1** may be mounted independently of the assembly. Said base plate **5** is supported by three adjustable legs **8**, suitably of stainless steel. Said adjustable legs **8** are countersunk to fit steel balls **9**. Said steel balls **9** rest on solid metal blocks **10** bolted to the top plate **11** of the commercial diffraction unit. Said base plate **5** is suitably provided in the shape of a quadrant of a circle. However, other shapes may be satisfactorily used. The rim of the upper face of said base plate **5** is preferably provided with a graduation scale **12** in degrees as shown to facilitate rotational alignment of the upper platform **13** around axis O. A pointer (not shown) attached to the rear of said upper platform **13** indicates on said graduation scale **12** the desired rotational position of said upper platform **13**. Said upper platform **13**, suitably of stainless steel, is supported on said base plate **5** by a shaft **14** and two outer rollers **15**. Said shaft **14** passes through said base plate **5** and is supported by a bearing (not shown) so as to allow only rotational movement. Said upper platform **13** can be rotated around the vertical axis O of shaft **14**. Passing through said shaft **14** and supported by a tapered bearing **16** is a smaller shaft **17**. On the upper end of said shaft **17** the first crystal holder **18** is mounted by means of set screws. On the lower end of said shaft **17** is mounted a drum **19** on which fits a collar and arm **20** which may be locked in position on the drum **19** by locking screw **21**. Said arm **20** is spring loaded **22** against a micrometer spindle **23** which may be locked by locking screw **24**. Said crystal holder **18** comprises a cylinder **25** upon the face of which is mounted the monochromatizing crystal **26**. Said cylinder **25** is mounted within a barrel **27**. Said cylinder **25** is retractable by means of the adjustment screw **28** permitting the monochromatizing crystal **26** to be retracted if the direct primary X-ray beam P is used as the incident beam. Said barrel **27** can be rotated around an axis J which is perpendicular to said axis O for exact adjustment of the monochromatizing crystal **26**. Said adjustment is particularly desirable if the reflecting (hkl) planes and the surface of the monochromatizing crystal **26** do not coincide. Said barrel **27** can be locked by locking screw **29** if the desired adjustment has been made. If said monochromatizing crystal **26** has been adjusted for the proper Bragg reflection by action of member **23** the reflected beam M will be monochromatized. **30** represents a spectroscopic slit system mounted on said upper platform **13**. Said spectroscopic slit system **30** comprises a vertical slit controlled by adjustment screws **31** and a horizontal V-shaped slit controlled by adjustment screw **32** which activates a gear and rack arrangement **33**. The positions of said slits are locked by locking screws **34**. By means of said spectroscopic slit system **30** the cross section of the incident X-ray beam is controlled and parasitic X-ray scattering is prevented. It will be understood, however, that various other slit systems may achieve the same effect. **35** is a specimen holder. Said specimen holder is fitted with dove-tailed cross slides **36** and **37** for horizontal adjustment of the position of specimen **38**. Said adjustment is obtained by means of adjustment screws **39** and **40**. Said specimen holder **35** is mounted by means of set screws on a shaft **41**. Said shaft **41** is supported by a tapered bearing **42** mounted beneath said upper platform **13**. A handwheel **43** is fitted to the lower end of said shaft **41** to facilitate rough angular positioning of the specimen **38**. An arm and collar **44** is fitted to the shaft **41**. Arm **44** is spring-loaded **45** against a spindle **46** of a differential micrometer **47** or against the spindle **48** of a standard micrometer **49**. By means of the differential micrometer **47** specimen rotations are obtained that are of the order of magnitude of seconds of arc, whereas by means of the standard micrometer **49** specimen rotations are obtained of the order of magnitude of minutes of arc. Arm **44** can be locked to shaft **41** by means of locking screw **50**. **51** represents a cylindrical camera which is inserted into a circular groove (not shown) of the upper platform **13**. The cylindrical camera **51** is made of transparent plastic material, suitably Lucite, and has its center of curvature coincident with the vertical axis S of shaft **41**. The inside of the cylindrical camera **51** is fitted with a groove **52** in which a film (not shown) covered with black paper can be inserted and said film can be fastened by means of two locking screws **53**. Said film will record the X-rays R reflected from the crystallites of the specimen **38**. The cylindrical camera **51** is provided with an aperture **54** to let the primary beam enter the camera **51**. The cylindrical camera **51** can be

rotated around an axis coincident with axis S by means of a nut 55 and screw 56. The screw 56 is supported by a bracket 57. Said bracket is bolted to the upper platform 13. Soldered to the end of the screw 56 is a metal strip 58 which is screwed to the outside of the cylindrical camera 51. 59 represents a slot into which a trap (not shown) for the incident X-ray beam may be inserted if required. Said trap is fastened to the upper platform 13 by means of a locking screw (not shown). The use of said trap is only required if the specimen investigated does not absorb the incident X-ray beam entirely. 60 represents a circular track which is bolted to the upper platform 13 by means of three supporting radial arms 61. A grooved block 62 slides on track 60. Two rods 63 fit into two holes of block 62. Said rods support the radiation detector 64 by means of a block 65 and sleeve 66 into which said radiation detector 64 can be clamped by means of locking screw 67. The height of radiation detector 64 is adjustable by means of rods 63 and can be locked by means of locking screws 68. The outer rim of the upper face of track 60 is provided with a graduation scale 69 in degrees, as shown. Angular alignment of radiation detector 64 is facilitated by means of graduation scale 69 and a pointer 70 fastened to the side of the grooved block 62. The angular position of said radiation detector is locked by locking screw 71. Radiation detector 64 is fitted with a receiving slit 72 limiting the amount of X-radiation recorded by the detector. A film may be placed into a slot 73 located behind the receiving slit 72. Said film when exposed to the reflected X-radiation entering receiving slit 71 may function as a guide as to how many reflections are being recorded thus indicating how the receiving slit 72 should be adjusted to eliminate the undesirable reflections from recording.

The precision photographic film holder assembly 74 is attachable to the specimen holder 35 by means of a split clamp 75 and lock screws 76. A film or photographic plate sensitive to X-radiation R is clamped against an adjustable plate 78 of said film holder by means of a clamping plate 79 and lock screw 80. The adjustable plate 78 is mounted within a shouldered ring 81. The shouldered ring 81 is supported by a split collar 82 and locked in place by lock screw 83. Split collar 82 permits height adjustment and rotation around an axis parallel to S of film or photographic plate 77. The split collar 82 is screwed on a column 84 which slides in a grooved block 85. It may be locked in position by means of locking screw 86 and locking plate 87. The grooved block 85 fits in a slot 88 in the filmholder base plate 89. The position of block 85 may be adjusted by means of adjusting screw 90. The relative position of block 85 is determined to an accuracy of $\frac{1}{1000}$ of an inch by means of linear scale 91 and micrometer dial 92. The block 85 is locked in position by means of locking screw 93. The position of the photographic plate 77 in a plane perpendicular to the axis S is determined by the relative movements between members 84 and 85 and members 85 and 89 whereby these movements are perpendicular to each other.

A more comprehensive understanding of the instant method will be greatly aided by reference to Figures 1, 2, 3, 4 and 5.

The polycrystalline test specimen 38 is irradiated at a small grazing angle by a divergent, preferably unfiltered X-ray beam P passing through a collimator 1 and a spectroscopic slit system 30 which controls the cross-section of the incident X-ray beam P. A tube voltage is applied which is considerably above the excitation potential of the characteristic radiation of the target material of said X-ray tube (e.g. 35 kv. for copper radiation) so that the primary incident X-ray beam consists of continuous and characteristic radiation. An X-ray micrograph of the crystallites reflecting from the surface of said test specimen 38 is obtained by recording the reflected images of said crystallites on a fine-grained X-ray film suitably held in a film holder placed in close contact to the specimen surface as schematically shown in Figure 5. The X-ray micrograph obtained in this manner supplies a great deal of information concerning the surface texture and topography of the crystallites since due to the employment of the divergent and unfiltered incident X-ray beam P a great many crystallites will be in Bragg reflection and will thus be recorded. However, fine-structural details of the crystallites cannot be disclosed by said micrograph because of the limitation of resolution imposed by the application of said divergent, polychromatic radiation P. Furthermore, said X-ray micrograph is useless from the viewpoint of X-ray analysis since the great number of reflections emerging from the surface of the specimen can not be traced outward and subjected to an X-ray analysis without losing the identity of the images. To increase the resolution of the structural details and make X-ray diffraction analysis of said crystallites possible, following steps have to be taken. A monochromatizing crystal 26 is interposed in the path of the primary beam P by means of the adjustment screw 28 of the crystal-holder 18 and rotated around axis O into Bragg reflection as schematically shown in Figure 5. The parallel crystal monochromatized radiation M subtends with the primary beam P and angle of 2θ where θ is the Bragg angle of the reflecting (hkl) planes of the monochromatizing crystal 26. The upper platform 13 is rotated around axis O by an angle of exactly 2θ as shown in Figure 5 keeping said monochromatizing crystal 26 rigidly locked in reflecting position by means of locking screws 24 and 29. The experimental arrangement thus obtained is indicated by dotted lines in Figure 5. Since upper platform 13, specimen 38 mounted on the specimen holder 35, and film holder assembly 73, as well as the spectroscopic slit system 30 rotates as a unit around axis O, the surface of specimen 38 is now impinged by the crystal monochromatizing radiation M. After the tube voltage has been increased an X-ray micrograph is obtained which, although registering fewer reflections than that obtained by unfiltered radiation due to the stringent reflecting conditions imposed, will exhibit a greatly increased resolution for fine-structural details. Furthermore, whereas the continuous radiation, contained in the unfiltered beam P, reveals lattice curvature as dark images, since each misaligned lattice region will reflect a wave-length of the spectrum corresponding to its Bragg condition of reflection, the irradiation with crystal monochromatized radiation M pins down lattice regions of the same orientation. Thus the alternate applications of continuous and crystal monochromatized radiation for the investigation of fine-structural details and surface texture are mutually complementary.

Depending on the target material of the X-ray tube and on the reflection angles of the monochromatizing crystal 26 monochromatic radiation corresponding to the different wavelengths of the characteristic spectrum can be obtained. Consequently, by appropriate rotation of the monochromatizing crystal 26 to the various Bragg reflections and the corresponding rotation of said upper platform 13 around axis O, as shown in Figure 5, different portions of the characteristic X-ray spectrum may be used as the X-ray beam incident on the surface of test specimen 38, thus enhancing on the X-ray micrographs the resolution for structural details of the reflecting crystallites of said specimen.

To identify and correlate the images of the reflecting crystallites recorded on an X-ray micrograph employing crystal monochromatized radiation M to the corresponding images of the reflecting crystallites recorded on an X-ray micrograph employing a divergent, unfiltered beam P, the X-ray beam is artificially decomposed in steps. First it is decomposed into a beam consisting of continuous (heterogeneous) radiation only, and subsequently, into a beam of crystal monochromatized radiation,

whereby the X-ray micrograph employing the latter type of radiation is superimposed over the X-ray micrograph employing the former type of radiation. To achieve this end an X-ray micrograph of the surface of said specimen **38** is taken with the identical experimental arrangement that was used when the X-ray micrograph was obtained employing the divergent, unfiltered primary X-ray beam P, as shown in Figure 5 by solid lines, except that now the tube voltage employed is below the critical excitation potential of the characteristic radiation. Consequently, a beam consisting only of continuous radiation impinges on specimen **38**. Actually two X-ray micrographs are taken with said arrangement. One X-ray micrograph is developed and kept for the purpose of reference and comparison, while the second exposed but undeveloped micrograph is rigidly retained in close contact with the specimen surface. The X-ray micrograph obtained in this manner is identical with the one previously obtained employing the primary beam P, except that due to the low operating voltage the contribution of the characteristic radiation and also that of the very short wave-length portion of the spectrum is missing.

The next step for the identification of the images of said crystallites involves the interposition of the monochromatizing crystal **26**, set in reflection position, into the path of the primary beam P, rotation of upper platform **13** around O by 2θ thus obtaining the experimental arrangement shown in Figure 5 by dotted lines, and superimposing on the X-ray micrograph obtained with continuous radiation, the X-ray micrograph obtained with crystal monochromatized radiation M. The exposure time using crystal monochromatized radiation M is purposely prolonged and the operating voltage is set considerably above the excitation potential of the characteristic radiation so that the contribution of the monochromatized radiation to the superimposed X-ray micrograph is clearly tagged. By inspection and comparison of all X-ray micrographs hereinabove obtained the identity of the reflecting crystallites is clearly established.

The same method of X-ray micrography can be applied to the investigation of single test crystals except that in order to obtain an X-ray micrograph the single test crystal must first be oriented to give a Bragg reflection. Instead of investigating the topography of crystallites, as in the case of polycrystalline materials, one investigates in the case of single crystals the topography of substructural entities.

It is understood that if said X-ray micrograph method is employed without the diffraction analysis which will be described in detail below, the apparatus to accomplish this end can be greatly simplified. For this purpose it is necessary to retain only: the base plate **5** with all the members attached to it intact (members **1** till and including **12**), the upper platform **13** with all the members which insure rotation around the axis O (members **14** and **15**), the crystal holder **18** with the monochromatizing crystal **26** and all the members which aid in adjusting and locking said crystal **26** (members **16**, **17**, **19**, **20**, **21**, **22**, **23**, **24**, **25**, **27**, **28**, **29**), the spectroscope slit system **30** with its members (**31**, **32**, **33**, **34**). However, instead of the specimen holder **35** a commercial X-ray goniometer head can be used and instead of the film holder assembly **74** any fixture which brings the X-ray film in close contact with the specimen surface will suffice.

All the other members described in said apparatus are not essential for the method involving X-ray micrography alone.

Pursuant to a special embodiment of my method the polycrystalline specimen, investigated by said method of X-ray micrography, may be further analyzed for chemical constitution, crystal structure, inclusions either singly or in combination. In case of single crystals it is principally applicable to inclusion studies. To achieve this end the images of the crystallites obtained on the X-ray micrograph employing crystal monochromatized radiation are traced outward by recording the images photographically at increasing distances from the specimen surface until they are recorded on the Debye-Sherrer lines of the cylindrical film inserted in the cylindrical camera **51**. The outward tracing of said images is achieved by means of the photographic film holder assembly **74** which can be attached to the specimen holder **35** by means of a split clamp **75**. When the images are traced outward to the circumference of the cylindrical camera **51** the film holder assembly **74** is removed and the images are recorded on said cylindrical film. From the analysis of the Debye-Sherrer lines by methods well known in the art, the crystal structure and chemical constitution of the crystallites can be determined. Likewise, the presence of inclusions and precipitates can be disclosed by my method of X-ray micrography and correlated by means of the above described photographic tracer technique to the reflection images of the corresponding Debye-Scherrer lines and can, therefore, be analyzed for their chemical constitution and crystal structure.

In a preferred embodiment of my invention X-ray micrography is combined with X-ray diffraction analysis to obtain qualitative as well as quantitative information regarding lattice inhomogeneities and substructure characteristics. Polycrystalline materials as well as single crystals may be investigated by said method.

According to said method the polycrystalline materials may be first investigated by my method of X-ray micrography and correlated by the above described photographic tracer technique to the corresponding reflection images of the Debye-Scherrer lines on the cylindrical film, and subsequently further analyzed by a method which is based on the principle of the X-ray double crystal diffractometer, also known in the art as X-ray double crystal spectrometer. In accordance with that principle each reflecting crystallite or substructural entity may independently be regarded to function as the second crystal of a double crystal diffractometer if the incident beam has been monochromatized by reflection from a single crystal. Consequently, if the crystallites are impinged by crystal monochromatized radiation and if said crystallites are rotated through their angular range of reflection and if the reflected intensities of said crystallites are recorded as a function of specimen rotation, one obtains reflection curves, also known in the art as rocking curves. These reflection curves are a criterion of the lattice misalignment of said crystallites and by measuring the width at half maximum, known in the art as half-width, or by measuring the integrated intensity which is equivalent to the area under the reflection curve, one obtains valuable quantitative information of the lattice misalignment of said crystallites. To achieve this end the specimen **38** impinged by parallel crystal monochromatized radiation M is rotated in discrete angular intervals by means of the differential micrometer **47** if very small specimen rotations of the order of seconds of arc are required or by means of the standard micrometer **49** if specimen rotations of the order of minutes of arc are required. If the recording of the intensities of the reflections is carried out on a cylindrical X-ray film then the cylindrical camera **51** in which the said film is inserted is rotated by means of nut **55** and screw **56** a small amount between each said discrete angular specimen rotation, so that the reflections corresponding to each angular rotation of the specimen **38** are clearly separated. As a result of this procedure one obtains for each reflection of a crystallite an array of spots on the Debye-Scherrer lines of said cylindrical film which represents a visual manifestation of the reflection curve of said crystallite. The intensities of the array of spots can be obtained either visually by comparing the spot intensities with a calibrated photographic strip or by means of a microdensitometer or quickly by means of a

positive-print method. (D. L. Evans and S. Weissmann, J. Opt. Soc. Americ. 43, 1183 (1953)).

If the intensities of the arrays of spots are strong enough so that they can be recorded directly by means of an electronic radiation detector **64** the exposed cylindrical film on which the spot reflections have been recorded is developed and inserted back in camera **51**. Since camera **51** is optically transparent the location of the reflected images is visually aided and the radiation detector **64** may be properly positioned by means of the positioning devices (members **60**, **63**, **67**, **68**, **69**, **71**). The receiving slit **72** is adjusted in a manner that only one spot reflection is recorded. Subsequent to the positioning of the radiation detector **64** the cylindrical camera **51** and film are removed and the intensities as a function of specimen rotation are directly registered by said detector. The radiation detector **64** may be connected via an amplification system to an automatic recorder so that the reflection curve of the individual crystallite can be automatically charted. This procedure can be repeated for as many reflections of crystallites as is desired to record. If any doubt should arise that the receiving slit **72** has not been adequately adjusted so that more than one reflection is simultaneously recorded for any one angular specimen position, a film strip is placed in slot **73** located behind the receiving slit **72** and exposed to the radiation entering said receiving slit. By inspection of said film strip after development the exact adjustment of slit **73** is controlled. It should be understood that various experimental variations are possible. Thus the intensities of the reflected crystallites as film placed in front of the window of the radiation de- a function of specimen rotation may be recorded by a tector **64** as well as by the radiation detector **64** itself. Said combination of recording by film and radiation detector is mutually complementary since small substructural entities of the crystallites are resolved by said film, whereas the radiation detector **64** will integrate the intensity contribution of said small substructural entities, thereby permitting to disclose substructural entities of a larger order of magnitude.

It is also possible to record the intensities reflected by the crystallites from the surface of specimen **38** as a function of specimen rotation by means of an X-ray sensitive film placed in close contact with said surface and to correlate the X-ray micrographs thus obtained by means of the above described photographic tracer technique to the corresponding images obtained on the Debye-Scherrer lines of the cylindrical film. Said technique will permit to disclose the variation of surface texture of said crystallites as a function of specimen rotation and simultaneously permit obtaining quantitative data of the lattice misalignment of said crystallites since the reflection curves of said crystallites can be obtained from the analysis of the corresponding array of spots on the Debye-Scherrer lines, as previously described. Likewise, said X-ray micrographs obtained as a function of specimen rotation can be correlated by means of said photographic tracer technique to the corresponding intensity values registered by the radiation detector **64**.

It will also be understood that the above described methods for investigating the substructural characteristics and lattice inhomogeneities of crystallites in polycrystalline materials are equally applicable to single crystal test specimens. Thus it is equally possible to study the surface texture of single crystals by X-ray micrography irradiating the single test crystal successively with heterogeneous and crystal monochromatized radiation, permitting thereby to disclose surface texture and fine-structural details of the irradiated surface, and to subject said single test crystal to a quantitative analysis for lattice misalignment by rotating said single test crystal around an axis S by means of the differential micrometer **47**, if small angular rotations are desired, or by means of a standard micrometer **49**, if larger angular rotations are desired, and recording the intensities reflected by said single test crystal as a function of specimen rotation by means of a film or preferably by means of a radiation detector **64**.

The variation of the adjoined method of X-ray micrography and diffraction analysis, based on the double crystal diffractometer principle, as applied to the crystallities of a polycrystalline material may equally be applied to single test crystals. Thus X-ray micrographs may be obtained by recording on a film close to the specimen surface the intensities reflected by the substructural entities of said single test crystal as a function of specimen rotation and to correlate said recorded X-ray micrograph to the corresponding intensities registered by radiation detector **64**. Said correlation is obtained by means of the photographic tracer technique utilizing for that purpose the film holder assembly **74**. Said method is very useful in disclosing the presence of various orders of magnitude of substructural entities since the X-ray micrographs can disclose the smallest entities, whereas the radiation detector integrates the intensity contributions of said smallest entities and may thus permit to obtain quantitative data of the largest order of magnitude of substructural entities comprising said smaller substructural entities. The recordings of reflected intensities by film combined with the radiation detector may, therefore, be mutually complementary.

It will be understood that the invention is susceptible to numerous variations, which will be readily apparent to those skilled in the art, without departing from the spirit and scope thereof.

The invention described herein may be manufactured and used by or for the Government of the United States of America for governmental purposes without the payment of any royalties thereon and therefor.

I claim:

1. The method for recording the topographical characteristics of a crystalline material which comprises impinging divergent X-radiation on a surface of a crystalline test specimen, successively impinging said specimen with radiation corresponding with various portions of the X-ray spectrum including crystal monochromatized radiation and continuous radiation while recording on X-ray sensitive film reflected images emanating from the irradiated area of said specimen, maintaining the identity of the reflecting portion of the irradiated surface by superposition of images recorded on X-ray sensitive film obtained by utilizing crystal monochromatized and continuous radiation.

2. The method for recording the topographical characteristics of a polycrystalline material which comprises impinging divergent heterogeneous X-radiation at a small grazing angle onto the surface of a polycrystalline test specimen utilizing an X-ray tube voltage substantially in excess of that of the excitation potential of the characteristic radiation, placing film sensitive to X-radiation close to the surface of said test specimen, recording on said film images of the crystallites of said specimen reflecting said X-radiation, lowering the tube voltage below the excitation potential of the characteristic radiation thereof, thereby to obtain continuous X-radiation, impinging continuous X-radiation at a small grazing angle onto said surface of said polycrystalline test specimen, placing a second film sensitive to X-radiation close to said surface of said specimen, recording on said second film images of crystallites of said specimen reflecting continuous radiation, interposing into the path of the primary X-radiation a monochromatizing crystal in reflecting position, impinging parallel crystal monochromatized radiation on said surface of said test specimen, superimposing the images obtained utilizing crystal monochromatized radiation on the images obtained utilizing said continuous radiation and recording on a separate film placed at the same position relative to said surface of said test specimen the images of the crystallites reflect-

ing parallel monochromatized X-radiation, whereby identification of the many crystallites reflecting divergent X-radiation with the few crystallites registered on the film utilizing crystal monochromatized radiation permits disclosure of fine-structural details of said polycrystalline test specimen.

3. The method of claim 2 wherein said divergent X-radiation is unfiltered.

4. The method of claim 2 involving the additional steps of positioning a cylindrical film to record spot reflections of crystallites on said specimen surface and, utilizing crystal monochromatized X-radiation, recording the reflection of crystallites of said surface of said specimen at successive distances from said surface of said specimen until reflections of said crystallites are recorded on said cylindrical film in the form of spot reflections on the Debye-Scherrer lines thereof, whereby the spot reflections on the Debye-Scherrer lines and the crystallites on the specimen surface giving rise to said spot reflections may be correlated.

5. The method of claim 4 involving the subsequent step of successively varying the angular position of said surface of said specimen relative to the incident X-radiation and, after each angular change, recording the intensity variation of the reflections of the individual crystallites as a function of specimen rotation, thereby to permit measurement of lattice misalignment of the individual crystallites.

6. The method of claim 4 involving the subsequent step of successively varying the angular position of said surface of said specimen relative to the incident X-radiation and, after each angular change recording by a radiation detector the intensity variation of the reflection of the individual crystallities as a function of specimen rotation, thereby to permit measurement of the lattice misalignment of individual crystallites.

7. An assembly wherein X-ray micrography is combined with X-ray diffraction to permit the qualitative and quantitative analysis of a polycrystalline test specimen for lattice inhomogeneities and substructural characteristics which comprises in combination with an X-ray tube to provide X-radiation and means for controlling the X-ray output thereof, means for mounting a polycrystalline test specimen for irradiation by said X-ray beam, means for successively impinging the same area of said polycrystalline test specimen with irradiation corresponding to various portions of the X-ray spectrum from said X-ray tube including crystal monochromatized radiation and heterogeneous radiation, X-ray sensitive film means proximate the irradiated surface of said test specimen in a manner such as to record at a constant distance from the irradiated surface the images of crystallites of said test specimen reflecting radiation corresponding to each of said portions of the spectrum from said X-ray tube, means for superimposing images obtained utilizing crystal monochromatized radiation and continuous radiation, whereby identification of the many crystallites reflecting heterogeneous radiation with the few crystallites registered on the film utilizing crystal monochromatized radiation permits disclosure of fine-structural details of said polycrystalline test solid, a cylindrical insert camera adapted to station cylindrical film mounted for recording in the form of spot reflections on Debye-Scherrer lines the images of crystallites of the same surface of said polycrystalline test solid, means for moving X-ray sensitive film progressively outward from said surface of said polycrystalline test solid in a manner such that images of crystallites of said surface of said polycrystalline test solid may be traced outward until they are recorded on the Debye-Scherrer lines of said cylindrical film, whereby spot reflections on the Debye-Scherrer lines and the crystallites on the specimen surface giving rise to said spot reflections may be correlated, means for angularly rotating said test specimen relative to incident radiation, and means for recording the intensities reflected by the individual crystallites as a function of specimen rotation, thereby permitting reflection curves to be obtained.

8. The assembly of claim 7 wherein said means for recording the intensity reflected by the individual crystallites as a function of specimen rotation comprises means for shifting said cylindrical film between each angular rotation of said test specimen whereby there is successively recorded on said cylindrical film the individual intensity contribution of each of said crystallites to the reflection curves.

9. The apparatus of claim 7 wherein said means for recording the intensity variation of the individual crystallites as a function of specimen rotation comprises a radiation detector and means for positioning said radiation detector at locations corresponding to the reflection images recorded on said cylindrical film when developed, whereby the reflected intensities of the individual crystallites of said specimen can be directly recorded as a function of specimen rotation by said radiation detector.

10. The apparatus of claim 7 wherein said cylindrical camera is optically transparent and wherein said means for recording the intensity variations of the individual crystallites as a function of specimen rotation comprises an electronic radiation detector mounted for movement relative to said camera in a manner such that said radiation detector may be positioned at locations corresponding to the reflection images recorded on said cylindrical film when developed, whereby the reflected intensities of the individual crystallites of said specimen can be directly recorded by said radiation detector after the removal of said cylindrical camera.

11. An assembly wherein X-ray micrography is combined with X-ray diffraction to permit the qualitative and quantitative analysis of a single crystal test specimen for lattice inhomogeneities and substructural characteristics which comprises in combination with an X-ray tube to provide X-radiation and means for controlling the X-ray output thereof, means for mounting said crystalline test specimen in position to reflect X-radiation from said tube, means for successively impinging the same area of said test specimen with radiation corresponding to the various portions of the X-ray spectrum including monochromatized radiation and heterogeneous radiation, X-ray sensitive film means proximate the irradiated surface of said test specimen in a manner such as to record at a constant distance from the irradiated surface the images of substructural entities and lattice inhomogeneities of said test specimen reflecting radiation corresponding to each of said portion of the spectrum from said X-ray tube, means of superimposing images obtained utilizing crystal monochromatized radiation and continuous radiation, thereby to reveal surface texture of said test specimen, means for controlled rotation of said test crystal relative to the incident beam whereby the misaligned lattice regions of said test specimen are successively brought into reflecting position, a radiation detector and means for positioning said detector relative to the beam reflected by said test specimen, whereby the intensities reflected by said test crystal can be recorded as a function of specimen rotation.

12. The assembly of claim 11 including means for moving said X-ray sensitive film progressively outward from the surface of said test specimen in a manner such that images of substructural entities of said test specimen may be traced outward until they may be recorded by said radiation detector whereby the intensity reflected by said substructural entities and lattice inhomogeneities can be directly recorded by said detector as a function of specimen rotation.

13. The assembly of claim 12 including means for placing X-ray sensitive film in front of the window of said radiation detector in a manner such that intensities reflected by substuctural entities and lattice inhomogeneities of said specimen surface can be registered as a function of specimen rotation by both said film and said radia-

tion detector thereby to permit disclosure of various orders of magnitude of substructural entities and lattice inhomogeneities.

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