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Bragg's Influence on Progress in Optical Computations

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An attempt is made in this paper to indicate the impact that one small part of Sir Lawrence Bragg's gross contributions have had upon crystallography during his lifetime up to the age of eighty. The small part referred to is the growing field of optical computation. Since this paper is partly for the enlightenment of the reader as well as a salute to history, descriptions of five basic optical operations are given: I, addition; II, multiplication; III, reciprocation; IV, convolution and V, diffraction. The role of Bragg and his co-workers is woven into each description. Finally as an extrapolation, a further process is described: VI, inverse imaging.

In a Festschrift in honor of a man having Sir Lawrence Bragg's long list of achievements, dating from the very beginning of X-ray diffraction to the present, there might be at least one report, giving in a brief manner, the high points of his scientific career; but it would require a large volume to do the subject justice in detail. It seems appropriate, however, to trace just one thread through his contributions, namely, optical computation.

This discussion is divided into six parts: I, addition; II, multiplication; III, reciprocation; IV, convolution; V, diffraction and VI, inverse imaging. All of these are basic mathematical processes capable of being performed by means of optics. The first five are closely associated with the works and inspirations of Bragg and his associates, while the sixth is really an extrapolation to illustrate what such pioneering work can lead to.

For the sake of brevity in symbolism throughout this report, the term $\varrho(xy)$ is used to designate the density, ϱ as a function of the distances x and y in the pattern, regardless of whether or not the pattern belongs to a repeating array such as is found in the projection of a crystal structure; the term $f(hk)$ designates the transform of one unit, whether it be one atom, a molecule, the content of an isolated unit cell, or any design or figure such as a portrait or a swastika [where h and k need not be restricted to integers (Wrinch, 1946)]; and the term $F(hk)$ designates the Fourier transform or structure factor of any array of designs that are positioned in a crystallographic manner (naturally h and k have only integral values).

I. Addition

Optical addition is illustrated schematically in Fig. 1(*a*), where O_1 and O_2 are evenly illuminated areas having designs on them such that the intensity of light reflected from them (or transmitted through them) is $I_1(xy)$ and $I_2(xy)$, proportional to $\varrho_1(xy)$ and $\varrho_2(xy)$ respectively. The lenses, L_1 and L_2 bring the images of O_1 and O_2 upon the screen Sc in register, with an intensity distribution $I_{12}(xy)$ on Sc proportional to the sum, $I_1(xy)+I_2(xy)$, or $\varrho_{12}(xy)=\varrho_1(xy)+\varrho_2(xy)$. Any number, N, of patterns, O_1, $O_2 \ldots O_j \ldots O_N$ can be added in this way. Or, if one wishes, one can use just one single lens and expose the O_j patterns one at a time in succession and the resulting multiple exposure of a photographic film placed at Sc can be developed to reveal the final sum.

The method of successive exposures was used by Bragg (1939) for the synthesis of Fourier terms of the kind, $F(hk) \cos 2\pi(hx+ky)$ and $F(hk) \sin 2\pi(hx+ky)$ where the O_j patterns were photographic films whose transparencies $T_j(xy)$ were proportional to $\cos 2\pi(hx+ky)$ or $\sin 2\pi(hx+ky)$ and the times of exposure were proportional to $F(hk)$. Thus Fourier summations of crystal structures were made.

The enthusiasm stimulated by this development by Bragg is indicated by the large number of related papers that followed. Huggins (1944) proceeded to put the method on a commercial basis, thus making available to the public the 'Bragg–Huggins Masks'. Wooley & McLachlan (1951) using the simultaneous exposure method shown in Fig. 1(*a*) developed what they called the 'multiple projector' (without lenses). Later Howell, Christensen & McLachlan (1951) produced masks that were more accurately reproducible photographically, and Howell & McLachlan (1955) built a table model projector with a Polaroid camera attached. Von Eller (1951) using opaque metallic strips that were rotatable and having axial motion (to change the size of their shadows) simulated masks, one at a time as they were needed during the summation. Von Eller's apparatus was produced commercially and was very successful. Similarly McLachlan (1957) built a 'synthesizer for triangular wave functions' using moiré patterns, which also simulated the masks O_j as needed during the synthesis.

Realizing that light is not the only medium that can be added in the Bragg manner, two other sets of authors came forward with new ideas. McLachlan & Champaygne (1946) used sand patterns to simulate the $\cos 2\pi(hx+ky)$ terms and Pepinsky (1947) developed a very usable machine (XRAC) using patterns of electrons striking an electroscope tube.

These means of simple addition are incorporated in other more complex processes which are to follow, but before going on, we should point out that a more general form of addition, recognized as integration is shown in Fig. 1(*b*). Here, all the points of the object patterns at O are to be added together through the action of the lens L which concentrates the radiation from O upon an area smaller than the face of the photocell PS.

It is interesting that if the patterns O_1 and O_2 in Fig. 1(*a*) were Patterson maps, and if they were shifted out of register by an amount **uv** corresponding to a through center vector on the Patterson map, a 'shifted Patterson sum' would be visible on the screen S_c. The literature on shifted Patterson maps in general, including the shifted Patterson sum, the shifted Patterson product, minimum function, vector convergence, image seeking function, *etc.* has been adequately recorded by Buerger (1959) and Lipson & Cochran (1953).

II. Multiplication

Optical multiplication is shown schematically in Fig. 2(*a*) where parallel light striking the transparent film, O_1 having transparency $T_1(xy) \simeq \varrho(xy)$ and on through film O_2 having transparency $T_2(xy) \simeq \varrho(xy)$, is brought to focus (through the lens L_2) upon the screen Sc. The image on Sc will have a brightness $B(xy)$ proportional to the product $T_1(xy)\ T_2(xy) \simeq \varrho_1(xy)\ \varrho_2(xy)$. A simpler way to get optical multiplication is shown in Fig. 2(*b*), where the films O_1 and O_2 are in contact with one another and with the screen Sc.

If for example O_1 and O_2 in Fig. 2 were identical and represented the electron density $\varrho(xy)$ in a unit cell of a crystal, then a 'squared' structure would be visible at S_c. The squared structure we have in mind is the one mentioned by Sayre (1952) in his paper on the squaring method of sign determination. Or if identical weighted reciprocal lattices, $F(hk)$ were at O_1 and O_2, then $|F(hk)|^2$ would appear on Sc. While both of these illustrations of multiplication might be considered trivial, one that is not trivial is the suggestion that if two identical Patterson maps $A(uv)$ were placed at O_1 and O_2 and shifted with respect to one another an amount equal to a through center vector $\mathbf{u}_{ij}\mathbf{v}_{ij}$, then at the screen Sc would be seen a shifted Patterson product (see references in Buerger, 1959).

Bragg & Lipson (1936) reported on some longhand computations in which optical multiplication could have been used. They computed maps for such terms as $\cos 2\pi hx \cos 2\pi ky$, $\cos 2\pi hx \sin 2\pi ky$, *etc.* which are really products of the Bragg masks which he developed three years later as mentioned above (Bragg, 1939). As Bragg points out, not many of these product maps need be computed. This means that if transparent masks of these product maps were used, not so many would have been required for the Fourier synthesis. However, this first paper by Bragg (Bragg & Lipson, 1936) was a pioneer work which was later carried on and applied to molecules by Knott (1940) and other workers. Molecular transforms and transforms of point sets in general are elegantly treated in a monograph by Wrinch (1946).

III. Reciprocation and division

The procedure for finding the reciprocal of a pattern $\varrho(xy)$, that is, finding a pattern whose density if $R(xy) = 1/\varrho(xy)$ is almost trivial from the standpoint of operation, but is of significance for purposes to be discussed later. Fig. 3 shows a floodlight, FL, evenly illuminating a transparent pattern at O and a transparent film, TF in contact with it. The transparency, $R(xy)$, of the film, TF, after development should be proportional to the reciprocal $(1/T(xy))$ of the film at O whose transparency is $T(xy)$. This is because of the nature of the Hurter–Druffield curve (shown on page 151 of Goodman, 1968) which exhibits a straight line portion obeying the equation relating density, D, and exposure, E

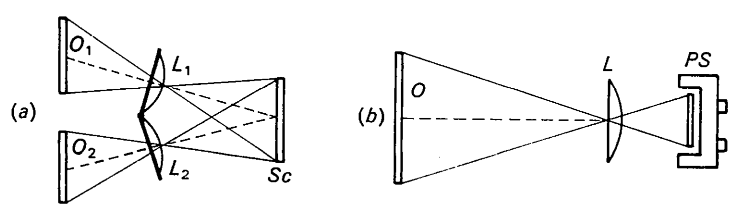

Fig. 1. (*a*) Showing optical addition by the superposition of images and (*b*) showing optical integration with a lens and photocell.

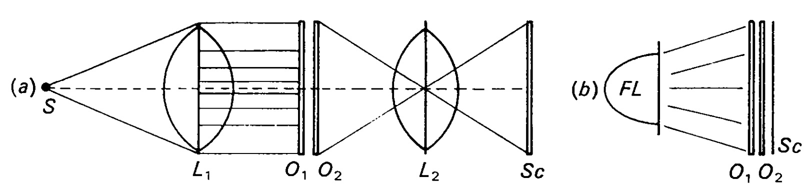

Fig. 2. (*a*) Showing optical multiplication using lenses and, (*b*) showing multiplication by contact printing of two patterns.

$$D \equiv \gamma \log E$$

or

$$\log \frac{I_0}{I_{tr}} = \gamma \log I_e t \, .$$

Letting

$$I_e = I_0 T(xy), \; I_{tr} = I_0 \, R(xy)$$

and solving for $R(xy)$:

$$R(xy) = \frac{I_0}{[I_0 t \; T(xy)]^\gamma} \, .$$

When

$$\gamma = 1$$
$$R(xy) = K/T(xy) \, .$$

With proper care one should be able to use photographic transparencies to obtain maps of $1/\varrho(xy)$, $1/f(hk)$ or $1/F(hk)$ which we will use in a later section of this paper to get inverse images.

This reciprocation procedure can be used in preparation for the process of carrying out optical division. Since the division of one number by another is equivalent to multiplying one of the numbers by the reciprocal of the other, one can place a transparency $T(xy)$ at O_1 in Fig. 2 and the transparent negative of $T_2(xy)$ at O_2 and get $T_1(xy)/T_2(xy)$ on screen S_c. When $T_1(xy) = T_2(xy)$, a uniform field should be obtained. But a popular stunt by professional as well as amateur photographers is to shift the two films slightly out of register to get a bas-relief kind of picture.

IV. Convolutions

For future reference we write here the mathematical expression for the cross convolution process in two dimensions,

$$A(uv) = \frac{1}{A} \int_0^a \int_0^b \varrho(xy) \, \varrho'(x+u, y+v) \, dxdy$$

which becomes an auto- or self-convolution when $\varrho(xy) = \varrho'(xy)$, familiarly recognized as a Patterson map. It involves the basic operations of multiplication and integration [(see Fig. 2(*a*) and (*b*)]. Fig. 4(*a*) shows at O_1 a map or film whose scattering power or transparency is proportional to $\varrho(xy)$, and at O_2 at one-half scale, a film whose transparency is proportional to $\varrho_1(xy)$. The convolution map shows on the screen S_c.

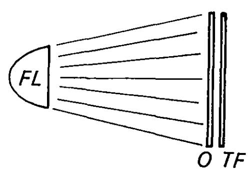

Fig. 3. Optical reciprocation by contact printing on a transparent negative film.

The first synthetic map to be produced optically was made by Robertson (1943) who used an arrangement of separate electric lights at O_1 to represent the planar configuration of atoms and holes similarly arranged in an opaque sheet at O_2. Bragg (1944) introduced a lens as shown in Fig. 4(*b*) which avoided the necessity of reducing the scale at O_2. Vand (1944) made two other modifications using mirrors which enabled one to produce Patterson maps with only one copy of the $\varrho(xy)$ map. Also Hägg (1944) and Booth & Wrinch (1946) contributed to the development of synthetic Patterson maps, while Philips & McLachlan (1954) considered the Robertson method in relation to other operations.

The general application of convolutions is much broader in scope than is generally realized. For example, the ordinary pin-hole camera is really a means of performing a convolution of an object O_1 against a pin-hole at O_2 [Fig. 4(*a*)] and in which the resolution of the picture at C is a function of the size of the pin-hole, the distance from O_2 to C, and the wavelength of the light used. The multiple pin-hole camera of Bragg (1944) also involved convolutions in a minor role. The shifted Patterson sum need not be done by straight addition as described earlier, but can be simulated reasonably accurately by placing an $A(uv)$ as Patterson map at O_1 [Fig. 4(*a*)] and two pinholes at O_2 spaced in accord with the desired shift.

Remembering that the maps O_1 and O_2 need not always represent electron densities, $\varrho(xy)$, but can also represent Fourier transforms $f(hk)$ and $F(hk)$, one can think of other uses for them. For example, they could be used to compute $F(H')\,F(H'-H)$ which Sayre (1952) called 'inner products'. Also, the concept of convolutions is not confined to crystallographic problems. For example, McLachlan (1962) proposed the use of optical convolutions as a means of pattern recognition, based upon the fact that two identical patterns (whether they be numerals, letters, portraits, fingerprints, *etc.*) give self-convolutions which have a pronounced peak at the center, while unlike patterns placed at O_1 and O_2 of Fig. 4(*a*) produce no such central peak. It will be shown in the last section of this paper that the concept of inverse images, derived through optical means is an even better approach to the growing and urgent problems of pattern recognition and retrieval.

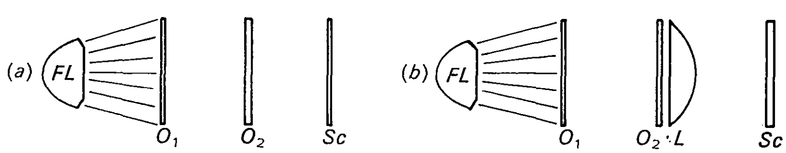

Fig. 4. (*a*) Optical convoluting without lenses (Robertson, 1943) and, (*b*) using a lens (Bragg, 1944).

V. Optical diffraction

Although spectroscopists have been applying diffraction phenomena for many decades to determine the intensities of the various wavelengths of radiation using a known pattern for their diffractor, the converse, *i.e.* the use of radiations of a known wavelength to analyze the diffracting object, is much newer. Before any Fourier transforms had been computed or purposely simulated, Abbe (1873) showed the role that diffraction plays in image formation by lenses. Many papers have been written (see references in Wrinch, 1946; Taylor & Lipson, 1965; and Lipson & Cochran, 1953) on the computation of Fourier transforms of atoms, molecules and projected crystal unit cells. According to Taylor & Lipson (1965, page 2) the propagandist (chiefly optical) for simulating X-ray diffraction effects 'was undoubtedly W. L. Bragg,' starting with a paper in *Nature* (Bragg, 1939).

It is of interest to point out that besides the book by Wrinch (1946) on computation methods and that by Taylor & Lipson (1965)* on optical methods, there is a newer book by Goodman (1968) devoted strictly to Fourier optics showing the viewpoint of physicists who are specialists in optics.

There have been two types of optical diffractometers put to use under different modifications by the various authors. One type is based entirely on lenses for focusing purposes and the other uses a mirror. Both of these have been described in the book by Taylor & Lipson (1965) with adequate detail for anyone to build a diffractometer for himself from their descriptions. The lens focusing optical diffractometer is shown briefly in Fig. 5, where light from the source S is concentrated on a pin-hole, PH, by the lens, L. The pinhole, acting as a coherent source, illuminates the pattern, O, after being collimated by the lens, L_2, and the modulated radiation from O is brought to focus on the plane D by means of the lens L_3. If a transparent pattern, $\varrho(xy)$ is placed at O_1 then the screen D reveals a diffraction pattern, $F^2(hk)$ of $\varrho(xy)$. For early suggestions, see Ewald (1940) and Knott (1940).

When the Fourier transform of an atom is desired, the $\varrho(xy)$ or $\varrho(r)$ of the atom is usually represented as a round hole in a thin opaque sheet; or if the $\varrho(xy)$ of a single unit cell of a crystal, or a single molecule is desired, the holes in the sheet at O are arranged according to the arrangement of atoms in the unit cell or molecule. To get the phases or signs of $f(hk)$ in $f^2(hk)$, one can apply methods suggested on pages 77 to 79 of Taylor & Lipson (1965) or one can give considerations regarding symmetry according to Wilson (1942) and others. When the diffraction from a projected crystal is desired, a two-dimensional array of $\varrho(xy)$ for a unit cell can be produced using the 'fly's eye' (Bragg, 1944; Crowfoot, Bunn, Rogers-Low & Turner-Jones, 1945) and the $|F(hk)|^2$ pattern appears at D in Fig. 5 showing only integral values of h and k. These methods are quick ways of checking trial structures.

Remembering that the diffraction from a diffraction pattern produces an image of the original pattern, one can see that a transform $f(hk)$ or $F(hk)$ can be placed at O in Fig. 5 and a $\varrho(xy)$ will appear at D. Bragg (1939) produced a projected image of the structure of the mineral diopside in this way using thin disks over the holes in O (Fig. 5) to regulate the phases by wave retardation. Buerger (1950) used tiltable mica discs over the holes. These Fourier syntheses, according to the reasoning of Abbe (1873) and Goodman (1968) are applicable to any kind of a pattern whatsoever.

VI. Inverse images

As far as is known, the first introduction to the concept of inverse images was by McLachlan (1969). It can be defined by reference to Fig. 4(*a*) which is a set up for cross convolutions. It is postulated that, for each and every pattern $\varrho(xy)$ that is placed at O_1 in Fig. 4(*a*) there can be found a pattern $\varrho^{-1}(xy)$ to be placed at O_2 which when convoluted against the pattern at O_1, will produce upon the screen Sc, one single point at the center. Also, if there are a number of identical patterns on O_1, then one inverse image of that pattern placed on O_2 will convolute to produce an equal number of points on C. Or, if the structure of a unit cell were placed on O_1 and an inverse image of an atom were placed on O_2, then a point atom structure would appear on C.

In the original paper, McLachlan treated the problem as a matrix problem. An imaginary one-dimensional cell containing six Gaussian atoms was divided into eight equal parts and a histogram $P(n)$ was drawn in. Then a histogram of a single Gaussian atom was similarly drawn. The inverse of an eight by eight matrix had to be solved by computer to give the histogram of the 'inverse Gaussian atom'. A numerical convolution of the histogram of the inverse Gaussian atom was performed against the six atom histogram of the cell, the results of which gave a highly satisfactory positioning of the centers of the six Gaussian atoms in the original

* Taylor & Lipson's book alone has references to eleven of Bragg's papers.

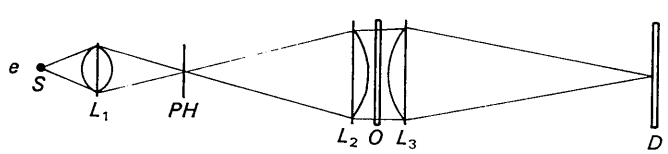

Fig. 5. Optical diffraction using lenses.

cell. This resolution of the atomic positions operated in spite of the fact that there was only one maximum in the original $\varrho(xy)$ and it was not at an atomic position.

In order to lead up to optical methods for studying inverse images, let us look at the well-known cross convolution equation:

$$B(uv) = \frac{1}{A}\int_0^a\int_0^b \varrho(xy)\varrho'(x+u, y+v)\,dxdy$$
$$= \frac{1}{A}\sum_h\sum_k F(hk)\,F'(hk)\exp -2\pi(hu+kv)] \quad (1)$$

where $F(hk)$ is the Fourier transform of $\varrho(xy)$ and $F^1(hk)$ is the Fourier transform of $\varrho^1(xy)$. In comparison, the people who get point atoms in the structure by dividing $F(hk)$ by $f(kh)$ (see Harker & Kasper, 1948) use an equation of the form:

$$\Pi(xy) = \frac{1}{A}\sum_h\sum_k F(hk)\frac{1}{f(hk)}\exp[-2\pi i(hx+ky)]\,. \quad (2)$$

A comparison of equations (1) and (2) reveals that $1/f(hk)$ is the Fourier transform of some structure $\varrho^1(xy)$ and that the structure $\Pi(xy)$ of point atoms is inadvertently a cross convolution $B(uv)$ of the structure $\varrho(xy)$ and some other structure $\varrho^1(xy)$ which in turn is the structure of an inverse atom, $\varrho^{-1}(xy)$. The structure of the inverse atom is then:

$$P^{-1}(xy) = \frac{1}{A}\sum_h\sum_k \frac{1}{f(hk)}\exp[-2\pi i(hx+ky)]$$

According to the discussion of reciprocation in §III above, the term $1/f(hk)$ can be obtained from $f(hk)$ by making a transparent negative of $f(hk)$ using film with matching γ's. This should be useful in sharpening Patterson projections, in fact, in the original paper it was shown that it resolves atoms that are totally hidden by overlaps.

Much discussion could be given to this subject, but at this time, it is best to only state that, at The Ohio State University, optical apparatus is being built to carry on investigations with atoms, molecules and patterns in general for help in problem in informations retrieval. Beyond this single projected suggestion, it is very difficult to predict the many future uses that optical computations will be put to in aid of the various branches of science, based on the work of pioneers such as Bragg and his co-workers.

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