A Contour Generator for X-ray Crystal Structure Analysis

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An electronic device has been developed for the presentation of a two-dimensional function as a contour map on the screen of a cathode ray oscillograph, to be used in conjunction with an analogue computer for X-ray crystal structure analysis. The function to be plotted is fed as an electrical voltage to one pair of plates of a cathode ray tube in front of which a system of slits and a photomultiplier are placed. The pulse output from the photomultiplier intensifies the spot on a second oscillograph whose *x* and *y* time bases are properly synchronized with the scanning of the function. The image photographed from the second cathode ray tube is the contour map.

THE contour generator described in this paper is part of an analogue computer designed by Suryan[1]. The aim of the computer is to sum a double Fourier series of the form

$$\rho_{x,y} = \sum_{h=0}^{\pm 12} \sum_{k=0}^{+12} F_{h,k} \cos 2\pi \frac{hx}{a} \cdot \cos 2\pi \frac{ky}{b}, \text{ etc.}$$

where x, y are the co-ordinates in a unit cell of dimension a, b, and $F_{(h,k)}$ are the observed structure factors got from X-ray data. The computer, in operation, simplifies tedious calculations of the above type involved in X-ray crystallography. The function $\rho_{(x,y)}$ represents the electron density at any point (x, y) within the crystal, and it is of interest to find the points of maximum electron density in order to identify them with the positions of the atoms in the crystal. The computer plots the above function on the screen of a cathode ray oscillograph, as a contour map. If a method of identifying the levels represented by the various contour lines is available, it is easy to locate the points of maxima.

For purposes of using electrical analogies, the variables x, y are transformed into the single time variable t according to the relations

$$x = pt \text{ and } y = p't \, (p \gg p') \quad \ldots\ldots (1)$$

which correspond to fast and slow scan in the x and y directions respectively. The values of p and p' used in the computer are $p = 50$ sec.$^{-1}$ and $p' = 0{\cdot}05$ sec.$^{-1}$, i.e. in the x-direction, the fundamental frequency is 50 c/s. and in the y-direction, 0·05 c/s. The ratio p/p' is, therefore, 1000, i.e. there are 1000 scanning lines almost parallel to the x-axis. This number of scanning lines is enough to give continuous contour lines because of the finite size of the spot (about 0·1 mm.) on a cathode ray tube.

The output of the amplifiers, a voltage V_t, at any instant t (i.e. t sec. after starting) represents, with the proper scale factor, the function $\rho_{(x,y)}$ at the point x, y, t being connected to x and y by the relation (1) and in time $t = 1/p'$ a complete x, y scan is made. In other words, the output voltage of the amplifier represents $\rho_{(x,y)}$ practically over the entire x, y domain.

The basic requirements of an electronic contour generator are met with if a c.r.t. spot at any point x, y on the screen is intensified whenever the corresponding $\rho_{(x,y)}$ is equal to any one of a set of suitably spaced values and the spot scans all the points x, y in the domain $x = 0$ to a, and $y = 0$ to b. If the scan is close enough these spots merge to form continuous contour lines. These requirements can be realized in many ways. Pepinsky[2] used a number of trigger circuits actuated by the voltage representing $\rho_{(x,y)}$ such that they give a pulse whenever $\rho_{(x,y)}$ crosses the pre-set values; these pulses intensify the spot on a cathode ray tube. He obtained very good contour maps in this manner. The circuit, however, requires the use of a large number of tubes and associated components. The contour generator described in this paper (Fig. 1) uses an auxiliary cathode ray tube in place of trigger circuits, to produce the intensifying pulses.

Corresponding to the value of V_t, the spot will assume a definite position on a vertical line. In front of the screen is a system of horizontal slits cut on an opaque material and behind it is placed a photomultiplier assembly (Fig. 1). Thus, whenever the spot is against any one of the slits light passes through to the photomultiplier and a voltage pulse is obtained. This is amplified and fed to the grid of the recording cathode ray tube. Thus we get the intensifying pulse.

The principle of working of the instrument is indicated in Fig. 2. A waveform *a b c d e f g* ... is applied to the contour generating cathode ray tube (C.G.C.R.T.). At *a*, *b* ... *h*, the spot crosses any one of the three slits denoted by lines 1, 2, 3 and corresponding pulses *A*, *B* ... *H* are got from the photomultiplier, which intensify the spot on the recording oscilloscope at points *A*, *B* ... *H* in Fig. 2(b). As the waveform changes even slightly, the relative positions of these spots also change and join to form the apparently continuous contour lines.

Description of the instrument

One of the primary requirements of the C.G.C.R.T. is that it should have a short persistence. With the fundamental frequency 50 c/s., harmonics up to the twelfth are of importance (i.e. 600 c/s.). When the spot is traversing the screen at this rate, the time taken by the spot to cross from one slit to an adjacent one can be as low as a fraction of a millisecond and within this time the screen illumination due to the first spot should have reduced to a negligible value, if there is not to be merging of the various pulses from the photomultiplier. A P-5 or a P-15 screen would be suitable; but in the present experiments, a P-11 screen was used with somewhat longer (5 millisec.) persistence (the time for the illumination to fall to 1 per cent of peak) which is too large for the higher frequency components contained. Another important consideration is the size of the spot and of the slit. To get narrow contour lines it is essential that the duration of the pulse from the photomultiplier be short. It is also necessary that the shape of the individual pulses be symmetrical about their centre.

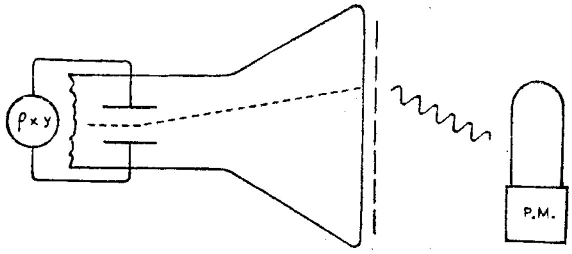

FIG. 1 — ARRANGEMENT OF CATHODE RAY TUBE, SLITS AND PHOTOMULTIPLIER

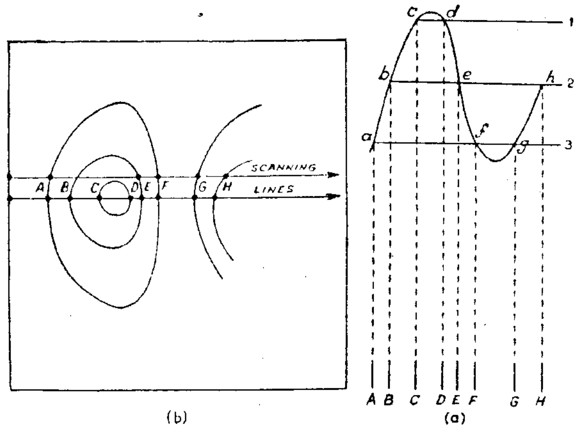

FIG. 2 — FORMATION OF CONTOUR LINE PULSES FROM WAVE *a*, *b* .... *h* [Pulses *A*, *B*, *C* .... *H* in (a) intensify CRT beam to produce spots at *A*, *B*, *C* .... *H* in (b)]

The width and height of pulses due to the various slits are, in general, unequal because of the variable velocity of the trace on the screen and the finite rise-time of the screen. If a sine-wave of frequency $\omega/2\pi$ and of amplitude A (as a length on the cathode ray tube) is applied to the tube, and the slit width is a_1 and spot diameter is a_2, it is seen that the time duration of the pulses varies between $p_{min.}$ and $p_{max.}$ as given by

$$p_{min.} \approx \frac{a_1 + a_2}{A\omega} \quad \ldots\ldots\ldots \text{(2a)}$$

$$p_{max.} \approx \frac{2 \cdot 8}{\omega} \sqrt{\frac{a_1 + a_2}{A}} \quad \ldots\ldots \text{(2b)}$$

In deriving equations (2a) and (2b) it has been assumed that the screen response is instantaneous and the persistence zero. It is not profitable to reduce the slit width indefinitely without reducing the spot size. The effect of variation of slit width with constant spot size is shown qualitatively in Fig. 3.

In the experiments conducted the pulses obtained with 400 c/s. wave were found to be very much smaller in amplitude than those

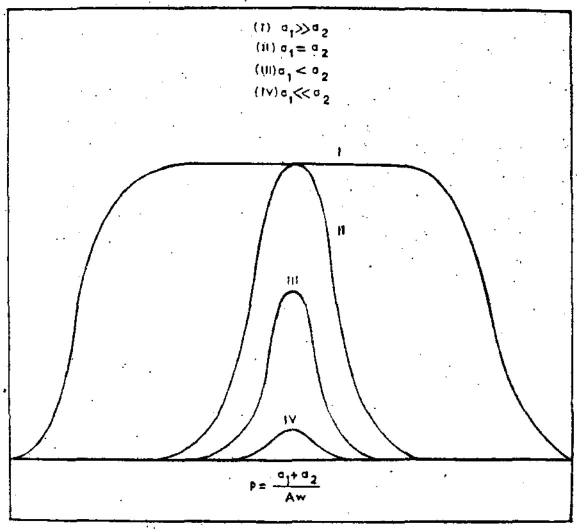

FIG. 3 — DEPENDENCE OF PULSE WIDTH ON SLIT SIZE [The spot size is assumed to be the same in all cases]

with 50 c/s. This was possibly due to the rather large rise-time of the P-11 screen used. Further, when the trace on the C.G.C.R.T. was expanded considerably over the screen diameter, adjacent pulses tended to merge, due obviously to the persistence of the screen being large. A P-5 screen is likely to improve the performance considerably.

The requirements for realization of sharp contour lines are a fine spot on the cathode ray tube which can be secured by using fairly large accelerating voltage and extremely short persistence of the screen of the contour generating oscilloscope. It is also desirable to have for the final cathode ray tube a screen having very short rise-time (time required to excite screen to a reasonably constant light output), since otherwise, the narrow pulses may fail to illuminate the trace, or produce only a faint trace.

Equalization of the heights of pulses — If the photomultiplier receives the same amount of light from each slit and if each slit is larger than the spot on the C.G.C.R.T., it would appear that the pulses will all be of equal height. This, however, is not true when it is considered that the instantaneous velocity of the spot does also contribute to lessening the illumination from the screen by not energizing it for a sufficiently long time. Hence it becomes necessary to equalize the pulses before they are applied to the final or recording cathode ray tube. This is done by an ordinary clipper circuit.

If the pulses are directly amplified and clipped, the higher ones and the corresponding portions of the contours would be disproportionately broad. One method of overcoming this difficulty is to use the photomultiplier pulses to trigger a standard pulse. The method is not suitable, however, because the triggered pulse does not coincide with the centre of the triggering pulse, especially when this is broad, thereby producing broken contour lines. However, a simple way of approximately equalizing the pulses before clipping is to apply the amplified pulse to X deflection plates of the C.G.C.R.T. whereby the spot, whenever it is before one of the slits, is driven to, say, the extreme right edge of the slit. If the slit edges are in a line parallel to the trace on the cathode ray tube, the output voltage remains constant. The principle is analogous to the photoformer. This method was also found to increase the resolution of pulses.

Pulse amplifier — The requirements on the pulse amplifier are not exacting since the pulse duration is at least a few hundredths of a millisecond. A feedback amplifier employing three stages of amplification and having a rise-time of 1 μsec. was constructed. The gain was made adjustable to a maximum of 100 and the output voltage could be clipped at any level from 15 to 80 V., the amplifier having good fidelity for output voltages of this magnitude. The output stage was a 6AG7 tube working into a 3 K ohm load. The clipping circuit is shown in Fig. 4. In this arrangement a cathode follower is used as a low impedance variable voltage d.c. source for clipping. The performance of this clipper was very satisfactory. Diode D_2 was used for d.c. restoration. Diode connected 6AC7 was used as

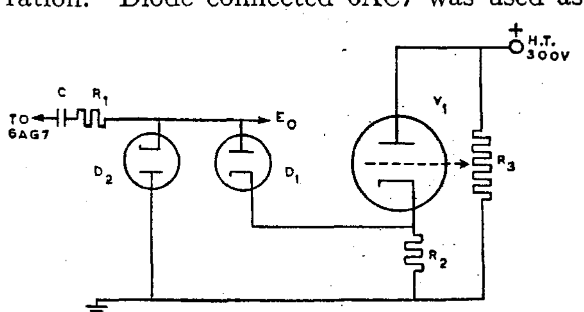

FIG. 4 — CLIPPER CIRCUIT SHOWING USE OF CATHODE FOLLOWER AS A LOW IMPEDANCE SOURCE [V_1, 6AC7 (triode-connected); D_1, D_2, 6AC7 (diode-connected); R_1, 100K Ω ; R_2, 10K Ω ; R_3, 82K Ω + 47K Ω pot; C, 0·05 μF]

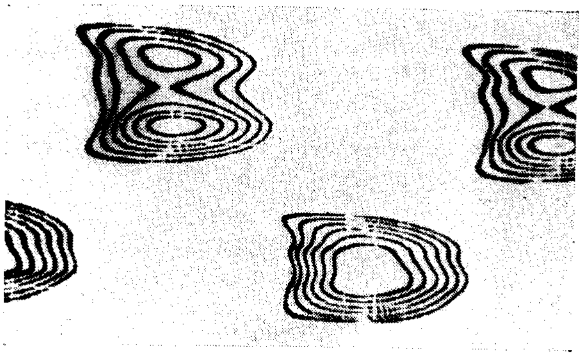

FIG. 5 — CONTOUR MAP FOR AN ARBITRARILY CONTROLLED WAVEFORM REPRESENTING $\rho_{x,y}$

its characteristics are superior to those of 6H6.

Six kV. accelerating potential was applied to the C.G.C.R.T. and the spot size was definitely less than 0·2 mm. (diam.). A general purpose commercial oscilloscope of good quality with a P_1 screen was used for photography.

Distinguishing of contour lines — In X-ray crystallography, especially in difference Fourier synthesis, it is necessary to distinguish between positive and negative contour levels. Also in the case of overcrowding of lines it is useful to omit the alternate levels. This can be easily accomplished in the present contour generator by having different sets of slits with the appropriate slits closed or open as the case may be, and shifting the trace to the proper set of slits. By placing a vibrating reed in front of the proper slits it is possible to get dotted contour lines. A photograph taken with a contour generator constructed on the above principles is shown in Fig. 5.

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