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An Integrating Weissenberg Apparatus for X-ray Analysis*

By E. H. Wiebenga and D. W. Smits

Laboratorium voor Anorganische en Physische Chemie, University Groningen, Netherlands

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A modification of the ordinary Weissenberg apparatus is described. With the modified instrument it is possible to measure the integrated reflexion intensities with an accuracy equal to, or even better than, that obtained with an integrating photometer, and in a time comparable with that required for visual estimation. The modification consists of a simple mechanical device by which at the end of every translation the camera is given a small additional movement in the direction of its axis and a small rotation. Each reflexion is thereby recorded with a series of slight displacements in two directions. The overlap generates a small parallelogram, the density distribution in which shows a flat plateau in the centre. From this the integrated reflexion intensity can be directly obtained. Constructional details of the integrating mechanism are given and some results are shown.

1. Introduction

To measure accurately the intensities of a great number of single-crystal X-ray reflexions requires a time which is comparable to the total time of the determination of the crystal structure in cases where, by the presence of heavy atoms or other methods, direct information on the phases can be obtained.

A modification of the ordinary Weissenberg apparatus has been designed to reduce the time required for the intensity measurements without loss of accuracy. The principle of this photographic method is to carry out the integration of the intensity in each spot automatically during the exposure, thus avoiding the tedious integration with an integrating photometer.

2. Integrating procedure

An ordinary Weissenberg apparatus is provided with a simple mechanical device by which at the end of every translation the camera performs the following two additional movements: (1) a small translation parallel to its axis (always in the same sense), and (2) a small rotation around this axis. The mechanism returns the camera to its original position with regard to its rotational movement after, say, every 13 of these displacements. During the whole exposure, which involves a multiple of 14 camera translations, 140, for example, each reflexion is thus recorded consecutively 140 times as a series of slightly displaced spots, lying within a small parallelogram (Fig. 1). When the total displacements in the two directions exceed the dimensions of the spot, a plateau of constant density will be found in the centre of the parallelogram. This density represents the *integrated* intensity of the spot, irrespective of its shape, since at each point of the plateau the intensities within the spot are superimposed at a great number of different equally spaced points.†

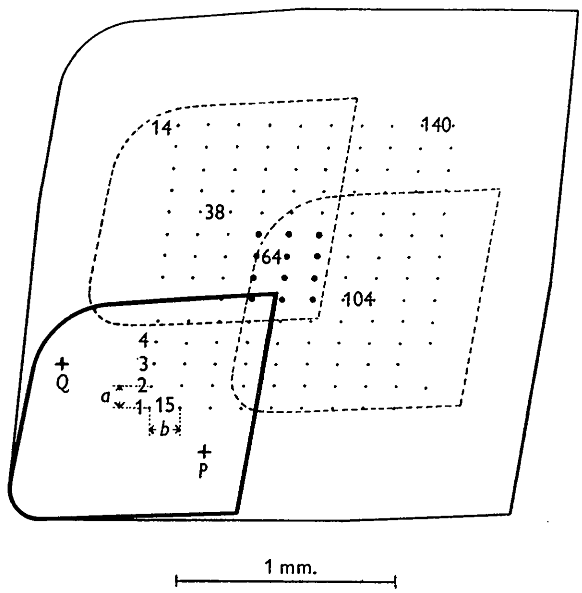


Fig. 1. Successive positions of the spot on the film. The spot is drawn in the positions 1, 38 and 104; in the other positions only its centre is indicated. In each point of the plateau, indicated by heavy spots, the total integrated intensity of the reflexion is measured, e.g. in point 64 point P of the spot is recorded when the spot is in position 38, point Q when the spot is in position 104, etc.

The intensity on the plateau relative to the background may be measured with an ordinary microphotometer, reducing the density to intensity afterwards, or with a double microphotometer, where the intensity on the plateau is directly obtained by comparison with a linear intensity scale.

* A preliminary account has been given by Wiebenga (1947).

† After the preliminary description of the apparatus we became aware of a note of Brentano & Froula (1945), giving an analogous method for powder diagrams.

The conditions for accurate integrating are:

(1) The steps a and b (Fig. 1), representing the displacements after every rotation and after every set of (14) additional translations respectively, must be chosen sufficiently small, namely, so small that a reduction of these steps has no appreciable influence on the (relative) integrated intensities. The choice of the magnitude of these steps will depend on the intensity distribution in the spots concerned.

(2) The total displacements in the rotational and the translational direction ($13\times a$ and $9\times b$ in Fig. 1) must exceed the dimensions of the spots to be measured. The excess, which determines the size of the flat plateau formed, should be at least 0·2 mm., so that the density of this plateau can be measured photometrically without inaccuracy due to the grain of the film.

(3) The output of the X-ray tube must be reasonably constant. It appeared, however, that variations in the output of the X-ray tube of about 10 % did not affect appreciably the values obtained for the integrated intensities.

With our instrument the total displacement in the rotational direction can be chosen between 0·8 and 2·5 mm. and involves generally 13 steps a. This number of steps, however, may be changed easily to 9 or 17. The translational step b is variable from 0 to 0·35 mm.; the total displacement in the translational direction is practically unlimited.

In most cases total displacements of about 1·3 mm. are sufficient to cover even those spots which are split into $\alpha_1\alpha_2$ doublets. The steps a and b are generally about 0·1 mm. However, from the possible variations indicated above, it is seen that much smaller steps may be chosen for small spots, namely, rotational steps down to 0·05 mm. and translational steps down to 0 mm. This is a distinct advantage over the integration of the intensity of spots by means of an integrating photometer, where the diameter of the scanning light beam cannot be reduced to less than about 0·1 mm. on account of the coarseness of the film grain. In case of a long regular needle-shaped crystal integration is required only in the rotational direction, and the additional translation is made equal to zero.

When taking a photograph with the integrating Weissenberg apparatus the minimum number of ordinary camera translations is fixed by the minimum number of integration steps. To obtain a correctly exposed photograph either the speed of the camera drive or the output of the X-ray tube must be adapted. Now, using a self-rectifying X-ray equipment, the former must be so low that the interval of time in which each reflexion takes place includes a sufficiently great number of cycles of the alternating current. This requirement gives a minimum time of approximately 90 sec. for each camera translation, assuming a 180° crystal rotation and an alternating current of 50 cyc./sec. The exposure time for an integrated Weissenberg diagram, involving, for example, 140 steps, thus amounts to approximately 4 hr. The output of the X-ray tube has to be adapted to this minimum exposure time.

The time of exposure required for taking an integrated Weissenberg diagram is somewhat longer than that for an ordinary Weissenberg photograph. As will be seen from Fig. 1, this increase is roughly determined by the ratio of the total number of steps (140) to the number of steps in which each spot is scanned (about 70 in Fig. 1). As a result of this increased exposure time the intensity of the general background on the film is increased by the same factor. This is the only—and not very serious—disadvantage as compared with the integration by an integrating photometer.

3. Results

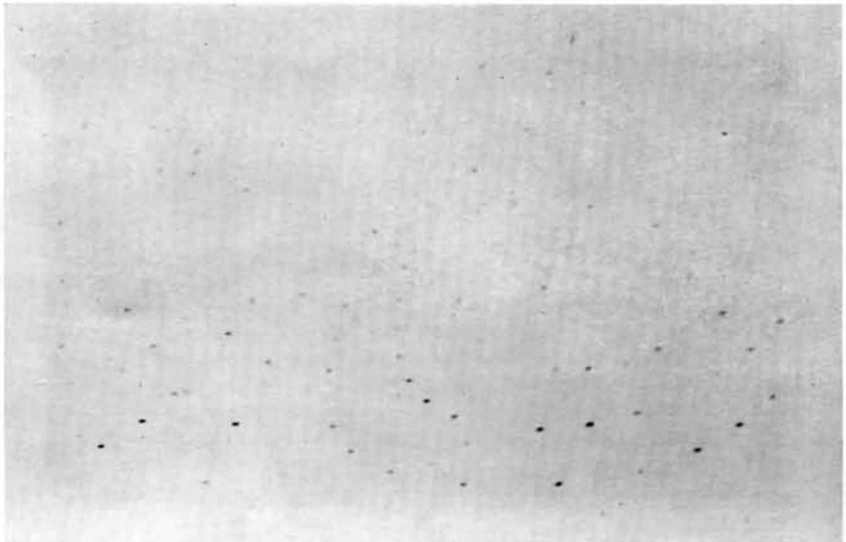
Examples of photographs made with the apparatus are given in Fig. 2. In Fig. 2 (*a*) an ordinary Weissenberg photograph is shown, taken by putting the integrating mechanism out of action. Fig. 2 (*b*) shows the corresponding integrated diagram. It may be seen that the spots on the ordinary Weissenberg photograph are of different shapes, so that a visual estimation of the intensities is difficult. Furthermore, the spots are so small that integration of the intensities by means of an integrating photometer would involve large inaccuracies.

The integrated intensities of eight spots of an integrated Weissenberg photograph were measured by determining photometrically the intensity difference between the plateau and the background around each spot. The result is given in arbitrary units in the first column of Table 1. To check these values the intensities of the same spots were measured with an integrating photometer (column 2). As may be seen, the ratio of the corresponding values in columns 1 and 2 (column 3) is constant to within 2·5 % on the average.

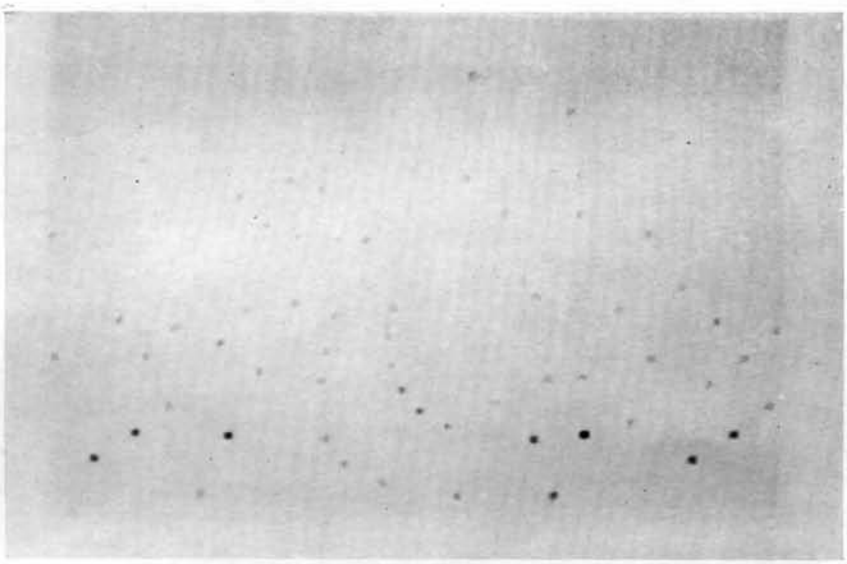
Table 1. *Comparison between the intensities of the flat plateaux of eight spots on an integrated Weissenberg photograph and the intensities of these spots as measured by means of an integrating photometer*

Flat plateau intensity I_1	Integrating photometer I_2	I_2/I_1	Discrepancy (%)
100·0	*100·0*	*1·00*	+1·5
94·6	97·4	1·03	+4·5
39·4	39·9	1·01	+2·5
40·6	37·8	0·93	−5·5
40·2	37·8	0·94	−4·5
37·8	37·8	1·00	+1·5
28·6	28·1	0·98	−0·5
14·9	14·7	0·99	+0·5
		Av. $0·98_5$	Av. 2·5

In the course of a three-dimensional Fourier synthesis it appeared that the reflexion intensities could be measured on integrated Weissenberg photographs in a time less than 1 min. per reflexion. This means that 1000 reflexions may be measured in 3 days. A visual estimation of the intensities of 1000 reflexions would take approximately the same time and give far less reliable results (compare Kaan & Cole (1949)).



(a)



(b)

Fig. 2. (a) Ordinary Weissenberg photograph. (b) Integrated Weissenberg photograph.

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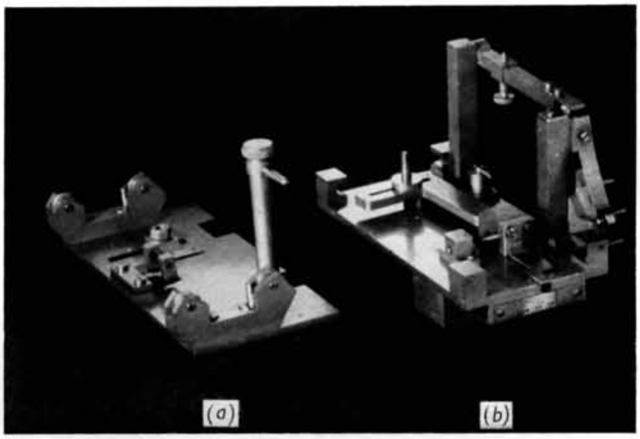

Fig. 3. (a) Upper camera support. (b) Lower camera carriage

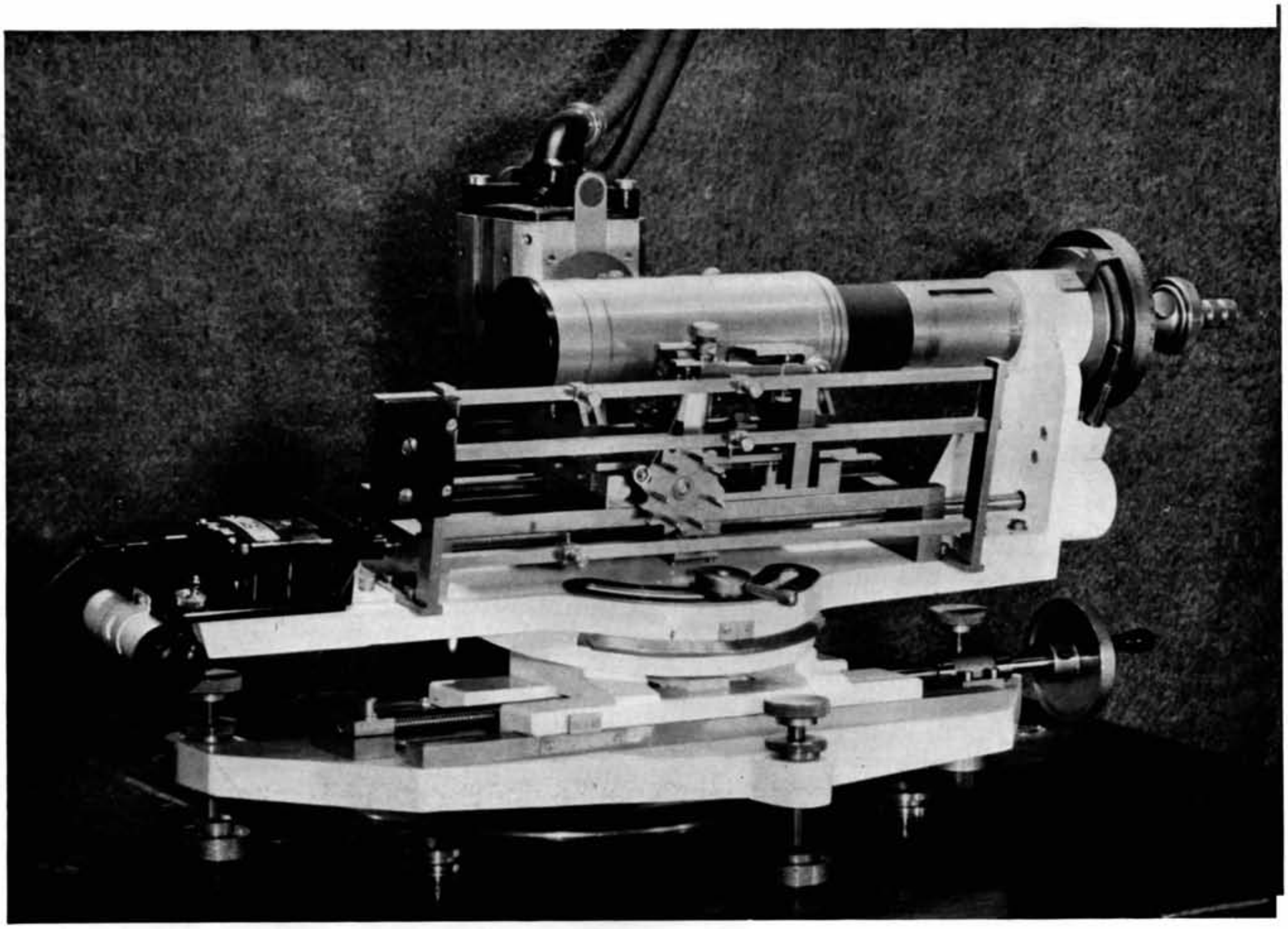

Fig. 4. The complete integrating Weissenberg apparatus.

4. Constructional details

The main features of the apparatus have been taken from a description by Buerger (1942) with the exception of the integrating mechanism and some minor details. (For instance, the direct-beam trap is not attached to the layer-line screen but directly to the spindle bearing in a stationary position. Furthermore, equi-inclination exposures are possible up to $\mu=45°$.)

The extra translations of the camera are obtained by mounting the camera on a support (Fig. 3 (*a*)) which is movable with respect to the camera-carriage proper (Fig. 3 (*b*)) in the direction of the camera axis. The support and the carriage are connected by four balls rolling between short grooved rails.

The small rotations of the camera are made possible as follows. The camera rests on four wheels, two of which fit in a groove in the camera to locate it in the translation direction. The camera is kept in place by a circular clasp gripping a roller (Fig. 3 (*a*)) which is attached to the support by spring blades.

The integrating mechanism is operated by a ratchet wheel provided with pins (Figs. 3 (*b*), 4 and 5). Towards the end of every ordinary camera translation one of the pins is caught by a stud, causing the wheel to rotate through a certain angle, always in the same sense. The wheel is locked in each new position by a spring blade and roller engaging with one of its notches. For most purposes a wheel with 7 pins and 14 notches may be used, the rotations of the wheel being 360/14 degrees.

As may be seen from Figs. 3 (*b*) and 5, the rotation of the ratchet wheel is converted into a displacement of a nut by means of a short screw mounted on the lower carriage. This displacement in turn is converted by an L-shaped lever into a small translation of the camera support with respect to the carriage. The connexion between the long arm of the lever and the nut consists of a ball-and-socket joint; the short arm is connected to the camera support by a pin, attached to the support by a nut and fitting into a slot in the short arm of the lever. The effective length of the short arm, and thus the magnitude of the extra camera translations, can be varied by changing the position of this pin.

The rotation of the camera is also a result of the rotation of the ratchet wheel. A rod, moving vertically in a bearing, rests on a spiral cam (Figs. 3 (*b*) and 5) directly connected to the ratchet wheel. The vertical displacement of the rod, resulting from every fractional rotation of the cam, is converted into a small camera rotation by means of a lever, seen at the top of Fig. 3 (*b*) and in Fig. 5. A wide lug on the camera (Fig. 4) is pressed on a stud on this lever by a helical spring (concealed in the vertical tube, Fig. 3 (*a*)) operating on the second, narrow lug. The leverage, and thus the magnitude of the small camera rotations, is variable by changing the position of the stud on the lever.

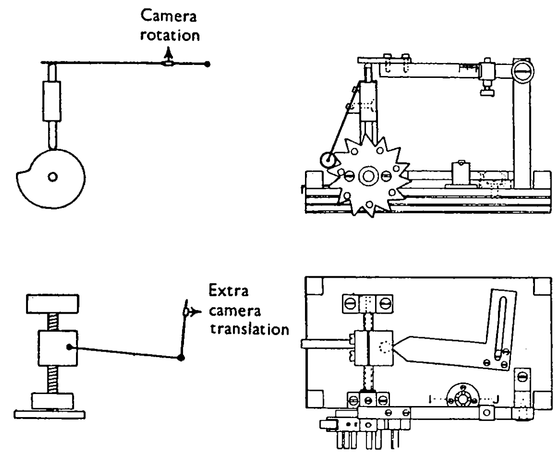

Fig. 5. Schematic diagram of the integrating mechanism.

The apparatus was made in our workshop by Messrs J. C. Heinen and A. v. d. Meulen, to whom we are much indebted for their careful work. The instrument can be obtained commercially from the N. V. Nederlandsche Instrumentenfabriek 'Nonius', Van Leeuwenhoek-singel, Delft, Netherlands. Our thanks are due to the Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek i.o. for stimulating the further work with the apparatus by a grant, and to Prof. Dr J. M. Bijvoet, Utrecht, who kindly put the integrating photometer of his laboratory at our disposal.

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