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Fluorescent Sources for X-ray Diffractometry

BY W. PARRISH, K. LOWITZSCH AND N. SPIELBERG

Philips Laboratories, Irvington-on-Hudson, New York, U.S.A.

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Various experimental arrangements were tried, using fluorescent X-ray sources. Their brightness was extremely low and for conditions of comparable resolution the intensities were about a few tenths of one percent of those obtained with the usual direct X-ray tube source. The effect of instrumental factors on line width and the reasons for the large decrease in intensity are outlined.

1. Introduction

Fluorescent radiation has been used for monochromatic X-ray sources since the very early days of X-ray analysis (Barkla, 1911). In recent years fluorescent sources have been used in a variety of applications, such as absorptiometry (Engström, 1947), microradiography (Rogers, 1952; Splettstosser & Seeman, 1952), and as wide-beam sources (Larson, Myers & Roesch, 1955). In a recent communication, Weiss, DeMarco & Weremchuk (1954) compared the results obtained using a fluorescent source with the usual X-ray tube source for powder diffractometry.

At first thought the use of a fluorescent source would seem to offer several attractive advantages for powder diffractometry: There is practically no limit to the choice of radiation, the use of a β-filter with the fluorescent source gives nearly monochromatic radiation without the continuous spectrum inherent in efficient direct electron excitation, sub-harmonics reflected by crystal monochromators are not present, and a source with a more uniform intensity distribution is readily obtained.

On the other hand, fluorescent sources of X-rays are of such low inherent brightness compared to conventional X-ray tube sources that it is necessary to sacrifice resolution in order to obtain usable intensities for powder diffractometry. There are many factors in the geometry of the instrument, the size and brightness of the X-ray source, and the characteristics of the X-ray detector system that combine in a complex manner to determine the intensity, resolution and peak-to-background ratio of the X-ray patterns. To properly evaluate the fluorescent source, it is necessary to separately determine the contribution of each of these factors. In this paper we will describe two types of fluorescent sources and compare the results with those obtained using a standard diffractometer with conventional X-ray tube source.

2. Experimental arrangements

The usual diffractometer apparatus (Fig. 1(*a*)) employs the line focus of the X-ray tube, *X*, as the geometrical source of the focusing arrangement. The

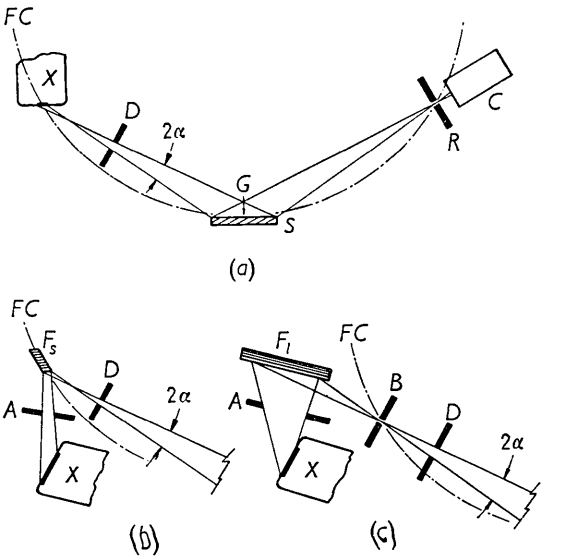

Fig. 1. (*a*) Conventional diffractometer arrangement; (*b*) small fluorescent source, Method I; (*c*) large fluorescent source, Method II. *FC*, focusing circle; *X*, X-ray tube; *D*, divergence slit; 2α, full angular aperture in focusing plane; *S*, specimen; *G*, goniometer axis of rotation; *R*, receiving slit; *C*, counter tube; F_s, small fluorescent source; F_l, large fluorescent source; *A*, slit limiting aperture of primary beam; *B*, source slit on *FC*. The metal foils of the parallel-slit assemblies (not shown) are parallel to the plane of the drawings.

divergence slit, *D*, limits the angular aperture 2α of the primary beam in the focusing plane (i.e., the plane of the drawing). After reflection by the specimen, *S*, the beam converges at the receiving slit, *R*, and is measured by the counter tube, *C*. The focusing circle, *FC*, passes through *X*, the goniometer axis of rotation *G*, and *R*.

Two devices were built to use fluorescent sources with the standard Norelco diffractometer (Parrish, Hamacher & Lowitzsch, 1954). In place of the standard X-ray diffraction tube, a Machlett OEG-50 tungsten-target tube was used. The latter has a 5×10 mm. focal spot, and hence may be operated at about 2·5 times greater power than the standard copper-target diffraction tube. The window transmission of both

tubes is about the same in the wavelength region considered here (Taylor & Parrish, 1955).

Method I

In this arrangement (Fig. 1(*b*)), a rectangular block of polished pure copper, F_s, was mounted at the focusing circle position. The end of the copper exposed to the primary X-ray beam was $1{\cdot}5\times10$ mm., approximately the same size as the line focus of a standard diffraction tube. A viewing angle $\psi = 3°$ (the angle between the surface of the target and a ray from the center of the focal spot to G, as shown in Fig. 2)

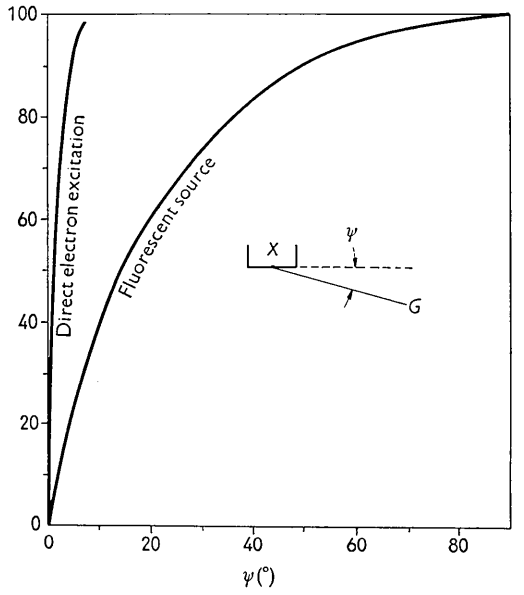

Fig. 2. Intensity as a function of angle-of-view ψ of the target surface for direct electron excitation used in X-ray tubes, and X-ray induced excitation as in fluorescent sources.

was used. The projected size was thus the same as in standard diffractometry. To minimize scattering, a lead slit, A, was placed on the X-ray tube window in order to reduce the primary beam to slightly larger than the exposed copper area. The copper was bevelled at a 5° angle so that the side facing the goniometer could not be irradiated by the primary beam.

Method II

In this arrangement (Fig. 1(*c*)) large flat polished pieces of pure copper or cobalt, F_l, were irradiated by the primary X-ray beam, and the fluorescent X-rays passed through a source slit, B, placed on the focusing circle. By rotating F_l, ψ could be varied from 0° to 25° and read from a scale. Method II was used by Weiss *et al.*, and the major differences between this arrangement and the standard diffractometer are listed in Table 1.

In both methods the X-ray tube and fluorescent source were enclosed in a ray-proof assembly to eliminate scatter.

Table 1. *Comparison of standard and fluorescence diffractometer*

	Standard diffractometer(*a*)	Fluorescent conversion(*b*)
Source size	$0{\cdot}08\times10$ mm., $\psi=3°$ $0{\cdot}16\times10$ mm., $\psi=6°$	$1\times15{\cdot}9$ mm. —
Goniometer radius	17 cm.	14 cm.
Angular aperture 2α	1°	1·68°(*c*)
Parallel slits, $2\delta=4{\cdot}5°$	Two sets	One set(*d*)
Receiving-slit height	0·3 mm.(*e*)	1 mm.

(*a*) Norelco, described by Parrish, Hamacher & Lowitzsch (1954).
(*b*) Arrangement described by Weiss, DeMarco & Weremchuk (1954).
(*c*) This is incorrectly given as 1·25° in their Fig. 2.
(*d*) The Norelco parallel-slit assembly is 10·5 mm. wide and hence the full width of the source was not effective.
(*e*) Better resolution is obtained with a receiving slit 0·15 mm. high.

For given excitation conditions the fluorescent intensity obeys the inverse square law, and it is therefore essential to minimize the distance between the X-ray target and fluorescence source. However, there must be enough separation between the X-ray tube window and the fluorescent source to allow taking off the fluorescent beam. To meet these conditions in Method I, the minimum distance between the center of the target and surface of the source was 2·5 cm., and in Method II, 3·0 cm. The distance between center of the target to the front of the X-ray tube beryllium window was 2·1 cm.

3. Source factors

(*a*) *Intensity versus angle-of-view*

The observed intensity of X-rays generated by direct electron excitation increases rapidly above $\psi = 0°$, approaching its maximum value asymptotically, as shown in Fig. 2. The slope of the intensity versus ψ curve depends on the applied voltage and density of the target material. For the usual conditions employed in diffractometry, the maximum intensity is reached at about $\psi = 6\pm2°$ because the electrons are stopped so close to the target surface that only a very thin layer acts as the X-ray source. This layer is so thin that it is practically transparent to the X-rays generated within it, even at small grazing angles.

In the fluorescent source the primary X-rays penetrate the source material to a relatively much greater depth before being absorbed, and the material emits fluorescent X-rays like a Lambert-type source. Hence the intensity continues to increase up to large viewing angles, $\psi = 30$–$75°$, depending upon the spectral distribution of the primary beam and the absorption characteristics of the irradiated material.

(*b*) *Size factor*

In the case of the large fluorescent source used in Method II, the value of ψ that gives the maximum

intensity is dependent upon the apparent size of the source, the position and size of the source slit and the angular aperture. This effect was measured using the following conditions: large copper block, source slit $1{\cdot}65\times10$ mm., $2\alpha = 2°$, two parallel slit assemblies each with an angular aperture $2\delta = 4{\cdot}5°$, 40 kV.p., unfiltered and the (111) peak of silicon powder. With $\psi < 4°$, the source slit saw a greater area of the fluorescent copper source than was irradiated by the primary beam, and hence the intensity fell off rapidly with decreasing ψ. At $\psi = 4$–$6°$, X-rays from the entire illuminated copper source passed through the source slit and the intensity was at a maximum. As ψ was increased beyond 6° the source slit saw a smaller area of the illuminated copper source and the peak intensity gradually diminished. The peak intensity in the region of $\psi = 4$–$6°$ was approximately one-third greater than at $\psi = 2{\cdot}5°$ or $\psi = 20°$. The scattered background continued to increase with ψ and was three times greater at 20° than at 2°.

(*c*) *Brightness*

The brightness of a source of X-rays is the intensity per unit projected area of the source in the direction of observation. The intensity of X-rays passing through the divergence slit D of Fig. 3 is determined by 2α

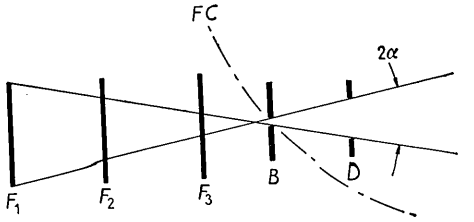

Fig. 3. The required sizes of fluorescent sources F of equal brightness placed at different positions to produce the same total intensity at D are shown by the 2α lines.

and the brightness of the source. The sizes of fluorescent sources of *equal brightness* at F_1, F_2 and F_3 required to give the same intensity at D are shown by the 2α lines. The total intensity passing through D is the same when a large fluorescent source is placed at F_1, or a small fluorescent source the size of the opening at B is placed on the focusing circle. Therefore, other conditions being equal, Methods I and II give the same intensity.

The brightness of the direct electron excited or the fluorescent X-ray source is obtained simply by dividing the observed intensity by $\sin\psi$. This is shown in Fig. 4 for the I versus ψ curves of Fig. 2. The rapid increase in brightness with diminishing ψ for the direct electron-excited X-rays is in marked contrast to the small increase obtained with fluorescent sources.

(*d*) *Intensity*

An approximate order-of-magnitude comparison of the direct and X-ray excited sources will be helpful in understanding the origin of the large difference of intensity produced by these sources.

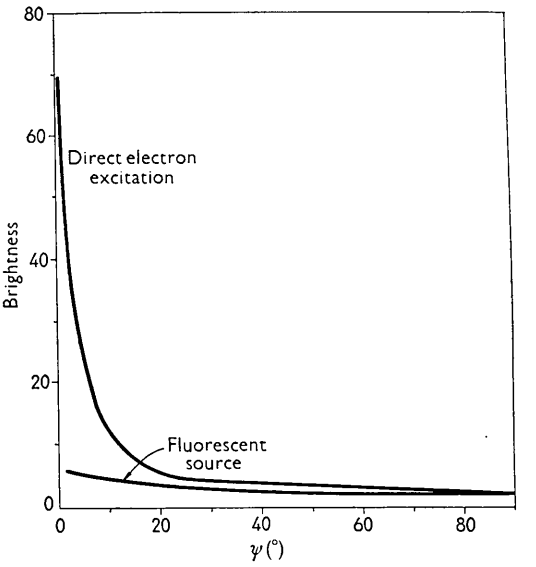

Fig. 4. Brightness as a function of angle-of-view ψ of direct electron excited and fluorescent sources. These were derived by dividing the data of Fig. 2 by $\sin\psi$.

In all X-ray sources the brightness for a given ψ angle is determined by the number of excited atoms per unit time per unit area. In the direct electron-excited case this is, of course, dependent upon the applied voltage and current. In the fluorescent case several additional factors reduce the brightness by a large amount. The fluorescent source receives only $\Gamma/4\pi$ of the primary radiation, where Γ is the solid angle the source subtends at the X-ray tube (Fig. 5). In the case of a source $1{\cdot}5\times10$ mm. and a 25 mm.

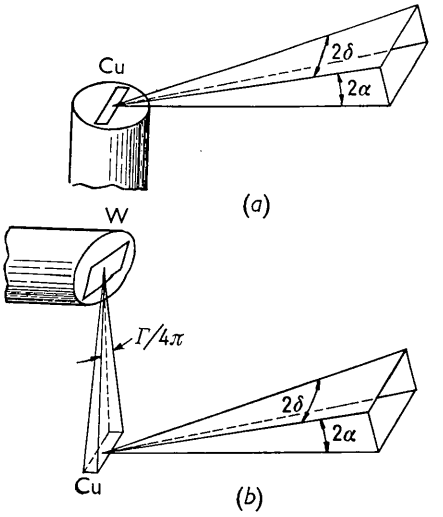

Fig. 5. Comparison of direct electron excited X-ray source (*a*), with fluorescent X-ray source (*b*), showing the large solid-angle loss.

distance between the primary X-ray target and fluorescent source, the latter receives only about 1/500 of the total primary radiation. Moreover, only a portion of the primary spectrum striking the specimen is useful in exciting fluorescence. The very high energy portion penetrates too deeply, causing losses due to self-absorption in the source, and a portion has wavelengths greater than the absorption edge of the source element. If we add to this the fluorescent yield (Compton & Allison, 1935), a factor of about one-fifth must be introduced. The only gain is the 2·5 times greater permissible power loading of the X-ray tube for the fluorescent source.* Taking all these factors into consideration, the intensity of the fluorescent source is lower by a factor of about 1000, and the largest factor by far is the solid-angle loss.

(*e*) *Peak-to-background ratio*

The total intensity was so low with the fluorescent sources that it became extremely difficult to obtain significant measurements of the background or peak-to-background ratio, and therefore these data are omitted from the tables. Even if the background were eliminated, the great reduction of peak intensity would cause a much more unfavorable intensity statistics problem than occurs in normal diffractometry.

We were unable to completely eliminate the background. Although the fluorescent source container was carefully designed, anti-scatter slits were used on the collimator system and the X-ray tube, and extra lead shielding surrounded the tube, there was always some continuous spectrum from the primary tube scattered by the fluorescent source. In addition, a recording of the 10.1 line of a single crystal quartz plate, using an unfiltered Cu fluorescent source, showed the W $L\alpha$ lines from the tungsten target of the primary tube (which were scattered by the Cu source) to be about 1% of the Cu $K\alpha$ lines.

In conventional electron-excited X-ray tubes the detector system also plays an important role in determining the recorded peak-to-background ratio (Parrish & Kohler, 1956*a*, *b*). If pulse-height discrimination is used with scintillation or proportional counters for measuring powder patterns, the peak intensity is reduced by less than 10%, and there is a large reduction in the recorded scattered background. The peak-to-background ratios are then comparable to those obtained when highly efficient focusing crystal monochromators are added to the diffractometer. The background remaining after discrimination contains at least as much characteristic line radiation as non-characteristic wavelengths so that the greatest further improvement would be a reduction of the background by a factor of about 2 or less. It is therefore unnecessary to use fluorescent sources to achieve a low background.

4. Instrumental factors

The major instrumental factors will be briefly described in the following sections, and the use of a well-crystallized specimen with no appreciable specimen fluorescence, as well as a perfectly aligned goniometer, will be assumed.

(*a*) *Source size*

In standard diffractometry, using the conditions given in Table 1, and a receiving slit 0·15 mm. high,* the line width, W, measured at one-half peak height is 0·10° 2θ for the $K\alpha_1$ component in the front-reflection region. W increased only slightly ($< 0{\cdot}01° 2\theta$) when the projected height of the line source was increased from 0·08 to 0·16 mm.; this was done simply by increasing ψ from 3° to 6°. The intensity then also increased by about 25% (see § III(*a*)). When the spot source was used, the projected height was 1·1 mm. at $\psi = 6°$, $W = 0{\cdot}31°$, and the intensity was about the same as the line source. Thus increasing the source height by a factor of 2 caused practically no increase in W; but a further increase of source height by an additional factor of 5 increased the line width by a factor of 3.

(*b*) *Goniometer radius*

When R is decreased from 17 to 14 cm., a small increase of intensity may be expected due to decreased air absorption and the larger chord of the ring intercepted by a receiving slit of fixed width. These factors are 72·5/67·8 (for Cu $K\alpha$) and 17/14 respectively, but the linear dispersion is decreased by 14/17.

(*c*) *Aperture*

If the source has a uniform intensity distribution, the intensity increases directly with 2α. However, the specimen must be sufficiently long to intercept the full length of primary beam at the smallest 2θ angle to be used. The length of specimen illuminated, L, may be calculated from $L = 2\alpha R/\sin\theta$ radians. Assuming a specimen 2·5 cm. long for both cases, the minimum 2θ angle that can be reached before the primary-beam length exceeds the specimen length is 13·6° for the standard diffractometer and 18·9° for the conversion. Considered another way, one could increase 2α in the standard diffractometer by the ratio of the radii, 17/14, thereby obtaining a 21% increase in primary intensity.

* Currently available commercial sealed-off water-cooled stationary target tubes have a rated specific loading of 25–75 W.mm.$^{-2}$, depending on the target. In the case of cobalt the permissible specific loading is about one-half that of copper so that a five times greater loading is permissible with the large focal spot fluorescence X-ray tube.

* To avoid confusion we will define the *width* of a slit or specimen as the dimension measured in a direction parallel to the goniometer axis of rotation G, the *length* of a specimen as normal to G, and the height of a slit as the dimension measured in the goniometer scanning plane (i.e., vertical plane in the Norelco diffractometer).

The use of a flat rather than a curved specimen causes asymmetrical line broadening (Wilson, 1950). This aberration may be neglected in the present discussion except to note that it would be larger at all 2θ angles for the conversion because L is longer.

In the standard diffractometer two sets of parallel slits limit the aperture 2δ of the beam in the plane normal to Bragg focusing (i.e., normal to the plane of Fig. 1). One set is placed between source and specimen, and the other between specimen and counter tube. By limiting this 'vertical' divergence, more symmetrical line profiles are obtained with an extended line source than would otherwise be possible. Each set with $2\delta = 4{\cdot}5°$ reduces the intensity by a factor of 2, but removing one set causes appreciable asymmetric broadening at small and very large 2θ.

(d) Receiving slit

The intensity, background and line width increase with receiving-slit height. The 111 peak of 10–20μ silicon powder was measured, using a standard diffractometer with $2\alpha = 1°$ and Cu $K\beta$ radiation to avoid the $K\alpha$ doublet separation. Increasing the receiving-slit height from 0·3 to 1·0 mm. (all other conditions remaining unchanged) increased the peak intensity by a factor of 1·5, the line width from 0·16° to 0·36° 2θ and the background by a factor of about 4, so that the peak-to-background ratio was reduced (Parrish, 1956*a*).

(e) Summary of instrumental factors

The effects of some instrumental factors on intensity, resolution and peak-to-background ratio are listed in Table 2. From the analysis given in §§ IV(*a*)–(*d*), we see that increasing 2α from 1·0° to 1·7°, removing one set of parallel slits, increasing the receiving-slit height from 0·3 to 1 mm., and decreasing the goniometer radius from 17 to 14 cm. gives an increased intensity by the product of the individual factors, namely, $1{\cdot}7 \times 2 \times 1{\cdot}5 \times 1{\cdot}07$ (air transmission) $\times 1{\cdot}2$ (halo length intercepted by slit) = 6·4. At the same time, the line width would be greatly increased and asymmetrically broadened, and the peak-to-background ratio and dispersion would be decreased. Coyle & Garrod (1956) reached similar conclusions on line breadth but did not take up the intensity problem.

Table 2. *Effect of slit heights on intensity and line width(a)*

Source slit	2α	Receiving slit	Peak intensity(*b*)	Line width at half peak height
1·65 mm.	2·1°	4·6 mm.	154 counts sec.$^{-1}$	1·23° (2θ)
0·83	1·6	1·7	88	0·54
0·45	1·4	0·8	38	0·35
0·45	0·8	0·8	22	0·30
0·45	0·8	0·3	12	0·20

(*a*) Measurements obtained with large Cu source, Method II, 40 kV.p., 40 ma., unfiltered, scintillation counter without pulse-height discrimination, silicon powder specimen, 111α line.
(*b*) Background subtracted.

5. Results

A few typical measurements with fluorescent sources are listed in Table 3. These measurements were made on very strong powder lines, and it is doubtful if a complete powder pattern, including the weak reflections, can be obtained with the usual type of ratemeter recording. The highest intensity of a powder line was 175 counts sec.$^{-1}$, and it would require about 1 sec. to obtain a probable error of 5%, and 26 sec. for 1%, assuming no background. For the same probable errors, the recording of lines 1/25 as strong, which are not uncommon in powder patterns, would require 26 and 641 sec., respectively. Because these statistical factors are always present in counter-tube measurements, it would require very unusual conditions to make it feasible to eliminate the background at the expense of so large a loss of intensity.

These results may be compared with those obtained with the standard diffractometer equipped with a scintillation counter and pulse-height discrimination,

Table 3. *Data obtained with fluorescent source(a)*

Method	II	II	II	II	II	I
Fluorescent source	Co	Co	Cu	Cu	Cu	Cu
ψ	10	5	5	5	5	3
Filter (mm.)	0·015 Fe	0·015 Fe	None	0·018 Ni	0·018 Ni	None
Source slit height (mm.)	1	1	1	1	0·15	—
2α	1·7	1·7	1·7	1·7	1·2	1·2
Receiving-slit height (mm.)	0·83	1·0	1·0	1·0	0·60	0·83
Specimen, *hkl*	Fe, 110	Q, 10.1(*d*)	Ni	Si, 111	Si, 111	Si, 111
Peak intensity (counts sec.$^{-1}$)(*b*)	175	2430	170	34	9(*e*)	19
Line width(*c*)	0·60	0·60	0·50	0·40	0·20	0·29

(*a*) Machlett OEG-50 tungsten target primary source, 50 kV.p., 40 ma., R = 14 cm., 1 set of parallel slits, argon-filled Geiger counter. The experimental arrangement was essentially a duplicate of that used by Weiss *et al.* (1954).
(*b*) Background subtracted.
(*c*) Expressed in °2θ measured at half peak height.
(*d*) Quartz single crystal plate cut parallel to 10.1.
(*e*) Scintillation counter with pulse-height discrimination.

a copper-target X-ray tube operated at 1000 kV.p.×ma., 0·018 mm. Ni filter and 0·15 mm. receiving slit. The peak intensity of the 111 line of a silicon powder specimen was over 9000 counts sec.$^{-1}$, the peak-to-background ratio was 134 and the line width 0·11° 2θ.

The intensities with the fluorescent source are thus about a few tenths of one percent of those obtained with the standard diffractometer under conditions of comparable resolution. When the slit sizes and apertures were increased, the intensity of the fluorescent source was still only 1% of the standard diffractometer and the resolution decreased by a factor of 4. These results are completely different from those of Weiss *et al.*, who stated, 'With only a 20% decrease in resolution the intensities differed by less than a factor of 2 while the background was reduced by a factor of about 25'.*

Large single crystal plates gave considerably greater intensities, but the fluorescence method is not feasible for the tiny crystals employed in structure work.

* Weiss *et al.* operated their Co diffraction tube at 30 kV.p. and 10 ma. but the tube may be safely operated at 50 kV.p., 10 ma. Thus they could have obtained about 3–4 times more intensity with the direct electron-excited X-ray tube.

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La Probabilité Élémentaire des Positions Atomiques

PAR E. F. BERTAUT

Laboratoire d'Electrostatique et de Physique du Métal, Institut Fourier, Grenoble, France

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When nothing is known about a structure, the elementary positional probability of atoms is the elementary hypervolume of coordinates $dx_1 \ldots dx_t$. If any information about the structure is gained (interatomic distances, forbidden volumes, known signs of structure factors), the positional probability has the more complete form $\psi(x_1, \ldots, x_t)dx_1 \ldots dx_t$. $\psi(x_1, \ldots, x_t)$ is Fourier-analyzed in its most general form, for which some examples are given.

Introduction

Nous rappelons explicitement le théorème* suivant (Bertaut, 1955*a*): 'La probabilité composée $P(A_1, \ldots, A_m)dA_1 \ldots dA_m$ pour que les fonctions $E_k(x_1, \ldots, x_t)$ $(k = 1, \ldots, m)$ prennent des valeurs comprises entre A_k et A_k+dA_k est telle que

$$P(A_1, \ldots, A_m) = \int_{\ldots}\int \delta(E_1-A_1)\ldots\delta(E_m-A_m)d\pi(\mathbf{x}_1, \ldots, \mathbf{x}_t) , \quad (1)$$

où les $\delta(E_k-A_k)$ sont des fonctions de Dirac et

$$d\pi = \psi(\mathbf{x}_1, \ldots, \mathbf{x}_t)d\mathbf{x}_1, \ldots, d\mathbf{x}_t \quad (2)$$

est la probabilité élémentaire de trouver le point $(x_1, \ldots, x_t)$ dans l'élément de volume $dx_1 \ldots dx_t$.' On doit avoir

$$\int d\pi = 1 . \quad (3)$$

En cristallographie, les fonctions E_k sont des facteurs de structure normalisés sous forme analytique, les A_k sont les valeurs observées de ces facteurs de structure et les variables $\mathbf{x}_j$ $(j = 1, \ldots, t)$ sont les coordonnées atomiques.

* Une démonstration mathématiquement complète, accompagnée d'exemples d'application de ce théorème a été récemment donnée (Sponsler, 1957).