In demonstrating the advantageous use of graphics in a heuristic approach to problem solving, the deciphering of unknown crystal structures is used as an environment. The interactive graphic console lends concreteness to the crystallographer's abstract perception of three-dimensional structures.

This paper describes how the crystallographer uses graphics in determining crystal structure by matching a known spatial molecular model with an experimental crystallographic model and by direct theoretical procedures when no model is available. Also presented are computer rendering techniques that make the man-machine relationship more productive.

In conclusion, the author ponders the possibilities of more efficient and productive scientific research through the evolution of computer-aided data libraries.

INTERACTIVE GRAPHICS IN DATA PROCESSING

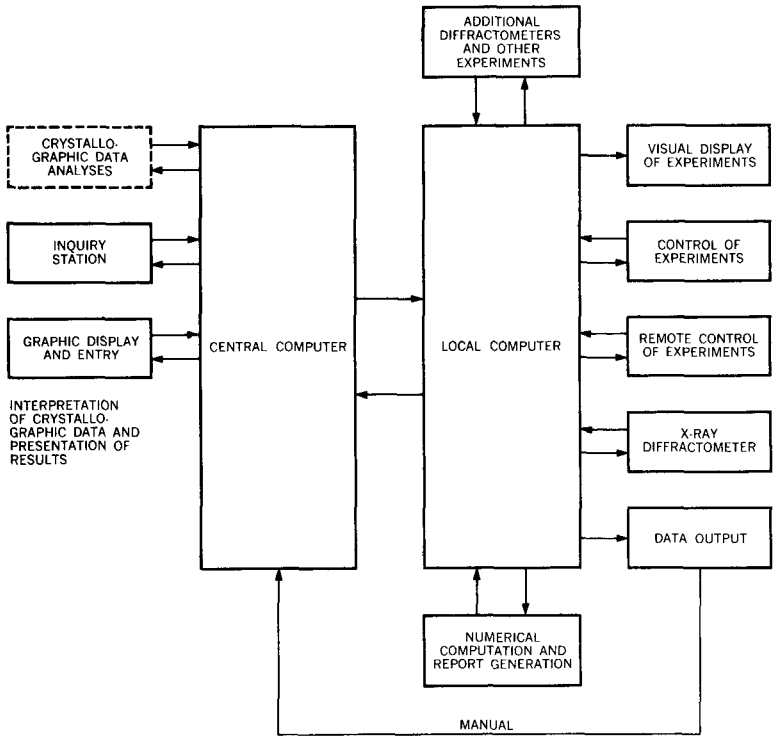
Interactive aspects of crystal structure analysis

by Y. Okaya

Graphic display and entry facilitate the dynamic exchange of information between the computer and human intelligence—they provide efficient man-machine interaction. Human interaction with the computer is useful in crystal structure analysis because man's ability to recognize crystallographic patterns far surpasses his ability to deal with digitized information such as printed number lists or bit configurations. The full capability of graphic systems, in conjunction with the time-sharing of computers, continues to be explored.

During the last decade, uses of computers in crystallography have developed extensively, encompassing such diversified areas as large-scale computation, control of data collection processes, display of crystal structures, and preparation of structural results for publication. The close kinship between the crystallographic techniques and computers is shown in Figure 1. The left side of the figure deals with data analyses using mainly high-speed computers with large storage capacity. The data analyses can be classified as relatively large-scale numerical computations, such as least-squares treatments of 200 unknowns with 2,000 nonlinear equations. The right side illustrates the role of computers in obtaining diffraction data from crystals. Developments in this area are relatively new, but it has already been established that the control of data collection systems can be successfully carried out.[1]

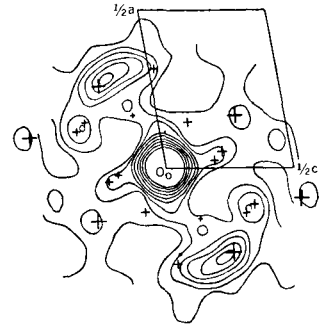
Figure 1 Crystallography and computers



The object of crystal structure analyses by the x-ray and neutron diffraction methods is to determine the arrangements of atoms in crystals. Unless experimental difficulty arises, it is also possible to derive amplitudes of thermal vibration of atoms to a certain degree of accuracy.

Crystal structure determination can be divided into four distinct stages. The first stage is the measurement of the intensity of each reflection of the incident x-ray beam at the diffraction angle which can be made automatically by a computer-controlled diffractometer. In the second stage, reflection-intensity data are corrected for geometrical and absorption factors, and an approximate model of the crystal structure is derived by means of the Patterson function, an example display of which is shown in Figure 2. Such a function represents the distribution of interatomic vectors derived from the actual electron-density distribution within a crystal. Since there is no truly automatic way of deciphering an interatomic vector map, the experience and knowledge of crystallographers are required here. Another approach is called the "direct method," and one of the relationships used extensively is called the "symbolic addition method." The third stage is a refinement of the approximate structure model, obtained from the electron-density function, to produce a quantitatively accurate picture of the crystal being studied.

Figure 2 Display of a Patterson function



The final stage is the interpretation and presentation of results using parameters derived in the previous stages. Here, computers are used for such activities as evaluating interatomic distances and angles, deciding the nature of thermal vibrations of atoms, data management, and graphic outputs.

A discussion of the disciplines of crystallography and mathematical methods, which are implicit in the above four stages, is not our intention here. Rather, this paper focuses on the graphic output of a time-shared computer system.[2] Here, we confront one of the ubiquitous graphics problems—how to effectively display three-dimensional objects, such as crystal structures, on a two-dimensional surface.

Computer graphics intensifies the crystallographer's perception of crystal structure.[3] To convey some of this perception to the reader, we describe the man-machine interactions involved and the results obtained.

Displaying three-dimensional objects

There are a number of systems that can be used to convert digital information into recognizable and aesthetic displays. One system that has been used in drawing crystal structures is the x-y plotter. Another display device especially useful because of its data entry capability is the IBM 2250 graphic display console.

The 2250 is most useful in the dynamic display and interpretation of results in time-shared environments—an example of man-computer interaction. This console has a display screen that generates displays by a combination of general strokes and modulation strokes (for alphanumeric characters). Alphanumeric data may be entered through a keyboard; these entries may be data, commands, or other inputs.

Another important, and probably more convenient means of entry to the 2250 is the light pen by which a point on the cathode-ray tube can be identified to the computer as a set of x-y coordinates. With proper programming, the light pen can be used to indicate corrections, insertions, deletions, and modifications to the display. Given such a capability, the ingenuity of the crystallographer can suggest an unlimited number of ways in which the system can augment his professional ability.

In considering methods of projecting three-dimensional objects on two-dimensional media, some limitations of the computer in comparison to the human being are apparent. A human being sees an object and recognizes its three-dimensional quality by his own detection system—a combination of biological I/O devices (the eyes) and a decoding logic organ (the brain). He can easily decide which part of the object is hidden from view by other parts simply because he cannot see the hidden part. However, if the computer draws a view of an object, entered as digital information, the user must provide appropriate programs for the computer to determine which parts of the object are hidden by another part.

The visibility, or hidden-line, problem is a relatively difficult one to solve, and it seems that there is no good universal method. However, in the case of crystal structures, the problem may not be as difficult as in the general case. We can represent crystal structures by combining such relatively simple objects as circles, ellipses, and lines.

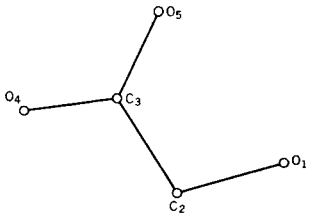
There are various methods of expressing a three-dimensional feeling on two-dimensional media. Although a full discussion of such problems is beyond the scope of this paper, we can indicate several methods that are applicable to computational methods.

1. For a relatively complicated object, select a view that gives a moderate amount of overlap of the various parts.
2. If the object is composed of several parts of the same or similar shapes, create perspective by changing their size according to the distance from the viewer.
3. In addition to changing size, also change shade, intensity, or brightness of the image to emphasize the impression of distance by psychological cues.
4. Use combinations of methods 1 to 3.
5. Rotate and/or translate the object in such a way as to show relationships among its parts.
6. As an improvement over 5 alone, use stereoscopic methods in conjunction with 5.

Graphic display in crystallography

We now turn to the use of graphic display systems in crystallography. At various stages in our work, we deal with problems of matching two three-dimensional objects. For example, one of the objects may be a part of the Patterson function while the other may be part of a molecule of known configuration. This is a typical method when one is establishing the orientation of a group of atoms in an unknown crystal. The problem of matching the two objects—Figures 2 and 3, for example[4]—is a very difficult task for the computer that requires strict adherence to an algorithm. The problem is especially difficult when one object is a combination of more than one component, and when both objects are further distorted by various experimental restrictions. This is a so-called heuristic[5] problem where human intelligence, although weak in repeating an established algorithm, demonstrates its superiority.

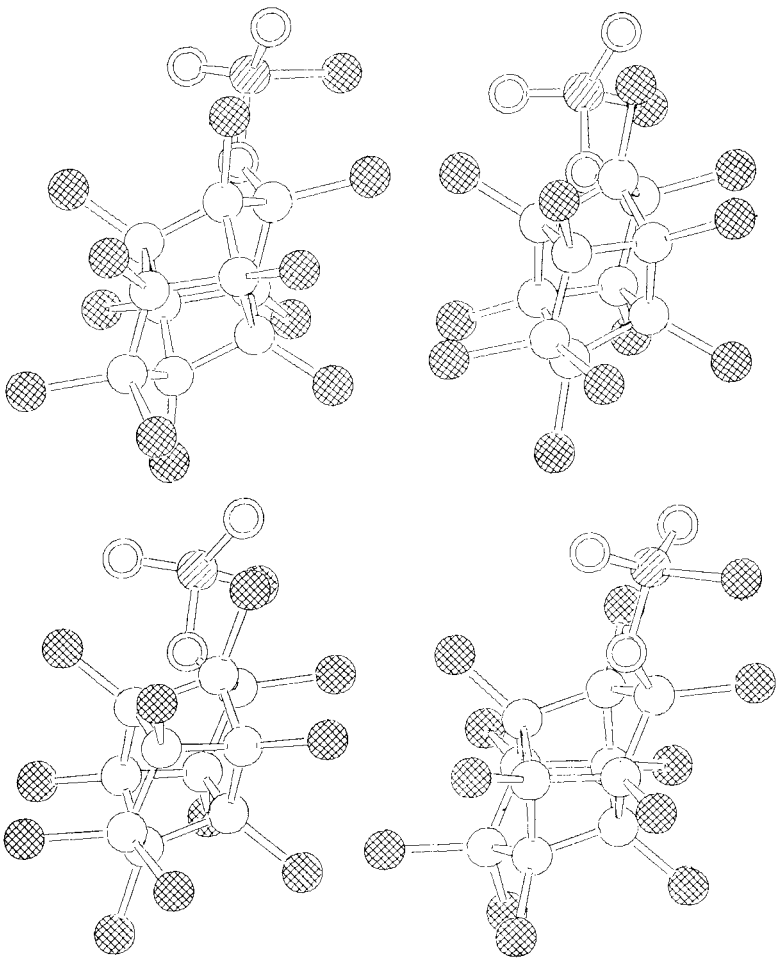
Figure 3 Chemical representation of a molecule



known molecular model

The interesting part of the three-dimensional Patterson function can be presented on the display console. In cases of functions with contours, one of the useful methods of presentation is to dim the contours of sections of less interest to the viewer by reducing the frequency of sweeps. Also, known parts of the molecule may be rotated under computer control and results displayed dynamically. Whenever the crystallographer decides that a reasonable fit—not always the rigorous one—has been obtained, he can enter his decision into the computer through the keyboard in interrupt mode.

Figure 4 Calculated views of a chlorosulfonate molecule

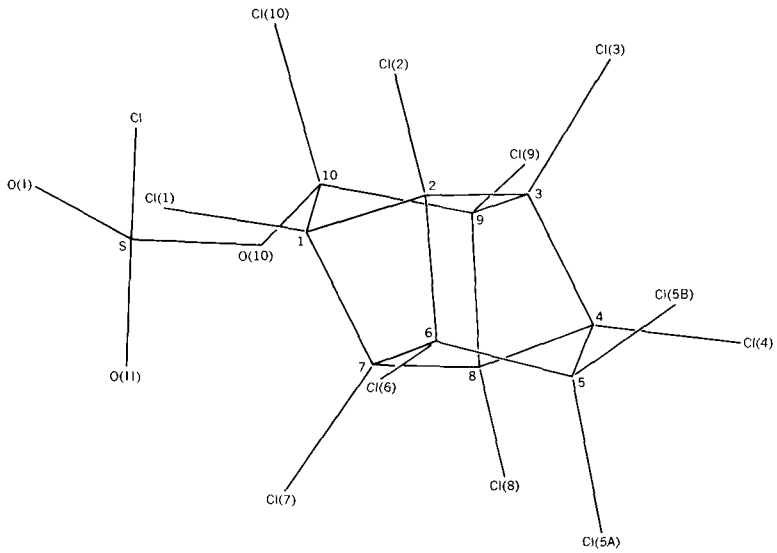


There is a program to perform this matching task on the IBM 7090 using strictly a computer algorithm, and it is of interest to compare the efficiency of the computer's recognition capability with our own rather obscure, compromise-making recognition capability. Computations for a 2250 display system are only those required for the linear transformation of the shape of the molecule. The matching of chemical models with electron-density functions of large molecules, such as proteins, may be similarly treated. Using a computer, it is relatively easy to construct a model of at least part of a protein molecule by means of its amino-acid sequence with certain constraints;[6] these results can again be compared to approximate density functions. The help given by the system may make protein structure determination more feasible than it presently is.

direct structure determination

Thus far, we have discussed the case where the shape of the molecule is known. Let us now consider the direct determination of unknown crystal structures using the symbolic addition method.[7]

Figure 5 Chemical representation of a chlorosulfonate molecule



Recent analysis of a chlorosulfonate structure, having the novel cage chlorocarbon[8] configuration shown in Figure 4, might serve as a good example. It was not possible to postulate the molecular configuration at the outset, because the molecule belongs to a new class of compounds. Therefore, when a reasonable electron-density function was obtained by the symbolic method, we were confronted with the problem of interpreting the results by deciphering the function. Usually, under such circumstances, it is an appropriate procedure to construct a model by hand using the peak heights and peak positions in an approximate density map—a rather time-consuming method. However, we may decide to let the computer draw the molecule, as viewed from several directions, using the electron-density data.

Figure 4 shows four views of the molecule calculated on the IBM 7094 and drawn by an x-y plotter. Inspection of these views immediately led to a good understanding of the shape and size of the molecule, a chemical representation of which is shown in Figure 5. The views in Figure 4 could also be displayed dynamically on the 2250 console.

In Figure 4, notice that the computer eliminates the overlapped part of the bonds and atoms. The atomic coordinates used in making these drawings were obtained by an automatic peak-search program for scanning the electron-density function and giving peak heights and the peak locations. The shading of the atoms depends upon the peak heights. Thus, in a sense, the drawings were obtained automatically from the three-dimensional electron-density function deduced by the symbolic addition method.

Since the method has proved to be effective in determining crystal structures, there is no doubt that more and more structures, especially those with unknown chemical configurations, will

be studied by the direct method. The display-entry capability of the 2250 console is expected to be a great help in this method of analysis. The trend toward using the direct method should be accelerated by the recent work that extends the technique to noncentrosymmetric structures.

other crystallographic applications

In connection with, or in addition to activities in structure solving, computer display methods are advantageously used for drawing clear, well-understood, and aesthetic representations of crystal structures. One of the most time-consuming parts of crystallographic studies, probably the worst bottleneck, is the hand drawing of adequate illustrations for publication. The graphic display system, be it a display screen or a much slower x-y plotter, can be used extensively for such purposes. For example, Figure 6 gives several views of an optically active silicon compound[9] (α-naphthylphenylmethylfluorosilane, $Si^* \cdot FCH_3C_6H_5C_{10}H_7$). These views were drawn by an x-y plotter, based on calculations made on a 7094, which computationally rotated the molecule around two perpendicular axes. One may choose the view or views that best suit the purpose. This figure, if done by hand, might take so long that the whole operation would be unfeasible. However, when done by a computer, it is a relatively simple mathematical procedure to compose these views.

Another application of computational rotation is the generation of stereoscopic pairs to illustrate crystal structures.[10] Figure 7 is an example of such a stereoscopic pair drawn by using an Oak Ridge National Laboratory program. Such figures can also be made on a display screen, and viewed directly with a proper mirror system. A stereoscopic motion picture of a rotating molecule was produced at the Lawrence Radiation Laboratory. It should be emphasized that the effect of good, easily recognizable illustrations of results is important and striking to the general scientific world.

Concluding remarks

On the basis of our experience in computer-aided crystallography, one might envision a hypothetical scientific data library system—or a method of handling the results of scientific work. A number of studies are being made of ways to place data in machine-readable form. Once in this form, data can be retrieved, corrected, correlated, and further analyzed. With the display capability of a computer-data retrieval system there is no need to store graphics and curves directly.

Take the example of putting the results of a thermal study into such a form. Suppose we have access to data that gives specific heat at constant pressure (C_p) as a function to temperature (T). When we call the data into the library computing system, we can easily compute and display the curve of C_p versus T. However, we can go beyond this point and, if it seems desirable to plot C_p versus T^2 and test the dependence, we can easily do this through a keyboard entry.

Figure 6 A molecule computationally rotated about two axes

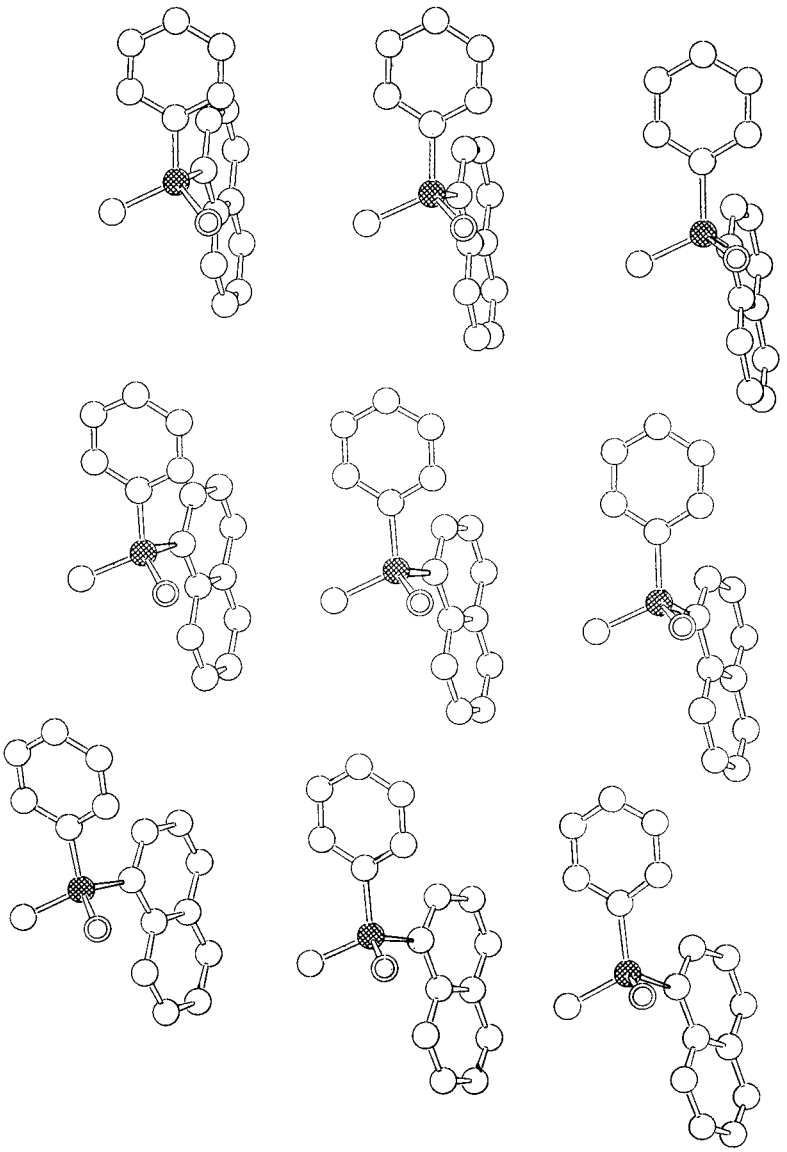
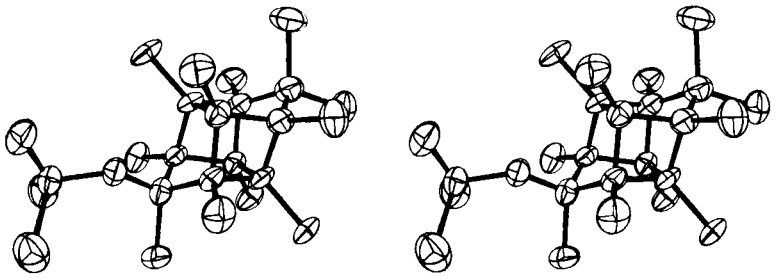


Figure 7 A computer-produced stereoscopic pair



Other useful computer facilities might be proposed for such a data library. Light pens can be used in describing or indicating the desired work. In this way, we can perform many more numerical analyses and display the results in a dynamic fashion. The library system could also include the projecting of slides and the playing of recorded messages simultaneously, with or without computer control, and might resemble computer-aided instruction systems. A programming system for such library usage would have to be developed, especially for easy manipulation of stored data and display of the results. Naturally, the envisioned system would have some capability of compiling programming language statements. It should also be able to address specified data in the literature as required for computation.

For the present, it might be useful simply to suggest how such a library computing system with machine-readable crystal structure data may be used. For example, one might compute interatomic distances and angles that were not mentioned in a publication or, similarly, produce structural views not included in a publication. Also, any part of the structure of interest might be expanded and studied more carefully.

Thus, we are imagining, by means of an example familiar to the author, how computer-aided displays might evolve to make scientific research more efficient and more productive.

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