DOUBLE FOURIER-SYNTHESIS. PUNCHED CARD METHOD

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The use of "Punched Card" methods in crystal structure analysis has the advantage of speeding up the enormous amount of computational work involved in these investigations. In this paper, two methods are suggested for the evaluation of electron density distributions using double Fourier-Synthesis. Of these two methods, complete details have been worked out for the latter one using the Tabulator for summing up the series.

1. Introduction

Punched Card methods have been used both in U.S.A. and in Great Britain for crystal structure calculations like the evaluation of electron density by Fourier Synthesis (Shaffer *et al.*, 1946; Hodgson *et al.*, 1949), structure factor calculations (Donohue and Schomaker, 1949; Grems and Kasper, 1949), refinement of co-ordinates by differential Fourier Synthesis (Cox *et al.*, 1949), least square and steepest descent methods, etc., during the past few years. Although, it might be possible to design *ad hoc* machines which are more efficient than the Hollerith for a particular type of calculation, the Hollerith system has the advantage that it is well established, reliable and widely distributed throughout the world. Since the use of interchangeable control panels makes it possible to divert the machine from one type of calculation to another without delay or inconvenience to the main work for which the system is installed, it is possible to use any available spare time of the machine for scientific computations.

2. Two-Dimensional Fourier Synthesis

Descriptions of the use of punched cards and their advantages in crystal structure calculations have already been given by different workers (Shaffer *et al.*, 1946; Hodgson *et al.*, 1949), so that only such details as are relevant to the present method will be given here. The previous punched card methods of evaluating the Fourier series are, in principle, mechanization of Beevers-Lipson strip method. Here a multi-dimensional series

$$\rho\,(x\,y\,z_1) = \sum_h \sum_k A_{hk} \cos 2\pi hx \cos 2\pi ky + \textit{similar terms} \tag{1}$$

for electron density $\rho(x\,y\,z_1)$ over the plane $z = z_1$, is split into one dimensional series and the summation done in steps using card files. These card files contain information about the functions $A \cos 2\pi hx$ and $A \sin 2\pi hx$ punched in them for a series of values for the amplitude A and frequency h, each set of cards giving the values $A \cos 2\pi hx$ and $A \sin 2\pi hx$ in steps corresponding to 1/500 or 1/120 of the unit cell dimensions.

The splitting of a multi-dimensional series into steps, however, increases the amount of manual labour needed in picking the correct cards for each step with a corresponding increase in the chance of errors creeping in. What is more important, one has to wait for the results of the first one-dimensional synthesis before one could pick up the cards for the second step and this causes delay, especially if the Hollerith equipment is not always available at hand for the investigator. Because of these difficulties, it was considered desirable to evolve a punched card method where the two-dimensional Fourier synthesis could be performed in one step.

3. The Multiplier Method

One very simple method that suggests itself to evaluate (1) is in principle, analogous to the method of using the 'Two-dimensional phase factor tables' of Beauclair and Sinogowitz (1949). Here, card files of $\cos 2\pi hx \cos 2\pi ky$ and similar functions are prepared corresponding to every point (x, y) over a suitable net and for integral values of frequency h and k (say 0–20). Actually, using standard eighty column cards it would be possible to cover a net in steps of 1/60 the unit cell with about 5000 cards for the function $\cos 2\pi hx \cos 2\pi ky$, since for each point (x, y) about 20 cards would be sufficient to give two figure accuracy. A similar number would be necessary for the functions cos sin, sin cos and sin sin. The number of cards needed would, of course, increase rapidly if a finer net, larger ranges for h and k or a higher degree of accuracy are needed. These cards form the master cards which have to be used in all subsequent computations. The actual procedure for evaluating a particular series would be somewhat as follows:—

Firstly, we have to punch a few, say 20 or 40, amplitude cards corresponding to the coefficients of the series for the frequency (h, k). Now, these A_{hk} cards are interspersed with the master cards for a particular point —say, (x_1, y_1) and the multiplication done between the corresponding entries in adjacent cards and the sums $\sum_h \sum_k A_{hk} \cos 2\pi hx_1 \cos 2\pi k y_1$, etc., obtained. All these steps could be done on the machine. After the evaluation is done for the point (x_1, y_1), the A_{hk} cards are extracted from the set using the sorter and the procedure repeated with the master cards corresponding to the point (x_2, y_2) and so on till the whole quarter of the unit cell is covered.

This method was first tried out, but it had the disadvantage that since the multiplier which is the slowest of Hollerith machines has to be used, the machine time needed for one calculation is comparatively large. Further, everyone of the master cards has to be used in each calculation and in addition there is the trouble of having to punch a few amplitude cards for every calculation. Because of these difficulties, the recurring cost for each calculation would be rather large.

4. THE TABULATOR METHOD

Because of the difficulties of the multiplier method, the method was modified into a more suitable form, avoiding the operations of multiplication and using only addition, so that the tabulator which is much faster than the multiplier could be used for the purpose. This method is an extension of the Beevers-Lipson strip method (1936) such that two-dimensional synthesis could be made directly.

The punched card files contain the functions of the form $A \cos(hM6°) \times \cos(kN6°)$ for

$$A = \pm 1, \pm 2, \pm 5, \pm 10, \pm 20, \pm 50, \pm 100, \pm 200 \text{ and } \pm 500$$

$$h, k = 0, 1 \quad \ldots\ldots, 20$$

$$M, N = 0, 1 \quad \ldots\ldots, 15.$$

Hence, the total number of cards needed to make up the complete file (about 1,00,000) is much more than in the previous case. A specimen of the card design is shown in Fig. 1. In the standard 80 column cards, the columns 1–64 are divided into 16 fields of 4 columns each. These 16 fields on a single card contain the values of the function $A_{hk} \cos(hM6°) \cos(kN6°)$, for a given A_{hk} and M, for the 16 values of N ranging from zero to 15. The columns 65–68 give the totals of the numbers in the 16 previous fields and this is used to give a check on the summation. The columns 69–80 are descriptive or indicative fields used to give data for identification of cards or for giving instructions for control operations, sorting, etc. The columns 69–70 give the values of M for a given A_{hk}, and the column 71 indicates to which group (odd-odd, odd-even, etc.) the frequency belongs. Columns 72–75 give the amplitude A, the two fields 76–77 and 78–79 give the values of the frequencies h, k and finally the column 80 gives the nature of the trigonometrical function, *i.e.*, whether cos cos cos. sin, etc. The data in the cards necessary for help in filing or in picking out by visual inspection like

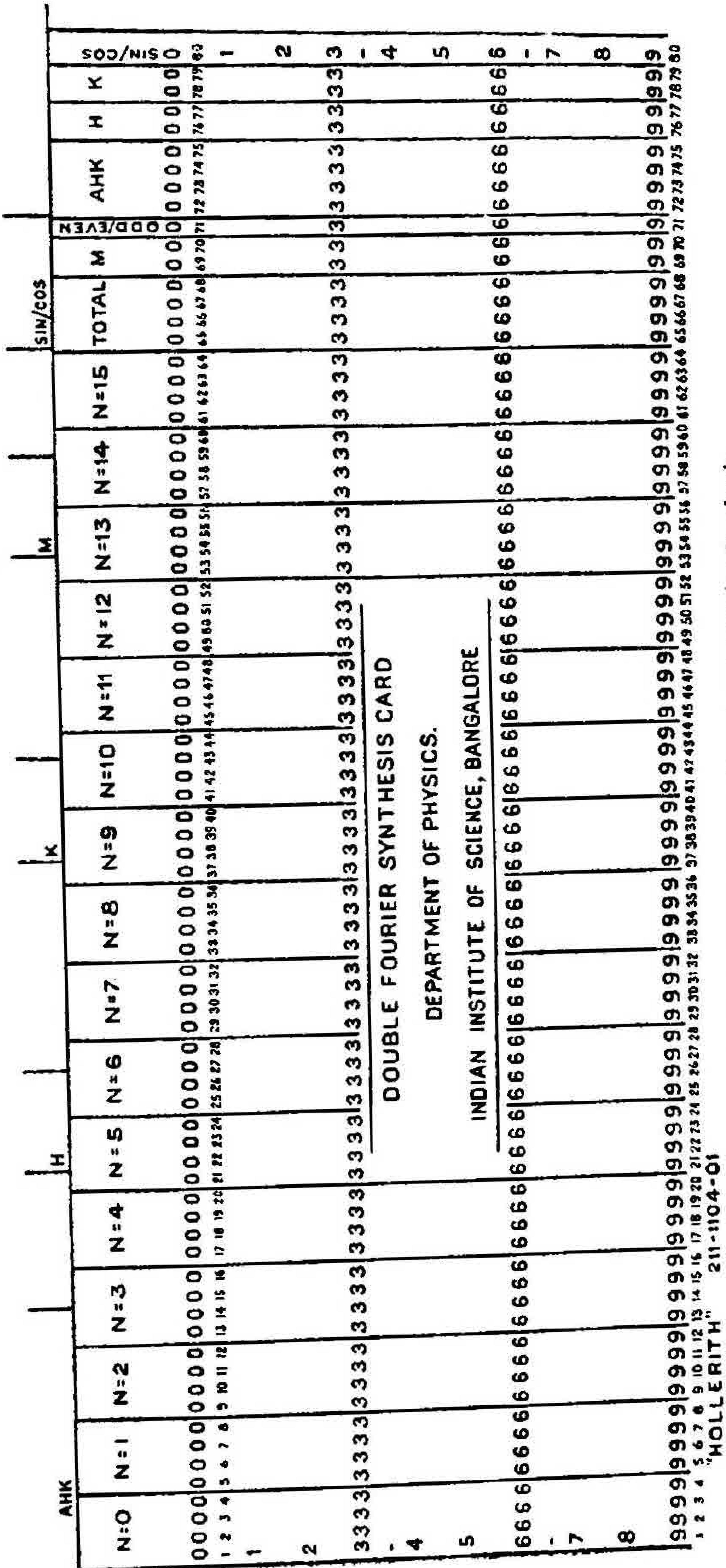

FIG. 1. Card design for two-dimensional Fourier Synthesis.

amplitude, frequency, etc., could be listed at the top of the cards. Different coloured streaks drawn along the edges of the cards help one to distinguish the different classes of cards and to identify cards which are misplaced.

The cards for the value $A = + 100$ could be punched manually using the key punch from standard trigonometrical tables. In fact, it is possible to make the machine itself choose the necessary set of values from 100 $\cos M6°$ $\cos N6°$ cards and punch the values of 100 $\cos (hM6°) \cos (kN6°)$ for different values of h and k by appropriate plug board wiring. Since the negative values of the function are given as Hollerith complements (*i.e.*, complements of 9999) only 3 figure accuracy is obtained by use of 4 column figure fields. From the cards for $A = 100$, cards for other values of A could easily be obtained automatically by means of the tabulator coupled to a summary-punch. A card of $A = + 200$ could, for instance, be obtained by tabulating two $A = + 100$ cards and summary-punching on to a fresh card. Similarly a card of $A = + 10$ could be obtained by reproducing the card $A = + 100$ by shifting 1 column to the right, after rounding off by adding 5 to the last decimal place.

5. USE OF THE TABULATOR METHOD IN FOURIER SYNTHESIS

To evaluate a particular double series (1), the following procedure could be adopted.

Corresponding to each value A for the frequency (h, k), we see that there are 16 cards for various values of M which are always kept together in a single packet. It may be necessary to take more than one packet to make up a particular amplitude A for the given frequency (h, k). The various packets for the different values of (h, k) are picked out from the card files, once and for all, by visual inspection. This is the only manual step in the synthesis.

All the cards are now put into the sorter and the cards divided into groups depending on the values of M by sorting along the columns 69 and 70. These 16 sets are now introduced one after another into the tabulator, which is made to print the sums every time the 'M' value changes by using control breaks on columns 69 and 70. The passage of the pack of cards through the machine 3 times, with the necessary changes in plug board wiring, is enough to give the sums in the 16 figure fields as well as the check columns. The final results are printed in the following form.

$M \downarrow$ \ $N \rightarrow$	0	1	2	. . .	15
0	×	×	×	. . .	×
1	×	×	×	. . .	×
2	×	×	×	. . .	×
.	.	.			
.	.	.	.	.	.
.	.	.	.	.	.
.	.	.	.	.	.
.	.	.	.	.	.
.	.	.	.	.	.
15	×	×	×	. . .	×

The value corresponding to (M_1, N_1) gives the sum of the series $\Sigma_h \Sigma_k A_{hk} \cos(hM_1 6^\circ) \cos(kN_1 6^\circ)$ for the point $(M_1/60, N_1/60)$. By keeping the odd and even frequencies separate, by sorting along column 71 and summing separately, we could get the values of the series over the whole of the unit cell, *i.e.*, for values of M and N from 0–60. If in addition to cos cos function, the series (1) also contains others of the form cos. sin, etc., these cards could be tabulated separately after sorting along the column 80 and the results may be suitably combined to give the final sum (1). When the number of such combinations becomes very large the computations could be made mechanical by summary-punching the results at every break in control columns to obtain cards which give the values (say, A, B, C, D) of the separate summations over the quarter cycle. Using the tabulator, various combinations of the values $(\pm A \pm B \pm C \pm D)$, depending on the frequencies and phases, could be obtained from these cards to give the final sum (1) over the whole of the unit cell. The accuracy of the summation could be tested by comparing the sum of the results for different values of N with the value given as the sum of the columns 65–68. Other checks like card counts, taking the sum of columns in some of the indicative fields, etc., could also easily be made to ensure correctness of the computations and of the cards used.

The essential steps in the method have been tested and found satisfactory.* Once the preliminary card files have been prepared, the

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computation of the series in any particular case is very simple, rapid and cheap. The preparation of the initial card files of over a 1,00,000 cards, however, is rather costly and hence even though the complete details have been worked out and the necessary " Electro " prepared, the project had to be set aside for the time being. However, it was thought worthwhile to present a concise report of the project in this paper.

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