Dec. 28, 1948. F. E. HAWORTH 2,457,555

APPARATUS FOR ORIENTING QUARTZ CRYSTALS BY X-RAYS

Filed July 9, 1945 3 Sheets-Sheet 1

FIG. 1

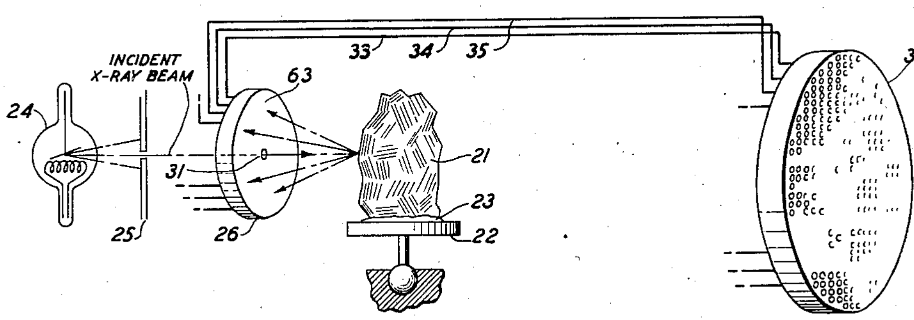


FIG. 2

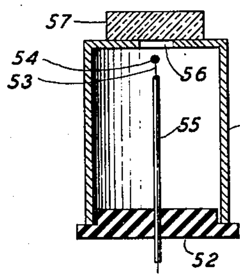


FIG. 3

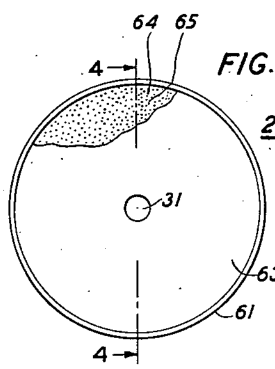
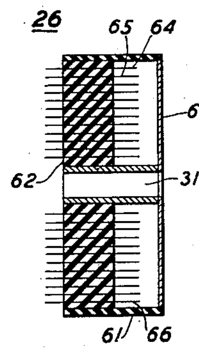


FIG. 4



INVENTOR
F. E. HAWORTH

BY E. V. Griggs
ATTORNEY

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FIG. 5

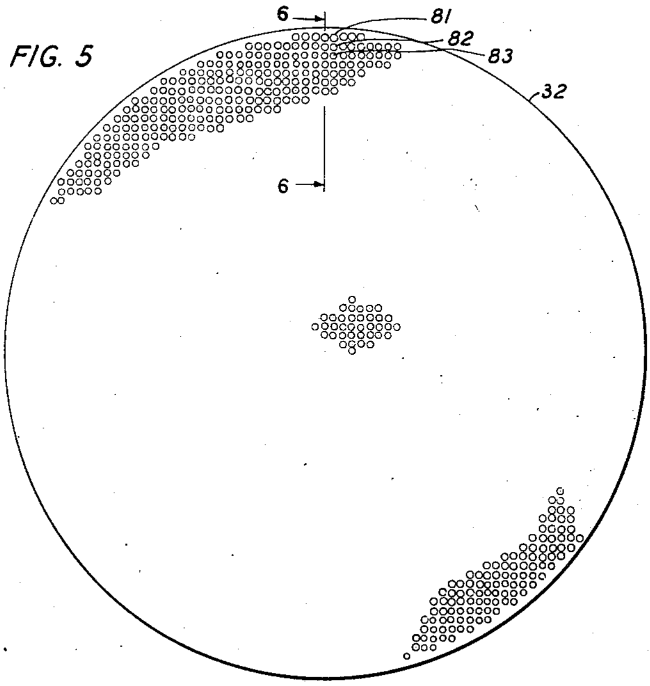


FIG. 6

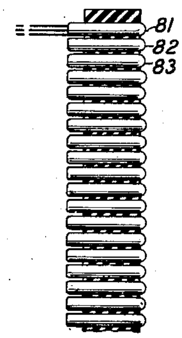
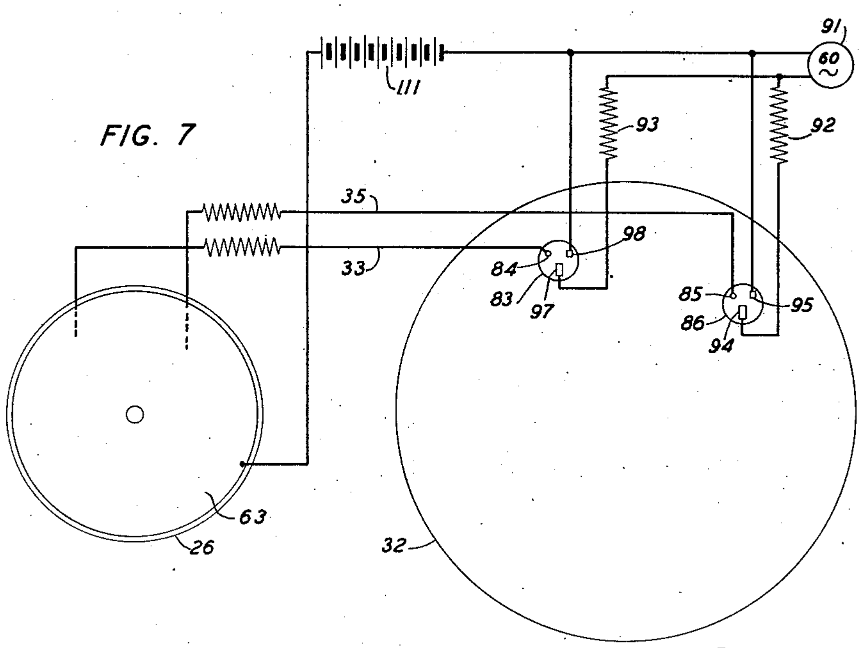


FIG. 7



INVENTOR
F. E. HAWORTH

BY W. Griggs
ATTORNEY

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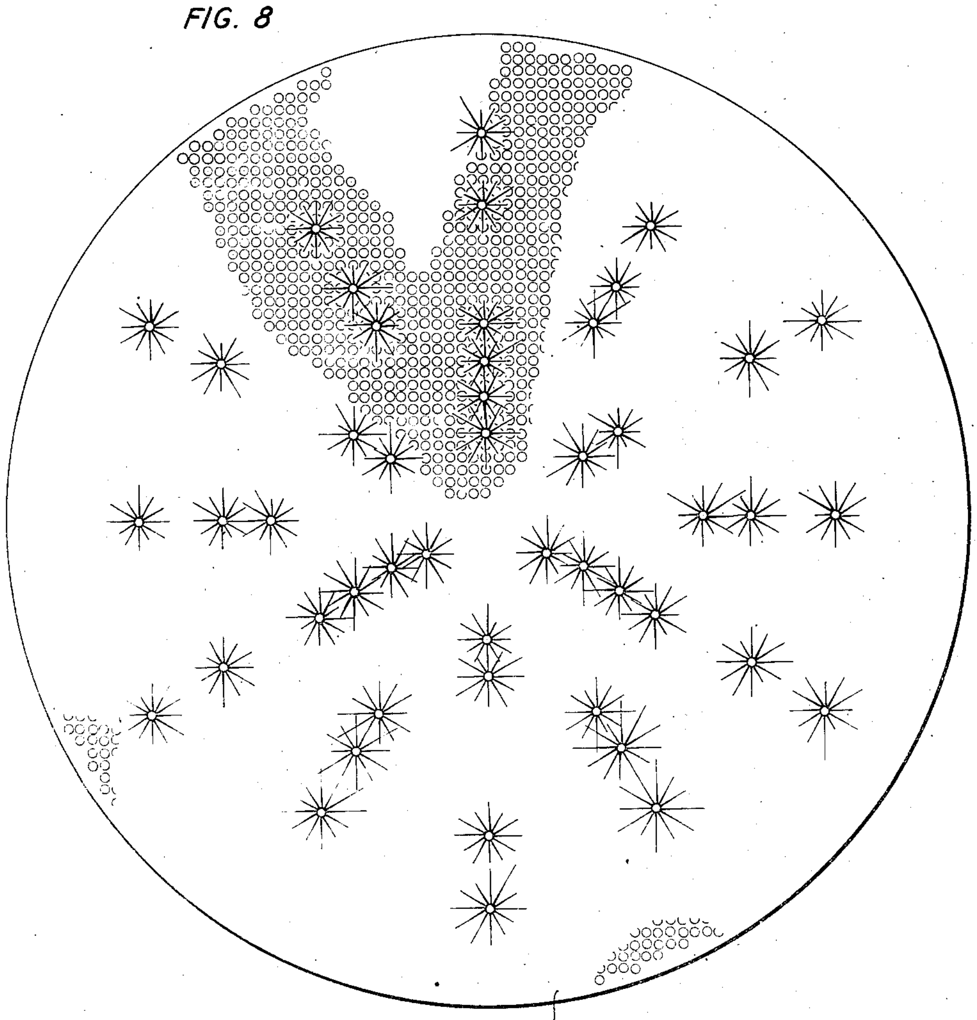
FIG. 8



INVENTOR
F. E. HAWORTH
E. V. Griggs
ATTORNEY

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APPARATUS FOR ORIENTING QUARTZ CRYSTALS BY X-RAYS

Francis E. Haworth, Summit, N. J., assignor to Bell Telephone Laboratories, Incorporated, New York, N. Y., a corporation of New York

Application July 9, 1945, Serial No. 603,938

8 Claims. (Cl. 250—53)

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This invention relates to a method of and apparatus for examining crystals and, more particularly, to a method of and apparatus for examining quartz piezoelectric crystals by X-rays.

An object of the invention is to facilitate the examination and analysis of crystals.

Another object of the invention is to facilitate the production of quartz piezoelectric crystal plates and to improve the quality thereof.

A still further object of the invention is to simplify and facilitate the process of orienting quartz crystals by X-rays.

Quartz elements, commonly in the form of wafers and generally referred to as "crystal plates" or "crystals," are used in large numbers at present in many important connections. For example, such elements are used in radio transmitters to maintain the output thereof at its exact assigned frequency value, in electrical wave filters, radio receivers, frequency standards and many other similar applications. These plates are usually cut from the "mother" crystal, i. e. the natural quartz, at various orientations with respect to one or more of the three sets of crystallographic axes thereof. For example, one such plate is produced, as described in detail in Patent 2,268,365, issued December 30, 1941, to G. W. Willard, by cutting the plate from the natural crystal so oriented that the major surfaces of the plate are inclined +38 degrees with respect to the Z, or optic, axis of the natural crystal and are parallel to an X, or electric, axis thereof. Other plates may be inclined various amounts in one or the other direction with respect to other axes of the mother crystal depending upon the particular characteristics desired in the finished plate.

It is obvious from the above that the production of crystal plates involves analysis of the natural crystal in order to determine various facts relative to the internal structure, particularly the direction of the crystallographic axes. If the quartz be well faced, i. e. if the natural faces have been retained, it is a fairly simple procedure to locate the several sets of axes. Crystals of this type are now quite rare however and most of the crystals encountered are of the so-called "river quartz" variety, i. e. crystals the natural faces of which have become invisible due to wear, handling, etc. In the case of such material the locating of the various axes is a somewhat involved procedure and may comprehend, for example, first the examination of the crystal in an immersion conoscope for location of the Z or optic axis and, secondly, additional examinations by X-rays and by optical instruments for location

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of the electrical (X-) and mechanical (Y) axes. While these procedures have been so perfected that highly accurate and otherwise satisfactory results are obtained, they are of course somewhat painstaking and time consuming.

It has been known for many years that a beam of X-rays passed through a quartz crystal will produce an orderly arrangement of spots, indicative of the X-ray diffraction pattern, on a photographic film, this type of photograph being known as a Laue photograph. Laue photographs provide a means of studying the internal structure of a crystal preparatory to orientation but the Laue method, involving, as it does, photography with the attendant development and printing processes, is generally considered too slow for commercial production.

It has also been known for some time (see for example page 642 of "Crystallography and Practical Crystal Measurement," volume 1, by A. E. H. Tutton) that the Laue pattern may be detected by a fluorescent screen and this has suggested a method of orientation wherein the crystal is moved about while observing the pattern on a fluorescent screen until the desired pattern has been obtained. While this method of instantaneous observation appears attractive in view of time saved, actual trials have proven somewhat unsatisfactory as it has been found that only a relatively few spots are sufficiently prominent for observation and analysis even by a trained observer and even when the process is carried out in a darkened room. The latter fact, i. e. the necessity for operating in a darkened room, is in itself a serious drawback.

The novel arrangement of the present invention provides the advantages resulting from instantaneous observation as previously sought after without any of the attendant disadvantages previously prevailing such as inadequate pattern, necessity of operating in a darkened room, etc.

A feature of the present invention is a multi-element pattern receiving device for reflected X-ray beams, each separate element of the device being individually responsive to the specific beam of X-rays reflected thereto entirely independent of X-ray beams reflected to any of the other elements, even the immediately adjacent elements.

A further feature of the invention is a multi-element indicator screen, each element of the screen being individually responsive to the particular energizing force transmitted thereto entirely independent of the energizing forces transmitted to any of the other elements, even the immediately adjacent elements.

In accordance with a specific embodiment of the invention an arrangement for orienting a quartz crystal is provided which may comprise a suitable source of X-rays and means for movably supporting the crystal in the path of the X-rays. The diffraction (Laue) pattern of the the crystal is displayed on an indicator screen which is in the form of a bank (large number, of order of 4,400) of cold cathode glow tubes which are selectively energized, in accordance with the diffraction of the X-rays from various points in the crystal, by pulses of current from a multi-element pattern receiver. The pattern receiving device contains a large number (corresponding to the number of glow tubes) of sensitive points, each of which may be thought of as a separate Geiger counter. As a diffracted X-ray beam strikes one of the points an impulse is transmitted to the corresponding glow tube in the display bank, which corresponding tube will be illuminated in substantial accordance with the intensity of the diffracted X-rays; i. e. for very low diffractions, the tube will merely flicker while for stronger diffractions the tube will glow more frequently with a miximum possible number of 60 illuminations a second, this being the frequency of the alternating current power supply utilized. The crystal may be manipulated, while the diffraction pattern displayed on the bank of tubes is observed, until the desired pattern for a typical orientation of the crystal is obtained. Use of this novel arrangement results in a pattern sufficiently extended to give a complete picture of the crystal structure and through use of the particular type of novel indicator screen, the pattern may be easily observed under normal conditions of illumination.

The term "reflection" is quite commonly used in the literature interchangeably with "diffraction" in referring to this general subject-matter since X-ray diffraction is like light reflection in that the entering and leaving beams make equal angles with the reflecting or diffracting atomic planes; it is contemplated in the present instance that the two terms may be used interchangeably.

A thorough understanding of the arrangement contemplated by the present invention as well as appreciation of the various valuable features thereof may be gained from consideration of the following detailed description together with the annexed drawings in which:

Fig. 1 is a schematic representation of an arrangement, which embodies features of the present invention, being used in examination of a crystalline specimen;

Fig. 2 is a sectional view of a Geiger-Muller counter of a type known in the art;

Fig. 3 is a plan view of a multi-element pattern receiving device, which embodies features of the present invention, a portion of the face being broken away to show a number of the individual, sensitive "points" of the device;

Fig. 4 is a sectional view, taken on line 4—4 of Fig. 3, showing the internal arrangement of the receiver;

Fig. 5 is a plan view of a multi-element indicator screen, which embodies features of the present invention, a sufficient number of the individual glow tubes being shown so that the arrangement of the entire bank of tubes will be readily appreciated;

Fig. 6 is a sectional view, taken on line 6—6 of Fig. 5, showing the arrangement of the individual glow tubes;

Fig. 7 is a schematic representation of a portion of the electrical circuit utilized in the arrangement contemplated by the present invention; and

Fig. 8 shows a diffraction pattern as displayed on the multi-element indicator screen contemplated by the invention.

Referring now to Fig. 1, a crystal 21, which, we will assume, is a specimen of river quartz with the natural faces missing and which, we will further assume, is to be analyzed preparatory to cutting definitely oriented plates therefrom, is shown mounted on platform 22 being temporarily attached thereto by a suitable agent 23 which may be, for example, sealing wax. By virtue of the ball and socket type of mounting illustrated, platform 22 is rotatable and tiltable in a plurality of directions whereby crystal 21 may be adjusted in various directions during analysis until the desired position is achieved.

A suitable source of X-rays, such as X-ray tube 24, is provided, together with the usual mounting and shielding appliances and power supply which it does not appear necessary to describe, for projecting an X-ray beam to crystal 21. Preferably all wavelengths of the "white" radiation are utilized. A suitable apertured shield 25 is provided for collimating the incident beam directed to the crystal and, as the "back-reflection" method (which will be described subsequently) is utilized, the beam is passed through pattern receiver 26 on its passage to the crystal, receiver 26 being provided with a centrally located aperture 31 to permit passage of the X-ray beam.

A plurality of diffracted, or reflected, X-ray beams results from the incident beam, the distribution and relative intensity of these beams being in accordance with the internal structure of crystal 21, that is these reflected beams, if projected onto a sensitive film, would indicate the diffraction pattern of the crystal in accordance with the Laue method.

As will be described in detail subsequently, receiving device 26, includes a plurality of distinctive sensitive points, each of which is connected by a suitable electrical transmission line to a corresponding tube in the bank of glow tubes provided on indicator screen 32. Three of these connecting lines 33, 34 and 35, are shown in Fig. 1. In a manner that will be described in detail subsequently, each sensitive point of receiver device 26 that is "struck" by a diffracted X-ray beam causes the transmission of an impulse over its respective line to the corresponding glow tube in screen 32 to cause energization of said corresponding glow tube in the manner previously referred to. A pattern caused by the illumination of a plurality of the tubes of screen 32 results therefore which is indicative of the diffraction pattern of crystal 21. The crystal may then be moved about in various directions on support 22, while the changing pattern on screen 32 is kept under observation, until the particular pattern is achieved which is indicative of a desired orientation of crystal 21.

Coming now to a more detailed description of certain of the devices contemplated by the invention, there is shown in Fig. 2, for purposes of comparison, a Geiger-Muller point counter of a type previously described in the literature (Zeitung für Phys. 71, pages 422–430, 1931, Max Steenbeck; and Journal of the Franklin Institute, 237, pages 371–384, 1944, Joseph Morgan, for example). Metal housing 51 is provided with a closure or base 52 of suitable insulating material. A suitable lead-in conductor 53, which is terminated by platinum point 54, is led into housing 51 through sleeve 55 which is sealed through base 52. Aperture 56, provided in the upper surface of housing 51 is closed by quartz plate 57, platinum point 54 being positioned opposite plate 57. The housing is filled with a suitable gas, use of air at a pressure of 63 millimeters being described in the article referred to above.

The counter may be operated under alternating current or direct current potential, the voltage being such that an extraneous source of ionization is required to initiate the discharge within the counter. The counter described in the article referred to operated to give a current of 1.6×10^{-7} coulomb per discharge.

The underlying theory of operation of the novel multi-element pick-up device or pattern receiver contemplated by the present invention is generally similar to that of the Geiger point counter of Fig. 2. The contemplated arrangement of the novel receiving device is shown in detail in Figs. 3 and 4. As illustrated, pattern receiver 26 comprises a suitable gas-tight chamber formed by cylindrical shell 61 of suitable insulating material, a closure block 62, also of suitable insulating material, and a face plate 63 which, in the present embodiment of the invention, is of beryllium. A plurality of separated wires which may be steel, platinum or other suitable material, as wires 64, 65 and 66, are sealed through block 62 and extend into the chamber to within a short distance from the inner surface of plate 63. As pointed out above, a centrally located aperture 31 extends through the chamber, providing a path therethrough for the incident X-ray beam.

The chamber is filled with air or another suitable gas at a predetermined suitable pressure. The wires are closely spaced, for example, one wire for each square millimeter of surface and the number of individual wires may be of the order of 4,400. Each of the individual wires acts as a separate Geiger point, corresponding to platinum point 54 of the device of Fig. 2, while beryllium sheet 63 acts as a common plate element for all of the "points."

Each of the individual wires of receiver 26 is connected by a separate conductor to a respectively associated glow tube of indicator screen 32. Three of these conductors, 33, 34 and 35. are shown in Fig. 1; actually, of course, if receiver 26 comprises 4,400 individual wires or points, there will likewise be 4,400 individual conductors connecting the points to the corresponding tubes of screen 32.

As stated above, the back-reflection method is preferably, although not necessarily, used, i. e. instead of placing the pattern receiver beyond the crystal and receiving the pattern after the X-ray beam passes through the crystal, receiving device 26 is placed on the same side of crystal 21 as the source of X-rays, tube 24, and the pattern is received as reflected back from the crystal. Use of the back-reflection method substantially reduces the time necessary for exposure and eliminates varying results due to varying crystal thicknesses.

Now as the Laue diffraction pattern of crystal 21 falls upon plate 63 of receiver 26 (Fig. 1) ions will be formed at those positions in the gas within the chamber which are irradiated by the reflected X-rays of the pattern. The potential across plate 63 and the "points" is so fixed that an instantaneous discharge or breakdown will occur between the plate and each point corresponding to the position at which an ion is formed in the gas. A series of impulses are therefore transmitted over the connecting lines to screen 32, an impulse being transmitted from each of the individual wires, or points, of device 26 which is opposite to an "ionized position" of the device, i. e., opposite to a point on plate 63 upon which a diffracted X-ray impinges. The series of impulses transmitted to device 32 at any moment therefore, is in accordance with the X-ray pattern reflected by crystal 21 and picked up by device 26 at the same moment.

Indicator device, or screen 32, in one form contemplated by the present invention, is illustrated in detail in Figs. 5 and 6. A plurality of lamps as 81, 82 and 83, which may be in the form of cold cathode gas-filled tubes of the general type described in an article, "Cold-Cathode Gas-Filled Tubes as Circuit Elements," S. B. Ingram, Electrical Engineering (Transactions), July 1939, vol. 58, page 342, are mounted in a suitable circular block of insulating material. The number and position of the tubes in the bank of screen 32 corresponds to the number and position of the individual wires, or sensitive points, of receiver 26; that is, there is a tube of screen 32 corresponding to, and electrically connected to, each individual wire of receiver 26.

The tubes, as pointed out, above may take the form of cold cathode vacuum tubes of the three element type, i. e., provided with a cathode, a control anode and a main anode. The conduction path between the cathode and the control anode is known as the control gap and the conduction path between the cathode and the main anode is known as the main gap.

For the present purpose, the tubes are preferably somewhat smaller than the ordinary commercial vacuum tube. The exact size of the tubes is not controlling but it is contemplated, by way of example, that tubes approximately one-half inch in diameter might be used in which the over-all diameter of the screen, with a total of 4,400 tubes would be of the order of three feet. It is apparent, of course, that the pattern displayed on screen 32, while an exact reproduction of that projected onto receiver 26, will be a considerably enlarged version thereof. This is a highly desirable feature.

The operation of the glow tubes of indicator screen 32 under control of the pulses set up in pattern receiving device 26 may best be understood by reference to the schematic circuit of Fig. 7. (For purposes of simplicity of disclosure, only two of the glow tubes of indicator screen 32 and two of the sensitive points, or wires, of pattern receiver 26 have been illustrated.) The control anode of each tube is connected over the individual line provided to the respectively associated sensitive point of receiver 26, i. e., as illustrated, control anode 84 of tube 83 is connected by line 33 to its associated point in receiver 26 (for example, this may be point 64 [Fig. 4]) and control anode 85 of tube 86 is connected over line 35 to its associated point of the pattern receiver.

A voltage from alternating current source 91, the frequency of which will be assumed to be 60 cycles per second, is applied, through respective resistances 92 and 93, across the main gap of each tube, for example across main anode 94 and cathode 95 of glow tube 86 and across main anode 97 and cathode 98 of glow tube 83.

A direct current voltage from battery 111 is applied across the control gap of each glow tube and the respective gap of receiver 26 in series. For example, the negative terminal of battery 111 is connected to respective cathodes 98 and 95 of tubes 83 and 86 while the positive terminal of the battery is connected to plate 63 of pattern receiver 26; the respective sensitive points of the receiver are connected to control anodes 84 and 85 of tubes 83 and 86 respectively. This voltage is not, in itself, quite sufficient to cause the glow tubes to trip, or break down, but, when augmented by the additional energy transmitted from pattern receiver 26 when a point is ionized, the voltage applied to the control gap of the respective glow tube will then be sufficient to cause ionization and subsequent breakdown of the main gap of the tube which will then light up or glow. Thus, those sensitive points of pattern receiver 26 which are receiving X-rays will trip-off the corresponding glow tubes of screen 32 and cause a display thereon corresponding to the diffraction pattern of crystal 21 (Fig. 1). The frequency with which a respective tube is tripped-off will depend upon the intensity of the X-ray diffraction from the corresponding portion of the specimen being analyzed; for very weak diffractions a tube may merely flicker while, as the intensity of the diffracted X-rays increases, the frequency of the tube breakdowns will increase with a possible maximum of 60 per second, the frequency of power supply 91, for the stronger diffractions.

There is illustrated in Fig. 8 the pattern displayed on screen 32 for a typical orientation of a crystal. That is the observer starts the test with the knowledge that the particular orientation of the crystal that he is seeking will result in the symmerical pattern of Fig. 8. He therefore manipulates the crystal on its mounting (Fig. 1) while observing the pattern on screen 32 until the desired pattern of Fig. 8 is attained.

From the "pattern" of a crystal displayed on screen 32 there may readily be determined various pertinent facts concerning the crystal, such as the location and sense of the various crystallographic axes and the handedness of the crystal. This is true, not only in the case of the mother crystal, but in the cases of finished and partially completed "cuts" or plates as well.

The novel arrangement contemplated by the present invention, one specific embodiment of which has just been described in detail above, makes possible the accomplishment in the matter of a few seconds of that which by previous methods takes considerably longer. The orientation of a quartz "stone" can be determined immediately with respect to all of the natural crystallographic axes without the necessity of making any cuts in the surface or immersion in a liquid.

While a specific embodiment of the invention has been selected for illustration and detailed description, the invention is not, of course, limited in its application to such embodiment. For example, the arrangement is applicable to analysis of crystalline materials other than quartz and types of pattern receivers and display screens other than those described may be utilized. While certain advantages appear to reside in use of the back-reflection method, this method need not necessarily be used. Alternating current power supplies of frequency other than 60 cycles per second may be utilized with corresponding change in the maximum possible number of times per second that the respective glow tubes may be tripped-off. In short the embodiment described should be taken as illustrative of the invention and not as restrictive thereof.

What is claimed is:

1. In the analyzation of a quartz crystal, apparatus for displaying the diffraction pattern of the crystal which comprises means for movably supporting said crystal, means for projecting an X-ray beam to said crystal, means for receiving the diffracted X-rays on a sensitive receiver, and means operating substantially in step with said receiving means for selectively energizing glow tubes under control of said sensitive receiver whereby the visual pattern produced by the energized glow tubes at any instant corresponds to the Laue pattern of X-rays diffracted by said crystal and received by the sensitive receiver at the same instant.

2. Apparatus for orienting a specimen of crystalline material which comprises means for projecting a beam of X-rays onto said specimen, means for receiving the Laue pattern of X-rays reflected by said specimen on a selective pattern receiver, a group of individual light sources, and means for selectively energizing the light sources of said group in accordance with the pattern received by said pattern receiver, said receiving means and said energizing means operating substantially simultaneously and in step.

3. Apparatus for analyzing a crystal of the type capable of emitting a plurality of distinct diffracted X-rays from a single incident X-ray beam, comprising means for adjustably supporting a crystal being analyzed, means for projecting an X-ray beam to said crystal, a pattern receiver for receiving the distinct X-rays diffracted by said crystal, said pattern receiver including a relatively large number of separate sensitive areas each of which is activated by contact of diffracted X-rays whereby said sensitive areas of said receiver are activated in accordance with the Laue pattern of said crystal, a viewing screen comprising a plurality of separate visual indicators, the number of visual indicators corresponding to the number of the sensitive areas of said pattern receiver, and means for energizing selected ones of said visual indicators in accordance with the diffraction pattern of said crystal received by said pattern receiver, said energizing means operating substantially simultaneously with respect to activation of respective areas of said receiver.

4. Apparatus for analyzing a quartz crystal comprising a source of X-rays, means for adjustably supporting a crystal being analyzed, means for projecting an X-ray beam from said source to said crystal, a pattern receiver for receiving the X-rays diffracted by said crystal, said receiver comprising a number of the order of 4,400 of individual sensitive areas each of which is capable of being independently activated by contact of diffracted X-rays whereby said sensitive areas are activated at any one instant in accordance with the Laue pattern of X-rays diffracted at that same instant by said crystal being analyzed, a viewing screen comprising a plurality of glow tubes, the arrangement of said glow tubes comprising said screen corresponding with respect to number and relative positioning to the arrangement of said sensitive areas of said pattern receiver, and means for activating said glow tubes under instantaneous control of said pattern receiver whereby the visual pattern resulting at any one instant from activation of glow tubes of said screen corresponds to the pattern of X-rays diffracted at the same instant by said crystal and received by said pattern receiver.

5. Apparatus for analysis of a crystalline specimen capable of emitting a plurality of distinct

diffracted X-rays from a single incident X-ray beam, comprising means for projecting an X-ray beam, means for adjustably supporting a crystal to be analyzed in the path of the projected beam, a diffraction pattern receiver comprising a plurality of separate Geiger points, means for supporting said pattern receiver in position to receive the X-ray pattern diffracted by said crystal, a screen of cold cathode glow tubes, means for selectively activating said Geiger points in accordance with the diffraction pattern received and means for individually connecting each of said Geiger points to a respective one of said glow tubes whereby to selectively control the activation of said glow tubes in accordance with the activation of said Geiger points.

6. In apparatus for analysis of a crystalline substance capable of emitting a plurality of distinct diffracted X-rays from a single incident X-ray beam, a Laue pattern receiver for receiving the diffracted X-rays and for translating them into a series of corresponding electrical pulses, said pattern receiver comprising a hermetically sealed chamber, a conductive sheet forming one closure member for said chamber, a plurality of distinct conductive wire-like members within said chamber, all of said wire-like members being spaced a like distance from said conductive sheet and also being spaced a like distance from each other, a gas within said chamber filling the space between said wire-like members and said conductive sheet, and means for applying a potential across said conductive sheet and the individual wire-like members, the magnitude of said applied potential being insufficient in itself to cause a discharge between respective ones of said members and said sheet but being sufficient to cause such a discharge in response to movement of an ionizing particle between said sheet and a respective one of said members.

7. In apparatus for analysis of a crystalline specimen capable of emitting a plurality of distinct diffracted X-rays from a single incident X-ray beam, a receiver for reception of the reflected X-rays and for translation thereof into a corresponding series of electrical pulses, said receiver comprising a gas-tight chamber, a plurality of equally spaced wires within said chamber, a beryllium plate forming one closure member of said chamber and exposed on one surface to reflected X-rays, the projecting ends of each of said wires being spaced an equal distance from said beryllium plate, a gas within said chamber filling the space between said beryllium plate and the projecting ends of said wires, and means for applying a potential across said beryllium plate and said wires, the magnitude of said applied voltage being insufficient in itself to cause breakdown between said plate and respective ones of said wires but being sufficient to cause such breakdown in response to movement of an ionizing particle between said plate and a respective one of said wires.

8. Apparatus for orienting a quartz crystal comprising means for projecting an X-ray beam to said crystal, means for intercepting the Laue pattern of X-rays reflected by said crystal, a bank of glow tubes, means controlled by said intercepting means for activating selected ones of the glow tubes to reproduce an enlarged version of the Laue pattern intercepted by said intercepting means, and means for adjusting said crystal in the path of the projected X-ray beam in order to bring the Laue pattern reproduced on said bank of glow tubes into substantial accord with a predetermined pattern.

FRANCIS E. HAWORTH.

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