

65

# ACTA

GEOLOGICA ET GEOGRAPHICA  
UNIVERSITATIS COMENIANAE

## GEOLOGICA

NR. 14

SEPARÁTNY VÝTLAČOK

1968

SLOVENSKÉ PEDAGOGICKÉ NAKLADATELSTVO BRATISLAVA

## X-RAY TOPOGRAPHIC TECHNIQUES

*Andrew Richard Lang*

## I. Review of Topographic Techniques

X-ray topography is one of a group of techniques which record point-by-point the distribution of certain properties it is desired to measure in a specimen. Experimental arrangements for diffraction topography are related to those of absorption topography, and combined diffraction and absorption topographic studies of natural<sup>(1)</sup> and synthetic<sup>(2)</sup> diamond, and of amethyst quartz<sup>(3)</sup>, have proved informative. Similarly, studies combining X-ray diffraction, birefringence and ultra-violet absorption topographs of the same specimens have been performed<sup>(4)</sup>, with the production of more instructive records than could have been obtained from the use of each technique separately. It is, however, with the technique of transmission electron microscopy, which has been so powerfully developed in the last decade by Hirsch and his colleagues<sup>(5)</sup>, that X-ray diffraction topography is most closely linked theoretically. From the many resemblances between X-ray topographic and electron micrographic images, and even more from the significant differences between them, a powerful stimulus to new investigations in diffraction theory has been derived. Of this work the many papers of Kato form an eminent record. On the practical side, the sad fact that all X-ray topographic methods have to labour without the benefit of magnification through X-ray lenses means that even with the greatest care the magnifications realisable are only about one thousandth of those achieved very easily in electron microscopy. Indeed, several fundamental factors conspire to limit the resolution achievable in X-ray topography to about one micron. It follows that rather different aspects of lattice defects are studied by transmission electron microscopy and by X-ray topography, respectively. Some offset to the disadvantage of the low resolution of X-ray topography is gained by its ability to examine the distribution of imperfections within quite large crystals, and to make repetitive studies of the same specimen after it has undergone various treatments. The introduction of the high-voltage electron microscope, operating at 500 kV or more, with which specimens several microns thick can be penetrated, will close the gap between the specimen thickness ranges in which X-ray and electron diffraction contrast are observable. The fields of application of both techniques will be enlarged thereby.

The discovery that individual dislocations could be detected by X-ray topography<sup>(6-9)</sup> (which took place in four laboratories independently, and

with the use of quite different techniques) generated increased interest in these methods and led to their application to a variety of problems concerning crystal imperfections. X-ray topographic techniques, and some of their applications, have been discussed in several reviews<sup>(10-13)</sup>. The basic geometry of topographic methods will now be outlined, following generally the classification previously adopted.<sup>(11)</sup>

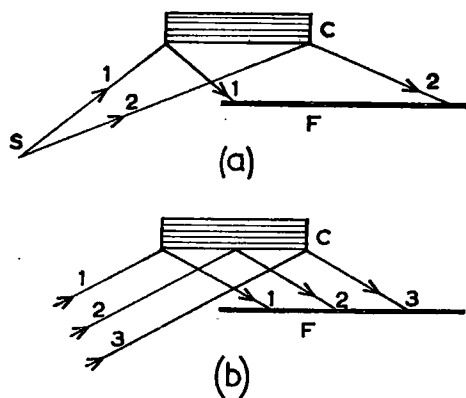


Fig. 1. X-ray source *S*, crystal *C*, and photographic plate *F*. Geometry of rays forming image of crystal surface with (a) continuous radiation and (b) characteristic radiation.

The term 'X-ray topograph' was introduced by Ramachandran<sup>(14)</sup>, whose transmission specimen technique, reported in 1944, is described below; but the earlier work of Berg<sup>(15)</sup> can reasonably be regarded as the starting point of X-ray topography. Berg pointed out the difference between forming an image of a crystal surface by specular reflection of light and by Bragg reflection of characteristic X-rays. Consider the arrangements shown in Figure 1. Reflection is to be obtained from planes parallel to the surface of the crystal *C*. If *S* is a point source of light (Fig. 1(a)), and the crystal surface reflects specularly, then all rays between the limiting rays 1 and 2 can be reflected at the crystal surface and they form an image on the film *F*. If, on the other hand, an image of all of the face of *C* is to be obtained by characteristic X-rays (ideally including a single wavelength only) which are Bragg-reflected by the planes parallel to the surface of *C*, then, in order to satisfy the Bragg condition simultaneously with all rays impinging upon *C*, the incident rays (such as 1, 2 and 3 in Fig. 1(b)) must all make the same angle with *C*. Thus they must either come from different points of an extended source (this was the arrangement used by Berg), or they must come from a small source so far away that the angle between 1, 2 and 3 does not exceed the angular range of reflection by the crystal *C*.

Another alternative is that 1, 2 and 3 are rendered parallel by prior Bragg-reflection at the surface of a large perfect crystal; this gives the strictest control over the range of divergence of 1, 2 and 3. If, on the other hand, an image of *C* is to be recorded with continuous X-radiation then the conditions imposed on angular range of incidence are completely relaxed and the geometry of rays is again as shown in Fig 1(a). A convenient basis for classifying topographic techniques is the nature of the incident radiation used, whether continuous, or characteristic with varying degrees of collimation. The nature of the incident radiation, and also the ratio of distance from source to specimen (a) to that from specimen to photographic plate (b),

determining the topographic point-by-point. The latter of the image being characteristic is obtained (10). The



lattice in Figure 2, which is a misorientation. finite misorientation is as perfect where the or it could energetic rest of the of Fig. 1 is quite clear show up would not the image like Fig 2 ation is s the angular orientation appear as as it does can be of image such tion due t due to the ristic radiation specimen-to-continuous radiation bounded t

rest in these  
concerning  
their appli-  
geometry of  
the classifi-

determine the form the record of various types of lattice imperfection takes on the topograph. Now the essential quantities that topographs can record are point-by-point differences in local lattice *orientation* and local lattice *perfection*. The latter may be indicated sensitively by recording point-by-point the values of the integrated reflecting power of the crystal, the experimental conditions being chosen so that the variation of integrated reflection with degree of perfection is large. (In the case of transmission specimens high sensitivity is obtained when the absorption is either quite low,  $\mu t \leq 1$ , or quite high,  $\mu t \geq 10$ ). The different responses of various topographic arrangements to certain

crystal  
late *F*.  
ng ima-  
with  
on and  
ation.

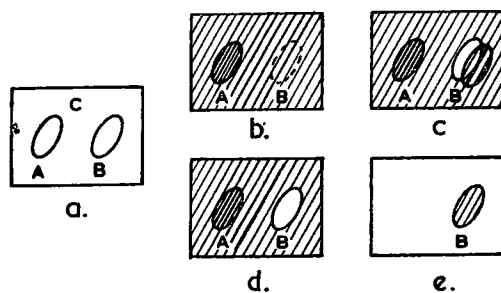


Fig. 2. (a) Crystal surface (*C*) with imperfect region (*A*) and misoriented region (*B*), (b) continuous radiation topograph with *b* very small, (c) continuous radiation topograph with *b* increased, (d) characteristic radiation topograph, main reflection, (e) characteristic radiation topograph, only *B* reflecting.

(14), whose  
below; but  
starting point  
forming an  
agg reflection  
Figure 1. Re-  
e crystal *C*.  
reflects spe-  
reflected at  
other hand,  
istic X-rays  
reflected by  
the Bragg  
cident rays  
. Thus they  
his was the  
ource so far  
lar range of

d by prior  
s gives the  
If, on the  
liation then  
letely rela-  
convenient  
incident ra-  
degrees of  
of distance  
ic plate (*b*),

lattice imperfections can be illustrated by the idealised example shown in Figure 2. The surface of the crystal *C* (Fig. 2(a)) is supposed to have an area *A* which is more imperfect than the surrounding crystal but which has no gross misorientation with respect to its surrounds, and an area *B* which has a definite misorientation with respect to its surrounds but within which the lattice is as perfect as that surrounding it. In practice, *A* might represent a region where there was a relatively high density of randomly distributed dislocations or it could be a region of localised radiation damage by a beam of X-rays or energetic particles. Area *B* might be a sub-grain slightly misoriented from the rest of the crystal. When continuous radiation is used, with the geometry of Fig. 1(a), the image of *C* would appear as in Fig. 2(b) when the plate *F* is quite close to the crystal *C*, i. e. with  $b \ll a$ . The imperfection in *A* would show up by increased diffracted intensity, but the slight misorientation of *B* would not be obvious. If the distance *b* is increased then the displacement of the image of *B* will be visible and measurable, and the topograph will look like Fig 2(c). When characteristic radiation is used, and the degree of collimation is such that the incident beam has a divergence *greater* than that of the angular range of reflection by a perfect crystal but *less* than the misorientation of *B* with respect to the rest of the crystal then the topograph will appear as in Fig. 2(d): area *A* will produce greater blackening on the plate, as it does in Fig. 2(b), but the image of *B* will be absent. Reflection from *B* can be obtained by appropriate change of the angular setting of *C*, and an image such as Fig. 2(e) will then be recorded. Apart from some loss of resolution due to geometrical factors and from the angular spread of diffracted rays due to the angular range of reflection and wavelength spread in the characteristic radiation, the form of the images (d) and (e) will not change as the specimen-to-plate distance *b* is increased, in contrast to the situation with continuous radiation (Fig. 2(b) and 2(c)). If the misoriented region *B* is sharply bounded then the direction and magnitude of its misorientation with respect to

the rest of the crystal may be measured from the shift of its image when  $b$  is increased, in the continuous-radiation technique. When characteristic radiation is used, and a succession of topographs is taken at various angular settings covering a range sufficient to bring  $B$  into the reflecting position, the component of its misorientation about the normal to the plane of incident and diffracted rays may be measured. Several specimen orientations or Bragg reflections must be used to find the total misorientation of  $B$ , but the precision obtainable is high. Even with the incident beam collimated only by slits, misorientations may be measured to about 1 second of arc. With crystal-reflected radiation, in the double-crystal spectrometer arrangement, misorientations of 1/10 second of arc can be measured. In practice, local variations of reflecting power are invariably associated with local tilts of the crystal lattice and interpretation of the images obtained by the simple techniques using continuous radiation is often difficult. For directness of interpretation, and for high resolution, it is preferable to use the techniques employing characteristic radiation.

A second division of topographic techniques is into those in which the diffracted rays are reflected from the specimen surface and those in which they are transmitted through it. It is certainly arguable that it is in the transmission techniques that the potentialities of X-ray topography are best realised, for they then provide a nondestructive method for studying the imperfections within the interior of crystals, in regions sufficiently far from crystal surfaces so that the imperfection distribution truly represents conditions in the bulk material. This distribution may be unobservable by any other technique: with X-rays, on the other hand, its three-dimensional configuration may be easily recorded. In transmission techniques it is usually necessary to impose slits between the specimen and the plate in order to prevent the image of directly transmitted X-rays overlapping that of the diffracted rays. When characteristic radiation is used, both with reflection and transmission specimens, it is strongly desirable to register only the images produced by the  $K\alpha_1$ -component of the  $K\alpha$  doublet. This demands collimation of the incident beam to an angular divergence of not more than about 1 to 2 minutes of arc. This collimation is usually achieved by use of a fine-focus X-ray tube and a narrow incident-beam-defining slit placed close to the specimen. It follows then that some means must be used for scanning the specimen with the collimated beam: this need led to the development of the moving-specimen techniques. Thus a further division of topographic techniques may be made between those that have the specimen stationary and those in which it moves. A brief outline of the various techniques will now be given. For experimental details the original papers should be consulted. The classification shown in Table I will be followed.

#### (a) Continuous radiation, stationary reflection specimen

This is the simplest of all techniques, but as developed by Schulz<sup>(16)</sup> it produced topographs of high quality and resolution. The geometry is that of Fig. 1(a). Schulz used a microfocussed X-ray tube with a focus whose projected size was about 3 microns by 30 microns. With this small focus good topographic resolution is obtained without having to place the plate very close to the spe-

Specimen
Reflection stationary
Transmission stationary
Reflection moving
Transmission moving

cimen. If the plate is moved but if the specimen is lost unless (a) and (b) reflection feature c:

The geometry is that of Fig. 1(a). The method is the same as that of Schulz's method for Diamond specimens since the planes are not the diffracting

**Table I**  
**X-ray Topographic Techniques**

Specimen	Radiation Incident		
	Continuous	Slit-collimated characteristic	Crystal-reflected characteristic
Reflection, stationary	(a) Schulz <sup>(16)</sup>	(c) Berg <sup>(15)</sup> , Barrett <sup>(17)</sup>	(g) Bond and Andrus <sup>(25)</sup> Bonse and Kappler <sup>(8)</sup>
Transmission, stationary	(b) Ramachandran <sup>(14)</sup>	(d) Lang <sup>(19)</sup>	(h) Authier <sup>(26)</sup> Kohra et al. <sup>(27)</sup>
Reflection, moving	—	(e) Wooster and Wooster <sup>(21)</sup> Merlini and Guinier <sup>(22)</sup> Lang <sup>(23)</sup>	(i) (Lang)
Transmission, moving	—	(f) Lang <sup>(24)</sup>	(j) Hart and Lang <sup>(28)</sup>

specimen. If an almost undistorted image of the crystal surface is required then the plate should be kept parallel to the crystal surface, as shown in Fig. 1; but if the X-rays pass through the emulsion obliquely, resolution will be lost unless the emulsion is very thin. The continuous radiation methods (a) and (b) will generally produce several images simultaneously, since reflection from various Bragg planes will occur, as in a Laue photograph. This feature can be used to advantage in the analysis of misorientations.

**(b) Continuous radiation, stationary transmission specimen**

The geometry of this early topographic technique was thoroughly investigated by Ramachandran<sup>(14)</sup>. It is the transmission-specimen analogue of the reflection-specimen arrangement shown in Fig. 1(a). Ramachandran used the method for the study of polished plates of diamonds up to 1 cm<sup>2</sup> in area. Diamond is an eminently suitable specimen for study by transmission techniques since its X-ray absorption is very low. Ramachandran used reflecting planes nearly normal to the plate surface and he set the plate inclination to the diffracted beam so as to produce an undistorted image of the plate. Ra-

machandran's experiments were performed with the ratio between distance source-to-specimen and distance specimen-to-plate (i. e. ratio  $a/b$ ) equal to 12. Hence his arrangement had the characteristics of Fig. 2(b) and his topographs were sensitive to variations in reflecting power but not to misorientation.

(c) Slit-collimated characteristic radiation,  
stationary reflection specimen

The topographs produced by Barrett<sup>(17)</sup> were a great advance upon the early attempts of Berg and they set the standard for future work with reflection specimens. The arrangement is that of Fig. 1(b). Users of this technique generally choose values of  $a$  between 20 and 100 cm. Barrett, and Newkirk<sup>(18)</sup> following him, used fine-grain photographic emulsions and made special efforts to reduce  $b$ . The latter condition is best satisfied by using Bragg reflections in which the diffracted beam leaves the crystal nearly perpendicularly. This requires large diffraction angles, and the softer radiations such as  $\text{CoK}\alpha$  and  $\text{CrK}\alpha$  are favoured for use in the Berg-Barrett technique.

(d) Slit-collimated characteristic radiation,  
stationary transmission specimen

This is the method of 'section topographs' which was developed for observing imperfections in the interior of lightly absorbing crystals<sup>(19)</sup> and also for studying the geometry of simultaneous reflections<sup>(20)</sup>. It may be understood by reference to Fig. 3, disregarding the double-headed arrow on this drawing.

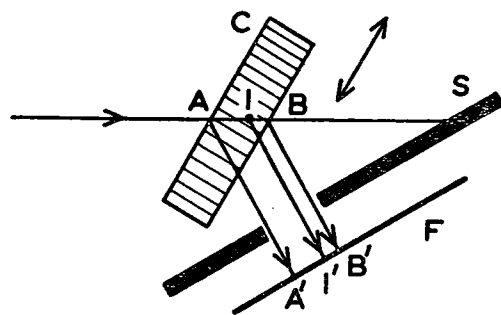


Fig. 3. Geometry of 'section topograph' and 'projection topograph' techniques.

Bragg reflection occurs at lattice planes which are normal, or nearly normal, to the crystal plate  $C$ . A narrow ribbon incident X-ray beam  $AB$  cuts through the crystal as shown. In the arrangement currently in use this beam comes from an X-ray source about 100 microns wide distant about 45 cm from the specimen, and it is defined by a slit (not drawn) placed close to the specimen. The slit has a minimum width of 12 microns, and this width defines the precision with which the position of an interior defect such as  $I$  can be located. The diffracted beam leaving the crystal falls on the photographic emulsion  $F$ , which is set perpendicular to the diffracted beam because thick emulsions, sensitive to the harder radiations, are generally used. The relation between the

width of given<sup>(19)</sup> imperfect screen  $S$  to the in

Several from an In the sc is bathed are fixed angular: the conc sensitive

A diff by Merli Film an a narrow ned by t rays 1, 2 beam.

$F_1$   
du

Another diffracted emulsion undistorted A well-collimated crystal is double an width  $A''$  of the crystal position  $L$  cannot be X-ray foc

width of the section topograph  $A'B'$  and the thickness of the crystal has been given<sup>(19)</sup>. If the absorption is low, i. e.  $\mu t$  not greater than a few units, the imperfection  $I$  will form a localised spot  $I'$  of extra blackening on  $F$ . The screen  $S$ , which intercepts the beam leaving the crystal in the direction parallel to the incident beam, is necessary when diffraction angles are small.

(e) Slit-collimated characteristic radiation, moving reflection specimen

Several techniques have been invented for obtaining Bragg reflections from an extended crystal surface without using an extended X-ray source. In the scheme devised by N. Wooster and W. A. Wooster<sup>(21)</sup> the crystal surface is bathed in radiation diverging from a small source. The crystal and the film are fixed relative to each other and are together rocked through a sufficient angular range to allow all points on the crystal surface to reflect. This simulates the conditions of a continuous-radiation topograph and the method is not sensitive to small changes in lattice orientation.

A different arrangement, capable of high angular resolution, was described by Merlini and Guinier<sup>(22)</sup>. The film is mounted parallel to the specimen surface. Film and specimen are translated together in a direction perpendicular to a narrow incident beam so that the whole surface of a large specimen is scanned by the beam. The geometry is similar to that of Fig. 1(b) where now the rays 1, 2 and 3 represent successive positions in time of the narrow incident beam.

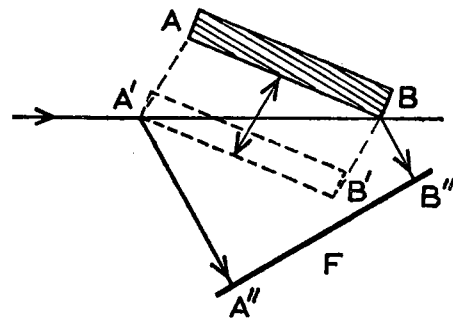


Fig. 4. Arrangement for producing undistorted image with moving reflection specimen.

Another technique has been used by Lang<sup>(23)</sup>. This is designed to allow diffracted rays to fall perpendicularly on the photographic plate, so that a thick emulsion can be used without loss of resolution. At the same time it gives an undistorted image of the crystal surface. The geometry is shown in Fig. 4. A well-collimated incident beam is used and the plate  $F$  is stationary. The crystal is translated back and forth during the exposure in the direction of the double arrow. The direction of translation is that calculated to give an image width  $A''B''$  equal to the width of the crystal face  $AB$ . In the figure, the edge  $B$  of the crystal is Bragg-reflecting to  $B''$ . When the crystal is translated to the position  $A'B'$ , the other edge of the crystal reflects, along  $A'A''$ . In this method  $b$  cannot be made very small. Hence a large value of  $a$  and/or a very small X-ray focus must be used to give good topographic resolution.



(f) Slit-collimated characteristic radiation,  
moving transmission specimen

This is the method of the 'projection topograph'<sup>(24)</sup>. The geometry is the same as that shown in Fig. 3 but now the specimen and photographic plate are together translated back and forth in the direction of the double arrow so that an image of a large volume of crystal is produced on *F*. This image can be regarded as a superimposition of many section topographs, and it is a projection along the diffracted-beam direction of the volume of crystal scanned and of its imperfection content. This projection does not show directly the depth within the specimen of a particular imperfection, but this information can be recovered by taking stereo-pairs of projection topographs. The slit defining the incident beam can be made much wider than the 12 micron width used in taking section topographs. For projection topographs it is only necessary that the incident beam be sufficiently well collimated to allow reflection of the  $K \alpha_1$  component alone to take place.

(g) Crystal-reflected characteristic radiation,  
stationary reflection specimen

The double crystal spectrometer arrangement provides a most sensitive technique for measuring lattice misorientations. In the techniques developed independently by Bond and Andrus<sup>(25)</sup> and by Bonse and Kappler<sup>(6)</sup> the dispersionless 'parallel' double crystal spectrometer arrangement is used, as shown in Fig. 5. An extended X-ray source *S* is employed, and characteristic

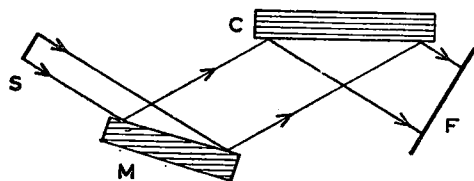


Fig. 5. Double-crystal spectrometer topographic technique.

radiation from it is reflected by the first crystal *M*. By using an asymmetric reflection as shown, the beam impinging upon *C* may be made wider than the beam leaving *S*. Consequently, there is no need to translate the specimen crystal unless a relatively small source *S* is used and the area of *C* to be surveyed is large. The first crystal *M* is assumed to be perfect. Then a series of topographs taken with the angular setting of *C* changed in small steps between each exposure allows differently oriented parts of *C* to be brought in turn into the orientation for peak Bragg reflection and thus to produce maximum blackening on the plate *F*. By rotating *C* through 180° about the Bragg-plane normal and repeating the series of topographs it is possible to separate the effects of misorientation from those due to changes in interplanar spacing. Since the beam leaving the perfect crystal *M* possesses only the divergence corresponding to the angular range of reflection of a perfect crystal it will not permit imperfect regions in *C* to give their complete integrated reflection when *C* is set at a fixed angle. Thus imperfections such as dislocations outcropping at the surface of *C*

will not  
to (f). O.  
rons from

Authic  
with bot  
between  
only a s  
crystals.  
with inci  
waves w  
produced  
Up to no  
but it co  
slightly c

Kohra  
from a p  
in place  
beam as  
-parallel  
each case  
graphs co  
ing spec  
of produ  
may have

As indi  
parallel u  
beams fo  
has been  
mination

will not produce the strong extra blackening that they do in the methods (a) to (f). On the other hand, the small lattice tilts at distances of up to 100 microns from the dislocation core can be detected and measured by this technique.

**(h) Crystal-reflected characteristic radiation,  
stationary transmission specimen**

Authier<sup>(26)</sup> used a 'parallel' double-crystal spectrometer arrangement with both crystals set for symmetrical transmission. A narrow slit placed between the two crystals selected rays whose angular range corresponded to only a small fraction of the full angular range of Bragg reflection by the crystals. In this way he produced 'section topographs' of the second crystal with incident waves which were effectively plane waves instead of the spherical waves which are obtained with slit collimation only. The patterns Authier produced by this ingenious technique are of considerable theoretical interest. Up to now only perfect silicon crystals have been studied by this technique, but it could be used for studying the trajectories of Bragg-reflected rays in slightly distorted crystals.

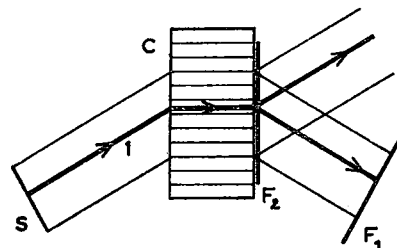


Fig. 6. Arrangements using anomalous transmission.

Kohra et al<sup>(27)</sup> used the wide, parallel beam produced by Bragg reflection from a perfect crystal as a source of X-rays for taking projection topographs, in place of traversing the specimen with respect to a narrow slit-collimated beam as in the usual technique<sup>(24)</sup>. They employed both the parallel and anti-parallel settings for monochromator and specimen crystal and reported in each case an improvement in resolution and contrast of the projection topographs compared with those obtained with the usual slit-collimation and moving specimen. This method suggests interesting possibilities in the way of producing projection topographs with truly plane incident waves which may have useful applications in the study of certain lattice defects.

**(i) and (j) Crystal-reflected characteristic radiation,  
moving specimen, reflection or transmission**

As indicated above, specimen movement is generally redundant when wide, parallel uniform beams of crystal-reflected radiation can be used as the incident beams for producing reflection or transmission topographs. The method (i) has been used experimentally (Lang, unpublished) to ensure the uniform illumination of a large specimen when neither the X-ray source nor the monochro-